APPENDIX II
Cellular Radiobiology

INTRODUCTION

Cellular radiobiology is a well-developed discipline, dating to almost 90 yr ago, when it was first recognized that exposure to ionizing radiation had biological consequences. Much information has since been accumulated from studies with irradiated animals and plants and, over the last 50 yr in particular, from studies of effects at the cellular, physical, and chemical levels. Molecular and cellular radiobiology have contributed greatly both to our understanding of the physical and chemical processes involved in the induction of radiation effects and to our understanding of the responses observed in whole organisms. From the vast store of radiobiological information, it is possible to draw a number of general principles.[26,28,35,36] These principles sometime constitute our only rational basis for making human risk assessments, for example, when direct empirical observations on which to base such assessments are not available.

DOSE-RESPONSE CURVES

Many models have been developed to describe radiobiological responses, but one broad generalization is that empirical dose-response curves for cellular radiobiological effects, whether in vivo or in vitro, take the general form:

$$Y = (a + \alpha_1 D + \beta_1 D^2) \exp(-\alpha_2 D - \beta_2 D^2), \qquad \text{(II-1)}$$

where Y is the response observed in a population, a is the spontaneous incidence, α_1 and β_1 are the coefficients for the induction of the observed

effect, D is the dose, and the exponential is a term expressing the loss of observed response due to competing effects, such as cell killing, with α_2 and β_2 as the coefficients for cell killing. For doses well below that at which competing effects are important, the simpler expression, $Y = a + \alpha D + \beta D^2$, suffices. The relationship is often referred to as mixed quadratic or linear-quadratic, although the expression is simply quadratic. Such an expression adequately describes the dose-response relationship at low to moderate doses of low linear energy transfer (LET) radiation for a wide variety of cellular radiobiological endpoints, including induction of mutations and induction of the various classes of chromosomal aberrations. As described in more detail below, the quadratic expression also describes adequately the responses of such systems to high-LET radiation.

Cell killing is often analyzed according to the following expression for survival:

$$S = 1 - (1 - e^{-kD^n}), \tag{II-2}$$

where k is the coefficient for killing, and n is an exponent often called the hit number or extrapolation number. The survival fraction can also be expressed by an $\alpha D + \beta D^2$ model of the form:

$$S = \exp(-\alpha D - \beta D^2), \tag{II-3}$$

the same as the last term in the complete quadratic with saturation model given above in Equation II-1.

Although the quadratic model can be derived entirely empirically from response data, there appears to be a rational biophysical basis. What such dose-effect curves imply is that some of the effects of radiation of any given class are induced by single ionizing events (i.e., the passage of a single photon or of a single particle), whereas others result from the interaction of two or more statistically independent ionizing events. The dose-squared term can be understood in another way: If P is the probability of hitting a target and causing a sublethal amount of damage, then the probability that target will be hit twice (or more), to give an effective hit, is $(P)(P)$, or P^2. The quadratic expression can yield curves ranging from linear (or nearly linear), in the case where β is so small in relation to α that the αD dominates, to essentially dose-squared, in the case where α is negligibly small in relation to β. Mutation induction in simple prokaryotic cells is an example of linear response, and the induction of two-break chromosomal aberrations of the exchange type in higher eukaryotic cells exposed to acute doses of low-LET radiation is an example of quadratic response. Many dose-response curves, however, are somewhere in between.

EFFECT OF DOSE RATE AND FRACTIONATION

The expressions presented above apply specifically to low-LET radiation, such as x or gamma rays, delivered at a high (acute) rate. It was noticed early, however, that when low-LET doses were protracted or appreciable periods passed between successive dose fractions, the effectiveness of the total dose was likely to diminish. In fact, as dose rate decreased, the radiobiological endpoints that at higher dose rates had response curves with an appreciable βD^2 term began to lose that term. Eventually, little was left of the βD^2 term, and the dose-response curve was essentially linear, represented simply by αD. With increasing interfraction time, much the same thing happened, with the βD^2 term for the total dose decreasing until, instead of what is often called complete interaction [i.e., $\beta(D_1 + D_2 + D_3 \ldots)^2$], one observed the sum of βD^2 terms for the individual fractions (i.e., $\beta {D_1}^2 + \beta {D_2}^2 + \beta {D_3}^2 \ldots$). This implied that somehow the subeffective partial lesions left at the end of the first dose fraction were becoming unavailable, or repaired, with time; thus, if enough time elapsed between fractions (a few hours), none were left from the first dose fraction to interact with those from the second. Loss of subeffective partial lesions also accounts for the simple dose-rate effect. Lea[35] understood this early and created a correction factor that he called the G factor to correct for dose-rate effects:

$$G = 2(\tau/T)^2(T/\tau - 1 + e^{-T/\tau}), \qquad \text{(II-4)}$$

where τ is the average time between breakage and restitution, and T is the duration of treatment.

EFFECT OF LET

As the LET of radiation (usually calculated as track-average LET) increases, two things happen. First, for radiobiological endpoints that have a substantial βD^2 term with acute doses of low-LET radiation, the curves begin to straighten, losing the D^2 component and tending toward linearity. Second, the effectiveness of the αD component also increases, indicating that this is not the same phenomenon as that observed for dose rate or fractionation with low-LET radiation. In sum, the higher-LET radiation seems to be capable, if it deposits energy in a target at all, of causing fully effective events. In other words, the production of subeffective lesions becomes less and less likely, while the production of fully effective lesions becomes more likely as LET increases. This makes sense, because as the ionization per unit track length increases, a target becomes more likely to suffer substantial damage, if it is affected at all. In fact, one might expect that if the LET of radiation becomes high enough, the effectiveness with

increasing LET would saturate at some point and then fall off as the LET continued to increase. That is what is observed, particularly in prokaryotic systems. For the induction of radiobiological effects in mammalian cells, the target volume appears to be large enough for the effect to occur at a higher LET, with the maximally effective LET being around 100 keV/μm.

MICROSCOPIC DOSE DISTRIBUTION

With energetic photon irradiation, the quantity dose, used in calculating a dose-effect model, has an easily understood meaning and is easy to measure empirically. However, the situation often is not so simple for particle irradiation from internally deposited radionuclides, particularly when the mean path length of the charged particle is small in relation to the diameter of the average cell or subcellular structure of interest and when the distribution of the radionuclide is very nonuniform. The problem is particularly acute when, as in the case of tritium incorporated into the DNA of a cell in the form of tritiated purines or pyrimidines, only parts of cells have incorporated the radionuclide and only some of the potential target cells have incorporated any radionuclide at all. In such cases, dose expressed in the usual terms of the average amount of energy deposited per unit of tissue mass over the mass of the organs (or parts of organs) becomes meaningless, if not actually misleading. Here it is important to consider the microscopic distribution of dose and the doses accumulated by individual targets (usually taken to be individual cell nuclei).

In the case of the alpha-emitting radionuclides of interest here, we must consider the problem presented by the inadequacy of simply measuring body or even organ burdens of a radionuclide, and try instead to identify the target cells at risk for a particular possible health impact and then to determine the average dose to the nuclei of all these cells. The distribution factor will otherwise be confounded with the relative biological effectiveness (RBE) factor, and that can lead to large uncertainties and ambiguities. In fact, if the RBE for a given particle seems to be reasonably well established, deviations from that RBE can be used to infer the magnitude that must be attributed to the microscopic dose-distribution factor.

MODELS OTHER THAN THE QUADRATIC

Models designed to describe and, thus, to permit prediction of the response of cell populations to radiation with different LETs can be divided broadly into two categories: those derived empirically from observed cellular dose-response curves (observation obviates assumptions about or dependence on underlying molecular or subcellular biological effects or mechanisms), and those built on assumptions about underlying subcellular

mechanisms (which might or might not be shown to be applicable). Although the two approaches must ultimately converge, there can be little confidence that this will occur soon.

The first category includes two models that are related to the original target theory:[35] the quadratic model and the model of Bond et al.[8] As noted above, the former is applicable to a wide variety of endpoints in many systems, particularly with low-level exposure to radiation (i.e., either small doses at any dose rate or larger doses at very low dose rates). It also adequately accommodates dose-rate and protraction effects and accurately describes most responses as a function of LET in tissue in terms of an increasing αD component and a decreasing βD^2 contribution as the LET increases.

Thus, with very low doses, the mean absorbed dose to the population of cells increases only because the fraction of cells hit increases; the mean dose (specific energy) delivered stochastically to the hit cells remains constant. However, as the fraction of cells hit approaches unity, the absorbed dose to the cell population can increase only if the number of hits per cell increases.[8] At high doses and high dose rates, where each cell has received many hits, the variance of the mean decreases; so the dose to each cell, the mean dose to the cell population, and the mean dose to the organ or other medium approach equality.

The other model that belongs in this category and that combines some elements of hit theory with those of microdosimetry is under development.[8] The unique addition is an empirically derived hit size effectiveness factor. This factor gives the probability that a cell with a certain amount of energy deposited within it will respond as a function of the energy deposited. Additional testing with different endpoints and biological systems is necessary before the degree of applicability of the model can be ascertained.

The second category includes several models that cover a wide variety of assumptions, some of which are commented on below. The early dual-action model[33] combines microdosimetry with assumptions about the stochastic interactions of microdosimetric events to derive the basic formulation:

$$E = K(\zeta D + D^2), \tag{II-5}$$

where ζ is a physical quantity equal to the average specific energy (dose to a subcellular target volume in single events), and K is the sensitivity coefficient. This expression is equivalent to the more general $\alpha D + \beta D^2$ formulation, with $K\zeta$ equivalent to α and K equivalent to β.

A recently developed thesis is the lethal and potentially lethal model,[24] which, with the earlier repair-nonrepair model of cell survival[46] and other models, is built on the added assumptions of irreparable lethal lesions,

repairable potentially lethal lesions, first-order kinetics for correct repair, and second-order kinetics for misrepair. Although the models have been applied only to cell lethality in plateau-phase (stationary-phase) cells in culture, they do take LET and dose rate into account. The principal mathematical formulation is complex and embraces a power expansion, but the first two terms also yield the $\alpha D + \beta D^2$ expression. Such models constitute valuable contributions in attempting to unify the most attractive features of a number of others; the extent of their universality remains to he determined.

Other models have also been developed, for example, the molecular theory of Chadwick and Leenhouts,[31] the kinetic model of Dienes,[25] the cybernetic model of Kappos and Pohlit,[32] the incomplete-repair model of Thames,[45] and the repair model of Braby and Roesch.[19] Many are limited, in that they deal either with only a small fraction of the many radiobiological factors that must be taken into account (e.g., some are limited to high dose rates and others to the question of repair, possible misrepair, and dose rate), or in encompassing only particular endpoints (e.g., some deal only with cell lethality, and others exclude it).

There is evidence that the various models can be reconciled, but it seems unlikely that there will be general agreement or that the mechanistic assumptions behind any of them will soon be proved.

None of the models in this category has the simplicity or the generality of application of the quadratic model, and none is at the point of applicability to the problems of predicting either genetic or carcinogenic responses in mammalian systems. Thus, although they contribute to our general understanding and appreciation of problems remaining to be solved, they cannot yet be applied in practical exercises that require the prediction of risk associated with exposure to principally low-level radiation. Therefore, it appears reasonable that the quadratic model be used.

APPLICATION TO RISK ESTIMATION FOR INCORPORATED ALPHA-EMITTING RADIONUCLIDES

With some notable exceptions—such as Thorotrast-exposed patients, radium-dial painters, or uranium miners—there is generally little direct information on the human health effects of the internally incorporated alpha-emitting radionuclides. However, we can derive several generalizations that can be applied to the problem of extrapolating available empirical evidence of the induction of human health effects by low-LET radiation.

First, even though the question of whether the low-LET effects best fit a simple linear dose-response model or a dose-squared model is unsettled, as far as the epidemiological data on radiation carcinogenesis are concerned (see Committee on the Biological Effects of Ionising Radiations [BEIR

III[38]), we can be reasonably sure that the dose-effect curves of alpha particles are linear (or essentially linear), at least for the low to moderate doses of interest, for which saturation effects can be ignored. Second, the evidence is that there are no dose-rate effects for high-LET alpha particles (with the one possible exception of in vitro cell transformation as discussed below). Third, we can be sure that the yields of effects per unit dose of alpha particles are greater than those per unit dose for low-LET x or gamma rays, that is, that the RBE for alpha particles is greater than unity. Althongh the appropriate RBE for a given alpha particle might not have been empirically measured in an appropriate target-cell system, generalizations on the basis of LET seem reasonable, provided that they are based on RBEs determined in cells likely to have similar cell nucleus and target volumes and based on doses that produce similar hit fractions per rad. Thus, in considering the hazard of induction of a malignancy of, say, the liver by deposition of plutonium, it seems reasonable to multiply the low-dose risk coefficients for low-LET radiation available in the BEIR III report[38] by an appropriate RBE factor to derive a new estimate that can be useful in the absence of empirical information on the overall risk associated with the radionuclide of interest.

We present below the empirical evidence from cellular radiobiology on RBE for alpha particles, with necessary background information, on which to base estimates of expected effects of exposure of human populations to alpha-emitting radionuclides.

MAMMALIAN CELL SURVIVAL

Although much work had been done earlier with prokaryotes and unicellular eukaryotes, mammalian cell survival studies became possible only with the development by Puck et al.[39] of a practical clonal assay for the survival of single mammalian cells in culture. The general principles already discussed in this appendix were elucidated mainly with low-LET x and gamma rays and have been reviewed elsewhere.[26,28] Much work has been done with high-LET irradiation, particularly with neutrons, protons, and beams of heavy ions, largely because of interest in their potential application to radiation oncology. Much less has been done on cell killing by alpha particles from the radionuclides of interest here. Therefore, it is necessary to consider the larger body of data from other high-LET radiation.

Extensive studies with fast neutrons of various energies have been done by Broerse et al.,[20] Berry,[6] and by the Radiological Research Laboratory group at Columbia University.[30,34] The last studies are of particular interest because of the essentially monoenergetic neutron beams that the group at Columbia University was able to use. Survival curves for Chinese

hamster cells irradiated with neutrons of 0.1–50 MeV have been obtained. As expected, neutron energies below a few million electron volts produced exponential survival curves without the shoulder characteristic of low-LET survival curves. At energies above a few million electron volts, the LET is lower and a small shoulder becomes evident.

RBE varied with neutron energy, exhibiting a rather broad peak at approximately 0.4 MeV. Because of the difference in survival-curve shapes, RBE was a function of the survival fraction at which the comparison was made with the reference curve, in this case for 250-kV x rays, peaking at about 9 for 80% survival and falling to approximately 3 at 0.001% survival.

Blakely et al.[7] recently reviewed extensive mammalian cell-survival studies with heavy ions, ^{12}C to ^{40}Ar, which used a variety of tissue-culture cell types. RBE was found to increase from about 1.0 at a track-average LET of 10 keV/μm to about 2.5 at a track-average LET of around 100 keV/μm. Survival curves lost their shoulders as LET increased, becoming simple exponentials for a higher LET. Thus, the results of both the fast-neutron and the heavy-ion experiments are in general agreement as to the modification of curve shape and increase in RBE with increasing LET. The maximal RBE observed in the neutron experiments, about 5 for 5% survival, is somewhat higher than the maximum observed in the heavy-ion experiments, probably because the heavy ions, with their high velocities, deposit a larger fraction of their energy via lower-LET secondary electrons and are thus less monochromatic with respect to LET.[43] The results of the heavy-ion experiments clearly demonstrate the falloff in efficiency (dose wastage) at average LETs greater than approximately 100 keV/μm.

Barendsen and coworkers[1–4] have studied mammalian cell-survival curves for cyclotron-accelerated alpha particles (and deuterons) and for ^{210}Po alpha particles. Polonium-210 alpha particles induced simple exponential survival curves with an RBE ranging from 2.5 (at high acute doses) to around 6.0 (at low doses). With cyclotron-accelerated alpha particles at LETs of 25–86 keV/μm, survival curves for the higher-LET alpha particles were again simple exponentials, although that for 25-keV/μm, alpha particles did show some evidence of a shoulder. The RBE, in comparison with the curve for 200-kV x rays, was dose dependent but was in the range of 3–5 for survival below about 20%, with the peak for any degree of survival at about 100 keV/μm.

Lloyd et al.[36] also determined mammalian cell-survival curves for accelerator-produced alpha particles. Essentially exponential survival curves were found for alpha particles with LETs of about 85 keV/μm; the slope agreed well with that determined by Barendsen and coworkers.[1–4]

Barnhart and Cox[5] and Thacker et al.[43] have determined survival curves for Chinese hamster cells in tissue culture exposed to alpha particles from ^{238}Pu. Again, the survival curves were exponential. With the

irradiation geometry used, the ranges of LET through the cells were 127–165 keV/μm and 100–135 keV/μm, respectively, in the two studies. RBE values of 7–10 were observed for high survival fractions.

Thus, there is general agreement that cell-survival curves for alpha particles, as well as other high-LET irradiation, are exponential with high RBE, peaking at around 100 keV/μm; are dose dependent; and reach maximal calculated values of about 5–10 at low doses. Low-dose-rate, low-LET ionizing radiation seems appropriate for comparison for the purposes of this report (i.e., comparison of the α terms of the quadratic relationship seems appropriate). Therefore, the lowest dose and, consequently, the highest RBE observed seem appropriate as a basis for extrapolating from the BEIR III report[38] and similar risk estimates to alpha-particle estimates when direct human data are not available.

MUTATION IN VITRO

Mutation induction at the hypoxanthine-guanine phosphoribosyl transferase (HGPRT) locus can be studied in vitro in Chinese hamster or human cells by using selection in a thioguanine-containing medium. Dose-effect curves have been determined in this way for both high- and low-LET radiations. Thacker et al.[43] and Cox and Masson[22] determined HGPRT mutation by heavy ions in Chinese hamster cells and human diploid fibroblasts, respectively. Helium, nitrogen, and boron ions with LETs of 28–470 keV/μm in the irradiated cells were used. Mutation-induction curves were essentially linear, giving maximal RBEs of about 6 at 90–200 keV/μm when calculated in terms of mutants per survivor and compared with the initial slope of the curve for gamma rays.

Both Barnhart and Cox[5] and Thacker et al.[43] have reported on the mutagenicity of alpha particles from ^{238}Pu in Chinese hamster cells. The study by Barnhart and Cox,[5] however, appears to have suffered from technical difficulties; as noted by Thacker et al., the latter found an RBE of about 2 for the alpha particles, which had an LET range of 127–165 keV/μm as they traversed the cells, compared with acute exposure to 250-kV x rays.

TRANSFORMATION IN VITRO

A few types of mammalian cells growing in tissue culture can be transformed in vitro from the growth patterns that characterize fairly normal cultured cells to a new phenotype that more closely resembles that of cancer cells. The basic change is from an orderly growth pattern exhibiting contact inhibition on the culture vessel surface to a pattern resulting from the loss of contact inhibition. That loss causes the cells

to pile up and overgrow each other and to produce foci of a distinct and recognizable morphology. Examples are shown in Figure II-1.

The development of cell-culture systems has made it possible to study the cellular and molecular mechanisms involved in radiation transformation under defined conditions devoid of host-mediated homeostatic modulating factors, and to assess the underlying mechanisms qualitatively and quantitatively. These systems afford the opportunity to study dose-related and time-dependent interactions of radiation with single cells and to identify factors and conditions that can prevent or increase cellular transformation by radiation. Because such cells transformed in vitro give rise to tumors when injected into hamsters, whereas untreated cells show no spontaneous transformation,[14,15] the system has obvious implications with respect to in vivo carcinogenesis. The details of the system and its utility in cellular radiation biology were recently reviewed by Borek.[11,12]

The role of DNA as a target in radiation transformation was suggested early by the requirement of DNA metabolism for fixation of the transformed state.[15] The ability of genomic high-molecular-weight DNA purified from cells transformed in vitro to transmit the transformed phenotype to normal cells constitutes an important criterion for the neoplastic state of the cells transformed after exposure to a carcinogen,[12,13,17,18,41] indicating that the transformed phenotype of the cells exposed to the carcinogen in vitro is encoded in the DNA. This criterion aids in mechanistic studies of transformation that attempt to analyze the specific transforming genes activated as a result of exposure to the carcinogen and to elucidate genetic changes.[17,18]

For both low- and high-LET radiation, transformants are produced as a function of increasing dose up to approximately 1–5 surviving cells/100 exposed cells, at which point the curves saturate. Transformants are produced more efficiently by fission neutrons than by 250-kVp x rays, with an RBE of approximately 10 in the region below saturation.[16,31] Figure II-2 shows an example for 430-keV neutrons.

Data on alpha-particle-induced in vitro transformation are sparse and often incomplete (there are no data on the tumorigenicity of the transformed cells). The effect of low-energy alpha particles in transforming the C3H/10T-1/2 cells was evaluated by Lloyd et al.[37] Transformation frequency per surviving cell increased as the cube of the dose, peaking at a fluence of 1.5×10^7–2.5×10^7 alpha particles/cm^2 (205–342 rad). Maximal transformation frequency reached 4%. No parallel experiments were carried out with x rays; thus, no RBE for alpha radiation was determined. By taking the dose required to reach the peak transformation region and comparing it with x-ray data determined for the same cell line by Terzaghi and Little,[42] an RBE of approximately 2 is obtained.

A study carried out by Robertson et al.[40] evaluated the effects of ^{238}Pu alpha particles in mouse BALB/3T3 cells. The A31-11 mouse BALB/3T3

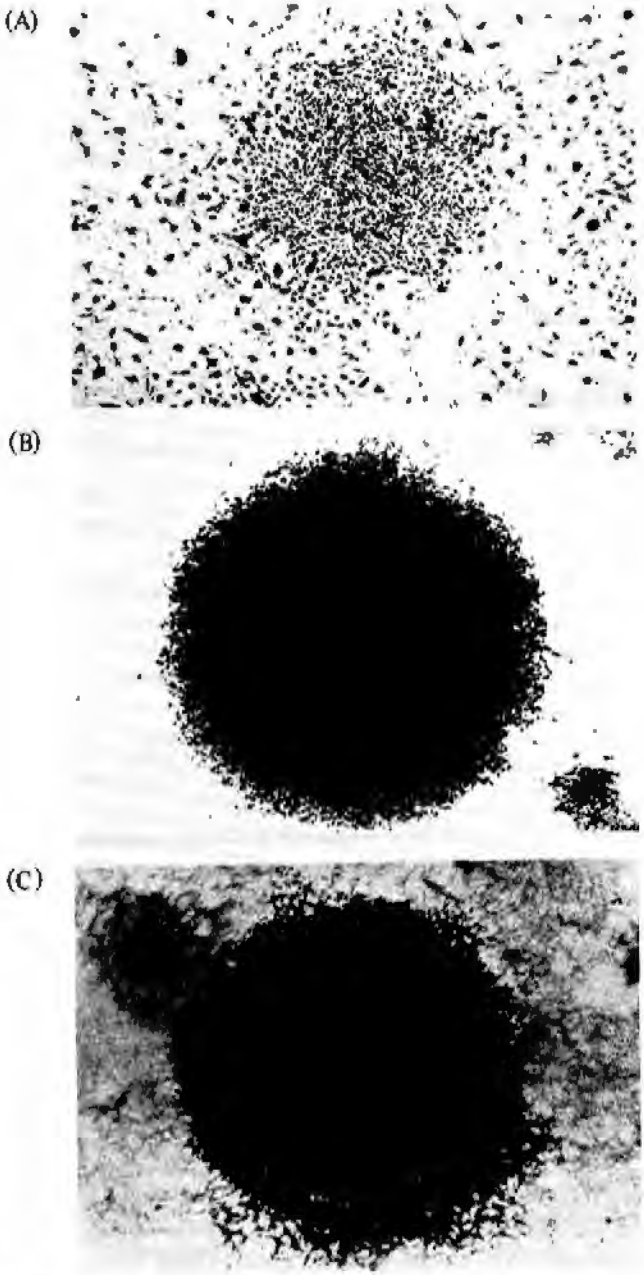


FIGURE II-1 (A) Normal colony of hamster embryo cells. (B) Colony of x-ray-transformed hamster embryo cells. (C) Focus of C3H/10T-1/2 cells transformed by x rays growing over normal cells. Morphology of transformed cells is the same as that after exposure to high-LET radiation. SOURCE: Borek.[12]

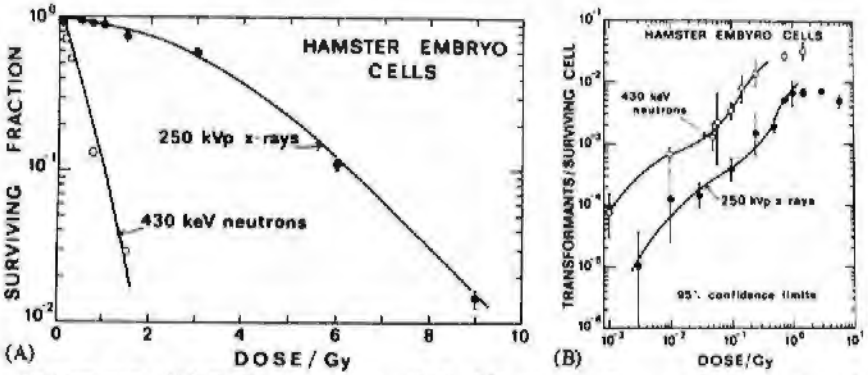


FIGURE II-2 (A) Pooled data on survival of hamster embryo cells irradiated with 250-kVp x rays (solid circles) or 430-keV monoenergetic neutrons (open circles). Error bars show estimated standard deviations. 1 Gy = 100 rad. (B) Pooled data for hamster embryo cells on number of transformants per surviving cell after irradiation with 250-kVp x rays (solid circles) or 430-keV monoenergetic neutrons (open circles) at the Radiological Research Accelerator Facility. Error bars show 95% confidence intervals for estimates. SOURCE: Borek et al.[16]

cell system was used, and in vitro transformation was induced by 5.3-MeV alpha particles from a specially constructed ^{238}Pu source. The biological effects were compared with those of 220-kVp x rays. The alpha-radiation survival curve gave an RBE of 3.5 at 50% survival. The transformation frequency increased exponentially with dose in the range examined (25–250 rad); the maximal RBE for the induction of transformation in growing cells was approximately 3.

However, the RBE for alpha transformation in nonproliferating cells appeared to be much higher; the yield of transformants among x-irradiated cells that were held in the stationary phase of growth for 6–220 h after irradiation declined by a factor of nearly 50, whereas no decrease occurred in alpha-irradiated cells. The findings suggest that carcinogenic damage induced by high-LET radiation in mammalian cells is very inefficiently repaired, compared with that induced by x rays, and that the intracellular carcinogenic effect of exposures to high-LET radiation can be cumulative. They also suggest that the effective RBE for alpha radiation in nonproliferating cell populations in vivo might be much higher than one would predict on the basis of measurements in dividing cells.

Work by Hall and Hei[29] with C3H/10T-1/2 cells compared the transforming action of radiation, from a source of ^{241}Am, delivered at 10 rad/min with that of gamma rays from a ^{137}Cs source with an absorbed dose rate of 137 rad/min at equivalent doses. Alpha particles were substantially more cytotoxic and more efficient than gamma rays in inducing oncogenic transformation. The calculated RBE ranged from 2.3 to 9 for the transformation frequencies examined.

REFERENCES

1. Barendsen, G. W. 1962. Dose-survival curves of human cells in tissue culture irradiated with alpha-, beta-, 20-KV.X- and 200-KV. X-radiation. Nature 193:1153–1155.
2. Barendsen, G. W., and T. L. J. Beusker. 1960. Effects of different ionising radiations on human cells in tissue culture. I. Irradiation techniques and dosimetry. Radiat. Res. 13:832–840.
3. Barendsen, G. W., T. L. J. Beusker, A. J. Vergroesen, and L. Burke. 1960. Effects of ionising radiations on human cells in tissue culture. II. Biological experiment. Radiat. Res. 13:841–849.
4. Barendsen, G. W., H. M. D. Walter, J. F. Fowler, and D. K. Bawley. 1963. Effects of different ionising radiations on human cells in tissue culture. III. Experiments with cyclotron-accelerated alpha-particles and deuterons. Radiat. Res. 18:106–119.
5. Barnhart, B. J., and S. H. Cox. 1979. Mutagenicity and cytotoxicity of 4.4-MeV α-particles emitted by plutonium-238. Radiat. Res. 80:542–548.
6. Berry, R. J. 1974. Modification of neutron effects upon cells by repair, and by physical and chemical means. Pp. 257–271 in Biological Effects of Neutron Irradiation. TI/PUB/352. Vienna: International Atomic Energy Agency.
7. Blakely, E. A., F. Q. H. Ng, S. B. Curtis, and C. A. Tobias. 1984. Heavy-ion radiobiology: Cellular studies. Adv. Radiat. Biol. 11:295–389.
8. Bond, V. P., M. N. Varma, C. A. Sondhaus, and L. E. Feinendegen. 1985. An alternative to absorbed dose, quality, and RBE at low doses. Radiat. Res. 104:S52–S57.
9. Boos, J. 1978. Mapping of fast neutron radiation quality. Proceedings of the Third Symposium on Neutron Dosimetry in Biology and Medicine, G. Burger and H. G. Ebert, eds. EUR 5845 DE/EN/FR:499-514. Brussels: Euratom.
10. Borek, C. 1980. X-ray induced in vitro neoplastic transformation of human diploid cells. Nature 282:776–778.
11. Borek, C. 1982. Radiation oncogenesis in cell culture. Adv. Cancer Res. 37:159–232.
12. Borek, C. 1985. The induction and control of radiogenic transformation in vitro: Cellular and molecular mechanisms. J. Pharmacol. Ther. 27:99–142.
13. Borek, C. 1985. Cellular and molecular mechanisms in malignant transformation of diploid rodent and human cells by radiation. Pp. 365–378 in Mechanisms in Carcinogenesis, Vol. 9, J. Barrett and R. W. Tennant, eds. New York: Raven.
14. Borek, C., and L. Sachs. 1966. In vitro cell transformation by x-irradiation. Nature 210:276–278.
15. Borek, C., and L. Sachs. 1967. Cell susceptibility to transformation by x-irradiation and fixation of the transformed state. Proc. Natl. Acad. Sci. USA 57:1522–1527.
16. Borek, C., E. J. Hall, and H. H. Rossi. 1980. Malignant transformation in cultured hamster embryo cells produced by x rays, 430-KeV monoenergetic neutrons, and heavy ions. Cancer Res. 38:2997–3005.
17. Borek, C., A. Ong, J. Bresser, and D. Gillespie. 1984. Transforming activity of DNA of radiation transformed mouse cells and identification of activated oncogenes in the donor cells. Proc. Am. Assoc. Cancer Res. 25:100.
18. Borek, C., A. Ong, and H. Mason. In press. Distinctive transforming genes in x-ray transformed mammalian cells. Proc. Natl. Acad. Sci. USA.
19. Braby, L. A., and W. C. Roesch. 1978. Testing of dose-rate models with *Chlamydomonas reinhardi*. Radiat. Res. 76:259–270.

20. Broerse, J. J., G. W. Barendsen, and G. R. van Kersen. 1967. Survival of cultured human cells after irradiation with fast neutrons of different energies in hypoxic and oxygenated conditions. Int. J. Radiat. Biol. 13:559–572.
21. Chadwick, K. H., and H. P. Leenhouts. 1973. The molecular target theory. Int. J. Radiat. Biol. 23:185.
22. Cox, R., and W. K. Masson. 1979. Mutation and inactivation of cultured mammalian cells exposed to beams of accelerated heavy ions. III. Human diploid fibroblasts. Int. J. Radiat. Biol. 36:149–160.
23. Cox, R., J. Thacker, D. T. Goodhead, and R. J. Munson. 1977. Mutation and inactivation of mammalian cells by various ionising radiations. Nature 267:425–427.
24. Curtis, S. B. 1986. Lethal and potentially lethal lesions induced by radiation. A unified repair model. Radiat. Res. 106:252–270.
25. Dienes, G. 1966. A kinetic model of biological radiation response. Radiat. Res. 28:183–202.
26. Elkind, M. M., and G. F. Whitmore. 1967. The radiobiology of cultured mammalian cells. New York: Gordon and Breach.
27. Goodhead, D. T., R. J. Munson, J. Thacker, and R. Cox. 1980. Mutation and inactivation of cultured mammalian cells exposed to beams of accelerated heavy ions. IV. Biophysical interpretation. Int. J. Radiat. Biol. 37:135–167.
28. Hall, E. J. 1978. Radiobiology for the Radiologists, 2nd ed. New York: Harper and Row.
29. Hall, E. J. and T. K. Hei. 1985. Oncogenic transformation *in vitro* by radiations of varying LET. Radiat. Prot. Dosimetry 13:149–151.
30. Hall, E. J., J. K. Novak, A. M. Kellerer, H. H. Rossi, S. Marino, and L. J. Goodman. 1975. RBE as a function of neutron energy. I. Experimental observations. Radiat. Res. 64:245–255.
31. Han, A., and M. M. Elkind. 1979. Transformation of mouse C3H/10T-1/2 cells by single and fractionated doses of x-rays and fission-spectrum neutrons. Cancer Res. 39:123–130.
32. A. Kappos and W. Pohlit. 1972. A cybernetic model for radiation reactions in living cells. I. Sparsely ionizing radiations; stationary cells. Int. J. Radiat. Biol. 22:51–65.
33. Kellerer, A. M., and H. H. Rossi. 1972. The theory of dual radiation action. Curr. Top. Radiat. Res. Q. 8:85–158.
34. Kellerer, A. M., E. J. Hall, H. Rossi, and P. Teedle. 1976. RBE as a function of neutron energy. II. Statistical analysis. Radiat. Res. 65:172–186.
35. Lea, D. E. 1946. Actions of Radiations on Living Cells. Cambridge: Cambridge University Press.
36. Lloyd, E. L., M. A. Gemmell, C. B. Henning, D. S. Gemmell, and B. J. Zabransky. 1979. Cell survival following multiple-track alpha particle irradiation. Int. J. Radiat. Biol. 35:23–31.
37. Lloyd, E. L., M. A. Gemmell, C. B. Henning, D. S. Gemmell, and B. J. Zabransky. 1979. Transformation of mammalian cells by alpha particles. Int. J. Radiat. Biol. 36:467–478.
38. National Research Council, Committee on the Biological Effects of Ionizing Radiations (BEIR). 1980. The Effects on Populations of Exposures to Low Levels of Ionizing Radiation. Washington, D.C.: National Academy Press. 524 pp.

39. Puck, T. T., D. Morkorn, P. I. Marcus, and S. J. Cieciura. 1957. Action of x-rays on mammalian cells. II. Survival curves of cells from normal human tissues. J. Exp. Med. 106:484–500.
40. Robertson, J. B., A. Keehler, J. George, and J. B. Little. 1983. Oncogenic transformation of mouse BALB/3T3 cells by plutonium-238 alpha particles. Radiat. Res. 96:261–274.
41. Shilo, B. Z., and R. A. Weinberg. 1981. Unique transforming gene in carcinogen-transformed mouse cells. Nature 289:607–609.
42. Terzaghi, M., and J. B. Little. 1976. X-radiation-induced transformation in a C3H mouse embryo-derived cell line. Cancer Res. 36:1367–1374.
43. Thacker, J., A. Stretch, and M. A. Stephens. 1979. Mutation and inactivation of cultured mammalian cells exposed to beams of accelerated heavy ions. II. Chinese hamster V79 cells. Int. J. Radiat. Biol. 36:137–148.
44. Thacker, J., A. Stretch, and D. T. Goodhead. 1982. The mutagenicity of particles from plutonium-238. Radiat. Res. 92:343–352.
45. Thames, H. D. 1985. An incomplete repair model for fractionated and continuous irradiations. Int. J. Radiat. Biol. 47:319–339.
46. Tobias, C. A. 1985. The repair-misrepair model in radiobiology: Comparison to other models. Radiat. Res. 104:S77–S95.

APPENDIX III

The Effects of Radon Progeny on Laboratory Animals

Animal studies have been conducted for over 50 yr to examine the respiratory effects of pollutants in the air of mines. This work, emphasizing respiratory cancer, has provided important data on exposure-response relationships and the interactions among the harmful agents to which miners are exposed. Many of the initial studies were concerned with early effects or short-term pathological changes.[21,22,29] In many of the studies, exposures were based primarily on radon-gas concentrations, with little or no consideration of radon-daughter concentrations, which have been shown to contribute the greatest radiation dose to the lung. Two American research centers—the University of Rochester and the Pacific Northwest Laboratory (PNL)—and the Compagnie Generale des Matieres Nucleaires (COGEMA) laboratory in France have contributed most of the experimental data on radon-daughter inhalation by laboratory animals.

INHALATION STUDIES AT THE UNIVERSITY OF ROCHESTER

Beginning in the 1950s, investigators examined the biological and physical behaviors of radon daughters and the dosimetry of radon daughters in the respiratory tract.[1,20,25] Shapiro[31] exposed rats and dogs to radon alone at several concentrations and to radon with radon daughters attached to room-dust aerosols. The degree of attachment of radon daughters to carrier dust particles was shown to be an important determinant of the alpha-radiation dose to the airway epithelium and that more than 95% of the dose to the airway epithelium was dne to the short-lived radon daughters radium A (^{218}Po) and radium C′ (^{214}Po), rather than to the parent radon. In 1953, Cohn et al.[9] reported the relative levels of

radioactivity found in the nasal passages, the trachea and major bronchi, and the other portions of rat lungs after exposure to radon or radon daughters. The respiratory tracts of animals that inhaled radon with its daughters contained 125 times more activity than those of animals that inhaled radon alone. Beginning in the mid-1950s, Morken,[25-27] and Morken and Scott[28] initiated a series of experiments to evaluate the biological effects of inhaled radon and radon daughters in mice; later experiments also used rats and beagles. The negative results of these studies suggested that alpha irradiation was inefficient in producing tumors in the respiratory system.

These experiments were noteworthy in describing exposure-dose relationships in the whole lung, in regions of the lung, and in other organs. The paucity of pathological effects did not permit examination of exposure-response relationships for carcinogenesis, as demonstrated later by experiments at COGEMA and PNL. In the early experiments, the only apparent late, permanent changes occurred in the alveolar and possibly the bronchiolar regions of the lung. They were observed for a wide range of doses and developed after 3 yr in the dog and 1 and 2 yr in the rat and mouse, respectively. Some of these changes might have been preneoplastic, but the high-level exposures (associated with life-span shortening) and the early termination of experiments precluded further development to neoplasia. The influence of the radon-daughter carrier aerosol (laboratory air) on the results of these experiments is uncertain, but it might have led to more rapid solubilization of the daughters into blood and a resulting decrease in irritation or fibrosis, in comparison with ore-dust and silica aerosols.

INHALATION STUDIES AT COGEMA

The studies by Chameaud and colleagues[2-8] were begun in the late 1960s and early 1970s to determine whether radon and its daughters induced tumors in rats and to provide experimental data to support the epidemiological data on radon-daughter carcinogenesis. Before 1972, rats were exposed to ambient air that was enriched with radon after passage through trays of finely ground ore containing 25% uranium. Resulting radon concentrations were 0.75 μCi/liter; radon-daughter equilibrium factors were about 30%. With filters and electrostatic purifiers, the equilibrium factor was reduced to about 1%. Radon-daughter concentrations were calculated to be around 2,300 and 75 working levels (WL), respectively, for the two radon-daughter equilibrium conditions.

After 1972, animals were exposed to radon derived from underground barrels of radium-rich lead sulfate. Radon was pumped by a closed circuit into a 1-m^3 equilibration container and then to two 10-m^3 metal inhalation chambers. Up to 600 rats could be exposed for as long as 16 h when oxygen

was added to the inhalation chambers. The maximum radon concentration was 1.25 μCi/liter, generally at 100% equilibrium with radon daughters. By calculation, the maximum radon-daughter concentration was 12,500 WL. Because of radon-daughter deposition on the cages and the hairs of rats, the disequilibrium of the radon daughters increased as the number of animals in the inhalation chambers increased. Exposure periods ranged from about 1 to 10 months; exposure rates ranged from less than 10 to hundreds of working-level months (WLM)/wk, the majority averaging approximately 200–400 WLM/wk.*

In two major experiments,[2] rats were exposed by inhalation to stable cerium hydroxide or to uranium-ore dust concentrations with and without radon daughters, at 130 mg/m^3, to determine whether the presence of dust altered the carcinogenic effect of radon daughters. Exposure to stable cerium hydroxide before exposure to radon daughters shortened the induction latent period by 2–3 months. Uranium-ore dust (given on days alternating with days of radon-daughter exposure) appeared to have little influence on the tumorigenic process, although too few animals were used to permit a firm conclusion.[5] Radon-daughter exposures varied from 500 to 8,500 WLM. The effect of the radon daughters did not change with the various equilibrium ratios. These experiments confirmed that radon daughters alone induced tumors in rats.

Other changes were observed in these experiments. These are given below.

- After large radon-daughter exposures, large areas of diffuse interstitial pneumonia with hyaline membrane formation and with severe fibrosis of interalveolar septa surrounding capillaries were noted. Death generally occurred within a few weeks to a few months if exposure exceeded 6,000 WLM. No lung cancers were produced.
- Animals lived longer after smaller radon-daughter exposures, with lung carcinomas appearing 12–24 months after the beginning of exposure. The time to appearance of tumors increased with decreasing cumulative radon-daughter exposure. Exposures of 2,000–5,000 WLM, delivered over 300–500 h (during 3–4 months), produced the highest incidence of tumors.
- Bronchiolar metaplasia occurred at the bronchioloalveolar junction and in neighboring alveoli. It consisted of large columnar cells with basal

*Some of the exposure values in these French studies have been supplied by COGEMA investigators and might be different from previously published values. (J. Chameaud, personal communication to F. T. Cross, 1986.)

nuclei and light-colored protoplasm that were often ciliated. Alveolar metaplasia of cuboidal cells, with darker protoplasm, appeared in peripheral regions of the lungs.

- Adenomatous lesions of varied size and cell layers covered areas of the alveolar septa. Adenomas consisting of round tumors with cells often clustered together occurred. Some adenomas showed malignant characteristics.
- Malignant tumors of several different types occurred, often in the same animal. These included epidermoid carcinomas, not always clearly differentiated, often keratinized or necrosed, and occasionally extending into the mediastinum; bronchiolar adenocarcinomas, sometimes mucus-producing, containing numerous cellular anomalies, and characterized by a high number of mitoses and invasion of other lung lobes, but seldom metastatic; and bronchioloalveolar adenocarcinomas with few mitoses, but later invading the mediastinum, diaphragm, and thoracic wall.
- The relationship of exposure to tumor incidence, uncorrected for life-span shortening, was not linear over a wide range of exposures; the incidence per unit exposure increased with decreasing high cumulative exposure.

Later experiments, which confirmed these pathological findings, extended the radon-daughter exposures to approximately 20–50 WLM.[5,7,8] Tumor-incidence and survival-time data and lifetime lung-tumor risk coefficients are shown in Table III-1. Although the risk data are uncorrected for life-span shortening, hazard-function analysis demonstrated that when the data are adjusted for competing causes of death, the excess risk of developing pulmonary tumors is approximately linearly related to exposure throughout the range of exposures studied.[19] Further findings are given below.

- The tumor latent period, defined as the interval between the start of radon-daughter exposure and death or killing, of the animal increased with decreasing cumulative WLM. Mean latent periods of tumor-bearing animals were around 750 days for exposures of less than 300 WLM and 650 days for exposures of over 1,000 WLM.
- Lung cancers in rats invaded pulmonary lymph nodes, but metastases to other tissues were rare. Tumor size increased with increasing cumulative WLM.
- No radiation-induced small-cell carcinomas were observed in rats; however, other histological types of lung carcinomas were similar to those observed in humans.

TABLE III-1 Summary of Tumors Primary to Lungs of Rats, Median Survival Times,[a] and Lung-Tumor Risk Coefficients for COGEMA Radon-Daughter Exposures

Group Mean Exposure (WLM)	Nominal Exposure Rate (WLM/wk)	No. of Animals Examined	No. of Animals with Tumors	% Animals with Tumors	Group Median Survival Time (days)	Mean Lifetime Risk Coefficient (10^{-4}/WLM)[b]
20–25	2–4	~1,500	25	1.7	684	7.5[c]
50	2–8	~1,000	30	2.9	687	5.8[c]
290	9	21	2	10	610	3.3
860	370	20	4	20	672	2.8[d]
1,470	370	20	5	25	606	1.7
1,800	200	50	17	34	600	1.9
1,900	310	20	7	35	548	1.8
2,100	220	54	23	43	593	2.0
2,800	310	180	74	41	560	1.5
3,000	370	40	17	43	670	1.4
4,500	370	40	29	73	644	1.6

[a]Data from Chameaud et al.[7-10]

[b]Values are uncorrected for life-span differences from control animals. Lifetime risk coefficients based on raw incidence at very low exposures are considered to define accurately the initial slope of the risk-coefficient curve.

[c]Value is corrected for lung-tumor incidence in control rats of the low-exposure group (0.83%); normal incidence in the absence of appreciable background radon exposure is about 0.1–0.2%. Median survival time of control rats of the two lowest exposure groups was 752 d.

[d]Calculated value at this exposure level is 2.3.

- Cutaneous epitheliomas of the upper lip and cancers of the urinary system were the only two sites other than the lungs where cancers were noted in exposed rats.
- The incidence of lung cancer increased with decreasing high radon-daughter exposure rate. The greatest effect was noted in exposure-fractionation experiments. Rats exposed to radon daughters for approximately 3,000 WLM, at 1,500 WL for 7 h/day or 1 or 5 days/wk (average exposure rates are calculated to be above 50 and 300 WLM/wk) had a nearly fourfold increase in cancer incidence with exposure protraction.
- While the latency period decreased, the lung-cancer incidence did not change with increasing age at first exposure. For 3,000-WLM exposures, the latent periods for ages at first exposure of 150, 280, 400, and 520 days were 640, 510, 450, and 305 days, respectively.
- Synergism was observed between exposure to radon progeny and whole-body cigarette-smoke exposures if the exposure to smoke followed the exposure to radon daughters. However, if the cumulative cigarette-smoke exposure preceded the radon-daughter exposure, no increase in cancer incidence was noted over that produced by radon daughters alone. Thus, the effect of cigarette smoke depended on the sequence of exposures and was attributed to its promoting action.[5] The histological types of cancers observed were not altered by cigarette-smoke exposures. The investigators have not reported whether the latent period for cancer was influenced by smoke exposure; the observation that tumors in the radon-daughter- and smoke-exposed animals were larger and more invasive than those in animals exposed only to radon daughters might be indicative of a shorter latent period for smoking-related tumors.

The COGEMA studies have produced more than 800 lung cancers in about 10,000 rats exposed to radon daughters with ambient aerosols and in mixtures with other pollutants. The exposure-response relationship data shown in Table III-1 therefore constitute only a portion of the data from these experiments. The derived range in mean lifetime risk coefficients, uncorrected for life-span differences from control animals, is about 1.5×10^{-4}–7.5×10^{-4}/WLM for exposures between about 20 and 5,000 WLM. The risk decreases at larger exposures because of life-span shortening. No evidence of a threshold below 20 WLM was apparent.[8]

INHALATION STUDIES AT THE PACIFIC NORTHWEST LABORATORY

Exposures of dogs and rodents to uranium-mine air contaminants were begun in the late 1960s and early 1970s to identify agents and the magnitude of exposures to them that were responsible for producing lesions of the respiratory tract similar to those observed in uranium miners. The early experiments concentrated on lifetime inhalation exposures of hamsters and beagles to mixed aerosols of radon, radon daughters, carnotite uranium-ore dust, diesel-engine exhaust, and cigarette smoke. Most of the final data from these early experiments have been published.[11–13] To provide data that were missing from the earlier dog study, follow-up studies have included exposures of beagles to uranium-ore dust alone (but not to radon daughters alone) and exposures of rats to mixtures of radon, radon daughters, and uranium-ore dust.[10,14–18,33] Because the studies in rats were designed to develop exposure-response relationships, the exposures were truncated rather than extended through the animals' lifetimes. They were also designed to study the roles of carnotite uranium-ore dust concentration and radon-daughter exposure rate, unattachment fraction, and disequilibrium in the production of lung lesions. Histopathological examination, clinical pathological examination, and pulmonary physiology tests were the primary means of measuring response. Urinalyses have recently supplemented serum tests as more sensitive evaluations for kidney damage. Radiometric analyses of tissues have been used to determine mean radon-daughter tissue doses and the body distribution of long-lived radioactivity from the ore dust.

Lifetime exposures of hamsters to radon daughters alone or in combination with uranium-ore dust and diesel-engine exhaust caused no significant ($P > 0.05$) changes in mortality patterns compared with those of controls. The mean radon-daughter exposure in the hamster experiments was about 10,000 WLM. Lifetime exposures of beagles to mixtures of radon daughters, uranium-ore dust, and cigarette smoke caused significant life-span shortening compared with that of controls. Mean survival times of the dogs exposed to mixtures of radon daughters and ore dust, with or without cigarette smoke, were 4–5 yr. Mean survival times of controls and dogs exposed to smoke only were equivalent during the same period. The mean radon-daughter exposure of the dogs was about 13,000 WLM.

Studies in progress show that chronic exposure of rats to mixtures of radon daughters and uranium-ore dust shortens the life span. The data thus far generally show no significant differences in mortality patterns compared with those of controls for exposures up to about 2,500 WLM. Exposures exceeding 5,000 WLM have caused significant life-span shortening, with

the effect increasing with exposure. In general, rats that showed life-span shortening also showed weight loss.

Thus far, two life-span-shortening anomalies have been noted in the rat experiments. First, in an interim study to determine any influence of radon-daughter exposure rate, rats exposed to about 640 WLM at the lowest rate (about 44 WLM/wk) died earlier than other animals given comparable cumulative exposures. Second, in a study to determine the influence of unattached radon daughters versus that of attached radon daughters, rats exposed to about 5,100 WLM with the highest unattachment fraction ($f_a = 24\%$) died earlier than other animals given comparable cumulative exposures. Life-table analyses of the survival-time data in the unattachment-fraction study[18] showed that the estimated probabilities that a rat would die with a lung tumor before 600 days were 0.42, 0.65, and 0.75 for 6, 10, and 24% ^{218}Po (radium A) unattachment, respectively. Expressed as percentages of radon concentration, rather than radium A concentration, the unattachment was 1.3, 5.2, and 9.5%. Later experiments at 640 and 53 WLM/wk showed no appreciable life-span shortening.

The mean survival time of tumor-bearing rats (as in the COGEMA data) was always significantly longer than that of non-tumor-bearing rats. The latent period of lung tumors is a large fraction of the rat life span, and tumors must grow to a size sufficient for detection; the shorter-lived animals might have died too soon for tumors, if any, to be detected.

In the life-span studies with dogs, animals with tumors of the respiratory tract generally had cumulative radon-daughter exposures exceeding 13,000 WLM; the exposure rate was 71 WLM/wk. Concomitant exposure to cigarette smoke had a mitigating effect on radon-daughter-induced tumors, possibly because smoking caused thickening of the mucus layer and stimulated mucociliary clearance. The overall incidence of lung primary tumors was 21% for a mean exposure of 13,100 WLM to radon daughters, 37% in the group exposed to radon daughters and uranium-ore dust, but only 5% in the comparable group that was also exposed to cigarette smoke. The overall incidence of nasal carcinoma was 8%. The lung cancers were about 70% bronchogenic carcinomas and 30% bronchioloalveolar carcinomas.[15] The simplified convention used was that squamous cell carcinomas and mucus-staining adenocarcinomas were bronchogenic carcinomas and that tumors of Clara cell or type II alveolar cell origin and non-mucus-staining adenocarcinomas were bronchioloalveolar carcinomas.

Lifetime inhalation exposures of hamsters produced severe radiation pneumonitis but only four squamous cell carcinomas (three in the radon daughters-only group and one in the group exposed to radon daughters and uranium-ore dust) in 306 radon-daughter-exposed animals (1.3%

incidence). Squamous cell carcinoma occurred only in association with squamous metaplasia of the alveolar epithelium, which was found only in hamsters exposed to radon daughters. Thus, it appears that after exposure to radon daughters, the development of squamous metaplasia and the development of carcinoma were related. Because so few lung cancers were produced in these high-exposure experiments, it was concluded that the hamster was an inappropriate surrogate for further study of the carcinogenic potential of inhaled (as opposed to instilled) mine-air pollutants.

Over 4,000 male rats have received chronic exposures to ambient air or to mixtures of radon daughters and uranium-ore dust since 1978. Data are still accumulating, but some general trends can be observed. Lung-cancer risk tended to increase (sometimes significantly) with decreasing radon-daughter exposure rate, increasing unattached fraction of radon daughters, and increasing radon-daughter disequilibrium. The lung cancers induced after exposures of approximately 300–5,000 WLM were about 70% bronchogenic carcinomas and 30% bronchioloalveolar carcinomas. The tumors were most often estimated (by sizing associated bronchi and bronchioles) to be about 50% proximal (bronchus-associated) and 50% distal (bronchiole- and alveolus-associated), in contrast with the greater proportion of proximal lung cancers in humans.[30] The prevalence of squamous metaplasia, and generally carcinoma, of the respiratory tract increased with an increasing unattached fraction of radon daughters.

The PNL data are inadequate for firm conclusions regarding the effect of radon-daughter exposure rate and the magnitude of the lifetime risk coefficient below 100 WLM. However, the data to date indicate an increasing lifetime lung-tumor risk coefficient with decreasing cumulative radon-daughter exposure. Like the COGEMA data, the PNL risk-coefficient data have not been corrected for life-span shortening due to competing causes of death, such as radiation pneumonitis (see Table III-1). It cannot be concluded that the increase in the risk coefficient continues with further decreases in cumulative exposure and exposure rate. The PNL experiments include exposures as low as 20 WLM. The tumor-incidence data, particularly those derived from high-exposure-rate experiments, are similar not only to those from COGEMA but also to present estimated lung-tumor incidence data in humans.

Animal exposure studies show that the tumorigenic efficiency of radon daughters varies with cumulative exposure, exposure rate, unattached fraction, disequilibrium, and concomitant exposures to other pollutants (i.e., cigarette smoke). The COGEMA and PNL data indicate that tumor incidence increases with an increase in radon-daughter cumulative exposure and a decrease in radon-daughter exposure rate. Chameaud et al.[5]

concluded that lung-cancer incidence at comparable cumulative exposures increased as the radon-daughter concentration decreased from 12,000 to less than 3,000 WL. In a related dose-fractionation study with a cumulative exposure of 3,000 WLM and a radon-daughter concentration of 1,500 WL, an approximately fourfold increase in lung cancers was observed when the exposure rate decreased from about 300 to 50 WLM/wk; it is not known whether this exposure-rate dependence persists at the far lower rates. A trend toward increasing a lung-tumor risk with decreasing exposure rate was noted in the earlier PNL rat experiments[14,18] when the rates changed from 180 to 88 and to 44 WLM/wk. Inasmuch as the increase was not significant and results were uncertain at 44 WLM/wk as a result of life-span shortening in that group, the exposure-rate dependence in rats might be lessened at the lower weekly rates of exposure. However, more recent data confirm the increase in lung-tumor risk with decreased exposure rate down to 53 WLM/wk.

Data from the PNL rat experiments also indicate an increase in the risk of lung tumors with increases in radon-daughter unattached fraction and disequilibrium.[18] The risk increase from 1.6 to 10% unattached radium A is significant ($P < 0.05$), but the positive trend reverses at 24% unattachment as a result of life-span shortening in that exposure group. In contrast with the results of the COGEMA experiments, the increase is also significant with radon-daughter disequilibrium (an equilibrium of 10 versus 40%) when the total numbers of lung cancers are compared. However, the trend is of borderline significance ($P = 0.10$) when the total numbers of rats with lung tumors are compared. The data on nasal carcinoma show an increasing trend with increasing unattachment and, as with the neoplastic lesions of the lung, a reverse trend at 24% unattachment. There is no indication that high-disequilibrium radon-daughter exposures, without concomitant high unattachments, produce more nasal carcinomas than do low-disequilibrium exposures.

The role of concomitant exposures to other pollutants depends not only on the nature of those pollutants but also on the sequence of exposures. Simultaneous or same-day exposure to radon daughters and uranium-ore dust, diesel-engine exhaust, or cigarette smoke increased the incidence of preneoplastic lesions but, except for cigarette smoke, did not change the incidence of lung tumors in the PNL experiments. In the COGEMA rat experiments, cigarette smoke was cocarcinogenic with radon daughters if exposure to smoke followed completion of exposure to the radon daughters,[4] but not if smoking preceded the radon-daughter exposures. In the PNL dog experiments, lung-tumor incidence decreased when animals were exposed to radon daughters and cigarette smoke alternately on the same day.

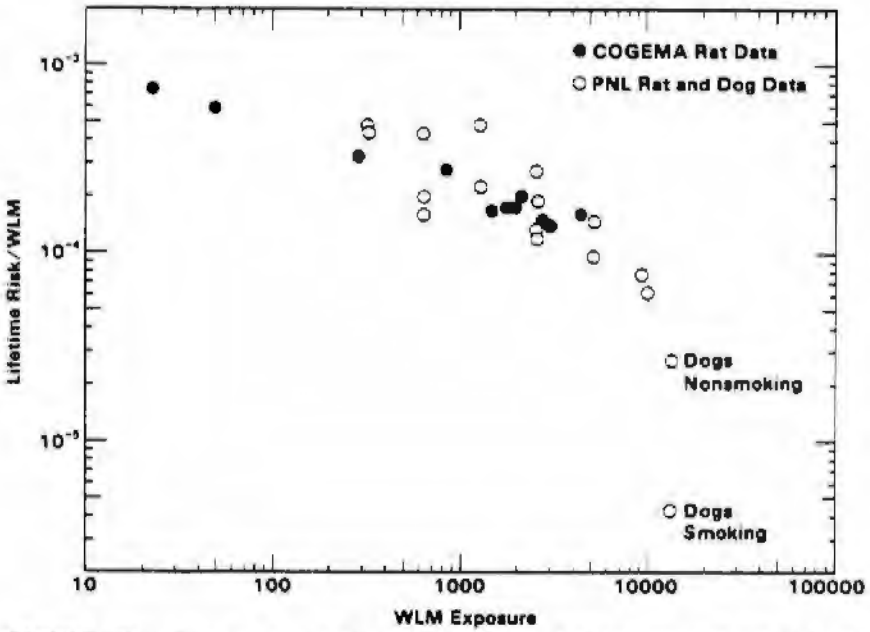


FIGURE III-1 Lifetime risk coefficients for radon-daughter exposure for PNL rat and dog data and COGEMA rat data; error bars are omitted. SOURCE: Personal communication, Dr. F. Cross, Pacific Northwest Laboratories.

LUNG CANCER

In Figure III-1 the mean lifetime lung-tumor risk per WLM (uncorrected for life-span differences from control animals) is plotted against the radon-daughter exposure (WLM) for PNL rats and dogs and COGEMA rats. The higher tumor efficiencies in the PNL studies (in contrast with the COGEMA studies) are probably due to the lower average exposure rates of the PNL experiments.

The uncertainties in the PNL lung-cancer incidence and risk-coefficient data are considered to be due mainly to uncertainties in the exposure data (standard deviations were generally well within ±20% of the means). Whenever PNL exposures were repeated, reproducibility of tumor-incidence data was generally within ±20% of the mean tumor incidence, which included the statistical uncertainties in the exposure data. Because the normal lung-tumor incidence in the absence of appreciable background radon exposures is very low (<0.2%) in the COGEMA and PNL rats, the risk-coefficient data, except for the 20- to 50-WLM COGEMA group of rats, have not been corrected for the incidence in control animals.

Current experiments at PNL, which involve mixtures of radon daughters and uranium-ore dust, will further define the shape of the risk-coefficient curve for very low exposures and exposure rates. For the present, COGEMA data on low exposures and low exposure rates indicate a leveling-off of the risk to a value of 6×10^{-4}–8×10^{-4}/WLM.

Kushneva[23] reported that rats given 50 mg of silica by instillation with inhalation exposures to radon at 8 μCi/liter developed many more pulmonary effects, including both adenomas and carcinomas, than did animals exposed to silica alone; the number of tumors and control animals was small. When silica dusts were included in the exposures, the radon-daughter inhalation studies at COGEMA and PNL showed no increased tumorigenic efficiency over exposures to radon daughters alone if these exposures exceeded a few hundred WLM. However, in contrast with the rat data of Kushneva[23] and the dog data from PNL, Chameaud et al.[5] have not found the silicotic process to be accelerated by the presence of radon daughters.

Little et al.[24,32] have shown in hamsters that when benzo(a)pyrene or saline instillations followed low-dose ^{210}Po instillations, the carcinogenic action of polonium was increased. Because radioactivity appears to be the initiator of the lung cancer, as in all the animal experiments with radon described here, any later exposure to an irritant that stimulates cell proliferation appears to increase the incidence of cancer.

SUMMARY AND CONCLUSIONS

Laboratory animal research programs on the effects of radon-daughter inhalation are being carried out in laboratories in both the United States and France. While much of the early work explored acute effects, more recent experiments involving chronic exposure have resulted in the induction of lung cancer in both rats and dogs. It should be noted, however, that the location and histopathology of such cancers are not analogous to humans, and caution is warranted in extrapolating from experiments with laboratory animals to humans. Nevertheless, substantial information has accumulated that provides insights into radon-daughter carcinogenesis. Table III-2 summarizes recent findings in animal studies of lung-cancer induction by radon decay products.

In rats, lung tumors have been induced at relatively low exposures (20 WLM).[7] As yet, experiments with dogs do not extend to this low-dose range, but tumors have been observed for exposures at the 600-WLM level.[13] It is of interest that lung-cancer incidence in animals increases with a decreasing rate of exposure for fixed cumulative exposure—a finding that has yet to be confirmed in studies of exposed underground miners (Annex 2A). The difficulty of documenting exposure rate for the miners may explain the failure to find a dose-rate effect in the epidemiological studies.

TABLE III-2 Summary of Factors Influencing the Tumorigenic Efficiency of Radon-Daughter Exposure

Factor	Effect on Respiratory Tract Tumor Incidence
Radon-daughter cumulative exposure	Increases approximately linearly with exposure
Radon-daughter exposure rate	Increases with decrease in exposure rate (~200 to 400% increase from about 500 to 50 WLM/wk)
Radon-daughter unattached fraction	Increases with increase in unattached fraction, f_a (~50% increase per WLM exposure from 2 to 10% f_a)
Radon-daughter disequilibrium	Increases with increase in disequilibrium (~30% increase per WLM exposure [borderline significance] from 0.4 to 0.1 equilibrium).
Concomitant exposure to cigarette smoke	Decreases if smoking alternates on same day with radon-daughter exposures
	Increases if smoking follows cumulative radon-daughter exposures
	No effect if smoking precedes cumulative radon-daughter exposures

Large-scale animal studies may become useful for elucidating the interactions between radon daughters and other inhaled pollutants. Information on the extent and duration of smoking is incomplete for human studies, but smoking can be controlled in experiments with animals. It is clear from such experiments that the interactions between smoking and lung cancer induced by radon decay products reflect a complex interplay of these agents in the host. Well thought out experiments with dogs and rats can provide models that aid our understanding of how smoking modulates radiogenic lung cancer. Nevertheless, application to humans is indirect, and confirming experiments with primates may be necessary. However, findings in humans and animals to date are generally parallel for short-half-life radon progeny.

REFERENCES

1. Bale, W. F. 1951 Hazards associated with radon and thoron. Division of Biology and Medicine, U.S. Atomic Energy Commission, Washington, D.C.: March 14. Memorandum. (Also found in Health Phys. 38:1061, 1951.)
2. Chameaud, J., R. Perraud, J. Lafuma, R. Masse, and J. Pradel. 1974. Lesions and lung cancers induced in rats by inhaled radon-222 at various equilibriums with radon daughters. P. 411 in Experimental Lung Cancer. Carcinogenesis and Bioassays, E. Karbe and J. F. Park, eds. New York: Springer-Verlag.
3. Chameaud, J., R. Perraud, J. Chretien, R. Masse, and J. Lafuma. 1976. Lung cancer induced in rats by radon and its daughter nuclides at different concentrations. P. 223 in Biological and Environmental Effects of Low Level Radiation, Vol. II. Publication STI/PUB/409. Vienna: International Atomic Energy Agency.
4. Chameaud, J., R. Perraud, J. Chretien, R. Masse, and J. Lafuma. 1980. Combined effects of inhalation of radon daughter products and tobacco smoke.

P. 551 in Pulmonary Toxicology of Respirable Particles, CONF-791002, C. L. Sanders, F. T. Cross, G. E. Dagle, and J. A. Mahaffey, eds. Springfield, Va.: National Technical Information Service.

5. Chameaud, J., R. Perraud, R. Masse, and J. Lafuma. 1981. Contribution of animal experimentation to the interpretation of human epidemiological data. In Proceedings of the International Conference on Radiation Hazards in Mining: Control, Measurement and Medical Aspects, M. Gomez, ed. Kingsport, Tenn.: Kingsport Press, Inc.
6. Chameaud, J., R. Perraud, and J. Lafuma. 1982. Cancers induced by Rn-222 in the rat. In Proceedings of Specialist Meeting on Assessment of Radon and Daughter Exposure and Related Biological Effects, G. F. Clemente, A. V. Nero, F. Steinhausler, and M. E. Wrenn, eds. Salt Lake City, Utah: RD Press.
7. Chameaud, J., R. Masse, and J. Lafuma. 1984. Influence of radon daughter exposure at low doses on occurrence of lung cancer in rats. Radiat. Prot. Dosimetry 7:385–388.
8. Chameaud, J., R. Masse, M. Morin, and J. Lafuma. 1985. Lung cancer induction by radon daughters in rats. Pp. 350–353 in Proceedings of the International Conference on Occupational Radiation Safety in Mining, Vol. 1, H. Stocker, ed. Toronto: Canadian Nuclear Association.
9. Cohn, S. H., R. K. Skow, and J. K. Gong. 1953. Radon inhalation studies in rats. Arch. Ind. Hyg. Occup. Med. 7:508.
10. Cross, F. T. 1984. A Review of Radon Inhalation Studies in Animals with Reference to Epidemiological Data. Research Report for SENES Consultants, Ltd., Ontario, Canada. Richland, Wash.: Battelle, Pacific Northwest Laboratories.
11. Cross, F. T, R. F. Palmer, R. E. Filipy, R. H. Busch, and B. O. Stuart. 1978. Study of the Combined Effects of Smoking and Inhalation of Uranium Ore Dust, Radon Daughters and Diesel Oil Exhaust Fumes in Hamsters and Dogs. PNL-2744. Springfield, Va.: National Technical Information Service.
12. Cross, F. T., R. F. Palmer, R. H. Busch, R. E. Filipy, and B. O. Stuart. 1981. Development of lesions in Syrian golden hamsters following exposure to radon daughters and uranium ore dust. Health Phys. 41:135–153.
13. Cross, F. T., R. F. Palmer, G. E. Dagle, R. E. Filipy, and B. O. Stuart. 1982. Carcinogenic effects of radon daughters in uranium ore dust and cigarette smoke in beagle dogs. Health Phys. 42:35–52.
14. Cross, F. T., R. F. Palmer, R. H. Busch, and R. L. Buschbom. 1982. Influence of radon daughter exposure rate and uranium ore dust concentration on occurrence of lung tumors. Pp. 189–197 in Proceedings of Specialist Meeting on Assessment of Radon and Daughter Exposure and Related Biological Effects. Salt Lake City: Salt Lake City Press.
15. Cross, F. T., R. F. Palmer, R. H. Busch, G. E. Dagle, R. E. Filipy, and H. A. Ragan. 1983. An overview of the PNL radon experiments with reference to epidemiological data. In Proceedings of the 22nd Hanford Life Sciences Symposium. Richland, Wash.: Batelle, Pacific Northwest Laboratory.
16. Cross, F. T., R. L. Buschbom, G. E. Dagle, R. E. Filipy, P. O. Jackson, S. M. Loscutoff, and R. F. Palmer. 1983. Inhalation hazards to uranium miners. P. 77 in Pacific Northwest Laboratory Annual Report for 1982 to the Department of Energy Assistant Secretary for Environment, Part 1. PNL-4600. Richland, Wash.: Battelle, Pacific Northwest Laboratory.
17. Cross, F. T., R. L. Buschbom, G. E. Dagle, P. O. Jackson, R. F. Palmer, and H. A. Dagan. 1984. Inhalation hazards to uranium miners. P. 41 in Pacific Northwest Laboratory Annual Report for 1983 to the Department of Energy Assistant Secretary for Environment, Part 1. PNL-5000. Richland, Wash.: Battelle, Pacific Northwest Laboratory.

Battelle, Pacific Northwest Laboratory.

18. Cross, F. T., R. F. Palmer, G. E. Dagle, R. E. Busch, and R. L. Buschbom. 1984. Influence of radon daughter exposure rate, unattachment fraction, and disequilibrium on occurrence of lung tumors. Radiat. Prot. Dosimetry 7:381–384.
19. Gray, R. G., J. Lafuma, S. E. Parish, and R. Peto. In press. Radon inhalation by rats: An examination of the dose-response relationship for induced pulmonary tumours and of the exposure parameters and cofactors affecting risk. In Proceedings of the 22nd Hanford Life Sciences Symposium. Richland, Wash.: Battelle, Pacific Northwest Laboratories.
20. Harris, S. J. 1954. Radon levels in mines in New York State. Arch. Ind. Hyg. Occup. Med. 10:54–60.
21. Jackson, M. L. 1940. The Biological Effects of Inhaled Radon. M.S. Thesis. Massachusetts Institute of Technology.
22. Jansen, H., and P. Schultzer. 1926. Experimental investigations into the internal radium emanation therapy. I. Emanatorium experiments with rats. Acta Radiol. 6:631.
23. Kushneva, V. S. 1959. P. 21 in On the Problem of the Long-Term Effects of Combinad Injury to Animals of Silicon Dioxide and Radon. TR-4473. Washington, D.C.: U.S. Atomic Energy Commission.
24. Little, J. B., R. B. McGandy, and A. R. Kennedy. 1978. Interactions between polonium-210, α-radiation, benzo(a)pyrene, and 0.9 NaCl solution instillations in the induction of experimental lung cancer. Cancer Res. 38:1929.
25. Morken, D. A. 1955. Acute toxicity of radon. Arch. Ind. Health 12:485.
26. Morken, D. A. 1973. The biological effects of the radioactive noble gases. P. 469 in Noble Gases, R. Stanley and A. A. Moghissi, eds. CONF-730915. U.S. Energy Development and Research Agency, Washington D.C.: National Environmental Research Center.
27. Morken, D. A. 1973. The biological effects of radon on the lung. In Noble Gases, R. Stanley and A. A. Moghissi, eds. CONF-730915. U.S. Energy Development and Research Agency, Washington D.C.: National Environmental Research Center.
28. Morken, D. A., and J. K. Scott. 1966. Effects on Mice of Continual Exposure to Radon and Its Decay Products on Dust. University of Rochester Atomic Energy Project Report UR-669. Springfield, Va.: National Technical Information Service.
29. Read, J., and J. C. Mottram. 1939. The "tolerance concentration" of radon in the atmosphere. Br. J. Radiol. 12:54.
30. Schlesinger, R. B., and M. Lippman. 1978. Selective particle deposition and bronchogenic carcinoma. Environ. Res. 15:424.
31. Shapiro, J. 1954. An Evaluation of the Pulmonary Radiation Dosage from Radon and Its Daughter Products. University of Rochester Atomic Energy Project UR-298. Rochester, N.Y.: University of Rochester.
32. Shami, S. G., L. A. Thibodeau, A. R. Kennedy, and J. B. Little. 1982. Proliferative and morphological changes in the pulmonary epithelium of the Syrian golden hamster during carcinogenesis initiated by ^{210}Po α-Radiation. Cancer Res. 42:1405.
33. Stuart, B. O., R. F. Palmer, R. E. Filipy, and J. Gaven. 1978. Inhaled radon daughters and uranium ore dust in rodents. In Pacific Northwest Laboratory Annual Report for 1977 to the Department of Energy Assistant Secretary for Environment, Part 1. PNL-2500. Richland, Wash.: Battelle, Pacific Northwest Laboratory.

APPENDIX IV

Epidemiological Studies of Persons Exposed to Radon Progeny

INTRODUCTION

The mining of radioactive ores in the Erz Mountains in eastern Europe was the first occupation associated with an increased risk of lung cancer. Metal ores were mined in Schneeberg, on the German side of the mountains, beginning in the fifteenth century, and in Joachimsthal, on what is now the Czechoslovakian side, beginning in the sixteenth century.[26,30] Both areas were later mined for radioactive ores. As early as the sixteenth century, Agricola[1] described exceptionally high mortality from respiratory diseases in miners in this region. The lung-cancer hazard was first recognized by Harting and Hesse[19] and was reported in 1879. Their report provided clinical and autopsy descriptions of intrathoracic neoplasms in miners, which they classified as lymphosarcoma. In a work force of about 650 men, Harting and Hesse counted 150 deaths from "miner's disease" between 1869 and 1877; in retrospect, most of these deaths were probably from lung cancer. During the early twentieth century, histopathological review of a series of cases established that the malignancy prevalent among miners in the Erz Mountains was primary cancer of the lung.[5,49]

The problem was not recognized in the miners on the Czechoslovakian side of the Erz Mountains until 1929, when two cases of lung cancer were reported in Joachimsthal miners. In 1932, Pirchan and Sikl[46] described the autopsy findings in nine miners with lung cancer. These 9 miners were among 19 miners in Joachimsthal who died during 1929–1930. Formal epidemiological studies of the Schneeberg and Joachimsthal miners were not carried out, but published reports documented that about 50% of the miners eventually died from lung cancer.[53] Peller[44] calculated lung-cancer

mortality rates for the Schneeberg miners during 1875–1912 and found that they were about 50 times those in Vienna males during 1932–1936.

Many authors offered explanations of the excess cancer in the Schneeberg and Joachimsthal miners (see references 26, 30, and 63 for reviews). Early theories emphasized dust exposure, metals in the ore (particularly arsenic), and increased susceptibility as a result of inbreeding in small mining communities. In 1924, Ludwig and Lorenser[31] reported that radioactivity could be measured in the air and water in the mines of Schneeberg and might contribute to the development of lung cancer. Pirchan and Sikl[46] suggested in 1932 that radioactivity was the most probable cause of the Joachimsthal cancers, on the basis of the finding of radioactivity in both Schneeberg and Joachimsthal mines, the occurrence of lung-cancer in both locations, and the long exposure of underground miners to radioactivity. Teleky's opinion in 1937 was similar.[63] He could find no other satisfactory explanation and concluded that the high level of radioactivity, thought not to be present in other mines, led to the apparently nnique lung-cancer problem of Schneeberg and Joachimsthal miners. In 1944, Lorenz[30] argued that radon alone could not be the cause of lung cancer and proposed that genetic susceptibility to lung cancer might be unusually high in the miners. However, during the 1950s and 1960s, as the biological basis of respiratory carcinogenesis became better understood and additional mining groups were studied, it came to be accepted that inhaled radon progeny were the cause of lung cancer in the Schneeberg and Joachimsthal miners and other exposed miners.[26,33,53]

After World War II, several new epidemiological studies were initiated to determine the safety of exposure to radon progeny in mines. Unlike early studies, the newer surveys addressed such important biological questions such as the shape of the dose-response curve, the influence on risk of age at exposure, the effect of dose rate, the temporal expression of risk after exposure, and the interaction of radon daughters with other substances associated with lung cancer. This appendix reviews the epidemiological literature that is now available for addressing these issues.

COLORADO PLATEAU STUDY

Beginning in the late 1940s, the American uranium industry grew rapidly in the Colorado Plateau, a mountainous region of southwestern Colorado and southeastern Utah. In 1949, in response to concerns about the health hazards to workers in this industry and with awareness of the high lung-cancer incidence in European miners in Joachimsthal and Schneeberg,[31] the U.S. Public Health Service (PHS) began to investigate the uranium mines and mills in the Colorado Plateau region. The investigation combined an industrial-hygiene survey with a medical study of the

workers. A prospective cohort study of miners and millers was carried out later, first by the PHS and then by the National Institute for Occupational Safety and Health (NIOSH). Until recently, this study offered one of the few epidemiological data bases for estimating the lung-cancer risk associated with exposure to radon progeny.

Field teams from PHS periodically conducted medical surveys of miners and millers between 1950 and 1960.[33] Before 1954, the teams did not attempt to examine all workers, but during 1954–1960, they tried to attain complete coverage. From among the examined miners, a group was assembled for follow-up that included miners who had worked at least a month underground in a uranium mine by January 1, 1964.[33] The number of subjects varied in the reports of this investigation (Table IV-1); in 1971, Lundin et al.[33] provided data on 3,366 white and 780 nonwhite subjects.

An exposure data base was developed from diverse sources: PHS, state agencies, and the mining companies. Holaday (quoted by Lundin et al.[33]) has provided a chronology (Table IV-2). During the period 1951–1968, for which cumulative exposure in working-level months (WLM) was initially calculated, nearly 43,000 measurements of radon-daughter concentrations were made in the approximately 2,500 mines that were worked (Table IV-3).[33] In discussing sources of potential inaccuracy in the working-level (WL) data, Holaday (quoted by Lundin et al.[33]) pointed out that the measurements taken after 1960 were primarily for control purposes and might have led to overestimates of the exposures to miners.

Because coverage was not comprehensive for all mines in all years, several different estimation procedures were used to fill the gaps in the exposure data. These estimation procedures were more important in the earlier years, when exposures to radon daughters were higher and fewer measurements were available.

To make estimates for missing data in the temporal series of WL measurements for a particular mine, the investigators interpolated and extrapolated earlier and later concentrations of radon daughters. When gaps in the data were too wide, area averages by locality, district, and state were used. For 1950 and earlier years, WL values were estimated on the basis of the few available radon measurements and the investigators' knowledge of the mining conditions. Many of the miners worked in other types of hard-rock mines before becoming uranium miners. For exposures to radon daughters in the hard-rock mines, WL values were based on calendar year: 1.0 WL for years before 1935, 0.5 WL for 1935–1939, and 0.3 WL for years 1940 and later.[33]

The arithmetic average of the individual WL measurements made within a mine in a given calendar year was assigned to the mine for that year. Use of the arithmetic average implicitly weighted all measurements equally; error would have been introduced if the numbers of workers

TABLE IV-1 Results of Colorado Plateau Study (Summarized from Principal Reports) of Male Uranium Miners

Followup Cutoff Date	No. of Subjects	No. of Lung-Cancer Deaths (Observed/Expected)	Comment	Reference
1959	2,666	6/3	Increase not statistically significant	2
1959	907	5/1.1[a]	Cohort members with at least 3 yr of experience	2
1962	3,656	15/4.2[a]	Includes 1,156 workers with surface, open-pit, or occasional underground work, respectively, through 1960	65
1963	3,415	22/5.7[b]	Response increases with cumulative WLM	66
1967	3,414	62/10.0[b]	Excess lung cancer in all exposure categories from < 120 WLM to 3,720 WLM	32
1968	3,366	70/11.7[b]	Most comprehensive report	33
1974	3,366	144/29.8[b]	Response increases with cumulative WLM in all smoking groups	4
1977	3,362	185/38.4[c]	WLM not considered in analysis	67

[a] $P < 0.05$.
[b] $P < 0.01$.
[c] SMR is 482, 95% lower confidence limit is 425.

TABLE IV-2 Chronology of Radon and Radon-Daughter Measurements in Colorado Plateau Study[a]

Time	Source and Type of Measurement
Before 1950	Few radon measurements; earliest 1949
1950	Few radon samples by the PHS and Colorado State Health Department
1951	Radon and radon-daughter samples by PHS, state agencies, and U.S. Bureau of Mines
1952	Radon and radon-daughter samples taken by PHS in attempt to survey all mines
1953-1954	Scant data collection by PHS and states; Utah tried to survey every mine
1955	Scant PHS coverage; variable among states; mining companies begin measurements
1956-1958	Mine survey work primarily by companies
1959-1960	Colorado, Utah, and New Mexico conducted surveys; company measurements continued
After 1960	State and mining-company programs

[a]Based on data from Lundin et al.[33]

exposed at the concentrations indicated by the measurement were not uniform. The arithmetic average could also be strongly influenced by outlying high values.

To calculate WLM, the WL estimates were combined with work-history information obtained from annual censuses of active miners and from questionnaires. Apparently, a 170-h work month was assumed; and time for vacations, sick leave, or other absences from work was not subtracted from the number of underground hours estimated from the work history.[52] However, cumulative exposures were also not adjusted for time worked beyond 170 h/month, a common practice in the early years of the industry.[52] The investigators did not have enough information to consider work location within a specific mine or job classification, which might have influenced ventilatory demands.

Because WL measurements were sparse in relation to the numbers of mines that were worked, the WLMs accumulated by most miners were based on both measurements and estimates. In fact, WLM totals were calculated solely from measurements on only 10.3% of the white miners. For 36.1% of the white miners, some type of estimation was involved in the calculation of all WLM values; for the remainder, some WLM estimates were based on WL values derived by one of the estimation procedures.[33] In reports published to date, the WLM estimates have extended through September 1969. Information on cigarette smoking was obtained during the survey examinations, at the annual censuses of miners, and from mailed questionnaires.[33,69] As described by Whittemore and McMillan,[69]

TABLE IV-3 Number of Mines Visited and Number of Measurements Made in Colorado Plateau Study

Calendar Year	No. of Mines Visited	No. of Measurements	No. of Measurements/ Mine Visited
1951	5	21	4.2
1952	151	242	1.6
1953	56	474	8.5
1954	33	143	4.3
1955	4	15	3.8
1956	101	1,434	14.2
1957	147	848	5.8
1958	54	475	8.8
1959	281	1,867	6.6
1960	179	1,785	10.0
1961	330	2,952	8.9
1962	336	4,362	13.0
1963	315	2,648	8.4
1964	268	4,196	15.7
1965	268	4,856	18.1
1966	274	5,084	18.6
1967	266	5,696	21.4
1968	259	5,691	22.0
1969	149	1,683	11.3

SOURCE: Dr. Richard W. Hornung, National Institute of Occupational Safety and Health, Cincinnati, Ohio, personal communication.

information on smoking was obtained on one to four occasions between 1950 and 1960, when the surveys were conducted, and at other times between 1963 and 1969.

Mortality in the cohort was determined with follow-up techniques that included records of the Social Security Administration and the Internal Revenue Service, direct contact, and other approaches.[33,67] Only a few subjects could not be traced, and nearly all death certificates were obtained. Most published reports are based on analysis with a modified life-table approach, which is a conventional method for longitudinal studies that compares observed with expected numbers of deaths by cause. More recently, several investigators have applied modeling techniques to the data.[23,24,34,69] In cohort analyses based on an external referent population, expected numbers of deaths were calculated with mortality rates for the western states where the mines were or with the rates for all U.S. white males.

Table IV-1 summarizes the principal reports for the white male miners. At all follow-up intervals, statistically significant excesses of lung-cancer deaths were reported; the standardized mortality ratios (SMRs), which

TABLE IV-4 Lung-Cancer Deaths by Cumulative WLM in White Underground Miners in Colorado Plateau Study[a]

Cumulative WLM	No. of Lung-Cancer Deaths		
	Observed	Expected	Ratio of Observed/Expected
< 120	1	1.81	0.55
120-359	12	2.57	4.67[b]
360-839	14	2.95	4.75[b]
840-1,799	12	2.52	4.76[b]
1,800-3,719	21	1.43	14.69[b]
≥ 3,720	10	0.42	23.81[b]

[a]Based on data from Lundin et al.[33]
[b]$P < 0.01$.

are age- and calendar-year-adjusted ratios of observed to expected deaths, ranged from approximately 4 to 6, without an obvious temporal trend. In several reports, the investigators used stratified analysis to examine the exposure-response relationship of lung-cancer mortality with cumulative WLM by calculating standardized mortality ratios within strata of increasing WLM.[3,4,32,33,66] In one report,[32] the mortality rates were standardized for cigarette smoking; in another,[4] they were stratified by cumulative WLM and smoking. Lundin et al.[33] adjusted the expected numbers of lung-cancer deaths for cigarette smoking. The investigators usually provided tables stratified by the interval after the start of employment in uranium mining.

Lundin et al.[33] compared observed with expected numbers of lung-cancer deaths in six strata of lifetime cumulative WLM (Table IV-4). A statistically significant excess was present in all categories of exposure, except in the category of less than 120 WLM. Archer et al.[4] provided mortality rates by exposure and cigarette smoking but did not include expected numbers of deaths.

Mortality from causes other than lung cancer was also examined. Significant excesses were not observed for cancers at sites other than the respiratory system.[4,33,67] Greater than expected numbers of deaths occurred from tuberculosis, nonmalignant respiratory diseases, accidents, and suicides. The 1981 report by Waxweiler et al.[67] showed a statistically significant excess of deaths (SMR, 262) attributable to the grouping of chronic and unspecified nephritis and renal sclerosis.

The data on the white uuderground miners have also been analyzed with other statistical approaches. Lundin and coworkers[33,34] developed a descriptive model for the development of lung cancer after radon-daughter

exposure; the model was based on the assumption of a time-latency distribution with the same shape and dispersion as that of leukemia incidence after a single radiation exposure. They used the model to examine the effects of latent period, age at exposure, dose rate, and cigarette smoking and to compare absolute- and relative-risk models for the effect of radon-daughter exposure. They found that the relative-risk model was preferable to the absolute-risk model and that a 10-yr latent period gave the best fit. Effects of age at first exposure and of exposure rate on lung-cancer risk were not demonstrated. With regard to cigarette smoking, Lundin et al.[33,34] concluded that nonsmokers had much less radiation-induced lung cancer and that the excess radiation-induced lung cancer in smokers was not heavily influenced by the extent of smoking.

Assuming an exponential form for the relative hazard, Hornung and Samuels[24] used the Cox proportional-hazards model on data accumulated through the 1977 follow-up date. They found that a lag period of 6–11 yr for exposure was most compatible with the data. The modeling also showed that the exposure-response curve was downward at higher doses; that is, lower exposure rates led to greater effects. On a multiplicative scale for assessing the effects of exposures on lung-cancer risk, smoking and radon-daughter exposure had statistically significant effects, but a cross-product term of the two exposures was not statistically significant. These analyses were limited, however, to examination of only the exponential form of the relative risk.

More recently, Hornung and Meinhardt[23] reported on a proportional-hazards analysis of data based on follow-up of the cohort through December 31, 1982. A total of 255 deaths from lung cancer was identified by that date. Hornung and Meinhardt considered exponential, linear, and power-function models of risk and chose the power-function model, because it provided the best fit to the data. The model was developed with a stepwise approach; the data were best fitted by variables for cumulative WLM, cumulative smoking (in packs), and age at initial exposure. In the power-function model, the coefficient for the interaction of radon-daughter exposure and cigarette smoking was negative, although it was of borderline statistical significance ($P = 0.058$). This finding implies a submultiplicative, rather than purely multiplicative, interaction between cigarette smoking and radon-daughter exposure.

Hornung and Meinhardt[23] assessed the effects of several temporal factors: exposure rate, calendar year, age at exposure, and cessation of exposure. They found increasing risk with decreasing exposure rate, greater risk for more recent birth, greater risk for those first exposed at a greater age, and decreasing risk with cessation. The last two effects were thought to suggest a late-stage action of radon daughters, in the context of a multistage model.

Hornung and Meinhardt[23] used their power-function model to develop risk estimates for occupational exposures. Quantitative relative-risk estimates were made for occupational exposure beyond an assumed background exposure rate of 0.4 WLM/yr. For a 30-yr working lifetime, risk estimates were made for exposures of 30–120 WLM (1–4 WLM/yr). The relative risks ranged from 1.42 at 30 WLM to 2.07 at 120 WLM.

Whittemore and McMillan[69] used a case-control approach to examine additive and multiplicative models for the relationship of lung-cancer mortality to radon-daughter exposure and cigarette smoking. The results of their analyses are discussed briefly here and more fully in Appendix VII. A multiplicative linear model, with excess relative risk given by the product of the risk associated with radon-daughter exposure and that associated with cigarette smoking, fitted the data better than an excess-relative-risk model in which excess risks associated with radon and smoking were added. A series of multiplicative relative-risk models was evaluated by the investigators. They found a better fit for a model that incorporated the effects of smoking and WLM on relative risk as simple linear variables than for one that included exponential representations of these factors. Cumulative exposure variables fitted the data better than measures of exposure rate. Risk was not affected by age at the start of underground mining.

The PHS study cohort also includes nonwhite male miners, primarily American Indians. These subjects are of particular interest because of the low incidence of lung cancer in American Indians of the Southwest—a pattern probably attributable to a low prevalence of cigarette smoking.[4,50] Less information has been reported on the nonwhite subjects (Table IV-5). No cases of lung cancer among American Indians were observed initially, but a statistically significant excess was present in the 1974 follow-up.[4] In fact, the expected numbers of cases were probably overestimated because of the use of mortality rates for all nonwhites rather than for American Indians alone. In New Mexico during 1969–1977, for example, the average annual lung-cancer mortality rate in American Indian males was 8.6/100,000, whereas the rate for non-Hispanic white males was 60.8/100,000.[50] Lung-cancer mortality rates for black males have generally been equal to or higher than rates for white males.

Two other reports have addressed lung-cancer risks in American Indians employed in the Colorado Plateau mines. Gottlieb and Husen[18] reported a case series of 17 Navajo males diagnosed as having lung cancer at the Shiprock Indian Health Service Hospital. All but one had worked as a uranium miner, and only two had smoked cigarettes; cumulative WLM ranged from 59 to 2,125. Samet et al.[51] conducted a population-based case-control study to assess the association between uranium mining and lung cancer in Navajo males. Of 32 lung-cancer cases diagnosed between 1969 and 1982, 23 had a documented history of uranium mining. None of

TABLE IV-5 Data on Nonwhite Male Underground Uranium Miners in Colorado Plateau Study

Followup Cutoff Date	No. of Subjects	No. of Lung-Cancer Deaths, Observed/ Expected	Comment	Reference
1959	640	0/[a]	Total of 11 deaths	2
1962	1,103	0/0.8	Comparison rates from state data	65
1974	780	11/2[b]	Comparison with male nonwhite population of Arizona and New Mexico	4

[a]Not reported.
[b]$P < 0.01$.

the 64 matched controls had been uranium miners. The results imply an extremely high relative risk in this nonsmoking population, but individual WLM estimates were not available for all miners, and the data cannot be used for quantitative risk estimation.

The Colorado Plateau study was designed and implemented 35 yr ago. Its strengths include the size of the cohort, the long duration of follow-up, the estimation of WLM for individual subjects, and the availability of cigarette-smoking histories. Application of new techniques to the data set has helped to explore the interaction between cigarette smoking and radon daughters and the effects of time-dependent factors such as dose rate and lag times. Even though investigators have dealt pragmatically with the severely limited number of WL measurements in calculating WLM estimates, the quality of the exposure information must be considered in interpreting the results of the study. Both random error and systematic bias might affect WLM estimates. Much of the exposure occurred before extensive measurement procedures were in place. For example, 36.1% of the total WLM ultimately accumulated by the cohort of white miners occurred before 1956 (Richard W. Hornung, NIOSH, personal communication, 1986). Few measurements were taken during the early years, when exposure rates were highest, so the higher exposures were probably estimated less accurately than later ones. If higher exposures were subject to a greater misclassification, the risk coefficients that have been calculated for the higher WLM values might be artificially low. Bias could also have been introduced by the investigators' decision to rely on measurements taken for control purposes after 1960, in that such measurements can over-represent higher exposures. Finally, the cohort had relatively high exposures and thus provides little information on the results of cumulative exposures of less than 100 WLM.

CZECHOSLOVAKIAN URANIUM MINERS

The retrospective cohort mortality study of the Czechoslovakian uranium miners was initially reported in 1971,[55] and periodic updates have been published.[22,28,29,47,54,56,58] The cohort consisted of miners who began mining uranium ore in 1948–1957. However, the results in the more recent reports are limited to 2,433 miners[55] who began in 1948–1952. The selection criteria for the cohort have not been specifically described. The investigators have not reported whether the study cohort included all eligible miners in a particular geographic area or only a sample, what procedure was used and what records were reviewed to identify the cohort, the total number of miners who died from any causes other than lung cancer, and the distribution of the cohort members by birth year, age, or age when first exposed.

Individual work histories were abstracted from payroll cards for all miners (Langon Swent, personal communication, 1984) from 1948. For each miner, WLM was estimated from radon gas measurements and the number of months of employment at each mine in each calendar year. Since 1948, more than 120,000 radon gas measurements were made by measuring ionization current in an ionization chamber by electrometer. Yearly numbers of radon measurements were not given, but the lowest reported mean number of measurements for a year was 101 ± 8/mine. The range of coefficients of variation of average yearly radon concentrations in mines was 3.5–20.0%. Radon gas concentrations were converted to WL on the basis of ventilation conditions and practices, emanation rates from different types of ores, and after 1959, radon-daughter measurements. Since 1968, each miner's WLM has been determined from individual personal dosimetry cards. Assessment of dosimetry errors was based on the magnitudes of coefficients of variation, which do not provide information on the validity of the dosimetry data.

The cohort was followed with lung-cancer registrations administered by the authors in health facilities, the records of the hygiene service in the uranium industry, and oncology notification cards from throughout the country. The latter two served as independent follow-up sources after 1960. Until 1960, only 12 deaths due to lung cancer occurred. The success of this approach for identifying lung-cancer cases is not established, and the number of persons lost to follow-up is not given in the 1976 report by Sevc et al.[56] Except for a paper on skin cancer, health effects other than lung cancer have not been reported.

In analyses of this cohort, observed lung-cancer mortality was compared with that expected on the basis of age- and calendar-period-specific rates of the male population in Czechoslovakia. In the 1976 report by Sevc et al.,[56] person-years at risk for each subject were classified by the

final cumulative WLM category, rather than being distributed across the appropriate WLM categories as they accumulated. This error was corrected in later analyses,[28,29] and only the later analyses are considered here. According to Swent (personal communication, 1984), a miner must have worked at least 4 yr underground to be eligible for inclusion in the cohort. However, person-years at risk were counted from the first date underground, rather than from the date of eligibility, so expected deaths were slightly overestimated.

Cigarette smoking was not assessed for all cohort members individually, but results of studies on a random group of 700 miners indicated that about 70% of the uranium miners were smokers. Data were not given on the amount smoked or the age when smoking started. According to Sevc et al.,[56] the prevalence of smoking in the general male population of Czechoslovakia was comparable with that in the sample of miners. Radon-daughter and other exposures from prior hard-rock mining were not evaluated, because less than 2% of the cohort miners had previously mined nonuranium ores. Other characteristics of this sample have not been reported.

The most recent and thorough analyses were based on follow-up through 1975 of miners who began exposure in 1948–1952. Follow-up averaged 26 yr.[29] In these modified life-table analyses, observed minus expected (based on the male population in Czechoslovakia) lung-cancer deaths were calculated for five categories of cumulative WLM (less than 100, 100–199, 200–399, 400–599, and 600 and over) and, with further stratification, for three categories of duration of exposure (0–7.9 yr; mean, 5.6 yr; 8–11.9 yr; mean, 9.5 yr; and 12 yr or longer; mean, 14 yr) or for three temporal exposure patterns. A temporal pattern of exposure was modeled for each miner individually by the regression, cumulative WLM $= a^b$, where cumulative WLM was calculated for each year of work, t. The cohort was then divided into three groups based on individual members' value of b: group A, b significantly less than 1, implying a high rate followed by a low rate of exposure; group B, b not significantly different from 1, implying a fairly constant rate of exposure; and group C, b significantly greater than 1, implying a low rate followed by a high rate of exposure.

The authors reported two general findings. First, the analyses indicated significant effects of cumulative WLM, duration of exposure, and their interaction. In this study, excess risk is expressed as the excess number of lung cancers per 1,000 miners (Table IV-6) and not per person-year, as is reported in most other cohort studies reviewed here. For those exposed 12 yr or longer, this risk was linearly related to cumulative WLM for miners overall (Figure IV-1), but not for the two shorter exposure periods. Second, excess risk was linearly related (Figure IV-2) to cumulative WLM for miners in groups A and B, but not group C (low followed by

TABLE IV-6 Lung-Cancer Mortality among Czechoslovakian Uranium Miners[a]

Cumulative WLM	Mean WLM[b]	No. of Excess Lung Cancers/1,000 Miners[c]
<100	72 ± 1.8	13.5 (−8.5, 54.3)
100–199	150 ± 1.5	46.6 (28.1, 69.8)
200–399	285 ± 2.6	87.3 (66.9, 109.8)
400–599	570 ± 6.0	116.4 (82.2, 154.2)
≥600		137.3 (89.0, 199.8)
Total	—	80.9 (68.4, 94.1)

[a]Based on data from Kunz et al.[29]
[b]Values are means ± standard deviation.
[c]The 95% confidence limits are given in parentheses.

high exposure rate). Other analyses of the data[29] indicated a significant effect of cumulative WLM on excess risk, but not of exposure pattern or their interaction. From Figures IV-1 and IV-2, it appears that the 95% Poisson-based confidence intervals are wide enough to allow nonlinear interpretations of the relationship between excess risk and cumulative WLM within separate groups of exposure duration or temporal pattern.

An earlier report[28] of follow-up through 1973 is the only report on the cohort of Czechoslovakian uranium miners that provided observed and expected mortality rates per 10,000 person-year and observed to expected lung-cancer mortality ratios, in addition to excess lung-cancer deaths. However, only one independent variable, cumulative WLM (<100, 100–199, 200–399, and ≥400), was reported (Table IV-7).

ONTARIO URANIUM MINERS

A retrospective cohort study of Ontario miners[37–39] engaged in various types of mining included a subcohort of uranium miners who met the following criteria:

- received a miner's physical examination required annually by the company any time in 1955–1977 (uranium mining began in 1955 in Ontario);
- worked at least 1 month as an underground uranium miner; and
- had not worked in a job with any known asbestos exposure, in uranium processing (except in mills), or in any uranium mining in another province as an employee of Eldorado Nuclear.

Radon-daughter exposure was estimated by different methods for 1967 and earlier and for 1968 and later. For 1968 and later, exposure records of WLM maintained by the mining companies were used. For 1957–1967, the

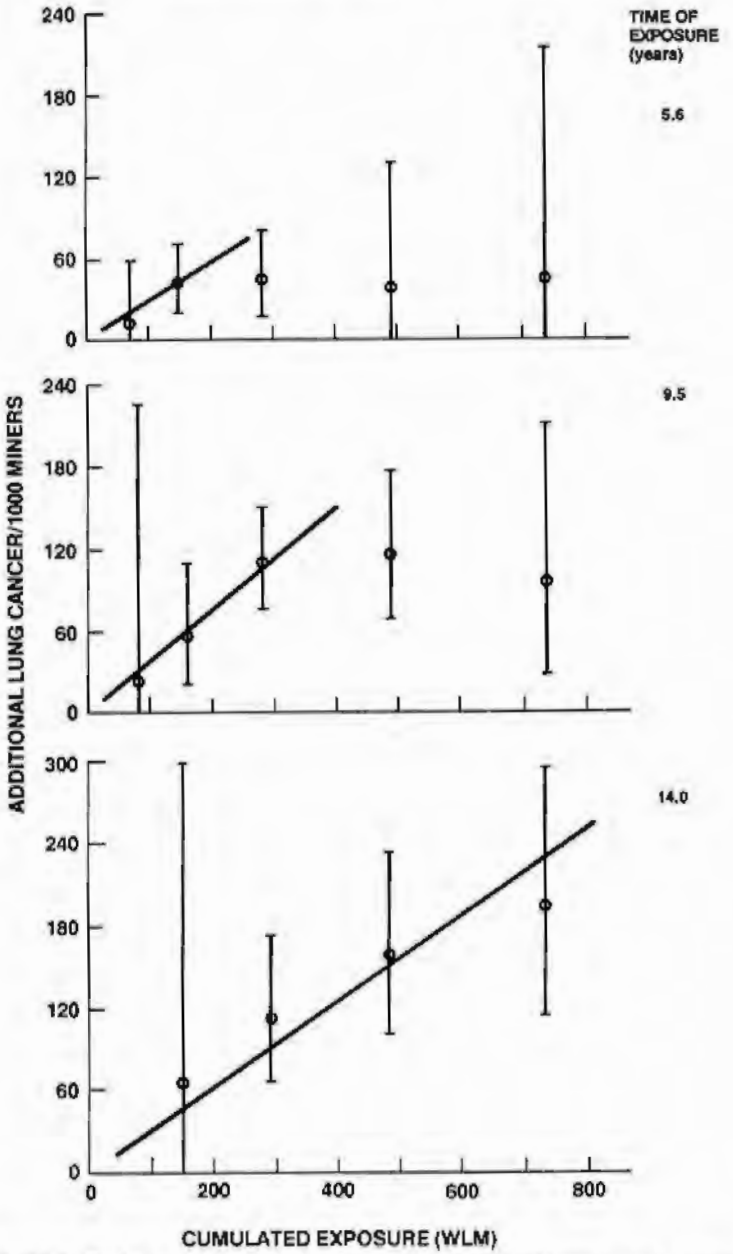


FIGURE IV-1 Relation between additional lung-cancer frequency and cumulative radiation exposure in three groups of Czechoslovakian uranium miners by mean duration of exposure. SOURCE: Kunz et al.[29]

investigators calculated WLM by combining WL information with work histories.[38] Because of the variability of radon-daughter concentrations, the investigators developed two separate sets of WL values for this earlier period. The standard (or lower) WL values were the averages of the four quarterly averages or three 4-month averages for a particular year. To calculate the special (or upper) WL values, the investigators weighted the average of the four highest quarterly measurements or the three highest 4-month measurements in headings, stoops, and raises (a total of 12 or 9 measurements, respectively) by 0.8 and the average of the four highest quarterly or three highest 4-month measurements in travel ways by 0.2. The difference between the standard and special WL values varied with mine and year;[38] for some mines in some years, the special and standard values were equivalent, but the special values were up to 4 times the standard WL estimates in the years and mines for which both were available.[38] The investigators considered that the true exposure of each man lies within this range. During 1958–1967, 13,081 measurements were taken (Table IV-8). For one large mine, WL data for the 4 yr from 1957, when the mine started operating, through 1960 had to be rejected, because they were shown to be unreliable. The values for the missing years were estimated by taking into account tonnage mined, ventilation, and dust concentrations at various times.

Work-history information was obtained primarily from records of preemployment and yearly examinations carried out by Ontario government agencies.[38] Additional information related to the first 5 yr of employment in the mining industry was collected from work-history cards.

The WLM values for 1955–1967 were calculated by combining the work-history information with a matrix of annual WL values for each mine in each year. Adjustment was made for deviations from normal working hours in a mine, considered to be 2,000 h/yr. No estimates of WL were made for prior gold-mining experience, but persons with such experience were analyzed separately, because Ontario gold miners were at increased risk of lung cancer.[40] It should be noted that the committee's analysis of the Ontario miners, described in Annex 2A, excluded miners with previous gold-mining experience.

Because the WL measurements did not cover the complete working experience of the cohort, some estimation of exposures before 1954 was necessary. These years included the period of highest exposures and, as Muller et al.[38] reported, 22% of the total WLM accumulated by the cohort is based on extrapolation from measured values, with account taken of, for example, ventilation. For one large mine, this percentage includes extrapolation up to 1960. The period of extrapolation weighted by WLM is, however, less than 2 yr.

Follow-up through 1981 was carried out by computer linkage with national mortality data bases combined with manual cross-checking to resolve problems. The investigators did not report on the percentage lost to

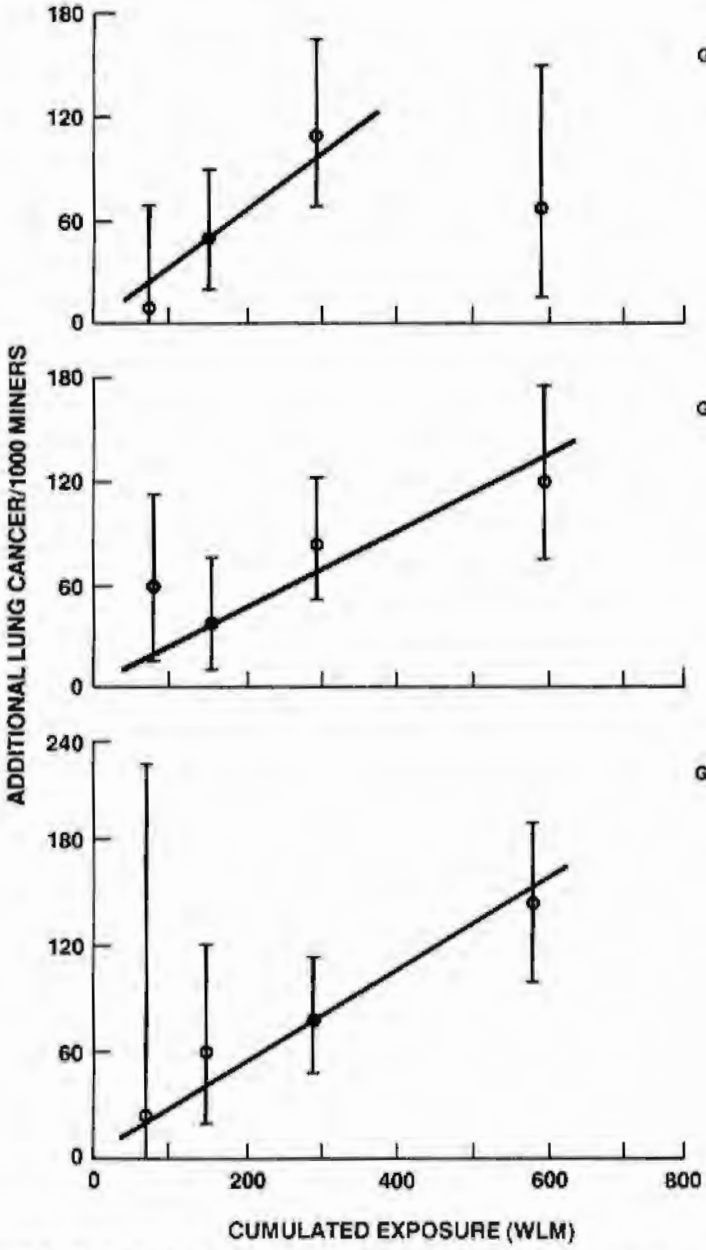


FIGURE IV-2 Relation between additional lung-cancer frequency and cumulative radiation exposure in three groups of Czechoslovakian uranium miners by time course of exposure accumulation (see text). SOURCE: Kunz et al.[29]

follow-up or on the percentage of death certificates not obtained. However, on the basis of a sample of known deaths, 6.3% were not identifiable as deceased with the same follow-up method. Death certificates were the only source of information on cause of death.

Using the modified life-table technique, Muller et al.[38] compared observed with expected mortality (based on the Ontario general male population rates with adjustment for age and calendar period). Results for causes of death other than lung cancer were available only for 1955–1977. The authors did not have information on cigarette-smoking habits of the miners.

The mean cumulative WLM of miners with no previous gold-mining experience was 40 (lower estimate) to 90 (upper estimate). All other descriptions of the cohort included those who had previously mined gold. The median year of birth of the cohort was 1932, and the median year first employed in a mine in Ontario was 1957; thus, the median age at first employment in a mine in Ontario was probably about 25 yr. The median duration of work in a mine was 1.5 yr.

Among uranium miners without any gold-mining experience, Muller et al.[38] found that observed to expected ratios for lung-cancer deaths increased across the six categories of cumulative WLM (Table IV-9). When the upper estimated exposures were used, the first definite excess occurred at a cumulative WLM of 100–170 (mean, 130), with 14 observed and 6.9 expected lung cancers. When the lower estimated exposures were used, there was a definite excess at a cumulative WLM of 40–70 (mean, 53), with 13 observed and 7.0 expected lung cancers. Muller et al.[39] reported that linear regression of the dose-response relationship, weighted by number of person-years at risk (PYAR), showed similar fits for the excess- and relative-risk models.

A 5- and 10-yr exposure lag did not change the slope of the relative-risk model (0.5% excess relative risk per WLM for the upper exposure estimates and 1.3% for the lower ones), but slightly increased the slope of the excess-risk model (from 4.8 to 7.2/million WLM for the lower exposure estimates and from 2.0 to 2.8 for the upper ones). With either model, the use of the upper exposure estimates decreased the dose-response slope by more than 50%. However, dose-response analysis for two age groups of PYAR indicated that the slopes for the relative-risk model were age-independent, whereas the slopes for the excess risk model were not.

ELDORADO URANIUM MINERS

Howe et al.[35] conducted a retrospective cohort mortality study of all 10,945 male employees who had worked at the Eldorado Uranium Mine in Beaverlodge, Saskatchewan, anytime between 1948 (when the mine opened) and December 31, 1980. The cohort was identified from company employment and payroll records. The final study group included

TABLE IV-7 Lung Cancer Among Czechoslovakian Uranium Miners in Relation to Cumulative Radon-Daughter Exposure[a] Based on Modified Life-Table Method[b]

		Calculated No. of Lung-Cancer Deaths/10,000 Person-Years			
Cumulative WLM	No. of Person-Years at Risk	Observed (95% Confidence Limits)	Expected	Additional (Observed − Expected)	Observed/Expected Ratio
<100	9,380	(2.3)- 6.4-(13.9)	5.5	0.9	1.2
100–199	16,131	(17.7)-24.8-(33.8)	7.6	17.2	3.3
200–399	19,614	(34.0)-42.8-(52.4)	7.7	35.1	5.6
≥400	11,830	(54.8)-69.3-(85.5)	8.4	60.9	8.2
Total	56,955	(34.7)-37.2-(42.5)	7.5	29.7	5.0

[a]Start of exposure was 1948–1952; the cutoff date for analysis was December 31, 1973.
[b]Based on date from Kunz et al.[28]

TABLE IV-8 Numbers of Mines and Measurements in Study of Ontario Uranium Miners[a]

Year	No. of Mines	Total No. of Measurements	No. of Measurements/Mine
1958	15	696	46
1959	14	2,145	153
1960	12	1,879	157
1961	7	1,446	207
1962	6	1,563	260
1963	6	1,170	195
1964	5	776	155
1965	4	985	246
1966	3	1,135	378
1967	4	1,286	322

[a]Personal communication, J. Muller, M.D., 1986.

TABLE IV-9 Observed and Expected Lung-Cancer Deaths by Cumulative WLM among Ontario Uranium Miners with No Gold-Mining Experience[a]

Exposure Group	Mean Cumulative Exposure[b] (WLM)	No. of Lung-Cancer Deaths		Observed/ Expected Ratio	No. of Person-Years at Risk
		Observed	Expected		
Cumulative Special WLM (upper estimates)					
0.1-10	5	14	9.5	1.47	45,055
10.1-40	22	15	17.4	0.86	62,173
40.1-100	64	12	13.2	0.91	47,154
100.1-170	130	14	6.9	2.03	22,041
170.1-340	235	13	6.4	2.03	18,249
340.1+	510	14	3.4	4.1	8,124
Cumulative Standard (lower estimates)					
0.1-6	3	14	11.7	1.20	51,356
6.1-20	12	13	17.2	0.76	61,823
20.1-40	29	15	11.0	1.36	38,751
40.1-70	53	13	7.0	1.86	23,313
70.1-140	98	12	6.0	2.00	17,345
140+	200	15	4.1	3.66	10,208

[a]Based on data from Muller et al.[34]

[b]No exposure lag or minimum latency period was used in estimating the WLM.

8,487 subjects; 1,782 (16%) persons were excluded because of missing or incorrect information, and another 676 (6%) were excluded because they had worked at other company sites. The authors were unable to detect any bias due to these exclusions. Follow-up from 1950 through 1980 was carried

out by linkage with a national mortality data base. Only one person was lost to follow-up.

The WLM values for Beaverlodge uranium miners were calculated by Eldorado Resources Ltd., which operated the mine. Different approaches were used for 1966 and earlier years and for 1967 and later years. For 1966 and earlier, the WL estimates were based on all available measurements of radon and radon daughters (Table IV-10). Equilibrium between radon and its daughters was estimated by comparing paired measurements of radon and radon-daughter concentrations. When paired measurements were unavailable for a particular year, the average of the equilibrium factors for adjoining years was interpolated. Because the distribution of measurements was strongly skewed toward higher values, the annual median, rather than the mean, was used to calculate exposure for each year.* For 1967 and later, radon-daughter measurements were generally available. Geometric means or averages of geometric means were used for the calculations. For some locations, adjustments were made on the basis of working conditions.

In calculating the WLM for the work force, the WL values for each year were adjusted for the extent of underground exposure sustained by workers in eight occupational categories. Dates of employment were used to determine the number of weeks worked in each year. Four weeks of holiday time each year were assumed, and adjustments were made for the changing duration of working hours over the study.

Silica exposures to this cohort were always very low, and diesel machinery was never used underground. Potential confounding from other mining exposures was addressed in one analysis by excluding the 540 men who were included in the Ontario miner study[36] and by excluding miners who had reported previous mining experience elsewhere. No measures of cigarette smoking were reported for cohort members individually.

The final cohort consisted of three groups: surface workers only (48%), underground workers only (45%), and both surface and underground workers (7%). The mean years of first exposure for these three groups were 1966, 1966, and 1963, respectively. The mean ages at first employment were 27.7, 28.8, and unreported, respectively. The mean periods of follow-up were 13.9, 13.5, and 17.3 yr, respectively. The mean durations employed

*A recent review of these calculations submitted to the committee, "Beaverlodge Working Level Month Calculations," Draft 4 by S. E. Frost, has suggested possible underestimation of exposures. New calculations for some years indicate that the choice of the median WL value and the method used to determine equilibrium factors might have resulted in bias toward low WLM estimates. For the years reviewed, use of the arithmetic mean, rather than the median, increases the annual WL value.

TABLE IV-10 Numbers of Radon-Daughter and Radon Measurements in Eldorado Beaverlodge Uranium Miner Study, 1954–1966[a]

Year	No. of WL Samples in 1977 Estimates	Total No. of WL Samples Available	No. of Radon Samples Available
1954	—	20[b]	139
1955	—	—	123
1956	38	33	382
1957	—	—	299
1958	—	—	373
1959	17	58	522
1960	—	4	952
1961	122	108	743[c]
1962	179	181	6
1963	160	210	5
1964	171	163	—
1965	286	304	—
1966	459	413	526

[a]Based on data from Beaverlodge Working Level Month Calculations, Draft 4, by S. E. Frost.
[b]Derived from RaA and RaC′ measurements.
[c]Does not include additional 203 shaft Rn measurements.

were 22.2, 15.0, and 43.9 months, respectively. The means of cumulative WLM were 2.8, 16.6, and 28.9, respectively.

A modified life-table analysis was carried out. Comparisons were made with 5-yr, age- and calendar-period-specific mortality rates for the general male population of Canada.

The finding of no lung-cancer excess among those with less than 5 WLM (19 observed versus 18.36 expected) was interpreted as evidence against strong confounding by cigarette smoking in the entire cohort. Furthermore, among those with greater than 5 WLM, no excess of lung cancer was found within the first 5 yr after exposure began. The authors excluded the first 10 yr of follow-up from further analyses, to be consistent with procedures in other studies, although an excess risk of lung cancer was found at higher doses within 5–9 yr after first exposure (6 observed versus 1.54 expected).

The SMRs for lung cancer increased monotonically (Table IV-11) from the lowest to the highest category of cumulative WLM (0–4, 5–24, 25–49, 50–99, 100–149, 150–249, and 250+). The authors used weighted least-squares regression to describe the exposure-response relationship. Exposure within each category was represented by the mean cumulative WLM, and PYAR was used for weighting. The addition of a quadratic term did not significantly improve the fit of the linear model to the data. When the authors multiplied simple linear functions by exponential terms

to represent a cell-killing parameter, they were unable to fit a biologically appropriate model to the data. Furthermore, a 5-yr lag of exposures changed the linear-regression coefficients by less than 10%, compared with no lag. Howe et al.[25] also investigated the effects of age at first exposure and age at observation. In both cases, the attributable risk was found to be much more dependent on age than was the relative risk.

The Beaverlodge miners have also been included in a larger study of Eldorado Resources Ltd. employees. Nair et al.[41] conducted a retrospective cohort mortality study of all males employed before 1981 at four major operations: a pitchblende mine at Port Radium from 1932 to 1940 (time period excluded from study) that was later a uranium mine during 1942–1960; a refinery at Port Hope, Ontario, which opened in 1932, refined radium until 1954, and refined uranium and converted it to uranium dioxide and uranium hexaflouride until the present; a uranium mine at Beaverlodge from 1953 until the present; and other sites.

The cohort was assembled from a company employee roll that included full name, sex, place and date of birth, and last year known alive. The Port Radium cohort was divided into those who ever and those who never worked underground. Follow-up was limited to computer linkage with a national mortality data base for 1950–1980. Bias might have been introduced by the rejection of a large percentage of each cohort (Port Hope, 38%; Port Radium, 44%; and Beaverlodge, 13%) because of inadequacy in personal data or loss to follow-up before 1950.

Preliminary findings on lung-cancer mortality were reported only as observed deaths due to lung cancer for each work group versus those expected based on national rates (Table IV-12).

Later examination determined that the Port Radium surface cohort included a number of underground miners. These results are not useful for assessing dose-response relationships, because data on WLM were not available.

FRENCH URANIUM MINERS

Tirmarche et al.[64] carried out a retrospective cohort study of all men who began underground uranium mining during 1947–1972 in any of 12 French mines and worked a minimum of 3 months. For 1947–1955, WLM values were based on a few radon measurements, ventilation conditions, ore characteristics, and working methods. Extensive radon measurements were taken later; there were an average of 20–30 values taken per mine/year during 1957–1970 and twice that during 1971–1980. WL was estimated retrospectively using the current equilibrium factor

TABLE IV-11 Observed and Expected Lung-Cancer Deaths by Cumulative WLM, 1950–1980 (First 10 Years of Followup Excluded) Among Eldorado Beaverlodge Uranium Miners[a]

Cumulative WLM	Mean CWLM[b]	Person-Years	No. of Lung-Cancer Deaths		RR[c]	AR[d]
			Observed	Expected		
0–4	0.9	29,818	14	14.46	0.97 (0.53, 1.62)	−15 (−288, 303)
5–24	11.7	14,815	12	6.48	1.85 (0.96, 3.24)	373 (−19, 978)
25–49	35.6	5,554	5	2.64	1.89 (0.61, 4.42)	425 (−183, 1,625)
50–99	69.8	3,755	6	2.48	2.42 (0.89, 5.26)	937 (−75, 2,817)
100–149	121.1	1,607	7	1.17	5.98 (2.41, 12.35)	3,628 (1,024, 8,248)
150–249	187.4	1,051	6	0.76	7.89 (2.88, 17.10)	4,986 (1,269, 11,705)
250+	294.9	342	4	0.28	14.29 (3.87, 36.35)	10,877 (2,366, 29,165)
Total	20.2	56,942	54	28.27	1.91 (1.43, 2.49)	452 (216, 741)

[a]Based on data from Howe et al.[25] Observed and expected deaths denote the number of deaths based on age-specific and calendar-year-specific Canadian national mortality rates, 1950–1980.

[b]Weighted by person-years at risk.

[c]Relative risk: observed/expected (with 95% confidence limits).

[d]Attributable risk: [(Observed − Expected)/PQ] × 10^6 (with 95% confidence limits).

TABLE IV-12 Standard Mortality Ratios for Lung Cancer among Eldorado Employees[a]

Site	No. of Lung-Cancer Deaths		SMR
	Observed	Expected	
Beaverlodge	112	60.87	184
Port Hope	14	17.16	82
Port Radium (surface)	28	15.97	175
Port Radium (underground)	55	14.67	375
Other sites	5	4.53	110

[a]Based on data from Nair et al.[41]

of 0.22. The only epidemiological results were for lung-cancer mortality: 36 observed versus 18.77 expected among the entire cohort without any lag in exposure or latency considerations. No dose-response results were reported. It appears that PYAR for each miner began inappropriately at the date of first employment and not after 3 months of mining. National mortality rates were used for comparison; however, the mines are all in agricultural areas. The WLM data in this study are potentially limited by the lack of measurements for 1947–1955 and the retrospective estimation of the equilibrium factor. Furthermore, cause of death was not known for 25% of the deceased subjects.

CORNISH TIN MINERS

High concentrations of radon and its daughters have been measured in tin mines in Cornwall, England. Fox et al.[17] conducted a retrospective cohort study of mortality in 1,333 men employed in two tin mines in Cornwall during 1939. In comparison with mortality rates for England and Wales, lung-cancer mortality was increased in the underground miners (SMR, 211), but not in surface workers (SMR, 74) or in workers who were not classifiable into either of these two categories (SMR, 94). WLM were not estimated for the subjects. The authors reported government estimates of 25 WLM and 15 WLM, respectively, annually for the two mines.

CHINESE TIN MINERS

Tin has been mined in the Yunnan region of China for centuries,[62] and the miners in this region are known to have arsenic and radon-daughter exposures. Wang et al.[68] identified a cohort of 12,243 underground miners

TABLE IV-13 Lung-Cancer Mortality by Cumulative Radiation Exposure among Chinese Tin Miners[a]

Cumulative WLM	No. of Person-Years at Risk	No. of Lung-Cancer Deaths		Observed/Expected Ratio
		Observed	Expected[b]	
<140	33,302	31	7.1	4.4
140–279	28,468	44	6.0	7.3
280–559	19,111	106	8.2	13.0
560–839	6,436	92	3.8	24.3
840–1,399	7,045	115	3.7	30.7
≥1,400	1,774	45	1.0	43.6
Total cohort	86,136	433	29.8	14.5

[a]Based on data from Wang et al.[66]
[b]Based on Shanghai population, apparently without adjustment for age, sex, or calendar period.

and followed them from 1975 to 1981 for lung-cancer incidence and mortality. Information has not been reported on the selection of study subjects, their duration of work, latency distribution, smoking distribution, follow-up methods, or losses to follow-up. The age distribution of the cohort was not given, but it has been reported that many persons began underground mine work between the ages of 8 and 14 yr;[62] this practice was phased out around 1949. WLM were calculated from detailed individual work histories and systematic radon-daughter measurements at underground work boxes during 1972–1980. Only natural ventilation was used in the mines in 1953–1972, so exposures were assumed to be constant during this interval. Before 1953, some of the mines were smaller, no wet-mining methods were used, and proportionate adjustments were made. Another adjustment was made for exposures before 1949, when more primitive mining methods, including back-carrying of ore through narrow tunnels, were used.

During the follow-up period, lung-cancer incidence was 515/100,000 (499 cases) among underground miners, 41.3/100,000 (59 cases) among surface workers. The investigators did not report the incidence data by dose, years worked, or latency. Lung-cancer mortality for the underground miners was also compared with that of the Shanghai population (Table IV-13), but apparently without adjustment for age, sex, calendar period, or smoking.

In addition to radon daughters, exposure to arsenic was considered to play an etiological role in the lung-cancer excess. Ore samples contained 1.5–3.5% arsenic; the investigators estimated that a miner's respiratory

tract was exposed to 1.97–7.43 mg of arsenic/yr during the years immediately after 1949. Total dust concentrations were estimated at 30 mg/m^3, with peaks during dry drilling of 344 mg/m^3 in the earlier years.

Sun et al.[62] described 929 lung-cancer cases (755 deaths) ascertained during 1954–1978 among workers at three mines of the Geiju Tin Mine Company in Yunnan. Death certificates, histological-cytological reports, and chest x rays were cross-checked to confirm the cases. The little available information on relative risks was based on a crude cohort study that calculated expected deaths using the age distribution of the workers in one of the mines in 1975. The cohort analyses that controlled for duration of mining indicated significant differences in SMRs by age at which the miners began mining. However, for those who began mining before age 14, risk did not increase with duration of mining. The smoking habits of 17,287 miners were recorded. The authors reported that the relative risk for lung cancer in smoking miners was about 20 times higher than that in nonsmokers. There was no significant relationship between latent period and degree of smoking.

The age distribution of the work force in the three mines in 1973 was trimodal, with peaks at 20, 25, and 40 yr, which is a reflection of temporal changes in hiring practices. This unusual age distribution and the emphasis on case ascertainment (i.e., follow-up) during 1971–1978 obscures the relationships among age at which miners started mining, latency period, length of follow-up, and risk. However, of the large number of persons who began work underground before the age of 14, few developed lung cancer before the age of 35.

CANADIAN FLUORSPAR MINERS

The open-pit mining of fluorspar (calcium fluoride) in St. Lawrence, Newfoundland, began in 1933. Underground mining began in 1936 and has been carried out in 12 mines. After the discovery in the 1950s of an unusual excess of lung-cancer deaths among the miners, a retrospective-prospective cohort study was undertaken.[13,14,35,36] The ore itself is not radioactive,[14] but the substantial amounts of water seeping through the mines contain radon gas.

Exposures have been estimated on the basis of occupational histories that include type and place of work and hours of work by year. For years before and to 1960, work hours were converted to working months (167 or 170 h) and used to calculate WLM. WL values were estimated retrospectively from measurements made in only one mine in one year, 1959. Before 1960, the mines were ventilated primarily by natural draft,

TABLE IV-14 Lung-Cancer Mortality by Cumulative Radiation Exposure among Canadian Fluorspar Miners[a]

Cumulative WLM	No. of Person-Years at Risk	No. of Lung-Cancer Deaths		Observed/Expected Ratio	*P* Value
		Observed	Expected		
0	13,657.8	7	7	1.00	—
1-9	3,045.5	3	2.02	1.49	0.50
10-239	9,510.5	13	7.22	1.80	0.09
240-599	5,105.5	10	3.87	2.58	0.06
600-1,979	7,107.0	6	1.71	3.51	0.03
1,980-2,039	2,415.5	25	1.54	16.23	<0.001
≥2,040	1,889.0	40	1.07	37.38	0.001

[a]Based on data from Morrison et al.[36]

occasionally aided by small blowers, and the ventilation varied greatly in each mine, as did the amount of water seepage. Radiation measurements were infrequent during 1960–1968, but were taken daily after 1968.

Recent follow-up of the cohort has been primarily through linkage with the nationwide mortality data base[36] but has also included a small number of deaths certified by local clergy, parish records, hospitals, and relatives, rather than medically. All persons not definitely identified as deceased were assumed to be alive. Initial analyses of lung-cancer mortality in 1952–1960 showed 21 deaths among the miners, compared with 0.7 expected from age-adjusted rates for the remainder of Newfoundland. Recent analyses[35,36] used standard modified life tables of PYAR and age-specific lung-cancer rates among the surface workers for comparison. This analysis is limited by the small number of lung-cancer deaths (seven) in the comparison group and possibly by migration between surface and underground work. Follow-up was accomplished with the use of the company, union, and medical files. Analysis based on follow-up through 1978 (ignoring the first 10 yr of risk after hiring) showed a strong dose-response relationship between lung-cancer risk and cumulative WLM (Table IV-14).[36] Smoking-specific findings are not reviewed because of the lack of adequate ascertainment of smoking status. The authors found that latency periods decreased for men first exposed when older and for men exposed during the earlier years, when exposures were presumably higher.

SWEDISH IRON MINERS: MALMBERGET

A retrospective cohort mortality study by Radford and Renard[48] included miners from two iron mines (in Malmberget and Koskoskulle)

owned by one company (LKAB). Selected for study were the 1,415 men born in 1880–1919 who were alive in 1930 and who worked underground for more than 1 calendar year during 1897–1976. The cohort was identified principally from company and union records of active and pensioned miners that dated back to 1900. Additional men were identified from medical surveys and a few were identified from parish records. Time worked underground was determined from company and union records and medical files. Work histories appear to contain data only by year; July 2 was assumed as a starting and stopping date for underground work. For those who stopped and restarted in 1 yr or started and died in 1 yr, April 1 and October 1 were assumed, respectively. The extent to which the cohort covered all employed miners was evaluated for the years 1942–1946 by comparing person-years underground from two sources: the work histories of the cohort and company records.

The WLM values for this analysis were those calculated by Radford and Renard.[48] As described in their 1984 report, radon dissolved in water was assumed to be a major source of radon daughters in the mines. Comparison of radon measurements in water taken in 1915 with data from 1972 and 1975 indicated constant radon concentrations in groundwater. The first measurements of radon in mine air were obtained in 1968. Radon and radon daughters were later measured by the mining company and by the National Radiological Institute. Past concentrations were then reconstructed on the basis of these measurements in combination with information on ventilation conditions. Radford and Renard assumed that ventilation conditions in 1968–1972, when the measurements were made, were not greatly different from those in the past. In support of this assumption, they cited a pattern of natural convection and data on quartz dust concentrations that extended to the 1930s. Radon daughters were found to be at about 70% equilibrium with radon.*

*A report submitted to the committee, "Comments to the U.S. Mine Safety Health Administration for the American Mining Congress," by Swent and Chambers, questioned some assumptions underlying the historical reconstruction of the exposures for the years before measurements were taken. Because Radford and Renard[48] assumed that water was the major source of radon in the mines and its strength was constant, Swent and Chambers argued that changing mining practices might alter radon influx into a mine, even in the face of a constant concentration in water. In addition, changing ventilation practices over the years could have influenced exposures. In discussing potential bias in the exposure estimates used by Radford and Renard,[48] Swent and Chambers suggested that the direction of changes in exposures would have been downward. If the exposures were, in fact, underestimated, the estimated risk coefficients would exaggerate the actual risk.

Time worked underground was determined from company and union records and from medical files. Adjustments were made for variation in the average number of hours worked underground in a month. Average yearly WLM were calculated for each 10-yr calendar period from the average number of hours per month underground and from radon-daughter concentration in each area, with weighting by the company data on the numbers of person-hours worked underground in each section of the mines.

Follow-up of the cohort depended on parish records and was thorough (only one person was untraceable through 1976). Of the lung-cancer deaths, 70% had been confirmed by autopsy or thoracotomy, but only death-certificate information was used for comparisons. The expected number of cases was based on age- and calendar-year-specific national mortality rates for males since 1951. Accordingly, PYAR and expected deaths were calculated from the later of two dates: January 1, 1951, and January 1 of the year after a miner began work underground. Induction-latency periods were considered in two ways: by excluding PYAR for each miner for 10.5 yr after mining was begun and by lagging the cumulative WLM by 5 yr.

Information on cigarette smoking was not reported for all cohort members individually, but only for a sample of the responses to a 1972–1973 survey of active miners and surface workers and from a 1977 survey of pensioners in the study cohort. In addition, smoking histories were obtained for each lung-cancer death. The authors estimated smoking-specific lung-cancer SMRs for two categories: smokers combined with recent ex-smokers and all others. They based these SMRs on the ratios between a sample of the responses from the surveyed miners and a national population study of the age-specific proportion of smokers and the amount smoked. Interpretation of the SMRs must be constrained by the lack of information for all cohort members on smoking as presently reported for the surveyed miners (556 of 1,294, or 43%), by differences in the periods associated with the questionnaire data from the miners (1972 and 1977) and from the national population sample (1963–1972), and by the use of information provided by the next of kin for deceased lung-cancer cases.

Other potentially confounding variables for lung cancer were considered. Silicosis, examined in a case-control study nested within the cohort, was found to be equally severe and prevalent in lung-cancer victims (14/50) and in age- and work-period-matched controls (26/100). Diesel equipment, with its exhaust, was not introduced into the mines until the 1960s, by which time 70% of the persons who later developed lung cancer had terminated work. Arsenic, chromium, and nickel—known respiratory

carcinogens—were virtually absent in analyses of bedrock. X-ray diffraction of airborne dust samples from the mine showed no identifiable asbestos fibers. Indoor radon concentrations in miners' homes ranged from 0.002 to 0.03 WL, but had been measured in a sample of homes selected because of potentially high concentrations. The lung-cancer rates among nonminers in this region are lower than Swedish national rates.

Of the mining groups exposed to radon daughters, this cohort offers one of the longer follow-up experiences. Over 41% of the cohort (532/1,294) were deceased. The average year first employed underground was 1932, the average age at first employment was 27.8, and the average duration underground was 19.5 yr. The average exposure rate was 4.8 WLM/yr, resulting in an average cumulative WLM of 93.7 (range, 2–300 WLM). Cause-specific and total mortality were assessed with a modified life-table analysis. Excesses of observed deaths were found for total mortality, lung cancer (50 observed versus 12.8 expected), stomach cancer (28 observed versus 15.1 expected), and all causes except cancer combined (393 observed versus 312.6 expected). The latter excess was due to silicosis, occupational accidents, and cardiovascular disease, according to Radford and Renard.[48]

Lung-cancer mortality was studied in detail. Excess risk was not evident until at least 20 yr after the start of underground work. Significantly increased risks were found for both smokers (32 observed versus 11.0 expected; SMR, 291) and nonsmokers (18 observed versus 1.8 expected; SMR, 1,000). The excess-risk coefficient for smokers was 21.8/million person-yr WLM, and for nonsmokers, 16.3. The combined effect of smoking and radon-daughter exposure was reported as nearly additive,[48] although formal statistical testing, as described in Appendix VII, was not carried out. (The rate ratio for smokers versus nonsmokers based on the Swedish population study was estimated by the authors to be 7.4.)

Dose-response relationships were evaluated for five categories of lagged cumulative WLM (0–49, 50–99, 100–149, 150–199, and over 199). An excess of lung cancer was found even in the lowest dose category (8 observed versus 3.4 expected), and the dose-response data were equally consistent with absolute- and relative-risk models, as measured by weighted correlation coefficients. Assessments of effects of age at beginning of work, year of beginning work, and age at risk were undertaken separately and not by multivariate modeling.

SWEDISH IRON MINERS: KIRUNA

A proportionate-mortality-ratio study was carried out in Kiruna, Sweden, to compare cause-specific mortality distributions among underground

iron miners from two companies (LKAB and TGA), surface miners and workers (LKAB), and all other male deaths in Kiruna.[27] Selected for study were all deaths registered in Kiruna that occurred in 1950–1970 among men aged 30–74. Because rates of emigration from Kiruna were very small, the authors considered that nearly all deaths among the miners would have been registered there. Lung-cancer deaths were verified from hospital records for 41 of 42 cases, and autopsies were performed in all 13 cases among miners. An additional analysis attempted to calculate SMRs among active employees of the mines (surface and underground combined), but was limited by the absence of data.

After age adjustment, underground miners experienced 13 lung-cancer deaths (12 were after 1957) versus 4.5 expected based on the cause-specific distribution of deaths among all other residents of Kiruna and versus 4.2 based on the cause-specific distribution of deaths among the entire Swedish male population in 1951–1966. Analyses were not presented on dose-response relationships, latency, or interaction of cigarette smoking and exposure to radon daughters.

The iron mine was an open-pit mine until the 1950s, when underground mining began. Diesels were introduced in the late 1950s. Quartz concentrations were around 7% of the particles smaller than 5 μ. The concentration of radon daughters, measured only since 1970, was 10–30 pCi/liter in most places and much higher in some unventilated areas. In 1966, a survey of all employees showed that about two-thirds of both underground and surface workers were smokers. Information from coworkers and next of kin indicated that 12 of the 13 lung-cancer cases among underground miners were smokers (four of these smoked only a pipe).

SWEDISH IRON MINERS: KIRUNA AND GALLIVARE

A case-control study[11] was carried out on 604 lung-cancer victims who died during 1972–1977 in three counties in northern Sweden. These counties contained two major iron mines, which were in two separate municipalities, Kiruna (containing the Kirunavaara mine) and Gallivare (containing the Malmberget mine). The investigators used next-of-kin interviews to determine underground mining and smoking histories. No estimates were made of WLM or duration of mining. The investigators concluded that their data showed that relative risks for smoking and underground iron mining were between additive and multiplicative in their combined effect. However, the wide confidence intervals in their data are consistent with an additive, a multiplicative, or a more extreme interpretation.

A recent extension of this case-control study[12] included 69 deaths during 1972–1982, but was limited to Kiruna and Gallivare. The median age of the subjects was 66. WLM were not estimated, but lung-cancer risks by duration of underground iron mining and lifetime number of cigarettes smoked were found to fit a multiplicative-risk model based on linear logistic regression. Unfortunately, statistical testing of the model was not reported, and the data were limited by the small numbers of nonsmoking miners (four) and nonsmoking nonminers (two) among the cases.

SWEDISH IRON MINERS: GRANGESBERG

Edling[15] carried out a case-control study of all male residents known to have died of lung cancer during 1957–1977 in the iron-mining town of Grangesberg, Sweden. The unmatched controls (897), who all died of other causes, and cases were submitted to the local iron-mining company for identification of history of underground mining. The author found an age-adjusted rate ratio for lung-cancer deaths associated with employment at a mine (16.6) that was significant (95% confidence interval, 7.7–35.3). Strikingly, 42 of the 47 lung-cancer cases had mined underground.

A separate analysis in the same report of the effect of cigarette smoking[15] added cases through 1980, but included only persons who had been underground miners. A new set of controls (individually matched for age, sex, and year of death) who died from causes other than malignancy and had been underground miners was selected (44 pairs). Smoking histories were obtained from next of kin by telephone. A risk ratio of 2.0 (95% confidence interval, 0.7–5.7) was found for smoking and lung cancer; the author interpreted that as not fully consistent with the general experience of at least a 5-fold to 10-fold risk ratio.

A second case-control analysis on the same population[16] used only controls who died during 1966–1977 at ages over 50. The authors found a lower age-standardized lung-cancer death rate ratio than in the previous analysis (relative risk, 11.7; 95% confidence interval, 5.3–26.0). A separate analysis of smoking similar to the one described above resulted in a risk ratio of 1.5 (95% confidence interval, 0.4–5.3) for smoking and lung cancer, on the basis of 28 matched pairs.

Edling and Axelson[16] also estimated a lung-cancer excess risk per million person-years WLM for miners aged 50–64 (26 excess cases) and aged 65 or greater (54 excess cases). These estimates were made by multiplying the number of miner person-years at risk in Grangesberg during 1966–1977, as estimated from town censuses, by the proportion of controls

from the case-control study who had previously been underground miners. Cumulative WLM was estimated by multiplying the average number of years worked underground by the product of number of cases and 0.5 WL—an exposure assumed to apply to the entire period, according to 1969 mine measurements. Although these risk estimates were based on extensive assumptions, the authors noted that they were in agreement with estimates in the report by the Committee on the Biological Effects of Ionizing Radiations (BEIR III).[42]

GENERAL SWEDISH MINERS

Snihs[59] presented an epidemiological study of miners in all districts of Sweden. Sweden had 60 underground mines; all mined ferrous and sulfide ores, and none mined uranium. Radon measurements were made in all mines in 1969–1970 with 4.80-liter propane containers and ionization chambers. Radon daughters were sampled with conventional glass-fiber filters and analyzed by the Kusnetz method. In March 1972, exposures were limited by regulation to an annual average of 30 pCi/liter. Snihs[59] reported that air brought into the miners for ventilation, rather than water or rocks in the working areas, was the predominant source of exposure in 17 of 22 mines.

A nationwide cohort of all miners aged 20–64 and employed during 1961–1968 was followed during 1961–1971. The follow-up and analytical methods were not described. It is unclear whether follow-up extended beyond 5 yr after employment ended.

Observed and expected lung-cancer deaths during 1961–1968 were compared among underground miners, aboveground miners, and nonminers in the mining districts. The methods were not fully described, but it appears that the estimated annual number of active miners aged 20–64 during this period was multiplied by district age-specific lung-cancer rates to estimate the expected number of deaths. Observed deaths were included if they occurred within 5 yr of cessation of mining. Annual WL was estimated from measurements only after 1969. Cumulative WLM was estimated from the WL estimates and duration of exposure estimates for all workers based on data on those dying of lung cancer. Limitations of the study noted above make it difficult to judge the validity of the dose-response relationship results.

SWEDISH LEAD-ZINC MINERS: HAMMAR

Axelson and Sundell[7] studied Kiruna (Hammar parish) lead-zinc miners with a case-control study embedded in a crude cohort study. In the

case-control study, the 29 cases included all men who died from lung cancer during 1956–1976 in the parish surrounding two physically connected lead-zinc mines. Controls (174) consisted of the first three deaths other than from lung cancer listed before and after the case in the chronologically ordered parish registry, but matching was dropped in the analysis. The authors believed that the registry included fairly complete diagnoses from the death certificates. The local mining company assessed the underground work experience of all subjects.

Smoking habits of the miners were learned from medical files and interviews with two retired foremen. For 2 of 10 subjects on whom smoking information was independently obtained from more than one source, the information was conflicting.

The age-staudardized rate ratio for lung cancer among lead-zinc miners was 16.3 (90% confidence interval, 7.8–35.3). Among underground miners, those who had never smoked (nine) appeared to have longer work-related induction latent periods than smokers (nine) (respective medians of 49 versus 37 yr) and to have a greater risk of developing lung cancer.

NORWEGIAN NIOBIUM MINERS

Solli et al.[60] followed a cohort of employees at a niobium-mining company that operated from 1951 to 1965. Niobium itself is not considered to be carcinogenic, but the ore also contained ^{238}U (0.3–2 ppm) and ^{232}Th (50–300 ppm). Exposure estimates for the cohort were of questionable quality. The WLM from both radon and thoronium progeny was calculated for the employees on the basis of measurements of alpha activity during 2 days in 1959. Among the employees, a strong dose-response relationship was found between lung-cancer risk and cumulative WLM (Table IV-15). Poor dosimetry probably resulted in underestimation of exposures by about a factor of 2, according to the authors. Lifetime occupational histories indicated that three of the subjects had been previously exposed to asbestos and one had mined iron. In addition, 75% of the employees were smokers, compared with 60% of the Norwegian population.

FLORIDA PHOSPHATE WORKERS

Some U.S. phosphate ore contains uranium and radium. Workers involved in the mining and the processing of the ore might be exposed to radon and radon daughters. Two retrospective cohort studies of mortality in Florida phosphate workers have been conducted recently; each was performed because of concern raised by apparent clusters of lung cancer.

TABLE IV-15 Lung-Cancer Mortality among Norwegian Niobium Mine Workers[a]

Cumulative WLM (Corrected/Twofold Underestimate)	No. of Person-Years at Risk	No. of Lung-Cancer Deaths		Observed/Expected Ratio
		Observed	Expected	
0	4,622	0	1.73	0
1–38	1,343	3	0.50	6.0
40–158	1,312	4	0.58	6.9
160–238	147	2	0.07	28.6
≥240	169	3	0.08	37.5

[a]Based on data from Solli et al.[60]

Stayner et al.[61] conducted a study of 3,199 workers employed at a phosphate fertilizer plant. Seven samples were taken for radon progeny; the range was 0.00–0.02 WL. Overall respiratory-cancer mortality was not significantly increased (SMR, 113). Further analysis did not show trends of respiratory-cancer mortality with duration of employment or length of follow-up in white men. In black men, respiratory-cancer mortality was significantly increased in those with more than 20 yr of employment. However, only five cases were identified in black men, and two were in the index cluster.

In a larger study, Checkoway et al.[10] examined mortality in 17,601 white and 4,722 nonwhite male employees of the Florida phosphate industry. Lung-cancer mortality was not significantly increased in either group, in comparison with rates for Florida. When mortality from lung cancer was examined in the workers considered to have potential exposure to alpha radiation, a significant excess was apparent (SMR, 1.08).

These studies do not have sufficiently detailed information on exposure for risk estimation. Individual exposures to radon progeny were not estimated, and information on cigarette smoking was not collected. Furthermore, the limited measurements that have been made indicate radon-daughter concentrations only slightly above background concentrations.

RESIDENTIAL EXPOSURE

Within a building, radon-progeny concentrations are determined by the strength of the source and the rate of air exchange with the outside. Most of the radon in buildings enters from the underlying soil and building materials, although water and utility gas can also contribute radon progeny to indoor air.[43] A wide range of radon-daughter concentrations in dwellings

has been demonstrated, with different radium concentrations in soil and building materials and different air-exchange rates largely explaining the size of the range.

Epidemiological investigations of domestic radon progeny as a risk factor for lung cancer are still preliminary. Both descriptive and analytical approaches have been used to examine the association between radon-daughter exposure in the home and lung cancer. Techniques for estimating lifetime exposure of people to radon daughters from indoor air are not yet available, and surrogates based on residence type or a few limited measurements have been used in the analytical studies. The available studies are insufficient for the development of quantitative risk estimates for associating exposure to radon progeny in the home and lung cancer.

In the descriptive studies, incidence or mortality rates for lung cancer within geographic units have been correlated with measures of exposure for inhabitants of the units. Edling[15a] compared mortality rates for different Swedish counties with background gamma radiation, described as being correlated with indoor exposure to radon and its daughters. For lung-cancer mortality, the correlation coefficients were 0.46 for males and 0.55 for females. Hess and colleagues[20] performed a similar analysis for lung-cancer mortality during 1950–1969 in the 16 counties of Maine. Using average radon concentrations in water as the measure of exposure, they calculated correlation coefficients of 0.46 for males and 0.65 for females. In a study of 28 Iowa towns served by deep wells, lung-cancer incidence increased with the concentration of ^{226}Ra, a possible surrogate for the radon concentration in the water.[9] These descriptive studies, which did not consider the exposures of people to radon daughters and other agents, provided only suggestive evidence that radon progeny exposure in the home increases lung-cancer risk.

The association has been more directly tested in case-control and cohort studies. Axelson et al.[8] conducted a case-control study in a rural area of Sweden. The investigation included 37 cases and 178 controls. Exposure to radon progeny was inferred from the characteristics of the subjects' residences at the time of death. Those who lived in stone houses were assumed to be most heavily exposed to radon daughters, and those who lived in wooden houses were assumed to be least exposed; other types of dwellings were considered to be sources of intermediate exposure. In spite of the crudeness of this exposure classification, residence in stone houses was associated with a significantly increased odds ratio, in comparison with the reference category of wooden houses (by Mantel-Haenszel method; odds ratio, 5.4; 90% confidence interval, 1.5–19). Data concerning cigarette smoking and residence history were not obtained.

Edling and Axelson[16a] conducted a similar case-control study in a rural area of Sweden. The study subjects were residents of the island of Oeland who died during 1960–1978. The geological characteristics of this island were thought to result in strong differences in background radon concentrations within a small area. Inclusion in the study population required at least 30 yr of residence at the same address before death; 23 lung-cancer cases and 202 controls who died from causes other than lung cancer met this criterion. Most of the dwellings were monitored for radon daughters during 3 months of summer and 1 month of winter. The dwellings were also classified on the basis of structural characteristics, as in the earlier study by Axelson et al.,[8] and cigarette-smoking information was obtained from next of kin. Lung-cancer risk was significantly associated with radon-daughter exposure, as assessed by either the measured concentration or the characteristics of the dwelling, and both crude and smoking-adjusted risk estimates were significantly increased. Logistic analysis yielded smoking-adjusted odds ratio, comparing most with least exposed, of 3.9, and the 90% confidence interval was 1.5–10.0.

Pershagen et al.[45] reported the findings of two small case-control studies in Sweden on domestic radon-daughter exposure, one drawn from a larger study in northern Sweden and the other from a twin registry. The investigators assembled each series with 30 case-control pairs, divided equally between smokers and nonsmokers. Exposure to radon was estimated from information on dwelling type; the investigators attempted to consider all residences lived in by the subjects. In the study group from northern Sweden, imputed radon exposures were significantly higher in smokers than in their smoking controls. Estimated exposures to radon progeny were similar in the nonsmoking cases and controls in the series from northern Sweden and in the smoking and nonsmoking cases and controls in the second series (selected from the twin registry).

In the United States, Simpson and Comstock[57] examined the relationship between lung-cancer incidence and housing characteristics. During a 12-yr period in Washington County, Maryland, lung-cancer incidence was not significantly affected by the type of basement construction or building materials. No measurements of radon or its daughters were made. Rather, dwelling-related variables were assumed to be surrogates for radon-daughter exposure.

SUMMARY

Cause-specific mortality risks for a number of the miner groups discussed above are listed in Table IV-16. Without exception, these studies

TABLE IV-16 Cause-Specific Risks of Mortality among Miners Exposed to Radon Daughters[a]

Study	All Causes			Lung Cancer			Tuberculosis			Other NMRD			Nephritis and Nephrosis		
	Obs.	Exp.	SMR	Obs.	Exp.	SMR	Obs.	Exp.	SMR	Obs.	Exp.	SMR	Obs.	Exp.	SMR
Colorado Plateau Uranium Miners[67]	950	600	158	185	38.4	482	14	3.4	409	83	16.6	499	9	3.7	243
Ontario[38,39c]	1,316	1,113	118	119	65.8	181	4	3.0	132	18	9.5	195			
Elliot Lake Ontario Uranium Miners[38]	999	854	117	81	50	162	1	2.3	44	13	7.2	180			
Bancroft Ontario Uranium Miners[38]	244	203	120	30	12.4	241	2	0.6	333	4	1.8	215			
Eldorado and Ontario[38]	198	116	171	33	7.2	458	1	0.4	250	2	1.0	200			
Eldorado–Port radium[41]															
underground	361	225	160	55	14.7	375	3	1.7	176	3	4.4	68	2	1.0	199
surface	340	259	131	28	16.0	175	9	1.8	0	8	5.6	143	0	1.2	
Eldorado, Beaverlodge															
underground	600	487	123	84	30.0	280	1	2.8	36	6	8.5	71	3	2.1	140
surface[41]	515	529	97	28	30.8	91	3	3.0	100	9	10.4	87	1	2.3	44
uranium miners[25]	604	582	104	65	34.2	190									
Swedish iron miners Malmberget[48]	532	409	130	50	14.6	342									
Norwegian niobium workers[61]	78	67.9	115	12	3.0	405									
Newfoundland fluorspar miners (rate ratios)[35]	244	173	(141)	65	6.5	1,000	24[e]	4.4	550	9[h]	5.4	167	1[i]	2.0	50
Cornish tin miners underground surface[17]	276	—	183	28	—	211	31[f]	—	—	41[k]	—	—			
Colorado Plateau uranium miners[68]	9	6.0	150	1	3.0	33	5	2.3	216	1	0.6	153	9	12.0	75
Ontario[38,39c]	21	16.1	130	6	5.8	104	4	4.9	82	2	1.4	145	26	27.4	95

TABLE IV-16 (*Continued*)

Study	Stomach 151[b]			Kidney 180[b]			Skin 190, 191[b]			Bone 196[b]			Lymph. and Hemo. 200-205		
	Obs.	Exp.	SMR	Obs.	Exp.	SMR	Obs.	Exp.	SMR	Obs.	Exp.	SMR	Obs.	Exp.	SMR
Elliot Lake Ontario uranium miners[38]	15	12.2	131	4	4.4	90	2	1.1	187	22	21.1	104			
Bancroft Ontario uranium miners[38]	4	3.1	129	2	1.1	188	1	0.8	122	0	0.2	0	3	4.8	
Eldorado and Ontario[36]	4	1.8	219	0	0.6	0	0	0.5	0	0	0.1	0	2	2.7	74
Eldorado-Port radium[41]															
underground	5	4.2	119	1	1.3	79	1	0.7	147	0	0.3	0	10	9.4	107
surface	2	5.2	39	2	1.4	145	0	0.7	0	0	0.3	0	14	11.6	121
Eldorado, Beaverlodge															
underground	10	8.4	120	2	2.6	76	0	1.6	0	0	0.6	0	9	17.6	51
surface[41]	8	9.5	84	1	2.7	38	2	1.6	127	1	0.7	148	13	21.2	61
uranium miners[25]	*														
Swedish iron miners Malmberget[48]	28	15.1	189										7[d]	4.7	149
Norwegian niobium workers[61]	1			1			0			0					
Newfoundland fluorspar miners (rate ratios)[35]	24[f]	12	(200)												
Cornish tin miners underground surface[17]	10	—	200												

[a]Abbreviations: Obs., observed; Exp., expected; NMRD, Non malignant Respiratory Disease.
[b]Numbers are disease identifiers from ICD, 7th revision.
[c]Including previous gold miners, excluding Eldorado uranium miners.
[d]Lymphoma only.
[e]No significant excess.
[f]Digestive system.
[g]Includes silicosis.
[h]Respiratory disease.
[i]Genitourinary disease.
[j]Silico tuberculosis.
[k]Silicosis alone.

indicate an excess probability of death due to lung cancer and, in many cases, other causes of death as well. Continued follow-up of these miner groups will provide additional information on the association of radon-daughter exposure to lung cancer and perhaps other diseases. As discussed in Chapter 2 and Appendix VII, epidemiological information that includes the smoking status of each participant is of paramount value. The committee suggests that every effort be made to collect and report such information for the studies described in this appendix.

REFERENCES

1. Agricola, G. De Re Metallica. Basel, 1556. New York: Dover Publications. English reprint (Hoover translation). 1950, p. 214.
2. Archer, V. E., H. J. Magnuson, D. A. Holaday, and P. A. Lawrence. 1962. Hazards to health in uranium mining and milling. J. Occup. Med. 4:55–60.
3. Archer, V. E., J. K. Wagoner, and F. E. Lundin, Jr. 1973. Lung cancer among uranium miners in the United States. Health Phys. 25:351–371.
4. Archer, V. E., J. D. Gillam, and J. L. Wagoner. 1976. Respiratory disease mortality among uranium miners. Ann. N.Y. Acad. Sci. 271:280–293.
5. Arnstein, A. 1913. Sozialhygienische untersuchungen über die Bergleute in den Schneeberger Kobaltgruben. Wein. Arbeit. Geb. Soz. Med. 5:64–83.
6. Axelson, O., and C. Edling. 1980. Health hazards from radon daughters in dwellings in Sweden, Pp. 79–87 in Health Implications of New Energy Technologies, R. Rom and V. Archer eds. Ann Arbor, Mich.: Ann Arbor Science Publishers, Inc.
7. Axelson, O., and L. Sundell. 1978. Mining, lung cancer and smoking. Scan. J. Work Environ. Health 4:46–52.
8. Axelson, O., C. Edling, and H. Kling. 1979. Lung cancer and residency: A case-referent study on the possible impact of exposure to radon and its daughters in dwellings. Scand. J. Work Environ. Health 5:10–15.
9. Bean, J. A., P. Isacson, R. M. A. Hahne, and J. Kohler. 1982. Drinking water and cancer incidence in Iowa. Am. J. Epidemiol. 116(6):924–932.
10. Checkoway, J., R. M. Mathew, J. L. S. Hickey, C. M. Shy, R. L. Harris, E. W. Hunt, and G. T. Waldman. 1985. Mortality among workers in the Florida phosphate industry. I. Industry-wide cause-specific mortality patterns. J. Occup. Med. 27:885–892.
11. Damber, L., and L. G. Larsson. 1982. Combined effects of mining and smoking in the causation of lung carcinoma-case-control study in northern Sweden. Acta Rad. Oncol. 21:305–313.
12. Damber, L., and L. G. Larsson. 1985. Underground mining, smoking and lung cancer: A case-control study in the iron ore municipalities in northern Sweden. J. Natl. Cancer Inst. 74(6):1207–1213.
13. deVilliers, A. J., and J. P. Windish. 1964. Lung cancer in a fluorspan mining community. Br. J. Ind. Med. 21:94–109.
14. deVilliers, A. J., J. P. Windish, F. de N. Brent, B. Holloywood, C. Walsh, J. W. Fisher, and W. D. Parsons. 1971. Mortality experience of the community and of the fluorspan mining employees at St. Lawrence, Newfoundland. Occup. Health Rev. 22:1–15.

15. Edling, C. 1982. Lung cancer and smoking in a group of iron ore miners. Am. J. Ind. Med. 3:191–199.
15a. Edling, C., P. Combs, O. Axelson, and V. Flodin. 1982. Effects of low-dose radiation—a correlation study. Scand. J. Work Environ. Health 8(Suppl. 1):59–64.
16. Edling, C., and O. Axelson. 1983. Quantitative aspects of radon daughter exposure and lung cancer in underground miners. Br. J. Ind. Med. 40:182–187.
16a. Edling, C., H. Kling, and O. Axelson. 1984. Radon in houses—a possible cause of lung cancer. Scand. J. Work Environ. Health 10:25–34.
17. Fox, A. J., P. Goldblatt, and L. J. Kinlen. 1981. A study of the mortality of Cornish tin miners. Br. J. Ind. Med. 38:378.
18. Gottlieb, L. S., and L. A. Husen. 1982. Lung cancer among Navajo uranium miners. Chest 81:449–452.
19. Harting, F. H., and W. Hesse. 1979. Der lungenkrebs, die Bergkrankheit in den Schneeberger gruben. Vjschr. Gerichtl. Med. Offentl. Gesundheitswesen 31:102–132, 313–337.
20. Hess, C. T., C. V. Weiffenbach, and S. A. Norton. 1983. Environmental radon and cancer correlations in Maine. Health Phys. 45:339–348.
21. Holaday, D. A., W. D. David, and H. N. Doyle. 1952. An Interim Report of a Health Study of the Uranium Mines and Mills by the Federal Security Agency, Public Health Service, Division of Occupational Health, and the Colorado State Department of Public Health. Washington D.C.: U.S. Public Health Service.
22. Horaceck J., V. Placek, and J. Sevc. 1977. Histologic types of bronchogenic cancer in relation to different conditions of radiation exposure. Cancer 40:832–835.
23. Hornung, R. W., and T. J. Meinhardt. 1987. Quantitative risk assessment of lung cancer in U.S. uranium miners. Health Phys. 52:417–430.
24. Hornung, R. W., and S. Samuels. 1981. Survivorship models for lung cancer mortality in uranium miners—is cumulative dose an appropriate measure of exposure? Pp. 363–368 in International Conference, Radiation Hazards in Mining: Control, Measurement, and Medical Aspects, M. Gomez, ed. New York: Society of Mining Engineers of the American Institute of Mining, Metallurgical, and Petroleum Engineers, Inc.
25. Howe, G. R., R. C. Nair, H. B. Newcombe, A. B. Miller, S. E. Frost, and J. D. Abbatt. 1986. Lung cancer mortality (1950–1980) in relation to radon daughter exposure in a cohort of workers at the Eldorado Beaverlodge uranium mine. J. Natl. Cancer 77(2):357–362.
26. Hueper, W. C. 1966. Occupational and Environmental Cancers of the Respiratory System. New York: Springer-Verlag.
27. Jorgensen, H. S. 1973. A study of mortality from lung cancer among miners in Kiruna 1950–1970. Work Environ. Health 10:126–133.
28. Kunz, E., J. Sevc, and V. Placek. 1978. Lung cancer mortality in uranium miners. Health Phys. 35:579–580.
29. Kunz, E., J. Sevc, V. Placek, and J. Horacek. 1979. Lung cancer in man in relation to different time distributions of radiation exposure. Health Phys. 36:699.
30. Lorenz, E. 1944. Radioactivity and lung cancer; a critical review of lung cancer in the miners of Schneeberg and Joachimsthal. J. Natl. Cancer Inst. 5:1–13.
31. Ludwig, P., and E. Lorenser. 1924. Untersuchungen der Grubenluft in den Schneeberger gruben auf den Gehalt and Radiumemanation. Strahlentherapie 19:428–435.

32. Lundin, F. D., Jr., J. W. Lloyd, E. A. Smith, V. E. Archer, and D. A. Holaday. 1969. Mortality of uranium miners in relation to radiation exposure, hardrock mining and cigarette smoking—1950 through September 1967. Health Phys. 16:571–578.
33. Lundin, F. D., Jr., J. K. Wagoner, and V. E. Archer. 1971. Radon Daughter Exposure and Respiratory Cancer, Quantitative and Temporal Aspects. Joint Monograph No. 1. Washington, D.C.: U.S. Public Health Service.
34. Lundin, F. D., Jr., V. E. Archer, and J. K. Wagoner. 1979. An exposure-time-response model for lung cancer mortality in uranium miners—effects of radiation exposure, age and cigarette smoking. Pp. 243–264 in Proceedings of the Work Group at the Second Conference of the Society for Industrial and Applied Mathematics, N. E. Breslow and A. S. Whittemore, eds. Philadelphia: Society for Industrial and Applied Mathematics.
35. Morrison, H. I., D. T. Wigle, H. Stocker, and A. J. deVilliers. 1981. Lung cancer mortality and radiation exposure among the Newfoundland fluorspar miners. Pp. 372–376 in International Conference on Radiation Hazards in Mining: Control, Measurements and Medical Aspects, M. Gomez, ed. New York: Society of Mining Engineers of the American Institute of Mining, Metallurgical, and Petroleum Engineers, Inc.
36. Morrison, H. I., R. M. Semenciw, Y. Mao, D. A. Corkill, A. B. Dory, A. J. deVilliers, H. Stocker, and D. T. Wigle. 1985. Lung cancer mortality and radiation exposure among the Newfoundland fluorspar miners. Pp. 354–364 in Occupational Radiation Safety in Mining, H. Stocker, ed. Proceedings of the International Conference. Toronto: Canadian Nuclear Association.
37. Muller, J., W. C. Wheeler, J. F. Gentleman, G. Suranyi, and R. Kusiak. Pp. 359–362 in The Ontario Miners Mortality Study, General Outline and Progress Report: Radiation Hazards in Mining, M. Gomez, ed. New York: Society of Mining Engineers of the American Institute of Mining, Metallurgical, and Petroleum Engineers, Inc.
38. Muller, J., W. C. Wheeler, J. F. Gentleman, G. Suranyi, and R. A. Kusiak. 1983. Study of Mortality of Ontario Miners, 1955–1977, Part I. Toronto: Ontario Ministry of Labour, Ontario Workers Compensation Board.
39. Muller, J., W. C. Wheeler, J. F. Gentleman, G. Suranyi, and R. Kusiak. 1985. Study of mortality of Ontario miners. Pp. 335–343 in Occupational Radiation Safety in Mining, Proceedings of the International Conference, E. Stocker, ed. Toronto: Canadian Nuclear Association.
40. Muller, J., R. A. Kusiak, G. Suranyi, and A. C. Richie. 1986. Study of Mortality of Ontario Gold Miners, 1955–1957, Part II. Ottawa, Canada: Ontario Ministry of Labour.
41. Nair, R. C., J. D. Abbott, G. R. Howe, H. B. Newcombe, and S. E. Frost. 1985. Mortality experience among workers in the uranium industry. Pp. 154–164 in Occupational Radiation Safety in Mining, H. Stocker, ed. Proceedings of the International Conference. Toronto: Canadian Nuclear Association.
42. National Research Council, Committee on the Biological Effects of Ionizing Radiations (BEIR). 1980. The Effects on Populations of Exposure to Low Levels of Ionizing Radiation. Washington, D.C.: National Academy Press. 524 pp.
43. Nero, A. V. 1983. Airborne radionuclides and radiation in buildings: A review. Health Phys. 45:303–322.
44. Peller, S. 1939. Lung cancer among mine workers in Joachimsthal. Human Biol. 11:130–143.

45. Pershagen, L. D., and R. Falk. 1984. Exposure to radon in dwellings and lung cancer: A pilot study. Pp. 73–78 in Indoor Air, Vol. 2. Radon, Passive Smoking, Particulates and Housing Epidemiology, B. Berglung, T. Lindvall, and J. Sundell, eds. Stockholm: Swedish Council for Building Research.
46. Pirchan, A., and H. Sikl. 1980. Cancer of the lung in the miners of Jachymov (Joachimsthal). Am. J. Cancer 4:681–722, 1932.
47. Placek, V., A. Smid, J. Sevc, L. Tomasek, and P. Vernerova. 1983. Late effects at high and very low exposure levels of the radon daughters. In Proceedings of the 7th International Congress of Radiation Research. Amsterdam: Martinus Nijhoff Publishers.
48. Radford, E. P., and K. G. St. Clair Renard. 1984. Lung cancer in Swedish iron miners exposed to low doses of radon daughters. N. Engl. J. Med. 310(23):1485–1494.
49. Rostoski, O., E. Saupe, and G. Schmorl. 1926. Die bergkrankheit der Erzbergleute in Schneeberg in Sachsen ("Schneeberger Lungenkrebs"). Z. Krebforsch. 23:360–384.
50. Samet, J. M., C. R. Key, D. M. Kutvirt, and C. L. Wiggins. 1980. Respiratory disease mortality in New Mexico's American Indians and Hispanics. Am. J. Public Health 70:492–497.
51. Samet, J. M., D. M. Kutvirt, R. J. Waxweiler, and C. R. Key. 1984. Uranium mining and lung cancer in Navajo men. N. Engl. J. Med. 310:1481–1484.
52. Schiager, K. J., and L. W. Hersloff. 1984. Review of radon daughter exposure measurements in U.S. uranium mines—past and present. In Assessment of the Scientific Basis for Existing Federal Limitations on Radiation Exposure to Underground Uranium Miners, Appendix F. Toronto: SENES Consultants Ltd.
53. Seltser, R. 1965. Lung cancer and uranium mining: A critique. Arch. Environ. Health 10:923–935.
54. Sevc, J., and V. Placek. 1973. Radiation induced lung cancer: Relation between lung cancer and long-term exposure to radon daughters. Pp. 305–310 in Proceedings of the 6th Conference on Radiation Hygiene. Jasna pod Chopkom, CSSR.
55. Sevc, J., V. Placek, and J. Jerabek. 1971. Lung cancer risk in relation to long-term radiation exposure in uranium miners. Pp. 315–326 in Proceedings of the 4th Conference on Radiation Hygiene. Pareszka, Lhota, CSSR.
56. Sevc, J., E. Kunz, and V. Placek. 1976. Lung cancer mortality in uranium miners and long-term exposure to radon daughter products. Health Phys. 30:433–437.
57. Simpson, S. G., and G. W. Comstock. 1983. Lung cancer and housing characteristics. Arch. Environ. Health 38:248–251.
58. Smid, A., J. Sevc, V. Placek, and E. Kunz. 1983. Lung Cancer in Exposed Human Populations and Dose/Effect Relationship. Presented at the 7th International Congress on Radiation Research. Amsterdam: Martinus Nijhoff Publishers.
59. Snihs, J. O. 1974. The approach to radon problems in non-radium uranium miners in Sweden. Proceedings of the 3rd International Congress of the International Radiation Protection Association. CONF-730907. Oak Ridge, Tenn.: U.S. Atomic Energy Commission.
60. Solli, M., A. Andersen, E. Straden, and S Langand. 1985. Cancer incidence among workers exposed to radon and thoron daughters at a niobium mine. Scan. J. Work Environ. Health 11:7–13.
61. Stayner, L. T., T. Meinhardt, R. Lemen, D. Bayliss, R. Herrick, G. R. Reeve, A. B. Smith, and W. Halperin. 1985. A restrospective cohort mortality study of a phosphate fertilizer production facility. Arch. Environ. Health 40:133–138.

62. Sun, S., X. Yang, Y. Lan, M. Xionyu, L. Shengen, and Y. Zhanyun. 1984. Latent period and temporal aspects of lung cancer among miners. Radiat. Prot. 4(5) (English translation).
63. Teleky, L. 1937. Occupational cancer of the lung. J. Ind. Hyg. Toxicol. 2:73–85.
64. Tirmarche, M., J. Brenot, J. Piechowski, J. Chameaud, and J. Pradel. 1985. The present state of an epidemiological study of uranium miners in France. Pp. 344–349 in Proceedings of the International Conference, Occupational Radiation Safety in Mining, Vol. 1. Canadian Nuclear Association, Toronto, Ontario, Canada.
65. Wagoner, J. K., V. E. Archer, B. E. Carroll. 1964. Cancer mortality patterns among U.S. uranium miners and millers, 1950 through 1962. J. Natl. Cancer Inst. 32:787–801.
66. Wagoner, J. K., V. E. Archer, F. E. Lundin, D. A. Holaday, and J. W. Lloyd. 1965. Radiation as the cause of lung cancer among uranium miners. N. Engl. J. Med. 273:181–188.
67. Waxweiler, R. J., R. J. Roscoe, V. E. Archer, M. J. Thun, J. K. Wagoner, and F. E. Lundin, Jr. 1981. Mortality follow-up through 1977 of the white underground uranium miners cohort examined by the United States Public Health Service. Pp. 823–830 in International Conference, Radiation Hazards in Mining: Control, Measurement and Medical Aspects, M. Gomez, ed. New York: Society of Mining Engineers of the American Institute of Mining, Metallurgical, and Petroleum Engineers, Inc.
68. Wang, X., X. Huang, et al. 1984. Radon and miners lung cancer, Zhonghua Fangshe Yixue yu Fanghu Zazhi 4:10–14 (English translation).
69. Whittemore, A. S., and A. McMillan. 1983. Lung cancer mortality among U.S. uranium miners: A reappraisal. J. Natl. Cancer Inst. 71(3):489–499.

APPENDIX V

Nonmalignant Respiratory and Other Diseases Among Miners Exposed to Radon

Epidemiological evidence on radon progeny as a potential risk factor for nonmalignant respiratory diseases is restricted to uranium miners. As discussed in Appendix III, animal studies are consistent with an association between exposure to radon progeny and nonmalignant respiratory diseases. Animals so exposed develop emphysema and interstitial fibrosis. Pulmonary fibrosis and, to a lesser extent, emphysema are common findings in hamsters, rats, and dogs exposed to radon progeny alone and in mixtures with uranium-ore dust.[7–9,18] These effects are not produced to any appreciable extent in groups of animals until exposures to radon daughters exceed several thousand working-level months (WLM). Thus, the clinical diseases of interest are chronic obstructive pulmonary disease, in which airflow obstruction results from emphysema and airway changes, and interstitial processes such as pulmonary fibrosis.

The occurrence of nonmalignant respiratory diseases has been examined in miners from the Colorado Plateau region and from Ontario, Canada. Several reports from the U.S. Public Health Service study described excess mortality from nonmalignant respiratory diseases.[4,26] Between 1950 and 1977, a fivefold excess of death occurred from nonmalignant respiratory diseases, exclusive of tuberculosis, bronchitis, influenza, and pneumonia.[26] Causes of death were primarily emphysema, fibrosis, and silicosis. The effects of cigarette smoking were not considered.

In the Ontario miners, mortality from influenza, pneumonia, bronchitis, and asthma was not increased. However, mortality from silicosis and chronic interstitial pneumonia was significantly elevated (11 deaths observed, with 2.14 expected).

As part of the U.S. Public Health Service study of Colorado Plateau miners, physical examinations and lung function testing were performed in 1957 and 1960.[2] Data were collected for 2,349 white males, but only 277 participated in both years. Spirometry was carried out at both examinations, and the peak expiratory flow rate was measured in 1960. Three different measures of radiation exposure were used: (1) years of underground uranium mining, (2) a radiation index based on the working level at the time of examination, and (3) a cumulative, ordinal measure of exposure. The analyses were interpreted as showing loss of ventilatory function with increasing cumulative exposure. However, the data were neither collected nor analyzed with techniqnes that are currently accepted for lung function parameters. Further, the accuracy of the exposure measures that were used is uncertain.

In a later paper, Archer et al.[3] used the same U.S. Public Health Service study data and demonstrated an increasing prevalence of emphysema, as diagnosed by a physician's examination, with increasing WLM. However, the diagnosis of emphysema, a histopathologically identified disease, cannot be established by physical examination. Trapp et al.[23] performed more detailed studies on Colorado Plateau uranium miners and found evidence of pulmonary dysfunction, both restrictive and obstructive. The design of the investigation did not permit assessment of exposure-response relationships with lung function measures.

More recently, Samet et al.[20] surveyed 192 long-term New Mexico uranium miners. The survey procedures included spirometry, completion of a respiratory symptoms questionnaire, physical examination of the chest, and interpretation of chest x rays. Total duration of nnderground uranium mining, not WLM, was used as the exposure index. The design of the investigation did not permit assessment of the effects of each potentially hazardous agents as radon daughters, silica, and diesel exhaust. With linear multiple-regression analysis that controlled for cigarette smoking, duration of mining was associated with reduction of the forced expiratory volume in 1 s and reduction of the midmaximum expiratory flow rate, but not with reduction of the forced vital capacity. Chest x-ray abnormalities compatible with silicosis were found in 9% of the uranium miners examined for this survey.

These investigations have not separated the effects of radon-daughter exposure from those of other atmospheric contaminants, such as silica, diesel-engine exhaust, and blasting fumes, found in a uranium mine. Given the inadequacies of available exposure data, epidemiological methods cannot assess the individual contributions of all harmful agents to which uranium miners are exposed or are potentially exposed.

OTHER DISEASES AMONG MINERS EXPOSED TO RADON PROGENY

Mortality from selected causes among miners exposed to radon daughters is detailed in Appendix IV, Table IV-16. In addition to the excess risk from lung cancer, a number of the mining cohorts have experienced an excess risk of mortality due to tuberculosis and to "other nonmalignant respiratory diseases," (ICD Code 510-527, 7th revision). Because of the past levels of silica in the mine atmospheres, most of these excesses are believed to be due, in fact, to silicosis, which has been often diagnosed on death certificates as silicotuberculosis, tuberculosis, or other forms of nonmalignant respiratory disease.

Results from many studies of the mining populations have suggested that there is a slight excess risk of stomach cancer, which has an elevated incidence among other mining groups, including gold miners in Ontario without uranium mining experience[15] and coal miners in the United States.[11] Among the Ontario uranium miners, no excess risk existed for those without prior gold mining experience (9 observed versus 9.55 expected).[17] The cases are few, and the risks are low in most studies, but the occurrence of this excess risk in eight different mining populations lends credibility to the causation hypothesis that the excess has resulted from a common occupational risk factor. From the reported analyses, however, it is difficult to determine if that risk factor is radon progeny because exposure-response analyses have not been reported.

Two studies, both based on small numbers, have found nonsignificant excesses of skin cancer. While Sevcova et al.[21] found that basal cell carcinomas predominated, four of the five miners in the Colorado Plateau study who died of skin cancer had melanomas.[26] Since the other mortality studies have not shown any significant skin-cancer excess, it is unlikely that alpha radiation in the mines accounted for these excesses. However, the occurrence of skin cancers, particularly nonmelanomas, cannot readily be evaluated with a mortality study. Most skin cancers other than malignant melanoma can be readily cured and rarely lead to death. No other sites of malignancy appear to be consistently elevated among these mining populations. Despite the fact that airborne radon daughters deposit in the nasal passages, no cases of nasal cancer have been reported in any of the epidemiological studies. Only Muller et al.[17] reported an expected value of 0.8 for nasal cancer mortality among all Ontario uranium miners.

Excess risk of mortality due to nonmalignant renal disease was found in a recent analysis of the Colorado Plateau study.[26] In that survey, chronic and unspecified nephritis was particularly elevated after 10 yr latency (7 observed versus 1.9 expected; standardized mortality ratio, 362). The committee considered that this excess may be either a chance

finding or indicative of an occupational risk factor, possibly alpha radiation or uranium. The nephrotoxicity of soluble uranium in animals is well documented in the experimental literature,[24] but most of the uranium in mines occurs as less soluble oxides.

Findings from relevant animal experiments should also be considered in interpreting the epidemiological data. In radon-daughter-exposed animals, lesions observed in organs other than the lung are considered spontaneous, or only indirectly exposure-related, in contrast to the case for most alpha-emitters, which translocate from the lung to irradiate other organs. Because of the extremely short half-life of the radon daughters, their alpha emissions occur before they move to other organs. In animals exposed to high concentrations of uranium-ore dust alone (and presumably to radon daughters and uranium-ore dust mixtures) sufficient long-lived radioactivity from the precursors of radon can concentrate in the kidneys to impair their function. However, direct evidence of renal function impairment from exposure to radon daughters alone is lacking.

CYTOGENETIC STUDIES

The frequency of chromosome aberrations in blood cells has been examined in uranium miners and other underground workers as a marker of injury due to ionizing radiation. Brandom and colleagues[5,6] have reported on chromosome aberrations in uranium miners from the Colorado Plateau region of the United States. In their 1972 report,[5] cytogenetic abnormalities in peripheral lymphocytes from 15 miners were compared with the findings in 15 age-matched nonminer controls; 5 of the miners had lung cancer at the time of the study. Most of the aberrations were more prevalent in the miners, and many of the differences between the two groups attained statistical significance.

A subsequent report by Brandom et al.[6] included 80 underground uranium miners and 20 controls, frequency-matched for age and smoking habits. Again, the various types of chromosomal aberrations were more prevalent in the uranium miners than in the controls. Exposure-response relationships were evident up to a cumulative exposure of 3,000 WLM; however, chromosonal abnormalities were less frequent in those with greater than 3,000 WLM than in those with 1,740–2,890 WLM, the next lowest exposure category.

Badgastein is a spa in Austria with thermal springs that discharge water with a high concentration of radon. At Badgastein, patients are also treated in a former gold mine that has a mean radon concentration of 3,000 pCi/liter. Pohl-Ruling and Fischer[19] evaluated cytogenetic abnormalities in inhabitants of the community, bath attendants, and personnel exposed underground. The investigators estimated blood doses from alpha and

gamma radiation and used the dose estimates for assessing dose-response relationships. The analyses did not provide a clear estimate of the effect of alpha radiation, though they concluded that occupational alpha doses flattened the dose-response relationships for radiation.

Relevant data are also provided by a study of cytogenetic abnormalities in persons presumed to be exposed to high concentrations of radon in household water.[22] Chromosome aberrations were evaluated in 18 exposed persons and 9 controls. Dicentrics, chromosome breaks, and cells with chromosome change were significantly more frequent in the 18 exposed subjects. However, exposures to radon daughters were not estimated, and the suitability of the control group was not satisfactorily established.

To date, only the above-mentioned limited data are available on cytogenetic abnormalities in radon-daughter-exposed populations. The study of Colorado Plateau uranium miners indicates exposure-response relationships for chromosome aberrations. However, confirming evidence is not available from other populations, and the biological significance of these observations bas not been established.

EFFECTS ON REPRODUCTIVE OUTCOME

Recent and primarily descriptive data have renewed speculation that uranium mining is associated with adverse reproductive outcomes. Muller and colleagues[13,14,16] made the first reports on this subject in a series of papers on Czechoslovakian uranium miners that were published during the 1960s. For 1,000 underground male workers, the numbers of children in relationship to age did not deviate from that expected from nationwide data.[13] However, in tbis sample and in another with 415 uranium miners,[16] the secondary sex ratio (male to female births) declined following the start of underground employment from 1.08 to 0.85 in the former sample and from 1.18 to 0.99 in the latter.

Potential reproductive effects of uranium mining received little further evaluation until the early 1980s. At that time, descriptive data from New Mexico were interpreted as suggesting the adverse reproductive effects caused by uranium mining, by affecting either uranium miners or those living in the vicinity of mines and mills.[27]

This more recent interest in reproductive effects caused by uranium mining followed reports of high rates of congenital malformations and spontaneous abortion at the Shiprock Indian Health Service Hospital, located in San Juan County, New Mexico, which serves Navajos in the northeastern portion of the Navajo nation. Goodman subsequently examined the secondary sex ratio in New Mexico and Navajo births.[12] His analyses showed a temporal decline in the secondary sex ratio for New Mexico, in comparison with nationwide data, that occurred during the period of

extensive uranium mining in the state. The decline in secondary sex ratio was greatest for counties with mining activity; further, the Navajo Area Indian Health Service units with the lowest sex ratios also encompassed areas of mining. A preliminary study of Grants, New Mexico, area miners also suggested effects on the secondary sex ratio, and a study of 11 miners showed distribution of Y bodies in their semen different from that in control populations.[10]

Waxweiler and Roscoe[25] reviewed the results of a 1965 questionnaire survey of Colorado Plateau miners; overall, the secondary sex ratio did not vary with cumulative WLM. When the participants were stratified at the population's median age of 24, the secondary sex ratio was significantly increased in the highest exposure category. This observation could not be readily explained.[1,25]

Two studies were implemented to follow-up on these hypothesis-generating observations: a survey of reproductive outcomes in wives of Navajo uranium miners and a case-control study of births at the Shiprock Hospital. Wiese and Skipper[28] have recently reported preliminary findings of the survey of reproductive outcomes. Questionnaires were distributed to uranium miners in the Grants, New Mexico, area and to potash miners employed in the southeastern portion of New Mexico. The study population included 491 uranium and 226 potash miners. The investigators did not find significant differences between the two groups in the frequency of low-birth-weight infants, sex ratio, miscarriages, or infertility. Birth weights were lower in children born after the men began underground mining, but the effect was present only in those births after 1970, when average exposures to radon daughters were lower in the mines. Findings from the case-control study in Shiprock have not yet been reported. To date, the evidence on the possible reproductive effects of uranium mining is largely descriptive and preliminary. The studies of uranium miners do not show a consistent and readily interpretable pattern of effect. The data related to possible effects of the uranium mining industry on the general population are fragmentary at present.

REFERENCES

1. Archer, V. E. 1981. Discussion of secondary sex ratio of first-born offspring of U.S. uranium miners. In Birth Defects in the Four Corners Area, W. H. Wiese, ed. Transcript of a meeting. Albuquerque, N.M.: University of New Mexico School of Medicine.
2. Archer, V. E., H. P. Brinton, and J. K. Wagoner. 1964. Pulmonary function of uranium miners. Health Phys. 10:1183–1194.
3. Archer, V. E., J. K. Wagoner, and F. E. Lundin, Jr. 1973. Lung cancer among uranium miners in the United States. Health Phys. 25:351–371.
4. Archer, V. E., J. D. Gillam, and J. W. Wagoner. 1976. Respiratory disease mortality among uranium miners. Ann. N.Y. Acad. Sci. 271:280–293.

5. Brandom, W. F., G. Saccomanno, V. E. Archer, P. G. Archer, and M. E. Coors. 1972. Chromosome aberrations in uranium miners occupationally exposed to ^{222}radon. Radiat. Res. 52:204–215.
6. Brandom, W. F., G. Saccomanno, V. E. Archer, P. G. Archer, and D. Bloom. 1978. Chromosome aberrations as a biological dose-response indicator of radiation exposure in uranium miners. Radiat. Res. 76:159–171.
7. Chameaud, J., R. Perraud, J. Chretien, R. Masse, and J. Lafume. 1982. Lung carcinogenesis during in vivo cigarette smoking and radon daughter exposure in rats. Rec. Results Cancer Res. 82:11–20.
8. Cross, F. T., R. F. Palmer, R. H. Busch, R. E. Filipy, and B. O. Stuart. 1981. Development of lesions in Syrian golden hamsters following exposure to radon daughters and uranium ore dust. Health Phys. 41:135–53.
9. Cross, F. T., R. F. Palmer, R. E. Filipy, G. E. Dagle, and B. O. Stuart. 1982. Carcinogenic effects of radon daughters, uranium ore dust and cigarette smoke in beagle dogs. Health Phys. 42:33–52.
10. Dean, R. G. 1981. Semen analysis among uranium miners. In Birth Defects in the Four Corners Area, W. H. Wiese, ed. Transcript of a meeting. Albuquerque: University of New Mexico School of Medicine.
11. Enterline, P. E. 1972. A review of mortality data for American coal miners. Ann. N.Y. Acad. Sci. 200:260–272.
12. Goodman, A. B. 1986. Sex ratio patterns; a biological indicator of environmental factors. Pp. 14, 36 in Birth Defects in the Four Corners Area, W. H. Wiese, ed. Transcript of a meeting. Albuquerque: University of New Mexico School of Medicine.
13. Muller, C., M. Kubat, and J. Marsalek. 1962. Study on fertility of the miners in Jochimstal. Z. Gynekol. 2:63–68.
14. Muller, C., L. Relicha, and M. Kubat. 1962. On the question of genetic effects of ionizing rays on the miners of Jochimstal. Z. Gynekol. 15:556–560.
15. Muller, J., R. A. Kusiak, G. Suranyi, and A. C. Richie. 1986. Study of mortality of Ontario Gold Miners, 1955–1957. Part II. Toronto: Ontario Ministry of Labour.
16. Muller, C., L. Ruzicka, and J. Bakstein. 1967. The sex ratio in offspring or uranium miners. Acta Univ. Carolinal Med. 13:549–609.
17. Muller, J., W. C. Wheeler, J. F. Gentlemen, G. Suranyi, and R. A. Kusiak. 1985. Study of mortality of Ontario miners. Pp. 335–343 in Occupational Radiation Safety in Mining, Proceedings of the International Conference. E. Stocker, ed. Toronto: Canadian Nuclear Association.
18. National Council on Radiation Protection and Measurements (NCRP). 1984. Evaluation of Occupational and Environmental Exposures to Radon and Radon Daughters in the United States. Report 78. Washington, D.C.: National Council on Radiation Protection and Measurements.
19. Pohl-Ruling, J., and P. Fischer. 1980. An epidemiological study of chromosome aberrations in a radon spa. In Proceedings of the Specialist Meeting on the Assessment of Radon and Daughter Exposure and Related Biological Effects, G. F. Clemente, A. V. Nero, F. Steinhausler, and M. E. Wrenn, eds. Salt Lake City: R.D. Press.
20. Samet, J. M., R. A. Young, M. V. Morgan, C. G. Humble, G. R. Epler, and T. C. McLoud. 1984. Prevalence survey of respiratory abnormalities in New Mexico uranium miners. Health Phys. 46:361–370.
21. Sevcova, M., J. Sevc, and J. Thomas. 1978. Alpha irradiation of the skin and the possibility of late effects. Health Phys. 5:803–806.

22. Stenstrand, K., M. Annanmäki, and T. Rytömaa. 1979. Cytogenic investigation of people in Finland using household water with high natural radioactivity. Health Phys. 36:441–443.
23. Trapp, E., A. D. Renzetti, Jr., T. Kabayashi, M. M. Mitchell, and A. Bigler. 1970. Cardiopulmonary function in uranium miners. Am. Rev. Respir. Dis. 101:27–43.
24. Voegtlin, C., and H. C. Hodge, ed. 1949. Pharmacology and toxicology of uranium compounds. National Nuclear Energy Series. Division IV. New York: McGraw-Hill.
25. Waxweiler, R. J., and R. J. Roscoe. 1981. Secondary sex ratio of first-born offspring of U.S. uranium miners. In Birth Defects in the Four Corners Area, W. H. Wiese, ed. Transcript of a meeting. Albuquerque, N.M.: University of New Mexico School of Medicine.
26. Waxweiler, R. J., R. J. Roscoe, V. E. Archer, M. J. Thun, J. K. Wagoner, and F. E. Lundin, Jr. 1981. Mortality followup through 1977 of the white underground uranium miners cohort examined by the United States Public Health Service. Pp. 823–830 in Radiation Hazards in Mining: Control, Measurement, and Medical Aspects, M. Gomez, ed. New York: Society of Mining Engineers, American Institute of Mining, Metallurgical, and Petroleum Engineers, Inc.
27. Wiese, W. H., ed. 1981. Birth Defects in the Four Corners Area. Transcript of a meeting. Albuquerque, N.M.: University of New Mexico School of Medicine.
28. Wiese, W. H., and B. J. Skipper. 1986. Survey of reproductive outcomes in uranium and potash mine workers: Results of first analysis. Ann. Am. Conf. Gov. Ind. Hyg. 14:187–192.

APPENDIX VI
Lung-Cancer Histopathology

Correlations between radon-daughter exposures and specific histopathological cell types of lung cancer in humans have been a subject of controversy for many years. Primary cancer of the lung comprises diverse and generally distinct histopathological cell types. The most common are squamous-cell carcinoma, small-cell carcinoma, adenocarcinoma, and large-cell carcinoma, representing, respectively, about 35, 17, 25, and 9% of lung cancers in the male population of the United States.[16] In nonsmokers, adenocarcinoma is the most common cell type and small-cell carcinoma is infrequent, accounting for less than 5%.[24] Clinically, these four cell types of lung cancers differ in their manner of clinical presentation, natural history, and response to therapy. At present, lung cancers are generally classified histologically by conventional light microscopy. Figure VI-1 shows examples of some of the common histological types of lung cancers. Numerous classification schemes have been published, with the most widely used being that developed by the World Health Organization and recently modified.[25] The accuracy of histopathological diagnoses is influenced by the quantity and quality of the tissue available for examination. Observer variability in the classification of lung-cancer histopathology has been well documented and may be of substantial magnitude.[14] Reliance on clinical reports may introduce substantial misclassification. Because of observer variability, classification of lung-cancer cell types for research purposes should incorporate a standardized review of original histological material by a panel of pathologists. However, use of review panels does not ensure comparability between studies.

The distribution of lung-cancer cell types has been examined in uranium and other miners exposed to radon daughters. The principal populations are listed in Table VI-1. Most are from from mining groups in which cigarette smoking was prevalent. Exceptions include early miners in Schneeberg and Joachimsthal, reported to have smoked little, and Navajo miners in the United States.[3,15]

In the 1879 report by Harting and Hesse,[12] the malignant disease in Schneeberg miners was identified as lymphosarcoma, a designation that may have reflected the similarity of cells in small-cell cancers and in some lymphomas when examined with a light microscope. Autopsy specimens from miners in nearby Joachimsthal showed a preponderance of small-cell carcinomas.[17,23] A later investigation of uranium miners in this same area showed that small-cell cancer remained the most common histological type. Horacek et al.[13] compared the histological distribution of 115 cases of lung cancer in Czechoslovakian uranium miners with that in 326 control cases. Diagnoses were made by one pathologist who was not informed about the exposure status of the cases under review. Only a small percentage of the lung-cancer cases in each group were nonsmokers. The percentages of squamous-cell carcinomas were similar in the miners and in the controls, but about 12% more small-cell cancers were observed in miners. The mortality rate of all major cell types was increased beyond that which would have been expected.

In the late 1950s and early 1960s a number of reports from France, which included crude proportionate mortality studies, were published describing the lung-cancer excess among iron miners in Lorraine. Roussel et al.,[18] in a histological review of a series of 225 lung-cancer cases among these miners, found that 44% were anaplastic versus an expected value of 28% for nonminers. Both groups included approximately 1% nonsmokers.

The studies by Saccomanno and colleagues of the Colorado Plateau uranium miners provide the most extensive data concerning lung-cancer cell types and radon-daughter exposure. Findings have been reported periodically since 1964.[2,3,19–21] The case material was derived from miners in the U.S. Public Health Service study and others who lived in the Colorado Plateau area. Review methods varied; most reports were based on a panel's consensus, but some reviews apparently involved only one pathologist. Most of the miners smoked cigarettes, and the total series has included only 14 nonsmokers.[20] Initially, the majority of cases reviewed were small-cell carcinomas. The proportion of this cell type declined from 76% in 1964 to 22% in the late 1970s, while squamous cell carcinomas increased concomitantly. In nonsmokers, eight cases were small-cell carcinomas and the remaining six were of other cell types.

The strong predominance of squamous-cell carcinomas in Newfoundland fluorspar miners appears anomalous (Table VI-1).[26] However, the histological diagnoses were made by sputum cytology, which results in

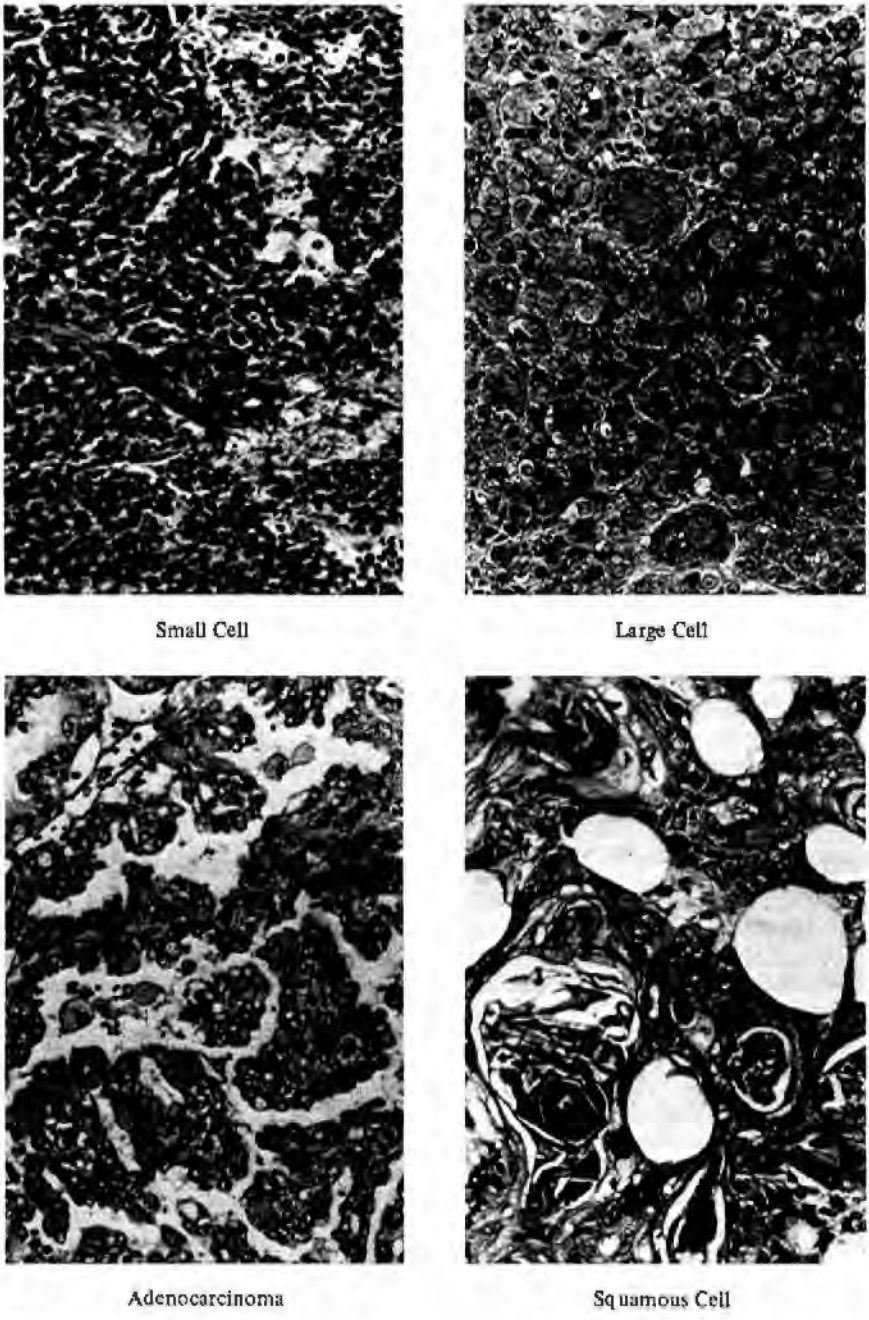


FIGURE VI-1 Histological types of lung cancer. SOURCE: Jonathan Samet, University of New Mexico, personal communication, 1987.

TABLE VI-1 Lung-Cancer Histopathology in Mining Groups Exposed to Radon Daughters

Population	Findings[a]	Comment
Joachimsthal miners[17,23]	28 cases: 16 SCC, 12 squamous	Autopsies 1929–1930 and 1933–1938; one pathologist
Colorado Plateau uranium miners[1,4,19,20,21]	SCC predominance, increasing with WLM	Cases seen at specific hospitals; panel review
Newfoundland fluorspar miners[26]	29 cases: 26 squamous, 2 SCC, 1 adenocarcinoma	All cases diagnosed by sputum cytology
British iron ore miners[5]	69 cases; 27 SCC	Histologic type from pathology report
Swedish iron ore miners[10]	36 cases: 26 SCC, 10 squamous	Histologic type from Swedish cancer registry
Swedish iron ore miners[9]	25 cases: 11 SCC, 11 squamous, 3 adenocarcinoma	Methods not given
Czechoslovakian uranium miners[13]	115 cases: 62 SCC, 40 squamous, 4 adenocarcinoma, 9 other types	One pathologist
Canadian uranium miners[8]	91 cases; 47 SCC	Selected from 134 cases; histologic type from pathology report
Navajo uranium miners[11]	16 cases: 10 SCC, 3 squamous, 3 other types	Methods not given; histologic type from pathology report
Navajo uranium miners[7]	21 cases: 7 SCC, 8 squamous, 4 adenocarcinoma, 2 LCC	Panel review
New Mexico uranium miners[6]	45 cases: 28 SCC, 15 squamous, 1 adenocarcinoma, 1 LCC	Panel review

[a]Abbreviations: SCC, small-cell carcinoma; LCC, large-cell carcinoma; Squamous, squamous cell carcinoma.

the over-representation of centrally located tumors. Both squamous- and small-cell carcinomas tend to be located in the larger airways, but at the time of this investigation the former may have been more readily diagnosed by cytology alone.

Navajo miners who worked in the Colorado Plateau are of interest because only a small proportion smoked cigarettes, with average consumption by the smokers of only a few cigarettes each day.[4,22] Gottlieb and Husen[11] described 16 Navajo miners diagnosed with lung cancer at the Shiprock Indian Health Service Hospital from 1965 through 1979. Based on record review, they reported that 10 of the cases were small-cell carcinomas. Butler et al.[7] reviewed histopathological material for 26 of 32 lung-cancer cases diagnosed among all Navajo males between 1969 and 1982. A panel of three pathologists examined all slides. In contrast with the earlier study of Gottlieb and Husen,[11] small-cell carcinomas did not predominate in the 21 cases of lung cancer occurring in Navajo uranium miners. Seven of these cancers were small-cell carcinoma, eight were squamous-cell carcinoma, four were adenocarcinoma, and two were large-cell carcinoma. While small-cell carcinoma was not the predominant cell type in this series, the proportion with this cell type (33%) is far greater than expected from the distribution of lung-cancer histopathology in nonsmokers. The discrepant findings of these two reports may reflect the use of medical records by Gottlieb and Husen[11] to determine the diagnoses.

Recent reports from Canada and New Mexico document a continued excess incidence of small-cell carcinomas in contemporary uranium miners.[6,8] In iron-ore miners in Great Britain and Sweden, also exposed to radon daughters, small-cell carcinoma has occurred in excess.[5,9,10] The pattern has been consistently observed in populations of miners who smoked cigarettes. Data for nonsmokers are sparse and conflicting. Saccomanno et al. [21] reported that most cases of lung cancer in nonsmokers from the Colorado Plateau region were small-cell carcinomas. Butler et al.[7] found a cell type distribution in Navajo miners comparable to the observed distribution in the general population. Thus, available information does not strongly support the association between uranium mining and small-cell cancers in nonsmokers, although this association in smokers is supported by the available data. This pattern appears to change as miners who smoke age and the interval since cessation of uranium mining exposure lengthens.

REFERENCES

1. Archer, V. E., H. P. Brinton, and J. K. Wagoner. 1964. Pulmonary function of uranium miners. Health Phys. 10:1183–1194.
2. Archer, V. E., J. K. Wagoner, and F. E. Lundin, Jr. 1973. Uranium mining and cigarette smoking effects on man. J. Occup. Med. 15:204–211.
3. Archer, V. E., G. Saccomanno, and J. H. Jones. 1974. Frequency of different histologic types of bronchogenic carcinoma as related to radiation exposure. Cancer 34:2056–2060.

4. Archer, V. E., J. D. Gillam, and J. W. Wagoner. 1976. Respiratory disease mortality among uranium miners. Ann. N.Y. Acad. Sci. 271:280–293.
5. Boyd, J. T., R. Doll, J. S. Faulds, and J. Leiper. 1970. Cancer of the lung in iron ore (haematite) miners. Br. J. Ind. Med. 27:97–105.
6. Butler, C., J. M. Samet, D. M. Kutvirt, C. R. Key, and W. C. Black. 1985. Cigarette smoking and lung cancer cell types in uranium miners. Am. Rev. Respir. Dis. 131(Pt. 2):A176 (abstract).
7. Butler, C., J. M. Samet, W. C. Black, C. R. Key, and D. M. Kutvirt. 1986. Histopathologic findings of lung cancer in Navajo men: Relations to uranium mining. Health Phys. 51:365–368.
8. Chovil, A., and B. Chir. 1981. The epidemiology of primary lung cancer in uranium miners in Ontario. J. Occup. Med. 23:417–421.
9. Damber, L., and L. G. Larsson. 1982. Combined effects of mining and smoking in the causation of lung carcinoma-case-control study in northern Sweden. Acta Rad. Oncol. 21:305–313.
10. Edling, C. 1982. Lung cancer and smoking in a group of iron ore miners. Am. J. Ind. Med. 3:191–199.
11. Gottlieb, L. S., and L. A. Husen. 1982. Lung cancer among Navajo uranium miners. Chest 81:449–452.
12. Harting, F. H., and W. Hesse. 1879. Der Lungenkrebs, die Bergkrankheit in den Schneeberger Gruben. Vierteljahrsschr Gerichtl. Med. Offentl. Gesundheitswesen 30:296–309; 31:102–129, 313–337.
13. Horacek, J., V. Placek, and J. Seve. 1977. Histologic types of bronchogenic cancer in relation to different conditions of radiation exposure. Cancer 40:832–835.
14. Ives, J. C., P. A. Buffler, and S. D. Greenberg. 1983. Environmental associations and histopathologic patterns of carcinoma of the lung: The challenge and dilemma in epidemiologic studies. Am. Rev. Respir. Dis. 128:195–209.
15. Myers, D. K., C. G. Stewart, and J. R. Johnson. 1981. Review of epidemiological studies on hazards of radon daughters. Pp. 513–524 in Radiation Hazards in Mining: Control, Measurement, and Medical Aspects, M. Gomez, ed. New York: Society of Mining Engineers, American Institute of Mining, Metallurgical, and Petroleum Engineers, Inc.
16. Percy, C., J. W. Horm, and T. E. Goffman. 1983. Trends in histologic types of lung cancer, SEER, 1973–1981. Pp. 153–159 in Lung Cancer: Causes and Prevention, M. Mizell, and P. Correa, eds. Deerfield Beach, Fla.: Verlag Chemie International.
17. Pirchan, A., and H. Sikl. 1932. Cancer of the lung in miners of Jachymov (Joachimsthal). Report of cases observed in 1929–1930. Am. J. Cancer 15:681–722.
18. Roussel, J., C. Pernot, P. Schoumacher, M. Pernot, and Y. Kessler. 1964. Considerations statistiques sur la cancer bronchique du mineur de fer du bassin de la Lorraine. J. Radiol. Electrol. 45:541–546.
19. Saccomanno, G. 1982. The contribution of uranium miners to lung cancer histogenesis. Rec. Results Cancer Res. 82:43–52.
20. Saccomanno, G., V. E. Archer, R. P. Saunders, L. A. James, and P. A. Beckler. 1964. Lung cancer of uranium miners on the Colorado Plateau. Health Phys. 10:1195–1201.
21. Saccomanno, G., V. E. Archer, O. Auerbach, M. Kuschner, R. P. Saunders, and M. G. Klein. 1971. Histologic types of lung cancer among uranium miners. Cancer 27:515–523.

22. Samet, J. M., D. M. Kutvirt, R. J. Waxweiler, and C. R. Key. 1984. Uranium mining and lung cancer in Navajo men. N. Engl. J. Med. 310: 1481–1484.
23. Sikl, H. 1950. The present status of knowledge about the Jachymov disease (cancer of the lungs in the miners of the radium mines). Acta Unio Intl. Contra Cancrum 6:1366–1375.
24. Stayner, L. T., and D. H. Wegman. 1983. Smoking, occupation and histopathology of lung cancer: A case-control study with the use of the Third National Cancer Survey. J. Natl. Cancer Inst. 70:421.
25. World Health Organization. 1982. The World Health Organization Histological Typing of Lung Tumors, 2nd ed. Am. J. Clin. Pathol. 77:123–136.
26. Wright, E. S., and C. M. Couves. 1977. Radiation-induced carcinoma of the lung—the St. Lawrence tragedy. J. Thorac. Cardiovasc. Surg. 74:495–498.

APPENDIX VII

The Combined Effects of Radon Daughters and Cigarette Smoking

Part 1 of this appendix reviews the epidemiological literature on the combined health effects of smoking and radiation. The studies reviewed by the committee are summarized in Table VII-1. Part 2 presents the committee's analyses of lung-cancer occurrence in persons exposed to both carcinogens. Part 3 summarizes the committee's views, including the possible effects of smoking on the validity of dose estimates and the need for further studies of the combined effects of radon daughters and smoking.

PART 1. Epidemiological Studies of Smoking and Radiation

STUDIES AMONG SWEDISH METAL MINERS

Studies of iron-ore miners in Northern Sweden reveal an excess of lung cancer that is related primarily to underground employment and exposure to radon.[17,28] In order to clarify the role of radon exposure combined with tobacco use on the occurrence of lung cancer, Damber and Larsson[10] carried out a case-control study in a three-county area of Northern Sweden, in which cases were ascertained in the years 1972–1977. As all iron mines were found within only two municipalities, succeeding studies were focused on the mining areas Kiruna and Gallivare and were extended to encompass the years 1972–1982.[11] Therefore, only the latest report is considered here.

The case group consisted of 69 lung-cancer cases who were reported to the Swedish cancer registry after 1972 and who were deceased before

July 1982. For each case, one deceased control was drawn from the National Registry for Cause of Death, matched by sex, year of death, age, and municipality; suicides and lung carcinomas were not included in the control group. A second living control for 60 cases aged 80 and under was selected from the Swedish National Population Registry and matched by sex, year of birth, and municipality.

As recognized by the authors, smoking-related risks that are based on deceased controls may be underestimated, since tobacco use was likely to have been greater by the deceased than by the general population. However, use of controls required to be alive until 1982 may overestimate relative risks since their smoking rates may be may have been *less* than those of the general population at risk.[21] Nevertheless, the results presented by Damber and Larrson[10] were generally comparable, regardless of control group.

Interviews were conducted with the index subject or the next of kin to obtain information on smoking practices and work history. Members of the study group who worked underground in an iron mine were considered exposed to radon. Since no accurate measurements of direct radon exposure were available, the surrogate variable, years underground, was used for analysis.

Smoking data consisted of the year tobacco use started the number of cigarettes smoked per day, and the year of cessation of smoking. Smokers were individuals who consumed one cigarette daily for at least 1 yr. For cigar and pipe smokers, 1 g of tobacco was equated to one cigarette.

Results were tabulated by three categories of lifetime tobacco use: nonsmoker, low (<150,000 cigarettes); and heavy (>150,000 cigarettes) consumption. For cases and deceased controls, relative risks rose from the baseline 1.0 for nonsmokers to 2.4 to 8.4 for aboveground workers and from 5.4 to 21.7 to 69.7 among underground miners. Similar results were reported for cases and the combined control group of all living and deceased subjects. Although based on small numbers (23 cases had no or low tobacco consumption, of which only 3 had no underground-mining experience), the results suggest that radon exposure and smoking combine multiplicatively rather than additively on a relative risk scale.

As outlined in Appendix IV, Radford and Renard[27] reported a historical cohort study of 1,415 miners from the Malmberget and Koskoskulle areas of Sweden. Data on current smoking habits were reported from 388 questionnaires administered in 1972–1973 to active miners and surface workers (35% of the contemporary work force) and from 168 pensioners. Pipe smoking was considered equivalent to cigarette smoking. Although pipe smoking has been related to lung-cancer risk, the affect of this assumption is difficult to assess since information on the percentage of pipe smokers and their inhalation patterns was not provided. The authors state

TABLE VII-1 Relative Risks from Selected Studies of Cigarette Use, Radiation Exposure, and Lung-Cancer Risk

Study Area	Design	Results	Comments
Kiruna and Gallivare, Sweden[11]	Cases (69) from death register 1972–1982; two types of controls: alive from general population (60) and deceased from register (67)	Cigarette Use[a] Underground miner 0 / < 150 / > 150 No 1 / 2.4 / 8.4 Yes 5.4 / 21.7 / 69.7	Smoking data from interviews of subjects or next-of-kin; results consistent with multiplicative, relative risk (RR) model, although formal testing not presented
Malmberget, Sweden[27]	Cohort study of 1,415 miners, with 50 cases of lung cancer	Nonsmoker / Smoker Nonminer 1 / 1 Miner 10.0 / 2.9	Results suggestive of submultiplicative model for RR, possibly additive; calculation of RRs not precisely described; formal model fitting not presented
Hammar, Sweden[2]	Cases (29) listed in death register 1957–1976; controls (174) also from register, matched on year of death	RR for mining 16.6 (90% confidence interval, 7.8–35.3); RR for smoking among miners 0.5 (90% confidence interval, 0.1–2.2)	Suggestive of a protective effect of smoking among miners; results subject to biases (see text)
Colorado[1]	Cohort study of uranium miners examined through 1960; followup from 1964–1967 with 39 cases of lung cancer	Lung Cancer Rate × 10^5 / Cigarette Use: No / Yes Miners 7.1 / 42.2 Expected[b] 1.1 / 4.2	Multiplicative combination is suggested; analysis of cases shows shorter latency period for smokers

Colorado[38]	Nested case control study from 3,362 miners followed from 1964–1977, with 194 cases and 776 controls; exposures lagged 10 years		Cigarette Use (pack yr)			Analyses formally reject additive RR model; data consistent with multiplicative model
		WLM	0–10	10–20	20–30	30+
		0–21	1	9.1	4.2	7.7
		22–119	1.1	13.6	6.5	19.0
		120–359	3.6	16.0	8.8	23.1
		360–839	7.8	5.2	16.2	46.8
		840–1,799	5.2	17.6	27.4	42.7
		1,800+	18.2	137.6	52.6	146.8
Colorado (Appendix VII, Part 2, this volume)	Cohort study of 3,362 miners followed through 1982, with 256 observed cases of lung cancer, exposures lagged 5 years		Cigarette Use (no./day)			Data fit well with multiplicative model ($P = 0.53$), while additive was rejected ($P = 0.03$); although not statistically superior to multiplicative model, best fitting power model was submultiplicative
		WLM	0–4	5–19	20–30	30+
		0–59	1[c]	2.7	7.8	2.9
		60–119	0.0	0.0	5.6	26.6
		120–239	2.4	9.1	15.3	9.8
		240–479	8.4	3.5	14.6	25.8
		480–959	17.8	12.6	32.0	34.0
		960+	27.6	36.0	63.6	90.3
Grand Junction Colorado[32]	Cases (489) and controls (992) drawn from cohort of 9,817 miners followed from 1960–1980, from whom sputum specimens were regularly obtained; cases defined as moderate or worst cell atypia		Cigarette Use (pack yr)			Study is of cell atypia; suggests multiplicative effects, although statistical testing not presented
		Yr Underground	0	1–20	21+	
		0	1	0.3	2.9	
		1–10	7.3	4.1	18.2	
		11+	9.6	9.8	26.0	

TABLE VII-1 (*Continued*)

Study Area	Design	Results	Comments
New Mexico (Appendix VII, Part 2, this volume)	Cases (52) and controls (222) extracted from cohort of uranium miners	Cigarette Use (no./day) Yr Mining Underground: <5 / 5–14 / 15–24 / >25 <10: 1 / 5.1 / 7.0 / 8.2 10–14: 1.0 / 12.0 / 6.7 / 6.2 15–19: 3.7 / 4.2 / 17.5 / 0.0 20+: 0.0 / 39.9 / 24.0 / 30.1	Both multiplicative and additive RR models consistent with data, although former exhibits better fit
Uranium City, Saskatchewan, Canada[4]	Followup for 3 yr of underground miners and controls who participated in lung cancer screening program; cases defined as moderate or worst cell atypia	Cigarette Use WLM: No / Yes 0: 1 / 2.7 <120: 2.6 / 3.7 ≥120: 1.2 / 12.6	Study is of cell atypia; few events among nonsmokers; data and analysis insufficient to assess interaction of exposures
Oeland, Sweden[14]	Cases (22) and controls (178) drawn from death registry 1960–1978; smoking habits obtained from next-of-kin using mail questionnaire	Cigarette Use Housing Type[d]: No / Yes 0: 1 / 2.7 1: 1.3 / 3.6 2: 4.4 / 9.3	Data were sparse, and no formal models were fit, but RRs suggest multiplicative interaction, or at least greater than additive
Japan[26]	Cohort study of 40,498 A-bomb survivors for whom smoking data are available; there were 281 lung-cancer deaths	Cigarette Use Radiation Exposure (rad): No / Yes <10: 1 / 2.4 10–99: 1.1 / 2.4 >100: 2.3 / 3.6	Both multiplicative and additive RR models fit data equally well

Japan (Appendix VII, Part 2, this volume)	Cases (485) and controls (1,089) identified during 1971–1980 from Life Span Study among A-bomb survivors		Cigarette Use (no./day)			Both multiplicative and additive RR models fit data equally well
		Radiation Exposure (rads)	0	1–10	11–20	>20
				Males		
		<10	1	3.7	6.9	26.5
		10–99	1.3	2.4	6.6	13.2
		>100	3.3	7.2	10.6	24.8
			Cigarette Use			
			0	1–10	>10	
				Females		
		<10	1	2.3	4.2	
		10–99	0.7	2.5	2.1	
		>100	5.2	5.2	—	

[a]Lifetime number in thousands.
[b]Incidence based on rates in mountain states.
[c]Baseline category based on 0.7 expected cases compared to 0 observed.
[d]See text for category definitions.

that approximately half the workers who were still living at the time of the study took part in the survey of smoking habits. Smoking histories for lung cancer patients were obtained from next of kin or, in a few instances, from the subject. Evaluation of the quality of tobacco consumption data is not possible, since no attempt was made to compare subjects from whom smoking data were obtained to those for whom data were unavailable. The smoking rate among miners is probably underestimated, since surveys covered only living workers.

The precise method of analysis of smoking is not completely clear in the published report. Among miners, smokers were defined as those who had stopped smoking within 10 yr of the interview or who were currently smoking, while nonsmokers were defined as subjects who stopped smoking 10 yr or more years to the interview or who had never smoked. The authors assumed that risk of lung cancer for smokers relative to that for nonsmokers is constant over age. The smoking status of miners was then compared with a national smoking survey of 25,000 men carried out in 1963.[30] It was determined that the miners had a higher proportion of smokers. Although apparently no adjustment was made for the different time periods of the two surveys, the mortality experience of the national survey[8] was applied to the miners and a relative risk of 7.4 for smokers versus nonsmokers was obtained. The method for deriving the relative risk of 7.4 was not explicitly described. The subsequent relative risks for miners to nonminers were 2.9 for smokers based on 32 lung-cancer cases and 10.0 for nonsmokers based on 18 cases. The authors concluded that mining- and smoking-related risks combine additively. This conclusion seems to go beyond the evidence as presented. Radford and Renard's[27] results, however, do tend to suggest that risks for the two exposures are submultiplicative.

Within the parish of Hammar, Sweden (population, 4,000), Axelson and Sundell[3] compared smoking and mining (zinc and lead) experiences in 29 lung-cancer cases deceased between 1956–1976 with 174 referents who died of causes other than lung cancer and who were matched to cases by time of death. A subject was exposed if he appeared on employee files of the mining company. For workers with mining experience (21 cases and 19 controls), foremen who were contemporaries of the subjects were contacted and queried about the smoking status of the subjects. Smoking status was not determined for nonminers.

Among miners, smoking appeared to be protective for lung cancer, although the 90% confidence interval was large (relative risk, 0.49; 90% confidence interval, 0.1–2.2). The authors explained this finding by suggesting that smokers have a lower radiation-induced risk because of a thickened mucus layer in critical bronchial segments.

Axelson and Sundell[3] did not evaluate the effects of smoking among nonminers or of mining exposure by smoking category. Because of this

lack of information on smoking in nonminers and on duration of radon exposure in miners, the study could not address the mode of interaction between radon and smoke exposures. Nevertheless, as noted in Table VII-1, the protective effect of smoking does suggest that an interaction could be additive or subadditive. However, potentially biased exposure assessment procedures (for example, inadequate company files and recall bias by the foremen), inappropriate control selection (inclusion of referents with tobacco-related causes of death), or simply the possibility that nonsmokers spent more time underground than smokers are alternative explanations.

STUDIES AMONG COLORADO PLATEAU MINERS

Several published reports based on the U.S. Public Health Service cohort of uranium miners of the Colorado Plateau have evaluated in detail the roles of radiation and cigarette smoking in the production of lung cancers.[1,22,24,32,38]

The earliest report, by Archer et al.,[1] included 39 cases of lung cancer that arose in a well-defined, physically examined special study group during a 4-yr observation period (1964–1967). Compared to lung-cancer rates among white male residents of mountain states, 1.1 and 4.4/10,000 person-yr for nonsmokers and smokers, respectively, the rates among uranium miners were 7.1 and 42.2/10,000, respectively. These comparisons, which show a 4-fold population-based excess for smoking and a 5.9-fold miner excess, suggest a multiplicative interaction of these agents.

Another analysis by Archer et al.,[1] reported in the same paper, focused on a larger sample of 207 cases, whose ascertainment of health status and population were less clearly defined but which included the 39 special study group cases. This second analysis relied solely on comparisons of age at lung-cancer diagnosis between groups of smoking and nonsmoking miners. Mine-related variables, such as age at start of mining, cumulative working-level months (WLM), and years of other hard-rock mining, were controlled through matching. The induction-latent period was shorter for smokers than for nonsmokers. The authors argue that the agents act synergistically. This analysis is questionable, however, regarding the form of the model, since in a survival model introduction of a second disease-related exposure, that is, radon, increases age-specific hazard rates and thus increases the probability of a tumor appearing earlier.

Lundin et al.[22] evaluated 62 lung-cancer cases that developed in the cohort of 3,366 white uranium miners followed from first medical examination through 1968. Smoking data came from periodic surveys carried out prior to 1970. Lundin et al. used tobacco consumption information from the last examination. Analysis was based on a log-normal model for estimating a yearly effective radiation dose, which weighted exposure in

each previous year, in order to account for disease latency. Although no formal statistical testing of hypotheses was carried out, the results suggest that the relative-risk model for WLM exposure, in comparison with the absolute-risk model, is more appropriate. The authors then analyzed the effect of smoking under their assumed effective-dose model and claimed a submultiplicative effect for smoking. The results suggest there is a greater amount of radiation-induced lung-cancer risk among smokers, with slight differences between heavy and light smokers, than in nonsmokers. It is difficult to evaluate conclusions from this analysis because of the lack of formal hypothesis testing.

A detailed study of the Colorado Plateau uranium miners in which 194 lung-cancer cases were used was carried out by Whittemore and McMillan.[38] A nested or synthetic case-control approach was used,[20,21] whereby each lung-cancer case was matched with four controls born within 8 months of the case and alive at the time the case died. Exposure histories for controls were adjusted to reflect values up to the time the case died (minus any lag time). With this type of analysis, relative hazard (or relative risk) is modeled in either a multiplicative or additive way. No direct information on disease rates are obtained, and hence, evaluation of absolute excess risk is not possible. Data were classified by four categories of cigarette pack-years (average cigarette packs smoked per day times duration, in years, of use), accumulated from the start of exposure to a predefined cutoff date, and six categories of WLM. A single 23-parameter relative-risk model was fit to the two-way classification. All subsequent models were then compared for goodness of fit to this saturated model. Several models for the relative risk with combined cigarette and WLM exposure were fit. Multiplicative and additive excess-risk models were fit, as well as other richer variants, for example, mixture models of additive and multiplicative terms for smoking and linear and quadratic terms for radiation; exponential models were also fit. Whittemore and McMillan[38] found substantial support for the multiplicative model, finding that it fit nearly as well as the saturated one. The authors rejected the additive model, which agrees with a preliminary analysis reported by Hornung and Samuels.[15] Further analyses found little improvement when smoking rate was added to the model, although this improvement might have been expected since pack-years incorporate cigarettes smoked per day and the subjects were matched by age. They also reported that the smoking effect did not interact with age.

The joint effect of smoking and radon-daughter exposure in this cohort was also addressed in a National Institute for Occupational Safety and Health (NIOSH) Report to the Mine Safety and Health Administration.[24] Using the Colorado Plateau miner cohort with follow-up through 1982, a synergistic effect between these two factors was reported, that is, combined

effects exceeding the sum of the separate effects (as would be predicted by an additive model). However, the data also snggested that the combined effect was less than multiplicative. It is generally difficult to compare these conclusions with other analyses of these data, since the authors relied on a power function relative risk model. Whittemore and McMillan[38] found that linear-relative-risk models for both smoking and radon exposure, individually, were preferrable to power-function relative-risk models.

An analysis of data from another group of Colorado Plateau workers has recently been reported by Saccomanno et al.[32] The cohort included 9,817 miners, underground and open pit, and millers who worked between 1960 and 1980 and who agreed to participate in the study.[31,32] Sputum samples were collected periodically, although irregularly, from 1957. Information on the number of workers lost to follow-up and on the completeness of sputum assessment was not reported. Although not explicitly stated, exposure measurements for radon and cigarette use were likely determined from periodic cohort surveys, as described previously for the other Colorado miner group.

Analyses were based on a selected case-control subsample from cohort members who had at least one sputum specimen taken between 1960–1980 and who had a current exposure history. Cases ($n = 489$) were defined as men who had at least one sputum cytology specimen classified as moderate or worse atypical squamous cell metaplasia. Controls ($n = 992$) were a 11% random sample of the noncase members of the cohort. Variables of interest were age, cumulative WLM, and pack-years. Because of case definition, this is a study of the determinants of moderate cell atypia or worse, and not of lung cancer.

The results suggested a multiplicative association for the combined effects of cigarette use and radon exposure, although formal testing procedures were not described. Based on unmatched analyses, increased age-adjusted relative risks with duration of underground uranium mining were similar within categories of pack-years, as were risks with cigarette consumption for categories of underground duration. The former increases from 1.0 for no underground experience to approximately 10.0 for more than 10 yr of underground experience, while the latter increases from 1.0 to approximately 3.0 for more than 20 pack-years of cigarette use. The authors also present results for logistic model fitting. Their interpretations are not clear, and may be statistically inappropriate.

This study is also subject to several potential biasing factors. Controls were substantially younger than cases (41.8 versus 58.2 yr, respectively) and were more likely to have been lost to follow-up (39% versus 23% respectively) or to have missing WLM data (33% versus 25%, respectively). Although one analysis was matched on age (±2 yr), the primary analysis was unmatched and relied on age adjustment, with either crude age

categories or a single age parameter in a logistic model. The adequacy of this adjustment is hard to assess. Bias is also possible from the method of control selection; controls were selected from all noncase members of the cohort, regardless of length of follow-up, instead of from cohort members at risk at the time of case ascertainment.[21] Controls were therefore likely to be healthier and to have received less exposure to radon and tobacco. Case selection bias, that is, more intense disease evaluation of higher-exposure workers, could have occurred, since workers who were more highly exposed to radon or cigarettes may have been more health conscious and therefore more likely to submit sputum specimens and ultimately categorized as a case. The authors did not give the mean number of specimens evaluated prior to ascertainment for cases or at equivalent follow-up for controls. Sputum specimens were obtained during follow-up and were used to define cases. However, men who were hospitalized or died with suspected lung cancer were apparently also classified as cases, although their atypia status should have been based on evaluated cytology records. Again, this deviation from the case definition criteria may have biased results of this study.

Using data from the 1982 follow-up of the Colorado Plateau cohort initiated by the U.S. PHS, this committee extended the analysis of radon daughters and cigarette use, which was carried out by Whittemore and McMillan.[38] The results of our analysis of 256 lung-cancer deaths are summarized in Table VII-1 and presented in detail in Part 2 of this appendix. They support Whittemore and McMillan's conclusions with some qualifications. The multiplicative relative-risk model fit the data quite well ($P = 0.48$), while the purely additive excess-relative-risk model was rejected ($P = 0.005$). To help clarify these results we studied a larger class of models, which were defined through a mixture of competing models,[34] in which both the additive and multiplicative models were nested. This investigation showed that the best-fitting model was submultiplicative, although it did not provide a statistically significant improvement in fit over the multiplicative model. The fitting of a sequence of models suggested that the data are consistent with a wide range of submultiplicative to supramultiplicative models, and there is no clear a prior reason to accept the multiplicative model, except parsimony.

STUDIES AMONG NEW MEXICO URANIUM MINERS

In the second part of this appendix, an evaluation is presented of the associations of cigarette smoking and duration of underground employment in a uranium mine with lung cancer in case-control data extracted from a cohort of New Mexico uranium miners. The results (Table VII-1) suggest that a multiplicative combination of the two exposures is more

compatible with the observed patterns of relative risks than an additive model, although there was not a statistically significant difference between the fit of the additive and relative-risk models.

STUDIES AMONG CANADIAN URANIUM MINERS

Band et al.[4] reported on a study initiated in 1974 of the combined effects of cigarette smoking and radon exposure among a group of miners and nonminers who were residents of Uranium City, a town in northern Saskatchewan, Canada. The miners were employed at the Eldorado Nuclear Mine. All residents were invited to participate in a lung-cancer screening program. Responses were obtained from 80% of the uranium workers (all males) and 50% of the total adult population. The study group consisted of 249 underground miners and 123 male controls. In a manner similar to that of Saccommano et al.,[32] outcome status was determined by degree of cellular atypia, as evaluated from two or three yearly sputum cytology samples. No information was given on whether the evaluations were carried out blindly or if more than one abnormal cytology (moderate or more severe atypia) during the 3-yr study period was required for an individual to be designated as a case.

Information on smoking and an occupational history was obtained by questionnaire. Based on work history, cumulative WLM exposure to radon through 1977 was determined for each underground miner. A nonsmoker was defined as one who never smoked cigarettes.

The results (Table VII-1) show an increasing risk with radon exposure and with cigarette use. However, there were too few nonsmokers with moderate or more severe atypia (three cases) to assess the interaction of these factors. The authors did not provide data on amount smoked by cumulative WLM exposure, but did show results from four separate (miners/nonminers, smokers/nonsmokers) logistic regressions, which included years of smoking and years underground as continuous variables. Although the parameter estimates for the smoking effect were very similar for the miner group and for the controls not exposed to radon (thus suggesting a multiplicative interaction), it would be unwise to infer support for either an additive or multiplicative relative-risk model because of sparse data and the lack of a more formal assessment of model fit.

STUDIES OF THE HOME ENVIRONMENT

Edling et al.[14] reported on a case-control study of the association among exposure to radon in homes, smoking, and lung-cancer mortality. Preliminary results were reported by Axelson.[2] The investigation was carried out on the island of Oeland, Sweden, which is located in the Baltic

Sea, where a narrow strip of uranium-containing alum shale is found on one side of the island. Cases of lung cancer and noncancer referents were obtained from death records between the years 1960 and 1978. In addition, all subjects were aged 40 yr or more and had to have lived for 30 yr or more at their death address. There were 22 cases and 178 controls for whom data were available.

For each subject, investigators classified blindly the type of housing as: wooden without a basement or on normal ground (category 0); wooden with a basement on radiation ground (alum shale) or stone, brick, and plaster with a basement on any ground or without a basement on radiation ground (category 2); or all other types (category 1) (e.g., wooden without basement on radiation ground). Next of kin provided information on smoking status through mail questionnaires.

Among nonsmokers, relative risks by the three categories of housing type were 1.0, 1.3, and 4.4, respectively, while among smokers risks were 2.7, 2.6, and 9.3. Although the data were sparse, a greater than additive interaction is suggested (Table VII-1).

STUDIES AMONG JAPANESE ATOMIC-BOMB SURVIVORS

Studies of lung-cancer mortality among atomic-bomb survivors in Hiroshima and Nagasaki offer information for assessing combined exposure to tobacco smoke and low linear energy transfer (LET) radiation received at a single time point. However, the relevance of such analyses for understanding the combined effect on lung-cancer risk of smoking and protracted, high-LET radiation is uncertain. In Part 2 of this appendix, the committee gives results of its own analysis of combined exposures to the atomic-bomb survivors.

Prentice et al.[26] combined data from several different surveys among atomic-bomb survivors: approximately 20,000 participants from the Adult Health Study who were interviewed in 1963–1964, in 1964–1968, or in 1968–1970;[5] a subset of males from the Life-Span Study cohort who were surveyed by mail in 1965;[18] and a subset of females from the Life-Span Study who were surveyed by mail in 1969–1970. A total of 40,498 subjects were available.

For analysis, Prentice et al.[26] used T65DR dose estimates for total radiation exposure. Information on tobacco use came from the various surveys. Although the questionnaires differed, current smoking pattern at the time of interview could be categorized into nonsmoking; about 5, 10, 20, or 30 cigarettes/day; and 0–4, 5–9, 10–14, 15–19, and 20 or more years of cigarette use. Smoking data, for subjects surveyed more than once, were taken from the earliest interview. To avoid bias resulting from healthy subjects surviving longer and hence having a greater likelihood of

interview, follow-up started at the initial interview and continued to death or the end of the study. A total of 281 lung-cancer deaths occurred. The Cox proportional hazards model was used for analysis.[9] Because of the relatively short 15-yr follow-up period, variation in radiation and smoking-induced lung-cancer risk with follow-up time was not evaluated.

Using categorical variables in the proportional hazards regression model and stratifying on city, sex, age at time of bombing, and survey date, relative risk of lung cancer among nonsmokers rose from 1.0 to 1.1 to 2.3 with exposure to <10, 10–99, >100 rad, respectively, while among smokers risks increased from 2.4 to 2.4 to 3.6, respectively (see Table VII-1). Additional analyses incorporating more detailed smoking information revealed no significant departures from either a multiplicative or an additive model, with the maximum likelihood values being nearly identical to each other.

Kopecky et al.[19] considered essentially the same data, but excluded those not in the city at the time of bombing and extended follow-up through 1980. A total of 29,332 subjects were in the study cohort; 351 lung-cancer deaths were observed. The results of Kopecky et al.[19] were similar to those of Prentice et al.[26] The additive-excess-risk model was shown to fit the data quite well, and neither superadditivity nor subadditivity was strongly suggested. However, Kopecky et al.[19] did not fit a multiplicative model so that results could be compared across models. Thus, while some preference for an additive model was suggested, these two analyses of atomic-bomb survivors are inconclusive in favoring a specific model.

In addition to these cohort studies, two case-control studies have been conducted. Lung-cancer cases, which were found among an autopsy series from the Life-Span Study cohort during 1961–1970, were paired with non-lung-cancer autopsy controls, matching on inclusion in the Adult Health Study, city, sex, age at death, and year of death.[16] Interviews with next of kin were conducted to ascertain information on tobacco use and occupation. A total of 180 case-control pairs were analyzed. Risk among lightly exposed (<1 rad) smokers was 3 times that of similarly exposed nonsmokers. Relative to lightly exposed nonsmokers, the risks to heavily exposed (200+ rad) smokers and nonsmokers were 8.6 and 6.2, respectively. Although sample size was small and detailed evaluation was missing, this suggests an additive model for the two effects.

Blot et al.[6] have presented preliminary results of a second case-control study of 582 lung cancers identified during the years 1971–1980 from members of the Life-Span Study cohort. Controls, also from the Life-Span Study cohort, were selected for each case and matched on date of birth, sex, city of participation in the Adult Health Study, and vital status. The 1,306 controls were selected from persons without cancer or chronic respiratory diseases. Interviews were conducted with 485 cases (83%) and

1,089 (83%) controls or next of kin (Table VII-1). Information was obtained on smoking status, passive exposure to tobacco smoke, occupation, and other factors. Among nonsmoking males, relative risks were 1.0, 2.1, and 6.2 for radiation dose categories 0–9, 10–99, and 100 rad or more, respectively, while among smoking males the corresponding risks were 9.7, 7.3, and 14.0, respectively. Among females the corresponding risks were 1.0, 0.7, and 5.3 for nonsmokers and 2.0, 2.1, and 4.5 for smokers, respectively. These patterns, based on the largest case series yet reported, appeared to suggest additive contributions for smoking and radiation, although Blot et al.[6] did not present formal significance tests in this preliminary report. The committee's analyses of related data and discussion of the fits of additive vis à vis multiplicative relative-risk model, are discussed below.

PART 2. The Committee's Analyses of Smoking and Radiation

In this portion of the appendix, we present the results of this committee's analyses of data from three populations, which address the combined effect of radiation exposure and cigarette consumption on the risk of lung cancer. The data sets include case-control studies of New Mexico uranium miners and Japanese atomic-bomb survivors and the cohort study of Colorado Plateau miners with follow-up through 1982.

METHODS

For the case-control data, models were fit using a conditional likelihood for matched data.[7] When data are matched on a time-related variable, such as age, the procedure is similar to a Cox survival time analysis for an entire cohort, and thus, the procedure is closely related to the Poisson methods that the committee employed for risk estimation.

In our modeling, it is assumed that the relative risk (RR) is the same for each matched set, regardless of level of exposure in controls (although variation with age and other matching factors can be evaluated). The RR is then modeled in several ways. Suppose the ranges for number of cigarettes smoked per day and years of mining are divided into categories; then, to estimate the relative risk associated with each variable, ignoring the other, one fits:

$$RR = 1 + \phi(\text{yr}) \text{ or} \qquad \text{(VII-1)}$$

$$RR = 1 + \phi(n/\text{day}), \qquad \text{(VII-2)}$$

where $\phi(\text{yr})$ and $\phi(n/\text{day})$ denote the individual excess RR estimates for categories of years of underground mining and number of cigarettes smoked per day, respectively, and are defined so that ϕ takes a value of zero at the baseline category.

More complex models that incorporate more than one variable can be defined. For example, for years of exposure and number of cigarettes per day, one can specify a multiplicative or additive combination of RR effects, namely:

$$RR = [1 + \phi(\text{yr})][1 + \phi(n/\text{day})] \text{ or} \quad \text{(VII-3)}$$

$$RR = [1 + \phi(\text{yr}) + \phi(n/\text{day})]. \quad \text{(VII-4)}$$

These are not nested models, since they involve the same number of parameters. However, each of these formulations can be imbedded in a richer RR model and compared to it.

In our evaluation, we also applied the transformation proposed by Thomas.[34] The relative risk for combined exposure is defined as follows:

$$RR = \{[1 + \phi(\text{yr})][1 + \phi(n/\text{day})]\}^{\lambda}[1 + \phi(\text{yr}) + \phi(n/\text{day})]^{1-\lambda}. \quad \text{(VII-5)}$$

At $\lambda = 1$, RR reduces to the multiplicative model, while at $\lambda = 0$, RR reduces to the additive model, as in Equations VII-3 and VII-4. Through the parameter λ, this richer model defines a smooth transformation in RR, which incorporates both additive and multiplicative models. Models given in Equations VII-3 to VII-5 can be compared to the saturated model given by:

$$RR = 1 + \phi(\text{yr}, n/\text{day}), \quad \text{(VII-6)}$$

were ϕ represents the excess RR in each cell of the cross-classification. If there are four categories of each variable, then $\phi(\text{yr}, n/\text{day})$ represents 15 free parameters, with the baseline parameter being fixed at zero.

Analogous to methodology described in Annex 2A, we fit Poisson regression models to the data on smoking and exposure to radiation described below. We assumed that the expected number of events in each cell of a cross-classification is the product of the person-years accrued times the lung-cancer disease rate, which is modeled as an age- and calendar-period-specific rate among nonexposed persons times a relative risk function, namely:

$$\text{person} - \text{yr} \times \alpha_a \times RR, \quad \text{(VII-7)}$$

where α_a represents age and year parameters. The models for RR are the same as defined above.

CASE-CONTROL STUDY AMONG NEW MEXICO URANIUM MINERS

The committee obtained access to data from an ongoing case-control study of lung cancer in a cohort of New Mexico uranium miners.[23] The cohort includes 4,051 subjects with at least 1 yr of documented underground employment in a New Mexico uranium mine. Large-scale uranium mining did not begin in New Mexico until the late 1950s, and exposures have generally been lower than those to the Colorado Plateau miners.

For assessment of the combined effects of cigarette smoking and uranium-mining exposure, a case-control study was conducted within this cohort. The cases included all Hispanic and non-Hispanic white males diagnosed with lung cancer, regardless of whether cause of death had been coded as lung cancer. The selection date of the case was the earlier of the date of diagnosis or death. For each of the 69 cases, four controls were selected. The controls were also Hispanic or non-Hispanic white males who (1) met the cohort entry criteria before the selection date of the case, (2) were alive and free of lung cancer at the selection date of the case, (3) had some follow-up information, and (4) had a record of a physical examination related to mine employment. From the pool of controls, the four controls closest in age to the case were selected.

Because the computation of cumulative exposures has not been completed for this cohort, the number of documented years of employment was used as the exposure variable. Cigarette-smoking information was available from one or more histories obtained at a pre-employment or annual physical examinations.

Table VII-2 gives the relevant data and shows that risks are elevated with the use of all types of tobacco products and increase with number of cigarettes smoked per day and with years spent in underground mining. The remaining analyses are restricted to nonsmokers and cigarette smokers; those who smoked cigars and/or pipes exclusively were dropped.

Matched RR regression models were fit to the 52 cases and 218 controls for whom the committee had data on both variables. Table VII-3 shows the distribution of cases and controls for the cross-classification of years of underground mining and smoking. Based on estimates from fitting a full 15-parameter model, risks are increased with years of underground mining within each cigarette-use category. RR estimates from the additive and multiplicative main effects models are also shown. The summary of the model fitting is given at the bottom of Table VII-3 and indicates that neither the multiplicative nor the additive model deviated significantly

TABLE VII-2 Data on Smoking Rate and Radiation Exposure from Case Control Study of New Mexico Uranium Miners by Various Variables[a]

	NS	FS	CS	KS	C+P/S	P/S	P+S	C+P+S	Total
Cases	2	0	33	14	8	1	0	5	63
Controls	49	3	117	56	28	1	3	9	266
RR[b]	1	—	7.4	6.6	7.8	22.6	—	13.7	

	No. of Cigarettes Smoked/day				
	<5	5–14	15–24	25+	Total
Cases	3	16	28	5	52
Controls	56	72	77	17	222
RR[b]	1	5.7	8.3	7.0	

	Years of Undergrounding Mining				
	<10	10–14	15–19	20+	Total
Cases	28	15	12	14	69
Controls	151	59	38	24	272
RR[b]	1	1.5	1.7	4.4	

[a]Abbreviations: NS, nonsmoker; FS, former smoker; CS, current smoker; KS, known to smoke: type, amount, and duration unknown; C+P/S, cigarette smoker who also used pipe or cigars; P/S, pipe or cigar smoker; P+S, smoked both pipe and cigars; C+P+S, smoked cigarettes, pipe, and cigars.
[b]Estimated from matched data.

from the full 15-parameter model. Although based on only 52 cases, the results suggest that the multiplicative model provides a better fit. The committee also fit the model defined by Equation VII-5 by fixing a sequence of λ values. The maximum log-likelihood (MLL) as a function of λ was rather flat, reaching a maximum at $\lambda = 4.0$ with $2 \times \text{MLL} = -127.2$, a value not much different from the simple multiplicative model (cf. Table VII-3).

CASE-CONTROL STUDY FROM THE LIFE-SPAN STUDY COHORT, JAPANESE ATOMIC-BOMB SURVIVORS

The committee's current analysis of radon-exposed miners has revealed substantial differences in the effects of radiation on lung cancer in miners in comparison with the Japanese atomic-bomb survivors. Most notable is the decline in excess risk by time since exposure, whereas there is little evidence of such decline in risk with time since exposure for atomic-bomb survivors. It is important to characterize differences and similarities in exposure effects among different radiation-exposed populations to provide insight into mechanisms of action for the exposures. The committee presents a

TABLE VII-3 Data from Case Control Study of New Mexico Uranium Miners

No. of cigarettes/day	Years of Underground Mining: <10		10–14		15–19		20+	
	No. of Cases	No. of Controls	No. of Cases	No. of Controls	No. of Cases	No. of Controls	No. of Cases	No. of Controls
<5	1	27	1	15	1	7	0	5
5–14	7	40	5	15	2	14	2	1
15–24	7	31	6	21	7	14	8	11
25+	2	8	1	4	0	1	2	4
Total	17	106	13	55	10	36	12	21

	Relative Risks					
	<10	10–14	15–19	20+	RR[a]	RR[b]
<5	1	1.0	3.7	0	1	1
5–14	5.1	12.0	4.2	39.9	6.8	5.7
15–24	7.0	6.7	17.5	24.0	8.6	6.6
25+	8.2	6.2	0.0	30.1	8.2	6.2
RR[a]	1	1.8	3.9	14.6		
RR[b]	1	1.3	1.6	3.8		

Regression Models	No. of Parameters	2×MLL	*P*-Value
1: $1 + \phi(yr, n/d)$	15	−121.8	
2: $[1 + \phi(yr)][1 + \phi(n/d)]$	6	−127.6	0.76
3: $1 + \phi(yr) + \phi(n/d)$	6	−129.6	0.55
4: $1 + \phi(yr)$	3	−135.9	0.29
5: $1 + \phi(n/d)$	3	−133.2	0.50

[a]Relative risks from additive model, Equation VII-2.
[b]Relative risks from multiplicative model, Equation VII-1.

new analysis of radiation exposure and cigarette use on lung-cancer risk, using data from a recent case-control study among atomic-bomb survivors, to formally evaluate their combined effects.

The details of this study have been described by Blot et al.[6] Cases include diagnosed lung cancers from participants of the Life-Span Study cohort (LSS) during 1971–1980. Death certificates were used to identify lung cancers among members of this cohort who resided outside of Hiroshima and Nagasaki, so that the 582 cases do not constitute, precisely, an incident series. Controls were selected from LSS members and matched by date of birth, sex, city, Radiation Effects Research Foundation sample status, and survival status. Two controls were selected for each Hiroshima case, and three were selected for each Nagasaki case. Smoking and other

information was obtained by direct interview of index subjects or their next of kin.

Table VII-4 presents the data by sex for categories of number of cigarettes smoked per day and radiation exposure. *RR*s obtained from a matched analysis are given in Table VII-5. Risks generally rise with amount of radiation received and number of cigarettes smoked. (Numbers differ slightly from those of Blot et al.[6] due to exclusion of cases who had suspect diagnoses.)

At first inspection, the data in Table VII-5 appear to support an additive model for these joint exposures. For example, by adding excess risks for males, the estimated relative risk in the highest category for each exposure based on separate exposures, $1 + (26.5 - 1) + (3.3 - 1) = 29.8$, is very close to the observed value of 24.8. Similarly, $1 + (6.9 - 1) + (3.3 - 1) = 9.2$ is close to 10.6. However, a specified model must hold throughout Table VII-5, so that the *RR* for a 20+ cigarette smoker who was exposed to less than 10 rad should be related to the 20+ per day and 10–99 rad category and the 0 per day should be related to the 10–99 rad category. Linking these, we have that $1 + (13.2 - 1) - (1.3 - 1) = 12.9$ should be the approximate risk for a 20+ per day smoker with <10 rad exposure. This is clearly quite different from the observed value of 26.5. Similar discrepancies can be found in other parts of the table. A more appropriate examination of Table VII-5 is provided by comparing the *RR*s with fitted estimates from additive and multiplicative main effects models, respectively:

TABLE VII-4 Data on Smoking Rate and Radiation Exposure from a Case Control Study of Lung Cancer among Japanese A-Bomb Survivors[a]

No. of Cigarettes/day	Radiation Exposure (rad)							
	<10		10–99		100+		Total	
	Cases	Controls	Cases	Controls	Cases	Controls	Cases	Controls
				Males				
0	6	65	3	19	3	8	12	92
1–10	21	73	7	33	4	7	32	113
11–20	49	111	17	43	7	15	73	169
20+	45	31	11	12	9	4	65	47
				Females				
0	51	151	15	59	16	14	82	224
1–10	16	24	5	9	6	5	27	38
11+	10	9	7	10	2	0	19	19

[a]Based on data from Blot et al.[6]

TABLE VII-5 Relative Risks from Matched Analysis for Radiation Exposure and Number of Cigarettes Smoked per Day by Japanese A-Bomb Survivors

No. of Cigarettes/day	Radiation Exposure (rad)				
	<10	10-99	100+	RR[a]	RR[b]
			Males		
0	1	1.3	3.3	1	1
1-10	3.7	2.4	7.2	3.0	2.7
11-20	6.9	6.6	10.6	6.0	5.5
20+	26.5	13.2	24.8	19.4	17.2
RR[a]	1	0.9	3.5		
RR[b]	1	0.8	1.6		
			Females		
0	1	0.7	5.2	1	1
1-10	2.3	2.5	5.2	2.4	2.2
11+	4.2	2.1	—	3.3	3.7
RR[a]	1	0.6	4.9		
RR[b]	1	0.6	4.0		

[a]Relative risks from additive model, Equation VII-2.
[b]Relative risks from multiplicative model, Equation VII-1.

$$RR = 1 + \phi(\text{rad}) + \phi(n/\text{day}) \text{ and} \quad \text{(VII-8)}$$

$$RR = [1 + \phi(\text{rad})][1 + \phi(n/\text{day})]. \quad \text{(VII-9)}$$

These estimates are included in Table VII-5. Visual inspection of the *RR*s based on the fitted estimates does not favor either model.

Tables VII-6 and VII-7 cross-classify data and *RR*s by radiation exposure and duration, in years, of cigarette use. Again, risks increase with both exposures. Formal model comparisons were carried out to assess adequacy relative to the 15 parameter model. Table VII-8 summarizes results and shows that for both sexes the multiplicative model with radiation and number of cigarettes smoked per day fits as well as the additive model. The lower half of Table VII-8 shows little difference between additive and multiplicative models when radiation and duration of cigarette use are included.

Several additional models were fit that included continuous exposures, and similar results were obtained. Because the MLL for the main effects models for females and males were almost the same as that for the full model, power models were not tested.

Prentice et al.[26] suggest that either multiplicative or additive relative-risk models fit the atomic-bomb survivor data for lung cancer, with little

TABLE VII-6 Data on Duration of Cigarette Smoking and Radiation Exposure from a Case Control Study of Lung Cancer among Japanese A-Bomb Survivors[a]

	Radiation Exposure (rads)							
	<10		10–99		100+		Total	
Years of Smoking	Cases	Controls	Cases	Controls	Cases	Controls	Cases	Controls
				Males				
0	6	65	3	19	3	8	12	92
1–34	9	24	47	12	1	3	14	39
35–44	6	38	5	16	4	4	15	58
45+	64	78	16	28	9	4	89	110
				Females				
0	51	151	15	59	16	14	82	224
1–34	5	17	2	8	3	1	10	26
35–44	7	3	4	5	0	1	11	9
45+	9	7	4	4	4	1	17	12

[a]Based on data from Blot et al.[6]

TABLE VII-7 Relative Risks from Matched Analysis for Radiation Exposure and Years of Cigarette Use among Japanese A-Bomb Survivors

	Radiation Exposure (rad)				
Years of Smoking	<10	10–99	100+	RR[a]	RR[b]
			Males		
0	1	1.0	8.8	1	1
1–34	1.8	2.0	1.2	1.6	1.1
35–44	1.6	4.9	9.6	2.0	1.9
45+	18.4	13.3	87.2	14.0	11.3
RR[a]	1	1.4	5.5		
RR[b]	1	1.0	3.4		
			Females		
0	1	0.6	4.6	1	1
1–34	0.9	1.2	8.5	1.0	1.1
35–44	14.9	2.6	0.0	5.6	7.3
45+	5.8	2.9	7.2	4.5	4.4
RR[a]	1	0.7	4.6		
RR[b]	1	0.6	4.1		

[a]Relative risks from additive model, Equation VII-2.
[b]Relative risks from multiplicative model, Equation VII-1.

TABLE VII-8 Results of Fitting Additive and Multiplicative Relative Risk Models to Evaluate Radiation Exposure and Number of Cigarettes Smoked per Day or Duration in Years of Cigarette Use among Japanese A-Bomb Survivors

	Males			Females		
Model	No. of Parameters	2 × MLL	*P* Value	No. of Parameters	2 × MLL	*P* Value
1: $1 + \phi(\text{rad}, n/d)$	11	−273.7		8	−219.1	
2: $[1 + \phi(\text{rad})][1 + \phi(n/d)]$	5	−276.0	0.89	4	−220.8	0.79
3: $1 + \phi(\text{rad}) + \phi(n/d)$	5	−275.9	0.90	4	−219.8	0.95
4: $1 + \phi(\text{rad})$	2	−335.4	<0.01	2	−234.3	0.02
5: $1 + \phi(n/d)$	3	−279.1	0.71	2	−236.7	0.01
1: $1 + \phi(\text{rad}, \text{dur})$	11	−140.8		11	−191.4	0.88
2: $[1 + \phi(\text{rad})][1 + \phi(\text{dur})]$	5	−144.7	0.69	5	−193.8	0.80
3: $1 + \phi(\text{rad}) + \phi(\text{dur})$	5	−146.1	0.51	5	−194.5	0.80
4: $1 + \phi(\text{rad})$	2	−187.9	<0.01	2	−212.5	0.01
5: $1 + \phi(\text{dur})$	3	−150.5	0.29	3	−208.8	0.03

preference. Kopecky et al.[19] fit only the additive model in their analysis. The committee's reanalysis of case-control data extracted from members of the LSS group agrees with the interpretation of Prentice et al.[26] in that the committee found no strong preference for an additive combination of radiation and smoking.

COHORT STUDY OF COLORADO PLATEAU URANIUM MINERS

The committee evaluated the combined effects of cigarette consumption and cumulative WLM exposure, using data from the cohort of Colorado Plateau uranium miners with follow-up through 1982. There were 256 observed lung-cancer cases in over 73,000 person-yr of observation.

Assignment of WLM exposures is described in Appendix IV. A 5-yr lag interval was used to determine exposure. Smoking information was obtained from initial medical examination, periodic surveys, and mail questionnaires. For this analysis, the committee used the mean number of cigarettes smoked per day as calculated from all available sources. Because the last update of cigarette use information was in 1969 and because precise details were not available, the committee did not attempt to evaluate other tobacco-related determinants of risk, such as duration of use, time since cessation of smoking, filter or nonfilter cigarette use, and intensity of inhalation. These factors are strongly related to lung-cancer risk and the inability of the committee to consider them may have an impact on this evaluation of combined effects of tobacco use and radon-daughter exposure. Thus, the results described below should be viewed cautiously, pending a more thorough evaluation.

For this analysis the same Poisson regression techniques were used as for the radon risk estimation. Data on lung-cancer events and person-years were cross-classified by categories of cumulative WLM exposure (<60, 60–119, 120–239, 240–479, 480–959, ≥ 960), number of cigarettes smoked per day (0–4, 5–19, 20–29, ≥ 30), age (<55, 55–59, 60–64, 65–69, ≥ 70), and calendar year (1950–1959, 1960–1964, 1965–1969, 1970–1974, 1975–1982). Table VII-9 shows the number of observed lung cancers, person-years, and crude disease rate by WLM exposure and cigarette use when data were collapsed across age and year categories. The numbers differ slightly from the tables in Annex 2A because of a different cross-classification. Note that cells with means exceeding 2,000 WLM have been excluded.

For these data we fit a more general relative risk model than that given in Equation VII-5, namely,

TABLE VII-9 Observed Lung-Cancer Mortality and Calculated Lung-Cancer Mortality Rate as a Function of Cumulative Exposure and Cigarette Consumption for the Colorado Plateau Miner Cohort[a]

Cumulative WLM	No. of Cigarettes/day	0-4	5-19	20-29	30+	Total
0-59	Observe	0	1	7	1	9
	Rate	12.3[b]	35.8	102.2	39.5	49.9
	P-yr[c]	5,878.8	2,790.5	6,848.3	2,530.5	18,048.0
60-119	Observed	0	0	2	3	5
	Rate	0	0	81.9	404.3	78.8
	P-yr	2,263.0	894.0	2,443.5	742.0	6,342.5
120-239	Observed	1	2	9	2	14
	Rate	34.8	138.9	232.0	157.3	148.0
	P-yr	2,872.0	1,439.0	3,879.0	1,271.5	9,461.5
240-479	Observed	6	1	12	8	27
	Rate	157.5	54.0	229.2	421.7	211.0
	P-yr	3,809.3	1,851.5	5,236.8	1,897.0	2,794.5
480-959	Observed	11	3	29	14	57
	Rate	323.1	216.0	523.8	651.7	456.8
	P-yr	3,404.5	1,389.0	5,536.5	2,148.3	12,478.3
960+	Observed	4	6	10	19	39
	Rate	289.5	554.0	457.5	1,189.0	625.0
	P-yr	1,381.8	1,083.0	2,186.0	1,598.0	6,239.8
Total	Observed	22	13	69	47	151
	Rate	112.2	137.6	264.1	461.8	231.0
	P-yr	19,609.3	9,447.5	26,130.0	10,178.3	65,365.0

[a]Cumulative exposure limited to 2,000 WLM.
[b]Baseline rate per 100,000 computed using expected number of cases, based on U.S. white male mortality rates for lung cancer adjusted to nonsmokers.
[c]Person years.

$$RR = R[w(a), n(a); \lambda] = \{[1 + \phi(w)\gamma(a)][1 + \phi(n)\delta(a)]\}^{\lambda} [1 + \phi(w)\gamma(a) + \phi(n)\delta(a)]^{1-\lambda}, \quad \text{(VII-10)}$$

where, as previously, $\phi(w)$ and $\phi(n)$ represent parameters for categories of cumulative WLM (w) and cigarettes per day (n), and where $\gamma(a)$ and $\delta(a)$ denote modifications of the $\phi(w)$ and $\phi(n)$ effects with age a. We have defined $\gamma(a) = \delta(a) = 1$ for age $a < 65$ yr and $\gamma(a) = \gamma$ and $\delta(a) = \delta$ for $a > 65$ yr. To reduce model complexity, we did not include a parameter for time since exposure. This factor is not significant in the Colorado data.

The marginally saturated model given in Equation VII-6 was generalized to include age as:

$$RR = 1 + \phi(w, n, a), \qquad \text{(VII-11)}$$

where ϕ denoted 46 parameters, including 23 parameters for the cross-classification of cumulative WLM and cigarettes per day for ages <65 yr and 23 parameters for exposures for ages >65 yr. The baseline rate $r_0(a)$ incorporates nine parameters for multiplicative effects of age and calendar year.

The class of models characterized by Equation VII-10 reduces to Equation VII-5 when $\gamma(a) = \delta(a) = 1$ for all a and includes the multiplicative ($\lambda = 1$) and additive ($\lambda = 0$) models. The inclusion of γ and δ (actually the exponential of each) permits formal likelihood ratio testing of age effects for cumulative WLM exposure and cigarettes per day. As seen in Annex 2A, the effects of cumulative WLM decline with age at risk.

Table VII-10 shows predicted relative risks by age group based on various models (risks relative to the lung-cancer rate in the entire cohort), while Table VII-11 gives results from model fittings. Table VII-11 indicates that age at risk is an important modifier for cumulative WLM exposure ($P = 0.005$), but not for cigarettes per day ($P = 0.8$). The estimate of the effect of age modification for cumulative WLM $\gamma(a)$ is 0.1 for ages 65 or over for both from $R[w(a),n(a);\lambda]$ and $R[w(a),n;\lambda]$, with $\lambda = 1.0$ and with the maximum likelihood estimates for λ. Note that this confirms the analyses of Annex 2A, in which cigarette use is not included.

Therefore, it is appropriate to consider a reduced form of Equation VII-10, $R[w(a),n;\lambda]$, where $\delta(a) = 1$ for all a. Focusing on models labeled 2 and 3 in Table VII-11, additive effects for WLM and cigarettes per day are rejected relative to the mixture model, while multiplicative effects are not. The maximum likelihood estimates for λ are 0.4 under $R[w(a),n(a);\lambda]$ and 0.6 under $R[w(a),n;\lambda]$. However, the maximized likelihood with $\lambda = 0.4$ (or $\lambda = 0.6$) is very similar to the likelihood with λ fixed at one, indicating a comparable fit. It should be noted that the likelihood in λ was very flat for $\lambda > 0.3$, which precludes precise specification of λ. The value $\lambda = 0.4$ (or $\lambda = 0.6$) does not indicate that the true model is halfway between additive and multiplicative models. Rather, because of the variability in λ and skewness of the distribution of possible values of λ, one cannot be more precise than to say that the data are consistent with a range of joint-effect models, from submultiplicative to supermultiplicative.

The committee's analyses of the interaction between smoking and cumulative exposure support the conclusions of Whittemore and McMillan.[38] An additive model is rejected in the committee's analysis, while a multiplicative combination of relative risks provides an acceptable fit. However, the committee also found that by embedding the simple models into a larger class of mixture models, a range of submultiplicative to supramultiplicative models was equally compatible with the data.

TABLE VII-10 Relative Risks for Lung Cancer among Colorado Plateau Miner's Cohort Based on Various Models for Cumulative WLM (w) and Cigarettes per day (n); Risks Relative to Rate in Entire Cohort

Cumulative WLM	Model[a]	No. of Cigarettes/day 0–4	5–19	20–29	30+
	Age < 65				
0–59	$[1 + \phi(w, n, a)]$	0.00	0.12	0.10	0.12
	$R[w(a), n; \lambda = 1.0]$	0.04	0.04	0.10	0.12
	$R[w(a), n; \lambda = 0.6]$	0.03	0.03	0.12	0.14
	$R[w(a), n; \lambda = 0.0]$	0.02	0.04	0.20	0.24
60–119	$[1 + \phi(w, n, a)]$	0.00	0.00	0.20	0.58
	$R[w(a), n; \lambda = 1.0]$	0.07	0.07	0.19	0.23
	$R[w(a), n; \lambda = 0.6]$	0.06	0.06	0.18	0.21
	$R[w(a), n; \lambda = 0.0]$	0.02	0.04	0.20	0.24
120–239	$[1 + \phi(w, n, a)]$	0.00	0.17	0.36	0.18
	$R[w(a), n; \lambda = 1.0]$	0.10	0.10	0.26	0.31
	$R[w(a), n; \lambda = 0.6]$	0.09	0.10	0.26	0.31
	$R[w(a), n; \lambda = 0.0]$	0.07	0.09	0.25	0.29
240–479	$[1 + \phi(w, n, a)]$	0.18	0.13	0.41	0.81
	$R[w(a), n; \lambda = 1.0]$	0.17	0.17	0.44	0.53
	$R[w(a), n; \lambda = 0.6]$	0.17	0.17	0.44	0.51
	$R[w(a), n; \lambda = 0.0]$	0.19	0.21	0.37	0.41
480–959	$[1 + \phi(w, n, a)]$	0.48	0.34	0.64	1.04
	$R[w(a), n; \lambda = 1.0]$	0.31	0.32	0.82	0.99
	$R[w(a), n; \lambda = 0.6]$	0.35	0.35	0.82	0.94
	$R[w(a), n; \lambda = 0.0]$	0.48	0.50	0.66	0.70
960+	$[1 + \phi(w, n, a)]$	0.80	1.00	2.20	2.87
	$R[w(a), n; \lambda = 1.0]$	0.79	0.81	2.07	2.51
	$R[w(a), n; \lambda = 0.6]$	0.90	0.91	2.03	2.30
	$R[w(a), n; \lambda = 0.0]$	1.25	1.27	1.44	1.47
	Age ≥ 65				
0–59	$[1 + \phi(w, n, a)]$	0.00	0.00	1.65	0.00
	$R[w(a), n; \lambda = 1.0]$	0.24	0.25	0.88	1.06
	$R[w(a), n; \lambda = 0.6]$	0.33	0.34	0.86	1.04
	$R[w(a), n; \lambda = 0.0]$	0.08	0.17	0.87	1.03
60–119	$[1 + \phi(w, n, a)]$	0.00	0.00	0.00	3.41
	$R[w(a), n; \lambda = 1.0]$	0.26	0.27	0.93	1.13
	$R[w(a), n; \lambda = 0.6]$	0.35	0.36	0.93	1.12
	$R[w(a), n; \lambda = 0.0]$	0.08	0.17	0.87	1.03
120–239	$[1 + \phi(w, n, a)]$	0.50	0.98	1.16	0.99
	$R[w(a), n; \lambda = 1.0]$	0.29	0.30	1.01	1.22
	$R[w(a), n; \lambda = 0.6]$	0.38	0.39	0.99	1.19
	$R[w(a), n; \lambda = 0.0]$	0.13	0.22	0.92	1.08

TABLE VII-10 (*Continued*)

Cumulative WLM	Model[a]	No. of Cigarettes/day 0-4	5-19	20-29	30+
240-479	$[1 + \phi(w, n, a)]$	0.60	0.00	0.77	0.74
	$R[w(a), n; \lambda = 1.0]$	0.35	0.36	1.16	1.40
	$R[w(a), n; \lambda = 0.6]$	0.43	0.44	1.13	1.36
	$R[w(a), n; \lambda = 0.0]$	0.24	0.34	1.04	1.20
480-959	$[1 + \phi(w, n, a)]$	1.00	0.78	3.08	1.17
	$R[w(a), n; \lambda = 1.0]$	0.49	0.50	1.50	1.79
	$R[w(a), n; \lambda = 0.6]$	0.54	0.56	1.42	1.72
	$R[w(a), n; \lambda = 0.0]$	0.52	0.62	1.32	1.47
960+	$[1 + \phi(w, n, a)]$	0.55	0.63	0.00	2.48
	$R[w(a), n; \lambda = 1.0]$	0.94	0.96	2.53	2.96
	$R[w(a), n; \lambda = 0.6]$	0.91	0.94	2.39	2.89
	$R[w(a), n; \lambda = 0.0]$	1.27	1.36	2.06	2.22

[a]Age-specific rates are $r_0(a)$ times the relative risk, as defined by the various models. The r_0 term includes nine parameters for age and calendar year. The $\phi(w, n, a)$ term denotes 46 free parameters, including 23 for the exposure cross-classification for age $a < 65$ and 23 for age $a \geq 65$ years. Other models are defined as follows;

$$R[w(a), n(a); \lambda] = \{[1 + \phi(w)\gamma(a)][1 + \phi(n)\delta(a)]\}^{\lambda}[1 + \phi(w)\gamma(a) + \phi(n)\gamma(a)]^{1-\lambda}$$

where $\phi(w)$ and $\phi(n)$ denote parameters for categories of cumulative WLM and cigarettes per day, and $\gamma(a)$ and $\delta(a)$ denote their respective age effects for $a < 65$ and $a \geq 65$. For the values in the table, $\delta(a) = 1$ for all a. At $\lambda = 1$, R specifies multiplicative effects for WLM and cigarettes per day for ages <65 and ≥ 65; and at $\lambda = 0$, R specifies additive effects.

APPLICATION OF THE MULTIPLICATIVE MODEL

In Chapter 2, the committee outlines how it applies the multiplicative model for the combined effects of smoking and exposure to radon progeny. Table 2.4 in Chapter 2 lists the estimated risk to smokers and nonsmokers of both sexes due to lifetime exposure. Exposures for shorter periods of time are also of interest since exposure to elevated levels of radon may occur and end at any age. Tables VII-12 to VII-14 and Tables VII-15 to VII-17 provide for male smokers and nonsmokers, respectively, the ratio of the lifetime risk of lung-cancer mortality due to exposure occurring within stated intervals of age. Included in the tables are two other measures of risk described in Chapter 2 and defined mathematically in Annex 2A. These are R_e, the lifetime risk of lung-cancer mortality, which includes the baseline risk R_0, for exposure between two age intervals; and $L_0 - L_e$, the number of years of life lost due to such exposures. Tables VII-18 to VII-20 and Tables VII-21 to VII-23 provide the same set of results for female smokers and nonsmokers.

TABLE VII-11 Results for Fitting Various Relative Risk Models to Colorado Plateau Miners' Cohort

	Model[a]	2 × MLL	No. of Parameters
1	$[1 + \phi(w, n, a)]$	−338.8	SS
2(a)	$R[w(a), n(a); \lambda = 0.4]$	−376.6	20
2(b)	$R[w(a), n(a); \lambda = 1.0]$	−377.4	19
2(c)	$R[w(a), n(a); \lambda = 0.0]$	−382.6	19
3(a)	$R[w(a), n; \lambda = 0.6]$	−377.0	19
3(b)	$R[w(a), n; \lambda = 1.0]$	−377.5	18
3(c)	$R[w(a), n; \lambda = 0.0]$	−384.8	18
4(a)	$R[w, n(a); \lambda = 0.2]$	−381.4	19
4(b)	$R[w, n(a); \lambda = 1.0]$	−385.4	18
4(c)	$R[w, n(a); \lambda = 0.0]$	−384.3	18
5(a)	$R[w, n; \lambda = 0.3]$	−383.4	18
5(b)	$R[w, n; \lambda = 1.0]$	−385.4	17
5(c)	$R[w, n; \lambda = 0.0]$	−388.2	17

	Tests of hypothesis: Chi-Sq (d.f.)	*P* value
Fit of mixture model:		
2(a) vs 1	37.8 (35)	0.343
3(a) vs 1	38.2 (36)	0.370
4(a) vs 1	42.6 (36)	0.208
5(a) vs 1	44.6 (37)	0.183
Age effects for WLM		
4(b) vs 2(b)	8.0 (1)	0.005
5(b) vs 3(b)	7.0 (1)	0.005
Age effects for cigarettes per day:		
3(b) vs 2(b)	0.1 (1)	0.752
5(b) vs 4(b)	0.0 (1)	0.841
Multiplicative fit:		
2(b) vs 2(a)	0.8 (1)	0.371
3(b) vs 3(a)	0.5 (1)	0.480
Additive fit:		
2(c) vs 2(a)	6.0 (1)	0.014
3(c) vs 3(a)	7.8 (1)	0.005

[a]See footnote *a* to Table VII-10.

TABLE VII-12 Ratio of Lifetime Risks (R_e/R_0) by Age Started and Age Exposure Ends for Various Rates of Annual Exposure[a] for Male Smokers[b]

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
					Exposure Rate = 0.10 (WLM/yr)				
0	1.007	1.015	1.022	1.030	1.038	1.045	1.049	1.050	1.050
10		1.007	1.015	1.022	1.031	1.038	1.041	1.043	1.043
20			1.007	1.015	1.024	1.031	1.034	1.035	1.036
30				1.008	1.016	1.023	1.027	1.028	1.028
40					1.009	1.015	1.019	1.020	1.021
50						1.007	1.010	1.012	1.012
60							1.004	1.005	1.005
					Exposure Rate = 0.20 (WLM/yr)				
0	1.015	1.029	1.044	1.059	1.076	1.090	1.097	1.099	1.100
10		1.015	1.029	1.045	1.062	1.075	1.082	1.085	1.085
20			1.015	1.030	1.047	1.061	1.068	1.071	1.071
30				1.016	1.033	1.046	1.053	1.056	1.056
40					1.017	1.031	1.038	1.040	1.041
50						1.014	1.021	1.024	1.024
60							1.007	1.010	1.010
					Exposure Rate = 0.50 (WLM/yr)				
0	1.036	1.072	1.108	1.147	1.188	1.221	1.238	1.244	1.245
10		1.036	1.073	1.111	1.153	1.186	1.203	1.210	1.211
20			1.037	1.075	1.117	1.151	1.168	1.174	1.176
30				1.039	1.081	1.115	1.132	1.139	1.140
40					1.043	1.077	1.094	1.101	1.102
50						1.034	1.052	1.059	1.060
60							1.018	1.024	1.025
					Exposure Rate = 1.00 (WLM/yr)				
0	1.072	1.144	1.215	1.289	1.370	1.433	1.465	1.477	1.479
10		1.072	1.144	1.220	1.301	1.366	1.399	1.410	1.412
20			1.073	1.150	1.232	1.298	1.331	1.343	1.345
30				1.078	1.161	1.228	1.261	1.274	1.276
40					1.085	1.152	1.186	1.199	1.201
50						1.069	1.103	1.116	1.118
60							1.035	1.048	1.051
					Exposure Rate = 4.00 (WLM/yr)				
0	1.283	1.552	1.809	2.068	2.334	2.534	2.628	2.660	2.665
10		1.283	1.555	1.828	2.109	2.321	2.421	2.455	2.460
20			1.286	1.575	1.871	2.095	2.201	2.238	2.244
30				1.305	1.617	1.854	1.968	2.007	2.014
40					1.331	1.583	1.704	1.747	1.754
50						1.269	1.399	1.445	1.453
60							1.139	1.189	1.197

TABLE VII-12 (*Continued*)

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
	Exposure Rate = 10.00 (WLM/yr)								
0	1.680	2.275	2.801	3.293	3.756	4.068	4.196	4.234	4.239
10		1.681	2.281	2.841	3.367	3.725	3.875	3.920	3.926
20			1.687	2.326	2.925	3.336	3.510	3.564	3.571
30				1.730	2.415	2.887	3.091	3.155	3.164
40					1.790	2.337	2.577	2.655	2.666
50						1.645	1.932	2.026	2.041
60							1.337	1.449	1.467
	Exposure Rate = 20.00 (WLM/yr)								
0	2.275	3.252	4.014	4.647	5.171	5.466	5.562	5.585	5.587
10		2.276	3.262	4.073	4.738	5.121	5.249	5.281	5.284
20			2.287	3.333	4.187	4.684	4.858	4.903	4.908
30				2.364	3.470	4.121	4.357	4.420	4.428
40					2.466	3.337	3.664	3.755	3.766
50						2.205	2.672	2.807	2.824
60							1.641	1.833	1.859

[a] Estimated with the committee's TSE model (Chapter 2) and a multiplicative interaction between smoking and exposure to radon progeny.

[b] R_0, the calculated lifetime risk for unexposed male smokers, is 0.123.

TABLE VII-13 Lifetime Risk (R_r) by Age Started and Age Exposure Ends for Various Rates of Annual Exposure[a] for Male Smokers

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
	Exposure Rate = 0.10 (WLM/yr)								
0	0.124	0.125	0.126	0.127	0.128	0.128	0.129	0.129	0.129
10		0.124	0.125	0.126	0.127	0.128	0.128	0.128	0.128
20			0.124	0.125	0.126	0.127	0.127	0.127	0.127
30				0.124	0.125	0.126	0.126	0.126	0.126
40					0.124	0.125	0.125	0.125	0.125
50						0.124	0.124	0.124	0.124
60							0.123	0.123	0.124
	Exposure Rate = 0.20 (WLM/yr)								
0	0.125	0.126	0.128	0.130	0.132	0.134	0.135	0.135	0.135
10		0.125	0.126	0.128	0.130	0.132	0.133	0.133	0.133
20			0.125	0.127	0.129	0.130	0.131	0.132	0.132
30				0.125	0.127	0.129	0.129	0.130	0.130
40					0.125	0.127	0.128	0.128	0.128
50						0.125	0.125	0.126	0.126
60							0.124	0.124	0.124
	Exposure Rate = 0.50 (WLM/yr)								
0	0.127	0.132	0.136	0.141	0.146	0.150	0.152	0.153	0.153
10		0.127	0.132	0.137	0.142	0.146	0.148	0.149	0.149
20			0.127	0.132	0.137	0.141	0.144	0.144	0.144
30				0.128	0.133	0.137	0.139	0.140	0.140
40					0.128	0.132	0.134	0.135	0.135
50						0.127	0.129	0.130	0.130
60							0.125	0.126	0.126
	Exposure Rate = 1.00 (WLM/yr)								
0	0.132	0.141	0.149	0.158	0.168	0.176	0.180	0.182	0.182
10		0.132	0.141	0.150	0.160	0.168	0.172	0.173	0.174
20			0.132	0.141	0.151	0.159	0.164	0.165	0.165
30				0.132	0.143	0.151	0.155	0.157	0.157
40					0.133	0.142	0.146	0.147	0.148
50						0.131	0.136	0.137	0.137
60							0.127	0.129	0.129
	Exposure Rate = 4.00 (WLM/yr)								
0	0.158	0.191	0.222	0.254	0.287	0.311	0.323	0.327	0.328
10		0.158	0.191	0.225	0.259	0.285	0.297	0.302	0.302
20			0.158	0.194	0.230	0.257	0.271	0.275	0.276
30				0.160	0.199	0.228	0.242	0.247	0.247
40					0.164	0.194	0.209	0.215	0.216
50						0.156	0.172	0.178	0.179
60							0.140	0.146	0.147

TABLE VII-13 (*Continued*)

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
	Exposure Rate = 10.00 (WLM/yr)								
0	0.206	0.280	0.344	0.405	0.462	0.500	0.516	0.520	0.521
10		0.207	0.280	0.349	0.414	0.458	0.476	0.482	0.482
20			0.207	0.286	0.359	0.410	0.431	0.438	0.439
30				0.213	0.297	0.355	0.380	0.388	0.389
40					0.220	0.287	0.317	0.326	0.328
50						0.202	0.237	0.249	0.251
60							0.164	0.178	0.180
	Exposure Rate = 20.00 (WLM/yr)								
0	0.280	0.400	0.493	0.571	0.635	0.672	0.684	0.686	0.687
10		0.280	0.401	0.500	0.582	0.629	0.645	0.649	0.649
20			0.281	0.410	0.515	0.576	0.597	0.602	0.603
30				0.290	0.426	0.506	0.535	0.543	0.544
40					0.303	0.410	0.450	0.461	0.463
50						0.271	0.328	0.345	0.347
60							0.202	0.225	0.228

[a] Estimated with the committee's TSE model (Chapter 2) and a multiplicative interaction between smoking and exposure to radon progeny. Note that R_e includes R_0, the calculated lifetime risk for unexposed male smokers, 0.123.

TABLE VII-14 Years of Life Lost, $(L_0 - L_e)$ by Age Started and Age Exposure Ends for Various Rates of Annual Exposure[a] for Male Smokers[b]

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
	Exposure Rate = 0.10 (WLM/yr)								
0	0.02	0.03	0.05	0.06	0.08	0.09	0.10	0.10	0.10
10		0.02	0.03	0.05	0.07	0.08	0.08	0.08	0.08
20			0.02	0.03	0.05	0.06	0.07	0.07	0.07
30				0.02	0.04	0.05	0.05	0.05	0.05
40					0.02	0.03	0.03	0.03	0.03
50						0.01	0.02	0.02	0.02
60							0.00	0.00	0.00
	Exposure Rate = 0.20 (WLM/yr)								
0	0.03	0.06	0.09	0.13	0.16	0.19	0.19	0.19	0.19
10		0.03	0.06	0.10	0.13	0.16	0.16	0.16	0.16
20			0.03	0.07	0.10	0.12	0.13	0.13	0.13
30				0.03	0.07	0.09	0.10	0.10	0.10
40					0.04	0.06	0.07	0.07	0.07
50						0.02	0.03	0.03	0.03
60							0.01	0.01	0.01
	Exposure Rate = 0.50 (WLM/yr)								
0	0.08	0.15	0.23	0.32	0.40	0.46	0.48	0.48	0.48
10		0.08	0.15	0.24	0.33	0.38	0.40	0.41	0.41
20			0.08	0.16	0.25	0.31	0.33	0.33	0.33
30				0.09	0.18	0.23	0.25	0.26	0.25
40					0.09	0.15	0.17	0.17	0.17
50						0.06	0.08	0.08	0.08
60							0.02	0.02	0.02
	Exposure Rate = 1.00 (WLM/yr)								
0	0.15	0.30	0.46	0.62	0.80	0.91	0.94	0.95	0.95
10		0.15	0.31	0.48	0.65	0.76	0.80	0.80	0.80
20			0.16	0.33	0.50	0.61	0.65	0.66	0.66
30				0.17	0.35	0.46	0.50	0.51	0.50
40					0.18	0.29	0.33	0.34	0.34
50						0.12	0.15	0.16	0.16
60							0.04	0.05	0.04
	Exposure Rate = 4.00 (WLM/yr)								
0	0.60	1.18	1.75	2.36	2.97	3.32	3.43	3.45	3.45
10		0.60	1.20	1.83	2.45	2.83	2.94	2.96	2.96
20			0.62	1.27	1.92	2.31	2.43	2.45	2.45
30				0.68	1.35	1.76	1.89	1.92	1.91
40					0.70	1.13	1.27	1.29	1.29
50						0.46	0.60	0.63	0.62
60							0.15	0.18	0.18

TABLE VII-14 (*Continued*)

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
					Exposure Rate = 10.00 (WLM/yr)				
0	1.46	2.80	4.05	5.32	6.48	7.09	7.25	7.28	7.28
10		1.47	2.83	4.21	5.48	6.17	6.35	6.38	6.38
20			1.50	3.00	4.40	5.16	5.37	5.40	5.40
30				1.65	3.18	4.03	4.27	4.31	4.31
40					1.70	2.66	2.93	2.99	2.98
50						1.10	1.42	1.48	1.48
60							0.37	0.44	0.43
					Exposure Rate = 20.00 (WLM/yr)				
0	2.80	5.14	7.15	9.06	10.61	11.28	11.41	11.43	11.43
10		2.80	5.20	7.43	9.26	10.09	10.26	10.28	10.28
20			2.86	5.50	7.68	8.70	8.92	8.96	8.96
30				3.14	5.76	7.03	7.32	7.37	7.37
40					3.21	4.82	5.20	5.27	5.27
50						2.09	2.63	2.72	2.72
60							0.70	0.83	0.83

[a] Estimated with the committee's TSE model (Chapter 2) and a multiplicative interaction between smoking and exposure to radon progeny.

[b] L_0, the calculated lifetime risk for unexposed male smokers, is 69.0 yr.

TABLE VII-15 Ratio of Lifetime Risks (R_e/R_0) by Age Started and Age Exposure Ends for Various Rates of Annual Exposure[a] for Male Nonsmokers[b]

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
					Exposure Rate = 0.10 (WLM/yr)				
0	1.008	1.016	1.024	1.032	1.041	1.049	1.053	1.054	1.055
10		1.008	1.016	1.024	1.033	1.041	1.045	1.047	1.047
20			1.008	1.016	1.026	1.033	1.037	1.039	1.039
30				1.008	1.018	1.025	1.029	1.031	1.031
40					1.009	1.017	1.021	1.022	1.023
50						1.008	1.012	1.013	1.013
60							1.004	1.006	1.006
					Exposure Rate = 0.20 (WLM/yr)				
0	1.016	1.031	1.047	1.064	1.083	1.098	1.106	1.109	1.109
10		1.016	1.032	1.049	1.067	1.082	1.090	1.093	1.094
20			1.016	1.033	1.051	1.066	1.074	1.077	1.078
30				1.017	1.035	1.050	1.058	1.061	1.062
40					1.018	1.033	1.041	1.044	1.045
50						1.015	1.023	1.026	1.027
60							1.008	1.011	1.012
					Exposure Rate = 0.50 (WLM/yr)				
0	1.039	1.079	1.118	1.161	1.206	1.244	1.264	1.271	1.273
10		1.039	1.079	1.121	1.167	1.205	1.225	1.232	1.234
20			1.040	1.082	1.128	1.166	1.185	1.193	1.194
30				1.042	1.088	1.126	1.146	1.153	1.155
40					1.046	1.084	1.104	1.111	1.113
50						1.038	1.058	1.065	1.067
60							1.020	1.028	1.029
					Exposure Rate = 1.00 (WLM/yr)				
0	1.079	1.157	1.237	1.321	1.412	1.487	1.527	1.542	1.545
10		1.079	1.158	1.242	1.334	1.409	1.448	1.464	1.467
20			1.079	1.164	1.256	1.331	1.370	1.385	1.388
30				1.085	1.176	1.251	1.291	1.306	1.309
40					1.092	1.167	1.207	1.222	1.225
50						1.075	1.115	1.130	1.133
60							1.040	1.055	1.058
					Exposure Rate = 4.00 (WLM/yr)				
0	1.314	1.627	1.941	2.274	2.634	2.927	3.081	3.140	3.151
10		1.314	1.630	1.964	2.326	2.621	2.775	2.834	2.846
20			1.317	1.653	2.017	2.313	2.468	2.527	2.539
30				1.337	1.703	2.000	2.156	2.216	2.228
40					1.367	1.666	1.823	1.883	1.895
50						1.301	1.459	1.519	1.531
60							1.159	1.220	1.231

TABLE VII-15 (*Continued*)

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
	Exposure Rate = 10.00 (WLM/yr)								
0	1.783	2.556	3.328	4.140	5.012	5.716	6.080	6.218	6.244
10		1.783	2.564	3.385	4.267	4.979	5.349	5.488	5.515
20			1.790	2.621	3.513	4.234	4.608	4.750	4.777
30				1.840	2.743	3.473	3.852	3.996	4.023
40					1.914	2.653	3.038	3.184	3.212
50						1.749	2.140	2.289	2.317
60							1.396	1.547	1.576
	Exposure Rate = 20.00 (WLM/yr)								
0	2.556	4.077	5.577	7.135	8.789	10.102	10.771	11.018	11.064
10		2.556	4.091	5.687	7.378	8.723	9.410	9.665	9.712
20			2.571	4.204	5.934	7.312	8.017	8.280	8.329
30				2.671	4.442	5.854	6.578	6.849	6.899
40					2.816	4.264	5.009	5.288	5.341
50						2.489	3.257	3.546	3.601
60							1.789	2.087	2.143

[a] Estimated with the committee's TSE model (Chapter 2) and a multiplicative interaction between smoking and exposure to radon progeny.

[b] R_0, the calculated lifetime risk for unexposed male nonsmokers, is 0.0112.

TABLE VII-16 Lifetime Risk (R_e) by Age Started and Age Exposure Ends for Various Rates of Annual Exposure[a] for Male Nonsmokers

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
					Exposure Rate = 0.10 (WLM/yr)				
0	0.011	0.011	0.011	0.012	0.012	0.012	0.012	0.012	0.012
10		0.011	0.011	0.011	0.012	0.012	0.012	0.012	0.012
20			0.011	0.011	0.011	0.012	0.012	0.012	0.012
30				0.011	0.011	0.011	0.012	0.012	0.012
40					0.011	0.011	0.011	0.011	0.011
50						0.011	0.011	0.011	0.011
60							0.011	0.011	0.011
					Exposure Rate = 0.20 (WLM/yr)				
0	0.011	0.012	0.012	0.012	0.012	0.012	0.012	0.012	0.012
10		0.011	0.012	0.012	0.012	0.012	0.012	0.012	0.012
20			0.011	0.012	0.012	0.012	0.012	0.012	0.012
30				0.011	0.012	0.012	0.012	0.012	0.012
40					0.011	0.012	0.012	0.012	0.012
50						0.011	0.011	0.011	0.012
60							0.011	0.011	0.011
					Exposure Rate = 0.50 (WLM/yr)				
0	0.012	0.012	0.013	0.013	0.014	0.014	0.014	0.014	0.014
10		0.012	0.012	0.013	0.013	0.014	0.014	0.014	0.014
20			0.012	0.012	0.013	0.013	0.013	0.013	0.013
30				0.012	0.012	0.013	0.013	0.013	0.013
40					0.012	0.012	0.012	0.012	0.012
50						0.012	0.012	0.012	0.012
60							0.011	0.012	0.012
					Exposure Rate = 1.00 (WLM/yr)				
0	0.012	0.013	0.014	0.015	0.016	0.017	0.017	0.017	0.017
10		0.012	0.013	0.014	0.015	0.016	0.016	0.016	0.016
20			0.012	0.013	0.014	0.015	0.015	0.016	0.016
30				0.012	0.013	0.014	0.014	0.015	0.015
40					0.012	0.013	0.014	0.014	0.014
50						0.012	0.012	0.013	0.013
60							0.012	0.012	0.012
					Exposure Rate = 4.00 (WLM/yr)				
0	0.015	0.018	0.022	0.025	0.030	0.033	0.035	0.035	0.035
10		0.015	0.018	0.022	0.026	0.029	0.031	0.032	0.032
20			0.015	0.019	0.023	0.026	0.028	0.028	0.028
30				0.015	0.019	0.022	0.024	0.025	0.025
40					0.015	0.019	0.020	0.021	0.021
50						0.015	0.016	0.017	0.017
60							0.013	0.014	0.014

TABLE VII-16 (*Continued*)

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
	Exposure Rate = 10.00 (WLM/yr)								
0	0.020	0.029	0.037	0.046	0.056	0.064	0.068	0.070	0.070
10		0.020	0.029	0.038	0.048	0.056	0.060	0.062	0.062
20			0.020	0.029	0.039	0.047	0.052	0.053	0.054
30				0.021	0.031	0.039	0.043	0.045	0.045
40					0.021	0.030	0.034	0.036	0.036
50						0.020	0.024	0.026	0.026
60							0.016	0.017	0.018
	Exposure Rate = 20.00 (WLM/yr)								
0	0.029	0.046	0.062	0.080	0.098	0.113	0.121	0.123	0.124
10		0.029	0.046	0.064	0.083	0.098	0.105	0.108	0.109
20			0.029	0.047	0.066	0.082	0.090	0.093	0.093
30				0.030	0.050	0.066	0.074	0.077	0.077
40					0.032	0.048	0.056	0.059	0.060
50						0.028	0.037	0.040	0.040
60							0.020	0.023	0.024

[a] Estimated with the committee's TSE model (Chapter 2) and a multiplicative interaction between smoking and exposure to radon progeny. Note that R_e includes R_0, the calculated lifetime for unexposed male nonsmokers, 0.0112.

TABLE VII-17 Years of Life Lost, $(L_0 - L_e)$ by Age Started and Age Exposure Ends for Various Rates of Annual Exposure[a] for Male Nonsmokers[b]

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
					Exposure Rate = 0.10 (WLM/yr)				
0	0.00	0.00	0.00	0.01	0.01	0.01	0.01	0.01	0.01
10		0.00	0.00	0.00	0.01	0.01	0.01	0.01	0.01
20			0.00	0.00	0.00	0.01	0.01	0.01	0.01
30				0.00	0.00	0.00	0.00	0.00	0.00
40					0.00	0.00	0.00	0.00	0.00
50						0.00	0.00	0.00	0.00
60							0.00	0.00	0.00
					Exposure Rate = 0.20 (WLM/yr)				
0	0.00	0.01	0.01	0.01	0.02	0.02	0.02	0.02	0.02
10		0.00	0.01	0.01	0.01	0.01	0.02	0.02	0.02
20			0.00	0.01	0.01	0.01	0.01	0.01	0.01
30				0.00	0.01	0.01	0.01	0.01	0.01
40					0.00	0.01	0.01	0.01	0.01
50						0.00	0.00	0.00	0.00
60							0.00	0.00	0.00
					Exposure Rate = 0.50 (WLM/yr)				
0	0.01	0.01	0.02	0.03	0.04	0.04	0.04	0.05	0.05
10		0.01	0.01	0.02	0.03	0.04	0.04	0.04	0.04
20			0.01	0.02	0.02	0.03	0.03	0.03	0.03
30				0.01	0.02	0.02	0.02	0.02	0.02
40					0.01	0.01	0.02	0.02	0.02
50						0.01	0.01	0.01	0.01
60							0.00	0.00	0.00
					Exposure Rate = 1.00 (WLM/yr)				
0	0.01	0.03	0.04	0.06	0.07	0.09	0.09	0.09	0.09
10		0.01	0.03	0.04	0.06	0.07	0.08	0.08	0.08
20			0.01	0.03	0.05	0.06	0.06	0.06	0.06
30				0.02	0.03	0.04	0.05	0.05	0.05
40					0.02	0.03	0.03	0.03	0.03
50						0.01	0.01	0.02	0.02
60							0.00	0.00	0.00
					Exposure Rate = 4.00 (WLM/yr)				
0	0.06	0.11	0.17	0.23	0.30	0.34	0.36	0.36	0.36
10		0.06	0.11	0.18	0.24	0.29	0.30	0.30	0.30
20			0.06	0.12	0.19	0.23	0.24	0.25	0.25
30				0.06	0.13	0.17	0.19	0.19	0.19
40					0.07	0.11	0.12	0.13	0.13
50						0.04	0.06	0.06	0.06
60							0.01	0.02	0.02

TABLE VII-17 (*Continued*)

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
					Exposure Rate = 10.00 (WLM/yr)				
0	0.14	0.28	0.42	0.58	0.74	0.84	0.87	0.88	0.88
10		0.14	0.28	0.44	0.60	0.70	0.74	0.75	0.74
20			0.14	0.30	0.46	0.57	0.60	0.61	0.61
30				0.16	0.32	0.43	0.46	0.47	0.47
40					0.16	0.27	0.31	0.32	0.32
50						0.11	0.15	0.15	0.15
60							0.04	0.05	0.04
					Exposure Rate = 20.00 (WLM/yr)				
0	0.28	0.56	0.83	1.13	1.44	1.64	1.70	1.71	1.71
10		0.28	0.56	0.87	1.18	1.38	1.44	1.46	1.46
20			0.29	0.60	0.91	1.12	1.18	1.20	1.20
30				0.31	0.64	0.84	0.91	0.93	0.93
40					0.33	0.54	0.61	0.63	0.62
50						0.22	0.29	0.30	0.30
60							0.07	0.09	0.09

[a] Estimated with the committee's TSE model (Chapter 2) and a multiplicative interaction between smoking and exposure to radon progeny.

[b] L_0, the calculated lifetime for unexposed male nonsmokers, is 70.5 yr.

TABLE VII-18 Ratio of Lifetime Risks (R_e/R_0) by Age Started and Age Exposure Ends for Various Rates of Annual Exposure[a] for Female Smokers[b]

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
					Exposure Rate = 0.10 (WLM/yr)				
0	1.008	1.016	1.024	1.033	1.043	1.050	1.053	1.055	1.055
10		1.008	1.016	1.025	1.035	1.042	1.045	1.047	1.047
20			1.008	1.017	1.026	1.034	1.037	1.039	1.039
30				1.009	1.018	1.025	1.029	1.030	1.031
40					1.009	1.016	1.020	1.021	1.022
50						1.007	1.011	1.012	1.013
60							1.003	1.005	1.006
					Exposure Rate = 0.20 (WLM/yr)				
0	1.016	1.032	1.049	1.066	1.085	1.099	1.106	1.109	1.110
10		1.016	1.033	1.050	1.069	1.083	1.090	1.093	1.094
20			1.016	1.034	1.053	1.067	1.074	1.077	1.078
30				1.018	1.036	1.051	1.058	1.061	1.062
40					1.019	1.033	1.040	1.043	1.044
50						1.014	1.021	1.024	1.025
60							1.007	1.010	1.011
					Exposure Rate = 0.50 (WLM/yr)				
0	1.040	1.081	1.122	1.166	1.212	1.247	1.264	1.271	1.274
10		1.040	1.081	1.125	1.172	1.207	1.224	1.232	1.234
20			1.041	1.085	1.132	1.167	1.184	1.192	1.194
30				1.044	1.091	1.126	1.143	1.151	1.154
40					1.047	1.082	1.099	1.107	1.110
50						1.035	1.053	1.060	1.063
60							1.017	1.025	1.028
					Exposure Rate = 1.00 (WLM/yr)				
0	1.081	1.161	1.242	1.329	1.421	1.489	1.522	1.537	1.542
10		1.081	1.162	1.250	1.342	1.411	1.444	1.459	1.464
20			1.082	1.170	1.262	1.332	1.366	1.381	1.386
30				1.089	1.181	1.251	1.285	1.300	1.306
40					1.094	1.164	1.198	1.213	1.218
50						1.071	1.105	1.121	1.126
60							1.035	1.050	1.055
					Exposure Rate = 4.00 (WLM/yr)				
0	1.320	1.633	1.944	2.272	2.609	2.857	2.975	3.027	3.044
10		1.320	1.638	1.973	2.319	2.573	2.694	2.747	2.765
20			1.325	1.668	2.021	2.281	2.406	2.460	2.478
30				1.351	1.712	1.979	2.106	2.162	2.181
40					1.371	1.644	1.775	1.833	1.852
50						1.281	1.416	1.475	1.495
60							1.138	1.199	1.219

TABLE VII-18 (*Continued*)

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
	Exposure Rate = 10.00 (WLM/yr)								
0	1.787	2.531	3.244	3.969	4.689	5.197	5.430	5.528	5.559
10		1.788	2.542	3.310	4.071	4.610	4.858	4.963	4.996
20			1.799	2.612	3.418	3.988	4.252	4.365	4.400
30				1.861	2.714	3.320	3.601	3.721	3.759
40					1.908	2.554	2.854	2.984	3.025
50						1.691	2.013	2.153	2.198
60							1.341	1.490	1.538
	Exposure Rate = 20.00 (WLM/yr)								
0	2.530	3.896	5.134	6.323	7.430	8.160	8.474	8.600	8.634
10		2.531	3.917	5.247	6.485	7.305	7.660	7.803	7.843
20			2.553	4.042	5.427	6.347	6.749	6.912	6.959
30				2.670	4.224	5.259	5.714	5.901	5.956
40					2.759	3.934	4.454	4.670	4.734
50						2.346	2.946	3.196	3.272
60							1.672	1.955	2.043

[a] Estimated with the committee's TSE model (Chapter 2) and a multiplicative interaction between smoking and exposure to radon progeny.

[b] R_0, the calculated lifetime risk for unexposed female smokers, is 0.0582.

TABLE VII-19 Lifetime Risk (R_e) by Age Started and Age Exposure Ends for Various Rates of Annual Exposure[a] for Female Smokers

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
	Exposure Rate = 0.10 (WLM/yr)								
0	0.059	0.059	0.060	0.060	0.061	0.061	0.061	0.061	0.061
10		0.059	0.059	0.060	0.060	0.061	0.061	0.061	0.061
20			0.059	0.059	0.060	0.060	0.060	0.060	0.060
30				0.059	0.059	0.060	0.060	0.060	0.060
40					0.059	0.059	0.059	0.059	0.059
50						0.059	0.059	0.059	0.059
60							0.058	0.058	0.059
	Exposure Rate = 0.20 (WLM/yr)								
0	0.059	0.060	0.061	0.062	0.063	0.064	0.064	0.065	0.065
10		0.059	0.060	0.061	0.062	0.063	0.063	0.064	0.064
20			0.059	0.060	0.061	0.062	0.063	0.063	0.063
30				0.059	0.060	0.061	0.062	0.062	0.062
40					0.059	0.060	0.061	0.061	0.061
50						0.059	0.059	0.060	0.060
60							0.059	0.059	0.059
	Exposure Rate = 0.50 (WLM/yr)								
0	0.061	0.063	0.065	0.068	0.071	0.073	0.074	0.074	0.074
10		0.061	0.063	0.066	0.068	0.070	0.071	0.072	0.072
20			0.061	0.063	0.066	0.068	0.069	0.069	0.070
30				0.061	0.064	0.066	0.067	0.067	0.067
40					0.061	0.063	0.064	0.064	0.065
50						0.060	0.061	0.062	0.062
60							0.059	0.060	0.060
	Exposure Rate = 1.00 (WLM/yr)								
0	0.063	0.068	0.072	0.077	0.083	0.087	0.089	0.089	0.090
10		0.063	0.068	0.073	0.078	0.082	0.084	0.085	0.085
20			0.063	0.068	0.073	0.078	0.079	0.080	0.081
30				0.063	0.069	0.073	0.075	0.076	0.076
40					0.064	0.068	0.070	0.071	0.071
50						0.062	0.064	0.065	0.066
60							0.060	0.061	0.061
	Exposure Rate = 4.00 (WLM/yr)								
0	0.077	0.095	0.113	0.132	0.152	0.166	0.173	0.176	0.177
10		0.077	0.095	0.115	0.135	0.150	0.157	0.160	0.161
20			0.077	0.097	0.118	0.133	0.140	0.143	0.144
30				0.079	0.100	0.115	0.123	0.126	0.127
40					0.080	0.096	0.103	0.107	0.108
50						0.075	0.082	0.086	0.087
60							0.066	0.070	0.071

TABLE VII-19 (*Continued*)

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
	Exposure Rate = 10.00 (WLM/yr)								
0	0.104	0.147	0.189	0.231	0.273	0.302	0.316	0.322	0.324
10		0.104	0.148	0.193	0.237	0.268	0.283	0.289	0.291
20			0.105	0.152	0.199	0.232	0.248	0.254	0.256
30				0.108	0.158	0.193	0.210	0.217	0.219
40					0.111	0.149	0.166	0.174	0.176
50						0.098	0.117	0.125	0.128
60							0.078	0.087	0.090
	Exposure Rate = 20.00 (WLM/yr)								
0	0.147	0.227	0.299	0.368	0.432	0.475	0.493	0.501	0.503
10		0.147	0.228	0.305	0.377	0.425	0.446	0.454	0.457
20			0.149	0.235	0.316	0.369	0.393	0.402	0.405
30				0.155	0.246	0.306	0.333	0.343	0.347
40					0.161	0.229	0.259	0.272	0.276
50						0.137	0.171	0.186	0.190
60							0.097	0.114	0.119

[a] Estimated with the committee's TSE model (Chapter 2) and a multiplicative interaction between smoking and exposure to radon progeny. Note that R_i includes R_0, the calculated lifetime risk for unexposed female smokers, 0.0582.

TABLE VII-20 Years of Life Lost ($L_0 - L_e$) by Age Started and Age Exposure Ends for Various Rates of Annual Exposure[a] for Female Smokers[b]

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
					Exposure Rate = 0.10 (WLM/yr)				
0	0.01	0.02	0.03	0.04	0.05	0.06	0.06	0.06	0.06
10		0.01	0.02	0.03	0.04	0.05	0.05	0.05	0.05
20			0.01	0.02	0.03	0.04	0.04	0.04	0.04
30				0.01	0.02	0.03	0.03	0.03	0.03
40					0.01	0.02	0.02	0.02	0.02
50						0.01	0.01	0.01	0.01
60							0.00	0.00	0.00
					Exposure Rate = 0.20 (WLM/yr)				
0	0.02	0.04	0.06	0.08	0.10	0.11	0.12	0.12	0.12
10		0.02	0.04	0.06	0.08	0.09	0.10	0.10	0.10
20			0.02	0.04	0.06	0.08	0.08	0.08	0.08
30				0.02	0.04	0.06	0.06	0.06	0.06
40					0.02	0.03	0.04	0.04	0.04
50						0.01	0.02	0.02	0.02
60							0.00	0.00	0.00
					Exposure Rate = 0.50 (WLM/yr)				
0	0.05	0.09	0.14	0.20	0.25	0.28	0.29	0.29	0.29
10		0.05	0.10	0.15	0.20	0.24	0.24	0.25	0.24
20			0.05	0.10	0.16	0.19	0.20	0.20	0.20
30				0.05	0.11	0.14	0.15	0.15	0.15
40					0.05	0.09	0.09	0.10	0.09
50						0.03	0.04	0.04	0.04
60							0.01	0.01	0.01
					Exposure Rate = 1.00 (WLM/yr)				
0	0.09	0.19	0.28	0.39	0.50	0.56	0.58	0.58	0.58
10		0.09	0.19	0.30	0.41	0.47	0.48	0.49	0.48
20			0.10	0.21	0.31	0.37	0.39	0.39	0.39
30				0.11	0.22	0.28	0.30	0.30	0.29
40					0.11	0.17	0.19	0.19	0.19
50						0.06	0.08	0.08	0.08
60							0.02	0.02	0.02
					Exposure Rate = 4.00 (WLM/yr)				
0	0.37	0.74	1.12	1.53	1.93	2.16	2.22	2.23	2.22
10		0.38	0.76	1.18	1.58	1.81	1.88	1.89	1.88
20			0.39	0.81	1.23	1.46	1.53	1.54	1.53
30				0.43	0.85	1.09	1.16	1.17	1.16
40					0.43	0.67	0.74	0.75	0.74
50						0.25	0.32	0.33	0.32
60							0.07	0.08	0.07

TABLE VII-20 (*Continued*)

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
					Exposure Rate = 10.00 (WLM/yr)				
0	0.93	1.82	2.70	3.65	4.55	5.04	5.17	5.19	5.17
10		0.93	1.85	2.84	3.76	4.28	4.41	4.43	4.41
20			0.96	1.98	2.95	3.48	3.63	3.65	3.63
30				1.07	2.07	2.64	2.78	2.81	2.78
40					1.05	1.64	1.80	1.83	1.80
50						0.62	0.79	0.82	0.79
60							0.17	0.20	0.17
					Exposure Rate = 20.00 (WLM/yr)				
0	1.82	3.50	5.11	6.78	8.26	9.01	9.20	9.23	9.22
10		1.82	3.56	5.35	6.95	7.78	7.99	8.02	8.00
20			1.87	3.81	5.54	6.45	6.68	6.71	6.69
30				2.09	3.97	4.96	5.21	5.26	5.22
40					2.06	3.16	3.44	3.48	3.45
50						1.22	1.54	1.59	1.54
60							0.34	0.40	0.34

[a] Estimated with the committee's TSE model (Chapter 2) and a multiplicative interaction between smoking and exposure to radon progeny.
[b] L_0, the calculated lifetime for unexposed female smokers, is 75.9 yr.

TABLE VII-21 Ratio of Lifetime Risk (R_e/R_0) by Age Started and Age Exposure Ends for Various Rates of Annual Exposure[a] for Female Nonsmokers[b]

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
					Exposure Rate = 0.10 (WLM/yr)				
0	1.008	1.017	1.025	1.034	1.044	1.051	1.055	1.057	1.057
10		1.008	1.017	1.026	1.036	1.043	1.047	1.048	1.049
20			1.008	1.018	1.027	1.035	1.038	1.040	1.040
30				1.009	1.019	1.026	1.030	1.031	1.032
40					1.010	1.017	1.021	1.022	1.023
50						1.007	1.011	1.013	1.013
60							1.004	1.005	1.006
					Exposure Rate = 0.20 (WLM/yr)				
0	1.017	1.033	1.050	1.069	1.088	1.103	1.110	1.113	1.114
10		1.017	1.034	1.052	1.071	1.086	1.093	1.096	1.097
20			1.017	1.035	1.055	1.069	1.076	1.080	1.081
30				1.018	1.038	1.052	1.059	1.063	1.064
40					1.019	1.034	1.041	1.044	1.046
50						1.015	1.022	1.025	1.026
60							1.007	1.010	1.012
					Exposure Rate = 0.50 (WLM/yr)				
0	1.042	1.083	1.126	1.171	1.220	1.256	1.274	1.282	1.285
10		1.042	1.084	1.130	1.178	1.215	1.233	1.241	1.244
20			1.042	1.088	1.136	1.173	1.191	1.199	1.202
30				1.046	1.094	1.131	1.149	1.157	1.160
40					1.048	1.085	1.103	1.111	1.114
50						1.037	1.055	1.063	1.066
60							1.018	1.026	1.029
					Exposure Rate = 1.00 (WLM/yr)				
0	1.083	1.167	1.251	1.343	1.439	1.512	1.548	1.564	1.570
10		1.083	1.168	1.259	1.356	1.429	1.465	1.481	1.487
20			1.085	1.176	1.273	1.346	1.382	1.398	1.403
30				1.091	1.188	1.261	1.297	1.313	1.319
40					1.097	1.170	1.206	1.222	1.228
50						1.073	1.109	1.126	1.131
60							1.036	1.052	1.058
					Exposure Rate = 4.00 (WLM/yr)				
0	1.334	1.666	2.003	2.366	2.748	3.037	3.179	3.243	3.265
10		1.334	1.671	2.035	2.418	2.708	2.850	2.914	2.936
20			1.339	1.703	2.087	2.377	2.520	2.584	2.607
30				1.365	1.750	2.041	2.184	2.249	2.271
40					1.386	1.678	1.821	1.886	1.908
50						1.293	1.436	1.501	1.524
60							1.144	1.209	1.232

TABLE VII-21 (*Continued*)

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
	Exposure Rate = 10.00 (WLM/yr)								
0	1.832	2.660	3.495	4.391	5.332	6.040	6.386	6.542	6.595
10		1.832	2.672	3.573	4.520	5.232	5.580	5.737	5.790
20			1.845	2.751	3.703	4.419	4.770	4.928	4.981
30				1.911	2.869	3.589	3.942	4.101	4.155
40					1.963	2.689	3.044	3.204	3.259
50						1.730	2.088	2.249	2.305
60							1.360	1.522	1.578
	Exposure Rate = 20.00 (WLM/yr)								
0	2.660	4.301	5.948	7.703	9.535	10.904	11.569	11.867	11.967
10		2.660	4.325	6.101	7.954	9.340	10.013	10.315	10.417
20			2.685	4.481	6.355	7.757	8.439	8.745	8.848
30				2.817	4.713	6.132	6.823	7.133	7.238
40					2.920	4.357	5.058	5.372	5.479
50						2.457	3.167	3.486	3.595
60							1.718	2.041	2.152

[a] Estimated with the committee's TSE model (Chapter 2) and a multiplicative interaction between smoking and exposure to radon progeny.

[b] R_0, the calculated lifetime risk for unexposed female nonsmokers, is 0.00602.

TABLE VII-22 Lifetime Risk (R_e) by Age Started and Age Exposure Ends for Various Rates of Annual Exposure[a] for Female Nonsmokers

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
					Exposure Rate = 0.10 (WLM/yr)				
0	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006
10		0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006
20			0.006	0.006	0.006	0.006	0.006	0.006	0.006
30				0.006	0.006	0.006	0.006	0.006	0.006
40					0.006	0.006	0.006	0.006	0.006
50						0.006	0.006	0.006	0.006
60							0.006	0.006	0.006
					Exposure Rate = 0.20 (WLM/yr)				
0	0.006	0.006	0.006	0.006	0.007	0.007	0.007	0.007	0.007
10		0.006	0.006	0.006	0.006	0.007	0.007	0.007	0.007
20			0.006	0.006	0.006	0.006	0.006	0.007	0.007
30				0.006	0.006	0.006	0.006	0.006	0.006
40					0.006	0.006	0.006	0.006	0.006
50						0.006	0.006	0.006	0.006
60							0.006	0.006	0.006
					Exposure Rate = 0.50 (WLM/yr)				
0	0.006	0.007	0.007	0.007	0.007	0.008	0.008	0.008	0.008
10		0.006	0.007	0.007	0.007	0.007	0.007	0.007	0.007
20			0.006	0.007	0.007	0.007	0.007	0.007	0.007
30				0.006	0.007	0.007	0.007	0.007	0.007
40					0.006	0.007	0.007	0.007	0.007
50						0.006	0.006	0.006	0.006
60							0.006	0.006	0.006
					Exposure Rate = 1.00 (WLM/yr)				
0	0.007	0.007	0.008	0.008	0.009	0.009	0.009	0.009	0.009
10		0.007	0.007	0.008	0.008	0.009	0.009	0.009	0.009
20			0.007	0.007	0.008	0.008	0.008	0.008	0.008
30				0.007	0.007	0.008	0.008	0.008	0.008
40					0.007	0.007	0.007	0.007	0.007
50						0.006	0.007	0.007	0.007
60							0.006	0.006	0.006
					Exposure Rate = 4.00 (WLM/yr)				
0	0.008	0.010	0.012	0.014	0.017	0.018	0.019	0.020	0.020
10		0.008	0.010	0.012	0.015	0.016	0.017	0.018	0.018
20			0.008	0.010	0.013	0.014	0.015	0.016	0.016
30				0.008	0.011	0.012	0.013	0.014	0.014
40					0.008	0.010	0.011	0.011	0.011
50						0.008	0.009	0.009	0.009
60							0.007	0.007	0.007

TABLE VII-22 (*Continued*)

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
	Exposure Rate = 10.00 (WLM/yr)								
0	0.011	0.016	0.021	0.026	0.032	0.036	0.038	0.039	0.040
10		0.011	0.016	0.022	0.027	0.032	0.034	0.035	0.035
20			0.011	0.017	0.022	0.027	0.029	0.030	0.030
30				0.012	0.017	0.022	0.024	0.025	0.025
40					0.012	0.016	0.018	0.019	0.020
50						0.010	0.013	0.014	0.014
60							0.008	0.009	0.010
	Exposure Rate = 20.00 (WLM/yr)								
0	0.016	0.026	0.036	0.046	0.057	0.066	0.070	0.071	0.072
10		0.016	0.026	0.037	0.048	0.056	0.060	0.062	0.063
20			0.016	0.027	0.038	0.047	0.051	0.053	0.053
30				0.017	0.028	0.037	0.041	0.043	0.044
40					0.018	0.026	0.030	0.032	0.033
50						0.015	0.019	0.021	0.022
60							0.010	0.012	0.013

[a] Estimated with the committee's TSE model (Chapter 2) and a multiplicative interaction between smoking and exposure to radon progeny. Note that R_r includes R_0, the calculated lifetime risk for unexposed female nonsmokers, 0.00602.

TABLE VII-23 Years of Life Lost ($L_0 - L_e$) by Age Started and Age Exposure Ends for Various Rates of Annual Exposure[a] for Female Nonsmokers[b]

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
	Exposure Rate = 0.10 (WLM/yr)								
0	0.00	0.00	0.00	0.00	0.01	0.01	0.01	0.01	0.01
10		0.00	0.00	0.00	0.00	0.00	0.01	0.01	0.01
20			0.00	0.00	0.00	0.00	0.00	0.00	0.00
30				0.00	0.00	0.00	0.00	0.00	0.00
40					0.00	0.00	0.00	0.00	0.00
50						0.00	0.00	0.00	0.00
60							0.00	0.00	0.00
	Exposure Rate = 0.20 (WLM/yr)								
0	0.00	0.00	0.01	0.01	0.01	0.01	0.01	0.01	0.01
10		0.00	0.00	0.01	0.01	0.01	0.01	0.01	0.01
20			0.00	0.00	0.01	0.01	0.01	0.01	0.01
30				0.00	0.00	0.01	0.01	0.01	0.01
40					0.00	0.00	0.00	0.00	0.00
50						0.00	0.00	0.00	0.00
60							0.00	0.00	0.00
	Exposure Rate = 0.50 (WLM/yr)								
0	0.00	0.01	0.01	0.02	0.03	0.03	0.03	0.03	0.03
10		0.00	0.01	0.02	0.02	0.02	0.03	0.03	0.03
20			0.01	0.01	0.02	0.02	0.02	0.02	0.02
30				0.01	0.01	0.01	0.02	0.02	0.02
40					0.01	0.01	0.01	0.01	0.01
50						0.00	0.00	0.00	0.00
60							0.00	0.00	0.00
	Exposure Rate = 1.00 (WLM/yr)								
0	0.01	0.02	0.03	0.04	0.05	0.06	0.06	0.06	0.06
10		0.01	0.02	0.03	0.04	0.05	0.05	0.05	0.05
20			0.01	0.02	0.03	0.04	0.04	0.04	0.04
30				0.01	0.02	0.03	0.03	0.03	0.03
40					0.01	0.02	0.02	0.02	0.02
50						0.01	0.01	0.01	0.01
60							0.00	0.00	0.00
	Exposure Rate = 4.00 (WLM/yr)								
0	0.04	0.08	0.12	0.16	0.21	0.23	0.24	0.24	0.24
10		0.04	0.08	0.12	0.17	0.19	0.20	0.20	0.20
20			0.04	0.09	0.13	0.16	0.16	0.16	0.16
30				0.05	0.09	0.12	0.12	0.12	0.12
40					0.04	0.07	0.08	0.08	0.08
50						0.03	0.03	0.03	0.03
60							0.01	0.01	0.01

TABLE VII-23 (*Continued*)

Age (yr) Started	Age (yr) Exposure Ends								
	10	20	30	40	50	60	70	80	110
					Exposure Rate = 10.00 (WLM/yr)				
0	0.10	0.20	0.29	0.41	0.52	0.58	0.60	0.60	0.60
10		0.10	0.20	0.31	0.42	0.48	0.50	0.50	0.50
20			0.10	0.21	0.32	0.39	0.41	0.41	0.40
30				0.11	0.22	0.29	0.31	0.31	0.31
40					0.11	0.18	0.20	0.20	0.19
50						0.07	0.08	0.09	0.08
60							0.02	0.02	0.02
					Exposure Rate = 20.00 (WLM/yr)				
0	0.20	0.39	0.59	0.81	1.02	1.15	1.18	1.19	1.18
10		0.19	0.39	0.62	0.83	0.96	0.99	1.00	0.99
20			0.20	0.42	0.64	0.77	0.81	0.81	0.80
30				0.22	0.45	0.57	0.61	0.62	0.61
40					0.22	0.35	0.39	0.39	0.39
50						0.13	0.17	0.17	0.17
60							0.04	0.04	0.03

[a] Estimated with the committee's TSE model (Chapter 2) and a multiplicative interaction between smoking and exposure to radon progeny.

[b] R_0, the calculated lifetime risk for unexposed female nonsmokers, is 76.7 yr.

PART 3. Further Considerations

EFFECTS OF CIGARETTE SMOKING ON THE RESPIRATORY TRACT

Cigarette smoking has well-characterized effects at all levels of the respiratory tract (Table VII-24).[36] Changes in the airways are most relevant for respiratory carcinogenesis. Cigarette smoking produces mucus gland hypertrophy and hyperplasia in the large airways and stimulates mucus production from goblet cells in the small airways. The clinical counterpart of these changes is chronic bronchitis, defined as regnlar sputum production. The bronchial epithelium develops dysplastic and metaplastic changes in smokers. Certain physiological changes accompany these structural abnormalities. Mucociliary clearance, which removes gases and particles from the large airways, is slowed in cigarette smokers. Increased permeability may facilitate passage of inhaled agents across the epithelium.

Proportionately greater central deposition of particles has been demonstrated in the airways of smokers, in comparison with nonsmokers. This deposition pattern may be a consequence of the abnormal small airway function commonly found in smokers. Impaired lung function can be demonstrated in many smokers, and perhaps 10 to 15% of sustained smokers develop disabling chronic airflow obstruction. The resulting physiological impairment leads to an increased respiratory rate for any particular level of activity.

TABLE VII-24 Histologic and Physiologic Changes in the Respiratory Tract, Other than Malignancy, Associated with Cigarette Smoking[36]

Large airways	Mucous gland hypertrophy and hyperplasia
	Dysplasia and metaplasia of epithelial cells
	Increased epithelial permeability
	Impaired mucociliary transport
	Inflammation
Small airways	Goblet cell metaplasia
	Epithelial cell metaplasia
	Increased mucus production
	Inflammation
Lung parenchyma	Fibrosis
	Increased cell numbers
	Altered cell populations
	Altered function of some cells
	Emphysema

In assessing the consequences of combined exposure to cigarette smoke and radon daughters, consideration must be given to these diverse effects of smoking (Table VII-24), as well as to interaction between the two agents in the process of carcinogenesis itself. Smoking-related changes in the lung's structure and function might alter the dose to target cells at any particular level of exposure. In comparison with nonsmokers, dose might be increased in smokers by the greater central deposition, the increased airways permeability, and the slowed mucociliary transport. Dose might be reduced in smokers by mucosal edema and the increased average mucus thickness due to the heightened mucus production in the airways of smokers. A conclusion concerning the net effect of these smoking-related changes on the dosimetry of radon daughters cannot be reached at present. Nevertheless, the effect of radon daughters in the presence of smoking must be interpreted in the context of the changes in lung structure and function, which can be readily demonstrated in many smokers.[36]

In this regard, several pulmonary disease processes resulting from cigarette smoking have been associated with increased lung-cancer risk: chronic bronchitis and chronic obstructive pulmonary disease. By epidemiological convention, chronic bronchitis refers to chronic sputum production. Clinical diagnosis of chronic obstructive pulmonary disease occurs in patients with disabling and irreversible airflow obstruction. At times, clinical diagnoses such as chronic bronchitis, emphysema, and chronic obstructive pulmonary disease may be applied to persons with irreversible airflow obstruction, regardless of other features.

Nevertheless, epidemiological studies show that these diagnoses are associated with increased risk of lung cancer, even with adjustment for cigarette smoking. In an early case-control study, Doll and Hill[13] found that lung-cancer cases yield a history of chronic bronchitis significantly more often than controls. In two subsequent case-control studies, diagnostic terms applied to patients with chronic airflow obstruction were also associated with lung cancer, even with control for cigarette smoking.[33,37] Davis[12] showed that the incidence of lung cancer in patients with chronic obstructive pulmonary disease was higher than expected in comparison with rates in smokers.

Two studies have demonstrated that mucus hypersecretion, as ascertained by a questionnaire, predicts increased lung-cancer occurrence. Rimington[29] determined lung-cancer incidence in male participants who had given information on their smoking habits and sputum production for a radiological screening program. In all categories of cigarette smoking, lung-cancer incidence was higher in those with a history of daily sputum production for 5 yr at the time of enrollment. Peto et al.[25] examined mortality of 2,518 British men during a 20- to 25-yr follow-up period.

Lung-cancer mortality was higher in those with a lower level of lung function and in those with chronic sputum production. The latter association persisted after adjustment for lung-function level and cigarette smoking. The finding of increased lung cancer in persons with underlying respiratory disease and mucus hypersecretion conflicts with the hypothesis that increased mucus production reduces penetration of alpha particles into the tracheobronchial epithelium and thus protects against cellular damage.[3]

THE ASSOCIATION BETWEEN LUNG CANCER, SMOKING, AND RADIATION

Exposure to radon progeny and cigarette consumption are each associated with lung cancer in a complex way. Because there are only a few studies on the combined effects of radiation exposure and tobacco smoke, the amount of information for their interaction is limited. The committee's analyses described in Chapter 2 and Annex 2A show that cancer risk associated with exposure to radon progeny depends on cumulative dose, age, and time since exposure. The actual biological relationship is undoubtedly more complex than the statistical model that the committee has developed and may be influenced by other factors that cannot be fully evaluated with the available data. These factors might include age at first exposure, dose rate, sex, diet, and genetic predisposition. Moreover, the association of tobacco consumption with lung cancer is also complex and depends on duration and number of cigarettes smoked per day, type of tobacco product, method of inhalation, and years since cessation of use for former smokers.[35] Assessment of the combined effects of cigarette smoking and radon progeny should account for the individual patterns of effect from both insults. Other aspects of the combined exposure may also be important, for example, the effect of the sequencing of exposures and the degree of their overlap in time.

In contrast, the studies of combined exposures, reported in the literature or analyzed by this committee in Part 2 of this appendix, have usually considered only cumulative WLM (or duration of employment or other surrogate) and duration or intensity of cigarette use, and not the effects of the other variables described above. Such assessments of the underlying relationship may be distorted by not accounting for other predictors of risk. Nevertheless, risk models are a useful method for describing patterns in the different data sets. With these complexities in mind, the data currently available on radon daughters and tobacco exposure suggest that risks do not combine additively on the relative-risk scale. Although there is great uncertainty regarding the relative impact of the two exposures, the multiplicative model appears to have greater support in the literature. The analyses by this committee suggest that a submultiplicative model

should not be dismissed and may provide a more accurate description of the underlying relationship.

A clear pattern of risk among studies of miner's exposure to radon and tobacco smoke has not yet emerged. A few small studies have shown mixed results, while the largest study of the issue by Whittemore and McMillan[38] indicates a multiplicative interaction. While the committee's analyses of the Colorado Plateau uranium miners in Part 2 of this appendix support this conclusion, the analyses also support submultiplicative and supramultiplicative relationships.

The committee's analysis of the Japanese atomic-bomb survivor data shows tbat for these data, neither an additive nor a multiplicative model can be rejected on statistical grounds; indeed, their maximum likelihoods are nearly identical. This is consistent witb the results of Prentice et al.[26] In summary, the atomic-bomb survivor data appear amenable to either a multiplicative or additive model for the relative risk. The most recent case-control study by Blot et al.[6] based on a large number of lung-cancer cases sustains this interpretation. The relevance of these studies of atomic-bomb survivors to the interaction of radon and smoking in their relationship to lung-cancer induction, however, must still be determined.

Our review suggests that tbis issue has yet to be resolved. Areas for further study that are needed to clarify the combined effect of these two exposures include the following:

- the impact of smoking rate (cigarettes per day) and smoking duration, as opposed to rate and/or the combined pack-years, on the radiation association with lung cancer;
- implications of low- versus high-LET radiation;
- the role of smoking cessation on the effect of radiation-associated lung cancer;
- the effect on interactions of tobacco use before and after radiation exposure;
- the role of cigarette use on the histological distribution of radiation-associated lung cancer;
- the relationship of smoking to other measures of radiation exposure, for example, working-level rate, cumulative WLM, and duration of exposure; and
- the role of other agents associated with lung diseases, such as asbestos, silica, and arsenic.

REFERENCES

1. Archer, V. E., J. K. Wagoner, and F. E. Lundin. 1973. Uranium mining and cigarette smoking effects on man. J. Occup. Med. 15:204–211.

2. Axelson, O. 1983. Experiences and concerns on lung cancer and radon daughter exposure in mines and dwellings in Sweden. Z. Erkrank. Atm.-Org. 161:232–239.
3. Axelson, O., and L. Sundell. 1978. Mining lung cancer and smoking. Scand. J. Work Environ. Health 4:46–52.
4. Band, P., M. Feldstein, G. Saccomanno, L. Watson, and O. King. 1980. Potentation of cigarette smoking and radiation: Evidence from a sputum cytology survey among uranium miners and controls. Cancer 45:1273–1277.
5. Beebe, W. G., and M. Usagawa. 1968. The major ABCC samples. Pp. 12–68 in Atomic Bomb Casualty Commission Technical Report. Hiroshima, Japan: Atomic Bomb Casualty Commission.
6. Blot, W. J., S. Akiba, and H. Kato. 1984. Ionizing radiation and lung cancer: A review including preliminary results from a case-control study among A-bomb survivors. In Atomic Bomb Survivor Data: Utilization and Analysis, R. L. Prentice and D. J. Thompson, eds. Philadelphia: Society for Industrial and Applied Mathematics.
7. Breslow, N. E., and B. E. Storer. 1985. General relative risk functions for case control studies. Am. J. Epidemiol. 122:149–162.
8. Cederlof, R., L. Friberg, Z. Hrubec, and U. Lorich. 1975. The relationship of smoking and some covariables to mortality and cancer morbidity. Report of the Department of Environmental Hygiene. Stockholm: Karolinska Institute.
9. Cox, D. R. 1972. Regression models and life tables (with discussion). J. R. Stat. Soc. Ser. B 34:187–220.
10. Damber, L., and L. G. Larsson. 1982. Combined effects of mining and smoking in the causation of lung carcinoma. Acta Radiol. Oncol. 21:305–313.
11. Damber, L., and L. G. Larsson. 1985. Underground mining, smoking, and lung cancer: A case-control study in the iron ore municipalities in Northern Sweden. J. Natl. Cancer Inst. 74:1207–1213.
12. Davis, A. L. 1976. Bronchogenic carcinoma in chronic obstructive pulmonary disease. J. Am. Med. Assoc. 235:612–622.
13. Doll, R., and A. B. Hill. 1952. A study of the aetiology of carcinoma of the lung. Br. Med. J. 2:1271–1286.
14. Edling, C., H. Kling, and O. Axelson. 1984. Radon in homes—a possible cause of lung cancer. Scand. J. Work Environ. Health 10:25–34.
15. Hornung, R. W., and S. Samuels. 1981. Survivorship models for lung cancer mortality in uranium miners—is cumulative dose an appropriate measure of exposure? Pp. 363–368 in Radiation Hazards in Mining: Control, Measurement and Medical Aspects, M. Gomez, ed. New York: Society of Mining Engineers of the American Institute of Mining Metallurgical and Petroleum Engineers.
16. Ishimaru, T., R. W. Cihak, C. E. Land, A. Steer, and A. Yamada. 1975. Lung cancer at autopsy in A-bomb survivors and controls, Hiroshima and Nagasaki, 1961–1970. II. Smoking, occupation and A-bomb exposure. Cancer 36:1723–1728.
17. Jorgensen, H. S. 1979. A study of mortality from lung cancer among miners in Kiruna 1950–1970. Scand. J. Work. Environ. Health 10:126–133.
18. Kato, H., K. G. Johnson, and K. Yano. 1966. Mail survey of cardiovascular disease study, Hiroshima and Nagasaki. Pp. 19–66 in Atomic Bomb Casualty Commission Technical Report. Hiroshima, Japan: Atomic Bomb Casualty Commission.
19. Kopecky, K. J., T. Yamamoto, T. Fujikura, S. Tokuoka, T. Monzen, I. Nishimari, E. Nakashima, and H. Kato. In press. Lung cancer, radiation exposure and smoking among A-bomb survivors, Hiroshima and Nagasaki, 1950–1980. In press. Techincal Report. Hiroshima, Japan: Radiation Effects Research Foundation.

20. Liddell, F. D. K., J. C. McDonald, and D. C. Thomas. 1977. Methods of cohort analysis: Appraisals by application to asbestos mining. J. R. Stat. Soc. Ser. A 140:469–491.
21. Lubin, J. H., and M. H. Gail. 1984. Biased selection of controls for case-control analyses of cohort studies. Biometrics 40:63–75.
22. Lundin, F. E., Jr., V. E. Archer, and J. K. Wagoner. 1979. An exposure-time-response model for lung cancer mortality in uranium miners—effects of radiation exposure, age and cigarette smoking. Pp. 243–264 in Proceedings of the Work Group at the Second Conference of the Society for Industrial and Applied Mathematics, N. E. Breslow, and A. Whittemore, eds.
23. Morgan, M.V. and J. M. Samet. 1986. Radon daughter exposures of New Mexico U miners, 1967–1982. Health Phys. 50:656–662.
24. National Institute for Occupational Safety and Health (NIOSH). 1986. Evaluation of Epidemiologic Studies Examining the Lung Cancer Mortality of Underground Miners. Division of Standards Development and Technology Transfer. Cincinnati, Ohio: Centers for Disease Control, National Institute for Occupational Safety and Health.
25. Peto, R., F. E. Speizer, A. L. Cochrane, F. Moore, C. M. Fletcher, C. M. Tinker, I. T. Higgins, R. G. Gray, S. M. Richards, J. Gilliland and B. Norman-Smith. 1983. The relevance in adults of air-flow obstruction, but not mucus hypersecretion to mortality from chronic lung disease: Results from 20 years of prospective observation. Am. Rev. Respir. Dis. 128:491–501.
26. Prentice, R. L., Y. Yoshimoto, and M. Mason. 1983. Relationship of cigarette smoking and radiation exposure to cancer mortality in Hiroshima and Nagasaki. J. Natl. Cancer Inst. 70:611–622.
27. Radford, E. P., and K. G. St. Clair Renard. 1984. Lung cancer in Swedish iron ore miners exposed to low doses of radon daughters. N. Engl. J. Med. 310(23):1485–1494.
28. Renard, K. G. St. Clair. 1974. Respiratory cancer mortality in an iron ore mine in Northern Sweden. Ambio. 3:67–69.
29. Rimington, J. 1971. Smoking chronic bronchitis and lung cancer. Br. Med. J. 2:373–375.
30. Rokvanor i Sverige. 1965. Report of the Central Statistical Bureau. Stockholm: Central Statistical Bureau.
31. Saccomanno, G., V. E. Archer, O. Auerbach, R. P. Saunders, and L. M. Brennan. 1974. Development of carcinoma of the lung as reflected in exfoliated cells. Cancer 33:256–270.
32. Saccomanno, G., C. Yale, W. Dixon, O. Auerbach, and G. C. Huth. 1986. An epidemiological analysis of the relationship between exposure to Rn progeny, smoking and bronchogenic carcinoma in the U-mining population of the Colorado Plateau—1960–1980. Health Phys. 50:605–618.
33. Samet J. M., C. G. Humble, and D. R. Pathok. 1986. Personal and family history of respiratory disease and lung cancer risk. Am. Rev. Respir. Dis. 134:466–470.
34. Thomas, D. C. 1981. General relative risk models for survival time in matched case-control analysis. Biometrics 37:673–686.
35. U.S. Department of Health and Human Services, U.S. Public Health Services Office of Smoking and Health. 1972. The Health Consequences of Smoking: Cancer. A report to the Surgeon General. Department of Health and Human Services (PHS) 82-50179 Washington D.C.: U.S. Government Printing Office.
36. U.S. Department of Health and Human Services, U.S. Public Health Service, Office of Smoking and Health. 1985. The Health Consequences of Smoking:

Chronic Obstructive Lung Disease. A Report to the Surgeon General. PHS 85-50207. Washington, D.C.: U.S. Government Printing Office.

37. Van der Wal, A.M., E. Huizingav, N.G. Orie, H.S. Sluiter, and K.de Vries. 1966. Cancer and chronic nonspecific lung disease (C.N.S.L.D.). Scand. J. Respir. Dis. 47:161–172.

38. Whittemore, A. S., and A. McMillan. 1983. Lung cancer mortality among U.S. uranium miners: A reappraisal. J. Natl. Cancer Inst. 71:489–499.

APPENDIX VIII

Previous Estimates of the Risk Due to Radon Progeny

Several expert groups and individual investigators have published estimates of the risk associated with exposure to radon progeny. In this appendix the committee examines some of the more widely cited studies both for their underlying assumptions and for the numerical value of the estimated risk.

Like the committee's lifetime risk estimates developed in Chapter 2, two steps are usually involved in estimating the risks from radon exposure: the development of an appropriate risk coefficient from epidemiological studies, and the projection of risks over a defined exposure and follow-up periods. Table VIII-1 lists risk coefficients developed in a number of epidemiological studies. Two types of risk coefficients are shown; those for absolute excess risk, the number of cases per person-years at risk per working-level month (WLM), and the excess relative risk, the proportional increase per 100 WLM. Estimates from Annex 2A, using a constant relative risk model are included in Table VIII-1 in cases in which the same cohorts were considered by this committee. Except for the Malmberget miners, the results of the Poisson regressions for internal and external controls used in Annex 2 are not too different from those obtained by other investigators using standardized mortality ratios.

As important as the risk coefficients are in estimating the risks associated with radon exposures, the assumptions in the projection models often have a larger numerical impact. The committee examines these assumptions for particular studies in the following sections.

TABLE VIII-1 Published Risk Coefficients for Exposure of Underground Miners to Radon Progeny

Cohort	Study	Attributable Excess Risk Deaths/10^4 Person-Year at Risk/WLM	Excess Relative Risk/100 WLM	Basis of Risk Estimate
Colorado Plateau	BEIR III[11]	3.5	0.45	Group average, 0–3,719 WLM
	Whittemore and McMillan[19]	6.0	0.8	Group Average, 0–360 WLM
			0.31	Regression, nonsmoker
			1.44	Regression, 20-pack year smoker
	NIOSH[4]		1.1	Proportional hazard regression at 120 WLM
	Annex 2A		0.6–0.6	Regression on exposures, $<2{,}000$ WLM[a]
Czechoslovakia	BEIR III[11]	19	1.8	Group average 0–300 WLM
Ontario	Muller[8]	7.2	1.3	Regression, standard WLM
		2.8	0.51	Regression, special WLM[b]
	Annex 2A		1.4–1.2	Regression, standard WLM[a]
Beaverlodge	Howe[3]	20.9	3.28	Regression
	Annex 2A		2.6–2.6	Regression[a]
Malmberget	Radford and Renard[12]	21	—	Group average for smokers
	Annex 2A	16.9	—	Group average for nonsmokers
		19	3.6	Group average
			1.4–1.6	Regression[a]
Newfoundland	BEIR III[11]	17.7	8.0	Group average
	Morrison et al.[7]	5.6	—	Regression

[a] Tables 2A-2 and 2A-3 for internal and external controls, respectively, using a constant relative risk model, not the time since exposure model recommended by this Committee.

[b] Maximum estimated exposure, see Appendix IV.

NCRP REPORT 78

Risk estimation in a 1984 report[10] by the National Council on Radiation Protection and Measurements (NCRP) relies on the Harley and Pasternack Model B of lung-cancer excess due to radon progeny.[2] The following assumptions formed the basis of the model.

- Following a latent period, the tumor rate is an exponentially decreasing function of the time since exposure.
- Disease rate excess associated with a single exposure increases with age at exposure.
- Lung cancer is rare before tbe age of 40 yr.
- Median age at lung cancer among miners is about 60 yr in nonsmokers and 50 yr or older in smokers.
- The minimal time for tumor growth, from initial cell transformation to clinical detection, is 5 yr.

From these postulated disease patterns, the Harley and Pasternack model specifies a 5-yr latent period for persons first exposed at the age of 35 yr or older and a $(40 - u)$ yr latent period for persons under the age of 35 yr, where u is age at first exposure. For a single annual exposure at age u, the excess radiation-associated risk above background at age $t > u$ (and $t > 40$) is taken to be

$$A(t, u) = Re^{-m(t-u)}S(t)/S(u),$$

where R is the attributable-risk coefficient per WLM, $S(t)$ and $S(u)$ are the probabilities of survival to the designated age, and m is the rate of removal of transformed stem cells due to repair or cell death. For risk projection, the NCRP task group fixed $m = \ln(2)/20\ \text{yr}^{-1}$, corresponding to a 20-yr half-life. For ages within the latent period or before initial exposure, the excess risk is zero. The exponential term allows for the excess risk to decline with time following exposure, and the survival ratio adjusts for competing causes of mortality. Given the parameters of this model, one integrates over t from age 40 to maximal assumed life (age 85) to obtain lifetime risk due to the single exposure at age u, or over years of exposure, $u_1, \ldots, u_n$, to obtain the excess risk at t due to all previous exposure. Lifetime excess risk from all exposures is the integral over t and u.

This model is extremely important, in that it postulates a modified effect with time since exposure. In this way, it is related to the TSE model recommended in Chapter 2 of this report and the latency models of Lundin et al.[6] and Thomas and McNeill,[16] all of which contrast with a relative-risk model constant in age at risk. Indeed, the distinction between a constant-relative-risk model and models that modify risk according to

time since exposure is more fundamental than discrimination among the latter types, which offer refinements in basically similar models.

The analysis presented in Annex 2A clearly suggests that risk effects are modulated by time since exposure. This is manifest in the declining parameter estimates of impact of exposures more distant in time. Therefore, the distinction between the Harley and Pasternack model and a relative-risk model that declines with time since exposure is related to the rate of decline in the relative risk. In light of the complexity of risk arising from chronic radiation exposure, substantial data would be required for an adequate evaluation of such subtle patterns of risk. An informal method of considering this issue is to examine additive excess risk after cessation of exposure. This committee's analysis indicated that the relative risk declines with time since cessation of exposure. However, the NCRP risk model requires that this decline be large enough for the attributable risk to decrease.

To test this hypothesis, data on observed and expected cancers and person-years of exposure from the four miner cohorts analyzed in Annex 2A were categorized by age, age at last exposure, and cumulative WLM. Figure VIII-1 presents for each of the four data sets age-specific attributable risks, (observed − expected)/person-years, for three age-at-last-exposure groups. In the figures, the excess risks were smoothed by graphing the mean of the observed excess and two adjacent values and weighting by the inverse variance. Data from these four worker populations do not show a consistent pattern of declining excess risk. In several cohorts, the excess risks generally increase; in others, the excess declines, but only 20 yr or more after the mean age at last exposure. The NCRP model would predict a declining excess shortly after cessation of exposure.

Patterns similar to those shown in Figure VIII-1 were observed after stratification by two categories of cumulative WLM. In addition, Poisson regression models were fit to the observed risk, where the attributable disease rate was postulated to be linear in age at last exposure and cumulative WLM. For each data set, after adjustment for WLM and age at last exposure, there was no significant improvement in model fit with the inclusion of age at risk. Parameter estimates for five age categories tended to increase, as suggested by Figure VIII-1. However, this effect is poorly estimated. Model fit did not improve significantly with inclusion of a continuous age variate, although the coefficients were generally positive.

A difficulty in the application of the NCRP model is the choice of m, the rate of removal of the transformed cell. Harley and Pasternack acknowledge the issue and select a 20-yr half-life as "representative for extrapolation," although they cite no formal data analysis or experimental results. Additional work in this area would be beneficial for refining the model.

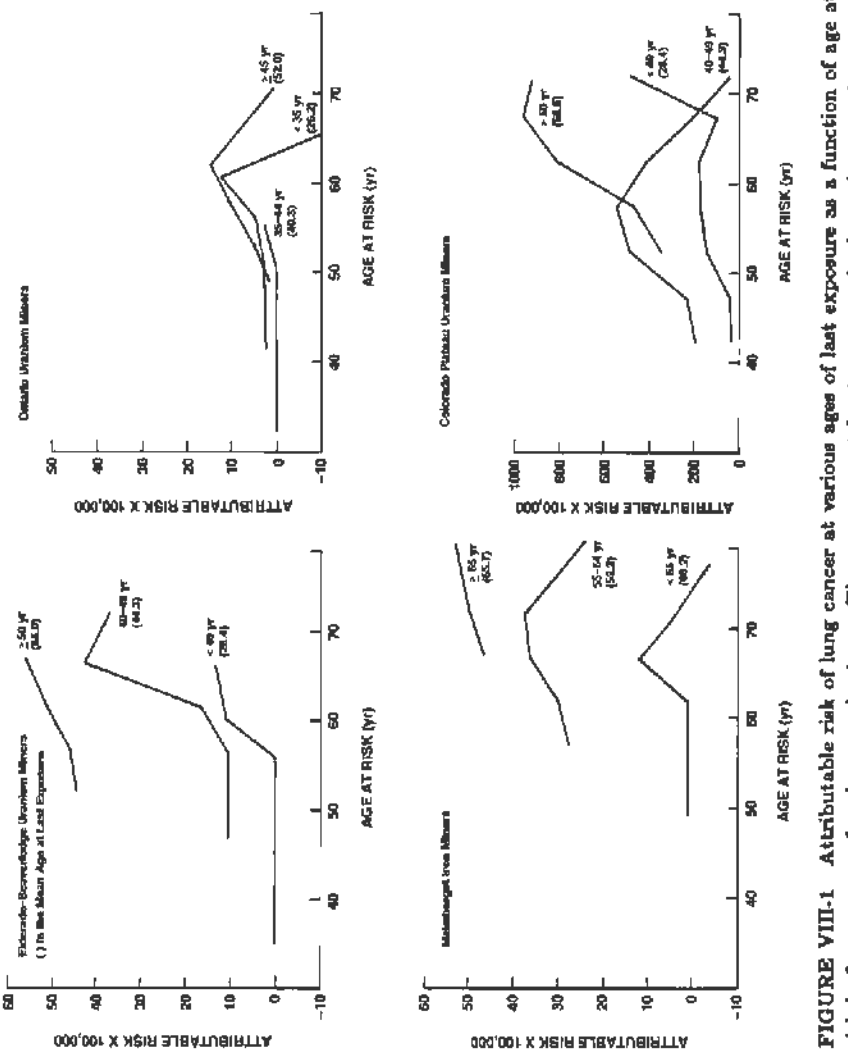


FIGURE VIII-1 Attributable risk of lung cancer at various ages of last exposure as a function of age at risk in four groups of underground miners. The mean age at least exposure is shown in parentheses.

Although it does not have much impact on NCRP lifetime risk estimates, their model limits the occurrence of radiation-induced lung cancer to the age of 40 yr and over, a restriction for which no biological mechanism is readily apparent. In contrast, several studies have observed lung cancers under the age of 40.[7,15] The failure to observe lung cancer in young persons in several other studies could be due to the very low background rates and few person-years. For example, Radford and Renard[12] reported that the mean age at first exposure of the Swedish miners was 28 yr. With a 5-yr latent period, 1,415 miners would accrue a maximum of some 10,000 person-yr by the age of 40, producing 0.5 expected cases if the population lung-cancer mortality rate for ages 35–39 were 5.1×10^{-5}. With this expected value, there is a 0.6 probability that no cases will occur before the age of 40.

A methodological issue concerns the manner in which the attributable risk is adjusted for competing causes of death. As defined, $S(t)$ is the probability that a person who is subject to disease rates of the standard population will survive to age t. For a 1-yr exposure at age $u < t$, the competing-cause adjustment $S(t)/S(u)$—which is the probability of survival of someone in the standard population to t, given survival to age u—does not incorporate the increased lung-cancer risk, and thus decreased survival, of someone exposed. This adjustment error is compounded as risk is integrated over age t and over yearly exposures but is unlikely to be important except at high dose rates.

BEIR III REPORT

The National Research Council's Committee on the Biological Effects of Ionizing Radiations (BEIR III)[11] assumed a linear relationship between exposure in WLM and the additive excess risk of lung cancer. The excess risk was estimated to vary with age at diagnosis, as shown in Table VIII-2. In addition to the minimal age at expression (similar to that in the NCRP model), a minimal latent period of 15–20 yr (for those exposed at age 15–34) or 10 yr (for those exposed above age 34) is assumed. Later risk is independent of latent period.

These risk values were based on the combined estimates from the epidemiology studies of U.S.[6] and Czechoslovakian[13] uranium miners, Swedish iron miners,[12] and Newfoundland fluorspar miners.[7] The techniques for combining the epidemiological data were not described and so cannot be evaluated. It appears that substantial weight was given to the results from the Swedish, Newfoundland, and Czechoslovakian miner surveys. The Colorado Plateau uranium miners had much lower lung-cancer risks, which the committee thought was due to their high dose rates. The Swedish metal miners had a higher risk, even with less prevalent cigarette smoking;

TABLE VIII-2 Excess Risk Estimated To Vary with Age and Diagnosis

Age (yr) at Diagnosis	Excess Cases (10^6 person-years at risk per WLM)
<35	0
35-49	10
50-65	20
>65	50

that difference was attributed to longer follow-up. No data are available to indicate whether these risk estimates apply to childhood irradiation.

The BEIR III report[11] discussed, but did not resolve, the effect of cigarette smoking on these radiation risks. The BEIR III report states that if the two exposures are additive, their risk estimates would apply to both smokers and nonsmokers. But if there is a multiplicative interaction (i.e., the lung-cancer risk estimates due to radiation are proportional to the smoking-specific rates), the estimates should be increased by 50% for smokers and reduced by a factor of 6 for nonsmokers.

REPORT OF THOMAS AND MCNEILL

The report of Thomas and McNeill and co-workers[16,17] reviewed epidemiological and animal data on lung cancer, bone and head sarcomas, and some other cancers, with an emphasis on lung cancer from radon progeny. To develop risk estimates, the authors considered data from the Czechoslovakian, Ontarian, and Colorado Plateau uranium miners; the Newfoundland fluorspar miners; the Swedish metal miners; and (for inferences regarding the shape of the dose-response curve, but not the magnitude of risk) the Japanese atomic-bomb survivors. Animal data were used primarily to investigate the effect of modifying factors, as opposed to estimation of magnitude of risk.

The comprehensive report reached qualitative and quantitative conclusions largely in accord with those in Chapter 2. Thomas and McNeill discussed at length the epidemiological and statistical principles underlying selection of a risk model (i.e., relative risk versus additive excess risk), the shape of the dose-response curve, and the role of modifying and confounding factors. We support and have repeated their approach of formally combining evidence from various cohorts. This committee concurs with their argument that simply comparing risk estimates from different cohorts in relation to average exposure of the cohorts is not suitable for studying the shape of the dose-response curve. Thomas and McNeill used a more statistically sound method; that is, they fit a single model to a

combination of data sets. They allowed the degree of risk to vary among studies, so that they could adjust for varied confounding factors, but incorporated parameters common to the data sets to model nonlinearities in dose-response relationships. The primary limitation of this analysis, as acknowledged, was the very limited form of the data that could be extracted from published reports concerning the various coborts.

Thomas and McNeill adopted a model with the relative-risk constant in age and, tentatively, linear in cumulative exposure, except at very high values. Their analysis indicated an estimated value of 2.28/100 WLM for the excess relative risk. In selecting this estimate, they discounted a substantially lower risk among the Colorado Plateau miners; and, to some extent, by using a cell-killing model, they compensated for the lower risks per unit exposure at very high levels of cumulative exposure. Inclusion of an exponential term to represent cell killing resulted in a final model that was nonlinear in dose; however, the decrease in slope caused by this cell-killing term was important only at very high doses. However, this allowance for a decrease in slope at very high exposures was statistically significant. They also considered models in which excess relative risk was proportional to an estimated power of dose; such models provide for a more general nonlinearity in dose. The fitted model, although not providing a statistically significant improvement over a simple linear model, resulted in a convex dose-response function, that is, a generally (but only slightly) decreasing slope of the response with increasing cumulative exposure. As noted above, however, they felt that a linear dose-response relationship at moderate to low doses was adequate for extrapolation, with data from very high doses discounted via the cell-killing model. Their interpretation of the possible curvilinearity was primarily that one should be less confident that low-dose extrapolations are conservative than in the case of low linear energy transfer (LET) radiation, where the curvilinearity is generally held to be of the opposite type (slope increasing with dose).

Although we emphasize that their conclusions are in accord with those drawn in this report, we believe that the adoption of a constant-relative-risk model at all ages for the effect of radon daughters is not well supported. The data available to Thomas and McNeill on this issue were sparse. The most relevant evidence was presented in Section 7.2.1 of their 1982 report,[16] where they argued that, with the meager data available, the additive excess risk increases substantially with age, at a given dose, whereas the relative risk is more stable. In Section 4.2.1.3 of the same report,[16] they attempted to discriminate between the "attributable-risk" (i.e., excess-risk) and relative-risk models, solely on the basis of the total (or average) risk over age (and time). This attempt may have been inappropriate, because information on age-specific risks was not available to Thomas and McNeill.

The present committee was fortunate to have access to much more detailed data on some populations and can confirm to some extent the conclusions drawn by Thomas and McNeill. Their average risk coefficient (2.28/100 WLM) is not very different from that found by this committee (1.5/100 WLM) using external controls and constant-relative-risk model. The difference is largely due to their exclusion of results from the Colorado Plateau cohort, which the committee's analysis includes.

The tentative conclusion of Thomas and McNeill regarding the linearity of the dose-response relationship was supported by the data available to them. We agree with the statistical approach that they used, and for two reasons concur with the tentativeness of their conclusion as to the shape of the dose-response curve. First, at very large doses, there is a suggestion of nonlinearity in specific cohorts, although it is not consistent enough among all the cohorts to be statistically significant. More important, there cannot be enough evidence from epidemiological studies to ascertain the effects at low doses.

On the critical issue of the interaction of cigarette smoking and radiation effects, Thomas and McNeill concluded[16] that the joint effect seemed to be "intermediate between additive and multiplicative, although on balance [they] would favor the multiplicative model." The evidence for this was moderately weak, inasmuch as the effects of other modifying factors—such as age at exposure, exposure rate, and time since cessation of exposure—were not controlled.

In conclusion, the reports of Thomas and McNeill[16,17] provide a strong discussion of principles and methods, but are limited by the data available to them. The present report is complementary in its approach, but more data were accessible to the committee. These were the data from the four cohorts in Eldorado-Beaverlodge, Ontario, Colorado, and Sweden described in Annex 2A. Although we disagree with the claim made in Thomas and McNeill's Appendix J[16] that grouping of doses tends to result in underestimation of risks, the general consistency of conclusions, both qualitative and quantitative, between the two reports is notable.

1981 REPORT OF EVANS ET AL.

In a brief report in the journal *Nature* in 1981, Evans et al.[1] provided an upper bound to the lifetime lung-cancer risk associated with radon-daughter exposure in the general population. The report originated in an international workshop on radiation protection principles for naturally occurring radionuclides. The authors primarily considered the epidemiological evidence in determining the risks of environmental radon. They cited a range of lifetime attributable-risk coefficients, developed by other authors, of 21–54 to 1,000 deaths/10^6 WLM. In their collective judgment,

the "most defensible upper bound of the lifetime risk to the general population is 100 lung cancer deaths per 10^6 WLM." This coefficient reflects a reduction in unit exposure for the general population, in comparison with miners, because of differing exposure conditions, smoking habits, and age and sex distributions of the two populations.

Evans et al. acknowledged the informality of their approach for determining a risk coefficient for the general population. They did not use models directly, either to derive a risk coefficient from the miner data or to extrapolate from miners to the general population. They also assumed an attributable-risk model and did not specifically address the effects of cigarette smoking.

1977 UNSCEAR REPORT

The 1977 report of the U.N. Scientific Committee on the Effects of Atomic Radiation (UNSCEAR)[18] provided an attributable-risk coefficient for lung-cancer incidence of 200–450/10^6 WLM, which described a full, for example, 40-yr, expression of the carcinogenic effect on lung tissue of radon and of its daughter products. The report reviewed data from American uranium miners, Swedish underground miners, Newfoundland fluorspar miners, iron-ore miners in the United Kingdom, and Czechoslovakian uranium miners. The upper bound of the attributable-risk range was clearly derived from analysis of the Czechoslovakian data; the derivation of the lower limit is unclear, although the Swedish data reported by Snihs[14] apparently were considered. The Colorado Plateau data do not appear to have been used in setting the range.

The UNSCEAR report emphasised the Czechoslovakian study, because of long latency after the onset of exposure and the availability of appropriate mortality rates. The authors cited the dose-response relationship of excess risk to exposure as 230×10^{-6}/WLM; this coefficient, however, was taken from the 1976 report[13] that was based on an incorrect method of analysis. To obtain the upper bound of 450×10^{-6}/WLM, the authors merely doubled the value reported by Sevc et al.[19] That calculation was justified by assuming that the average follow-up in the Czechoslovakian study (20 yr) represented the median latency for a 40-yr complete expression of the effects of exposure. The report did not provide evidence to support the biological model that is implicit in the doubling of the risk coefficient.

The Swedish data were also characterized as appropriate for consideration, although the original report by Snihs[14] did not provide complete information. The present committee does not regard these data as adequate for risk estimation. For a 40-yr period, Snihs estimated the attributable

risk as 140×10^{-6}/WLM, on the basis of the Swedish data. The derivation of the lower bound of 200×10^{-6}/WLM from this value was not described. The report did not make firm statements about the effects of cigarette smoking.

ICRP PUBLICATION 32

Publication 32 by the International Commission on Radiological Protection (ICRP)[5] published in 1981, provided a recommended limit for inhalation of radon progeny by workers. In developing this limit, ICRP considered both the epidemiological evidence and the results of a dosimetric analysis. This committee has focused on ICRP's epidemiological approach.

The ICRP group emphasized the findings of the Colorado Plateau and Czechoslovakian studies. Relying on reports from those studies and on the 1977 UNSCEAR[18] and 1980 BEIR III[11] reviews, it cited a range of attributable risk of 2–20 cases/10^6 person-yr/WLM. Because the effect of exposure was noted to vary with age at exposure, the group considered 5–15 cases/10^6 person-yr/WLM as "the most probable range," on the basis of averaging "over all age periods during occupational work." Over "a mean manifestation period of 30 years," the group translated the attributable-risk range of 5–15 cases/10^6 person-yr/WLM into a total lifetime risk of 1.5–4.5 excess cases/WLM. With adjustment for the higher breathing rate of miners, the excess risks were reduced by about 20%. The ICRP group noted that the risks for miners might be increased by the effects of other exposures and thus tend to overestimate the effects of radon daughters alone.

This committee could not fully critique ICRP's epidemiological approach, because some procedures were not fully described: the derivation of the range of 2–20 cases/10^6 person-yr/WLM, the averaging that reduced this range to 5–15 cases/10^6 person-yr/WLM, and the rationale for the 30-yr period for calculating lifetime risk. As discussed elsewhere, this committee finds a modified relative-risk model to be preferable to the attributable-risk model used by ICRP in 1981.[5]

SUMMARY

The descriptions of risk estimates given above make it clear that a number of approaches have been applied to estimating the risks due to radon-daughter exposure. Some are based largely on expert opinion, while others depend on analyses of limited data on lung-cancer cases associated with exposure to radon progeny. Results vary, as indicated in Table VIII-1 above and Table 2-13 in Chapter 2. There are at least three underlying causes for this lack of agreement between risk estimates.

1. As discussed in Chapter 2 and Appendix IV, there is a fair amount of variability between the results of the individual epidemiological studies. Although these differences are perhaps no greater than would be anticipated on statistical grounds, it is not unreasonable to believe that other factors enter as well. Since some risks estimators put greater weight on one set(s) of observations than another, differences between risk estimates are not surprising.

2. A variety of techniques must be used to project lifetime risk to a general population on the basis of relatively short-term occupational exposures to underground miners, a topic discussed at length in Chapter 2. Foremost among these is the modeling of age-specific lung-cancer risk. Risk projections which use models based on the relative risk depend critically on the age-specific background rates. As discussed in Chapter 2, differences in estimated lifetime risks occur if the relative risk is constant or if it is permitted to vary with time-related factors. Similarly, lifetime risks that are derived from models of additive excess risk depend on the modeling of time-related effects. The different models will produce approximately the same average risk for populations with similar age structure and follow-up such as the underground miners. However, projecting beyond the range of the miner cohort data can produce very different numerical estimates.

3. Finally, several of the risk projections described above seem to depend more on considerations of biological plausibility rather than data analyses by standard methods. Some investigators might perhaps argue that biological plausibility should be the main criteria for risk projections, but others are less sure. Lung cancers observed in the miner studies are largely due to two complete carcinogens, smoking and high-LET radiation, whose joint interaction is not well defined. The committee believes that until underlying processes of carcinogenesis are understood, an objective analysis of observational data is a surer path to valid estimates of radon risks.

REFERENCES

1. Evans, R. D., J. H. Harley, W. Jacobi, H. S. McLean, W. A. Mills, and C. G. Stewart. 1981. Estimate of risk from environmental exposure to Rn-222 and its decay products. Nature 290:98–100.
2. Harley, N. H., and B. S. Pasternack. 1981. A model for predicting lung cancer risks induced by environmental levels of radon daughters. Health Phys. 40:307–316.
3. Howe G. R., R. C. Nair, H. G. Hewcombe, A. B. Miller, and J. D. Abbett. Lung cancer mortality (1950–1980) in relation to radon daughter exposure in a cohort of workers in the Eldorado Beaverlodge uranium mine. J. Natl. Cancer Inst. 77(2):357–362.

4. Hornung, R. W., and T. J. Meinhardt. 1987. Quantitative risk assessment of lung cancer mortality in U.S. uranium miners. Health Phys. 52:417–430.
5. International Commission on Radiological Protection (ICRP). 1981. P. 24 in Limits for Inhalation of Radon Daughters by Workers. ICRP Publication 32. Oxford: Pergamon.
6. Lundin F. E., J. K. Wagoner, and V. E. Archer. Radon Daughter Exposure and Respiratory Cancer Quantitative and Temporal Aspects. 1971. Joint Monograph No. 1. Washington, D.C.: U.S. Public Health Service.
7. Morrison H. I., D. T. Wigle, and A. J. deVilliers. 1981. Lung cancer mortality and radiation exposure among the Newfoundland fluorspar miners. Pp. 372–376 in Proceedings of the International Conference on Radiation Hazards in Mining: Control, Measurements, and Medical Aspects, M. Gomez, ed. New York: Society of Mining Engineers of the American Institute of Mining, Metallurgical, and Petroleum Engineers, Inc.
8. Morrison, H. I., R. M. Semenciw, Y. Mao, D. A. Corkill, A. B. Dory, A. J. deVilliers, A. J. Stocker, and D. T. Wigle. 1985. Lung cancer mortality and radiation exposure among the Newfoundland fluorspar miners. Pp. 365–368 in Proceedings of the International Conference on Occupational Radiation Safety in Mining, E. Stocker, ed. Toronto: Canadian Nuclear Association.
9. Muller J., W. C. Wheeler, J. F. Gentleman, G. Suranyi, and R. A. Kusiak. 1985. Study of mortality of Ontario miners. Pp. 335–343 in Proceedings of the International Conference on Occupational Radiation Safety in Mining, A. Stocker, ed. Toronto: Canadian Nuclear Association.
10. National Council on Radiation Protection and Measurements (NCRP). 1984. Evaluation of Occupational and Environmental Exposure to Radon and Radon Daughters. NCRP Report 78. Washington, D.C.: National Council on Radiation Protection and Measurements. 204 pp.
11. National Research Council, Committee on the Biological Effects of Ionizing Radiations (BEIR). 1980. The Effects on Populations of Exposure to Low Levels of Ionizing Radiation. Washington, D.C.: National Academy Press. 524 pp.
12. Radford, E. P., and K. G. St. Clair Renard. 1984. Lung cancer in Swedish iron miners exposed to low doses of radon daughters. N. Engl. J. Med. 310:1485–1494.
13. Sevc, J., E. Kung, and V. Placek. 1976. Lung cancer in uranium miners and long-term exposure to radon daughter products. Health Phys. 30:433–437.
14. Snihs, J. O. 1973. The approach to Rn problems in non-uranium mines in Sweden. Pp. 900–911 in Proceedings of Third International Congress of the IRPA. CONF-730907-P2. Technical Information Center, U.S. Department of Energy. Oak Ridge, Tenn.: Oak Ridge Natural Laboratory.
15. Sun S., X. Yang, Y. Lan, M. Xionyu, L. Shengen, and Y. Zhanyun. 1984. Latent period and temporal aspects of lung cancer among miners. Radiat. Prot. 4(5) (English translation).
16. Thomas, D. C., and K. G. McNeill. 1982. P. 23 in Risk Estimates for the Health Effects of Alpha Radiation. INFO-0081. Ottawa, Canada: Atomic Energy Control Board.
17. Thomas, D. C., K. G. McNeill, and C. Dougherty. 1985. Estimates of lifetime lung cancer risks resulting from Rn progeny exposure. Health Phys. 49:835–846.
18. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCLEAR). 1977. P. 725 in Sources and Effects of Ionizing Radiation. Report E.77.IX.1., New York: United Nations.
19. Whittemore, A. S., and A. McMillan. 1983. Lung cancer mortality among U.S. uranium miners: A reappraisal. J. Natl. Cancer Inst. 71:489–499.

Glossary

Absorbed dose. The mean energy imparted to the irradiated medium, per unit mass, by ionizing radiation. Units: gray (Gy), rad.

Activity. The mean number of decays per unit time of a radioactive nuclide. Units: becquerel (Bq), curie (Ci).

Activity median aerodynamic diameter (AMAD). The diameter of a unit-density sphere with the same terminal settling velocity in air as that of the aerosol particulate whose activity is the median for the entire aerosol.

Additive interaction model. This model is used to find the combined risk for risk factors which have no interaction with each other. For example, the combined mortality risk of cigarette smoking and automobile accidents is the sum of the separate risks.

Adenosarcoma. A mixed tumor which consists of a substance like embroyonic connective tissue together with glandular elements.

Alpha particle. Two neutrons and two protons bound as a single particle that is emitted from the nucleus of certain radioactive isotopes in the process of decay or disintegration.

Aneuploid. Having numbers of chromosomes not equal to exact multiples of the haploid number. Down syndrome is an example.

Background radiation. Radiation arising from radioactive material other than that under consideration; background radiation due to cosmic rays and natural radioactivity is always present; there may also be background radiation due to the presence of radioactive substances in building material.

Bayesian analysis. Analysis in which Bayes' theorem is used to derive posterior probabilities from assumed prior knowledge together with observational data. For example, biological information on the relationship between species and hazardous substances can be combined with data on interspecies dose response to calculate the response of human populations.

Becquerel (Bq). SI unit of activity. (*See* Units.)

Bremsstrahlung. The production of electromagnetic radiation (photons) by the acceleration (positive or negative) that a fast, charged particle (usually an electron) undergoes from the effect of an electric or magnetic field; for instance, from the field of another charged particle (usually a nucleus).

Bronchioles. The small branches of the tracheobronchial tree of the lung.

Cell culture. The growing of cells in vitro, in such a manner that the cells are no longer organized into tissues.

Chromosomal nondisjunction. Either a gain or a loss of chromosomes that occurs when cell division leading to either egg or sperm production goes awry. This results in aneuploidy.

Ciliated mucosa. The mucous membrane in the lung covered with small hairlike structures which serve to move the mucus.

Competing risks. Other causes of death which affect the value of the risk being studied. Persons dying from other causes are not at risk of dying from the factor in question.

Constant-relative-risk model. A risk model which assumes that, after a certain time, the ratio of the risk at a specific dose to the risk in the absence of the dose does not change with time.

Contact inhibition. The cessation of migratory activity and sometimes other functions, including mitosis, when adjacent cells establish firm contact.

Cox proportional hazards model. A relative-risk model that permits the use of internal comparison groups as controls for confounding variables such as cigarette smoking and age.

Curie (Ci). A unit of activity equal to 3.7×10^{10} disintegrations/s. (*See* Units.)

Daughter product. An isotope formed as a result of radioactive decay. One daughter atom is formed for each particle emitted.

Decay chain or decay series. A sequence of radioactive decays of the same nucleus. An initial nucleus, the parent, decays into a daughter nucleus that differs from the first by whatever particles were emitted during the decay. If further decays take place, the subsequent nuclei are also usually called daughters. Sometimes, to distinguish the sequence, the daughter of the first daughter is called the granddaughter, etc.; ordinarily, however, this quickly becomes too complicated.

Diffusion. The random path followed by very small particles due to the impact of surrounding molecules. This Brownian motion governs the transport of submicrometer-size particles in air.

Dominant mutation. The mutation is dominant if it produces its effect in the presence of an equivalent normal gene from the other parent.

Dose-distribution factor. A factor which accounts for modification of the dose effectiveness in cases in which the radionuclide distribution is nonuniform.

Dose equivalent. A quantity that expresses, for the purposes of radiation protection and control, the assumed effectiveness of dose on a common scale for all kinds of ionizing radiation. SI unit is the Sievert. (*See* Units.)

Doubling dose. The amount of radiation needed to double the natural incidence of a genetic or somatic anomaly.

Electron volt (eV). A unit of energy = 1.6×10^{-12} ergs = 1.6×10^{-19} J; 1 eV is equivalent to the energy gained by an electron in passing through a potential difference of 1 V; 1 keV = 1,000 eV; 1 MeV = 1,000,000 eV.

Epithelium. A membranous cellular tissue that covers the surface of some organ or part of the body.

Equilibrium fraction. In equilibrium, the parents and daughters have equal radioactivity, that is, as many decay into a specific nuclide as decay out. When fresh radon enters a volume, the daughter products have not yet accumulated, and there is disequilibrium. The working-level definition of radon does not take into account the amount of equilibrium.

Equilibrium, radioactive secular. The condition in which the activities of a parent and daughter in a radioactive decay chain are (very nearly) equal.

Equilibrium, radiation. The condition in a radiation field where the energy of the radiations entering a volume equals the energy of the radiations leaving that volume.

Euploid. Having uniform exact multiples of the haploid number of chromosomes.

Gamma ray. Short-wavelength electromagnetic radiation of nuclear origin (range of energy, 10 keV to 9 MeV).

Gray (Gy). SI unit of absorbed dose. (*See* Units.)

Half-life, biologic. Time required for the body to eliminate half of an administered dose of any substance by regular processes of elimination; it is approximately the same for both stable and radioactive isotopes of a particular element.

Half-life, radioactive. Time required for a radioactive substance to lose 50% of its activity by decay.

Impaction. As air is taken into the lung, it follows a tortuous path, changing direction many times. At each change of direction, the momentum of the particles carried in the airstream causes them to impact on the bifurcations of the lung. The force on the particle causing it to move and impact on the lung surface is the Stokes force, which is proportional to the velocity of the air moving with respect to the particle. Impaction is important for particles with large aerodynamic diameters.

Incidence. The rate of occurrence of a disease within a specified period; usually expressed in number of cases per 100,000 persons per year.

Ionization. The process by which a neutral atom or molecule acquires a positive or negative charge.

Ionization density. Number of ion pairs per unit volume.

Ionization path (track). The trail of ion pairs produced by ionizing radiation in its passage through matter.

Isotopes. Nuclides that have the same number of protons in their nuclei, and hence the same atomic number, but that differ in the number of enutrons, and therefore in the mass number; chemical properties of osotopes of a particular element are almost identical. The term should not beused as a synonym for nuclide.

Latent period. The period of time between exposure and expression of the disease. After exposure to a dose of radiation, there is a delay of several years (the latent period) before any cancers are seen.

Life-span study (LSS). Life-span study of the Japanese atomic-bomb survivors; the sample consists of 120,000 persons, of whom 82,000 were exposed to the bombs, mostly at low doses.

Lifetime risk. The lifetime probability of dying of a specific disease.

Lifetime risk ratio. The ratio of the lifetime risk (R_e) of an exposed person to the lifetime risk of an unexposed person (R_0). This number minus 1 is the proportional increased risk associated with exposure ($R_e - R_0$).

Linear dose model. This model postulates that the excess risk is linearly proportional to the dose.

Linear energy transfer (LET). Average amount of energy lost per unit track length.

Low LET. Radiation characteristic of electrons, x rays, and gamma rays; the distance between ionizing events is large on the scale of a cellular nucleus.

High LET. Radiation characteristic of protons and fast neutrons; the distance between ionizing events is small on the scale of a cellular nucleus. Average LET is specified to even out the effect of a particle that is slowing down near the end of its path and to allow for the fact that secondary particles are not all of the same energy.

Lymphosarcoma. A sarcoma of the lymphoid tissue. This does not include Hodgkin's disease.

Minute volume. The amount of air moving through the lung per minute; the product of the breathing rate times the volume of air per breath.

Multiplicative interaction model. A model in which independent risk factors interact so that the combined risk is the product of the relative risks due to each factor alone.

Neoplasms. Any new and abnormal growth, such as a tumor; *neoplastic disease* refers to any disease that forms tumors, whether malignant or benign.

Nonstochastic. Describes effects whose severity is a function of dose; for these, a threshold may occur; some nonstochastic somatic

effects are cataract induction, nonmalignant damage to skin, hematological deficiencies, and impairment of fertility.

Nuclide. A species of atom characterized by the constitution of its nucleus, which is specified by its atomic mass and atomic number (Z), or by its number of protons (Z), number of neutrons (N), and energy content.

Oncogenes. Genes which carry the potential for cancer.

Person-gray. Unit of population exposure obtained by summing individual dose-equivalent values for all people in the exposed population. Thus, the number of person-grays contributed by 1 person exposed to 1 Gy is equal to that contributed by 100,000 people each exposed to 10 μGy.

Person-years-at-risk (PYAR). The number of persons exposed times the number of years after exposure minus some lag period during which the dose is assumed to be unexpressed (latent period).

Prevalence. The number of cases of a disease in existence at a given time per unit population, usually 100,000 persons.

Progeny. The decay products resulting after a series of radioactive decays. Progeny can also be radioactive, and the chain continues until a stable nuclide is formed.

Quadratic-dose model. A model which assumes that the excess risk is proportional to the square of the dose.

Quality factor (Q). A linear energy transfer dependent factor by which absorbed doses are multiplied to obtain (for radiation-protection purposes) a quantity which corresponds more closely to the degree of biological effect produced by x or low-energy gamma rays.

Rad. A unit of absorbed dose. Replaced by the gray in SI units. (*See* Units.)

Radioactivity. The property of some nuclides of spontaneously emitting particles or gamma radiation, emitting x radiation after orbital electron capture, or undergoing spontaneous fission.

Artificial radioactivity. Man-made radioactivity produced by fission, fusion, particle bombardment, or electromagnetic irradiation.

Natural radioactivity. The property of radioactivity exhibited by more than 50 naturally occurring radionuclides.

Radioisotopes. A radioactive atomic species of an element with the same atomic number and usually identical chemical properties.

Radionuclide. A radioactive species of an atom characterized by the constitution of its nucleus.

Radiosensitivity. Relative susceptibility of cells, tissues, organs, and organisms to the injurious action of radiation; radiosensitivity and its antonym, radioresistance, are used in a comparative sense rather than an absolute one.

Recessive gene disorder. This requires that a pair of genes, one from each parent, be present in order for the disease to be manifest. An example is cystic fibrosis.

Relative biological effectiveness (RBE). Biological potency of one radiation as compared with another to produce the same biological endpoint. It is numerically equal to the inverse of the ratio of absorbed doses of the two radiations required to produce equal biological effect. The reference radiation is often 200-kV x rays.

Relative mutation risk. The ratio of the risk of a genetic mutation among the exposed population to that in the absence of exposure.

Risk coefficient. The increase in the annual incidence or mortality rate per unit dose: (1) absolute risk coefficient is the observed minus the expected number of cases per person year at risk for a unit dose; (2) the relative-risk coefficient is the fractional increase in the baseline incidence or mortality rate for a unit dose.

Risk estimate. The number of cases (or deaths) that are projected to occur in a specified exposed population per unit dose for a defined exposure regime and expression period: number of cases per person-Gray or, for radon, the number of cases per person cumulative working-level month.

Rem. A unit of dose equivalent. Replaced by the sievert. (*See* Units.)

Sedimentation. The gravitational force on a particle is partially balanced by the viscous force of the air. The resultant velocity toward the earth is the sedimentation velocity. Important for particles with intermediate aerodynamic diameters.

Sex-linked mutation (or X-linked). A mutation associated with the X chromosome. It will usually only manifests its effect in males (who have only a single X chromosome).

SI units. The International System of Units as defined by the General Conference of Weights and Measures in 1960. These units are generally based on the meter/kilogram/second units, with special quantities for radiation including the becquerel, gray, and sievert.

Sievert. The SI unit of radiation dose equivalent. It is equal to dose in grays times a quality factor times other modifying factors, for example, a distribution factor; 1 sievert equals 100 rem.

Specific activity. Total activity of a given nuclide per gram of a compound, element, or radioactive nuclide.

Specific energy. The actual energy per unit mass deposited per unit volume in a given event. This is a stochastic quantity as opposed to the average value over a large number of instances (i.e., the absorbed dose).

Squamous cell carcinoma. A cancer composed of cells that are scaly or platelike.

Standard mortality ratio (SMR). Standard mortality ratio is the ratio of the disease or accident mortality rate in a certain specific population compared with that in a standard population. The ratio is based on 100 for the standard so that an SMR of 200 means that the test population has twice the mortality from that particular cause of death.

Stochastic. Describes random events leading to effects whose probability of occurrence in an exposed population (rather than severity in an affected individual) is a direct function of dose; these effects are commonly regarded as having no threshold; hereditary effects are regarded as being stochastic; some somatic effects, especially carcinogenesis, are regarded as being stochastic.

Stopping power. The average rate of energy loss of a charged particle per unit thickness of a material or per unit mass of material traversed.

Straggling. The statistical variation in the range of a particle caused by the large number of interactions and scatterings within the material being traversed.

Surface-seeking radionuclide. An internal emitter that is deposited and remains on the surface of bone for a long period of time. This contrasts with a volume seeker, which deposits more uniformly throughout the bone volume.

Target theory (hit theory). A theory explaining some biological effects of radiation on the basis that ionization, which occurrs in a discrete volume (the target) within the cell, directly causes a lesion that later results in a physiological response to the damage at that location; one, two, or more hits (ionizing events within the target) may be necessary to elicit the response.

Threshold hypothesis. The assumption that no radiation injury occurs below a specified dose.

Time-since-exposure (TSE) model. A model in which the relative risk is not constant but varies with the time after exposure.

Transformed cells. Tissue culture cells changed in vitro from growing in an orderly pattern and exhibiting contact inhibition to growing in a pattern more like that of cancer cells, resulting in the loss of contact inhibition.

Translocation. A chromosome aberration resulting from chromosome breakage and subsequent structural rearrangement of tbe parts between the same or different chromosomes.

Tumorigenicity. Ability of cells to proliferate into tumors wben inoculated into a specified host organism under specified conditions.

Unattached fraction. That fraction of the radon daughters, usually ^{218}Po (Radium A), which has not yet attached to a particle. As a free atom, it has a high probability of being retained within the lung and depositing alpha energy when it decays.

Units[a]	Conversion Factors
Becquerel (SI)	1 disintegration/s = 2.7×10^{-11} Ci
Curie	3.7×10^{10} disintegrations/s = 3.7×10^{10} Bq
Gray (SI)	1 J/kg = 100 rad
Rad	100 erg/g = 0.01 Gy
Rem	0.01 Sievert
Sievert (SI)	100 rem

[a]International Units are designated (SI).

Working level (WL). Any combination of short lived radon daughters in 1 liter of air that will result in the ultimate emission 1.3×10^5 MeV of potential alpha energy. This number was chosen because it is approximately the alpha energy released from the decay of daughters in equilibrium with 100 picocuries of ^{222}Ra.

Working-level month (WLM). Exposure resulting from inhalation of air with a concentration of 1 working level of radon daughters for 170 working hours.

Years of life lost. The expected years of life for a nonexposed person minus the expected lifetime of an exposed person.

Index

A

B

C

D

E

M

N

P

R

S

T

U

Z

EXHIBIT A-23

NORM/TENORM Information Sheet

OHIO DEPARTMENT OF HEALTH

NORM/TENORM INFORMATION SHEET

OhioEPA
Ohio Environmental Protection Agency

Ohio
Department of Health

OHIO DEPARTMENT OF HEALTH

Naturally Occurring Radioactive Material (NORM)
Technologically Enhanced Naturally Occurring Radioactive Material (TENORM)
INFORMATION SHEET

Naturally Occurring Radioactive Material (NORM) is radioactive material present in the environment; (i.e., soils, air and water) that is not man-made. NORM such as uranium (U), radium (Ra), and thorium (Th) emit low levels of naturally occurring radiation.

FAQs

What is NORM?

NORM, by definition, is naturally occurring and can be found everywhere. Since these materials are found in the natural environment, NORM is exempted from regulation by the U.S. Department of Energy, the U.S. Nuclear Regulatory Commission, and the State of Ohio.

Common examples of NORM include the following:

- radon gas that homeowners may detect in basements and living spaces;
- potassium-40 in all plants and animals, including humans;
- krypton-80 that is in the atmosphere and air we breathe;
- carbon-14 that is taken in by all organic matter and can be measured thousands of years later to determine its age in the process known as "carbon dating";
- uranium and thorium and their decay products commonly used in stone work, including granite countertops used in residential kitchens; marble used for cemetery markers, statues, and building veneers; and granite and limestone walls used in buildings;
- radium in deep drinking water aquifers that causes additional burdens to water treatment plants; and
- uranium ***while it is still in the ground,*** before it is mined and processed into fuel rods for use in nuclear reactors.

What is TENORM?

When NORM is used for commercial purposes, processed, separated, or in some other manner has its radioactivity concentrated (intentionally or unintentionally), it becomes another category of radioactive material called Technologically Enhanced Naturally Occurring Radioactive Material (TENORM), which is regulated by the Ohio Department of Health. TENORM is the same group of NORM radionuclides, but it has been modified or "technologically enhanced" resulting in a man-made concentration higher than NORM.

Common examples of TENORM include the following:

- phosphate industry wastes including phosphogypsum and slag;
- phosphate fertilizers that are commonly used;
- coal industry wastes including fly ash, bottom ash and slag;
- oil and gas industry wastes including scale and sludges;
- water treatment plant wastes including sludges and resin filtration systems;
- metal mining and processing industry wastes including rare earths, zirconium, hafnium, titanium, and tin;
- large volume industries including copper and iron; and
- geothermal energy production wastes.

Where can I find Ohio TENORM regulations?

ODH TENORM rules were finalized in April 2012 and can be viewed at the following web link:
http://www.odh.ohio.gov/en/rules/final/3701_1-43-TENORM.aspx

How are Oil & Gas drilling-related TENORM wastes regulated in Ohio?

The state of Ohio has some of the most restrictive regulations in the country regarding TENORM.

- Ohio does not allow hydraulic fracturing water, flow back water, produced water, or other liquid wastes defined as brine to be used as drinking water. Since brine may contain elevated levels of NORM and other chemical constituents, Ohio requires this material to be sent to a permitted underground injection control-well where it can be safely disposed underground and not come into contact with drinking water supplies or wells.
- Oil & gas drilling-related waste, other than brine, that is TENORM must be tested before leaving the well pad to determine the concentration of radium-226 and radium-228.
- ***Wastes containing TENORM cannot be disposed of at an oil and gas drill site.***
- Solid waste landfills can only accept TENORM wastes for disposal at concentrations less than 5 picocuries per gram above natural background.
- If a solid waste landfill or other facility wants to dilute TENORM wastes with concentrations greater than or equal to 5 picocuries per gram above natural background prior to disposal, this activity requires authorization from the Ohio Department of Health.

If solid wastes cannot be managed at a solid waste landfill because of elevated levels of TENORM, the waste must be sent to a low-level radioactive waste disposal facility.

Where can you find NORM & TENORM in the Oil & Gas Drilling Process?

The radioactive material categories found in Oil & Gas exploration and production (E&P) associated with drilling and hydraulic fracturing include the following:

(1) Exploration: preparing for production (includes drilling, hydraulic fracturing, produced fluids):

- "Earthen material" from the drilling process – **NORM**
- "Earthen material" with residual coating of refined-oil based muds - **NORM**
- Recycled drilling mud – **TENORM**
- Recycled hydraulic fracturing water / flow back water including some Brine (likely concentrated) – **TENORM**
- Spent tank bottoms - **TENORM**
- Filtrate, either liquid or solid, that results from, created during, processing and/or recycling of used hydraulic fracturing water, flow back water, or produced water – **TENORM**
- Used hydraulic fracturing sands – **TENORM**

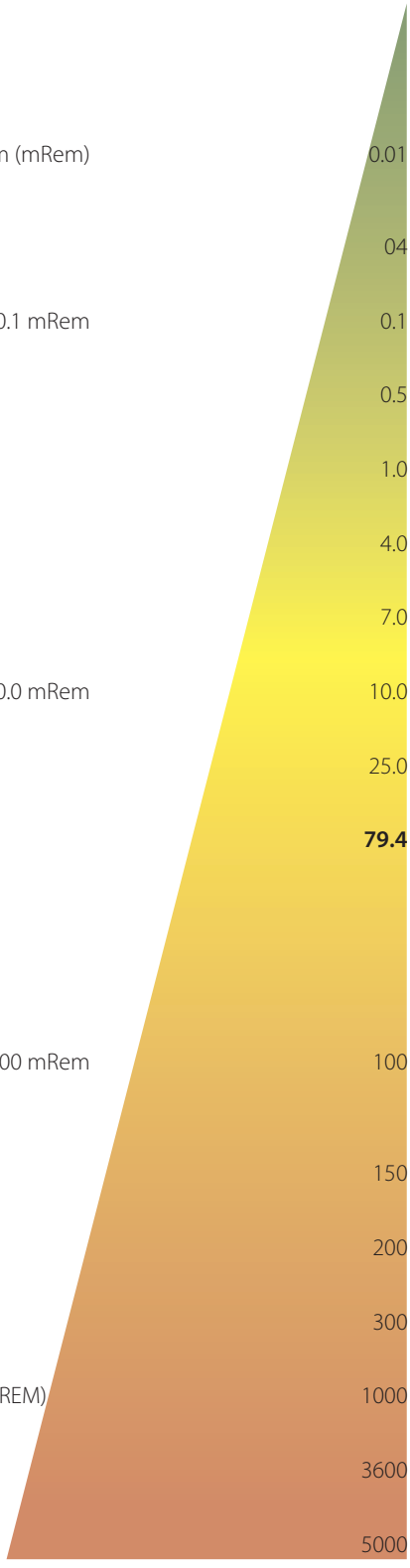
(2) Production: pumping out gas, oil, and brine (a separation station is used to separate the gas, oil, and brine.)

- Gas
- Oil
- Brine – **NORM**
- Pipe scale (buildup) - **TENORM**

What are the relative risks from radiation exposures to the Public?

Radiation exposures from TENORM will vary based on individual activities. The relative exposures from TENORM are low compared to the risks from other sources of radiation. See exposure comparisons below.

Comparison of Radiation Doses



0.01 millirem (mRem)	0.01	Eating a banana
	04	Natural radiation in the human body
0.1 mRem	0.1	Using a cathode ray tube monitor for a year
	0.5	Dental x-ray
	1.0	Background dose received by an average person on an average day
	4.0	Flight from New York to Los Angeles
	7.0	Living in a stone, brick or concrete building for a year
10.0 mRem	10.0	Chest x-ray
	25.0	Release limit for a nuclear power plant for a year
	79.4	**Annual radiation dose received by a resident growing their own food and living in a home built on land with a five meter thick layer of topsoil containing a concentration of combined Radium-226 and Radium-228 equal to 5 pCi/g, excluding natural background.[1]**
100 mRem	100	ODH yearly limit on radiation exposure to a member of the public resulting from licensed radioactive material activities
	150	Spinal x-ray
	200	Natural background radiation a human body is exposed to per year
	300	Mammogram
1000 mRem (1000 mRem = 1 REM)	1000	Average CT scan
	3600	Smoking 1.5 pack of cigarettes a day for a year
	5000	Maximum annual dose permitted for US radiation workers

[1]The 79.4 mRem value was generated using the U.S. NRC's RESRAD 6.5 software program developed by the U.S. Department of Energy's Argonne National Laboratory. The "Resident Farmer" scenario that was used is the most conservative model and will produce the highest radiological dose potential.

TENORM concentrations ≥ 5 pico-Curies per gram (5pCi/g) are not allowed in Ohio landfills.
REM (Roentgen Equivalent Man) is the standard unit of measure for absorbed dose or dose equivalent to humans. A millirem is one thousandth of a rem (1000 mrem = 1 rem)

Source: President's Blue Ribbon Commission on America's Nuclear Future Report to the Secretary of Energy January 2012

EXHIBIT A-24

Ohio Department of Health, Bureau of Radiation Protection, Acceptable TENORM Analytical Methods for Radium-226 and Radium-228

ACCEPTABLE TENORM ASSAY METHODS FOR RADIUM-226 AND RADIUM-228

At the request of the Ohio Department of Natural Resources (ODNR) and in coordination with the Ohio Environmental Protection Agency (OEPA), the Ohio Department of Health (ODH) has established this list of methods for assaying Radium-226 (Ra-226) and Radium-228 (Ra-228) to assist ODNR permit holders in determining the concentration of Ra-226 and Ra-228 in materials as required by Ohio Revised Code 1509.074 and to assist OEPA permitted solid waste disposal site operators in verifying that:

- All solid waste containing Technologically Enhanced Naturally Occurring Radioactive Material (TENORM) has been accurately assayed for Ra-226 and Ra-228 using an acceptable method; and

- The reported "combined Ra-226 / Ra-228" concentration for solid waste being received for disposal, satisfies the exempt TENORM concentration criteria listed in paragraph (A) of rule 3701:1-43-07 of the Ohio Administrative Code, making the waste acceptable for disposal at their facility.

NOTE:
Exempt TENORM concentrations for solids are <5 picocuries per gram (<5 pCi/g) combined Ra-226 / Ra-228 excluding natural background radiation. Statewide natural background for combined Ra-226 / Ra-228 is 2 pCi/g. ODH may approve a request for an alternate location-specific background on a case-by-case basis.

- TENORM loads accepted at solid waste landfills permitted by OEPA pursuant to Ohio Revised Code Chapter 3734 must be accompanied by lab results identifying the analytical method(s) used from Table 1 and/or Table 2 below.

- A representative composite sample[1] must be obtained from each conveyance of solid waste defined as TENORM that is presented for disposal at a landfill in Ohio.

[1] Note: Taking one sample for a production operation or geographic region is <u>not</u> acceptable.

TABLE 1 Analytical Methods for Radium-226 and Radium-228 in TENORM Solids

PARAMETER	METHOD	REFERENCE	METHODOLOGY
Ra-226	901.1M	EPA, 1980 www.epa.gov/waste/hazard/testmethods/sw846/index.htm	Gamma spectroscopy (after 21-day ingrowth period) - water method modified (M) for solids.
Ra-226	903.1	EPA, 1980 http://www.epa.gov/homeland-security-research/epa-method-9031-radium-226-drinking-water-radon-emanation-technique	Radiochemistry
Ra-226	9315M	Florida NELAP Accreditation #E87688 to Summit Environmental Technologies, Cuyahoga Falls, OH (330) 253-8211	Radiochemistry
Ra-226	Ra-03-RC	HASL-300, 28th edition, 1997 Volume 1, Section 4 http://www.orau.org/ptp/PTP%20Library/library/DOE/eml/hasl300/HASL300TOC.htm	Radiochemistry
PARAMETER	**METHOD**	**REFERENCE**	**METHODOLOGY**
Ra-226, Ra-228	EMSL-LV-0539-17	EPA, 1979 EMSL-LV-0539-17 (pgs. 19 - 32) http://nepis.epa.gov/Simple.html	Radiochemistry
Ra-228	904	EPA, 1980 www.epa.gov/waste/hazard/testmethods/sw846/index.htm	Radiochemistry
Ra-228	9320M	Florida NELAP Accreditation #E87688 to Summit Environmental Technologies, Cuyahoga Falls, OH (330) 253-8211	Radiochemistry
Ra-228/Ac-228, Pb-212, Bi-214	901.1M	EPA, 1980 www.epa.gov/waste/hazard/testmethods/sw846/index.htm	Gamma spectroscopy - water method modified (M) for solids.

TABLE 2 Screening Methods for Radium-226 and Radium-228 in TENORM Solid Waste for Disposal.

PARAMETER	METHOD	REFERENCE	METHODOLOGY
Ra-226, Ra-228	ODH 901.1M - Ra226 Direct	Approved method for ODH radioactive materials use licensee(s): • #03225150000 - Shale Testing Solutions LLC, Warren, OH (330) 502-7882 • #03122780029 - Summit Environmental Technologies, Inc., Cuyahoga Falls, OH (330) 253-8211 • #03123850006 - Microbac laboratories, Inc. Marietta, OH (740) 373-4071 x 4172 • #03219150000 – Armada EP OH, New Concord, OH (740) 255-0357 • #03219160000 – Buckeye Brine LLC, Uhrichsville, OH (740) 295-9331 • #03219150001 – K2 Environmental Leetonia, OH (330) 277-7686	Gamma spectroscopy - Ra-226 by 186.2 keV (sumpeak with U-235 185.7 keV), Ra-228 by Ac-228 peak
Ra-226, Ra-228	ODH In-Situ Radium	Approved method for ODH radioactive materials use licensee(s): • #03219070000 – Austin Master Services LLC, Martins Ferry, OH (740) 609-3800	Limited to in-situ Gamma spectroscopy of containerized TENORM solids.

Ra-226, Ra228	ND-DOH Screening for Disposal	North Dakota Department of Health Approved Screening for Disposal Method. For a list of ND-DOH approved labs, see: https://deq.nd.gov/aq/Radiation/TENORM.aspx	Gamma spectroscopy (utilizing Ra-226 186 keV peak with 0.571 correction factor), Ra-228 by Ac-228 peak

NOTE: Accredited laboratories located outside of Ohio may have approved screening for disposal methods equivalent to those identified in Table 2 above. To be added to Table 2, the out-of-state lab must submit a copy of the approval documentation from its state regulatory authority by email to BRadiation@odh.ohio.gov with the subject line "TENORM Method Approval". If the state regulatory authority's review and approval process is similar to or stricter than Ohio's review process, then the lab will be added to Table 2.

EXHIBIT A-25

List of List of Sites Authorized for Beneficial Use of Drill Cuttings from Marcellus Shale Gas Wells in the Commonwealth

Permit Number	Name of the Permittee	Address of the Permittee	Permitted Activity Authorized by the Permit	Location of the Permitted Activity (Processing Site)	Location of the Permitted Activity (Beneficial Reuse at)	Permit Status
WMGR097R017	Clean Earth, Inc.	334 S Warminster Rd Hatboro, PA 19040-3430	Research and development project to process and beneficially re-use drill cuttings and drilling solids/sediments from natural gas extraction wells as a construction material at Act 2 sites and as construction material for road subbase.	Williamsport Drilling Mud Processing Facility 212 Colvin Rd Williamsport, PA 17701	Canal Road 269 Canal Road Fairless Hills, PA 19030-4305 Falls Township, Bucks County NJ Zinc Brownfield, LLC 1120 Mauch Chunk Rd Palmerton, PA 18071-1110 Carbon County Hazelton Creek Properties, LLC Route 924 Mined Lands City of Hazleton, PA 18201 Luzerne County Bobst Mountain Hunting Club Little Gap Run North Abandoned Strip Mine Site Cogan House Township, Williamsport, PA Lycoming County	Effective 11/29/2011 Expired 03/01/2017
WMGR097R027	NJ Zinc Brownfield, LLC	1120 Mauch Chunk Rd Palmerton, PA 18071-1110	Research and development project to process and beneficially use drill cuttings and sediments generated from shale gas exploration to construct a protective cap at the former NJ Zinc Company West Plant remediation site.		NJ Zinc Brownfield, LLC 1120 Mauch Chunk Rd Palmerton, PA 18071-1110 Carbon County	Effective 08/18/2015 Rescinded 09/01/2016
WMGR097R025	Range Resources Appalachia LLC	3000 Town Center Blvd Canonsburg, PA 15317	Research and development project to process and beneficially use drill cuttings from natural gas wells as an aggregate in a stabilized soil pavement for construction of Marcellus Shale and Utica well pads and access roads.		Dog Run Hunting Club UNIT 4-6H OG ESCGP 725 Hickory Swale Rd Jersey Shore, PA 17740 Cummings Township, Lycoming County	Effective 08/01/2014 Expired 08/01/2016

EXHIBIT A-26

Pennsylvania Department of Environmental Protection (PADEP). 2024b. Management of Fill policy

DEPARTMENT OF ENVIRONMENTAL PROTECTION
Bureau of Waste Management

DOCUMENT NUMBER: 258-2182-773

EFFECTIVE DATE: January 16, 2021

TITLE: Management of Fill Policy

AUTHORITY: This document is established in accordance with the Solid Waste Management Act, 35 P.S. §§ 6018.101 *et seq.* (SWMA); the Clean Streams Law, 35 P.S. §§ 691.1 *et seq*.; Section 1917-A of the Administrative Code, 71 P.S. § 510-17; and the Land Recycling and Environmental Remediation Standards Act, 35 P.S. §§ 6026.101 *et seq.*

POLICY: A person placing solid waste onto the ground is generally required to obtain a disposal permit from the Department of Environmental Protection (Department or DEP). A person is not required to obtain a permit under SWMA if the person can demonstrate that the material qualifies as clean fill in accordance with the municipal and residual waste regulations, 25 Pa. Code § 271.101(b)(3) and § 287.101(b)(6).

PURPOSE: This policy provides DEP's procedures for determining whether fill is "clean fill," as defined in the municipal and residual waste regulations at 25 Pa. Code § 271.1 and § 287.1, respectively, or "regulated fill," as defined in this policy. Regulated fill may not be used outside of a project area or right-of-way of a project unless a SWMA permit has been issued to the person using the regulated fill.

APPLICABILITY: This policy shall be used to evaluate whether a person is required to obtain a permit under the SWMA for the use of fill in accordance with the municipal and residual waste regulations, 25 Pa. Code § 271.101(b)(3) and § 287.101(b)(6). This policy describes the type of fill that qualifies as clean fill or regulated fill. This policy does not apply to mine land reclamation activities subject to a permit or fill used within the same project area or project right-of-way. Excavation, movement or reuse of fill within a project area or right-of-way of a project is not an activity that requires a SWMA permit. This policy does not apply to fill that has been determined to be clean or regulated fill prior to the effective date of this policy, unless the fill is moved to a new receiving site or off the project area or project right-of-way after the effective date of this policy. This policy does not apply to fill that has been determined to be clean or regulated fill prior to the implementation of revised clean fill concentration limits or regulated fill concentration limits, unless the fill is moved to a new receiving site or off the project area or project right-of-way after the effective date of the revised limits.

DISCLAIMER: The policies and procedures outlined in this guidance document are intended to supplement existing requirements. Nothing in the policies or

procedures shall affect regulatory requirements. The policies and procedures herein are not an adjudication or a regulation. There is no intent on the part of the DEP to give the rules in these policies that weight or deference. This document establishes the framework within which DEP will exercise its administrative discretion in the future. DEP reserves the discretion to deviate from this policy statement if circumstances warrant.

PAGE LENGTH: 26 pages

DEFINITIONS:

Acid-producing rock – Stone, rock or other mineral materials that, when exposed to air and water, cause a low pH discharge that adversely affects or endangers public health, safety, welfare, or the environment or causes a public nuisance.

Act 2 – The Land Recycling and Environmental Remediation Standards Act, 35 P.S. §§ 6026.101 *et seq.*

Act 2 site – A site as defined in Section 103 of Act 2, 35 P.S. § 6026.103, for which a notice of intent to remediate has been submitted to DEP.

Background – The concentration of a regulated substance that is present at a site but not related to the release of regulated substances from a specific point source or activity at the site.

Background reference area – The area identified for sampling that: will be used to establish background; is sampled and analyzed to determine the concentration of regulated substances found at or within a close proximity to the donor site, at a depth comparable to that of the area to be excavated at the donor site, in the same soil layer as the donor fill; is unaffected by a release of regulated substances from a specific point source or activity at the site; and meets one of the following criteria:

i. The concentration of regulated substances in the soil is attributable to the parent material from which the soil was derived and the natural processes which produce soil, or

ii. The concentrations of regulated substances in the soil resulted from an atmospheric deposition, including lead or polynuclear aromatic hydrocarbons, but are not attributable to a specific point source or release of a regulated substance. For the purposes of this definition, "atmospheric deposition" refers only to the ubiquitous, widespread deposition of regulated substances from the air that is incapable of being traced to a specific point source or multiple point sources. For example, chromium that has condensed on the ground outside an electroplater air vent would not be due to "atmospheric deposition" because the presence of the chromium is a result of a discharge from a specific point source, even though the chromium was released into the air before being deposited on the ground. However, the presence of lead or benzo-a-pyrene (BAP) in an urban or industrial area that can be traced to the operation of motor vehicles may be due to atmospheric deposition if the concentration levels are demonstrated to be pervasive over the greater urban or industrial area.

Clean fill – Uncontaminated, nonwater-soluble, nondecomposable, inert solid material used to level an area or bring an area to grade. The term does not include materials placed in or on the waters of the Commonwealth. Although the placement of clean fill in or on waters of the Commonwealth cannot be managed under this policy, placement of clean fill in or on waters of the Commonwealth may be approved under a separate DEP authorization. The term includes only those materials that are identified as "fill," as the term is defined in this policy. The term does not include fill that has been blended, mixed or treated with the purpose of meeting the definition of "clean fill" and that without being blended, mixed or treated would fail to meet the numeric limits identified in the definition of "uncontaminated material" contained in this policy.

Clean fill concentration limits (CFCLs) – With the exception of PCBs and chloride, the concentrations of regulated substances that do not exceed the numeric values specified in Table 3 [Medium-Specific Concentrations (MSCs) for Organic Regulated Substances in Soil] and Table 4 [Medium-Specific Concentrations (MSCs) for Inorganic Regulated Substances in Soil] of Appendix A in 25 Pa. Code

Chapter 250 (relating to administration of land recycling program). The applicable numeric limit is determined by comparison of the Generic Soil to Groundwater Value[1] with the Direct Contact Residential Value[2] and selection of the lower of the two values. For PCBs, the sum total of the concentration of all PCB aroclors (total PCB concentration) may not exceed 50 ppm. Fill containing a concentration of total PCBs greater than 2 ppm may be subject to regulation under the Toxic Substances Control Act (TSCA), 15 U.S.C. Section 2601 et seq., and 40 C.F.R. Part 761, which is administered and implemented by the U.S. Environmental Protection Agency (EPA). EPA's TSCA requirements are independent of any use of fill that is otherwise in accordance with the Department's policy and regulations. An applicant should be aware that its characterization and handling of any soils through the guidance of the Management of Fill policy does not necessarily satisfy a potential EPA TSCA inquiry, and that an applicant may need a separate approval from EPA should EPA require it. For all such material, DEP recommends that you contact the PCB Coordinator for EPA Region 3 by email at R3_PCB_Coor@epa.gov to determine whether PCB-containing fill may be used and to obtain information relating to the associated EPA procedures for collecting and analyzing samples. For chloride, the value obtained using the Synthetic Precipitation Leaching Procedure, (SPLP, SW-846, Method 1312) may not exceed the numeric value specified in Table 2 [MSCs for Inorganic Regulated Substances in Groundwater] of Appendix A in 25 Pa. Code, Chapter 250.

Composite sample – A sample collected across a spatial range that typically consists of a set of discrete samples that are combined or "composited." A composite sample should not be confused with a discrete sample that is created from multiple increments taken at a single location to obtain a sample of the desired size, shape and orientation.

Discrete sample – A sample that represents material from a single location. A discrete sample can be composed of more than one increment.

Donor site – The area from which fill originates that is separate from a receiving site. Multiple donor sites may be identified on a single project area.

Environmental due diligence – Investigative techniques used to determine whether fill from a donor site has been affected by a release of a regulated substance. Examples of investigative techniques included in this term are visual property inspections, electronic data base searches, review of ownership and historical use of a property, Sanborn maps, environmental questionnaires, transaction screens, analytical testing, environmental assessments, audits, or procedures outlined in ASTM standard E1527-13. A single investigative technique may not be used as the basis for environmental due diligence. Environmental due diligence includes visual property inspection and a review of ownership and historical property use, at a minimum, unless analytical sampling is performed in lieu of a review of ownership and historical property use.

Fill – The term is limited to clean, regulated and historic fill that is soil, rock, stone, gravel, used asphalt, brick, block or concrete from construction and demolition activities that is separate from other waste and recognizable as such, and "dredged material," as the term is defined by the municipal and residual waste regulations, 25 Pa. Code §§ 271.1 and 287.1, whichever is applicable. The term does not include reclaimed asphalt pavement, naturally occurring asbestos, mine spoils or acid-producing rock.

Grab sample – A discrete sample, consisting of one increment, collected specifically for Volatile Organic Compounds (VOC) analysis.

---

[1] Numeric values based on generic leaching modeling for soils at residential properties overlying used aquifers with total dissolved solids at concentrations less than or equal to 2500 mg/L.

[2] Direct contact numeric values for soils at residential properties.

Historic fill – Material, excluding material disposed in landfills, waste piles and impoundments, used to bring an area to grade prior to 1988, and consisting of a conglomeration of soil and residuals, such as ashes from the residential burning of wood and coal, incinerator ash, coal ash, slag, dredged material and construction and demolition waste. The term does not include iron or steel slag that is separate from residuals if it is a coproduct, as the term is defined in 25 Pa. Code § 287.1 and satisfies the requirements of 25 Pa. Code § 287.8. The term does not include coal ash that is separate from residuals if it is beneficially used in accordance with 25 Pa. Code §§ 290.1 – 290.415.

Increment – Material collected in a single operation of the sampling device.

PCB – A chemical substance that is limited to the biphenyl molecule that has been chlorinated to varying degrees or a substance that contains that substance.

ppm – Parts per million.

Project area – The boundary within which earth disturbance activities occur, including areas in close proximity to the earthmoving activities that are necessary for the completion of a construction project, or other human activity which disturbs the surface of the land, including land clearing and grubbing; grading; excavations; embankments; land development; agricultural plowing or tilling; operation of animal heavy use areas; timber harvesting activities; road maintenance activities; linear projects such as utility line work; oil and gas activities; well drilling; mineral extraction; and the moving, depositing, stockpiling, or storing of soil, rock or earth materials. The term includes the boundary within which all earth disturbance activity, construction, materials storage, grading, landscaping and related activities occur.

Reclaimed asphalt pavement (RAP) – Small particles, typically less than one inch in size, of bitumen and inorganic materials produced by the mechanical grinding of bituminous pavement surfaces that have not been subject to a release of regulated substances or mixed with other solid waste. The term does not include "used asphalt," as the term is defined in this policy.

Receiving site – The area to which fill is proposed to be relocated. A receiving site is separate from a donor site and not part of a project area or right-of-way.

Regulated fill – "Fill," as the term is defined in this policy, that has been affected by release of a regulated substance and is not "uncontaminated material," as the term is defined in this policy. The term does not include fill that has been blended, mixed or treated with the purpose of meeting the definition of "regulated fill" and that without being blended, mixed or treated would fail to meet the regulated fill concentration limits, as the term is defined in this policy.

Regulated fill concentration limits (RFCLs) – With the exception of PCBs, the concentrations of regulated substances that do not exceed the numeric values specified in Table 3 [Medium-Specific Concentrations (MSCs) for Organic Regulated Substances in Soil] and Table 4 [Medium-Specific Concentrations (MSCs) for Inorganic Regulated Substances in Soil] of Appendix A in 25 Pa. Code Chapter 250 (relating to administration of land recycling program). The applicable numeric limit is determined by comparison of the Generic Soil to Groundwater Value[3] with the Direct Contact Non-Residential Value[4] and selection of the lower of the two values. For PCBs, the sum total of the

[3] Numeric values based on generic leaching modeling for soils at non-residential properties overlying used aquifers with total dissolved solids at concentrations less than or equal to 2500 mg/L.
[4] Direct contact numeric values for soils at non-residential properties.

concentration of all PCB aroclors (total PCB concentration) may not exceed 50 ppm. Fill containing a concentration of total PCBs greater than 2 ppm may be subject to regulation under the Toxic Substances Control Act (TSCA), 15 U.S.C. Section 2601 et seq., and 40 C.F.R. Part 761, which is administered and implemented by the EPA. EPA's TSCA requirements are independent of any use of fill that is otherwise in accordance with the Department's policy and regulations. An applicant should be aware that its characterization and handling of any soils through the guidance of the Management of Fill policy does not necessarily satisfy a potential EPA TSCA inquiry, and that an applicant may need a separate approval from EPA should EPA require it. For all such material, DEP recommends that you contact the PCB Coordinator for EPA Region 3 by email at R3_PCB_Coor@epa.gov to determine whether PCB-containing fill may be used and to obtain information relating to the associated EPA procedures for collecting and analyzing samples.

Regulated substance – The term includes hazardous substances and contaminants regulated under the Hazardous Sites Cleanup Act, 35 P.S. §§ 6020.101 *et seq.*; and substances regulated by the Clean Streams Law, 35 P.S. §§ 691.1 *et seq.*; the Air Pollution Control Act, 35 P.S. §§ 4001 *et seq.*; the Solid Waste Management Act, 35 P.S. §§ 6018.101 *et seq.*; the Infectious and Chemotherapeutic Waste Law, 35 P.S. §§ 6019.1 *et seq.*; and the Storage Tank and Spill Prevention Act, 35 P.S. §§ 6021.101 *et seq*.

Release – Spilling, leaking, pumping, pouring, emitting, emptying, discharging, injecting, escaping, leaching, dumping or disposing of a regulated substance into the environment in a manner not authorized by the Department. The term includes the abandonment or discarding of barrels, containers, vessels and other receptacles containing a regulated substance.

Uncontaminated or *Uncontaminated material* – The term means either of the following:

(1) Fill unaffected by a release of a regulated substance or,

(2) Fill affected by release of a regulated substance, if the concentrations of regulated substances in the fill do not exceed the clean fill concentration limits, as the term is defined in this policy. Analysis should be carried out for only those regulated substances that are suspected to be present due to a release.

The term does not include fill that has been blended, mixed or treated with the purpose of meeting the definition of "uncontaminated material."

Used asphalt – Pieces of bitumen and inorganic materials from the demolition of bituminous pavement. The term does not include "reclaimed asphalt pavement," as the term is defined by this policy.

REFERENCES:

25 Pa. Code Chapters 287 to 299 (residual waste regulations)
25 Pa. Code Chapters 271 to 285 (municipal waste regulations)
Solid Waste Management Act, 35 P.S. §§ 6018.101 *et seq.*
Land Recycling and Environmental Remediation Standards Act, 35 P.S. §§ 6026.101 *et seq.*
Section 1917-A of the Administrative Code, 71 P.S. § 510-17
The Clean Streams Law, 35 P.S. §§ 691.1 *et seq*.

TECHNICAL GUIDANCE:

A. Purpose and Applicability

Fill is used in construction or earthmoving projects across the Commonwealth to level an area or bring an area to grade. These projects may involve using fill as a subbase or to fill in low-lying areas. The manner in which fill may be used depends on whether the fill is clean fill or regulated fill. This policy provides procedures for determining whether fill is clean fill or regulated fill and describes how each category may be managed after a fill determination has been performed.

This policy does not apply to the following activities:

- Mine land reclamation activities subject to a permit.
- Management of waste from land clearing, grubbing and excavation, including trees, brush, stumps and vegetative material.[5]
- Movement or use of fill within a project area or right-of-way of a project.
- Use of reclaimed asphalt pavement in accordance with DEP's industry-wide coproduct determination.
- The use of clean fill or regulated fill prior to January 1, 2020, unless the fill is moved to another receiving site, project area or off the project right-of-way after January 1, 2020.

In general, fill that is demonstrated to be clean fill can be used in an unrestricted manner, provided it is not placed in waters of the Commonwealth; it is used in compliance with 25 Pa. Code, Chapters 102 and 105 (relating to erosion and sediment control; and dam safety and waterway management); and it is managed in accordance with Section D of this policy. Persons using fill must also comply with the fugitive emissions regulations under 25 Pa. Code, Chapter 123 (relating to standards for contaminants) issued under the Air Pollution Control Act, 35 P.S. § 4001, and shall comply with all the applicable provisions of 25 Pa. Code §§ 123.1 and 123.2 (relating to prohibition of certain fugitive emissions and fugitive particulate matter). Depending on the manner in which it is generated, clean fill may be a "waste," as that term is defined in the municipal or residual waste regulations, 25 Pa. Code § 271.1 and § 287.1, respectively, whichever is applicable.

This policy does not apply to fill that has been determined to be clean or regulated fill prior to the implementation of revised CFCLs or RFCLs, unless the fill is moved to a new receiving site or off the project area or project right-of-way after the effective date of the revised CFCLs or RFCLs.

Fill that is demonstrated to be regulated fill can be used by persons who have applied for and obtained coverage under the Department's General Permit No. WMGR096, Beneficial Use of

[5] In accordance with 25 Pa. Code § 271.101(b)(4), a person managing waste from land clearing, grubbing and excavation, including trees, brush, stumps and vegetative material, shall implement best management practices developed by the Department. Refer to Document No. 254-5400-001 - *Best Management Practices for the Management of Waste From Land Clearing, Grubbing, and Excavation (LCGE)*.

Regulated Fill. Coverage under General Permit No. WMGR096 is not required in the following instances:

- Remediation activities undertaken entirely on an Act 2 site, pursuant to the requirements of § 902 of Act 2.

- When fill from an Act 2 site is used as construction material at a receiving site that is being remediated to attain an Act 2 standard, provided the procedural and substantive requirements of Act 2 and the conditions specified in Section C.2.a. and b. of this policy are satisfied.

- Use of the regulated fill is limited to the excavation, movement or use of the regulated fill within a project area or right-of-way of a project.

Regulated fill is a "waste," as that term is defined in the municipal or residual waste regulations, 25 Pa. Code § 271.1 and § 287.1, respectively.

B. Procedure for Performing a Fill Determination

Prior to the movement of fill to a receiving site, either the person proposing to provide the fill from a donor site or the person proposing to receive the fill determines whether the fill is clean fill or regulated fill pursuant to this policy. Use the following steps to make that determination:

1. Determine Eligibility: A material is eligible for management as clean or regulated fill under this policy if it satisfies the following criteria:

    a. The material is "fill," as the term is defined in this policy. If the fill under consideration contains acid-producing rock, it is specifically excluded from the definition of fill. Appendix B contains information relevant to identifying acid-producing rock.

    b. The fill does not contain regulated substances that were intentionally released.

    c. The fill has not been blended, mixed or treated with the purpose of meeting the definition, or applicable numeric limits, of "uncontaminated material," "clean fill" or "regulated fill."

    d. The fill does not exhibit a characteristic of toxicity, as determined by 40 CFR § 261.24 (relating to toxicity characteristic). The toxicity characteristic is of concern only when environmental due diligence indicates that the fill being considered for use may have been affected by a release of a regulated substance that is included in Table 1 in 40 CFR § 261.24. If the total concentration of the substance exceeds the limit for the substance in Table 1 of 40 CFR § 261.24 by a factor of 20 or more, the issue regarding potential toxicity should be addressed either by performing the Toxic Characteristic Leaching Procedure (TCLP), in accordance with Method 1311, found in the most recent version of EPA's publication, *Test Methods for Evaluating Solid Waste, Physical/Chemical Methods*, also known as SW–846, or providing additional description of the fill, indicating that the substance is bound in the matrix and not leaching.

e. PCB-containing Fill: If the environmental due diligence indicates that the fill may have been subject to a release of PCBs, test it for the presence of PCBs. Fill containing a concentration of total PCBs greater than 2 ppm may be subject to regulation under the Toxic Substances Control Act (TSCA), 15 U.S.C. §§ 2601 *et seq.*, and 40 CFR Part 761, which is administered and implemented by the EPA. EPA's TSCA requirements are independent of any use of fill that is otherwise in accordance with the Department's policy and regulations. An applicant should be aware that its characterization and handling of any soils through the guidance of the Management of Fill policy does not necessarily satisfy a potential EPA TSCA inquiry, and that an applicant may need a separate approval from EPA should EPA require it. For all such material, DEP recommends that you contact the PCB Coordinator for EPA Region 3 by email at R3_PCB_Coor@epa.gov to determine whether PCB-containing fill may be used and to obtain information relating to the associated EPA procedures for collecting and analyzing samples.

2. Perform Environmental Due Diligence: Once determined that the fill is eligible for use under this policy, evaluate the fill to determine whether it has been affected by a release of a regulated substance by performing "environmental due diligence," as the term is defined in this policy. Except for historic fill, analytical testing of the fill is not necessary unless environmental due diligence indicates that the fill may have been affected by a release of a regulated substance. However, a person performing a fill determination may choose to perform analytical testing in lieu of conducting a review of ownership and historic property use to satisfy the minimum condition for performing environmental due diligence.

The use of historic fill as clean fill under this policy is limited to historic fill that is a conglomeration of soil, residuals and fill. Historic fill that is comprised primarily of residuals does not represent a conglomeration of soil, residuals, and fill and therefore, cannot be used as clean fill. Pockets of residuals, such as ash or slag, should be removed and managed separately from other historic fill prior to making a determination that the historic fill can be used as clean fill. Perform analytical testing to demonstrate that the historic fill meets the definition of uncontaminated material. To qualify for use as clean fill, historic fill should be tested for the parameters included in Table 1, below, as well as any additional parameters that are suspected based on historic property use or review of records. The placement of historic fill as clean fill may not contaminate groundwater. For regulated substances detected in the historic fill, the value obtained using the Synthetic Precipitation Leaching Procedure, (SPLP, SW-846, Method 1312) may not exceed the numeric value as identified in Table 1 [MSCs for Organic Regulated Substances in Groundwater] and Table 2 [MSCs for Inorganic Regulated Substances in Groundwater] of Appendix A in 25 Pa. Code, Chapter 250.

a. If due diligence shows no evidence that the fill may have been affected by a release of a regulated substance, the fill may be managed as clean fill in accordance with the Section D of this policy (relating to management of clean fill) unless during movement, transport or placement there are observable indications (such as appearance or odors) which indicate evidence of a release of a regulated substance.

b. If due diligence shows evidence that the fill may have been affected by a release of a regulated substance, test the fill to determine if it is clean fill or regulated fill. Perform the testing in accordance with Appendix A of this policy. Analysis should be carried out for only those regulated substances that are suspected to be present due to a release or based upon historic use of the donor site.

i. Except as provided elsewhere in this policy, if testing reveals that the fill contains regulated substances at concentrations that are below the CFCLs, the fill may be managed as clean fill in accordance with Section D of this policy (relating to management of clean fill). A person may not blend, mix or treat fill that would otherwise fail to meet the CFCLs with the purpose of meeting the definition of uncontaminated material or clean fill. For the purposes of completing Form FP-001 for the certification of clean fill, the CFCLs in effect on the date of submission should be used to evaluate whether the fill qualifies for use as clean fill.

ii. Except as provided elsewhere in this policy, if testing reveals that the fill contains regulated substances at concentrations that exceed the CFCLs but are at or below the RFCLs, the fill may be managed as regulated fill only if coverage under General Permit No. WMGR096 is obtained. A person may not blend, mix or treat fill that would otherwise fail to meet the RFCLs with the purpose of meeting the definition of regulated fill. Manage regulated fill in accordance with the Section C of this policy (relating to management of regulated fill).

iii. Except as provided elsewhere in this policy, if testing reveals that the fill contains regulated substances at concentrations that exceed the RFCLs, the fill may not be managed as clean fill or regulated fill. Fill exceeding the RFCLs may require disposal in accordance with the hazardous, municipal or residual waste regulations, 25 Pa. Code, Articles VII, VIII or IX, respectively, whichever is applicable.

TABLE 1: Screening Parameters for Historic Fill

Regulated Substance	CASRN	Regulated Substance	CASRN	Regulated Substance	CASRN
Aldrin	309-00-2	PCB-1254 (Aroclor)	11097-69-1	Copper	7440-50-8
Anthracene	120-12-7	Phenanthrene	85-01-8	Iron	7439-89-6
Benzene	71-43-2	Pyrene	129-00-0	Lead	7439-92-1
Benzo(a)anthracene	56-55-3	Toluene	108-88-3	Manganese	7439-96-5
Benzo(a)pyrene	50-32-8	Trichloroethane, 1,1,1-	71-55-6	Mercury	7439-97-6
Benzo(b)fluoranthene	205-99-2	Trichloroethylene (TCE)	79-01-6	Molybdenum	7439-98-7
Benzo(ghi)perylene	191-24-2	Xylenes (Total)	1330-20-7	Nickel	7440-02-0
Chrysene	218-01-9	Aluminum	7429-90-5	Selenium	7782-49-2
Cumene (Isopropyl benzene)	98-82-8	Antimony	7440-36-0	Silver	7440-22-4
DDD, 4,4	72-54-8	Arsenic	7440-38-2	Thallium	7440-28-0
DDE, 4,4	72-55-9	Barium	7440-39-3	Vanadium	7440-62-2
DDT, 4,4	50-29-3	Beryllium	7440-41-7	Zinc	7440-66-6

TABLE 1: Screening Parameters for Historic Fill

Regulated Substance	CASRN	Regulated Substance	CASRN	Regulated Substance	CASRN
Dichloroethylene, cis-1,2-	156-59-2	Boron	7440-42-8	Ammonia	7664-41-7
Dieldrin	60-57-1	Cadmium	7440-43-9	Chloride	7647-14-5
Ethylbenzene	100-41-4	Chromium(III)	16065-83-1	Fluoride	7681-49-4
Fluorene	86-73-7	Chromium(VI)	18540-29-9	Sulfate	7757-82-6
Ideno(1,2,3-cd) pyrene	193-39-5	Chromium (total)	7440-47-3		
Napthalene	91-20-3	Cobalt	7440-48-4		

C. Management of Regulated Fill

Regulated fill must be managed in accordance with the Department's municipal or residual waste regulations, 25 Pa. Code § 271.2 and § 287.2, respectively, whichever is applicable, and may be beneficially used in accordance with General Permit No. WMGR096.

Coverage under General Permit No. WMGR096 is not required in the following instances:

1. Remediation activities undertaken entirely on an Act 2 site, pursuant to the requirements of Section 902 of Act 2.

2. When fill from an Act 2 site is used as construction material at a receiving site that is being remediated to attain an Act 2 standard, provided the procedural and substantive requirements of Act 2 and the following are satisfied:

    a. Regulated substances contained in the fill are incorporated into the notice of intent to remediate and the final report for the remediation taking place at the receiving site.

    b. Movement of fill between Act 2 sites is documented in the final reports for both the donor site and receiving site.

    c. Except as provided elsewhere in this policy, placement of the fill does not and will not cause the receiving site undergoing remediation to exceed the selected Act 2 standard.

3. Use of the regulated fill is limited to the excavation, movement or use of the regulated fill within a project area or right-of-way of a project.

A person or municipality interested in obtaining coverage under General Permit No. WMGR096 must apply to the Department in accordance with the application instructions provided in the permit. The terms and conditions of General Permit No. WMGR096 are available on the Department's website.

D. Management of Clean Fill

Pursuant to 25 Pa. Code § 271.101(b)(3) and § 287.101(b)(6), use of clean fill does not require a permit under the SWMA or the municipal or residual waste regulations. Clean fill may be used in accordance with all applicable requirements governing the placement or use of clean fill,

including 25 Pa. Code Chapter 102 (relating to erosion and sediment control) and 25 Pa. Code Chapter 105 (relating to dam safety and waterway management). Persons using fill must also comply with the fugitive emissions regulations under 25 Pa. Code, Chapter 123 (relating to standards for contaminants) issued under the Air Pollution Control Act, 35 P.S. § 4001, and shall comply with all the applicable provisions of 25 Pa. Code §§ 123.1 and 123.2 (relating to prohibition of certain fugitive emissions and fugitive particulate matter). The use of clean fill may be regulated under other environmental laws and regulations.

If the uncontaminated brick, block or concrete from a construction or demolition activity is intended for use as clean fill, best management practices (BMPs) should be followed prior to demolition activities to remove from a building or structure all materials that do not meet the definition of clean fill, such as materials or surfaces covered with lead-based paint, friable asbestos, and hazardous materials such as mercury switches, PCB ballasts, tritium-containing exit signs, and fluorescent light bulbs.

Clean fill may not contain any free liquids based on visual inspection and cannot create a public nuisance (such as an objectionable odor) to users of the receiving site or adjacent properties.

If any person wants to use clean fill under this policy, complete Form FP-001, Certification of Clean Fill, and submit it to DEP electronically on the DEP website at https://www.dep.pa.gov/Business/Land/Waste/SolidWaste/Residual/Pages/default.aspx. Complete and submit the FP-001 prior to movement of clean fill to the receiving site. Complete and submit FP-001 regardless of whether sampling and analysis are performed as part of environmental due diligence.

If the donor site has undergone or is undergoing cleanup or remediation under a local, state or federal regulatory program that requires site characterization, or if the fill proposed to be managed as clean fill has otherwise been subject to analytical testing or other procedures identified in the definition of "environmental due diligence," attach the following to Form FP-001:

- Copies of the sampling plan developed for the fill,
- All laboratory reports,
- Documentation and data associated with a background determination and equivalent site evaluation conducted as part of the fill determination, including the identification and location of point sources, the proximity of identified point sources to the background reference area, identification of areas of imported fill other than imported clean fill, etc.

If a person receives fill from multiple donor sites, a separate Form FP-001 is necessary for each donor site. DEP will accept the completed FP-001 electronically via a link on the DEP website.

If a background demonstration is made, as described in Appendix A of this policy, use the FP-001 to include documentation of the background demonstration along with documentation demonstrating that an equivalent site evaluation has been performed and the provisions of Appendix A have been satisfied.

Both the donor site and the receiving site are responsible for maintaining copies of the completed Form FP-001 for a period of five (5) years. Copies of the form and all supporting documentation, including analytical test reports, should be made available and provided to DEP upon request.

Appendix A
Sample Collection and Analytical Testing Protocol for Performing Environmental Due Diligence

Prior to movement of fill to a receiving site, use Sections B-D of the Management of Fill policy to make a fill determination. Analytical testing of the fill is not necessary unless environmental due diligence indicates a release of a regulated substance. This Appendix provides guidelines for using analytical testing as part of the environmental due diligence.

A. Sampling Plan Development

The first step in a chemical evaluation of fill is to develop a plan for sampling. To use analytical testing as part of the environmental due diligence, develop and implement a scientifically credible sampling plan in accordance with the most recent version of the EPA's publication, *Test Methods for Evaluating Solid Waste, Physical/Chemical Methods,* also known and hereinafter referred to as SW-846, and the *RCRA Waste Sampling Draft Technical Guidance, EPA530-D-02-002*. Chapter 9 of SW-846 describes procedures for developing a sampling plan and the statistical treatment of data. Where there is disagreement between the procedures outlined in this Appendix and the referenced EPA documents, follow the procedures contained in this Appendix.

Employ a systematic planning process, such as the Data Quality Objectives Process identified in the *RCRA Waste Sampling Draft Technical Guidance*, to set objectives for the type, quantity and quality of data needed to demonstrate with a known level of assurance that the applicable standards for clean fill or regulated fill are achieved. The level of complexity and detail needed in the sampling plan are directly related to the size, scope and level of complexity of the donor site.

The following are the minimum scientific objectives of a sampling plan developed under this policy:

- Identify and quantify known or suspected contaminants in the fill.
- Collect samples that will allow measurements of the chemical properties of the fill that are both accurate and precise.
- Collect representative samples, which for the purposes of implementing this policy are samples exhibiting typical properties of the whole volume of fill.
- Collect enough samples, and in no case less than eight discrete samples or two composite samples, to sufficiently represent the variability of the fill.
- Obtain a statistically valid and reliable estimate of the fill's chemical properties.

Characterize the fill both horizontally and vertically to represent the entire volume of fill to be transported off the donor site and used at a receiving site. A thorough characterization will provide the following information:

- Identity of regulated substances associated with a release that are present in the fill, the concentration of each identified regulated substance, and the spatial variation in concentration of each regulated substance both horizontally and vertically.

- The physical characteristics of the fill in which the regulated substances associated with a release are present. Examples of these include the fill type (such as soil, rock, dredge), texture, dry bulk density, permeability, organic carbon content, porosity, and moisture content. Include documentation of physical characteristics and any significant variability over the donor site.

In the sampling plan include a summary of existing information about the donor site, including any previously performed sampling or analysis information, preliminary estimates of summary statistics such as the mean and standard deviation, process descriptions and materials used, spatial boundaries of the donor site to be managed under this policy, information about what is known or suspected at the donor site, releases, and release mechanisms. Document this information by written descriptions of site conditions supported by maps, cross-sections, site diagrams, or other descriptive, graphical, or tabular illustrations necessary to characterize the site conditions.

Sampling units for fill managed under this policy should represent the total volume of fill being characterized pursuant to Sections B and C of this Appendix. Sampling plans may include a combination of probability sampling and authoritative sampling designs depending on conditions at the donor site. Probability sampling should be used to characterize the fill as a whole. Some sites may require additional, more focused sampling, such as authoritative sampling, to evaluate problem areas, such as localized areas that are suspected to contain the highest levels of regulated substances, or "hot spots," or areas that may require further evaluation. For example, areas that housed an underground storage tank or experienced a release of regulated substances should be sampled authoritatively and more frequently than other areas of the donor site. The remaining area of the donor site should be sampled using probability sampling, in which all parts of the fill being characterized have a known probability of being included in the characterization. Samples collected to delineate a "hot spot" are typically in addition to those collected for the overall site characterization.

B. Sampling Procedures for Fill Stored in Piles

There are several variables involved in the sampling of fill stored in piles, including the size and shape of the pile, compactness of the fill, and physical properties of the fill. The size and shape of the pile should be used to calculate volume and plan for the correct number of samples to be taken. Simple random sampling or stratified random sampling should be used to obtain representative samples from a fill pile, in accordance with SW-846 and Sections 5.2.1 – 5.2.2, and 5.3 of the *RCRA Waste Sampling Draft Technical Guidance, EPA530-D-02-002*. A method of random sampling, such as simple random or stratified random sampling should be used unless one of the following conditions exists:

- There are known distinct strata.

- An objective of the sampling is to prove or disprove that there are distinct strata.

- The number of samples is limited, and an objective of the sampling is to statistically minimize the size of a "hot spot" that might not get sampled.

Stratified random sampling can be employed only if all points within the pile can be accessed. In such cases, the pile should be divided into a three-dimensional grid system. The grid cubes should be numbered, and the grid cubes to be sampled should be chosen by random number tables or generators.

Generally, stainless-steel shovels, trowels, or scoops should be used to clear away surface material before samples are collected. Depth samples may be collected using a decontaminated auger. For a sample core, thin-wall tube samplers or grain samplers may be used. Near surfaces, samples can be collected with a clean, stainless-steel spoon or trowel. All samples collected, except those for VOCs analysis, should be placed into a Teflon-lined or stainless-steel pail and mixed thoroughly before transfer to the appropriate sample container. Since volatilization of VOCs can occur rapidly once the matrix is disturbed, grab samples are necessary for VOCs analysis. Grab samples should be handled as intact cores and transferred immediately to the container that will be used for analysis. Refer to SW-846, Method 5035, for container and preservation details specific to samples for VOCs analysis.

The sampling and subsequent analysis of fill stored in piles may be performed by collecting composite or discrete samples.

1. Procedure for Using Composite Samples:

    a. Do not use composite sampling if the integrity of the individual sample changes because of the physical mixing of discrete samples.

    b. For up to 125 cubic yards of fill, collect and handle eight discrete samples (plus two grab samples for VOCs) as follows:

        i. Prior to compositing, field screen the eight discrete samples to identify the two that are most likely to contain the highest concentrations of VOCs.

        ii. In accordance with SW-846, Method 5035, collect grab samples for VOC analysis from the two points identified by the field screening described above.

        iii. For all other substances, combine the eight discrete samples collected into two composite samples comprised of four discrete samples each. Perform the analysis on the two composite samples in accordance with SW-846.

c. For greater than 125 cubic yards and up to and including 3,000 cubic yards, collect and handle 12 discrete samples (plus three grab samples for VOCs) as follows:

i. Prior to compositing, field screen the 12 discrete samples to identify the three samples that are most likely to contain the highest concentrations of VOCs.

ii. In accordance with SW-846, Method 5035, collect grab samples for VOC analysis from the same sampling points as the three discrete samples identified by field screening.

iii. For all other substances, combine the 12 discrete samples collected into three composite samples comprised of four discrete samples each. Perform the analysis on the three composite samples in accordance with SW-846.

d. For each additional 1,000 cubic yards of fill or part thereof over the initial 3,000 cubic yards, collect four additional discrete samples (plus one grab sample for VOCs). Composite and analyze the four discrete samples in accordance with SW-846.

2. Procedure for Using Discrete Samples:

a. For up to 125 cubic yards of fill, collect and analyze a minimum of eight discrete samples (plus two grab samples for VOCs). For volumes of fill greater than 125 cubic yards and up to and including 3,000 cubic yards, collect and analyze a minimum of 12 discrete samples (plus three grab samples for VOCs). For each additional 1,000 cubic yards of fill or part thereof over the initial 3,000 cubic yards, collect and analyze a minimum of four additional discrete samples (plus one grab sample for VOCs).

b. For VOCs analysis, perform grab sampling as described in subsection B.1 of this Appendix.

C. Sampling Procedures for In-situ Fill

For the purposes of this policy, "in-situ fill" refers to fill that is undisturbed in its original location at the donor site or fill that has previously been used as clean or regulated fill and will be subsequently excavated and moved to a receiving site. If conducting sampling on in-situ fill to evaluate whether that fill can be managed as clean or regulated fill, characterize both the vertical and horizontal extent of the fill to be transported and used at a receiving site. Where multiple zones of contamination are possible due to site-specific conditions, including separate and discrete releases or the manner in which fill was originally placed, the characterization and demonstration that the fill meets the CFCLs or RFCLs apply individually to the separate zones.

For in-situ sampling where the purpose of the sampling is to characterize a specific release at the donor site, discrete samples collected using a focused sampling technique, such as authoritative sampling, must be used for analysis. These areas may be:

- Localized areas that are known to contain levels of regulated substances that exceed the CFCLs or RFCLs, whichever is applicable, based on analytical results, or

- Localized areas suspected to contain levels of regulated substances that exceed the CFCLs or RFCLs from a specific release, whichever is applicable, based on the historic use of the site.

Once the specific release at the donor site has been characterized, composite samples may be used to confirm that the remaining fill to be excavated and transported to a receiving site and used as clean or regulated fill meets the CFCLs or RFCLs, respectively.

To characterize the remaining area, the area should be sampled using a method of random sampling, such as simple random or stratified random sampling. Composite samples can then be used to verify that the fill intended for excavation and transportation meet the CFCLs or RFCLs, whichever is applicable. When composite samples are utilized for in-situ samples, the sampling plan must demonstrate that localized areas that are known to contain regulated substances exceeding the CFCLs or RFCLs, whichever is applicable, are not included in the portion of the site evaluated using composite samples.

Apart from known hot spots, which may require further sampling and analysis, as discussed above, the donor site should be divided into a three-dimensional grid. Where possible, each grid unit should be of similar size and shape and be comprised of equal volumes of fill. A method of random sampling, such as simple random or stratified random sampling, should be chosen based on knowledge of the donor site as set forth in SW-846 or the *RCRA Waste Sampling Draft Technical Guidance, EPA530-D-02-002*.

The number of sample points is determined by the volume of fill being characterized. Sampling frequency should account for the depth of donor fill to be removed. If an area of donor fill will be excavated to more than one depth (for example, three feet in one part and six feet in another part), then the samples should be distributed accordingly at multiple depths to be representative of the full depth of each cut. Determine the minimum number of samples using the procedure outlined in subsection B.2 of this Appendix. Additional sampling may be necessary based on site-specific conditions.

D. Evaluation of Data

Evaluate sample data generated in accordance with Sections B and C of this Appendix in accordance with the following:

1. For a composite sample collected in accordance with subsection B.1, the measured numeric value for a parameter may not exceed the CFCL for that parameter for the fill to be managed as clean fill, or the RFCL for that parameter for the fill to be managed as regulated fill.

2. For a grab sample collected for VOC analysis in accordance with the above sections, the measured numeric value for a parameter may not exceed the CFCL for that parameter for the fill to be managed as clean fill, or the RFCL for that parameter for the fill to be managed as regulated fill.

3. For discrete samples collected in accordance with subsection B.2, the measured numeric values for a substance in 75% of the discrete samples may not exceed the CFCL for that parameter for the fill to be managed as clean fill, or the RFCL for that parameter for the fill to be managed as regulated fill. For persons using the discrete sampling method, no single sample may show regulated substances at a concentration that is more than twice the CFCL or RFCL, whichever is applicable, for any parameter.

E. Alternate Evaluation of Data

In lieu of Section D of this Appendix, a person may use the 95% Upper Confidence Limit (UCL) of the arithmetic mean to determine whether the fill meets the CFCL or RFCL, whichever is appropriate, for a parameter. The calculated 95% UCL of the arithmetic mean should be below the appropriate CFCL or RFCL for that parameter. Persons intending to use this method for the treatment of data should determine a minimum number of samples in accordance with SW-846 and the *RCRA Waste Sampling Draft Technical Guidance, EPA530-D-02-002*. The application of the 95% UCL of the arithmetic mean should comply with the following performance standards:

1. The null hypotheses (Ho) is that the true arithmetic average concentration is at or above the CFCL or RFCL for that parameter, whichever is appropriate, and the alternative hypothesis (Ha) is that the true arithmetic average concentration is below the CFCL or RFCL for that parameter, whichever is appropriate.

2. Meet the underlying assumptions of the statistical method, such as data distribution.

3. Compositing cannot be used for VOCs.

4. The censoring level for each non-detect is the assigned value randomly generated that is between zero and the limit related to the practical quantitation limit (PQL).

5. Tests should account for spatial variability, unless otherwise approved by the Department.

6. Statistical testing should be done individually for each parameter for which a single sample result or multiple results exceed(s) a limit.

7. Where a fill has distinct physical, chemical or biological characteristics, or originates from different areas, do the statistical testing separately.

8. Document the following information:

    a. A description of the original areas of the fill and physical, chemical and biological characteristics of the fill.

b. A description of the underlying assumptions of the statistical method.

c. Documentation showing that the sample data set meets the underlying assumptions of the statistical method.

d. Documentation of input and output data for the statistical test, presented in tables or figures, or both, as appropriate.

e. An interpretation and conclusion of the statistical test.

F. Use of the Synthetic Precipitation Leaching Procedure (SPLP, SW-846 Method 1312) to Establish an Alternative Soil-to-Groundwater Value

Fill may be analyzed using SPLP to provide an alternative soil-to-groundwater value for use in making a fill determination. The value obtained using the SPLP represents a concentration of a regulated substance in the fill that does not produce leachate in which the concentration of the regulated substance exceeds the applicable groundwater MSC identified in Table 1 [MSCs for Organic Regulated Substances in Groundwater] or 2 [MSCs for Inorganic Regulated Substances in Groundwater] of Appendix A in 25 Pa. Code, Chapter 250. For both clean and regulated fill, the groundwater MSC for used aquifers with TDS <2,500 mg/L should be used to compare the SPLP result to Tables 1 or 2. For clean fill, use the groundwater MSC for residential use ("R") for comparison. For regulated fill, use the groundwater MSC for non-residential use ("NR") for comparison. If SPLP is used to identify an alternative soil-to-groundwater value, the alternative value is only applicable to the fill that was tested using SPLP.

Use the following procedure to determine an alternative soil-to-groundwater value based upon the SPLP:

1. During characterization of the donor site, obtain a minimum of ten samples from the proposed fill. For volumes of fill less than 125 cubic yards, collection of a minimum of eight samples is acceptable. Submit the four samples with the highest total concentration of the regulated substance for SPLP analysis. Samples obtained will be representative of the soil type and horizon impacted by the release of the regulated substance.

2. Determine the lowest total concentration (TC) that generates a failing SPLP result. The alternative soil-to-groundwater value will be the next lowest TC.

3. If all samples result in a passing SPLP level, the alternative soil-to-groundwater value will be the TC corresponding to the highest SPLP result. Additional samples may be collected.

4. If none of the samples generates a passing SPLP, additional samples may be collected and concurrent TC/SPLP analyses performed to satisfy the above conditions for establishing an alternative soil-to-groundwater value.

5. The alternative soil-to-groundwater value is then compared to the direct contact residential value for clean fill or the direct contact non-residential value for regulated fill

found in Chapter 250, Appendix A, Tables 3[6] or 4[7]. The lower of the compared values is the applicable numeric limit.

G. Performing a Background Demonstration and Equivalent Site Evaluation

A background demonstration may be utilized for both clean fill and regulated fill determinations. For clean fill determinations, use the CFCLs. For regulated fill, use the RFCLs. If fill from the donor site contains regulated substances at concentrations exceeding the CFCL or RFCL, whichever is appropriate, for that parameter, a demonstration may be made to show that the exceedance is due to background at the donor site. If a successful background demonstration is made, perform an equivalent site evaluation prior to movement of fill to a receiving site. The equivalent site evaluation ensures that no new regulated substance is placed on the receiving site other than a regulated substance already determined to be present and that the concentration(s) of regulated substance(s) in the donor fill has been compared to the concentration(s) of the same regulated substance(s) at the receiving site in accordance with subparagraphs G.3.b.i-ii. of this Appendix. Regulated substances detected in the donor fill that are below the CFCL or RFCL, whichever is appropriate, for that parameter, do not require a background demonstration or an equivalent site evaluation.

Generally, only naturally occurring metals, lead and some ubiquitous organics, such as polynuclear aromatic hydrocarbons (PAHs), from widespread atmospheric deposition, are eligible for a background demonstration. When data or other information indicates that a regulated substance has migrated onto the donor site from the release of a regulated substance at another site, the regulated substance is not due to background of that substance at the donor site. Pathways for the migration of a regulated substance due to an offsite release include surface runoff from specific sources (such as runoff from parking lots and storage facilities where spills have occurred); spills at railroad facilities and in railroad rights-of-way; and air deposition of regulated substances from specific sources.

Previously collected background data published by an accredited source with appropriate peer review may be considered, provided the information is sufficiently focused and contains the level of detail on the area used to determine background necessary to legitimately compare it to the donor site. The description of the sampling and analysis performed should be detailed enough to provide statistical validity.

Use the following guidelines when performing a background demonstration under this policy:

1. Select a Background Reference Area:

   The first step in making a demonstration that the presence of a regulated substance is due to background at the donor site and is not due to a release is to select a background reference area, as the term is defined in this policy, to collect samples for the purpose of establishing background at the donor site. Samples may be collected from the background reference area to demonstrate that an exceedance of a CFCL or RFCL, as appropriate, can be attributed to background, as the term is defined in this policy. Background reference areas should not include areas affected by a known or suspected

[6] Direct contact numeric values for soils at residential properties
[7] Direct contact numeric values for soils at non-residential properties

release of a regulated substance, including areas impacted by road runoff, areas near railroads affected by engine exhaust contaminants, and areas near buildings contaminated by paint chips. In urban areas, background reference areas may include areas where widespread, ubiquitous contamination is present that cannot be traced to a specific source.

Background reference areas should be as similar as possible to the donor site. Every attempt should be made to reduce the factors that are different between the background reference area and the donor site. This does not mean that a sample collected at a location that is a considerable distance from an area known or suspected to have been affected by a release of a regulated substance is unacceptable merely because the known or suspected regulated substance is detected in the sample. The presence of regulated substance outside of the area known or suspected to have been affected by a release may indicate that the presence of the regulated substance is truly ubiquitous, widespread and incapable of being traced to a specific source. In this case, the regulated substance may be part of the background at the donor site.

A background reference area, as the term is defined in this policy, should be selected for use in the background demonstration.

2. Sampling, Analysis and Evaluation of Data:

Establish background by a sampling methodology that is statistically valid and consistent with the methodology used to perform the fill determination. Use the same analysis methods for the background samples that were used for performing the fill determination.

Compare the analytical results of the background samples with the results obtained from the fill determination. Use the following statistical methods for the comparison:

a. Demonstrate that the highest measurement from the donor site is not greater than the highest measurement from the background reference area. The Department may accept insignificant variances in numbers. The minimum number of samples to be collected is 10 from the background reference area and 10 from each donor site. Analysis should be carried out on discrete samples.

b. The Department may accept another appropriate statistical method if it meets the conditions below.

    i. For nonparametric and parametric methods, the false-positive rate for a set of data applied to a statistical test may not be greater than 0.05. The minimum number of samples to be collected is 10 from the background reference area and 10 from each donor site.

    ii. For parametric methods, the censoring level for each non-detect (ND) should be the assigned value randomly generated that is between zero and the limit related to the PQL.

3. Equivalent Site Evaluation:

The equivalent site evaluation ensures that no new regulated substance is placed on the receiving site other than a regulated substance that is already determined to be present and that the concentration(s) of regulated substance(s) in the donor fill has been compared to the concentration(s) of the same regulated substance(s) at the receiving site in accordance with subparagraphs G.3.b.i-ii. of this Appendix. Regulated substances detected in the donor fill that are below the CFCLs or RFCLs, as appropriate do not need to be included in the equivalent site evaluation. Perform the equivalent site evaluation prior to the movement of fill to a receiving site. Include documentation in the FP-001 demonstrating that the equivalent site evaluation has been performed and is satisfied in accordance with this section.

a. Develop a Plan for Sampling the Receiving Site.

Make a background determination on the receiving site to determine whether the same regulated substances present in the donor fill due to background are also present at the receiving site, and if so, determine the concentrations of the identified regulated substances. Development of a sampling plan in accordance with Section A of this Appendix is necessary to characterize the receiving site.

In the sampling plan include a summary of existing information about the receiving site, including any previously performed sampling or analysis information, process descriptions and materials used, spatial boundaries of the receiving site, information about what is known or suspected at the receiving site, releases, and release mechanisms. Document this information by written descriptions of site conditions and supported by maps, cross-sections, site diagrams, or other descriptive, graphical, or tabular illustrations necessary to characterize the site conditions.

The receiving site should be sampled using probability sampling, in which all parts of the site being characterized have a known probability of being included in the characterization, except for areas of the receiving site that are known to be or suspected of being affected by a release of a regulated substance, including areas impacted by road runoff, areas near railroads affected by engine exhaust contaminants, and areas near buildings contaminated by paint chips, unless the entire receiving site is part of a larger urban area where ubiquitous, widespread contamination is present that is incapable of being traced to a specific source.

Select the area of the receiving site used for the equivalent site evaluation in accordance with the following:

i. The area sampled is unaffected by a release of a regulated substance.

ii. The area sampled should be at a depth comparable to the area where donor fill is to be placed on the receiving site.

iii. The concentration of regulated substances in the area sampled is attributable to the parent material from which the soil was derived and the

natural processes which produce soil; or the concentrations of regulated substances resulted from an atmospheric deposition, as the term is described in the definition of "background reference area," but are not attributable to a specific point source or release of a regulate substance.

b. Sampling, Analysis and Evaluation of Data.

Establish the background by a sampling methodology that is statistically valid and consistent with the methodology used to perform the fill determination. Use the same analysis methods for background samples that were used for performing the fill determination.

Compare the analytical results of background samples for the receiving site with the results obtained from the donor fill. Use one of the following statistical methods for comparison:

i. Demonstrate that the highest measurement from the donor site is not greater than the highest measurement from the receiving site. The Department may accept insignificant variances in numbers. The minimum number of samples to be collected is 10 from the receiving site and 10 from each donor site.

ii. The Department may accept another appropriate statistical method if it meets the conditions below.

(A) For nonparametric and parametric methods, the false-positive rate for a set of data applied to a statistical test may not be greater than 0.05. The minimum number of samples to be collected is 10 from the receiving site and 10 from each donor site.

(B) For parametric methods, the censoring level for each non-detect (ND) should be the assigned value randomly generated that is between zero and the limit related to the PQL.

Appendix B
Recognition and Identification of Acid-Producing Rock

Pennsylvania's municipal and residual waste regulations define clean fill, in part, as inert solid material. Acid-producing rock reacts when exposed to air or water and therefore does not meet the regulatory definition of clean fill. In addition to presenting abrupt and adverse environmental concerns, exposed acid-producing rock can also have long-term damaging effects on highways and highway structures, including corrosion of concrete and steel structures; destabilization of cut slopes and fill slopes; ground heaving of structures and pavements; toxicity to roadside vegetation and aquatic life; and degradation of drinking water supplies.

Determining whether or not fill contains acid-producing rock begins with determining the presence of or likelihood of encountering acid-bearing rock (ABR), which is widespread in Pennsylvania. The primary source of acidity in Pennsylvania sedimentary rocks is sulfide minerals. Although there are many minerals that contain sulfur, those containing pyrite, or ferrous disulfide, are the major contributors to the release of acid. While pyrite minerals are not always large enough to be visible to the unaided eye, larger crystals have a yellowish, metallic appearance. Deposits containing pyrite concentrations greater than 0.5% have the potential to be significant sources of acid. Various other forms of sulfide minerals are of lesser concern due to their chemical stability, and include chalcopyrite (CuFeS2), galena (PbS) and sphalerite (ZnS), but can be problematic when present with pyrite.

Although there are more than 200 common minerals that contain sulfur, only those classified as iron sulfide are of potential concern due to the ability of these elements to promote oxidation, hydration and the release of acid. In Pennsylvania, there are four potential sulfide deposit types, listed as follows in descending order of pyrite oxidation reactivity:

- Veined Rock Deposits;
- Sedimentary Rock Deposits;
- Mine Spoils; and
- Acid Sulfate Soil Deposits.

Typically, the upper 25- to 35-feet of bedrock does not contain pyrite because pyrite is not stable under atmospheric conditions and will weather away. Therefore, if excavations are shallower than 30 feet, the risk of acid release is generally minimal. This is particularly true if a site is located south of the glacial margin. Within the glaciated regions of Pennsylvania, weathered bedrock may have been removed by glaciers and pyrite may exist closer to the surface. Unconsolidated sediments, such as glacial till, sand, and gravel, are not acid-producing and can be excavated without risk of acidic drainage. With regard to characterization of fill excavated to depths greater than 25 feet, environmental due diligence should include details demonstrating that the fill does not contain acid-producing rock.

The following publicly available resources may also assist in assessing the likelihood of encountering ABR:

- The Pennsylvania Geological Survey's a map of potentially acid bearing rocks (OFMI Report 05-01.1);
- The Pennsylvania Department of Transportation's (PennDOT) Geotechnical Engineering Manual, Publication 293 (4/18) (PUB 293 (4/18)); and
- DEP's Fact Sheet titled, "How to Avoid and Handle Acid-Producing Rock Formations Encountered During Well Site Development" (PA DEP Link)

If ABR is anticipated in the fill based on published information or identified during due diligence, testing should be done to estimate the acid-producing potential. For more information on testing procedures and acid-base accounting procedures, please refer to PennDOT's "Geotechnical Engineering Manual, Publication 293 (4/18) (PUB 293 (4/18))," DEP's "Coal Mine Drainage Prediction and Pollution Prevention in Pennsylvania (Coal Mine Drainage Prediction and Pollution Prevention)" or DEP's "Evaluation of Acid-Base Accounting Using Computer Spreadsheets (Evaluation of Acid-Base Accounting)."

EXHIBIT A-27

Pennsylvania Department of Environmental Protection (PADEP). 2016. Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) Study Report, Rev. 1. May 2016

TECHNOLOGICALLY ENHANCED NATURALLY OCCURRING RADIOACTIVE MATERIALS (TENORM) STUDY REPORT

Rev. 1

May 2016

Prepared for:

pennsylvania
DEPARTMENT OF ENVIRONMENTAL PROTECTION

Pennsylvania Department of Environmental Protection
Rachel Carson State Office Building
400 Market Street
Harrisburg, PA 17101

Prepared by:

PermaFix® environmental services
A Nuclear Services and Waste Management Company

Perma-Fix Environmental Services, Inc.
325 Beaver Street, Suite 3
Beaver, PA 15009

TECHNOLOGICALLY ENHANCED NATURALLY OCCURRING RADIOACTIVE MATERIALS (TENORM) STUDY REPORT

Rev. 1

May 2016

Prepared for:

Pennsylvania Department of Environmental Protection
Rachel Carson State Office Building
400 Market Street
Harrisburg, PA 17101

Prepared by:

Perma-Fix Environmental Services, Inc.
325 Beaver Street, Suite 3
Beaver, PA 15009

TABLE OF CONTENTS

LIST OF APPENDICES

LIST OF FIGURES

LIST OF TABLES

ABBREVIATIONS, ACRONYMS, AND SYMBOLS

Ac	Actinium
ALARA	As Low As Reasonably Achievable
α	Alpha
ANSI	American National Standards Institute
API	American Petroleum Institute
ATD	Alpha Track Detector
Ba	Barium
$BaCO_3$	Barium Carbonate
$BaSO_4$	Barium Sulfate
Bcf	billion cubic feet
β	Beta
Bi	Bismuth
BRP	Bureau of Radiation Protection
Ca	Calcium
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
CFR	Code of Federal Regulations
CLP	Contract Laboratory Program
cm	centimeter
cpm	counts per minute
CWT	Centralized Wastewater Treatment
DAC	Derived Air Concentration
DCNR	Department of Conservation and Natural Resources
DEP	Department of Environmental Protection
DEP Laboratory	DEP Bureau of Laboratories
DER	Duplicate Error Ratio
DOE	U.S. Department of Energy
DOT	U.S. Department of Transportation
dpm	disintegrations per minute
DQL	Data Quality Level
EIC	Electret Ion Chamber
EPA	U.S. Environmental Protection Agency
Fe	Iron
FSP	Field Sampling Plan
ft	foot/feet
ft^2	square foot
ft^3	cubic foot
g	gram
GIS	Geographic Information System
GM	Geiger-Muller
GIS	Geographic Information Systems
GPS	Global Positioning System
HASL	Health and Safety Laboratory
HCl	Hydrochloric Acid
HDPE	High Density Polyethylene
HNO_3	Nitric Acid
HPS	Health Physics Society

hr	hour
IAEA	International Atomic Energy Agency
ICP	Inductively Coupled Plasma
K	Potassium
keV	kilo-electron volt
l	liter
LLD	Lower Level of Detection
µR/hr	microroentgens per hour
µrem/hr	microroentgen equivalent man per hour
mcf	thousand cubic feet
MDC	Minimum Detectable Concentration
Mg	Magnesium
Mn	Manganese
mph	miles per hour
mrem	millirem
MS	Matrix Spike or Mass Spectrometry
MSD	Matrix Spike Duplicate
Na	Sodium
NaCl	Sodium Chloride
NaI	Sodium Iodide
NELAP	National Environmental Laboratory Accreditation Program
NIST	National Institute of Standards and Technology
NJDEP	New Jersey Department of Environmental Protection
NORM	Naturally Occurring Radioactive Material
NPDES	National Pollutant Discharge Elimination System
NRC	U.S. Nuclear Regulatory Commission
O&G	Oil and Gas
OSHA	Occupational Safety and Health Administration
%R	Percent Recovery
±	plus or minus
Pa	Protactinium
PA	Pennsylvania
Pa. C.S.	Consolidated Statutes
PASDA	Pennsylvania Spatial Data Access
Pb	Lead
pCi	picocuries
Perma-Fix	Perma-Fix Environmental Services, Inc.
pH	Potential Hydrogen
Po	Polonium
POTW	Publicly Owned Treatment Works
PPE	Personal Protective Equipment
ppm	parts per million
PSIA	pounds per square inch absolute
QA	Quality Assurance
QAM	Quality Assurance Manual
QAPP	Quality Assurance Project Plan
QC	Quality Control
Ra	Radium

RCRA	Resource Conservation and Recovery Act
RESRAD	Residual Radiation
RG	Regulatory Guide
Rn	Radon
RPD	Relative Percent Difference
SOP	Standard Operating Procedure
Sr	Strontium
Sv	Sievert
TDS	Total Dissolved Solids
TENORM	Technologically Enhanced Naturally Occurring Radioactive Materials
Th	Thorium
Tl	Thallium
TPU	Total Propagated Uncertainty
U	Uranium
µohm	microhm
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
U.S.	United States
USACE	U.S. Army Corps of Engineers
U.S.C.	United States Code
USGS	U.S. Geological Survey
WL	Working Level
WWTP	Wastewater Treatment Plant
XRF	X-ray Fluorescence
yr	year
ZLD	Zero Liquid Discharge

GLOSSARY*

Alpha – A positively charged particle consisting of two protons and two neutrons, emitted in radioactive decay or nuclear fission. They are generally produced in the process of alpha decay but may also be produced in other ways. They are designated by the Greek letter α.

Basic Sediment – Oil and gas production storage impurities/sediment from produced oil at storage tank battery.

Beta – High-energy, high-speed electrons or positrons emitted by certain types of radioactive nuclei. The beta particles emitted are a form of ionizing radiation also known as beta rays. The production of beta particles is termed beta decay. They are designated by the Greek letter β.

Brine – Water that is produced with oil and gas when a well is in production, typically water containing more dissolved inorganic salt than seawater.

Condensate – A low density, high American Petroleum Institute (API) gravity, mixture of hydrocarbons that is present in a gaseous state at formation temperatures and pressures but condenses out of the raw gas to a liquid form at standard temperature of 60 degrees Fahrenheit and pressure 14.7 pounds per square inch (PSIA).

Conventional Formation – A formation that is not an unconventional formation.

Conventional Well – A bore hole drilled or being drilled for the purpose of or to be used for construction of a well regulated under 58 Pa. C. S. § § 3201—3274 (relating to development) that is not an unconventional well, irrespective of technology or design. The term includes, but is not limited to:

- Wells drilled to produce oil.
- Wells drilled to produce natural gas from formations other than shale formations.
- Wells drilled to produce natural gas from shale formations located above the base of the Elk Group or its stratigraphic equivalent.
- Wells drilled to produce natural gas from shale formations located below the base of the Elk Group where natural gas can be produced at economic flow rates or in economic volumes without the use of vertical or nonvertical well bores stimulated by hydraulic fracture treatments or multilateral well bores or other techniques to expose more of the formation to the well bore.
- Irrespective of formation, wells drilled for collateral purposes, such as monitoring, geologic logging, secondary and tertiary recovery, or disposal injection.

Drill Cuttings – Rock cuttings and related mineral residues generated during the drilling of an oil or gas well.

Drilling Fluid Waste – Oil and gas drilling mud and other drilling fluids (other than fracturing fluid and spent lubricant).

Drilling Mud – A chemical, water-based, or oil-based mixture pumped into an oil well during drilling in order to seal off porous rock layers, equalize the pressure, cool the bit, and flush out the

cuttings. The mud is circulated down the drill pipe, out through the drill bit, across the rock face being drilled, then back to the surface carrying debris from the bottom of the well.

Flowback – The return flow of water and formation fluids recovered from the well bore of an oil or gas well following the release of pressures induced as part of the hydraulic fracture stimulation of a target geologic formation until the well is placed into production.

Flowback Fluid – Flowback fluid is a water based solution that flows back to the surface during and after the completion of hydraulic fracturing. It consists of the fluid used to fracture the target formation. The fluid contains clays, chemical additives, dissolved metal ions, and total dissolved solids (TDS).

Flowback Fracturing Sand – Oil and gas drilling flowback fracturing sand.

Fracturing Fluid Waste – Oil and gas fracturing/stimulation fluid waste and/or flowback.

Gamma – Electromagnetic radiation of an extremely high frequency and high energy. Gamma rays are ionizing radiation, and are thus biologically hazardous. They are classically produced by the decay of atomic nuclei as they transition from a high energy state to a lower state known as gamma decay, but may also be produced by other processes. Natural sources of gamma rays include gamma decay from naturally occurring radioisotopes, and secondary radiation from atmospheric interactions with cosmic ray particles. They are designated by the Greek letter γ.

Gas – A fluid, combustible or noncombustible, which is produced in a natural state from the earth and maintains a gaseous or rarified state at standard temperature of 60 degrees Fahrenheit and pressure of 14.7 PSIA. This product type must be reported in Mcf (1,000 cubic feet) at a standard temperature of 60 degrees Fahrenheit and pressure of 14.7 PSIA.

Horizontal Drill Cuttings – Drill cuttings from the horizontal portion of an oil or gas well.

Hydraulic Fracturing Fluid – Hydraulically pressurized liquid used to fracture rock in the hydraulic fracturing process. Hydraulic fracturing fluids are used to initiate and/or expand fractures, as well as to transport proppant into fractures. The U.S. O&G industry has used fluids for fracturing geologic formations since the early 1940s.

Leachate – A solution resulting from water that has percolated through solid, e.g., waste in landfill, and potentially leached out some of the soluble constituents.

Marinelli – A lightweight polypropylene sample container with snap-on lid used for gamma spectroscopy analysis.

Natural Gas – A fossil fuel consisting of a mixture of hydrocarbon gases, primarily methane, and possibly including ethane, propane, butane, pentane, carbon dioxide, oxygen, nitrogen, and hydrogen sulfide and other gas species. The term includes natural gas from oil fields known as associated gas or casing head gas, natural gas fields known as nonassociated gas, coal beds, shale beds, and other formations. The term does not include coal bed methane.

NORM – Naturally occurring radioactive material. It is a nuclide that is radioactive in its natural physical state, not man-made, but does not include source or special nuclear material.

Oil – Hydrocarbons in liquid form at formation temperatures and pressures that remain in liquid form at standard temperature of 60 degrees Fahrenheit and pressure 14.7 PSIA.

Produced Water – Water that is produced with oil and gas when the well is in production.

Proppant Sand – Solid treated sand suspended in water or other fluid designed to keep an induced hydraulic fracture open during or following a fracturing treatment.

Radiological Environmental Impact – Impact to the environment from the release and subsequent spreading of radionuclides and from the direct emission of radiation from facilities.

Removable Contamination – The fraction of total surface alpha/beta radioactive contamination easily removed by pressing a 47-mm diameter filter paper to the surface with moderate pressure, i.e., smear sampling. Usually expressed in units of dpm/100 cm^2 of surface area sampled.

Secular Equilibrium – A type of radioactive equilibrium in which the half-life of the precursor (parent) radionuclide is so much longer than that of the product (progeny) radionuclide(s) that the radioactivity of the progeny become equal to the parent over time equal to approximately 10 half-life's of the progeny.

Servicing Fluid – Oil and gas production well maintenance and work-over fluids and/or oil/water-based mud and foam.

Smear Sample – A sample of removable alpha and beta surface radioactivity collected by pressing a 47-mm diameter filter paper to 100 cm^2 of surface area sampled to obtain an assumed fraction of removable material. The filter paper is counted for alpha and beta radioactivity without any preparation.

Spent Lubricant – Oil and gas drilling and/or plug drilling lubricants that have exceeded their useful life.

Student t-test – A test for determining whether or not an observed sample mean differs significantly from a hypothetical normal population mean.

TENORM – Technologically enhanced naturally occurring radioactive materials. It is naturally occurring radioactive material not specifically subject to regulation under the laws of the Commonwealth of Pennsylvania or Atomic Energy Act of 1954 (42 U.S.C. §2011 et seq.), but whose radionuclide concentrations or potential for human exposure have been increased above levels encountered in the undisturbed natural environment by human activities.

Total Contamination – The surface alpha/beta radioactive contamination comprised of fixed and removable components. Total contamination is measured by placing an appropriate alpha/beta detector on the surface to be surveyed so that both the fixed and removable fractions are counted together. Usually expressed in units of dpm/100 cm^2 of surface area surveyed.

Unconventional Formation – A geological shale formation existing below the base of the Elk Sandstone or its geologic equivalent stratigraphic interval where natural gas generally cannot be produced at economic flow rates or in economic volumes except by vertical or horizontal well bores stimulated by hydraulic fracture treatments or by using multilateral wellbores or other techniques to expose more of the formation to the well bore.

Unconventional Well – A bore hole drilled or being drilled for the purpose of or to be used for the production of natural gas from an unconventional formation.

Vertical Drill Cuttings – Drill cuttings from the vertical portion of an oil or gas well.

Well Site – The area occupied by the equipment or facilities necessary for or incidental to the drilling, production, or plugging of a well.

*These definitions are for the purposes of this report only and are not necessarily regulatory definitions.

0.0 SYNOPSIS

In 2013, the Pennsylvania Department of Environmental Protection (DEP) initiated a study to collect data relating to technologically enhanced naturally occurring radioactive material (TENORM) associated with oil and gas (O&G) operations in Pennsylvania. This study included the assessment of potential worker and public radiation exposure, TENORM disposal, and other possible environmental impacts. The study encompassed radiological surveys at well sites, wastewater treatment plants, landfills, gas distribution and end use, and O&G brine-treated roads. The media sampled included solids, liquids, natural gas, ambient air, and surface radioactivity.

The observations and recommendations for future actions based on this peer-reviewed study are:

1. There is little potential for additional radon exposure to the public due to the use of natural gas extracted from geologic formations located in Pennsylvania.

2. There is little or limited potential for radiation exposure to workers and the public from the development, completion, production, transmission, processing, storage, and end use of natural gas. There are, however, potential radiological environmental impacts from O&G fluids if spilled. Radium should be added to the Pennsylvania spill protocol to ensure cleanups are adequately characterized. There are also site-specific circumstances and situations where the use of personal protective equipment by workers or other controls should be evaluated.

3. There is little potential for radiation exposure to workers and the public at facilities that treat O&G wastes. However, there are potential radiological environmental impacts that should be studied at all facilities in Pennsylvania that treat O&G wastes to determine if any areas require remediation. If elevated radiological impacts are found, the development of radiological discharge limitations and spill policies should be considered.

4. There is little potential for radiation exposure to workers and the public from landfills receiving waste from the O&G industry. However, filter cake from facilities treating O&G wastes are a potential radiological environmental impact if spilled, and there is also a potential long-term disposal issue. TENORM disposal protocols should be reviewed to ensure the safety of long-term disposal of waste containing TENORM.

5. While limited potential was found for radiation exposure to recreationists using roads treated with brine from conventional natural gas wells, further study of radiological environmental impacts from the use of brine from the O&G industry for dust suppression and road stabilization should be conducted.

1.0 INTRODUCTION

1.1 Purpose and Objectives of the Study

During the expansion of the Marcellus Shale Gas industry the Pennsylvania Department of Environmental Protection (DEP) staff observed a steady increase in the volume of waste containing technologically enhanced naturally occurring radioactive material (TENORM), generated by the oil and gas (O&G) industry, being disposed in Pennsylvania landfills. TENORM is naturally occurring radioactive material whose radionuclide concentrations or potential for human exposure have been increased above levels encountered in the undisturbed natural environment by human activities.

In 2013, DEP initiated a study to collect information and data needed to effectively manage TENORM from O&G operations for environmental and health protection. This study included the assessment of potential worker and public radiation exposure, evaluation of potential impacts from TENORM waste disposal, and the investigation of possible radiological environmental effects. The survey and sample data will be used to address potential radiological concerns from O&G operations, disposal of waste, and product use.

This study report includes recommendations for future actions to be taken to address issues of concern identified by the study, including additional investigations and surveys.

1.2 Background

The Marcellus Shale formation underlies much of Pennsylvania, with the exception of southeastern Pennsylvania. The organic-rich portion reaches its maximum thickness in the northeastern part of the state. The northwestern borders of Franklin, Cumberland, Lebanon, Berks, Lehigh, and Northampton counties provide the southeastern margin of the Marcellus Shale formation. Between this border and the approximate corridor with US 220/I 99, the Marcellus Shale formation crops out in the folded Ridge and Valley physiographic province where it may be a concern for indoor Radon (Rn). The type of gas found in most areas of the Marcellus Shale throughout Pennsylvania is geologically mature and consists of mostly methane that requires little processing prior to use. This gas is commonly called "dry gas." Marcellus Shale gas found along the westernmost border of Pennsylvania is less geologically mature; therefore, in addition to methane, the gas contains additional hydrocarbons such as ethane, propane, and butane. This gas is commonly called "wet gas" and can be used to produce plastics and other high-value petroleum-based products. **Figure 1-1** depicts the extent of the Marcellus Shale formation within Pennsylvania. **Figure 1-2** shows the approximate dividing line between the wet and dry gas zones in the state.

The Pennsylvania Department of Conservation and Natural Resources (DCNR) has documented that Marcellus Shale can contain from 10 to 100 parts per million (ppm) uranium (U). Typical crustal U concentrations in the United States (U.S.) average 3 ppm.

See **Appendix A** for additional geologic information on other natural gas-producing formations and on heavy metal content.

Figure 1-1. Marcellus Shale Formation in Pennsylvania

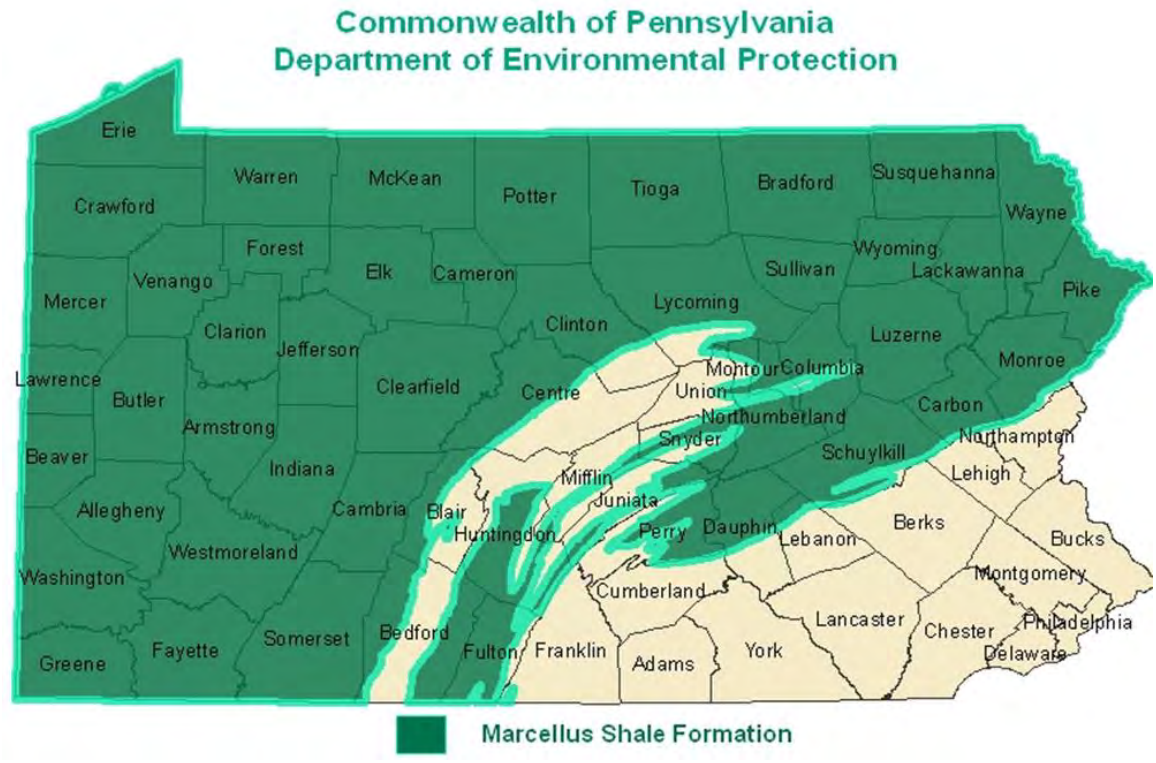
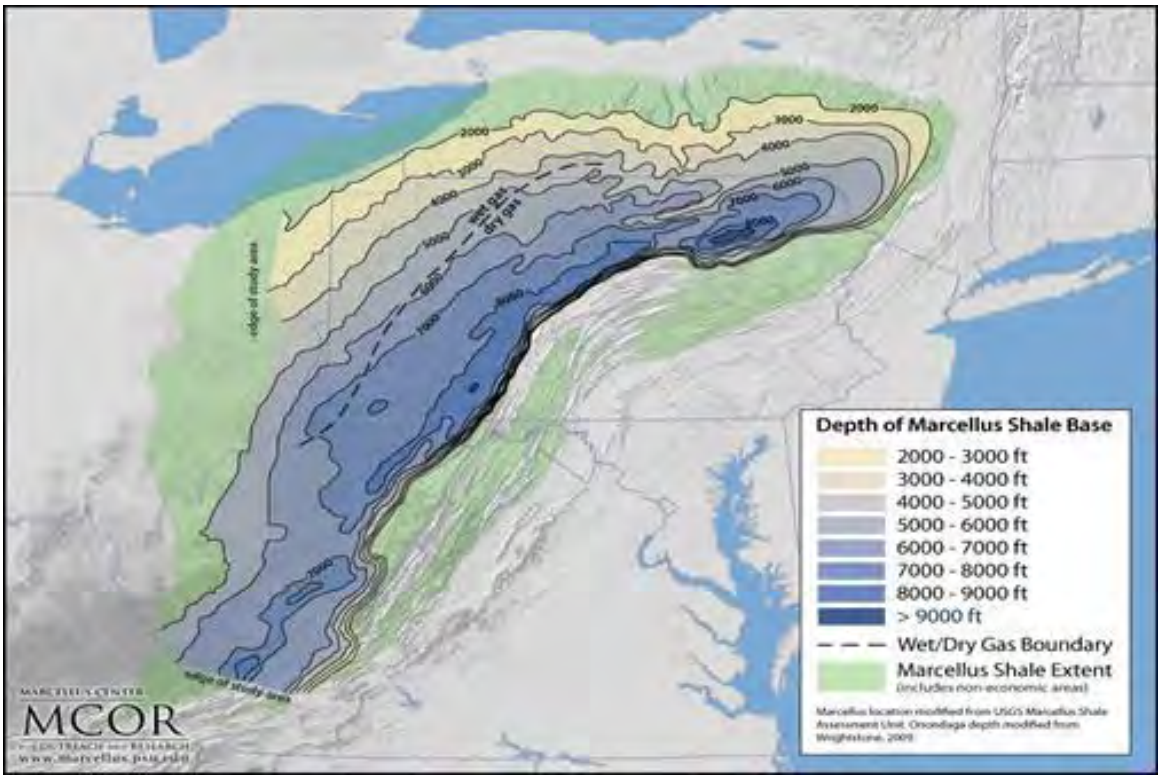


Figure 1-2. Marcellus Shale Formation "Wet" and "Dry" Areas



Source: PSU Marcellus Center for Outreach and Research (MCOR), www.marcellus.psu.edu

Marcellus Shale and other geologic formations rich in O&G resources may contain naturally occurring radioactive material (NORM), specifically U, U-238 parent and thorium (Th), Th-232 parent, and their decay progeny, as well as Potassium-40 (K-40). These series occur naturally and are the most prevalent of the three natural decay series, the third being the actinium (Ac), U-235 parent. The decay series of U and Th are illustrated in **Figures 1-3** and **1-4**, respectively. Surface soil typically contains approximately 1 to 2 picocuries per gram (pCi/g) of both the U and Th series radionuclides with all of the series members at approximately equal activity, i.e., secular equilibrium. The radioactive materials, including TENORM, are brought to the land surface by O&G activities.

Each of the natural decay series includes a Rn gas member. Radon and its progeny are the primary issue of concern associated with natural gas distribution and its end uses.

1.3 Pennsylvania Oil and Gas Operations (Conventional and Unconventional)

Natural gas wells are classified as either conventional or unconventional. Related statutory and regulatory definitions include the following:

Pennsylvania's 2012 Oil and Gas Act (58 Pa. C. S. § 2301)

"Unconventional formation." A geological shale formation existing below the base of the Elk Sandstone or its geologic equivalent stratigraphic interval where natural gas generally cannot be produced at economic flow rates or in economic volumes except by vertical or horizontal well bores stimulated by hydraulic fracture treatments or by using multilateral wellbores or other techniques to expose more of the formation to the well bore.

"Unconventional gas well." A bore hole drilled or being drilled for the purpose of or to be used for the production of natural gas from an unconventional formation.

25 Pa. Code § 78.1

"Conventional formation." A formation that is not an unconventional formation.

"Conventional well."

(i) A bore hole drilled or being drilled for the purpose of or to be used for construction of a well regulated under 58 Pa. C. S. §§ 3201—3274 (relating to development) that is not an unconventional well, irrespective of technology or design.

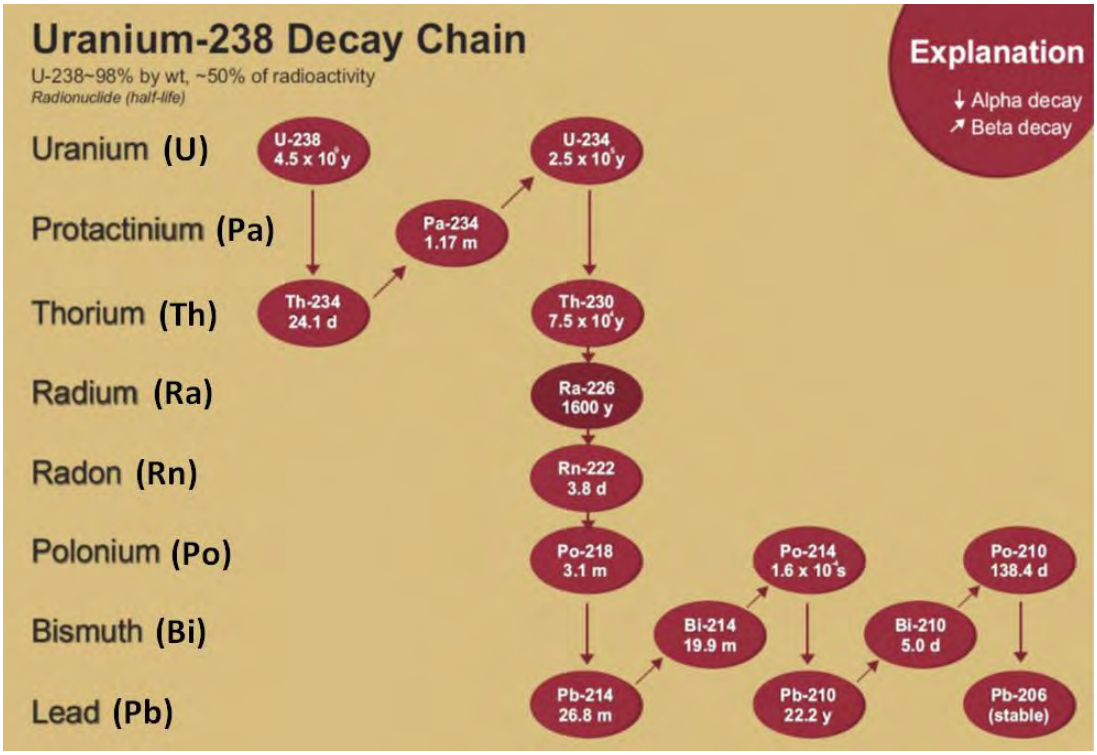
(ii) The term includes, but is not limited to:

(A) Wells drilled to produce oil.

(B) Wells drilled to produce natural gas from formations other than shale formations.

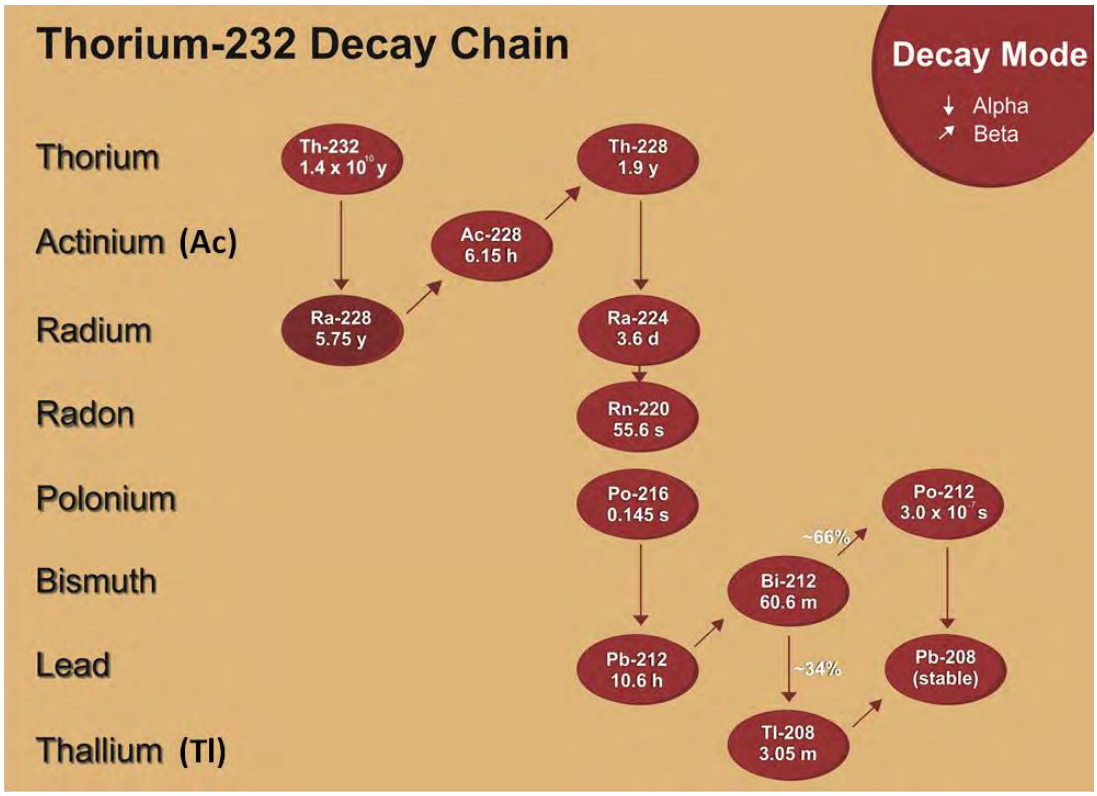
(C) Wells drilled to produce natural gas from shale formations located above the base of the Elk Group or its stratigraphic equivalent.

Figure 1-3. Uranium-238 Decay Chain



Note: y = years, d= days, h = hours, and m = minutes

Figure 1-4. Thorium-232 Decay Chain



Note: y = years, d= days, h = hours, and m = minutes

(D) Wells drilled to produce natural gas from shale formations located below the base of the Elk Group where natural gas can be produced at economic flow rates or in economic volumes without the use of vertical or nonvertical well bores stimulated by hydraulic fracture treatments or multilateral well bores or other techniques to expose more of the formation to the well bore.

(E) Irrespective of formation, wells drilled for collateral purposes, such as monitoring, geologic logging, secondary and tertiary recovery, or disposal injection.

1.4 Subject Media

The types of media evaluated as part of this study result from the product media that either contain TENORM or may be impacted by TENORM due to O&G operations. The product streams evaluated are natural gas and natural gas liquids, i.e., condensates. Other media evaluated includes solid and liquid wastes, soils, ambient air, and gaseous emission products associated with O&G operations.

1.4.1 Media Sampled

1.4.1.1 Solids

Natural gas exploration, extraction and production result in various types of solids that may contain TENORM or may be impacted by TENORM. These materials include drill cuttings, filter sock residuals, impoundment sludge, tank bottom sludge, pipe scale, wastewater treatment plant (WWTP) sludge, and soils. Drill cuttings are wastes brought to the surface during the drilling process. Filter sock residuals and WWTP sludge are generated during the processing of wastewaters generated by O&G activities. Impoundment and tank bottom sludge accumulates as a result of solid material settling out of well site wastewater.

Other solids potentially impacted by radioactive isotopes include soils at WWTP discharge outfalls, soils in the proximity of dirt roads where brines from conventional O&G operations are used for dust suppression, and pipe scale on natural gas transmission infrastructure.

1.4.1.2 Liquids

There are various types of liquids generated during the development and operating life of a gas well including drilling muds, used hydraulic fracturing fluid, brine, and other wastewaters. Liquid wastes are processed at WWTPs for reuse on well sites or to meet National Pollutant Discharge Elimination System (NPDES) criteria prior to discharge to waters of the Commonwealth.

The study classified WWTPs into three categories:

1) Publicly Owned Treatment Works (POTWs) are the most common type of WWTPs. These facilities are designed to process sewage and wastewater from residences and businesses and may take industrial wastewater under specific circumstances. After the wastewater is processed and meets specified chemical criteria, the processed water may be discharged to streams under an NPDES permit.

2) Centralized Waste Treatment (CWT) facilities are designed to process commercial and industrial liquid wastes prior to discharge to receiving streams under an NPDES permit. Additionally, there are some industrial facilities that process wastewater prior to discharge to POTWs for final processing and discharge (pre-treatment).

3) Zero Liquid Discharge (ZLD) facilities are the most modern and utilize distillation and chemical technologies to remove solids from the wastewater. The processed wastewater is returned for reuse at natural gas well sites for hydraulic fracturing of new wells. All centralized ZLD facilities that recycle water to be used for hydraulic fracturing must be permitted by DEP.

Landfill leachate is liquid waste generated by the movement of precipitation through the disposed waste and by the compaction and decomposition of the waste itself. As liquid moves through the waste, contaminants are leached from the disposed material. Landfills are designed to ensure leachate does not enter the groundwater and is collected for treatment. Upon meeting NPDES water quality standards, the treated leachate may be discharged to surface waters. Some landfills operate onsite treatment systems while others are connected to local POTWs, which treat landfill leachate prior to discharge. Because landfills accept natural gas industry wastes such as drill cuttings and treatment sludge that may contain TENORM, there is a potential for leachate from those facilities to also contain TENORM.

1.4.1.3 Natural Gas

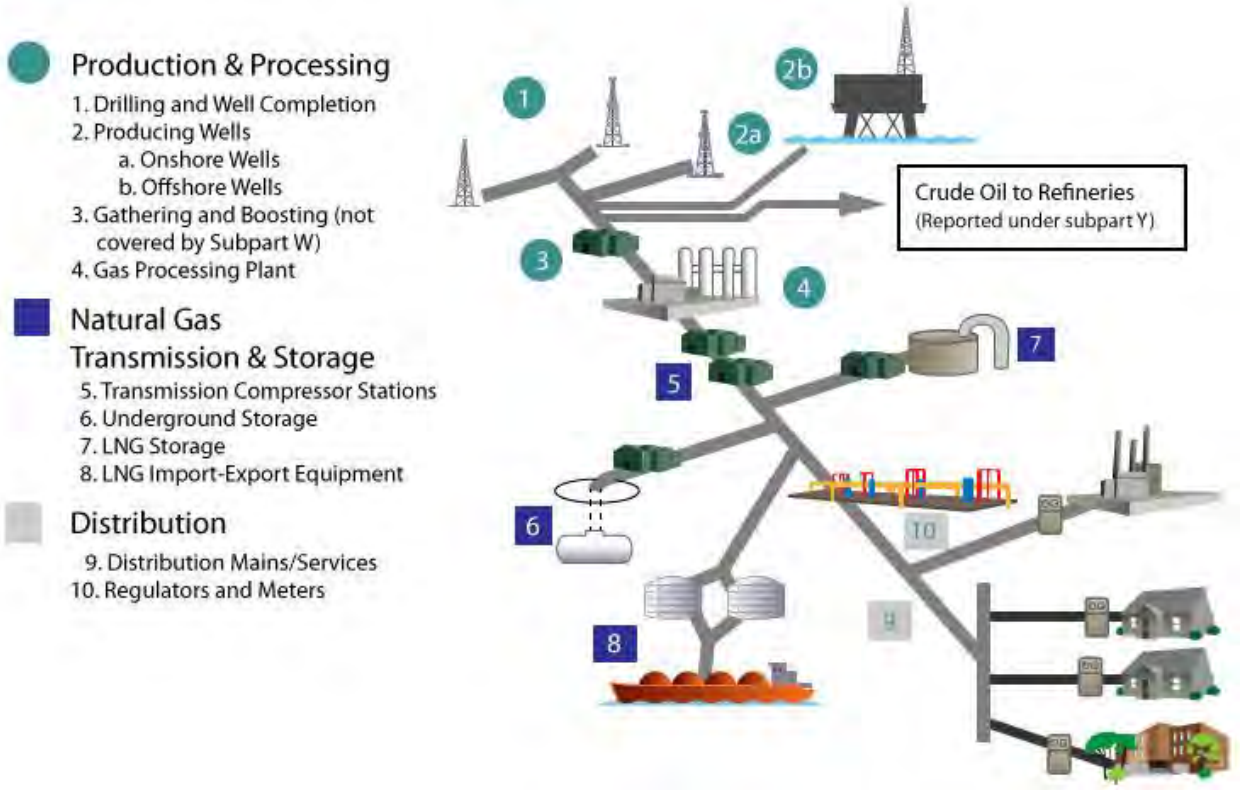
Many facilities, structures, and systems are utilized during the exploration, extraction, and production of natural gas before the product is distributed to the residential, industrial, and commercial end users.

Natural gas samples were collected and evaluated for Rn at compressor stations, natural gas processing plants, and underground storage facilities. Ambient air samples were also collected and evaluated for Rn at well sites, WWTPs, gathering compressor stations, natural gas-fired power plants, and landfills.

Natural gas passes through gathering lines, compressor stations, transmission lines, natural gas processing plants, underground storage facilities, and a network of pipes and valves (see **Figure 1-5**).

Gathering Compressor Stations:
Gathering compressor stations compress the natural gas from the well sites to transport the product to the transmission line network. These facilities include large internal combustion engines and may also include dewatering equipment such as glycol dehydrators and liquid storage tanks. Geographically, they are typically located at a nexus of piping from well sites.

Figure 1-5. Natural Gas Operations



Source: US EPA, http://www.epa.gov/methane/gasstar/basic-information/index.html

Natural Gas Processing:
Natural gas and condensate are also used as feedstock for the synthesis of other products. Natural gas enters a processing facility and undergoes a dehydration process, is refrigerated to remove condensable liquids, then goes through a series of other processes including de-ethanizing/de-propanizing and fractionation. These facilities can be quite large with very extensive piping networks. They also have several intermediate and final product storage tanks and vessels. The operations at these facilities necessitate opening of the product conveyance network for periodic cleaning and maintenance.

Transmission Line Compressor Stations:
These facilities are larger than their gathering station counterparts. Power to the compressors is supplied by natural gas turbine engines, similar to those found on jet aircraft. These facilities normally do not have dehydrating equipment or liquid storage tanks. Dehydration and condensate removal take place further upstream at the well sites and gathering compressor stations. The origin of the natural gas passing through these facilities can be difficult to ascertain. Transmission line compressor stations may be handling natural gas from Pennsylvania, other parts of the U.S., or international sources.

Underground Storage Facilities:
Some deep sandstone formations, such as the Oriskany Sandstone formation, are used for storing natural gas. These underground reservoirs are used to address fluctuations in demand for natural gas.

End Users:
The primary radionuclide of concern in natural gas is Rn-222. Radon is a noble gas and is not destroyed by combustion, nor is it removed by an air emission source control device. Consequently, Rn present in the fuel gas will remain after combustion. However, the process of combustion dilutes the concentration of Rn in the exhaust gas stream by a ratio of 10:1 of ambient air to natural gas when perfect combustion is achieved.

1.5 Facility Selection

Category-specific criteria were used to select specific facilities for inclusion in the study. The criteria differed based on the type of facility. The following lists the various selections.

1.5.1 Well Site Selection

1) A Marcellus Shale formation well site from the dry gas areas predominantly in the northern and central parts of the state.
2) A Marcellus Shale formation well site from the wet gas area found predominantly in the southwestern part of the state.
3) A Utica formation well site and other non-Marcellus Shale formations, e.g., Geneseo, Burket, and Rhinestreet that became available.
4) A conventional O&G well site.

1.5.2 Wastewater Treatment Plant Selection

1) The three types of WWTPs, including POTW facilities, CWT facilities, ZLD facilities.
2) WWTPs that accept wastewater from conventional and unconventional types of well sites.
3) WWTPs that accept waste material from unconventional well sites in the wet gas-producing area rather than the dry gas-producing area.
4) WWTPs where elevated radioactivity readings have been measured from the intake wastewater, produced sludge, effluent discharge, or discharge point stream/river sediments, etc.
5) WWTPs that DEP regional offices have indicated are of particular interest.

1.5.3 Landfill Facility Selection Criteria

1) All Pennsylvania landfills.
2) Nine landfills that accepted the largest amount of TENORM-containing waste during the past year.
3) Large-volume TENORM-containing waste disposal sites where onsite worker exposure measurements could be obtained and representative samples of solids could be collected.

1.5.4 Gas Distribution and End Use Operations Selection Criteria

1) Facilities that compress, carry, and distribute natural gas from the wet gas-producing area of the state.
2) Facilities that compress, carry, and distribute natural gas from the dry gas-producing area of the state.

3) Facilities that distribute or process natural gas produced in Pennsylvania rather than those that distribute or process natural gas from out of state.
4) Major natural gas users, e.g., electrical generator, processing, and storage facilities.

1.5.5 Road Sites Selection Criteria

1) Multiple locations in the southwestern, northwestern, and north-central regions of the state.
2) Roads where liquids from wells in the wet and dry gas-producing areas were applied for dust suppression and road stabilization.
3) Roads where liquids from wells in the wet and dry gas-producing areas were not applied for dust suppression and road stabilization.

1.5.6 Well Component Reconditioning Selection Criteria

Well casing/pipe reconditioning or de-scaling facilities in the state.

1.5.7 Centralized Impoundments

1) A facility in the wet gas-producing area.
2) A facility in the dry gas-producing area.

2.0 STUDY IMPLEMENTATION

2.1 Sampling and Survey Methods

The primary data for this study were gathered using radiological screening surveys and through the sampling and analysis of solid and liquid wastes, soils, ambient air, and gaseous emission products associated with O&G operations.

2.1.1 Field Surveys

2.1.1.1 Scope

Radiological surveys were performed to identify the possible presence and abundance of NORM and TENORM in locations that include the following:

- Well Sites (Section 3.0)
  - Offices and living quarters
  - Storage and maintenance areas
  - Drill rigs and associated equipment
  - Temporary wastewater storage tanks
  - Wastewater impoundments
  - Production equipment
  - Drill cutting pits (closed)
- Wastewater Treatment Plants (Section 4.0)
  - Wastewater off-load areas
  - Influent wastewater storage areas (untreated)
  - Effluent wastewater storage areas (treated)
  - Processing tanks and equipment
  - Offices, break rooms, laboratories
  - Discharge points where applicable
- Landfills (nine study landfills – details provided in Section 5.0)
  - Offices and other occupied spaces
  - Storage and maintenance areas
  - Natural gas processing facilities
  - Leachate processing facilities
  - Earthmoving equipment
- Gas Distribution and End Use (Section 6.0)
  - Compressor stations
  - Natural gas-fired power plants
  - Natural gas processing facilities
- Oil and Gas Brine-Treated Roads (Section 7.0)

2.1.1.2 Instrumentation and Documentation

Radiological instrumentation used for field surveys included portable scalers/ratemeters with various scintillators for detection of alpha (α), beta (β), and/or gamma radiation; portable gamma

dose rate meters; portable gamma exposure rate meters; general purpose Geiger-Muller (GM) detectors; and field counters for low-level α and β radiation detection.

All instruments used were calibrated and their operation verified prior to use on each day they were used. The instruments were maintained and operated in accordance with Perma-Fix Environmental Services, Inc. (Perma-Fix) operating procedures by qualified health physics technicians. Records of calibration, daily quality control (QC) checks for the days used, survey results, logbooks, and various other records generated during field screening survey activities are included in **Appendix B**.

2.1.1.3 Activities

General descriptions of the various field surveys performed as part of this study are provided below.

2.1.1.3.1 Radiological Surveys of Facilities and Reference Background Areas

Gamma radiation exposure rates and gross gamma radioactivity surveys were performed at each facility included in the study. The gamma radiation exposure rates were measured using a Bicron Micro-Rem Meter recorded in micro-Roentgen equivalent man per hour (µrem/hr) or a Ludlum Model 19 Micro-R Meter recorded in units of micro-Roentgen per hour (µR/hr). The gross gamma radioactivity surveys were recorded in counts per minute (cpm) using a Ludlum Model 44-10 Sodium Iodide (NaI) detector. To properly evaluate survey data, surveys were also performed in areas outside the influence of the facility to establish natural background.

2.1.1.3.2 Radiological Surveys of Liquid Samples and Tanks

Liquid samples were collected at each of the three types of WWTPs and included influent, effluent, and in-stream discharge points where POTWs, and in limited cases CWTs, are permitted to discharge directly to a receiving stream.

During liquid sampling, gamma radiation exposure surveys were performed. In addition, gamma radiation exposure rates were performed on contact with tanks when possible. Otherwise, measurements were collected in the general proximity of the point of sample collection or tank. To properly evaluate survey data, surveys were also performed in areas outside the influence of the facility to establish natural background.

2.1.1.3.3 Radiological Surveys of Equipment and Structures

Equipment such as drill rigs, well development equipment, etc., was subject to field screening surveys including:

- Gamma radiation exposure rate surveys using a Bicron MicroRem Meter or Ludlum Model 19.
- Gross gamma radioactivity surveys using a Ludlum Model 44-10 NaI detector.
- Total α and β surface radioactivity using a direct frisk Ludlum Model 43-89 detector and/or a Ludlum Model 44-93 and cpm results converted to units of disintegrations per minute per 100 square centimeters (dpm/100 cm^2) of surface area surveyed.

- Removable α and β surface radioactivity by sample collection with smears. Smears were counted on a Ludlum 2929 with a Model 43-10-1 portable scaler/ratemeter and detector. Count results were converted to units of dpm/100 cm^2 of surface area smeared.

To properly evaluate survey data, surveys were also performed in areas outside the influence of the facility to establish natural background.

2.1.1.3.4 Radiological Surveys of Samples

All samples collected were surveyed prior to transportation to the laboratory. The surveys were performed on contact with the sample container and included:

- Gamma radiation exposure rate surveys using a Bicron MicroRem Meter or Ludlum Model 19.
- Gross gamma radioactivity surveys using a Ludlum Model 44-10 NaI detector.
- Total α and β surface radioactivity using a direct frisk Ludlum Model 43-89 detector or a Ludlum Model 44-93 detector.
- Removable α and β surface radioactivity by sample collection with smears. Smears were counted on a Ludlum 2929 with a Model 43-10-1 portable scaler/ratemeter and detector.

To properly evaluate survey data, surveys were also performed in areas outside the influence of the facility to determine natural background.

2.1.2 Field Sampling Activities

2.1.2.1 Scope

DEP sampled solids, liquids, and gas during the study to understand the movement and potential exposure pathways of TENORM from O&G operations. The sampling and analysis of environmental media provides data that are informative in determining radionuclides of concern as well as their potential mobility. The media sampled during this study included:

- Solid samples:
  - Drill cuttings
  - Wastewater treatment sludge/filter cake
  - Wastewater treatment discharge sediment
  - Soil samples
  - Filter sock residuals
- Liquid samples:
  - Flowback and produced water
  - Accumulated liquids from production equipment
  - Wastewater treatment influent and effluent
  - Landfill leachate influent and effluent
- Gas samples:
  - Natural gas (for Rn-222 concentration)
  - Ambient air (for Rn-222 concentration)
- Removable α/β radioactivity surface samples:
  - Removable α radioactivity by smear sampling

- Removable β radioactivity by smear sampling

Collected samples, with the exception of smear samples, were transported to the DEP Bureau of Laboratories (DEP Laboratory) under chain-of-custody control. Five percent of samples were split by Perma-Fix and forwarded by the DEP Laboratory to the independent QC laboratory (GEL Laboratory of Charleston, SC) for filtration, as needed, and analyses. Smear samples were transported to the Perma-Fix laboratory, and 10 percent of the smear samples were forwarded to the DEP Laboratory for duplicate analysis.

2.1.2.2 Solid Sample Methods

Solid samples were collected using clean sampling equipment. Samples were collected using stainless steel trowels and bowls, then promptly transferred into laboratory-approved containers and immediately labeled to maintain identification.

2.1.2.3 Liquid Sample Methods

When sampling tanks through a valve, samples were collected directly into the clean sample container. Otherwise, representative tank samples were collected using a clean high-density polyethylene (HDPE) dipper. The sampled liquids were transferred to clean, laboratory-approved containers. Two consecutive 4-liter (L) samples were obtained at each sample location.

When the samples were received at the DEP Laboratory, they were preserved. Sample preservation is the measure or measures taken to prevent reduction or loss of target analytes. Analyte loss can occur between sample collection and laboratory analysis because of physical, chemical, and biological processes that result in chemical precipitation, adsorption, oxidation, reduction, ion exchange, degassing, or degradation. Preservation stabilizes analyte concentrations for a limited period of time. The first sample was analyzed after preservation without filtration. The second sample was preserved and subsequently filtered in the laboratory using a 0.45-micron mixed cellulose ester filter. The filtered sample was placed into a clean container. The filtrates were maintained for analysis.

2.1.2.4 Gas Sample Methods

Radon concentration in ambient air was measured by various technologies. The technology used was dependent on several factors, including the location, the collection period/detector deployment period, and atmospheric conditions such as relative humidity. Sampling technologies used for this study included:

- Electret ion chambers (EICs)
- Alpha track detectors (ATDs)

Natural gas grab samples were also collected to measure Rn concentrations. Natural gas was collected directly into scintillation cells, referred to as Lucas cells. Two Lucas cells were connected in sequence, which provided a duplicate sample at each sample location. An in-line Millipore® Type HA, 0.45-micron glass fiber filter was used prior to natural gas entering the first cell. This filter prevents sample contamination by Rn particulate progeny.

The natural gas was flowed through the cells for 10 minutes. This provided for purging of the gas lines and the scintillation cells, resulting in the collection of new discrete samples for analysis.

2.1.2.5 Removable Alpha/Beta Surface Radioactivity Smear Sample Method

Smear samples of removable α and β surface radioactivity were collected by pressing a 47-millimeter diameter filter paper to the sampling surface and smearing with moderate pressure approximately 100 cm^2 of surface area.

2.2 Laboratory Methods

2.2.1 Solid Matrix

The following sample types were classified as solid matrices: surface soil impacted by sediments, filter cakes, soils, sludge, drill cuttings, drilling muds, proppant sand, and filter socks, including the materials inside the socks. Upon arrival at the DEP Laboratory, the samples were scanned for radiological activity using a GM pancake probe. The samples were logged with the appropriate standard analysis code that designated the requested radiological analyses.

2.2.1.1 Gamma Spectroscopy

The samples were dried in a Presier Scientific Model 91-2290-83 100°C oven, ground to a fine powder (~80 mesh), weighed into a new 0.5-L Marinelli, sealed with general purpose polyethylene tape, and analyzed by high purity germanium gamma spectroscopy. The following radionuclides were identified or inferred using gamma spectroscopy:

Ra-226	Direct Energy Line	186 keV
Ra-228	Inferred Energy Line	911 keV (Ac-228)
U-235	Direct Energy Line	143 keV
Ac-228	Direct Energy Line	911 keV
Th-232	Inferred Energy Line	911 keV (Ac-228)
U-238	Inferred Energy Line	63.3 keV (Th-234)
Pb-212	Direct Energy Line	238 keV
Pb-214	Direct Energy Line	351 keV
Bi-212	Direct Energy Line	727 keV
Bi-214	Direct Energy Line	609 keV
K-40	Direct Energy Line	1,460 keV

The sample was counted again using gamma spectroscopy after a minimum of 21 days from the first analysis date. The same radionuclides were identified or inferred. Prior to the start of analysis, a daily background and instrument QC check was completed, reviewed, and validated. The gamma spectroscopy reference method is U.S. Department of Energy (DOE) 4.5.2.3.

2.2.1.2 X-ray Fluorescence

After gamma spectroscopy analyses were complete, the dried solid samples were analyzed for various elements using X-ray fluorescence (XRF). The samples were weighed into XRF sample cups, covered with a Prolene® film, and analyzed using an X-ray spectrometer. Forty-eight

elements were analyzed using XRF. The XRF analyses were conducted using a DEP Laboratory-developed method. Standard QC calibration verification instrument checks were performed using National Institute of Standards and Technology (NIST) primary traceable standards.

2.2.1.3 Alpha Spectroscopy

One percent of solid samples analyzed by gamma spectroscopy were selected and analyzed using alpha spectroscopy for U-238, U-235, U-234, Th-232, Th-230, and Th-228. Prior to analysis, the samples were digested using Health and Environmental Chemistry: Analytical Techniques, Data Management, and Quality Assurance ER200 and ER230 sample preparation methods. A 10-gram (g) aliquot of the original solid sample matrix was digested and diluted to a final volume of 4 L, resulting in a concentration of 2.5 g/L. The isotopes and iron (Fe) carrier added were precipitated from the liquid as hydroxides, re-solubilized in hydrochloric acid (HCl), and then passed over a column of anion exchange resin, which removed the Fe and other interfering isotopes. Each isotopic fraction was concentrated, converted to the nitrate salt, and applied to a second anion exchange column. After washing the resin, the isotope was eluted, electrodeposited, and analyzed for isotopic U and Th. Instrument background, secondary, and pulser counts were obtained at the beginning and end of every sample batch. The alpha spectroscopy reference method is Standard Methods 7500-U C.

2.2.2 Liquid Matrix

The following sample types received at the DEP Laboratory were classified as liquid matrices:

- WWTP influent and effluent liquids
- Landfill leachates
- Well site liquids/fluids including:
  - Hydraulic fracturing fluid
  - Flowback fluid
  - Produced water

Based on solid content, a portion of the drilling mud samples were analyzed as liquids. Upon arrival at the DEP Laboratory, the samples were scanned for radiological activity using a GM pancake probe. The samples were preserved with nitric acid (HNO_3) to a potential hydrogen (pH) less than 2 and logged with the appropriate standard analysis code that designates the requested radiological analyses. After being acidified, samples were maintained a minimum of 16 hours prior to analysis. Samples were vacuum filtered using a 0.45-micron mixed cellulose ester filter. The filtrate was collected and transferred into a clean gallon cubitainer. The filtered solids were analyzed for gamma-emitting radionuclides using gamma spectroscopy (see solid matrix). The liquid samples were counted for gross α-, gross β-, and gamma-emitting radionuclides.

2.2.2.1 Gamma Spectroscopy

The liquid samples were measured to 3 L, placed into a clean 4-L Marinelli, sealed with general purpose polyethylene tape, and analyzed. The following radionuclides were identified or inferred using gamma spectroscopy:

Ra-226	Direct Energy Line	186 keV
Ra-228	Inferred Energy Line	911 keV (Ac-228)
U-235	Direct Energy Line	143 keV
Ac-228	Direct Energy Line	911 keV
Th-232	Inferred Energy Line	911 keV (Ac-228)
U-238	Inferred Energy Line	63.3 keV (Th-234)
Pb-212	Direct Energy Line	238 keV
Pb-214	Direct Energy Line	351 keV
Bi-212	Direct Energy Line	727 keV
Bi-214	Direct Energy Line	609 keV
K-40	Direct Energy Line	1,460 keV

The samples were counted again using gamma spectroscopy after a minimum of 21 days from the date of their first analysis. The same radionuclides were identified or inferred each day analyses were performed. Prior to the start of analysis, a background and standard QC calibration verification check was completed, reviewed, and validated.

2.2.2.2 Gross Alpha Gross Beta Analyses

An aliquot of sample was evaporated to less than 5 milliliters. The evaporated volume was transferred to a 2-inch diameter planchet using 10 percent HNO_3 and dried. The dried sample was placed in a desiccator for 72 hours. The samples were flamed to convert the hydroscopic salts to oxides. The samples were counted for gross α- and gross β-emitting radionuclides using a gas proportional counter. Standard QC calibration verification and daily background checks were completed, reviewed, and validated at the beginning and end of analysis. The gross α and gross β reference method is EPA 900.0.

2.2.2.3 X-Ray Fluorescence

The liquid samples were analyzed for various metals using XRF. The samples were weighed into XRF sample cups, covered with a Prolene® film, and analyzed using an X-ray spectrometer. Forty-eight elements were identified using XRF. The XRF analyses were conducted using a DEP Laboratory-developed method. Standard QC calibration verification instrument checks were performed using NIST primary traceable standards.

2.2.2.4 Inorganic Analyses

During the third round of sampling, additional analyses including basic inorganic analyses were included as part of the study. The samples were received by the DEP Laboratory and logged with the appropriate standard analysis code that designated the requested inorganic analyses. The analyses included hardness (SM2340 B), pH (SM4500H-B), specific conductance at 25.0°C (SM2510B), total chloride (SM4500-CL E), total sulfate (EPA 375.2), total dissolved solids at 180°C (USGS I-1750), and total suspended solids (USGS I-3765).

2.2.3 Gas Matrix

Natural gas samples were collected at various locations using scintillation cells and analyzed for Rn concentration. The scintillation cells were counted in one of two counters: the Pylon AB-5

Portable Radiation Monitor or the Ludlum Model 2200 Scaler-Ratemeter. The counter used was dependent upon the type of scintillation cell used to collect the sample. All samples were allowed to equilibrate for a minimum of four hours before being counted. In all cases, the first count was not used in the calculations to allow for "dark adaptation" of the instruments. The next three counts were each individually calculated and the average and standard deviation calculated. The average result, plus or minus (±) two standard deviations, and the minimum detectable activity are reported in the data tables.

Natural gas is composed mostly of methane, which is lighter and less dense than air. Alpha counting efficiency is directly proportional to the density of the gas counted. Because the scintillation cells were calibrated using a known concentration of Rn in ambient air, density correction was applied to all Rn in natural gas results. A correction factor (Jenkins et al., 2014) was used for this effect to prevent biasing the results. The final calculated Rn concentrations were divided by 1.054. This reduced all results by five percent to correct for the bias.

2.2.4 Filter Matrix – Smears

All smear samples were collected by Perma-Fix technicians and transported to the Perma-Fix Laboratory for analysis. All smear samples were counted for gross α and gross β radioactivity. Ten percent of those smear samples were then forwarded to the DEP Laboratory for duplicate analysis as a QC measure.

Upon arrival at the Perma-Fix laboratory, the samples were logged. The smear samples were placed on a 2-inch diameter planchet and analyzed for gross α and gross β particles using a Ludlum Model 2929 Meter equipped with a Ludlum Model 43-10-1 Smear Counter (zinc-sulfide scintillation detector). A standard QC background and calibration verification count was performed each day the smear counter was used.

Upon receipt at the DEP Laboratory, the samples were logged. The smear samples were placed on a 2-inch diameter planchet and analyzed for gross α and gross β particles using a gas proportional counter. Prior to the start of analysis, an instrument source check and background check were completed, reviewed, and validated. The gross α and gross β filter analyses were conducted using the DEP Laboratory-developed method. A standard QC calibration verification instrument check was performed with NIST traceable sources.

2.3 Survey and Sample Analyses Data Management

All of the solid and liquid samples were analyzed by the DEP Laboratory using gamma spectroscopy. The result, the standard two-sigma error (95 percent confidence level) and the minimum detectable concentration (MDC) were reviewed for each of the following radionuclides as reported:

- Natural Uranium Decay Series Results (U-238, Ra-226, Pb-214, and Bi-214)
- Natural Thorium Decay Series Results (Th-232, Ra-228, Ac-228, Pb-212, and Bi-212)
- Natural Actinium Decay Series Results (U-235)
- Miscellaneous (K-40)

2.3.1 Limitations on Gamma Spectroscopy Results

The following limitations on gamma spectroscopy of radioactive samples were considered when reviewing the analytical results for solid and liquid samples:

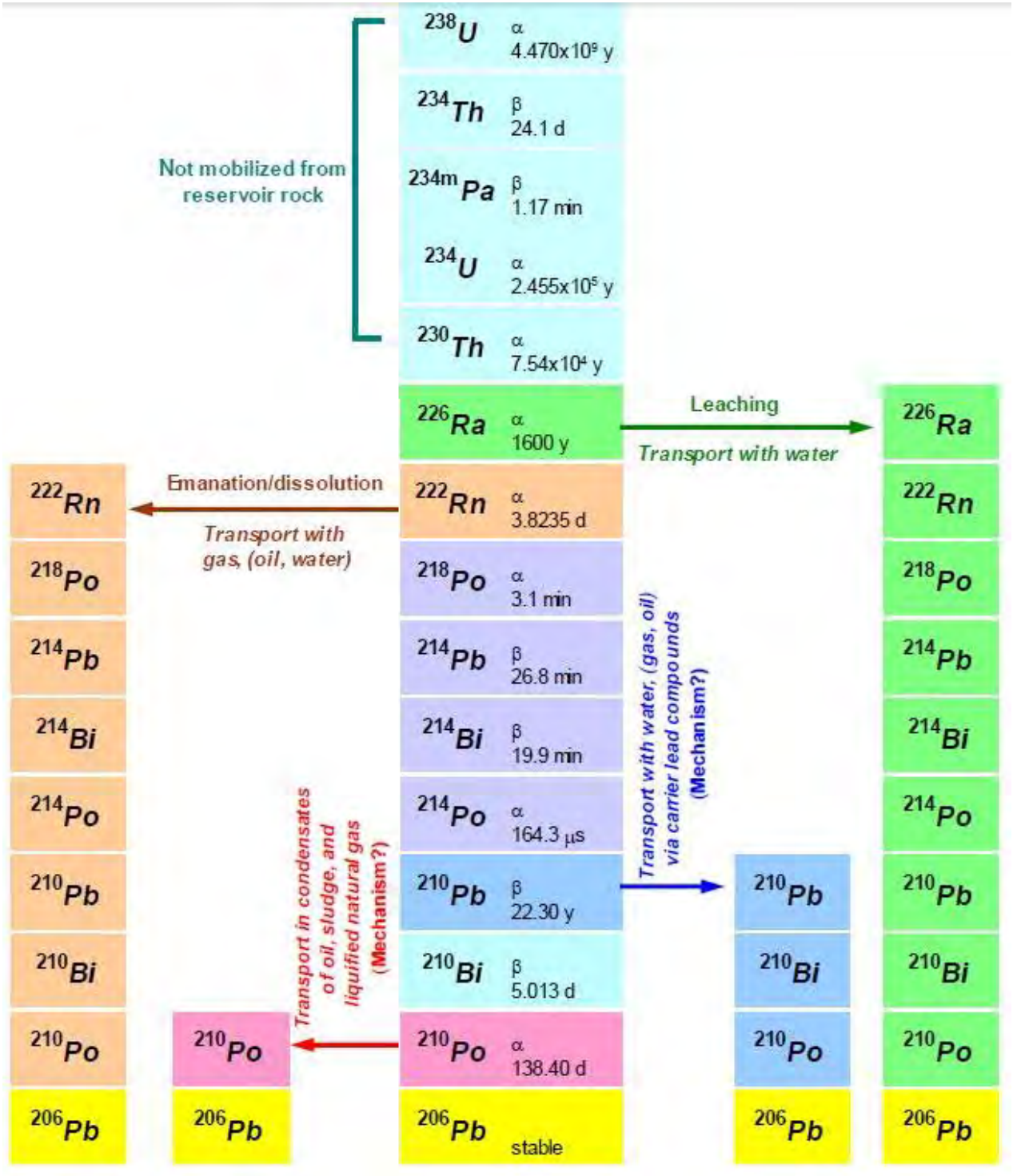
- Gamma spectroscopy cannot directly measure radium (Ra)-228. Rather, Ra-228 is inferred from a short-lived progeny of Ra-228, Ac-228, which is readily detected by gamma spectroscopy when the radionuclides are in secular equilibrium. Due to the relative half-lives of Ra-228 (5.8 years) and Ac-228 (6.1 hours) after 24 hours, this is always the case for the samples collected as part of the study.
- Gamma spectroscopy cannot directly measure Th-232. Consequently, Th-232 is inferred from the short-lived progeny of Th-232, Ac-228, when the radionuclides are in secular equilibrium. Due to the difference in solubility between Th and Ra, this is not the case in liquid samples or in solid samples of wastewater residue, sludge and filter cake. Only the soluble Ra and progeny of Ra are present in those samples. Consequently, knowledge of the status of the secular equilibrium of the Th decay series within the sample matrix is necessary to properly evaluate gamma spectroscopy results. **Figures 2-1** and **Figure 2-2** present the solubility of the Uranium and Thorium Series.
- Uranium-238 can be detected by gamma spectroscopy, but the gamma emission used is of low energy and low yield, resulting in a high MDC and high standard error compared to the other radionuclides in the environment. Consequently, the U-238 result is not used as positive identification of U-238 without knowledge of the status of U series secular equilibrium and the identification of additional, more statistically robust U progeny.
- Uranium is insoluble in water while Ra is water soluble. Therefore, wastewater, produced and flowback fluids, and wastewater treatment solids (sludge and filter cake) contain Ra and its progeny but do not include U.

Only the radionuclides present in a given sample are reported in the following sections. The average, median, standard deviation, and minimum and maximum values are also provided at the bottom of each table for each set of results. Please note:

- When the reported result is less than the MDC, a value equal to ½ the MDC is used in the derivation of average, median, standard deviation, and minimum and maximum values.
- When "<" precedes the reported result, the value is the MDC.

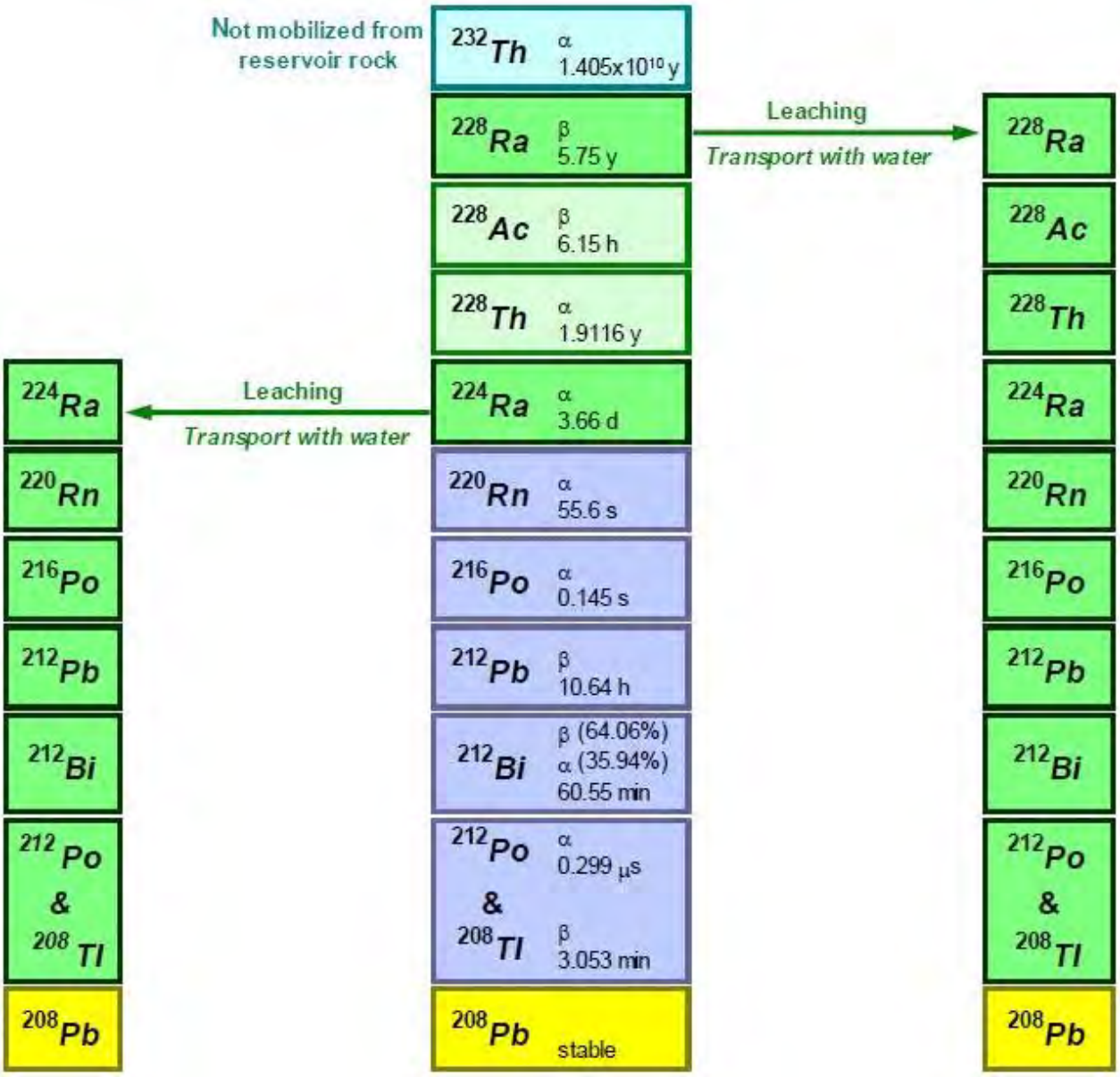
Appendix C contains the gamma spectroscopy analytical analysis results for each radionuclide identified along with their associated standard two-sigma counting error (error) and the MDC for the analyses.

Figure 2-1. Solubility of the Uranium Series in Oil and Gas Produced Water



Source: IAEA 2010.

Figure 2-2. Solubility of the Thorium Series in Oil and Gas Produced Water



Source: IAEA 2010.

2.3.2 Radium-226 Quantification by Gamma Spectroscopy

Radium-226 may be measured directly by detection of its 186.2 kilo-electron volt (keV) energy line, 3.28 percent yield. For liquid samples and sludge/filter cake samples that do not contain U, this yields an accurate Ra-226 result. However, in soil and drill cutting samples, the presence of U-235 causes interference with direct Ra-226 detection because one of its gamma lines is of similar energy, 185.7 keV at 54 percent yield. In solid samples where natural U including U-238 and Ra-226 are at equal activity and U-235 is at 1/22 the activity of U-238, the theoretical overestimation of Ra-226 was quantified assuming the gamma peaks for Ra-226 and U-235 completely overlap. The theoretical overestimation of Ra-226 is presented in **Table 2-1**.

The short-lived equilibrium progeny of Ra, Pb-214 and Bi-214, may be used to infer Ra-226 concentrations in soil or drill cuttings when U-235 is present in the sample. The parent of these progeny, Rn-222, is a gas and has a half-life of 3.8 days. When the soil or drill cuttings sample is collected, some of the Rn gas escapes the solid matrix. Therefore, samples are sealed to allow the Rn gas to in-grow to reestablish equilibrium after the sample has been sealed.

2.3.3 Criteria for Comparison to Analytical Analyses Results

Table 2-2 presents criteria against which the analytical results and assessments of this study were evaluated.

2.3.4 Normal Background Radioactivity Values

Table 2-3 presents average, minimum, and maximum background radioactivity values for soil in the U.S. used as a reference point when reviewing analytical results of solid samples.

2.3.5 Data Presentation

A large volume of survey and sample analytical analyses data were generated. The next five sections present the survey and sampling data for Well Sites, WWTPs, Landfills, Gas Distribution and End Use, and Brine-Treated Roads.

All numbers in this report have been rounded to three significant figures. Actual significant figures for each reported value can be found in **Appendix C**, *Gamma Spectroscopy Analytical Results*.

Table 2-1. Theoretical Overestimation of Ra-226 Activity in Solid Samples with Natural Uranium Analyzed by Gamma Spectroscopy

Radionuclide	(pCi/g)	(pCi/g)	(pCi/g)	(pCi/g)	(pCi/g)	(pCi/g)	(pCi/g)
U-238	1.00	2.00	3.00	4.00	5.00	10.0	20.0
U-235	0.05	0.09	0.14	0.18	0.23	0.45	0.91
Ra-226	1.00	2.00	3.00	4.00	5.00	10.0	20.0
Excess Ra-226[a]	0.75	1.51	2.26	3.02	3.77	7.54	15.1
Reported Ra-226	1.75	3.51	5.26	7.02	8.77	17.5	35.1
Excess U-235[b]	0.06	0.12	0.18	0.24	0.30	0.60	1.21
Reported U-235	0.11	0.21	0.32	0.42	0.53	1.06	2.11

[a]Excess Ra-226 is calculated by converting the U-235 value to Ra-226 activity by a factor equal to the ratio of the gamma yields, i.e., 50.4/3.28.
[b]Excess U-235 is calculated by converting the Ra-226 value to Ra-226 activity by a factor equal to the ratio of the gamma yields, i.e., 3.28/50.4.

Table 2-2. Criteria for Comparison

Parameter	Criteria	Reference	Potentially Apply to:
Volumetric Solids	3 pCi/g Total Radium (Ra-226 + Ra-228) above background	American National Standards Institute (ANSI)/Health Physics Society (HPS) N13.53-2009, Control and Release of Technologically Enhanced NORM (TENORM) (2009)	Sediment, Beneficial Use Surface Soil, Surface Soil on Well Sites
Volumetric Solids	5 pCi/g Total Radium (Ra-226 + Ra-228) above background	EPA Directive No. 9200.4-35, Remediation Goals for Radioactively Contaminated CERCLA Sites (2000)	Sediment, Beneficial Use Surface Soil, Surface Soil on Well Sites
Volumetric Solids	270 pCi/g Total Radium (Ra-226 + Ra-228)	U.S. Department of Transportation (DOT), 49 CFR 173.436, Radioactive Material (in regards to transportation)	Sludge, Filter Cake, Filter Socks, Scale, Cuttings
Volumetric Liquids	5 pCi/L Total Radium (Ra-226 + Ra-228) in drinking water	EPA Drinking Water Standard, 40 CFR 141.66	Effluent Water from Well Sites
Volumetric Liquids	60 pCi/L Total Radium (Ra-226 + Ra-228) direct discharge	U.S. Nuclear Regulatory Commission (NRC), 10 CFR Part 20 Appendix B, Table 2, Liquid Effluent	Effluent Water from Well Sites and Wastewater Facilities
Volumetric Liquids	600 pCi/L Total Radium (Ra-226 + Ra-228) discharge to sanitary sewer	U.S. NRC, 10 CFR Part 20 Appendix B, Table 2, Liquid Effluent (assumes dilution and solubility of Ra)	Effluent Water from Well Sites and Wastewater Facilities
Total Alpha Surface Contamination	100 dpm/100 cm^2	U.S. NRC, Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors (1974)—Criteria for Ra-226	Structural surfaces on well sites and within wastewater facilities, and equipment released from sites

Table 2-2. Criteria for Comparison

Parameter	Criteria	Reference	Potentially Apply to:
Total Beta Surface Contamination	1,000 dpm/100 cm^2	U.S. NRC, Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors (1974)—Criteria for natural Th including Ra-228	Structural surfaces on well sites and within wastewater facilities, and equipment released from sites
Removable Alpha Surface Contamination	20 dpm/100 cm^2 (of surface area smear sampled)	U.S. NRC, Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors (1974)—Criteria for Ra-226	Structural surfaces on well sites and within wastewater facilities, and equipment released from sites
Removable Beta Surface Contamination	200 dpm/100 cm^2 (of surface area smear sampled)	U.S. NRC, Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors (1974)—Criteria for natural Th including Ra-228	Structural surfaces on well sites and within wastewater facilities, and equipment released from sites
Volumetric Gas	4 pCi/L	EPA, 402/K-12/002, A Citizen's Guide to Radon (2012)	Buildings, General Public
Volumetric Gas	30 pCi/L Derived Air Concentration (DAC)	U.S. NRC, 10 CFR Part 20 Appendix B, Table 1, Col 3	Occupational Exposure
Volumetric Gas	100 pCi/L	Occupational Safety and Health Administration (OSHA) 29 CFR 1910.1096	General Public Workforce
Annual Exposure	25 mrem/year plus as low as reasonably achievable (ALARA)	U.S. NRC, 10 CFR 20.1402-20.1403, Radiological Criteria for Unrestricted Use	General Public
Annual Exposure	100 mrem/year	U.S. NRC, 10 CFR 20.1301, Radiation Dose Limits for Members of the Public	General Public Workers not trained as Radiation Workers, i.e., well site and water facilities workers

Table 2-2. Criteria for Comparison

Parameter	Criteria	Reference	Potentially Apply to:
Annual Exposure	5,000 mrem/year	U.S. NRC, 10 CFR 20.1201, Occupational Dose Limits for Adults	Radiation Workers

Table 2-3. Natural Background Radioactivity Values for U.S. Soil

Material	U-238 (pCi/g)	Ra-226 (pCi/g)	Th-232 (pCi/g)	K-40 (pCi/g)
Soil (Average)[a]	0.95	1.1	0.95	10
Soil (Minimum)[a]	0.11	0.22	0.11	2.7
Soil (Maximum) [a]	3.8	4.3	3.5	19

[a]UNSCEAR, Sources and Effects of Ionizing Radiation (UNSCEAR 2000).

3.0 WELL SITES

Thirty-eight well sites, including four conventional wells and 34 unconventional wells, were sampled from June 2013 through July 2014. Data from five phases of well development and completion were collected: vertical drilling, horizontal drilling, hydraulic fracturing, flowback, and production. A listing of the well types, formations, phases, and geographic regions is provided below.

- 4 Conventional Wells
  - Formations
    - 1 in the Lower Devonian/Oriskany
    - 3 in the Upper Devonian
  - Phase
    - Production Phase
- 34 Unconventional Wells
  - Formations
    - 29 in the Lower Devonian/Marcellus
    - 2 in the Lower Devonian/Marcellus Sandstone
    - 1 in the Upper Devonian/Burket
    - 2 in the Middle Ordovician/Utica
  - Phases
    - 10 sampled during the vertical drilling phase
    - 10 sampled during the horizontal drilling phase
    - 10 sampled during the hydraulic fracturing phase
    - 9 sampled during the flowback phase
    - 19 sampled during the production phase
      - 9 sampled for fluids and Rn
      - 10 sampled for just Rn
  - Regions
    - 1 in the Northeast Region
    - 17 in the North-central Region
    - 4 in the Northwest Region
    - 16 in the Southwest Region

3.1 Radiological Survey Results

Radiological surveys were conducted at each well site resulting in four data sets:

- *Removable α/β surface radioactivity* measurements recorded in units of dpm/100 cm^2
- *Total α/β surface radioactivity* measurements recorded in units of dpm/100 cm^2
- *Gross Gamma Radiation Scan* measurements recorded in units of cpm
- *Gamma Radiation Exposure Rate* measurements recorded in units of μR/hr

3.1.1 Removable Alpha/Beta Surface Radioactivity Measurement Results

Measurements of removable α/β surface radioactivity were performed to assess potential internal radiation worker exposure through ingestion and/or inhalation. The results were evaluated using

the NRC Regulatory Guide 1.86 (RG 1.86) guidelines. RG 1.86 Table 1 requires that α and β levels be evaluated separately. The primary α emitter of concern is Ra-226 with a removable criterion of 20 dpm α/100 cm^2. The primary β emitter of concern is Ra-228 of the natural Th decay series with a removable criterion of 200 dpm β/100 cm^2. The average removable α and β levels at each well site were below the RG 1.86 criteria. The maximum removable α and β levels were 14.9 dpm/100 cm^2 and 123 dpm/100 cm^2, respectively, also below the RG 1.86 criteria. The summary results of removable α/β radioactivity for each of the well sites surveyed are presented in **Table 3-1.** Individual smear sample removable α/β results are presented in **Appendix D.**

3.1.2 Total Alpha/Beta Surface Radioactivity Measurement Results

Measurements of total α/β surface radioactivity were performed to assess potential worker internal radiation exposure through ingestion and/or inhalation. The results were evaluated using the RG 1.86 Table 1 guidelines. RG 1.86 requires that α and β activity be evaluated separately. The primary α emitter of concern is Ra-226 with a total criterion of 100 dpm α/100 cm^2. The primary β emitter of concern is Ra-228 of the natural Th decay series with a total criterion of 1,000 dpm β/100 cm^2. The maximum average total α and β levels measured at any single well site were 93.0 dpm/100 cm^2 and 1,630 dpm/100 cm^2. The maximum total α and β levels measured were 754 dpm/100 cm^2 and 2,503 dpm/100 cm^2. The summary results of total α and β surface radioactivity for each of the well sites surveyed are presented in **Table 3-2**. Individual total α/β measurement results are presented in **Appendix D.**

3.1.3 Gross Gamma Radiation Scan Results

Gross gamma radiation scans recorded in cpm were performed on well sites to identify areas of radioactivity above local background levels. Summary results for each of the well sites surveyed and each phase surveyed are presented in **Table 3-3**. The highest average gross gamma radiation count rate was 14,519 cpm (approximately 18 μR/h), and the maximum gamma radiation scan result measured was 30,823 cpm (approximately 39 μR/h). A graphic display of the gamma radiation scan results (figures) at each facility was prepared using geographic information system (GIS) software. Figures are presented in **Appendix E**.

3.1.4 Gamma Radiation Exposure Rate Results

Gross gamma radiation scan results in units of cpm presented in **Table 3-3** were converted to μR/hr using the 800 cpm per μR/hr conversion factor appropriate for Ra-226 gamma energy as detected with 2-inch by 2-inch NaI detectors, rounded to one significant figure (Table 6.4, NaI Scintillation Detector Scan MDCs for Common Radiological Contaminants, NUREG-1507, Minimum Detectable Concentrations With Typical Radiation Survey Instruments for Various Contaminants and Field Conditions, USNRC June 1998). The exposure rate results for each well site are presented in **Table 3-4**. The highest average exposure rate measured at any single site was 18.1 μR/hr, and the maximum gamma exposure rate measured was 38.5 μR/hr.

3.2 Solid Sample Results

3.2.1 Vertical Phase Drill Cuttings

Vertical cuttings were sampled at 11 unconventional well sites and analyzed using gamma spectroscopy to identify gamma-emitting members of the natural U, Th, and Ac decay series. The gamma spectroscopy results are presented in **Table 3-5**. XRF analysis was also performed on the vertical drill cuttings to identify non-gamma-emitting isotopes of U-238 and Th-232. XRF ppm concentration data for Th was converted to pCi/g of Th-232 using the specific activity of 0.110 pCi/g Th-232 per ppm of Th. XRF ppm concentration data for U was converted to pCi/g of U-238 using the specific activity of 0.334 pCi/g U-238 per ppm of U. Both the ppm and the pCi/g results for 10 well sites are presented in **Table 3-6**. All of the XRF analytical results are presented in **Appendix F**.

There were two methods for managing drill cuttings at the well sites. The first method, called a "half round," accumulates cuttings in a large mixing container where the materials were stabilized prior to shipment to the landfill. This method does not provide an opportunity to collect samples at discrete depths; consequently, a composited sample was collected during vertical drilling. This method was used at nine of the 10 well sites.

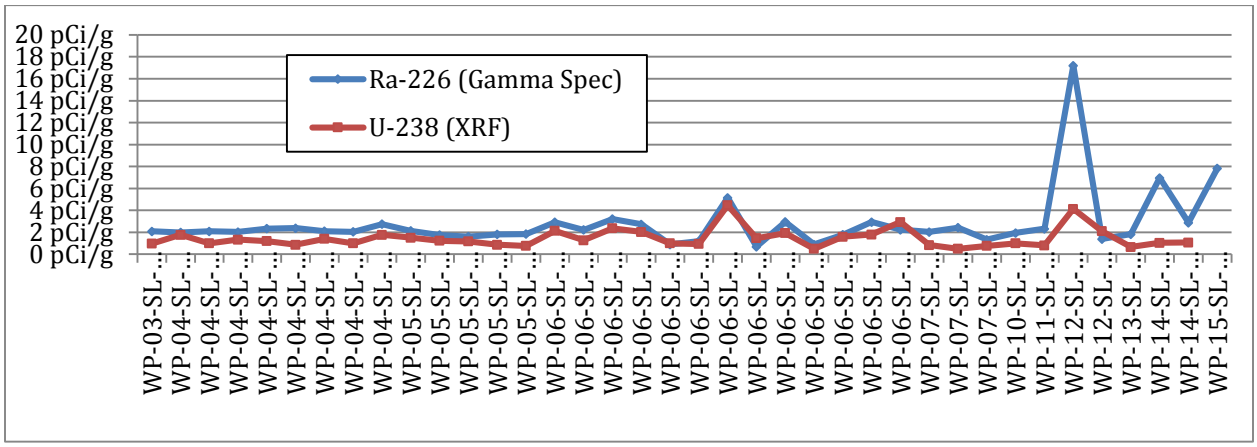
The second method loads the cuttings into roll-off containers from the shaker tables. This method enables sampling of cuttings from discrete depths. Each container was labeled with the start and end depth of the collected material. The formations sampled are presented in **Table 3-6** for these vertical drill cuttings. This method was used at one well site.

The U series activities are variable because the vertical cuttings represent different geologic formations lying above the target natural gas-containing shale. These vertical drill cuttings are mostly siltstones and sandstones. Potassium-40 (K-40) concentrations provide an indication of the type of formation. Shale has higher levels of K-40 than sandstone. Shale is typically in the range of 25-30 pCi/g of K-40 while sandstone typically contains approximately 5 pCi/g of K-40.

The U-238 measured using XRF and the Ra-226 measured using gamma spectroscopy were compared to confirm secular equilibrium of the U decay series within drill cuttings. **Figure 3-1** provides a graphic representation of this comparison and shows agreement between the two U series radionuclides, indicating secular equilibrium. Although the gamma spectroscopy results for Ra-226 are consistently higher than the XRF results for U-238, both values trend together, i.e., increase and decrease together. The high bias of the Ra-226 gamma spectroscopy results is due in part from the U-235 interference when identifying Ra-226 using gamma spectroscopy of the 186 keV gamma line. (Refer to Section 2.3.2 for a complete discussion of Ra-226 detection using gamma spectroscopy.) U-235, which is also present in drill cuttings, also emits gamma at 186 keV, causing a consistent positive bias of Ra-226 results.

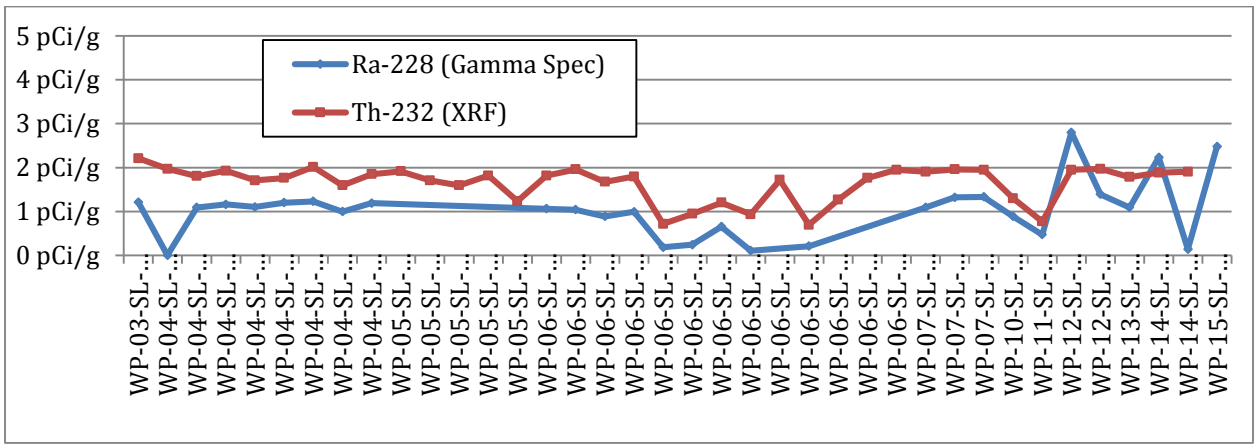
Th-232 and Ra-228 do not emit gamma rays identifiable by gamma spectroscopy; consequently, the levels were inferred from the Ac-228 gamma rays. The Th-232 series radionuclide activity levels all typify natural background for soil (reference Table 2-3).

Figure 3-1. Comparison of Ra-226 Gamma Spectroscopy Results to U-238 XRF Results in Vertical Drill Cuttings



The Th-232 identified using XRF and the Ra-228 inferred using gamma spectroscopy were compared to confirm secular equilibrium of the Th decay series within drill cuttings. **Figure 3-2** provides a graphic representation of this comparison and shows agreement between the two Th series radionuclides.

Figure 3-2. Comparison of Ra-228 Gamma Spectroscopy Results to Th-232 XRF Results in Vertical Drill Cuttings



The Th-232 to Ra-228 values for most samples trend together, i.e., when the activity concentration of one increases, there is a comparable increase in the other.

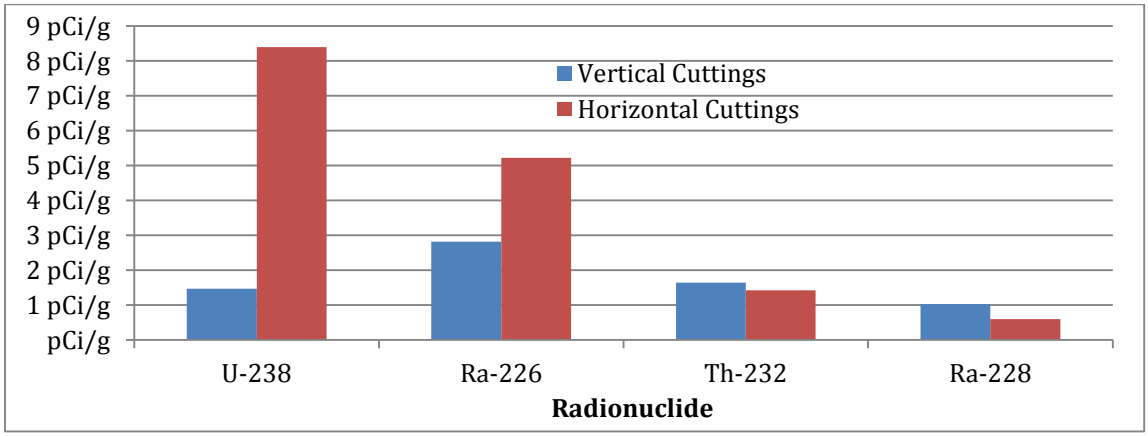
3.2.2 Horizontal Phase Drill Cuttings

The same two cuttings management methods described for vertical drill cuttings were also used for horizontal drill cuttings. A total of 18 samples were collected from the horizontal well bore target formations on 10 well sites. The gamma spectroscopy and XRF results are presented in **Tables 3-7** and **3-8**.

Figure 3-3 presents the analytical results for vertical and horizontal cutting samples. The horizontal drill cuttings had higher concentrations of Ra-226 than the vertical drill cuttings as

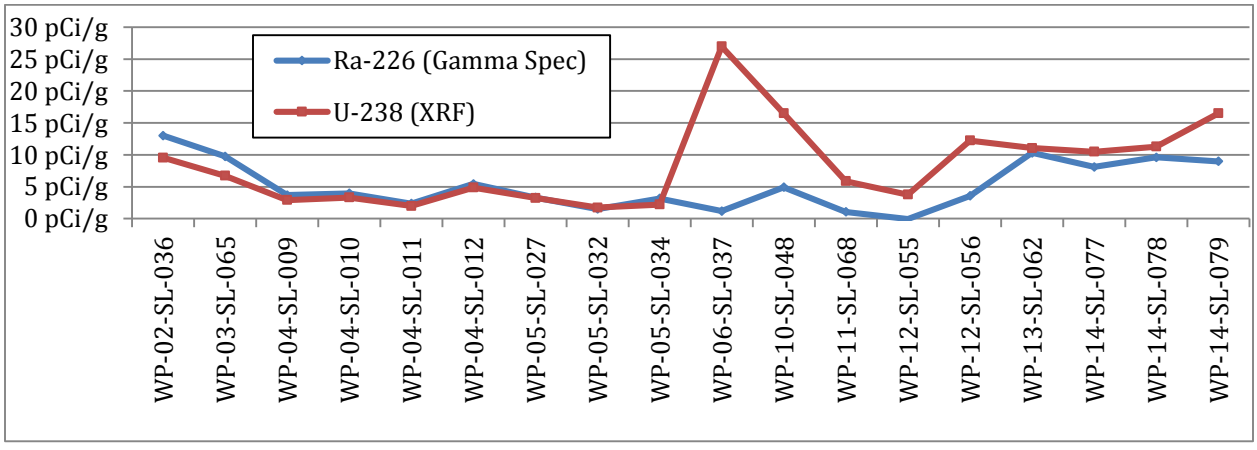
determined using a student t-test. The two-sample student t-test was used to compare the horizontal drill cuttings Ra-226 results with the vertical drill cuttings Ra-226 results. ProUCL version 5.0 was used to perform the student t-test on the data. The Null Hypothesis tested is that the mean value of the vertical drill cuttings Ra-226 results and the mean value of the horizontal drill cuttings Ra-226 results are statistically different at the 95 percent confidence level. The Null Hypothesis was accepted; mean values are statistically different at the 95 percent confidence level. The same t-test was run on the U-238 results for vertical and horizontal drill cuttings. Again, the difference between the mean values of U-238 for vertical and horizontal drill cuttings is statistically different at the 95 percent confidence level. **Appendix G** presents the t-test output files.

Figure 3-3. Comparison of Analytical Analyses Results for Horizontal and Vertical Drill Cutting Samples



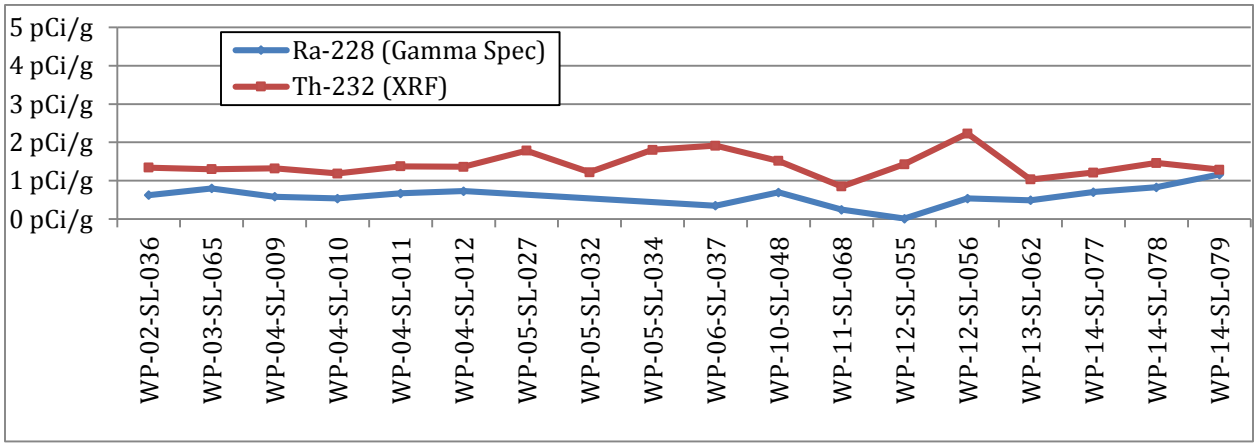
The U concentration (ppm) measured using XRF was converted to pCi/g of U-238 using the specific activity of 0.334 pCi/g U-238 per ppm of U. The U-238 measured using XRF and the Ra-226 measured using gamma spectroscopy were compared to confirm secular equilibrium of the U decay series within drill cuttings. **Figure 3-4** provides a graphic representation of this comparison and shows agreement between the two U series radionuclides, indicating secular equilibrium.

Figure 3-4. Comparison of Ra-226 Gamma Spectroscopy Results to U-238 XRF Results in Horizontal Drill Cuttings



The Th concentration (ppm) measured using XRF was converted to pCi/g of Th-232 using the specific activity of 0.110 pCi/g Th-232 per ppm of Th. The Th-232 measured using XRF and the Ra-228 inferred using gamma spectroscopy were compared to confirm secular equilibrium of the Th decay series within drill cuttings. **Figure 3-5** provides a graphic representation of this comparison.

Figure 3-5. Comparison of Ra-228 Gamma Spectroscopy Results to Th-232 XRF Results in Horizontal Drill Cuttings



The Th-232 to Ra-228 values trend together, i.e., when the activity concentration of one increases, there is a comparable increase in the other.

3.2.3 Drilling Mud

In addition to drill cuttings, drilling mud was also collected when in use on the sites. A total of 14 drilling mud samples were collected during both the vertical and horizontal phases of drilling. The drilling mud was evaluated as a drilling solid or a drilling liquid as determined when received by the laboratory. Nine of those samples were analyzed as solids and the other five as liquids. The gamma spectroscopy results for solids are presented in **Table 3-9**.

Analytical results for the drilling mud demonstrate secular equilibrium within the U and Th natural decay series, i.e., the activity concentrations within the natural series radionuclides identified are approximately equal. All results were within the range of typical natural background found in surface soils (reference Table 2-3), given the overestimation of Ra-226 in the presence of U-235 as discussed in Section 2.3.2.

3.2.4 Hydraulic Fracturing Proppant Sand

During hydraulic fracturing, 10 well sites were surveyed and sampled. The proppant sand was collected from the sand hoppers prior to being mixed with fluids and injected into the well. The gamma spectroscopy results are presented in **Table 3-10**.

The sand contained nominal concentrations of U and Th series. The sand did not contain radioactivity exceeding that of natural background levels found in surface soil (reference **Table 2-3**).

3.2.5 Flowback Solids

A total of eight well sites were surveyed and sampled during the flowback phase. From the eight well sites, sufficient volumes to perform analytical analysis of solids were only present at four of the eight well sites. The gamma spectroscopy results are presented in **Table 3-11**.

Uranium and Th are at or below background activity levels. Radium-226 was elevated above background levels for soil (reference Table 2-3) ranging from 0.763 to 7.73 pCi/g.

3.3 Liquid Sample Results

Liquid sampling included drilling mud, hydraulic fracturing fluids, flowback fluids, and produced water.

3.3.1 Drilling Liquid (Mud)

A total of 14 drilling mud samples were collected from both vertical and horizontal phases. The drilling mud was evaluated as a drilling solid or a drilling liquid as determined when received by the laboratory. Five of the samples were analyzed as liquids. Because of the large concentrations of solids in the samples, gross α and gross β analyses were performed on only two samples. The results for Ra-226, Ra-228, K-40, gross α and gross β are presented in **Table 3-12**.

3.3.2 Hydraulic Fracturing Fluid

Hydraulic fracturing fluid was sampled prior to injection into the well. The well sites sampled during the study utilized hydraulic fracturing fluid made up of either fresh water, reused flowback liquid, produced water, or a combination of the three to perform the hydraulic fracturing phase. If a combination of fluids was used for fracturing, only the produced water was collected as a sample because it was not possible to collect a sample after the hydraulic fracturing fluid had been mixed for injection. The results for Ra-226, Ra-228, K-40, gross α and gross β are presented in **Table 3-13**.

Radium-226 was detected within the hydraulic fracturing fluid ranging from 64.0 to 21,000 pCi/L. Ra-228 was also detected ranging from 4.50 to 1,640 pCi/L. Table 2-2 contains several volumetric liquids criteria for relative comparison: 5 pCi/L total Ra EPA maximum contaminant level for drinking water, 60 pCi/L total Ra USNRC direct discharge, and 600 pCi/L total Ra USNRC discharge to sanitary sewer.

3.3.3 Flowback Fluid

Flowback fluid is the injected hydraulic fracturing fluid and other fluids returning to the surface of the well prior to the well entering production. The results for Ra-226, Ra-228, K-40, gross α and gross β are presented in **Table 3-14**.

Radium-226 concentrations were elevated, ranging from 551 to 25,500 pCi/L. Radium-228 was also elevated, ranging from 248 to 1,740 pCi/L. Table 2-2 contains several volumetric liquids criteria for relative comparison: 5 pCi/L total Ra EPA drinking water, 60 pCi/L total Ra USNRC direct discharge, and 600 pCi/L total Ra USNRC discharge to sanitary sewer.

3.3.4 Produced Water

Twelve wells were sampled for produced water, including four conventional and eight unconventional wells. The results for unfiltered and filtered Ra-226, Ra-228, K-40, gross α and gross β are presented in **Tables 3-15** and **3-16**.

Radium-226 concentrations in unfiltered samples were elevated, ranging from 40.5 to 26,600 pCi/L. Radium-228 concentrations were also elevated, ranging from 26.0 to 1,900 pCi/L.

Radium-226 concentrations were also elevated in filtered samples, ranging from 87.0 to 24,100 pCi/L. Radium-228 concentrations were also elevated, ranging from 44.0 to 1,860 pCi/L.

3.4 Radon Sample Results

3.4.1 Ambient Air Samples During Flowback

Seventeen ambient air samples for evaluation of Rn concentration were collected during flowback at four different well sites. The EICs were distributed around the well site approximately 3 feet (ft) above grade and at available locations as close as 6 ft and as far as 40 ft from the well head. The EICs collected data from four to seven days. The results are presented in **Table 3-17**. The Rn analytical reports are presented in **Appendix H**.

The Rn measurement results during flowback in ambient air range from 0.200 to 1.70 pCi/L while typical ambient background Rn concentrations range from 0.00 to 1.11 pCi/L (with a median value of 0.39 pCi/L) in outdoor ambient air in the U.S., as reported by EPA.

3.4.2 Production Gas Radon

Twenty-two production site natural gas samples were collected in eight counties (Washington, Tioga, Lycoming, McKean, Forest, Sullivan, Bradford and Jefferson). Seventeen of the natural gas samples were collected from Marcellus Shale, and five natural gas samples were collected from other geologic formations.

The production site natural gas samples for Rn were collected between the well head and the separator unit(s). A typical sampling location is shown in **Figure 3-6**. All natural gas samples were collected directly into scintillation cells, referred to as Lucas Cells. Section 2.0 describes the sample collection in detail.

The sample results are presented in **Table 3-18.** The results ranged from 3.00 to 148 pCi/L. The median Rn concentration in natural gas is 41.8 pCi/L. The Rn analysis analytical reports are presented in **Appendix H**.

3.5 Well Site Worker Exposure Assessment

The study included radiation measurements collected on 21 well sites to provide a comprehensive evaluation of potential personnel radiation exposure from working on well sites. The measurements included:

Figure 3-6. Natural Gas Radon Sampling Location

- Gamma radiation count rate using a NaI detector (gross cpm), converted to exposure rate potential, to estimate potential external gamma exposure.
- Total α/β surface radioactivity measurements using a scintillation detector to evaluate potential β external exposure as well as α/β surface activity having the potential to become removable and, therefore, becoming a potential internal exposure.
- Removable α/β surface radioactivity measurements (dpm/100 cm^2) by smear samples counted on an α/β counter to estimate potential α and β internal exposure.
- Ambient air samples analyzed for Rn concentration to estimate Rn inhalation exposure.

The measurements were taken during four work phases on natural gas well sites to ensure appropriate evaluation of potential exposure to TENORM present on well sites. The phases are:

- Vertical/Horizontal Drilling – personnel are potentially exposed to drill cuttings while working on the site.
- Hydraulic Fracturing – personnel are potentially exposed to radioactivity in hydraulic fracturing fluid while working on the site.
- Flowback – personnel are potentially exposed to radioactivity in flowback water while working on the site.
- Production – personnel are potentially exposed to radioactivity in produced water while working on the site.

3.5.1 External Gamma Exposure

Gross gamma scan results in units of cpm presented in **Table 3-3** were converted to μR/hr using the 800 cpm per μR/hr conversion factor appropriate for Ra-226 gamma energy as detected with 2-inch by 2-inch NaI detectors [Table 6.3, NaI Scintillation Detector Count Rate Versus Exposure Rate (cpm/μR/hr), NUREG-1507, Minimum Detectable Concentrations With Typical Radiation Survey Instruments for Various Contaminants and Field Conditions, USNRC June 1998]. The local background gamma exposure rate across all well sites surveyed was measured at 5 μR/hr. The exposure rate results are presented in **Table 3-4**.

The lowest exposure rates measured and the maximum exposure time were during drilling. The highest exposure rates measured were in the proximity of holding tanks for produced water. The gamma dose rates during drilling ranged from background (measured at 5 μR/hr) to a maximum of 38.5 μR/hr, and the highest average exposure rate at any of the well sites was 18.1 μR/hr. Assuming the time period of exposure is a full occupational year of 2,000 hours, the average well site external gamma exposure was estimated as follows:

Maximum Average Well Site External Gamma Exposure Estimate

$$(18.1 - 5)\ \mu R/hr \times 2000\ hr/yr \times (1\ mrem/1{,}000\ \mu R\ gamma) = 26.2\ mrem/yr$$

The result is less than the 100 mrem/yr dose equivalent limit for a member of the public. Actual exposure is dependent upon the actual exposure rates and occupancy time for individual workers.

3.5.2 Internal Alpha/Beta Exposure

Results for α/β surface radioactivity measurements are provided in Sections 3.1.1 and 3.1.2. Ten of the 491 α measurements and 69 of the 491 β measurements of total surface radioactivity exceeded the RG 1.86 criteria. Only one of 493 α removable surface activity measurements and one of 493 β removable surface radioactivity measurements exceeded RG 1.86 criteria, indicating the total α/β surface radioactivity measured is fixed to the surface and not readily available for inhalation or ingestion.

3.5.3 Internal Radon Exposure

The Rn measurement results in ambient air during flowback range from 0.200 to 1.70 pCi/L, while typical ambient background Rn concentrations range from 0.00 to 1.11 pCi/L, with a median of 0.39 pCi/L in outdoor ambient air in the U.S., as reported by EPA.

3.6 Well Site Data Assessments

3.6.1 Comparison of Different Geological Formations Based on X-Ray Fluorescence Data

Eighteen drill cutting samples were collected and analyzed for Th and U using XRF. The samples were collected from the Lower Devonian/Marcellus, Upper Devonian/Burket, and the Middle Ordovician/Utica geologic formations. The data for the three geologic formations, including the average, median, standard deviation, and ratios of Th to U are presented in **Table 3-19**.

XRF ppm concentration data for Th was converted to pCi/g of Th-232 using the specific activity value of 0.110 pCi/g Th-232 per ppm of Th. XRF ppm concentration data for U was converted to pCi/g of U-238 using the specific activity value of 0.334 pCi/g of U-238 per ppm of U. Ratios of U/Th are also presented in **Table 3-19**.

3.6.2 Filtered Versus Unfiltered Sample Data Evaluation

Appendix I contains the assessment of filtered and unfiltered liquid sample results for the entire TENORM study. The conclusion from this evaluation is that there is no apparent trend or bias that filtering produces. There were some subsets of data where either the unfiltered results or the filtered results appear to be significantly higher. There was no statistically significant correlation found within any sample group. Because the liquid samples were preserved by addition of acid prior to filtering, the radioactive particulates may have entered solution and were therefore not removed by filtering.

3.6.3 Conventional Versus Unconventional Produced Water Data Evaluation

There was a significant difference observed in the produced water from conventional and unconventional O&G well sites. **Tables 3-15** and **3-16** present gamma spectroscopy results for conventional and unconventional produced water for both filtered and unfiltered samples. Two distinct differences in magnitude of activity and in the ratio of Ra-226 to Ra-228 are summarized in **Figure 3-7**.

Figure 3-7. Conventional vs Unconventional Produced Water Radium Concentrations

O&G Production	Filtered Samples	No. of Samples	Average Ra-226 (pCi/L)	Average Ra-228 (pCi/L)	Ratio of Ra-226/Ra-228
Conventional	No	4	336	295	1.14
Unconventional	No	9	8,340	986	8.46
Conventional	Yes	4	334	288	1.16
Unconventional	Yes	9	8,220	985	8.35

The Ra-226 activity in unconventional well site produced water is approximately 20 times greater than that observed in conventional well site produced water. The ratio of Ra-226 to Ra-228 in unconventional well site produced water is approximately eight times greater than that found in conventional well site produced water. The higher ratio of Ra-226 to Ra-228 for unconventional well site produced water reflects the higher ratio of U to Th observed in Marcellus Shale horizontal cuttings sample results. The U to Th ratio is approximately six. Filtering of the samples does not appreciably change the activity concentration or the relationship between Ra-226 and Ra-228.

3.7 Potential Offsite Environmental Impact

A potential offsite environmental impact could result from the removal of materials and/or equipment with total and/or removable α/β surface radioactivity above applicable guidelines. The highest total α surface radioactivity measurement was 754 dpm/100 cm^2. Additional measurements exceeded the RG 1.86 Ra-226 total surface contamination guideline of 100 dpm/100 cm^2. The highest total β measurement was 2,503 dpm/100 cm^2. This and several other measurements exceeded the RG 1.86 Th-232 total surface contamination guideline of

1,000 dpm/100 cm^2. These readings were on equipment associated with wastewater handling/storage, and this equipment is likely to be reused.

Table 3-1. Removable Alpha and Beta Surface Radioactivity Measurement Results Summary[a,b]

Study ID	No. of Data Points	Removable Alpha (dpm/100 cm^2)				Removable Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WP-01-FS-045	12	4.24	12.4	2.36	4.92	93.7	93.7	0.00	93.7
WP-01-FS-081	7	4.24	4.24	0.00	4.24	102	102	0.00	102
WP-01-FS-128	3	4.24	12.4	4.79	6.96	118	118	0.00	118
WP-02-FS-083	27	4.24	4.24	0.00	4.24	102	102	0.00	102
WP-03-FS-029	15	4.15	4.15	0.78	4.15	109	109	0.00	109
WP-03-FS-082	14	4.24	4.24	0.00	4.24	86.7	86.7	0.00	86.7
WP-04-FS-014	10	4.24	7.24	1.07	4.24	93.7	93.7	0.00	93.7
WP-04-FS-084	22	4.24	4.24	0.18	4.24	95.7	95.7	0.00	95.7
WP-04-FS-085	29	4.24	4.24	0.00	4.24	108	108	0.00	108
WP-05-FS-077	3	4.15	4.15	0.00	4.15	113	113	0.00	113
WP-05-FS-089	26	4.15	4.15	0.00	4.15	98.9	98.9	0.00	98.9
WP-06-FS-026	3	4.14	4.14	0.00	4.14	112	112	0.00	112
WP-06-FS-091	29	4.24	4.24	0.00	4.24	102	102	0.00	102
WP-06-FS-092	23	4.24	4.24	0.00	4.24	95.5	95.5	0.00	95.5
WP-06-FS-093	4	4.15	4.15	0.00	4.15	111	111	0.00	111
WP-07-FS-094	12	4.24	4.24	0.00	4.24	102	102	0.00	102
WP-08-FS-010	5	4.24	4.24	0.00	4.24	123	123	0.00	123
WP-08-FS-095	5	4.24	4.24	0.00	4.24	102	102	0.00	102
WP-09-FS-097	7	4.15	4.15	0.00	4.24	102	102	0.00	102
WP-09-FS-098	3	4.24	4.24	0.00	4.24	113	113	0.00	113
WP-10-FS-003	21	4.24	4.24	0.00	4.24	93.7	93.7	0.00	93.7
WP-10-FS-004	21	4.14	4.15	0.00	4.15	93.7	93.7	0.00	93.7
WP-10-FS-009	8	4.15	4.15	0.00	4.15	113	113	0.00	113
WP-11-FS-023	17	4.15	4.15	0.00	4.15	109	109	0.00	109
WP-11-FS-037	15	4.15	4.15	0.00	4.15	113	113	0.00	113

Table 3-1. Removable Alpha and Beta Surface Radioactivity Measurement Results Summary[a,b]

Study ID	No. of Data Points	Removable Alpha (dpm/100 cm^2)				Removable Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WP-11-FS-102	17	4.15	4.15	0.00	4.15	109	109	0.00	109
WP-12-FS-017	23	4.24	4.24	0.00	4.24	93.7	93.7	0.00	93.7
WP-12-FS-018	4	4.14	4.14	0.00	4.14	113	113	0.00	113
WP-12-FS-019	19	4.24	4.24	0.00	4.24	93.7	93.7	0.00	93.7
WP-13-FS-041	17	4.24	4.24	0.00	4.24	123	123	0.00	123
WP-13-FS-042	18	4.24	4.24	0.00	4.24	123	123	0.00	123
WP-14-FS-035	20	4.15	12.2	2.47	4.96	114	114	0.00	114
WP-14-FS-036	23	4.15	14.9	2.36	4.62	114	114	0.00	114
WP-14-FS-107	4	4.15	4.15	0.00	4.15	114	114	0.00	114
WP-15-FS-028	7	4.15	4.15	0.00	4.15	114	114	0.00	114

[a]Smear samples were performed on facility, system, and structure surfaces.
[b]During the calculations to convert from raw counts to dpm, the calculated value was compared to half of the MDC. If the value was below this number, half of the MDC was inserted into the tables. Where the standard deviation is zero and the minimum, maximum, and average are the same, then all measurements were below half of the MDC.

Table 3-2. Total Alpha and Beta Surface Radioactivity Measurement Results Summary[a,b]

Study ID	No. of Data Points	Total Alpha (dpm/100 cm^2)				Total Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WP-01-FS-045	12	30.5	30.5	0.00	30.5	364	2,503	618	1,190
WP-01-FS-081	7	19.0	19.0	0.00	19.0	279	1,710	460	777
WP-02-FS-083	27	7.44	14.9	2.84	8.82	288	676	75.5	305
WP-03-FS-029	16	30.5	30.5	0.00	30.5	357	884	137	587
WP-03-FS-082	14	7.44	79.0	19.2	16.0	266	364	28.6	282
WP-04-FS-014	10	30.5	30.5	0.00	30.5	364	2,220	812	1,170
WP-04-FS-084	22	7.46	69.6	16.4	13.7	325	325	0.00	325
WP-04-FS-085	29	7.46	29.8	7.06	11.3	317	651	69.6	337
WP-05-FS-077	3	30.5	30.5	0.00	30.5	513	698	96.0	592
WP-05-FS-089	26	7.46	164	46.6	26.0	280	542	59.0	299
WP-06-FS-026	30.5	30.5	30.5	0.00	30.5	646	1,920	652	1,200
WP-06-FS-091	29	7.46	24.9	4.00	8.92	297	297	0.00	297
WP-06-FS-092	23	7.44	44.6	8.00	9.38	278	527	54.0	292
WP-06-FS-093	4	30.5	30.5	0.00	30.5	268	268	0.00	268
WP-07-FS-094	12	7.44	19.8	4.16	9.71	291	988	201	349
WP-08-FS-010	5	30.5	30.5	0.00	30.5	557	721	65.0	624
WP-08-FS-095	5	19.0	19.0	0.00	19.0	279	279	0.00	279
WP-09-FS-097	7	7.44	29.8	8.69	13.5	285	285	0.00	285
WP-09-FS-098	3	30.5	30.5	0.00	30.5	268	268	0.00	268
WP-10-FS-003	21	30.5	754	167	93.0	268	1,580	417	676
WP-10-FS-004	21	30.5	258	69.4	60.0	268	1,580	410	709
WP-10-FS-009	8	30.5	30.5	0.00	30.5	1,390	1,890	145	1,630
WP-11-FS-023	17	30.5	30.5	0.00	30.5	364	1,410	294	966
WP-11-FS-037	15	30.5	30.5	0.00	30.5	268	1,020	223	583
WP-11-FS-102	17	30.5	30.5	0.00	30.5	268	1,410	294	960

Table 3-2. Total Alpha and Beta Surface Radioactivity Measurement Results Summary[a,b]

Study ID	No. of Data Points	Total Alpha (dpm/100 cm^2)				Total Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WP-12-FS-017	23	30.5	30.5	0.00	30.5	268	884	145	374
WP-12-FS-018	4	30.5	30.5	0.00	30.5	839	951	51.0	910
WP-12-FS-019	19	30.5	30.5	0.00	30.5	268	1,550	378	513
WP-13-FS-041	17	30.5	30.5	0.00	30.5	371	1,430	303	1,010
WP-13-FS-042	18	30.5	30.5	0.00	30.5	275	1,380	273	799
WP-14-FS-035	20	30.5	30.5	0.00	30.5	268	721	130	315
WP-14-FS-036	23	30.5	30.5	0.00	30.5	268	483	55.0	284
WP-14-FS-107	4	27.8	27.8	0.00	27.8	69.3	69.3	0.00	69.3
WP-15-FS-028	6	30.5	30.5	0.00	30.5	268	268	0.00	268

[a]Static measurements were performed on facility, system, and structure surfaces.
[b]During the calculations to convert from raw counts to dpm, the calculated value was compared to half of the MDC. If the value was below this number, half of the MDC was inserted into the tables. Where the standard deviation is zero and the minimum, maximum, and average are the same, then all measurements were below half of the MDC.

Table 3-3. Gross Gamma Scan Results Summary[a]

Site	Phase	Scan Max[b] (cpm)	Scan Min[b] (cpm)	Scan Average[b] (cpm)	Scan Std Dev (cpm)	No. Data Points
WP-01	Fracturing	16,608	7,209	13,028	1,349	4,857
WP-01	Flowback	17,299	6,653	14,519	1,246	4,474
WP-01	Production	16,641	9,019	13,787	1,075	4,891
WP-02	Horizontal	9,363	4,262	5,371	1,041	8,318
WP-03	Vertical	13,650	4,758	7,254	1,531	7,438
WP-04	Vertical	15,961	7,249	13,378	902	7,083
WP-04	Horizontal	16,099	7,210	13,260	1,139	6,470
WP-04	Fracturing	22,724	8,055	14,322	1,234	4,554
WP-04	Flowback	17,057	10,982	13,938	750	5,411
WP-04	Production	17,031	8,545	13,019	895	3,624
WP-05	Horizontal	9,394	3,181	7,236	724	5,552
WP-05	Fracturing	8,293	3,925	6,668	825	3,033
WP-06	Vertical	8,906	4,424	6,357	560	8,518
WP-06	Horizontal	8,280	4,756	6,097	356	8,562
WP-06	Flowback	8,231	4,722	6,014	464	5,037
WP-06	Fracturing	10,803	3,049	8,033	692	2,532
WP-07	Vertical	8,437	4,675	6,318	483	12,519
WP-08	Fracturing	7,454	3,710	5,387	470	4,602
WP-09	Fracturing	30,823	2,686	5,380	1,146	4,354
WP-10	Horizontal	15,258	8,924	12,916	970	3,440
WP-10	Flowback	16,013	8,508	13,817	790	1,856
WP-10	Production	16,528	10,447	13,257	835	2,946
WP-11	Vertical	15,603	10,050	12,412	771	3,091
WP-11	Horizontal	14,781	4,368	12,075	1,252	2,960
WP-11	Production	13,505	9,914	12,281	503	1,168
WP-12	Vertical	11,479	5,543	8,005	1,144	3,204
WP-12	Horizontal	11,360	5,328	8,034	1,073	3,525
WP-13	Vertical	15,088	8,068	13,096	628	2,924
WP-13	Horizontal	15,357	8,119	12,916	966	3,234
WP-14	Vertical	6,772	1,992	3,854	684	2,840
WP-14	Horizontal	5,891	2,302	3,449	468	1,821
WP-14	Flowback	7,421	3,181	4,421	648	3,273
WP-15	Vertical	8,557	4,398	6,093	573	2,230
WP-16	Production	10,833	4,623	7,753	1,361	290
WP-17	Production	8,797	4,183	6,179	907	277
WP-19	Production	7,046	2,494	4,314	1,013	238
WP-20	Production	5,422	2,790	4,166	537	366

Table 3-3. Gross Gamma Scan Results Summary[a]

Site	Phase	Scan Max[b] (cpm)	Scan Min[b] (cpm)	Scan Average[b] (cpm)	Scan Std Dev (cpm)	No. Data Points
WP-21	Production	5,307	2,677	3,870	572	182

[a] Gross gamma scans were performed on site ground surfaces outside facilities, structures, and systems, and include soil, asphalt, gravel, and concrete matrices.
[b]Convert count rate data to exposure rate by dividing count rate by 800 to yield µR/hr.

Table 3-4. Results Summary of NaI Count Rate Data Converted to Exposure Rates

Site	Phase	Scan Max (µR/hr)	Scan Min (µR/hr)	Scan Average (µR/hr)	Scan Std Dev (µR/hr)	No. Data Points
WP-01	Fracturing	20.8	9.00	16.3	1.70	4,857
WP-01	Flowback	21.6	8.30	18.1	1.60	4,474
WP-01	Production	20.8	11.3	17.2	1.30	4,891
WP-02	Horizontal	11.7	5.30	6.70	1.30	8,318
WP-03	Vertical	17.1	5.90	9.10	1.90	7,438
WP-04	Vertical	20.0	9.10	16.7	1.10	7,083
WP-04	Horizontal	20.1	9.00	16.6	1.40	6,470
WP-04	Fracturing	28.4	10.1	17.9	1.50	4,554
WP-04	Flowback	21.3	13.7	17.4	0.900	5,411
WP-04	Production	21.3	10.7	16.3	1.10	3,624
WP-05	Horizontal	11.7	4.00	9.00	0.900	5,552
WP-05	Fracturing	10.4	4.90	8.30	1.00	3,033
WP-06	Vertical	11.1	5.50	7.90	0.700	8,518
WP-06	Horizontal	10.4	5.90	7.60	0.400	8,562
WP-06	Flowback	10.3	5.90	7.50	0.600	5,037
WP-06	Fracturing	13.5	3.80	10.0	0.900	2,532
WP-07	Vertical	10.5	5.80	7.90	0.600	12,519
WP-08	Fracturing	9.30	4.60	6.70	0.600	4,602
WP-09	Fracturing	38.5	3.40	6.70	1.40	4,354
WP-10	Horizontal	19.1	11.2	16.1	1.20	3,440
WP-10	Flowback	20.0	10.6	17.3	1.00	1,856
WP-10	Production	20.7	13.1	16.6	1.00	2,946
WP-11	Vertical	19.5	12.6	15.5	1.00	3,091
WP-11	Horizontal	18.5	5.50	15.1	1.60	2,960
WP-11	Production	16.9	12.4	15.4	0.600	1,168
WP-12	Vertical	14.3	6.90	10.0	1.40	3,204
WP-12	Horizontal	14.2	6.70	10.0	1.30	3,525
WP-13	Vertical	18.9	10.1	16.4	0.800	2,924
WP-13	Horizontal	19.2	10.1	16.1	1.20	3,234
WP-14	Vertical	8.50	2.50	4.80	0.900	2,840

Table 3-4. Results Summary of NaI Count Rate Data Converted to Exposure Rates

Site	Phase	Scan Max (µR/hr)	Scan Min (µR/hr)	Scan Average (µR/hr)	Scan Std Dev (µR/hr)	No. Data Points
WP-14	Horizontal	7.40	2.90	4.30	0.600	1,821
WP-14	Flowback	9.30	4.00	5.50	0.800	3,273
WP-15	Vertical	10.7	5.50	7.60	0.700	2,230
WP-16	Production	13.5	5.80	9.70	1.70	290
WP-17	Production	11.0	5.20	7.70	1.10	277
WP-19	Production	8.80	3.10	5.40	1.30	238
WP-20	Production	6.80	3.50	5.20	0.700	366
WP-21	Production	6.60	3.30	4.80	0.700	182

Table 3-5. Vertical Solids, Drill Cuttings – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)	U-238 (pCi/g)	U-235 (pCi/g)	Th-232 (pCi/g)
WP-03-SL-038	2.09	1.21	23.4	< 1.27	0.127	1.18
WP-04-SL-001	1.99	1.06	9.01	< 1.50	< 0.173	1.06
WP-04-SL-002	2.09	1.09	20.2	1.86	< 0.149	1.07
WP-04-SL-003	2.04	1.16	20.3	< 1.43	< 0.146	1.14
WP-04-SL-004	2.34	1.10	18.1	1.85	< 0.181	1.08
WP-04-SL-005	2.39	1.20	20.2	1.67	< 0.158	1.18
WP-04-SL-006	2.11	1.23	24.4	0.827	< 0.061	1.20
WP-04-SL-007	2.05	0.994	22.5	< 0.934	< 0.070	0.971
WP-04-SL-008	2.75	1.19	23.6	1.30	0.097	1.16
WP-05-SL-028	2.13	1.08	21.6	1.56	< 0.138	1.05
WP-05-SL-029	1.75	1.07	17.3	< 1.31	0.198	1.05
WP-05-SL-030	1.61	0.939	15.9	< 0.565	< 0.092	0.920
WP-05-SL-031	1.81	1.05	21.7	0.835	< 0.107	1.03
WP-05-SL-033	1.84	0.701	12.6	< 1.62	< 0.136	0.687
WP-06-SL-014	2.93	1.06	22.7	1.27	0.178	1.05
WP-06-SL-015	2.22	1.04	21.0	1.52	< 0.165	1.03
WP-06-SL-016	3.21	0.885	26.9	2.07	< 0.140	0.871
WP-06-SL-017	2.73	0.991	24.0	1.64	0.166	0.976
WP-06-SL-018	0.900	0.181	3.26	< 1.13	< 0.081	0.177
WP-06-SL-019	1.19	0.242	6.81	0.469	< 0.058	0.238
WP-06-SL-020	5.15	0.654	8.90	< 0.923	< 0.096	0.642
WP-06-SL-021	0.698	0.107	18.8	0.164	0.016	0.110
WP-06-SL-022	2.96	0.802	18.4	1.29	< 0.121	0.782
WP-06-SL-023	0.899	0.208	4.97	< 1.29	< 0.097	0.197
WP-06-SL-024	1.79	0.416	12.3	< 0.790	< 0.067	0.407

Table 3-5. Vertical Solids, Drill Cuttings – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)	U-238 (pCi/g)	U-235 (pCi/g)	Th-232 (pCi/g)
WP-06-SL-025	2.94	0.769	18.4	0.987	< 0.169	0.751
WP-06-SL-026	2.24	0.592	14.2	< 1.21	< 0.171	0.578
WP-07-SL-039	2.03	1.09	20.1	< 1.45	< 0.194	1.07
WP-07-SL-040	2.43	1.32	23.6	0.788	0.147	1.29
WP-07-SL-041	1.33	1.33	20.8	< 0.869	< 0.172	1.30
WP-10-SL-045	1.94	0.885	16.5	0.959	< 0.106	0.866
WP-11-SL-047	2.32	0.472	12.7	< 0.949	< 0.082	0.191
WP-12-SL-052	17.2	2.80	17.6	< 3.01	< 0.311	2.74
WP-12-SL-053	1.39	1.39	16.6	< 2.25	< 0.302	1.37
WP-13-SL-059	1.83	1.09	20.4	< 1.75	< 0.231	1.07
WP-14-SL-073	6.97	2.23	20.9	< 1.54	< 0.210	2.18
WP-14-SL-074	2.88	0.140	22.2	1.41	0.104	1.37
WP-15-SL-075	7.82	2.48	19.5	< 1.39	< 0.126	2.45
Average	2.82	1.01	18.0	0.960	0.085	1.01
Std. Dev.	2.79	0.572	5.64	0.484	0.046	0.555
Median	2.10	1.06	19.8	0.819	0.074	1.05
Minimum	0.698	0.107	3.26	0.164	0.016	0.110
Maximum	17.2	2.80	26.9	2.07	0.198	2.74

[a] Values reported as < are the method MDC.

Table 3-6. XRF Uranium and Thorium for Vertical Cuttings

Study ID	Date	Formation	Thorium Result (ppm)	Thorium Error (ppm)	Th-232 (pCi/g)	Uranium Result (ppm)	Uranium Error (ppm)	U-238 (pCi/g)
WP-03-SL-038	07/26/13	Varies	20.1	0.400	2.21	2.90	0.200	0.969
WP-04-SL-001	06/17/13	Varies	17.9	0.400	1.97	5.30	0.300	1.77
WP-04-SL-002	06/17/13	Varies	16.4	0.400	1.80	3.00	0.200	1.00
WP-04-SL-003	06/17/13	Varies	17.5	0.400	1.93	4.00	0.300	1.34
WP-04-SL-004	06/17/13	Varies	15.5	0.400	1.71	3.60	0.200	1.20
WP-04-SL-005	06/17/13	Varies	16.0	0.400	1.76	2.60	0.200	0.868
WP-04-SL-006	06/17/13	Varies	18.3	0.400	2.01	4.20	0.300	1.40
WP-04-SL-007	06/17/13	Varies	14.5	0.400	1.60	3.00	0.200	1.00
WP-04-SL-008	06/17/13	Varies	16.8	0.400	1.85	5.30	0.300	1.77
WP-05-SL-028	07/08/13	Varies	17.4	0.400	1.91	4.50	0.300	1.50
WP-05-SL-029	07/08/13	Varies	15.5	0.400	1.71	3.70	0.200	1.24
WP-05-SL-030	07/08/13	Varies	14.5	0.400	1.60	3.50	0.200	1.17
WP-05-SL-031	07/08/13	Varies	16.5	0.400	1.82	2.60	0.200	0.868
WP-05-SL-033	07/08/13	Varies	11.2	0.400	1.23	2.30	0.200	0.768
WP-06-SL-014	07/01/13	Varies	16.5	0.400	1.82	6.40	0.300	2.14
WP-06-SL-015	07/01/13	Varies	17.8	0.400	1.96	3.80	0.300	1.27
WP-06-SL-016	07/01/13	Varies	15.2	0.400	1.67	7.10	0.300	2.37
WP-06-SL-017	07/01/13	Varies	16.3	0.400	1.79	6.10	0.300	2.04
WP-06-SL-018	07/01/13	Varies	6.50	0.400	0.715	3.00	0.200	1.00
WP-06-SL-019	07/01/13	Varies	8.60	0.400	0.946	2.80	0.200	0.935
WP-06-SL-020	07/01/13	Varies	10.9	0.400	1.20	13.4	0.500	4.48
WP-06-SL-021	07/01/13	Varies	8.50	0.400	0.935	4.40	0.200	1.47
WP-06-SL-022	07/01/13	Varies	15.6	0.400	1.72	5.80	0.300	1.94
WP-06-SL-023	07/01/13	Oriskany	6.30	0.300	0.693	1.50	0.100	0.501
WP-06-SL-024	07/08/13	Varies	11.5	0.400	1.27	4.80	0.300	1.60
WP-06-SL-025	07/08/13	Varies	16.0	0.400	1.76	5.40	0.300	1.80
WP-06-SL-026	07/08/13	Varies	17.7	0.500	1.95	8.80	0.500	2.94
WP-07-SL-039	08/05/13	Varies	17.3	0.400	1.90	2.50	0.200	0.835
WP-07-SL-040	08/05/13	Varies	17.8	0.400	1.96	1.50	0.100	0.501
WP-07-SL-041	08/05/13	Varies	17.7	0.400	1.95	2.30	0.200	0.768
WP-10-SL-045	08/26/13	Varies	11.8	0.400	1.30	3.00	0.200	1.00
WP-11-SL-047	08/27/13	Varies	7.00	0.400	0.770	2.40	0.100	0.802
WP-12-SL-052	09/05/13	Varies	17.7	0.500	1.95	12.4	0.500	4.14
WP-12-SL-053	09/05/13	Varies	17.9	0.400	1.97	6.30	0.300	2.10
WP-13-SL-059	10/15/13	Varies	16.2	0.400	1.78	2.00	0.200	0.668
WP-14-SL-073	01/31/14	Varies	17.1	0.400	1.88	3.10	0.200	1.04
WP-14-SL-074	01/31/14	Varies	17.3	0.400	1.90	3.20	0.200	1.07
		Average	15.0		1.64	4.39		1.47

Table 3-6. XRF Uranium and Thorium for Vertical Cuttings

Study ID	Date	Formation	Thorium Result (ppm)	Thorium Error (ppm)	Th-232 (pCi/g)	Uranium Result (ppm)	Uranium Error (ppm)	U-238 (pCi/g)
		Std. Dev.	3.66		0.403	2.64		0.881
		Median	16.3		1.79	3.60		1.20
		Minimum	6.30		0.693	1.50		0.501
		Maximum	20.1		2.21	13.4		4.48

Table 3-7. Horizontal Solids, Drill Cuttings – Uranium Series Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)	U-238 (pCi/g)	U-235 (pCi/g)	Th-232 (pCi/g)
WP-02-SL-036	13.0	0.621	18.3	4.96	0.789	0.608
WP-03-SL-065	9.76	0.797	26.2	4.19	0.265	0.786
WP-04-SL-009	3.69	0.581	12.6	0.803	0.130	0.568
WP-04-SL-010	3.96	0.535	12.6	0.917	0.240	0.524
WP-04-SL-011	2.37	0.668	16.8	0.575	0.144	0.654
WP-04-SL-012	5.43	0.727	15.3	< 2.53	0.220	0.712
WP-05-SL-027	3.31	0.772	18.3	1.88	0.201	0.755
WP-05-SL-032	1.50	0.711	14.2	< 2.09	< 0.158	0.696
WP-05-SL-034	3.17	0.861	20.1	< 1.32	< 0.152	0.841
WP-06-SL-037	1.17	0.346	6.33	0.830	< 0.085	0.339
WP-10-SL-048	4.92	0.694	31.5	< 2.30	< 0.250	0.680
WP-11-SL-068	1.06	0.241	7.41	< 0.835	< 0.091	0.237
WP-12-SL-055	< 0.183	< 0.031	1.47	< 0.485	< 0.058	< 0.031
WP-12-SL-056	3.56	0.535	11.7	1.57	0.153	0.527
WP-13-SL-062	10.3	0.487	8.70	3.11	0.391	0.478
WP-14-SL-077	8.09	0.702	17.5	2.78	0.384	0.689
WP-14-SL-078	9.60	0.828	20.4	3.09	0.302	0.813
WP-14-SL-079	8.97	1.16	16.7	2.24	0.277	1.14
Average	5.22	0.627	15.3	1.76	0.223	0.615
Std. Dev.	3.80	0.254	7.13	1.36	0.180	0.249
Median	3.83	0.681	16.0	1.21	0.211	0.667
Minimum	0.092	0.016	1.47	0.243	0.029	0.016
Maximum	13.0	1.16	31.5	4.96	0.789	1.14

Table 3-8. XRF Uranium and Thorium for Horizontal Cuttings

Study ID	Date	Target Formation / Gas Type	Thorium Result (ppm)	Thorium Error (ppm)	Th-232 (pCi/g)	Uranium Result (ppm)	Uranium Error (ppm)	U-238 (pCi/g)
WP-02-SL-036	07/24/13	Marcellus / Wet	12.2	0.400	1.34	28.6	0.500	9.55
WP-03-SL-065	11/08/13	Marcellus / Wet	11.8	0.400	1.30	20.1	0.600	6.71
WP-04-SL-009	06/20/13	Marcellus / Dry	12.0	0.500	1.32	8.70	0.400	2.91
WP-04-SL-010	06/20/13	Marcellus / Dry	10.8	0.500	1.19	9.90	0.400	3.31
WP-04-SL-011	06/20/13	Marcellus / Dry	12.5	0.400	1.38	5.90	0.300	1.97
WP-04-SL-012	06/20/13	Marcellus / Dry	12.4	0.400	1.36	14.6	0.500	4.88
WP-05-SL-027	07/08/13	Burkett / Wet	16.2	0.400	1.78	9.70	0.400	3.24
WP-05-SL-032	07/08/13	Burkett / Wet	11.1	0.400	1.22	5.20	0.300	1.74
WP-05-SL-034	07/08/13	Burkett / Wet	16.4	0.500	1.80	6.60	0.400	2.20
WP-06-SL-037	07/25/13	Utica / Wet	17.4	1.30	1.91	80.8	1.30	27.0
WP-10-SL-048	08/30/13	Marcellus / Dry	13.8	0.800	1.52	49.4	1.00	16.5
WP-11-SL-068	11/14/13	Utica / Dry	7.70	0.500	0.847	17.6	0.500	5.88
WP-12-SL-055	09/11/13	Marcellus / Dry	13.0	0.800	1.43	11.3	0.500	3.77
WP-12-SL-056	09/11/13	Marcellus / Dry	20.3	1.20	2.23	36.6	1.20	12.2
WP-13-SL-062	10/21/13	Marcellus / Dry	9.40	0.500	1.03	33.1	0.600	11.1
WP-14-SL-077	02/07/14	Marcellus / Dry	11.0	0.500	1.21	31.4	0.700	10.5
WP-14-SL-078	02/07/14	Marcellus / Dry	13.3	0.500	1.46	33.8	0.700	11.3
WP-14-SL-079	02/07/14	Marcellus / Dry	11.7	0.700	1.29	49.4	0.900	16.5
		Average	12.9		1.42	25.2		8.40
		Std. Dev.	3.01		0.331	20.0		6.70
		Median	12.3		1.35	18.9		6.30
		Minimum	7.70		0.847	5.20		1.74
		Maximum	20.3		2.23	80.8		27.0

Table 3-9. Drilling Solids, Mud – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)	U-238 (pCi/g)	U-235 (pCi/g)	Th-232 (pCi/g)
WP-03-SL-066	1.51	0.178	4.93	< 0.436	< 0.085	0.176
WP-04-SL-013	1.58	0.221	4.31	< 0.866	< 0.073	0.216
WP-05-SL-035	0.675	0.182	3.54	< 0.375	< 0.054	0.179
WP-10-SL-046	3.66	0.266	6.91	< 1.61	< 0.034	0.261
WP-10-SL-049	3.35	0.335	7.32	1.73	< 0.035	< 0.870
WP-11-SL-069	1.04	0.195	3.84	< 0.673	< 0.058	0.191
WP-12-SL-054	1.28	0.122	1.47	1.10	< 0.081	0.120
WP-13-SL-060	2.78	0.296	5.96	< 0.692	0.086	0.290
WP-13-SL-063	3.72	0.328	6.53	0.700	0.143	0.322
Average	2.18	0.236	4.98	0.651	0.063	0.243
Std. Dev.	1.20	0.074	1.89	0.504	0.038	0.095
Median	1.58	0.221	4.93	0.433	0.043	0.216
Minimum	0.675	0.122	1.47	0.188	0.017	0.120
Maximum	3.72	0.335	7.32	1.73	0.143	0.435

Table 3-10. Proppant Sand – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)	U-238 (pCi/g)	U-235 (pCi/g)	Th-232 (pCi/g)
WP-04-SL-050	0.180	0.053	0.733	0.139	< 0.025	0.047
WP-05-SL-058	0.225	0.135	7.25	< 0.200	< 0.037	0.115
WP-06-SL-070	0.170	0.026	0.069	0.323	< 0.018	0.025
WP-08-SL-044	0.246	0.065	0.162	< 0.020	< 0.004	0.045
WP-09-SL-043	0.301	0.045	0.199	< 0.426	< 0.050	0.044
WP-10-SL-067	0.218	0.018	0.136	< 0.369	< 0.036	0.018
WP-11-SL-072	0.275	0.025	0.070	< 0.203	< 0.033	0.025
WP-12-SL-064	0.358	0.038	0.386	< 0.426	< 0.042	0.037
WP-14-SL-081	0.266	< 0.026	4.99	< 0.442	< 0.035	0.102
WP-25-SL-042	0.188	0.018	< 0.061	< 0.267	< 0.029	< 0.013
Average	0.243	0.044	1.40	0.157	0.015	0.046
Std. Dev.	0.059	0.036	2.55	0.091	0.006	0.035
Median	0.236	0.032	0.181	0.159	0.017	0.041
Minimum	0.170	0.013	0.031	0.010	0.002	0.007
Maximum	0.358	0.135	7.25	0.323	0.025	0.115

Table 3-11. Flowback Solids, Sand – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)	U-238 (pCi/g)	U-235 (pCi/g)	Th-232 (pCi/g)
WP-04-SL-061	7.73	0.619	0.659	< 1.86	< 0.199	0.609
WP-09-SL-057	0.763	0.194	0.457	< 0.711	< 0.083	0.191
WP-11-SL-080	2.76	0.611	1.68	< 0.783	< 0.091	0.603
WP-12-SL-071	2.58	0.353	0.597	< 0.985	< 0.080	0.343
Average	3.46	0.444	0.848	0.542	0.057	0.437
Std. Dev.	2.99	0.208	0.561	0.265	0.029	0.205
Median	2.67	0.482	0.628	0.442	0.044	0.473
Minimum	0.763	0.194	0.457	0.356	0.040	0.191
Maximum	7.73	0.619	1.68	0.930	0.100	0.609

Table 3-12. Drilling Fluids – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha[a] (pCi/L)	Gross Beta[a] (pCi/L)
WP-02-LQ-002	4,690	372	9,910	ND	ND
WP-06-LQ-001	1,510	162	4,340	1,580	3,940
WP-06-LQ-003	2,010	216	5,220	ND	ND
WP-12-LQ-009	1,800	184	420	3,820	1,250
WP-14-LQ-026	4,940	466	11,400	ND	ND
Average	2,990	280	6,260	2,700	2,600
Std. Dev.	1,678	133	4,430	1,580	1,900
Median	2,010	216	5,220	2,700	2,600
Minimum	1,510	162	420	1,580	1,250
Maximum	4,940	466	11,400	3,820	3,940

[a]ND – Sample Matrix was not suitable for analysis.

Table 3-13. Fracturing Fluids – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WP-04-LQ-008	21,000	1,640	< 565	37,000	11,200
WT-05-LQ-013	872	78.0	195	1,870	398
WP-06-LQ-016	64.0	< 9.00	< 21.0	< 1.39	4.41
WP-08-LQ-007	3,080	723	444	5,020	1,610
WP-09-LQ-006	2,000	442	338	3,400	< 879
WP-10-LQ-015	10,300	600	< 298	13,500	2,310
WP-11-LQ-023	115	14.0	44.0	< 3.76	< 1.63
WP-14-LQ-046	2,270	189	456	5,760	1,200
WP-14-LQ-047	2,160	218	423	5,650	1,010
WP-19-LQ-004	16,200	1,250	435	54,100	14,900
WP-19-LQ-005	105	< 9.00	25.0	< 113	< 186
Average	5,290	469	255	11,500	3,020
Std. Dev.	7,250	547	178	17,700	5,080
Median	2,160	218	283	5,020	1,010
Minimum	64.0	4.50	10.5	0.695	0.815
Maximum	21,000	1,640	456	54,100	14,900

Table 3-14. Flowback Fluids – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WP-01-LQ-010	7,310	589	151	15,300	4,070
WP-04-LQ-014	25,500	1,740	500	71,000	21,300
WP-06-LQ-017	551	248	416	< 576	742
WP-08-LQ-012	4,280	1,140	500	7,270	1,820
WP-09-LQ-011	2,880	863	448	10,700	4,380
WP-10-LQ-045	8,690	633	2,630	11,100	1,960
WP-11-LQ-035	1,540	564	927	2,250	1,320
WP-12-LQ-022	4,550	507	< 177	10,100	2,440
WP-14-LQ-052	21,100	1,430	461	32,000	5,400
Average	8,490	857	680	17,800	4,830
Std. Dev.	8,840	486	769	21,900	6,370
Median	4,550	633	461	10,700	2,440
Minimum	551	248	88.5	288	742
Maximum	25,500	1,740	2,630	71,000	21,300

Table 3-15. Unfiltered Produced Waters – Gamma Spectroscopy and Miscellaneous Results

Study ID	Well Type	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WP-01-LQ-048	Unconventional	2,050	366	132	3,890	< 225
WP-04-LQ-039	Unconventional	26,600	1,900	328	30,000	7,600
WP-08-LQ-021	Unconventional	5,020	1,280	592	11,300	3,270
WP-09-LQ-019	Unconventional	4,490	1,140	571	9,760	2,570
WP-10-LQ-050	Unconventional	7,730	434	191	14,000	3,620
WP-10-LQ-055	Unconventional	6,710	470	149	41,700	4,560
WP-11-LQ-043	Unconventional	1,700	636	852	2,420	1,500
WP-12-LQ-041	Unconventional	14,500	1,710	408	21,800	6,810
WP-16-LQ-027	Conventional	819	896	220	< 2,570	1,140
WP-19-LQ-029	Conventional	< 81.0	26.0	103	< 465	< 402
WP-20-LQ-031	Conventional	145	42.0	129	< 2,440	< 987
WP-21-LQ-033	Conventional	340	214	< 31.0	< 1,230	< 830
WP-05-LQ-037	Unconventional	6,300	941	667	10,700	2,300
	Average	5,880	773	335	11,500	2,660
	Std. Dev.	7,450	604	260	12,800	2,460
	Median	4,490	636	220	9,760	2,300
	Minimum	40.5	26.0	15.5	233	113
	Maximum	26,600	1,900	852	41,700	7,600

Table 3-16. Filtered Produced Waters – Gamma Spectroscopy and Miscellaneous Results

Study ID	Well Type	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WP-01-LQ-049	Unconventional	1,930	373	129	2,750	933
WP-04-LQ-040	Unconventional	24,100	1,860	323	33,000	7,180
WP-08-LQ-020	Unconventional	4,940	1,350	518	11,200	4,050
WP-09-LQ-018	Unconventional	4,470	1,240	560	8,780	3,040
WP-10-LQ-051	Unconventional	8,060	466	164	19,900	4,050
WP-10-LQ-054	Unconventional	7,130	479	3,950	10,900	3,530
WP-11-LQ-044	Unconventional	1,520	602	751	2,440	1,500
WP-12-LQ-042	Unconventional	15,100	1,610	389	18,000	4,050
WP-16-LQ-028	Conventional	849	851	< 34.0	1,440	1,610
WP-19-LQ-030	Conventional	87.0	44.0	71.0	< 608	< 420
WP-20-LQ-032	Conventional	106	48.0	129	< 1,040	< 857
WP-21-LQ-034	Conventional	292	210	144	< 1,860	< 863
WP-05-LQ-038	Unconventional	6,720	883	485	11,400	3,370

Table 3-16. Filtered Produced Waters – Gamma Spectroscopy and Miscellaneous Results

Study ID	Well Type	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
	Average	5,790	770	587	9,350	2,650
	Std. Dev.	6,980	591	1,030	9,750	2,020
	Median	4,470	602	323	8,780	3,040
	Minimum	87.0	44.0	17.0	304	210
	Maximum	24,100	1,860	3,950	33,000	7,180

Table 3-17. Ambient Radon at Well Sites During Flowback

Study ID	County	Date	Radon Concentration (pCi/L)	Error (± Std. Dev.) (pCi/L)	MDC (pCi/L)
WP-01-RA	Sullivan	9/2013	< 0.300	0.000	0.300
			0.800	0.000	0.300
			0.500	0.400	0.300
			< 0.300	0.000	0.300
			< 0.300	0.000	0.300
WP-09-RA	Washington	9/2013	0.700	0.600	0.300
			0.600	0.200	0.300
			0.600	0.200	0.300
			1.70	1.60	0.300
WP-08-RA	Washington	9/2013	0.500	0.800	0.300
			0.200	0.200	0.300
			0.600	0.600	0.300
			0.700	0.400	0.300
WP-04-RA	Tioga	10/2013	0.500	0.200	0.300
			0.200	0.200	0.300
			0.500	0.600	0.300
			0.700	0.200	0.300

E-PERM samples with short-term electrets were deployed. MDC for a four-day exposure at 50 percent error is 0.300 pCi/L.

Table 3-18. Natural Gas Samples from Production Sites

Study ID	County	Gas Source	Radon Concentration (pCi/L)	Error (±2 Std. Dev.) (pCi/L)	MDA (pCi/L)
WP-08-RG	Washington	Marcellus Shale	79.6	0.800	0.300
WP-09-RG	Washington	Marcellus Shale	78.8	4.20	0.300
WP-22-RG	Tioga	Marcellus Shale	42.8	0.200	0.100
WP-23-RG	Tioga	Marcellus Shale	39.6	0.800	0.200
WP-24-RG	Tioga	Marcellus Shale	73.8	0.400	0.200
WP-25-RG	Tioga	Marcellus Shale	44.4	2.60	0.200
WP-26-RG	Lycoming	Oriskany Sandstone	19.9	0.200	0.200
WP-27-RG	Tioga	Marcellus Shale	38.4	3.40	0.300
WP-28-RG	Tioga	Marcellus Shale	40.8	5.20	0.400
WP-16-RG	Washington	Marcellus Shale	50.0	5.20	0.300
WP-17-RG	Washington	Marcellus Shale	49.5	5.80	0.500
WP-19-RG	McKean	Upper Devonian Shale	18.3	4.40	0.400
WP-20-RG	McKean	Upper Devonian Shale	88.2	10.6	0.700
WP-21-RG	Forest	Upper Devonian Shale	92.2	6.40	0.400
WP-04-RG	Tioga	Marcellus Shale	49.6	29.6	1.20
WP-05-RG	McKean	Marcellus Shale	148	15.6	1.50
WP-12-RG	Lycoming	Marcellus Shale	37.6	33.4	2.20
WP-11-RG	Tioga	Utica	5.70	1.20	0.500
WP-29-RG	Sullivan	Marcellus Shale	23.4	4.00	0.240
WP-30-RG	Bradford	Marcellus Shale	25.5	2.70	0.200
WP-31-RG	Bradford	Marcellus Shale	3.00	1.20	0.300
WP-14-RG	Jefferson	Marcellus Shale	5.60	0.100	0.140
		Average	47.9		
		Median	41.8		
		Standard Deviation	34.5		
		Minimum	3.00		
		Maximum	148		

Note: All results adjusted to account for the fact that Rn was counted in methane, but the scintillation cells were calibrated for Rn in air. Range of α particles is greater in methane than in air. All results divided by 1.054, according to Jenkins et. al., Health Physics, Vol. 106, No. 3, March 2014.

Table 3-19. Thorium and Uranium XRF Data for Drill Cuttings By Formation

Formation	Thorium Result (ppm)	Th-232 Concentration (pCi/g)	Uranium Result (ppm)	U-238 Concentration (pCi/g)	U/Th	U-238/ Th-232
Marcellus	13.8	1.52	49.4	16.5	3.58	10.9
Marcellus	13.0	1.43	11.3	3.77	0.870	2.64
Marcellus	20.3	2.23	36.6	12.2	1.80	5.48
Marcellus	9.40	1.03	33.1	11.1	3.52	10.7
Marcellus	11.8	1.30	20.1	6.71	1.70	5.16
Marcellus	12.0	1.32	8.70	2.91	0.730	2.20
Marcellus	10.8	1.19	9.90	3.31	0.920	2.78
Marcellus	12.5	1.38	5.90	1.97	0.470	1.43
Marcellus	12.4	1.36	14.6	4.88	1.18	3.59
Marcellus	11.7	1.29	49.4	16.5	4.22	12.8
Marcellus	13.3	1.46	33.8	11.3	2.54	7.73
Marcellus	11.0	1.21	31.4	10.5	2.85	8.67
Marcellus	12.2	1.34	28.6	9.55	2.34	7.13
Average	12.6	1.40	25.6	8.60	2.10	6.20
Median	12.2	1.30	28.6	9.60	1.80	5.50
Standard Deviation	2.57	0.280	15.0	5.01	1.23	3.72
Minimum	9.40	1.03	5.90	1.97	0.470	1.43
Maximum	20.3	2.23	49.4	16.5	4.22	12.8
Burket	16.2	1.78	9.70	3.24	0.600	1.82
Burket	16.4	1.80	6.60	2.20	0.400	1.22
Burket	11.1	1.22	5.20	1.74	0.470	1.42
Average	14.6	1.60	7.17	2.39	0.490	1.49
Median	16.2	1.78	6.60	2.20	0.470	1.42
Standard Deviation	3.00	0.330	2.30	0.770	0.100	0.300
Minimum	11.1	1.22	5.20	1.74	0.400	1.22
Maximum	16.4	1.80	9.70	3.24	0.600	1.82
Utica	7.70	0.850	17.6	5.88	2.29	6.92
Utica	17.4	1.91	80.8	27.0	4.64	14.1
Average	12.6	1.38	49.2	16.4	3.46	10.5
Median	12.6	1.38	49.2	16.4	3.46	10.5
Standard Deviation	6.86	0.750	44.7	14.9	1.67	5.10
Minimum	7.70	0.850	17.6	5.88	2.29	6.92
Maximum	17.4	1.91	80.8	27.0	4.64	14.1

4.0 WASTEWATER TREATMENT PLANTS

A total of 29 WWTPs were surveyed and/or sampled. This included 10 POTWs, 10 CWTs and nine ZLDs. The results, by wastewater facility, are presented in this section.

4.1 Publicly Owned Treatment Works

A total of 10 POTWs were surveyed and/or sampled. There were three rounds of surveys conducted over a seven-month period (April 2013 through October 2013); however, not all POTWs were sampled in all three rounds. Six of the 10 POTWs are considered ***influenced*** **(POTW-I)** by having received wastewater from the O&G industry, mainly the effluent of CWTs. Four POTWs are considered ***non-influenced*** (**POTW-N**) by having never received wastewater from the O&G industry. As such, surveying was conducted for the 10 POTWs as follows:

- 5 **POTW-I's** were surveyed in all three rounds,
- 1 **POTW-I** was surveyed in two rounds, and
- 4 **POTW-N's** were surveyed one time.

4.1.1 Radiological Survey Results

Radiological surveys were conducted at each **POTW-I**, resulting in four data sets:

- *Removable* α/β *surface radioactivity* measurements recorded in units of dpm/100 cm^2
- *Total* α/β *surface radioactivity* measurements recorded in units of dpm/100 cm^2
- *Gross Gamma Radiation Scan* measurements recorded in units of cpm
- *Gamma Radiation Exposure Rate* measurements recorded in units of μR/hr

4.1.1.1 Removable Alpha/Beta Surface Radioactivity Measurement Results

Measurements of removable radioactivity were performed to assess potential internal radiation exposures of workers through ingestion and/or inhalation. The results were evaluated using the RG 1.86 guidelines, Table 1. RG 1.86 requires that α and β surface radioactivity levels be evaluated separately. The primary α emitter of concern is Ra-226, with a removable criterion of 20 dpm α/100 cm^2. The primary β emitter of concern is Ra-228 of the natural Th decay series with a removable criterion of 200 dpm β/100 cm^2. The average removable α and β surface radioactivity levels at each WWTP were below the RG 1.86 criteria. The maximum removable α and β surface radioactivity levels were 22 dpm/100 cm^2 and 161 dpm/100 cm^2. The results of removable α and β surface radioactivity for the **POTW-I** plants are presented in **Table 4-1.** Individual removable α and β surface radioactivity measurement results are presented in **Appendix D**.

4.1.1.2 Total Alpha/Beta Surface Radioactivity Measurement Results

Measurements of total radioactivity were performed to assess potential internal radiation exposures of workers through ingestion and/or inhalation. The results were evaluated using the RG 1.86 guidelines, Table 1. RG 1.86 requires that α and β surface radioactivity levels be evaluated separately. The primary α emitter of concern is Ra-226, with a total surface radioactivity criterion

of 100 dpm α/100 cm^2. The primary β emitter of concern is Ra-228 of the natural Th decay series with a total surface radioactivity criterion of 1,000 dpm β/100 cm^2. The maximum average total α and β surface radioactivity measured at any single facility were 313 dpm/100 cm^2 and 10,000 dpm/100 cm^2, respectively. The maximum total α and β concentrations measured at any single facility were 1,190 dpm/100 cm^2 and 38,000 dpm/ 100 cm^2. The summary results of total α and β surface radioactivity for the **POTW-I** plants surveyed are presented in **Table 4-2**. Individual total α and β surface radioactivity measurement results are presented in **Appendix D**.

4.1.1.3 Gross Gamma Radiation Scan Results

Gross gamma radiation scans recorded in cpm were performed on open land areas and accessible areas of the WWTPs to identify areas with elevated gross gamma radiation levels. Summary results for the **POTW-I** are presented in **Table 4-3**. The highest average count rate for the plants was 29,034 cpm, and the maximum count rate recorded was 205,446 cpm. A graphic display of the gamma radiation scan results (figures) at each facility was prepared using geographic information system (GIS) software. Figures are presented in **Appendix E**.

4.1.1.4 Gamma Radiation Exposure Rate Results Summary

Gross gamma radiation scan results in units of cpm presented in **Table 4-3** were converted to μR/hr using 800 cpm per μR/hr, a conversion factor appropriate for Ra-226 gamma energy as detected with 2-inch by 2-inch NaI detectors, rounded to one significant figure (Table 6.4, NaI Scintillation Detector Scan MDCs for Common Radiological Contaminants, NUREG-1507, Minimum Detectable Concentrations With Typical Radiation Survey Instruments for Various Contaminants and Field Conditions, USNRC June 1998). **Table 4-4** presents statistical results for each **POTW-I**. The highest average gamma radiation exposure rate was 36.3 μR/hr, and the maximum gamma radiation exposure rate measured was 257 μR/hr.

4.1.2 Solid Sample Results

4.1.2.1 Filter Cake Samples

Filter cakes were sampled at **POTW-I** and **POTW-N** plants and analyzed using gamma spectroscopy for U, Th, and Ac series decay chains. The gamma spectroscopy results are presented in **Tables 4-5** and **4-6**.

The analytical results for **POTW-I** plants presented in **Table 4-5** show Ra-226 and Ra-228 are present above typical background concentrations in soil. The average Ra-226 result was 20.1 pCi/g with a large variance in the distribution, and the maximum result was 55.6 pCi/g. The average Ra-228 result was 7.63 pCi/g, and the maximum result was 32.0 pCi/g Ra-228.

The radioactivity levels at **POTW-N** plants presented in **Table 4-6** were also above typical background concentrations in soil with Ra-226 average and maximum results of 9.72 pCi/g and 35.4 pCi/g. The average and maximum Ra-228 results were 2.26 pCi/g and 7.26 pCi/g.

4.1.2.2 Sediment-Impacted Soil Samples

Sampling was performed at only three of the **POTW-I** plants due to limited accessibility at the other plants. A total of seven samples were collected at the effluent discharge points and analyzed for U, Th, and Ac series decay chains by gamma spectroscopy. The gamma spectroscopy results are presented in **Table 4-7**.

The analytical results for **POTW-I** sediment-impacted soil samples indicate Ra-226 and Ra-228 are present at concentrations above typical background in soil. The average Ra-226 result was 9.00 pCi/g, and the maximum result was 18.2 pCi/g. The average Ra-228 result was 3.52 pCi/g, and the maximum result was 6.25 pCi/g.

4.1.3 Liquid Sample Results

Influent and effluent liquid sampling was performed at six **POTW-I** plants and four **POTW-N** plants. Filtered and unfiltered samples were analyzed for U, Th, and Ac decay series, and for gross α/β radioactivity levels. The filtered and unfiltered analyses are presented separately in **Tables 4-8** through **4-15** for both influenced and non-influenced POTWs. A comparison of the influenced and non-influenced POTW results and the filtered and unfiltered sample results is presented in Section 4.1.5.1.

4.1.4 Indoor Radon Sampling Results

ATDs were deployed in the **POTW-I** plants at various indoor locations such as break rooms, labs, offices, etc., to measure Rn concentrations. The results were evaluated using the EPA action level of 4 pCi/L. The ATDs were deployed in late July or early August 2013 and were all recovered from the field in February 2014. The results ranged from 0.200 to 8.70 pCi/L. One result exceeded the action level. The results are presented in **Table 4-16**. The Rn analytical reports are presented in **Appendix H**.

4.1.5 POTW Data Comparisons

4.1.5.1 POTW-I / POTW-N Comparison

Thirty-two influent and effluent sample radionuclide and gross α/β concentration results from **POTW-I's** and **POTW-N's** were compared to determine if there was a difference in the radionuclide activity content. **Tables 4-17** through **4-20** present and compare the average Ra concentration results and gross α/β concentration results from all influent and effluent filtered and unfiltered samples for all **POTW-I** and **POTW-N** plants. Twenty-nine of the 32 average concentration results for both filtered and unfiltered influent and effluent samples were higher for **POTW-I** plants than the **POTW-N** plants.

4.1.5.2 Radium-226/Radium-228 Sediment-Impacted Soil and Effluent Results Comparison

The sediment-impacted soil radioactivity levels were compared to filtered and unfiltered effluent results for Ra-226 and Ra-228 and are presented in **Table 4-21**. In cases where no results were reported for a member of the data pair (sediment-effluent pair), or when a result was reported as less than MDC, the data pair comparison was not evaluated.

The sediment-impacted soil sample results are above typical background for soil. However, there is no readily apparent relationship between the sediment-impacted soil sample and effluent sample results. The effluent wastewater discharged over time may contribute to the activity in the sediment-impacted soil, but a correlation between the sediment-impacted soil activity and the effluent samples could not be made from the study as performed.

The ratio of Ra-226 to Ra-228 was also calculated for a variety of sample types including sediments, filtered effluents, and unfiltered effluents from POTWs and CWTs. The results are presented in **Table 4-22**. The average ratio ranged from 2.4 to 11.4.

4.1.6 POTW Worker Exposure Assessment

4.1.6.1 External Gamma Radiation Exposure

The gamma radiation exposure rate survey results are provided in Section 4.1.1.4. The maximum average gamma radiation exposure rate measured at any of the POTW plants was 36.3 μR/hr. The lowest background gamma radiation exposure rate measured at any of the sites was 5 μR/hr. Assuming the time period of exposure is a full occupational year of 2,000 hours, the maximum average POTW annual external gamma radiation exposure was estimated as follows:

Maximum Average POTW External Gamma Radiation Exposure Estimate

(36.3 – 5) μR/hr x 2,000 hr/yr x (1 mrem/1,000 μR gamma) = 62.6 mrem/yr

This is an estimate of the maximum average gamma radiation exposure at a single facility based on 2,000 hours in one year. The result is less than the 100 mrem/yr dose equivalent limit for a member of the public. Actual exposure is dependent upon the actual exposure rates and occupancy time for individual workers.

The maximum gamma radiation exposure rate measured at the POTWs was 257 μR/hr on contact with the outside of a wastewater tank. Consequently, the public dose limit of 100 mrem per year could potentially be reached by a person working 400 hours within the immediate proximity of the tank. Actual annual exposure for a POTW worker is dependent upon the exposure rates and time worked in proximity to the tank.

4.1.6.2 Internal Alpha/Beta Radiation Exposure

The total and removable α/β survey surface radioactivity summary results are provided in Sections 4.1.1.1 and 4.1.1.2. Nine of the 566 α measurements and 68 of the 566 β measurements of total surface radioactivity exceeded the RG 1.86 criteria. One of the 286 removable α measurements and none of the 286 removable β measurements exceeded the RG 1.86 criteria. Fixed or removable α and β surface radioactivity may present a potential inhalation or ingestion hazard if disturbed during routine system maintenance.

4.1.6.3 Internal Radon Exposure

The Rn measured in indoor air averaged 1.74 pCi/L. This average is below the EPA action level of 4 pCi/L, and very near the U.S. average indoor Rn level of 1.3 pCi/L, as reported by EPA.

4.1.7 POTW Radiological Environmental Impacts

Seven sediment-impacted soil samples were collected at the effluent discharge points of three of the **POTW-I's**. Radium-226 activity concentrations above typical soil background activity concentrations were identified in all sediment samples, with 18.2 pCi/g being the maximum reported result.

The presence of Ra in sediment-impacted soil at effluent discharge points indicates effluent wastewater contained Ra. Radium and gross α and β radioactivity were identified in effluent samples. **Table 4-21** presents filtered and unfiltered effluent average sample results and sediment-impacted soil results for POTWs sampled during the study.

4.2 Centralized Wastewater Treatment Plants

Three survey rounds were conducted at nine of the 10 CWTs. The 10th facility was added after the first survey round was completed, resulting in only two surveys at that facility.

4.2.1 Survey Results

Radiological surveys were conducted at each CWT resulting in four data sets:

- *Removable α/β surface radioactivity* measurements recorded in units of dpm/100 cm^2
- *Total α/β surface radioactivity* measurements recorded in units of dpm/100 cm^2
- *Gross Gamma Radiation Scan* measurements recorded in units of cpm
- *Gamma Radiation Exposure Rate* measurements recorded in units of μR/hr

4.2.1.1 Removable Alpha/Beta Surface Radioactivity Measurement Results

Measurements of removable radioactivity were performed to evaluate potential internal radiation exposures of workers through ingestion and/or inhalation. The results were evaluated using the RG 1.86 surface radioactivity guidelines, Table 1. RG 1.86 requires that α and β surface radioactivity levels be evaluated separately. The primary α emitter of concern is Ra-226, with a removable surface radioactivity criterion of 20 dpm α/100 cm^2. The primary β emitter of concern is Ra-228 of the natural Th decay series with a removable surface radioactivity criterion of 200 dpm β/100 cm^2.

The average removable α and β surface radioactivity levels were all below the RG 1.86 criteria. The maximum removable α and β surface radioactivity levels were 38.1 dpm/100 cm^2 and 133 dpm/100 cm^2. The summary results of removable α and β surface radioactivity are presented in **Table 4-23.** Individual removable α and β surface radioactivity measurement results are presented in **Appendix D**.

4.2.1.2 Total Alpha/Beta Surface Radioactivity Measurement Results

Measurements of total α and β surface radioactivity were performed to evaluate potential internal radiation exposures of workers through ingestion and/or inhalation. The results were evaluated using the RG 1.86 surface radioactivity guidelines, Table 1. RG 1.86 requires that α and β surface radioactivity levels be evaluated separately. The primary α emitter of concern is Ra-226, with a total surface radioactivity criterion of 100 dpm α/100 cm^2. The primary β emitter of concern is Ra-228 of the natural Th decay series with a total surface radioactivity criterion of 1,000 dpm β/100 cm^2.

Eighteen of the 28 average total α surface radioactivity measurements were below the RG 1.86 surface radioactivity criterion. Three of the 28 average total β surface radioactivity measurements were below the RG 1.86 surface radioactivity criterion. The maximum total α and β surface radioactivity levels were 3,220 dpm/100 cm^2 and 50,400 dpm/100 cm^2. The summary results of total α and β surface radioactivity measurements are presented in **Table 4-24**. Individual total α and β surface radioactivity measurement results are presented in **Appendix D**.

4.2.1.3 Gross Gamma Radiation Scan Results

Gross gamma radiation scans recorded in cpm were performed on open land areas and accessible areas of the CWT facilities to identify any areas with levels above local background. The summary results of the gross gamma radiation scans for each plant are presented in **Table 4-25**. The highest average count rate for the plants was 19,281 cpm, and the maximum count rate recorded was 401,688 cpm. A graphic display of the gamma radiation scan results at each facility was prepared using GIS software. The resulting figures are in **Appendix E**.

4.2.1.4 Gamma Radiation Exposure Rate Results Summary

Gross gamma radiation scan results in units of cpm presented in **Table 4-25** were converted to μR/hr by dividing by 800 cpm per μR/hr, a conversion factor appropriate for Ra-226 gamma energy as detected with 2-inch by 2-inch NaI detectors rounded to one significant figure (Table 6.4, NaI Scintillation Detector Scan MDCs for Common Radiological Contaminants, NUREG-1507, Minimum Detectable Concentrations With Typical Radiation Survey Instruments for Various Contaminants and Field Conditions, USNRC June 1998). **Table 4-26** presents statistical results for each CWT facility. The highest average gamma radiation exposure rate was 24.1 μR/hr, and the maximum gamma radiation exposure rate measured was 502 μR/hr.

4.2.2 Solid Sample Results

4.2.2.1 Filter Cake Samples

Three survey rounds were conducted at nine of the 10 CWTs. The 10th facility was added after the first survey round was completed, resulting in only two surveys at that facility. Also, the 10th facility is a primary treatment facility, so it does not produce a filter cake. A total of 25 filter cake samples were collected from the nine plants. The results are presented in **Table 4-27**. The analytical results indicate all the CWT filter cake samples contain elevated Ra-226 and Ra-228

above typical background levels for soil. The maximum results were 294 pCi/g of Ra-226 and 177 pCi/g of Ra-228.

4.2.2.2 Solids/Sediment Samples

Four of the CWTs surveyed and sampled as part of the study are permitted to discharge effluent wastewater to the environment. If the discharge point was accessible, surface soil impacted by sediment was sampled. The gamma spectroscopy results are presented in **Table 4-28**. The Ra-226 results ranged from 2.50 to 421 pCi/g. The Ra-228 results ranged from 0.978 to 86.9 pCi/g. Uranium and Th were also detected at surface soil typical background levels in some of the samples because of natural soil collected along with the sediment.

4.2.2.3 Solids/Biased Samples

Gamma radiation walkover scans identified areas with radioactivity above local background. At three of these locations, a biased soil sample was collected to determine the amount of activity at or near the surface. The gamma spectroscopy results are presented in **Table 4-29**. Radium above soil typical background levels to a maximum of 444 pCi/g Ra-226 and 83.1 pCi/g Ra-228 was identified in biased soil samples.

4.2.3 Liquid Samples

Samples of influent and effluent, both filtered and unfiltered, were analyzed. Three survey rounds were conducted at nine of the 10 CWTs. The 10th facility was added after the first survey round was completed, resulting in only two surveys at that facility. Also, the 10th facility is only a primary treatment facility, with the influent and the effluent essentially the same. Consequently, only the influent was sampled at the 10^{th} facility. A total of 31 effluent and 26 influent samples were collected for filtered and unfiltered analysis. The filtered and unfiltered analyses are presented separately. The gamma spectroscopy results, gross α, and gross β are presented in **Tables 4-30** through **4-33**. Radium (Ra-226 and Ra-228) was routinely detected in all sample types with little difference between influent and effluent or between filtered and unfiltered results as presented for Ra-226 in **Figure 4-1**.

Figure 4-1. CWT Influent and Effluent Liquid Ra-226 Minimum, Maximum, and Average

Wastewater Source	Filtered or Not	Min (pCi/L)	Max (pCi/L)	Ave (pCi/L)
Effluent	Filtered	18.0	14,900	2,100
Effluent	Unfiltered	42.0	15,500	1,840
Influent	Filtered	57.0	14,100	1,550
Influent	Unfiltered	17.5	13,400	1,870

4.2.4 Indoor Radon Sampling Results

ATDs were deployed in the CWT plants at various indoor locations such as break rooms, labs, offices, etc., and the results were evaluated using the EPA action level of 4.0 pCi/L. The results ranged from 0.900 to 5.00 pCi/L. Two results exceeded the action level. The results of the analyses are presented in **Table 4-34**. The Rn analytical reports are presented in **Appendix H**.

4.2.5 Filtered Versus Unfiltered Sample Data Evaluation

Appendix I presents a complete evaluation of filtered versus unfiltered liquid samples for the entire study. The conclusion from this evaluation is that there is no apparent trend or bias that filtering produces. There were some subsets of data where either the unfiltered results or the filtered results appear to be significantly higher. There was no statistically significant correlation found within any sample group. Because the liquid samples were preserved by addition of acid prior to filtering, the radioactive particulates may have entered solution and were therefore not removed by filtering.

4.2.6 CWT Exposure Assessment

4.2.6.1 CWT External Radiation Exposure

The maximum average gamma radiation exposure rate measured at any of the CWT plants was 24.1 μR/hr. The lowest background gamma radiation exposure rate measured at any of the sites was 5 μR/hr. Assuming the time period of exposure is a full occupational year of 2,000 hours, the maximum average CWT annual external gamma radiation exposure was estimated as follows:

Maximum Average CWT External Gamma Radiation Exposure Estimate

(24.1 – 5) μR/hr x 2,000 hr/yr x (1 mrem/1,000 μR gamma) = 38 mrem/yr

This is an estimate of the maximum average gamma radiation exposure based on 2,000 hours in one year. The result is less than the 100 mrem/yr dose equivalent limit for a member of the public. Actual exposure is dependent upon the actual exposure rates and occupancy time for individual workers.

The maximum gamma radiation exposure rate measured was 502 μrem/hr on contact with the outside of a wastewater tank. Work in proximity of the tank could potentially result in an exposure of 100 mrem in 200 hours of annual exposure or 10 percent of an employee's 2,000-hour occupational year. Actual annual exposure for a CWT worker is dependent upon actual exposure rates and actual time worked in the proximity of the tank.

4.2.6.2 CWT Potential Internal Alpha/Beta Radioactivity Exposure

The total and removable α/β surface radioactivity survey results are discussed in Sections 4.2.1.1 and 4.2.1.2. One hundred eighty-six of the 777 α measurements and 461 of the 777 β measurements of total surface radioactivity exceeded the RG 1.86 criteria. Seven of the 805 removable α measurements and 6 of the 805 removable β measurements exceeded the RG 1.86 criteria. The average of the β total surface radioactivity measurements exceeded the RG 1.86 criteria in 10 of the 11 CWT facilities surveyed. The average of the α total surface radioactivity measurements exceeded the RG 1.86 criteria in four of the 11 CWT facilities surveyed. The corresponding removable radioactivity measurements are mostly less than the RG 1.86 criteria, indicating the total radioactive contamination measured is fixed to the surface and not immediately available for inhalation or ingestion. Fixed α and β surface radioactivity may present a potential inhalation or ingestion hazard if disturbed during routine system maintenance.

4.2.6.3 Internal Radon Exposure

The Rn in indoor area air averaged 2.00 pCi/L. This average is below the EPA action level of 4 pCi/L and only slightly above the U.S. average indoor level of 1.3 pCi/L, as reported by EPA.

4.2.7 CWT Radiological Environmental Impacts

Sediment-impacted soil was collected at the accessible effluent discharge points at the CWTs. A total of nine samples were collected. Radium above typical soil background levels to a maximum of 508 pCi/g of total Ra was identified in the sediment-impacted soil samples. Effluent wastewater also contained Ra and is the likely source of the Ra in sediment-impacted soil above soil typical background levels.

4.3 Zero Liquid Discharge Plants

4.3.1 Survey Results

Radiological surveys were conducted at each ZLD facility resulting in four data sets:

- *Removable* α/β *surface radioactivity* measurements recorded in units of dpm/100 cm^2
- *Total* α/β *surface radioactivity* measurements recorded in units of dpm/100 cm^2
- *Gross Gamma Radiation Scan* measurements recorded in units of cpm
- *Gamma Radiation Exposure Rate* measurements recorded in units of μR/hr

4.3.1.1 Removable Alpha/Beta Surface Radioactivity Measurement Results

Measurements of removable surface radioactivity were performed to evaluate potential internal radiation exposures of workers through ingestion and/or inhalation. The results were evaluated using the RG 1.86 guidelines, Table 1. RG 1.86 requires that α and β surface radioactivity levels be evaluated separately. The primary α emitter of concern is Ra-226, with a removable surface radioactivity criterion of 20 dpm α/100 cm^2. The primary β emitter of concern is Ra-228 of the natural Th decay series with a removable surface radioactivity criterion of 200 dpm β/100 cm^2. The average removable α and β surface radioactivity levels were below the RG 1.86 criteria. The maximum removable α and β surface radioactivity levels were 294 dpm/100 cm^2 and 342 dpm/100 cm^2. The summary results of removable α and β surface radioactivity are presented in **Table 4-35.** Individual removable α and β surface radioactivity measurement results are presented in **Appendix D**.

4.3.1.2 Total Alpha/Beta Surface Radioactivity Measurement Results

Measurements of total α and β surface radioactivity were performed to evaluate potential internal radiation exposures of workers through ingestion and/or inhalation. The results were evaluated using the RG 1.86 guidelines, Table 1. RG 1.86 requires that α and β surface radioactivity levels be evaluated separately. The primary α emitter of concern is Ra-226, with a total surface radioactivity criterion of 100 dpm α/100 cm^2. The primary β emitter of concern is Ra-228 of the natural Th decay series with a total surface radioactivity criterion of 1,000 dpm β/100 cm^2. The highest average total α and β surface radioactivity levels were 239 dpm/100 cm^2 and

4,740 dpm/100 cm^2. The maximum total α and β surface radioactivity levels were 1,410 dpm/100 cm^2 and 49,700 dpm/100 cm^2. The summary results of total α and β surface radioactivity measurements are presented in **Table 4-36**. Individual total α and β surface radioactivity measurement results are presented in **Appendix D**.

4.3.1.3 Gross Gamma Radiation Scan Results

Gross gamma radiation scans recorded in cpm were performed on open land areas and accessible areas of the plant to identify levels of elevated gross gamma radiation. The results of the gross gamma radiation scans are presented in **Table 4-37**. The highest average count rate for the plants was 34,513 cpm, and the maximum count rate recorded was 356,274 cpm. A graphic display of the gamma radiation scan results (figures) at each facility was prepared using GIS software. The resulting figures are in **Appendix E**.

4.3.1.4 Gamma Radiation Exposure Rate Results Summary

Gross gamma radiation scan results in units of cpm presented in **Table 4-37** were converted to μR/hr by dividing by 800 cpm per μR/hr, a conversion factor appropriate for Ra-226 gamma energy as detected with 2-inch by 2-inch NaI detectors rounded to one significant figure (Table 6.4, NaI Scintillation Detector Scan MDCs for Common Radiological Contaminants, NUREG-1507, Minimum Detectable Concentrations With Typical Radiation Survey Instruments for Various Contaminants and Field Conditions, USNRC June 1998). **Table 4-38** presents statistical results for each ZLD facility. The highest average gamma radiation exposure rate was 43.1 μR/hr, and the maximum gamma radiation exposure rate measured was 445 μR/hr.

4.3.2 Solid Sample Results

4.3.2.1 Filter Cake Samples

Three survey rounds were conducted at each of the nine ZLD plants and a total of 31 filter cake samples were collected from the nine plants. The gamma spectroscopy results are presented in **Table 4-39**. Radium-226 and Ra-228 were measured in ZLD filter cake samples at concentrations above typical background levels for surface soils. Radium-226 concentrations ranged from 3.08 to 480 pCi/g, and Ra-228 concentrations ranged from 0.580 to 67.3 pCi/g.

4.3.2.2 Solids/Biased Samples

A single biased surface soil sample was collected. The gamma spectroscopy results are presented in **Table 4-40**. The Ra-226 and Ra-228 were measured in concentrations above typical background levels. The Ra-226 concentration was 37.1 pCi/g, and the Ra-228 concentration was 7.47 pCi/g.

4.3.3 Liquid Samples

Three survey and sample events were conducted at each of the nine ZLD plants. A total of 30 effluent samples and 26 influent samples were collected. The filtered and unfiltered sample analyses results are presented separately. The results of the U series, Th Series, and Ac series with K-40, gross α, and gross β are presented in **Tables 4-41** through **4-44**. Radium (Ra-226 and

Ra-228) was routinely detected in all sample types with an approximate 50 percent difference between influent and effluent, but little difference between filtered and unfiltered results, as presented for Ra-226 results below in **Figure 4-2**.

Figure 4-2. ZLD Influent and Effluent Liquid Ra-226 Minimum, Maximum, and Average

Wastewater Source	Filtered or Not	Min (pCi/L)	Max (pCi/L)	Ave (pCi/L)
Effluent	Filtered	29.0	12,500	2,780
Effluent	Unfiltered	33.0	11,900	2,610
Influent	Filtered	38.5	20,900	4,660
Influent	Unfiltered	134	17,100	4,710

4.3.4 Indoor Radon Sampling Results

ATDs were deployed in the ZLD plants at various indoor locations such as break rooms, laboratories, offices, etc., and the results were evaluated using the EPA action level of 4 pCi/L. The results ranged from 0.500 to 4.90 pCi/L. Two results exceeded the action level. The results of the analyses are presented in **Table 4-45**. The Rn analytical reports are presented in **Appendix H**.

4.3.5 Filtered Versus Unfiltered Sample Data Evaluation

Appendix I contains a complete evaluation of filtered versus unfiltered liquid samples for the entire study. The conclusion from this evaluation is that there is no apparent trend or bias that filtering produces. There were some subsets of data where either the unfiltered results or the filtered results appear to be significantly higher. There was no statistically significant correlation found within any sample group. Since the liquid samples were preserved by addition of acid prior to filtering, the radioactive particulates may have entered solution and were therefore not removed by filtering.

4.3.6 ZLD Worker Exposure Assessment

4.3.6.1 ZLD Worker Potential External Gamma Radiation Exposure

The maximum average gamma radiation exposure rate measured at any of the ZLD plants was 43.1 µR/hr. The lowest background gamma radiation exposure rate measured at any of the sites was 5 µR/hr. Assuming the time period of exposure is a full occupational year of 2,000 hours, the maximum average ZLD annual external gamma radiation exposure was estimated as follows:

Maximum Average ZLD External Gamma Radiation Exposure Estimate

(43.1 – 5) µR/hr x 2,000 hr/yr x (1 mrem/1,000 µR gamma) = 76 mrem/yr

This is an estimate of the maximum average gamma radiation exposure based on 2,000 hours in one year. The result is less than the 100 mrem/yr dose equivalent limit for a member of the public. Actual exposure is dependent upon the actual exposure rates and occupancy time for individual workers.

The maximum gamma radiation exposure rate measured was 445 μrem/hr on contact with the outside of a wastewater tank. Work performed in the immediate proximity to the tank could potentially result in an exposure of 100 mrem in 225 hours of annual exposure, or about 10 percent of an employee's 2,000-hour occupational year. Actual annual exposure for a ZLD worker is dependent upon actual exposure rates and actual time worked in the proximity of the tank.

4.3.6.2 ZLD Worker Potential Internal Alpha/Beta Exposure

The total and removable α/β survey surface radioactivity results are discussed in Sections 4.3.1.1 and 4.3.1.2. One hundred fifty-nine of the 566 α measurements and 175 of the 566 β measurements of total surface radioactivity exceeded the RG 1.86 criteria. Fourteen of the 589 removable α measurements and two of the 589 removable β measurements exceeded the RG 1.86 criteria. The highest average total α and β surface radioactivity levels were 239 dpm/100 cm^2 and 4,740 dpm/100 cm^2. The maximum total α and β surface radioactivity levels were 1,410 dpm/100 cm^2 and 49,700 dpm/100 cm^2. The corresponding removable surface radioactivity measurements are mostly less than the RG 1.86 criteria, indicating the total surface radioactivity measured is fixed to the surface and not immediately available for inhalation or ingestion. Fixed α and β surface radioactivity may present a potential inhalation or ingestion hazard if disturbed during routine system maintenance.

4.3.6.3 ZLD Worker Potential Internal Radon Exposure

The Rn in ambient indoor area air averaged 2.29 pCi/L. The average is above the average typical background indoor level of 1.30 pCi/L in the U.S. as reported by EPA.

4.3.6.4 Gamma Radiation Exposure during Transport of Wastewater and Wastewater Sludge

Gamma radiation exposure was estimated for the transport of wastewater from well sites to WWTPs, and sludge from WWTPs to landfills. This was done for the driver of the transport truck. The truck driver spends the most time near the TENORM-influenced wastewater during transport.

It was assumed a truck driver hauled full containers with either wastewater or sludge/filter cake for four hours per day and made return trips with empty containers for four hours per day. The driver was assumed to work 40 hours per week for 10 weeks per year hauling O&G wastewater or sludge. Therefore, the total exposure time was assumed to be 200 hours per year as calculated below:

Estimated Duration of Gamma Radiation Exposure for Truck Driver per Year

4 hr/day x 5 days/wk x 10 wks/yr = 200 hrs/yr

Radiation exposure rates to the driver were not measured; they were modeled using the computer program MicroShield®. The MicroShield® output files are presented in **Appendix J**. Two external exposure scenarios were evaluated:

1. Exposure rate to a driver hauling wastewater based on the maximum measured concentrations of Ra-226 and Ra-228 in wastewater.

2. Exposure rate to a driver hauling sludge or filter cake based on the maximum measured concentrations of Ra-226 and Ra-228 in sludge.

The input and output of MicroShield® based on the two scenarios are summarized in **Figure 4-3**.

Figure 4-3. MicroShield® External Exposure Scenarios Input/Output

Parameter	Scenario	
	Wastewater Truck Maximum Measured Concentration, Scenario 1	**Sludge/Filter Cake Roll-off Maximum Measured Concentration, Scenario 2**
Volume	3,800 gallons	20 cubic yards
Shielding Material	Stainless steel, 0.5 cm thick	Iron, 0.3 cm thick
Ra-226 and Progeny Input Concentration	18,400 pCi/L	480 pCi/g
Ra-228 and Progeny Input Concentration	1,440 pCi/L	183 pCi/g
Resulting Driver Exposure Rate (µrem/hr)	14.7	1,340
Exposure Rate per Radium Concentration	0.000741 µrem/hr / pCi/L of total Ra	2.02 µrem/hr / pCi/g of total Ra

Maximum Wastewater Truck Driver External Gamma Radiation Exposure Estimate

0.000741 µrem/hr / pCi/L x 2,380 pCi/L x 200 hr/yr x (1 mrem/1,000 µrem gamma) = 0.35 mrem/yr

This is an estimate of the maximum annual gamma radiation exposure based on the maximum total Ra activity concentration of influent wastewater measured and 200 hours exposure in one year. The result is less than the 100 mrem/yr dose equivalent limit for a member of the public. Actual exposure is dependent upon the actual exposure rates and occupancy time for individual workers.

Maximum Sludge Truck Driver External Gamma Radiation Exposure Estimate

2.02 µrem/hr / pCi/g x 129 pCi/g x 200 hr/yr x (1 mrem/1,000 µrem gamma) = 52 mrem/yr

This is an estimate of the maximum annual gamma radiation exposure based on the maximum total Ra activity concentration in sludge measured and 200 hours of exposure in one year. The result is less than the 100 mrem/yr dose equivalent limit for a member of the public. Actual exposure is dependent upon the actual exposure rates and occupancy time for individual workers.

The sludge truck driver assessment is conservative due to the following: solid samples were dried prior to gamma spectroscopy analysis, artificially increasing the activity concentration results in direct proportion to the moisture content of the sample, i.e., after removal of the weight of the wastewater within the sludge sample. In addition, the MicroShield® activity input includes all of the Ra progeny in secular equilibrium. Often the sludge is "fresh," i.e., progeny ingrowth has not progressed to secular equilibrium and the progeny activity is only a fraction of the Ra activity.

4.3.7 Alpha Spectroscopy Analysis of Filter Cake

Elevated Ra-226 and Ra-228 and progeny activity were detected in CWT and ZLD filter cake samples analyzed by gamma spectroscopy. Due to the low solubility in water of U and Th, relative to Ra, U and Th were not present in wastewater and resulting filter cake at the elevated levels observed for Ra. Because gamma spectroscopy analysis of solid and liquid samples is limited in regards to the quantification of U and Th isotopes (Section 2.3), α spectroscopy analysis to measure U (U-238, U-234, and U-235) and Th (Th-232, Th-230, and Th-228), isotope activity levels was performed on 10 filter cake samples. The results are presented in **Table 4-46**. The U-238, U-234, and Th-230, all members of the natural U decay series above Ra-226, were measured at approximately 1/3 of typical background activity in soil. Uranium-235 is only identified once > MDC. Th-232, a member of the natural Th decay series above Ra-228, was measured at approximately ¼ of typical background activity in soil. Only Th-228, a progeny of Ra-228, was measured at activity concentrations comparable to Ra-228 identified by gamma spectroscopy. The α spectroscopy results confirm the low solubility of U and Th, resulting in low activity levels in wastewater and sludge/filter cake.

Table 4-1. POTW-I Removable Alpha and Beta Surface Radioactivity Measurement Results Summary

Study ID	No. of Data Points	Removable Alpha (dpm/100 cm^2)				Removable Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WT-12-FS-024	10	8.15	8.15	0.000	8.15	38.0	38.0	0.000	38.0
WT-12-FS-074	19	6.90	6.90	0.000	6.90	60.5	60.5	0.000	60.5
WT-12-FS-075	17	9.15	9.15	0.000	9.15	34.8	34.8	0.000	34.8
WT-13-FS-034	17	9.15	9.15	0.000	9.15	38.5	38.5	0.000	38.5
WT-13-FS-119	32	6.40	16.4	1.76	6.71	56.0	56.0	0.000	56.0
WT-13-FS-120	20	9.10	9.10	0.000	9.10	34.8	34.8	0.000	34.8
WT-14-FS-027	12	9.10	9.10	0.000	9.10	41.5	41.5	0.000	41.5
WT-14-FS-121	20	4.25	4.25	0.000	4.25	65.0	65.0	0.000	65.0
WT-14-FS-122	20	8.85	8.85	0.000	8.85	30.0	30.0	0.000	30.0
WT-15-FS-031	8	8.85	8.85	0.000	8.85	30.0	30.0	0.000	30.0
WT-15-FS-032	14	6.40	22.0	4.93	6.00	56.0	161	27.9	63.5
WT-15-FS-033	5	9.15	9.15	0.000	9.15	38.5	38.5	0.000	38.5
WT-16-FS-043	16	9.10	9.10	0.000	9.10	41.5	41.5	0.000	41.5
WT-16-FS-123	19	7.30	7.30	0.000	7.30	65.5	65.5	0.000	65.5
WT-16-FS-124	22	9.10	9.10	0.000	9.10	35.0	35.0	0.000	35.0
WT-17-FS-051	20	8.00	8.00	0.000	8.00	30.8	30.8	0.000	30.8
WT-17-FS-125	15	8.70	8.70	0.000	8.70	38.3	38.3	0.000	38.3

Note: During the calculations to convert from raw counts to dpm, the calculated value was compared to half of the MDC. If the value was below this number, half of the MDC was inserted into the tables. Where the standard deviation is zero and the minimum, maximum, and average are the same, then all measurements were below half of the MDC.

Table 4-2. POTW-I Total Alpha and Beta Surface Radioactivity Measurement Results Summary

Study ID	No. of Data Points	Total Alpha (dpm/100 cm^2)				Total Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WT-12-FS-024	10	29.4	29.4	0.000	29.4	100	563	144	413
WT-12-FS-074	19	7.30	43.7	10.1	19.7	308	308	0.000	308
WT-12-FS-075	17	7.45	54.5	14.2	18.27	269	1,550	268	870
WT-13-FS-034	17	30.5	74.4	13.7	37.0	847	2,130	325	1,290
WT-13-FS-119	15	18.6	875	220	88.8	305	728	117	337
WT-13-FS-120	20	19.0	164	33.9	30.2	280	1,530	391	811
WT-14-FS-027	13	30.5	30.5	0.000	30.5	773	1,540	197	1,130
WT-14-FS-121	20	18.6	112	26.1	37.0	254	1,490	352	515
WT-14-FS-122	20	30.5	89.3	20.3	38.8	268	1,630	359	784
WT-15-FS-031	8	30.5	1,190	437	313	268	38,000	14,800	10,000
WT-15-FS-032	4	18.6	18.6	0.000	18.6	263	466	102	313
WT-15-FS-033	5	30.5	30.5	0.000	30.5	735	1,360	259	1,070
WT-16-FS-043	16	30.5	30.5	0.000	30.5	676	29,800	7,170	2,930
WT-16-FS-123	19	7.45	24.9	6.39	11.4	276	1,140	272	498
WT-16-FS-124	22	7.45	34.7	10.2	12.7	273	1,200	295	593
WT-17-FS-051	20	7.45	54.5	13.4	16.0	313	929	159	363
WT-17-FS-125	15	29.8	134	32.8	61.0	313	2,760	704	773

Note: During the calculations to convert from raw counts to dpm the calculated value was compared to half of the MDC. If the value was below this number, half of the MDC was inserted into the tables. Where the standard deviation is zero and the minimum, maximum, and average are the same then all measurements were below half of the MDC.

Table 4-3. POTW-I Gross Gamma Radiation Scan Results Summary

Site	GWS Max[a] (cpm)	GWS Min[a] (cpm)	GWS Average[a] (cpm)	GWS Std Dev (cpm)	No. Data Points
12	9,514	4,966	7,184	633	7,129
13	9,362	3,404	5,072	829	4,408
13	20,761	3,608	6,019	2,694	8,553
13	18,203	3,486	5,418	2,082	5,474
14	33,141	3,112	5,582	2,517	7,638
14	29,220	3,867	6,110	2,272	7,302
14	32,253	3,680	6,435	3,812	3,275
15	131,626	3,804	20,392	14,569	3,508
15	162,535	5,684	18,319	16,130	7,334
15	205,446	5,452	29,034	36,865	3,052
16	10,005	3,463	5,671	870	9,390
16	13,915	3,723	5,628	1,050	9,520
16	13,597	3,473	6,871	1,722	2,026
17	150,649	3,305	9,194	10,116	4,509
17	156,738	3,478	11,137	17,801	3,003

[a]Convert count rate data to exposure rate by dividing count rate by 800 to yield µR/hr.

Table 4-4. POTW-I Results Summary of NaI Count Rate Data Converted to Exposure Rates

Site	GWS Max (µR/hr)	GWS Min (µR/hr)	GWS Average (µR/hr)	GWS Std Dev (µR/hr)	No. Data Points
12	11.9	6.21	8.98	0.791	7,129
13	11.7	4.26	6.34	1.04	4,408
13	26.0	4.51	7.52	3.37	8,553
13	22.8	4.36	6.77	2.60	5,474
14	41.4	3.89	6.98	3.15	7,638
14	36.5	4.83	7.64	2.84	7,302
14	40.3	4.60	8.04	4.77	3,275
15	165	4.76	25.5	18.2	3,508
15	203	7.11	22.9	20.2	7,334
15	257	6.82	36.3	46.1	3,052
16	12.5	4.33	7.09	1.09	9,390
16	17.4	4.65	7.04	1.31	9,520
16	17.0	4.34	8.59	2.15	2,026
17	188	4.13	11.5	12.6	4,509
17	196	4.35	13.9	22.3	3,003

Table 4-5. POTW-I Filter Cake Results Summary – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)
WT-12-SL-030	6.37	1.56	4.04
WT-12-SL-048	9.75	1.87	6.94
WT-12-SL-085	5.16	0.854	2.69
WT-13-SL-021	6.50	3.08	3.96
WT-13-SL-060	21.3	2.99	9.38
WT-13-SL-065	17.4	8.69	3.93
WT-14-SL-017	55.6	32.0	7.77
WT-14-SL-052	9.27	2.80	14.3
WT-14-SL-068	13.1	6.73	6.71
WT-15-SL-057	41.9	19.7	12.9
WT-16-SL-026	5.01	1.29	6.95
WT-16-SL-044	52.6	5.21	7.78
WT-16-SL-073	2.71	0.894	0.822
WT-17-SL-059	35.1	19.2	6.14
Average	20.1	7.63	6.74
Std. Dev.	18.5	9.40	3.71
Median	11.4	3.04	6.83
Minimum	2.71	0.854	0.822
Maximum	55.6	32.0	14.3

Table 4-6. POTW-N Filter Cake Results Summary – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)
WT-26-SL-094	3.97	1.31	5.47
WT-26-SL-095	3.61	1.46	5.41
WT-27-SL-096	2.33	0.817	6.51
WT-27-SL-097	5.76	1.12	4.31
WT-28-SL-098	7.36	1.84	6.57
WT-28-SL-099	3.78	1.07	6.55
WT-29-SL-100	35.4	7.26	7.66
WT-29-SL-101	15.6	3.28	7.34
Average	9.72	2.26	6.23
Std. Dev.	11.2	2.16	1.10
Median	4.87	1.39	6.53
Minimum	2.33	0.817	4.31
Maximum	35.4	7.26	7.66

Table 4-7. POTW-I Sediment Sample Results Summary – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)
WT-14-SL-018	4.25	1.96	10.3
WT-14-SL-053	1.83	0.799	8.71
WT-14-SL-069	3.94	1.96	5.53
WT-15-SL-020	16.6	6.25	15.7
WT-15-SL-056	18.2	6.19	13.0
WT-15-SL-067	15.3	5.77	24.5
WT-17-SL-058	2.91	1.69	6.20
Average	9.00	3.52	12.0
Std. Dev.	7.29	2.42	6.58
Median	4.25	1.96	10.3
Minimum	1.83	0.799	5.53
Maximum	18.2	6.25	24.5

Table 4-8. POTW-I Filtered Effluent Results Summary – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WT-12-LQ-098	134	< 18.0	< 66.0	< 196	< 392
WT-12-LQ-159	< 127	< 25.0	81.0	< 5.77	10.6
WT-12-LQ-295	77.0	< 13.0	42.0	195	365
WT-13-LQ-054	< 126	< 22.0	73.0	< 29.6	< 18.9
WT-13-LQ-193	101	< 16.0	46.0	< 114	< 198
WT-13-LQ-209	363	< 10.0	53.0	< 123	< 203
WT-14-LQ-044	< 130	< 24.0	56.0	< 25.8	< 163
WT-14-LQ-171	87.0	< 12.0	60.0	< 111	< 186
WT-14-LQ-215	104	< 13.0	71.0	< 118	< 202
WT-15-LQ-052	191	< 24.0	< 81.0	< 21.3	< 16.2
WT-15-LQ-185	< 139	< 25.0	< 98.0	< 5.67	8.70
WT-15-LQ-223	120	25.0	52.0	< 161	< 198
WT-16-LQ-079	101	< 8.00	34.0	< 2.26	5.77
WT-16-LQ-145	57.0	< 6.00	55.0	< 6.96	11.3
WT-16-LQ-241	335	< 9.00	< 32.0	4.64	10.7
WT-17-LQ-191	154	< 18.0	< 48.0	< 121	< 187
WT-17-LQ-217	116	12.0	< 33.0	< 127	< 203
Average	129	9.34	48.1	42.9	75.0
Std. Dev.	93.1	5.35	19.0	49.6	88.8
Median	101	8.50	50.5	35.1	87.3
Minimum	57.0	3.00	16.0	1.13	5.77
Maximum	363	25.0	81.0	195	365

< – indicates a value less than the reported number which is the MDC.

Table 4-9. POTW-I Unfiltered Effluent Results Summary – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WT-12-LQ-097	< 67.0	< 10.0	51.1	< 284	< 396
WT-12-LQ-160	94.0	< 11.0	41.0	9.63	10.9
WT-12-LQ-296	59.0	< 5.00	40.0	< 192	< 207
WT-13-LQ-053	113	< 8.00	37.0	< 36.5	< 135
WT-13-LQ-194	82.0	< 5.00	55.0	< 117	< 187
WT-13-LQ-210	< 35.0	< 23.0	< 11.0	< 144	< 194
WT-14-LQ-043	122	< 18.0	80.0	< 84.2	< 158
WT-14-LQ-172	340	< 15.0	< 58.0	< 464	< 218
WT-14-LQ-216	< 128	< 27.0	< 106	< 136	< 193
WT-15-LQ-051	80.0	< 9.00	53.0	< 177	< 163
WT-15-LQ-186	135	< 9.00	< 27.0	11.0	9.60
WT-15-LQ-224	< 79.0	27.0	64.0	< 235	< 209
WT-16-LQ-080	100	< 9.00	33.0	< 3.13	7.16
WT-16-LQ-146	< 67.0	< 11.0	< 41.0	< 2.16	7.71
WT-16-LQ-242	107	< 9.00	44.0	< 2.51	10.5
WT-17-LQ-192	100	21.0	82.0	1,110	337
WT-17-LQ-218	156	35.0	31.0	< 152	< 197
Average	103	10.4	42.6	125	82.1
Std. Dev.	73.7	9.40	21.5	269	79.3
Median	97.0	5.75	40.5	63.3	87.5
Minimum	17.5	2.50	5.50	1.08	7.16
Maximum	340	35.0	82.0	1,110	337

< – indicates a value less than the reported number which is the MDC.

Table 4-10. POTW-N Filtered Effluent Results Summary – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WT-26-LQ-300	< 74.0	15.0	60.0	< 7.65	5.29
WT-27-LQ-304	< 44.0	< 5.00	42.0	< 10.8	5.72
WT-28-LQ-308	< 23.0	< 5.00	53.0	< 4.78	7.64
WT-29-LQ-312	116	17.0	56.0	< 4.83	14.6
Average	46.6	9.25	52.8	3.51	8.31
Std. Dev.	47.4	7.84	7.72	1.43	4.31
Median	29.5	8.75	54.5	3.12	6.68
Minimum	11.5	2.50	42.0	2.39	5.29
Maximum	116	17.0	60.0	5.40	14.6

< – indicates a value less than the reported number which is the MDC.

Table 4-11. POTW-N Unfiltered Effluent Results Summary – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WT-26-LQ-299	328	< 9.00	< 34.0	< 6.46	5.75
WT-27-LQ-303	115	< 7.00	57.0	< 7.48	7.48
WT-28-LQ-307	78.0	< 14.0	49.0	< 5.18	7.15
WT-29-LQ-311	59.0	5.00	66.0	< 191	< 209
Average	145	5.00	47.3	26.3	31.2
Std. Dev.	124	1.47	21.3	46.2	48.9
Median	96.5	4.75	53.0	3.49	7.32
Minimum	59.0	3.50	17.0	2.59	5.75
Maximum	328	7.00	66.0	95.5	105

< – indicates a value less than the reported number which is the MDC.

Table 4-12. POTW-I Filtered Influent Results Summary – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WT-12-LQ-096	66.0	8.00	49.0	< 5.64	< 7.91
WT-12-LQ-157	109	< 14.0	32.0	< 13.2	< 5.01
WT-12-LQ-293	100	8.00	63.0	< 290	< 230
WT-13-LQ-056	< 154	< 29.0	137	< 207	< 394
WT-13-LQ-195	115	< 20.0	< 68.0	< 183	< 201
WT-13-LQ-211	58.0	6.00	53.0	< 13.2	< 8.48
WT-14-LQ-042	260	< 48.0	< 171	< 16.8	< 15.5
WT-14-LQ-169	< 77.0	< 12.0	< 41.0	489	< 199
WT-14-LQ-213	82.0	10.0	63.0	< 323	< 230
WT-15-LQ-050	498	< 28.0	< 82.0	< 17.3	< 16.1
WT-15-LQ-183	245	103	< 141	11.0	9.60
WT-15-LQ-225	255	91.0	31.0	490	< 207
WT-16-LQ-077	< 84.0	< 17.0	119	< 2.63	6.24
WT-16-LQ-143	5,910	878	44.0	11,400	11,300
WT-16-LQ-243	66.0	5.00	43.0	< 3.31	6.75
WT-17-LQ-189	< 121	23.0	33.0	< 117	< 198
WT-17-LQ-219	< 74.0	20.0	49.0	< 154	< 196
Average	497	76.8	56.9	768	722
Std. Dev.	1,450	216	31.4	2,740	2,730
Median	91.0	12.0	49.0	58.5	98.0
Minimum	37.0	5.00	20.5	1.32	2.51
Maximum	5,910	878	137	11,400	11,300

< – indicates a value less than the reported number which is the MDC.

Table 4-13. POTW-I Unfiltered Influent Results Summary – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WT-12-LQ-095	< 113	< 19.0	< 59.0	< 220	< 392
WT-12-LQ-158	90.0	< 15.0	< 54.0	6.28	10.1
WT-12-LQ-294	345	< 7.00	< 21.0	< 110	< 201
WT-13-LQ-055	91.0	< 16.0	69.0	< 14.4	76.4
WT-13-LQ-196	95.0	< 15.0	72.0	< 287	< 224
WT-13-LQ-212	96.0	< 9.00	54.0	< 13.4	14.5
WT-14-LQ-041	259	< 48.0	< 171	< 14.8	17.2
WT-14-LQ-170	57.0	20.0	65.0	< 118	< 199
WT-14-LQ-214	120	9.00	47.0	< 301	< 227
WT-15-LQ-049	< 73.0	< 15.0	< 50.0	< 4.32	4.89
WT-15-LQ-184	514	48.0	< 67.0	240	< 196
WT-15-LQ-226	479	227	< 102	1,190	493
WT-16-LQ-078	343	< 9.00	< 5.00	< 1.85	7.50
WT-16-LQ-144	106	< 9.00	30.0	< 3.91	9.94
WT-16-LQ-244	131	41.0	65.0	< 7.48	9.64
WT-17-LQ-190	100	14.0	56.0	< 120	< 200
WT-17-LQ-220	178	20.0	45.0	< 125	< 203
Average	190	28.1	46.1	125	85.9
Std. Dev.	146	52.9	22.4	283	114
Median	120	9.00	47.0	55.0	92.0
Minimum	36.5	3.50	2.50	0.925	4.89
Maximum	514	227	85.5	1,190	493

< – indicates a value less than the reported number which is the MDC.

Table 4-14. POTW-N Filtered Influent Results Summary – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WT-26-LQ-298	134	10.0	30.0	13.0	6.62
WT-27-LQ-302	64.0	< 5.00	38.0	15.2	11.6
WT-28-LQ-306	84.0	< 14.0	62.0	4.57	12.4
WT-29-LQ-310	58.0	< 4.00	52.0	< 5.29	8.38
Average	85.0	5.38	45.5	8.85	9.75
Std. Dev.	34.5	3.82	14.3	6.17	2.71
Median	74.0	4.75	45.0	8.79	9.99
Minimum	58.0	2.00	30.0	2.65	6.62
Maximum	134	10.0	62.0	15.2	12.4

< – indicates a value less than the reported number which is the MDC.

Table 4-15. POTW-N Unfiltered Influent Results Summary – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WT-26-LQ-297	113	< 10.0	< 33.0	< 173	< 207
WT-27-LQ-301	92.0	32.0	44.0	< 192	< 209
WT-28-LQ-305	91.0	< 10.0	43.0	< 169	< 207
WT-29-LQ-309	114	< 9.00	< 29.0	< 4.21	8.63
Average	103	11.6	29.5	67.3	80.0
Std. Dev.	12.7	13.6	16.2	43.7	47.6
Median	103	5.00	29.8	85.5	104
Minimum	91.0	4.50	14.5	2.11	8.63
Maximum	114	32.0	44.0	96.0	105

< – indicates a value less than the reported number which is the MDC.

Table 4-16. POTW-I Ambient Radon

Facility	Location	Radon (pCi/L)	Percent Error
WT-17-RA-001	Lab	2.20	4%
WT-17-RA-002	Filter Press Room	3.10	3%
WT-17-RA-003	Not Given	0.200	12%
WT-15-RA-001	Old Lab	0.700	7%
WT-12-RA-001	Filter Press Room	0.500	8%
WT-12-RA-002	Break Room	0.500	8%
WT-14-RA-001	Press Room Shelf	0.700	7%
WT-14-RA-002	Break Room	8.70	2%
WT-16-RA-001	Filter Press Room	0.600	9%
WT-16-RA-002	Break Room	1.20	7%
WT-13-RA-001	Load and Filter	0.900	6%
WT-13-RA-002	Lab	1.60	5%
	Average	1.74	
	Median	0.800	
	St. Dev.	2.34	
	Minimum	0.200	
	Maximum	8.70	

Note: ATDs. Lower level of detection (LLD) for 10 pCi/L-day is 0.1 pCi/L for 90-day test, 0.3 pCi/L for 30-day test.

Table 4-17. POTW-I vs POTW-N Average Concentrations Comparison for Filtered

Filtered Sample Set Averages for:	Ra-226 (pCi/L)	Ra-228 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
POTW-I Effluent	129	9.34	42.9	75.0
POTW-N Effluent	46.6	9.25	3.51	8.31
POTW-I Influent	497	76.8	768	722
POTW-N Influent	85.0	5.38	8.85	9.75

Table 4-18. POTW-I vs POTW-N Average Concentrations Comparison for Unfiltered

Unfiltered Sample Set Averages for:	Ra-226 (pCi/L)	Ra-228 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
POTW-I Effluent	103	10.4	125	82.1
POTW-N Effluent	145	5.00	26.3	31.2
POTW-I Influent	190	28.1	125	85.9
POTW-N Influent	103	11.6	67.3[a]	80.0

[a]All sample results were < MDC value reported.

Table 4-19. Average Radium, Gross Alpha, and Gross Beta Concentrations for Filtered Influent and Effluent POTW Samples

Filtered Sample Set Averages for:	Ra-226 (pCi/L)	Ra-228 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
POTW-I Influent	497	76.8	768	722
POTW-I Effluent	129	9.34	42.9	75.0
POTW-N Influent	85.0	5.38	8.85	9.75
POTW-N Effluent	46.6	9.25	3.51	8.31

Table 4-20. Average Radium, Gross Alpha, and Gross Beta Concentrations for Unfiltered Influent and Effluent POTW Samples

Unfiltered Sample Set Averages for:	Ra-226 (pCi/L)	Ra-228 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
POTW-I Influent	190	28.1	125	85.9
POTW-I Effluent	103	10.4	125	82.1
POTW-N Influent	103	11.6	67.3[a]	80.0
POTW-N Effluent	145	5.00	26.3	31.2

[a]All sample results were < MDC value reported.

Table 4-21. POTW-I Sediment and Effluent Results for Ra-226 and Ra-228

Sample Set	Study ID	Sample Type	Ra-226	Units	Ra-228	Units	Ra-226/ Ra-228 Ratio
POTW 1 Round 2	WT-17-SL-058	Sediment	2.91	pCi/g	1.69	pCi/g	1.72
	WT-17-LQ-218	Effluent - Unfiltered	156	pCi/L	35.0	pCi/L	4.46
	WT-17-LQ-217	Effluent - Filtered	116	pCi/L	12.0	pCi/L	9.67
POTW 2 Round 1	WT-14-SL-018	Sediment	4.25	pCi/g	1.96	pCi/g	2.17
	WT-14-LQ-043[a]	Effluent - Unfiltered	122	pCi/L	9.00	pCi/L	13.6
	WT-14-LQ-044[a]	Effluent - Filtered	65.0	pCi/L	12.0	pCi/L	5.42
POTW 2 Round 2	WT-14-SL-053	Sediment	1.83	pCi/g	0.799	pCi/g	2.29
	WT-14-LQ-172[a]	Effluent - Unfiltered	340	pCi/L	7.50	pCi/L	45.3
	WT-14-LQ-171	Effluent - Filtered	87.0	pCi/L	6.00	pCi/L	14.5
POTW 2 Round 3	WT-14-SL-069	Sediment	3.94	pCi/g	1.96	pCi/g	2.01
	WT-14-LQ-216	Effluent - Unfiltered	64.0	pCi/L	13.5	pCi/L	4.74
	WT-14-LQ-215	Effluent - Filtered	104	pCi/L	6.50	pCi/L	16.0
POTW 3 Round 1	WT-15-SL-020	Sediment	16.6	pCi/g	6.25	pCi/g	2.66
	WT-15-LQ-051	Effluent - Unfiltered	80.0	pCi/L	4.50	pCi/L	17.8
	WT-15-LQ-052[a]	Effluent - Filtered	191	pCi/L	12.0	pCi/L	15.9
POTW 3 Round 2	WT-15-SL-056	Sediment	18.2	pCi/g	6.19	pCi/g	2.94
	WT-15-LQ-186[a]	Effluent - Unfiltered	135	pCi/L	4.50	pCi/L	30.0
	WT-15-LQ-185[a]	Effluent - Filtered	69.5	pCi/L	12.5	pCi/L	5.56
POTW 3 Round 3	WT-15-SL-067	Sediment	15.3	pCi/g	5.77	pCi/g	2.65
	WT-15-LQ-224	Effluent - Unfiltered	39.5	pCi/L	27.0	pCi/L	1.46
	WT-15-LQ-223	Effluent - Filtered	120	pCi/L	25.0	pCi/L	4.80

[a] Result was not detected, ½ of the reported MDC was presented.

Table 4-22. POTW Sediment and Effluent Ratios for Ra-226/Ra-228

Ratio Statistic	Sediments (CWT + POTW)	Sediments (CWT)	Sediments (POTW)	Unfiltered (CWT + POTW)	Unfiltered (CWT)	Unfiltered (POTW)	Filtered (CWT + POTW)	Filtered (CWT)	Filtered (POTW)
Average	3.00	3.40	2.40	8.40	11.4	5.30	5.70	3.80	8.30
Std Dev	0.900	0.900	0.400	6.70	8.30	3.40	3.90	3.60	3.00
Max	4.80	4.80	2.90	21.3	21.3	10.0	10.4	9.20	10.4
Min	1.70	2.30	1.70	1.00	1.00	2.00	1.10	1.10	4.80

Table 4-23. Summary of Removable Alpha and Beta Surface Contamination Results at CWT Plants

Study ID	No. of Data Points	Removable Alpha (dpm/100 cm^2)				Removable Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WT-01-FS-021	22	7.30	18.6	2.90	8.27	62.5	62.5	0.000	62.5
WT-01-FS-108	38	8.00	18.6	2.61	8.05	62.5	62.5	0.000	62.5
WT-01-FS-109	25	8.00	18.1	2.02	8.40	30.8	30.8	0.000	30.8
WT-02-FS-012	20	7.30	15.8	1.33	7.51	62.5	62.5	0.000	62.5
WT-02-FS-066	41	8.15	8.15	0.000	8.15	38.0	38.0	0.000	38.0
WT-02-FS-067	29	8.00	29.4	4.56	9.18	30.8	30.8	0.000	30.8
WT-03-FS-040	5	6.90	6.90	0.000	6.90	60.5	60.5	0.000	60.5
WT-03-FS-110	10	8.00	8.00	0.000	8.00	31.0	31.0	0.000	31.0
WT-03-FS-111	16	6.90	6.90	0.000	6.90	60.5	60.5	0.000	60.5
WT-04-FS-025	19	9.15	9.15	0.000	9.15	113	113	0.000	113
WT-04-FS-112	37	7.70	38.1	4.94	8.50	62.0	62.0	0.000	62.0
WT-04-FS-113	25	9.10	9.10	0.000	9.10	69.6	69.6	0.000	69.6
WT-05-FS-044	25	9.11	9.11	0.000	9.11	41.5	41.5	0.000	41.5
WT-05-FS-114	45	6.40	13.6	1.07	6.56	56.0	56.0	0.000	56.0
WT-05-FS-115	23	9.10	9.10	0.000	9.10	32.5	32.5	0.000	32.5
WT-07-FS-022	14	9.15	9.15	0.000	9.15	23.3	23.3	0.000	23.3
WT-07-FS-071	35	7.30	7.30	0.000	7.30	62.5	62.5	0.000	62.5
WT-07-FS-072	15	8.00	8.00	0.000	8.00	36.6	36.6	0.000	36.6
WT-08-FS-015	25	9.10	9.10	0.000	9.10	41.5	41.5	0.000	41.5
WT-08-FS-062	46	7.70	7.70	0.000	7.70	62.0	62.0	0.000	62.0
WT-08-FS-063	32	9.10	9.10	0.000	9.10	41.5	41.5	0.000	41.5
WT-09-FS-013	17	9.10	9.10	0.000	9.10	41.5	41.5	0.000	41.5
WT-09-FS-060	27	4.25	31.1	5.40	5.77	65.0	133	13.1	67.5
WT-09-FS-061	20	8.85	8.85	0.000	8.85	30.0	30.0	0.000	30.0
WT-10-FS-002	22	8.15	8.15	0.000	8.15	38.0	38.0	0.000	38.0
WT-10-FS-046	34	6.90	6.90	0.000	6.90	60.5	60.5	0.000	60.5

Table 4-23. Summary of Removable Alpha and Beta Surface Contamination Results at CWT Plants

Study ID	No. of Data Points	Removable Alpha (dpm/100 cm^2)				Removable Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WT-10-FS-047	20	9.10	9.10	0.000	9.10	32.5	32.5	0.000	32.5
WT-11-FS-005	15	8.15	8.15	0.000	8.15	36.0	36.0	0.000	36.0

Note: During the calculations to convert from raw counts to dpm, the calculated value was compared to half of the MDC. If the value was below this number, half of the MDC was inserted into the tables. Where the standard deviation is zero and the minimum, maximum, and average are the same, then all measurements were below half of the MDC.

Table 4-24. Summary of Total Alpha and Beta Surface Contamination Results at CWT Plants

Study ID	No. of Data Points	Total Alpha (dpm/100 cm^2)				Total Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WT-01-FS-021	22	30.5	1,540	315	211	929	50,400	10,900	8,780
WT-01-FS-108	30	7.30	476	133	172	283	32,700	7,030	5,310
WT-01-FS-109	25	14.9	448	113	152	287	13,200	3,870	4,090
WT-02-FS-012	20	30.5	332	77.9	58.9	268	8,220	1,710	1,690
WT-02-FS-066	41	7.30	403	120	92.6	240	8,260	2,060	1,590
WT-02-FS-067	29	19.1	473	114	74.5	286	9,040	1,900	1,140
WT-03-FS-040	5	19.0	347	144	115	334	6,310	2,710	2,410
WT-03-FS-110	10	7.45	487	204	194	288	7,120	2,070	1,940
WT-03-FS-111	13	18.6	3,220	877	348	249	30,200	8,170	3,150
WT-04-FS-025	20	30.5	565	157	123	268	8,560	2,290	3,210
WT-04-FS-112	38	18.6	540	137	142	297	14,600	3,720	3,200
WT-04-FS-113	25	7.45	1,600	310	144	291	14,200	3,940	3,480
WT-05-FS-044	25	7.44	179	44.6	61.5	325	3,370	771	1,230
WT-05-FS-114	32	7.30	180	45.1	53.2	257	3,060	829	1,340
WT-05-FS-115	23	19.0	243	82.2	71.9	306	7,380	1,480	1,290
WT-07-FS-022	14	30.5	922	250	132	891	6,650	1,490	2,480
WT-07-FS-071	36	18.6	1,000	206	130	249	5,330	1,210	1,140
WT-07-FS-072	13	19.0	1,390	399	213	310	6,620	1,990	1,740
WT-08-FS-015	25	30.5	208	43.9	50.9	572	3,270	780	1,920
WT-08-FS-062	46	19.1	194	39.5	56.0	284	3,880	1,010	1,370
WT-08-FS-063	32	7.45	94.2	27.5	40.8	290	2,580	696	1,050
WT-09-FS-013	18	30.5	258	56.0	51.0	728	11,900	2,540	2,260
WT-09-FS-060	26	18.6	117	27.3	35.3	354	7,120	1,600	1,280
WT-09-FS-061	20	35.7	35.7	0.000	35.7	286	6,640	1,540	1,690
WT-10-FS-002	22	29.4	224	54.9	53.1	121	2,730	623	395
WT-10-FS-046	34	18.6	476	81.9	44.0	288	5,770	972	623

Table 4-24. Summary of Total Alpha and Beta Surface Contamination Results at CWT Plants

Study ID	No. of Data Points	Total Alpha (dpm/100 cm^2)				Total Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WT-10-FS-047	21	7.45	174	37.6	24.0	297	1,760	366	482
WT-11-FS-005	15	30.5	114	26.6	49.0	617	3,380	746	1,350

Note: During the calculations to convert from raw counts to dpm, the calculated value was compared to half of the MDC. If the value was below this number, half of the MDC was inserted into the tables. Where the standard deviation is zero and the minimum, maximum, and average are the same, then all measurements were below half of the MDC.

Table 4-25. Summary of NaI Count Rate Data at CWTs

Site	GWS Max[a] (cpm)	GWS Min[a] (cpm)	GWS Average[a] (cpm)	GWS Std Dev (cpm)	No. Data Points
1	152,322	4,717	18,543	19,037	2,192
1	252,693	3,273	12,750	24,179	9,513
1	178,291	4,843	17,806	23,505	2,077
2	69,545	4,844	13,849	10,904	2,360
2	33,174	3,850	8,141	2,490	4,743
2	203,895	4,909	19,281	29,028	2,057
3	12,172	5,208	8,375	916	1,162
3	13,983	4,579	7,790	1,655	3,741
3	111,523	5,120	13,819	14,182	2,950
4	288,000	5,448	11,725	24,058	6,492
4	401,688	5,445	15,883	38,194	6,720
4	20,932	7,065	9,310	1,114	3,015
5	20,666	4,751	7,273	752	12,166
5	10,640	5,766	7,532	650	7,274
5	10,369	5,805	7,414	625	5,977
7	9,397	5,124	6,742	796	825
8	27,735	2,611	6,927	3,495	2,924
8	9,915	2,718	5,223	975	6,552
8	24,840	2,723	7,302	3,383	1,812
9	33,141	3,112	5,582	2,517	7,638
9	29,220	3,867	6,110	2,272	7,302
10	12,455	4,175	5,880	1,093	5,790
10	13,200	7,756	5,708	1,398	7,756
11	150,649	3,305	9,194	10,116	4,509
11	156,738	3,478	11,137	17,801	3,003

[a]Convert count rate data to exposure rate by dividing count rate by 800 to yield µrem/hr.

Table 4-26. Results Summary of NaI Count Rate Data Converted to Exposure Rates

Site	GWS Max (µR/hr)	GWS Min (µR/hr)	GWS Average (µR/hr)	GWS Std Dev (µR/hr)	No. Data Points
1	190	5.90	23.2	23.8	2,192
1	316	4.09	15.9	30.2	9,513
1	223	6.05	22.3	29.4	2,077
2	86.9	6.06	17.3	13.6	2,360
2	41.5	4.81	10.2	3.11	4,743
2	255	6.14	24.1	36.3	2,057
3	15.2	6.51	10.5	1.15	1,162
3	17.5	5.72	9.74	2.07	3,741
3	139	6.40	17.3	17.7	2,950
4	360	6.81	14.7	30.1	6,492
4	502	6.81	19.9	47.7	6,720
4	26.2	8.83	11.6	1.39	3,015
5	25.8	5.94	9.09	0.940	12,166
5	13.3	7.21	9.42	0.813	7,274
5	13.0	7.26	9.27	0.781	5,977
7	11.7	6.41	8.43	1.00	825
8	34.7	3.26	8.66	4.37	2,924
8	12.4	3.40	6.53	1.22	6,552
8	31.1	3.40	9.13	4.23	1,812
9	41.4	3.89	6.98	3.15	7,638
9	36.5	4.83	7.64	2.84	7,302
10	15.6	5.22	7.35	1.37	5,790
10	16.5	9.70	7.14	1.75	7,756
11	188	4.13	11.5	12.6	4,509
11	196	4.35	13.9	22.3	3,003

Table 4-27. CWT Solids, Filter Cake – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)
WT-01-SL-009	208	106	< 1.33
WT-01-SL-037	261	137	< 2.01
WT-01-SL-084	256	132	12.0
WT-02-SL-006	120	75.0	15.7
WT-02-SL-036	118	66.0	12.8
WT-02-SL-081	164	97.2	13.0
WT-03-SL-012	56.6	13.5	10.7
WT-04-SL-013	59.9	57.3	7.65
WT-04-SL-050	35.1	36.0	5.04
WT-04-SL-062	70.1	59.4	5.22
WT-04-SL-063	165	91.7	8.74
WT-05-SL-022	82.1	49.8	9.91
WT-05-SL-061	10.1	5.03	6.06
WT-05-SL-064	104	52.4	9.13
WT-08-SL-027	67.5	6.46	7.47
WT-08-SL-047	35.7	3.59	10.5
WT-08-SL-072	52.1	4.46	4.13
WT-08-SL-088	41.1	3.45	< 0.553
WT-08-SL-089	15.7	2.44	17.4
WT-09-SL-019	174	108	9.05
WT-09-SL-054	269	164	13.7
WT-09-SL-066	294	177	16.1
WT-10-SL-029	3.88	0.363	0.969
WT-10-SL-049	5.97	0.687	2.89
WT-06-SL-045	24.7	2.74	11.1
Average	108	58.1	8.45
Std. Dev.	91.0	55.7	5.03
Median	70.1	52.4	9.05
Minimum	3.88	0.363	0.277
Maximum	294	177	17.4

< – indicates a value less than the reported number which is the MDC.

Table 4-28. CWT Solids, Sediment – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)
WT-01-SL-010	105	29.7	8.44
WT-01-SL-038	37.2	12.4	7.17
WT-01-SL-083	76.8	20.0	8.31
WT-02-SL-007	5.86	2.59	4.55
WT-02-SL-035	3.60	1.37	4.67
WT-02-SL-082	2.50	0.978	9.26
WT-03-SL-011	4.72	1.54	6.34
WT-04-SL-014	101	22.7	10.1
WT-04-SL-051	421	86.9	10.0
Average	84.2	19.8	7.65
Std. Dev.	133	27.4	2.11
Median	37.2	12.4	8.31
Minimum	2.50	0.978	4.55
Maximum	421	86.9	10.1

Table 4-29. CWT Solids, Biased Soil – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)	U-238 (pCi/g)	U-235 (pCi/g)
WT-01-SL-008	117	30.6	17.0	< 2.46	1.83
WT-02-SL-034	13.3	4.26	5.06	< 3.14	< 0.331
WT-04-SL-015	444	83.1	10.5	< 3.37	< 0.774
Average	191	39.3	10.9	1.50	0.794
Std. Dev.	225	40.1	5.98	0.240	0.904
Median	117	30.6	10.5	1.57	0.387
Minimum	13.3	4.26	5.06	1.23	0.166
Maximum	444	83.1	17.0	1.69	1.83

< – indicates a value less than the reported number which is the MDC.

Table 4-30. CWT Filtered Effluent – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha[a] (pCi/L)	Gross Beta (pCi/L)
WT-01-LQ-023	110	< 19.0	334	< 1,270	< 847
WT-01-LQ-115	< 169	55.0	406	< 1,040	< 909
WT-01-LQ-281	287	< 18.0	235	< 2,040	< 879
WT-02-LQ-021	113	< 15.0	116	13.1	< 263
WT-02-LQ-111	86.0	< 16.0	140	< 1,340	< 872
WT-02-LQ-279	55.0	6.00	174	< 1,950	< 870
WT-03-LQ-029	< 36.0	< 5.00	52.0	< 50.1	45.7
WT-03-LQ-121	91.0	< 11.0	52.0	< 104	< 190
WT-03-LQ-287	86.0	< 9.00	62.0	< 192	< 208
WT-04-LQ-031	76.0	37.0	403	< 692	< 422
WT-04-LQ-165	104	94.0	618	< 2,200	< 940
WT-04-LQ-201	320	68.0	339	< 1,040	< 802
WT-05-LQ-058	215	118	595	< 762	504
WT-05-LQ-197	150	< 9.00	282	< 950	608
WT-05-LQ-207	181	80.0	607	< 1,810	< 938
WT-07-LQ-015	5,510	849	888	ND	7,660
WT-07-LQ-109	1,630	324	586	2,330	1,080
WT-07-LQ-273	8,810	1,740	360	21,400	8,700
WT-08-LQ-081	84.0	< 9.00	< 30.0	1.13	< 0.998
WT-08-LQ-085	12,700	1,110	304	22,800	5,810
WT-08-LQ-151	< 79.0	< 15.0	49.0	8.25	1.98
WT-08-LQ-153	14,900	1,300	598	22,700	4,570
WT-08-LQ-237	12,400	1,220	388	40,700	12,100
WT-09-LQ-046	< 73.0	< 12.0	148	ND	69.4
WT-09-LQ-175	503	319	181	< 1,120	< 895
WT-09-LQ-227	273	164	188	< 2,550	< 989
WT-10-LQ-094	150	< 17.0	< 96.0	< 204	< 393
WT-10-LQ-161	363	10.0	203	< 126	< 187
WT-10-LQ-291	77.0	< 13.0	55.0	< 161	< 196
WT-11-LQ-187	1,700	943	238	5,520	1,670
WT-11-LQ-221	2,090	976	228	4,160	1,730
Average	2,100	316	285	4,460	1,650
Std. Dev.	4,250	510	221	9,847	3,013
Median	166	37.0	232	540	444
Minimum	18.0	2.50	15.0	1.13	0.499
Maximum	14,900	1,740	888	40,700	12,100

[a]ND – Non-detectable; sample matrix was not suitable for analysis.
< – indicates a value less than the reported number which is the MDC.

Table 4-31. CWT Unfiltered Effluent – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228[a] (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WT-01-LQ-024	104	< 18.0	296	< 1,340	< 871
WT-01-LQ-116	< 196	26.0	381	< 1,130	< 844
WT-01-LQ-282	114	< 15.0	270	< 2,650	< 1,000
WT-02-LQ-022	64.0	< 5.00	113	< 689	< 444
WT-02-LQ-112	< 116	< 18.0	140	< 1,250	< 804
WT-02-LQ-280	108	< 10.0	162	< 2,600	< 994
WT-03-LQ-030	61.0	< 8.00	29.0	< 260	< 181
WT-03-LQ-122	126	< 13.0	36.0	< 142	< 191
WT-03-LQ-288	362	11.0	< 30.0	< 213	< 211
WT-04-LQ-032	124	84.0	406	ND	480
WT-04-LQ-166	117	112	568	< 1,030	1,280
WT-04-LQ-202	< 131	< 27.0	361	< 1,450	< 846
WT-05-LQ-057	357	133	565	< 595	< 453
WT-05-LQ-198	< 202	89.0	688	< 1,320	< 500
WT-05-LQ-208	240	92.0	648	< 912	< 845
WT-07-LQ-110	1,670	318	571	2,370	1,060
WT-07-LQ-274	8,050	1,740	1,450	33.6	5,380
WT-08-LQ-082	87.0	< 4.00	37.0	< 1.66	< 1.17
WT-08-LQ-086	10,300	912	371	18,900	4,900
WT-08-LQ-152	85.0	6.00	42.0	4.68	< 2.01
WT-08-LQ-154	15,500	1,250	414	17,100	4,440
WT-08-LQ-238	12,700	1,200	355	42,300	12,900
WT-09-LQ-045	161	28.0	118	0.260	< 341
WT-09-LQ-176	594	331	200	1,810	1,540
WT-09-LQ-228	404	166	233	1,410	< 869
WT-10-LQ-093	42.0	6.00	80.0	< 294	< 397
WT-10-LQ-162	< 138	< 27.0	217	< 205	202
WT-10-LQ-292	< 95.0	< 10.0	69.0	< 224	< 209
WT-11-LQ-188	1,840	996	264	3,460	1,410
WT-11-LQ-222	1,470	1,100	252	3,880	1,320
Average	1,840	289	312	3,430	1,330
Std. Dev.	4,070	486	291	8,750	2,610
Median	121	27.0	258	565	423
Minimum	42.0	2.00	15.0	0.260	0.585
Maximum	15,500	1,740	1,450	42,300	12,900

[a]ND – Non-detectable; sample matrix was not suitable for analysis.
< – indicates a value less than the reported number which is the MDC.

Table 4-32. CWT Filtered Influent – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha[a] (pCi/L)	Gross Beta (pCi/L)
WT-01-LQ-025	1,760	711	345	ND	3,040
WT-01-LQ-117	2,810	1,120	603	10,500	2,970
WT-01-LQ-283	1,900	961	304	3,940	1,950
WT-02-LQ-019	1,650	747	272	ND	2,810
WT-02-LQ-113	1,660	913	247	2,360	1,900
WT-02-LQ-277	1,770	962	300	3,930	2,760
WT-03-LQ-027	116	< 16.0	< 63.0	< 129	< 149
WT-03-LQ-119	121	< 19.0	< 54.0	< 205	< 202
WT-03-LQ-285	126	< 5.00	36.0	< 227	< 212
WT-04-LQ-033	175	172	419	< 369	276
WT-04-LQ-167	445	392	626	660	1,510
WT-04-LQ-203	216	173	394	< 1,450	< 846
WT-05-LQ-060	57.0	56.0	< 111	< 2,550	< 998
WT-05-LQ-199	118	48.0	547	< 579	587
WT-05-LQ-205	242	78.0	514	< 1,040	< 802
WT-07-LQ-013	1,390	203	163	2,290	1,310
WT-07-LQ-107	1,930	322	505	3,420	893
WT-07-LQ-275	1,410	203	219	1,920	853
WT-08-LQ-083	87.0	6.00	37.0	6,110	1,570
WT-08-LQ-155	14,100	1,520	526	22,200	4,640
WT-08-LQ-239	7,080	615	203	28,400	7,820
WT-09-LQ-047	469	247	121	1,310	< 811
WT-09-LQ-173	300	238	176	1,950	1,360
WT-10-LQ-092	97.0	< 15.0	95.0	< 220	< 392
WT-10-LQ-163	132	< 10.0	345	< 294	276
WT-10-LQ-289	102	8.00	55.0	< 312	< 231
Average	1,550	361	273	3,862	1,430
Std. Dev.	3,015	431	198	7,086	1,760
Median	300	203	247	1,293	853
Minimum	57.0	2.50	27.0	64.5	74.5
Maximum	14,100	1,520	626	28,400	7,820

[a]ND – Non-detectable; sample matrix was not suitable for analysis.
< – indicates a value less than the reported number which is the MDC.

Table 4-33. CWT Unfiltered Influent – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha[a] (pCi/L)	Gross Beta (pCi/L)
WT-01-LQ-026	1,430	740	333	4,830	1,780
WT-01-LQ-118	2,870	1,110	592	8,400	3,440
WT-01-LQ-284	1,820	984	243	2,940	1,420
WT-02-LQ-020	1,740	835	245	3,220	1,890
WT-02-LQ-114	3,630	1,920	< 373	47,100	12,800
WT-02-LQ-278	1,790	1,010	279	4,220	1,650
WT-03-LQ-028	100	< 8.00	33.0	< 188	< 163
WT-03-LQ-120	327	< 17.0	< 55.0	< 116	< 199
WT-03-LQ-286	66.0	6.00	48.0	< 158	< 212
WT-04-LQ-034	214	229	459	ND	1,030
WT-04-LQ-168	453	467	< 69.0	< 1,700	1,130
WT-04-LQ-204	286	228	433	< 883	< 842
WT-05-LQ-059	146	77.0	493	< 910	< 430
WT-05-LQ-200	492	86.0	550	< 575	591
WT-05-LQ-206	238	126	526	< 2,040	1,200
WT-07-LQ-014	1,330	188	171	1,890	485
WT-07-LQ-108	2,330	366	468	3,490	1,180
WT-07-LQ-276	1,030	203	227	1,740	638
WT-08-LQ-084	5,920	367	159	7,960	2,550
WT-08-LQ-156	13,400	1,520	544	27,700	6,870
WT-08-LQ-240	6,940	623	184	27,600	10,200
WT-09-LQ-048	950	328	< 99.0	< 746	343
WT-09-LQ-174	458	222	151	2,050	1,040
WT-10-LQ-091	< 37.0	< 6.00	67.0	< 198	< 393
WT-10-LQ-164	< 98.0	< 8.00	328	< 117	375
WT-10-LQ-290	< 35.0	< 6.00	59.0	< 123	< 203
Average	1,870	436	262	5,920	2,000
Std. Dev.	3,010	515	192	11,600	3,220
Median	492	228	227	1,380	1,030
Minimum	17.5	3.00	27.5	58.0	81.5
Maximum	13,400	1,920	592	47,100	12,800

[a]ND – Non-detectable; sample matrix was not suitable for analysis.
< – indicates a value less than the reported number which is the MDC.

Table 4-34. CWT Radon Sample Results

Facility	Location	Radon (pCi/L)	Percent Error
WT-05-RA-001	Conference Room	3.10	3%
WT-05-RA-002	Near Filter Press	0.900	6%
WT-04-RA-001	Filter Press 2	1.90	4%
WT-04-RA-002	2nd Fl. Office	1.60	5%
WT-04-RA-003	Break Room	1.60	5%
WT-08-RA-001	On fuse panel	4.00	4%
WT-08-RA-002	Lab	1.50	6%
WT-09-RA-001	Office	2.00	4%
WT-09-RA-002	Filter Press Area	3.00	3%
WT-10-RA-001	Under Filter Press	1.20	5%
WT-07-RA-001	Lab Fridge	1.40	7%
WT-07-RA-002	Clarifier Elec. Panel	0.900	8%
WT-03-RA-001	Influent Wastewater Pump	1.30	7%
WT-03-RA-002	Wastewater Receiving Office	1.20	8%
WT-02-RA-001	Office	1.20	7%
WT-02-RA-002	Filter Press	1.30	7%
WT-01-RA-001	Wastewater Receiving Off.	5.00	4%
WT-01-RA-002	Top of Filter Press	2.90	5%
	Average	2.00	
	Median	1.55	
	St. Dev.	1.14	
	Minimum	0.900	
	Maximum	5.00	

ATDs. LLD for 10 pCi/L-day is 0.1 pCi/L for 90-day test, 0.3 pCi/L for 30-day test.

Table 4-35. Summary of Removable Alpha and Beta Surface Contamination Results at ZLDs

Study ID	No. of Data Points	Removable Alpha (dpm/100 cm^2)				Removable Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WT-06-FS-039	24	7.30	7.30	0.000	7.30	61.5	61.5	0.000	61.5
WT-06-FS-116	46	7.70	18.4	1.57	7.92	62.0	62.0	0.000	34.0
WT-06-FS-117	33	9.35	25.1	3.22	10.1	34.0	34.0	0.000	34.0
WT-18-FS-011	15	6.90	6.90	0.000	6.90	123	123	0.000	123
WT-18-FS-058	31	7.30	7.30	0.000	7.30	65.5	193	22.8	69.6
WT-18-FS-059	20	6.40	22.0	3.77	7.54	32.8	32.8	0.000	32.8
WT-19-FS-078	13	9.15	9.15	0.000	9.15	38.5	38.5	0.000	38.5
WT-19-FS-079	17	6.40	6.40	0.000	6.40	56.0	56.0	0.000	56.0
WT-19-FS-080	18	6.40	6.40	0.000	6.40	32.8	32.8	0.000	32.8
WT-20-FS-020	16	9.10	9.10	0.000	9.10	41.6	41.6	0.000	41.6
WT-20-FS-068	39	6.40	30.5	4.91	7.93	56.0	56.0	0.000	56.0
WT-20-FS-069	32	6.40	22.0	3.07	7.25	33.0	33.0	0.000	33.0
WT-21-FS-030	4	7.85	17.7	2.05	8.28	36.4	36.4	0.000	36.4
WT-21-FS-126	45	7.30	35.6	5.45	8.68	62.5	62.5	0.000	62.5
WT-21-FS-127	39	8.00	294	3.42	8.55	36.6	36.6	0.000	36.6
WT-22-FS-001	10	7.15	7.15	0.000	7.15	37.5	37.5	0.000	37.5
WT-22-FS-048	28	7.30	38.4	5.88	8.41	63.0	342	52.8	72.5
WT-22-FS-049	18	8.00	8.00	0.000	8.00	30.8	30.8	0.000	30.8
WT-23-FS-007	23	8.70	30.4	4.53	9.64	76.5	76.5	0.000	76.5
WT-23-FS-054	33	4.24	4.24	0.000	4.24	65.0	65.0	0.000	65.0
WT-23-FS-055	25	9.10	9.10	0.000	9.10	32.5	32.5	0.000	32.5
WT-24-FS-016	20	7.85	7.85	7.85	0.000	36.5	36.5	0.000	36.5
WT-24-FS-064	21	8.00	8.00	0.000	8.00	30.8	30.8	0.000	30.8
WT-24-FS-065	41	6.90	6.90	0.000	6.90	60.5	307	39.5	68.0
WT-25-FS-006	23	7.15	70.7	17.6	12.7	37.5	37.5	0.000	37.5

Table 4-35. Summary of Removable Alpha and Beta Surface Contamination Results at ZLDs

Study ID	No. of Data Points	Removable Alpha (dpm/100 cm^2)				Removable Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WT-25-FS-052	25	8.85	22.9	2.81	9.41	60.0	60.0	0.000	60.0
WT-25-FS-053	25	6.40	36.2	8.18	9.58	32.8	65.9	6.61	34.1

Note: During the calculations to convert from raw counts to dpm, the calculated value was compared to half of the MDC. If the value was below this number, half of the MDC was inserted into the tables. Where the standard deviation is zero and the minimum, maximum, and average are the same, then all measurements were below half of the MDC.

Table 4-36. Summary of Total Alpha and Beta Surface Contamination Results at ZLDs

Study ID	No. of Data Points	Total Alpha (dpm/100 cm^2)				Total Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WT-06-FS-039	23	30.5	139	32.2	47.3	1,950	49,700	9,810	4,740
WT-06-FS-116	46	19.1	691	134	103	474	7,760	1,420	2,540
WT-06-FS-117	33	7.45	248	53.0	81.5	1,210	8,710	1,540	2,440
WT-18-FS-011	15	18.6	194	44.4	75.0	415	4,200	1,131	2,100
WT-18-FS-058	20	730	199	57.1	78.9	211	7,190	1,610	2,360
WT-18-FS-059	20	19.1	249	60.3	69.1	277	4,670	1,080	1,720
WT-19-FS-078	13	30.5	114	23.2	36.9	943	2,370	411	1,550
WT-19-FS-079	17	7.30	72.9	22.2	27.3	277	1,490	369	553
WT-19-FS-080	18	19.1	54.7	10.1	22.5	318	705	91.2	339
WT-20-FS-020	16	30.5	719	215	222	268	6,990	2,230	3,080
WT-20-FS-068	27	7.30	554	154	150	249	8,830	2,240	2,030
WT-20-FS-069	32	19.1	741	165	174	321	8,800	1,840	1,550
WT-21-FS-030	23	30.5	645	159	111	780	13,400	2,730	2,440
WT-21-FS-126	44	18.6	452	127	127	264	17,900	3,420	2,540
WT-21-FS-127	39	7.45	537	111	49.8	283	3,090	713	960
WT-22-FS-001	10	30.5	273	87.0	85.2	269	3,180	1,050	1,620
WT-22-FS-048	28	7.30	836	226	133	249	15,500	3,290	2,080
WT-22-FS-049	18	19.1	1,410	350	239	265	6,380	1,640	1,730
WT-23-FS-007	25	7.45	273	73.1	83.5	313	6,230	1,380	1,550
WT-23-FS-054	32	18.6	72.9	14.6	25.1	250	2,660	537	920
WT-23-FS-055	25	7.45	193	43.3	43.1	313	4,520	905	927
WT-24-FS-016	20	305	466	123	107	268	4,420	977	2,150
WT-24-FS-064	21	7.45	711	187	125	288	4,380	980	1,060
WT-24-FS-065	41	18.6	476	90.8	69.2	260	9,410	1,530	985
WT-25-FS-006	13	30.5	213	55.4	89.5	802	3,980	921	1,660

Table 4-36. Summary of Total Alpha and Beta Surface Contamination Results at ZLDs

Study ID	No. of Data Points	Total Alpha (dpm/100 cm^2)				Total Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
WT-25-FS-052	25	7.45	373	109	97.3	307	3,820	1,040	1,120
WT-25-FS-053	24	19.1	433	97.9	81.7	321	4,900	1,140	893

Note: During the calculations to convert from raw counts to dpm, the calculated value was compared to half of the MDC. If the value was below this number, half of the MDC was inserted into the tables. Where the standard deviation is zero and the minimum, maximum, and average are the same, then all measurements were below half of the MDC.

Table 4-37. Summary of NaI Count Rate Data at ZLDs

Site	GWS Max[a] (cpm)	GWS Min[a] (cpm)	GWS Average[a] (cpm)	GWS Std Dev (cpm)	No. Data Points
6	11,264	3,689	6,618	1,435	1,077
6	11,273	4,157	6,315	1,037	4,716
18	7,446	2,692	4,507	714	3,570
18	34,596	2,748	7,432	5,069	2,032
19	15,542	10,665	13,449	573	3,379
19	15,603	11,347	13,667	560	4,098
19	52,815	4,506	13,153	3,995	2,813
20	11,574	3,266	5,966	1,814	7,086
20	73,475	3,771	8,426	8,110	9,495
21	66,958	4,752	12,383	7,293	1,911
21	34,908	4,335	6,912	2,613	15,435
21	46,611	4,351	7,797	4,423	8,792
22	42,518	4,857	10,358	5,297	1,544
22	39,712	4,065	6,937	4,905	5,063
23	12,198	5,546	8,585	1,250	6,265
23	13,938	5,662	9,014	1,348	7,512
24	12,234	5,164	7,419	1,279	1,712
24	11,844	6,541	8,985	1,211	2,959
25	28,597	7,558	12,955	2,243	5,371
25	31,290	2,819	12,524	2,352	8,019
25	356,274	4,464	34,513	63,202	2,006

[a]Convert count rate data to exposure rate by dividing count rate by 800 to yield µR/hr.

Table 4-38. Results Summary of NaI Count Rate Data Converted to Exposure Rates

Site	GWS Max (µR/hr)	GWS Min (µR/hr)	GWS Average (µR/hr)	GWS Std Dev (µR/hr)	No. Data Points
6	14.1	4.61	8.27	1.79	1,077
6	14.1	5.20	7.89	1.30	4,716
18	9.31	3.37	5.63	0.893	3,570
18	43.2	3.44	9.29	6.34	2,032
19	19.4	13.3	16.8	0.716	3,379
19	19.5	14.2	17.1	0.700	4,098
19	66.0	5.63	16.4	4.99	2,813
20	14.5	4.08	7.46	2.27	7,086
20	91.8	4.71	10.5	10.1	9,495
21	83.7	5.94	15.5	9.12	1,911
21	43.6	5.42	8.64	3.27	15,435
21	58.3	5.44	9.75	5.53	8,792
22	53.1	6.07	12.9	6.62	1,544
22	49.6	5.08	8.67	6.13	5,063
23	15.2	6.93	10.7	1.56	6,265
23	17.4	7.08	11.3	1.69	7,512
24	15.3	6.46	9.27	1.60	1,712
24	14.8	8.18	11.2	1.51	2,959
25	35.7	9.45	16.2	2.80	5,371
25	39.1	3.52	15.7	2.94	8,019
25	445	5.58	43.1	79.0	2,006

Table 4-39. ZLD Solids, Filter Cake – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)
WT-06-SL-046	159	14.2	7.67
WT-06-SL-074	31.7	3.48	14.9
WT-18-SL-025	8.02	2.01	26.3
WT-18-SL-043	6.14	1.63	21.7
WT-18-SL-076	19.1	1.95	5.95
WT-19-SL-023	4.62	1.44	17.5
WT-19-SL-041	127	11.0	16.6
WT-19-SL-070	3.08	0.580	7.46
WT-20-SL-024	26.9	2.62	11.2
WT-20-SL-042	20.0	2.24	10.0
WT-20-SL-075	22.7	2.21	13.4
WT-20-SL-086	11.1	1.40	6.51
WT-20-SL-087	10.2	1.41	6.55
WT-21-SL-004	6.46	1.54	21.1
WT-21-SL-039	29.3	9.34	10.8
WT-21-SL-078	25.8	7.09	25.4
WT-21-SL-092	214	43.6	12.5
WT-21-SL-093	212	40.5	10.3
WT-22-SL-003	281	17.8	14.1
WT-22-SL-032	145	19.2	15.9
WT-22-SL-079	134	13.1	2.75
WT-23-SL-016	78.9	18.1	8.62
WT-23-SL-055	33.6	6.87	4.28
WT-23-SL-077	26.0	3.39	1.61
WT-24-SL-001	420	58.7	5.25
WT-24-SL-002	41.6	5.26	3.02
WT-24-SL-031	480	67.3	5.16
WT-24-SL-080	289	46.3	5.26
WT-25-SL-028	221	25.1	2.76
WT-25-SL-040	185	24.2	3.27
WT-25-SL-071	206	32.4	3.47
Average	112	15.7	8.53
Std. Dev.	128	18.6	6.09
Median	33.6	6.98	6.55
Minimum	3.08	0.580	1.61
Maximum	480	67.3	25.4

Table 4-40. ZLD Solids, Biased Soil – Uranium Series Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)	U-238 (pCi/g)	U-235 (pCi/g)
WT-21-SL-005	37.1	7.47	16.6	3.81	< 0.201

< – indicates a value less than the reported number which is the MDC.

Table 4-41. ZLD Filtered Effluent – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
WT-06-LQ-076	12,000	908	< 552	19,600	4,840
WT-06-LQ-149	11,200	806	385	13,300	3,340
WT-06-LQ-245	8,360	571	273	13,700	2,100
WT-18-LQ-070	335	< 16.0	159	< 485	< 413
WT-18-LQ-139	86.0	< 10.0	648	< 383	435
WT-18-LQ-253	94.0	< 10.0	149	701	< 832
WT-19-LQ-062	< 127	< 21.0	56.0	0.0970	135
WT-19-LQ-133	< 58.0	< 8.00	55.0	< 293	< 225
WT-19-LQ-229	126	< 11.0	338	< 412	< 234
WT-20-LQ-066	8,930	1,090	< 339	11,800	2,440
WT-20-LQ-135	12,500	941	206	31,100	6,190
WT-20-LQ-251	11,100	910	316	14,400	4,110
WT-21-LQ-011	3,470	503	807	6,830	2,160
WT-21-LQ-123	5,050	750	646	10,900	2,650
WT-21-LQ-261	4,690	725	885	10,200	2,890
WT-22-LQ-007	418	< 17.0	487	< 542	284
WT-22-LQ-105	3,280	241	738	5,040	1,530
WT-22-LQ-269	2,310	163	183	2,690	515
WT-23-LQ-038	580	111	186	1,660	602
WT-23-LQ-040	< 82.0	< 14.0	< 30.0	5.05	3.10
WT-23-LQ-177	110	12.0	54.0	< 145	< 191
WT-23-LQ-179	587	96.0	670	< 1,340	< 504
WT-23-LQ-257	< 69.0	< 7.00	< 41.0	23.6	< 4.03
WT-23-LQ-259	2,540	280	< 64.0	9,610	3,210
WT-24-LQ-001	1,830	277	429	2,540	655
WT-24-LQ-101	2,260	204	339	3,660	1,520
WT-24-LQ-265	292	120	799	< 2,090	< 967
WT-25-LQ-088	173	< 12.0	190	< 1,140	< 827
WT-25-LQ-127	163	15.0	113	< 1,100	< 475
WT-25-LQ-235	59.0	< 10.0	134	< 479	< 424
Average	2,780	272	327	5,250	1,370
Std. Dev.	3,880	348	270	7,220	1,560
Median	580	111	206	1,660	515
Minimum	29.0	3.50	15.0	0.0970	2.02
Maximum	12,500	1,090	885	31,100	6,190

< – indicates a value less than the reported number which is the MDC.

Table 4-42. ZLD Unfiltered Effluent – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha[a] (pCi/L)	Gross Beta (pCi/L)
WT-06-LQ-075	12,100	914	275	13,700	3,770
WT-06-LQ-150	11,300	866	326	27,300	6,530
WT-06-LQ-246	7,950	523	256	37,600	12,600
WT-18-LQ-069	5,490	875	982	14,100	3,820
WT-18-LQ-140	< 80.0	< 20.0	674	< 140	573
WT-18-LQ-254	106	< 10.0	143	< 641	< 780
WT-19-LQ-061	130	< 19.0	102	ND	92.6
WT-19-LQ-134	104	< 16.0	111	< 108	< 198
WT-19-LQ-230	< 66.0	< 11.0	333	< 280	231
WT-20-LQ-065	8,830	1,090	400	14,500	3,540
WT-20-LQ-136	1,580	221	4,310	40,900	8,340
WT-20-LQ-252	11,900	862	299	42,800	13,900
WT-21-LQ-012	3,770	552	821	5,540	1,850
WT-21-LQ-124	5,120	785	612	16,000	5,530
WT-21-LQ-262	4,370	721	926	13,100	4,020
WT-22-LQ-008	165	19.0	439	< 275	< 460
WT-22-LQ-106	2,730	250	723	8,940	1,630
WT-22-LQ-270	2,240	178	190	5,100	1,260
WT-23-LQ-037	531	121	160	1,570	358
WT-23-LQ-039	116	< 12.0	31.0	4.94	< 1.78
WT-23-LQ-178	< 85.0	< 16.0	< 60.0	< 217	< 203
WT-23-LQ-180	800	109	497	1,220	871
WT-23-LQ-258	87.0	< 12.0	< 42.0	5.12	26.1
WT-23-LQ-260	2,640	308	340	13,300	4,030
WT-24-LQ-002	2,040	269	431	2,750	< 424
WT-24-LQ-102	2,480	301	358	4,440	1,300
WT-24-LQ-266	293	102	748	< 810	< 836
WT-25-LQ-087	< 146	< 31.0	158	< 917	< 831
WT-25-LQ-128	601	305	4,840	< 448	< 417
WT-25-LQ-236	< 126	< 25.0	158	< 1,030	< 475
Average	2,610	295	670	8,990	2,510
Std. Dev.	3,470	337	1,120	13,000	3,697
Median	800	178	340	2,160	573
Minimum	33.0	5.00	21.0	4.94	0.890
Maximum	12,100	1,090	4,840	42,800	13,900

< – indicates a value less than the reported number which is the MDC.
[a]ND – Non-detectable; sample matrix was not suitable for analysis.

Table 4-43. ZLD Filtered Influent – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha[a] (pCi/L)	Gross Beta[a] (pCi/L)
WT-06-LQ-073	12,100	1,100	393	21,400	4,530
WT-06-LQ-147	11,300	1,290	302	23,500	5,630
WT-06-LQ-247	3,910	230	215	13,100	4,340
WT-18-LQ-072	278	< 24.0	234	< 427	< 412
WT-18-LQ-141	< 77.0	< 14.0	848	< 175	592
WT-19-LQ-064	950	901	16,600	ND	ND
WT-19-LQ-131	131	13.0	281	< 175	< 190
WT-19-LQ-231	1,140	91.0	718	4,770	1,860
WT-20-LQ-068	13,200	1,390	399	18,700	4,740
WT-20-LQ-137	20,900	603	< 187	59,400	10,700
WT-20-LQ-249	18,400	1,410	491	36,000	7,680
WT-21-LQ-009	2,580	338	517	ND	2,403
WT-21-LQ-125	3,360	515	584	4,750	1,340
WT-21-LQ-263	6,190	687	350	17,100	4,460
WT-22-LQ-005	106	10.0	299	< 257	2,400
WT-22-LQ-103	16,300	847	< 371	30,800	3,730
WT-22-LQ-271	590	51.0	105	754	< 198
WT-23-LQ-035	1,300	413	421	828	425
WT-23-LQ-181	564	94.0	135	2,080	492
WT-23-LQ-255	226	28.0	158	497	< 207
WT-24-LQ-003	2,580	332	552	3,630	1,530
WT-24-LQ-099	1,920	153	341	2,300	395
WT-24-LQ-267	832	380	568	< 1,330	< 838
WT-25-LQ-090	6,650	660	202	8,920	1,030
WT-25-LQ-129	2,100	181	187	2,290	396
WT-25-LQ-233	903	127	169	3,220	1,320
Average	4,660	431	998	10,200	2,350
Std. Dev.	6250	443	3,260	15,000	2,730
Median	1,920	332	302	3,220	1,330
Minimum	38.5	7.00	93.5	87.5	95.0
Maximum	20,900	1,410	16,600	59,400	10,700

[a]ND – Non-detectable; sample matrix was not suitable for analysis.
< – indicates a value less than the reported number which is the MDC.

Table 4-44. ZLD Unfiltered Influent – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha[a] (pCi/L)	Gross Beta[a] (pCi/L)
WT-06-LQ-074	12,200	1,090	7,210	17,700	5,920
WT-06-LQ-148	11,100	1,240	350	25,500	5,950
WT-06-LQ-248	4,300	250	243	7,700	1,570
WT-18-LQ-071	1,310	142	318	ND	ND
WT-18-LQ-142	134	< 21.0	761	497	806
WT-19-LQ-063	1,470	777	13,300	ND	ND
WT-19-LQ-132	11,700	1000	< 247	2,230	2,080
WT-19-LQ-232	1,600	81.0	701	2,800	1,180
WT-20-LQ-067	13,600	1,390	288	16,200	6,060
WT-20-LQ-138	210	19.0	123	49,200	10,600
WT-20-LQ-250	16,500	1,310	529	88,000	23,400
WT-21-LQ-010	3,030	429	605	6,590	1,610
WT-21-LQ-126	2,620	421	528	6,920	2,400
WT-21-LQ-264	6,560	727	415	18,900	4,530
WT-22-LQ-006	216	14.0	136	110	105
WT-22-LQ-104	17,100	903	332	52,400	11,500
WT-22-LQ-272	750	43.0	234	1,240	231
WT-23-LQ-036	1,280	437	410	ND	2,240
WT-23-LQ-182	665	95.0	160	1,300	535
WT-23-LQ-256	221	41.0	153	1,120	423
WT-24-LQ-004	2,700	457	651	3,640	1,320
WT-24-LQ-100	2,100	181	220	3,380	782
WT-24-LQ-268	632	388	558	< 1,470	1,060
WT-25-LQ-089	6,870	628	269	9,270	977
WT-25-LQ-130	1,560	140	114	1,810	466
WT-25-LQ-234	1,930	199	161	4,470	1,400
Average	4,710	453	867	13,800	3,530
Std. Dev.	5,310	433	2,600	22,100	5,340
Median	1,930	388	318	4,060	1,400
Minimum	134	10.5	114	110	105
Maximum	17,100	1,390	13,300	88,000	23,400

[a]ND – Non-detectable; sample matrix was not suitable for analysis.
< – indicates a value less than the reported number which is the MDC.

Table 4-45. ZLD Radon in Ambient Air Results

Facility	Location	Radon (pCi/L)	Percent Error
WT-06-RA-001	Filter Press	2.20	5%
WT-06-RA-002	Lab	2.40	5%
WT-18-RA-001	Centrifuge	0.900	8%
WT-18-RA-002	Lab	4.30	4%
WT-20-RA-001	Transfer Panel	1.90	5%
WT-20-RA-002	Break Area	2.60	5%
WT-23-RA-001	Break Room	0.500	8%
WT-23-RA-002	Ctrl Panel/Boiler Room	1.70	6%
WT-23-RA-003	First Floor	0.900	8%
WT-21-RA-001	Locker Room Shelf	3.70	4%
WT-21-RA-002	Back of Filter Cake Room	2.60	5%
WT-24-RA-001	Filter Press	2.90	5%
WT-24-RA-002	Office	1.90	6%
WT-22-RA-001	Filter Press Room	4.90	4%
WT-22-RA-002	Wastewater Receiving Office	0.900	8%
	Average	2.29	
	Median	2.20	
	St. Dev.	1.28	
	Minimum	0.500	
	Maximum	4.90	

Note: ATDs. LLD for 10 pCi/L-day is 0.1 pCi/L for 90-day test, 0.3 pCi/L for 30-day test.

Table 4-46. ZLD and CWT Filter Cake Sample Alpha Spectroscopy Results

Study ID	U-238 (pCi/g)	U-234 (pCi/g)	Th-230 (pCi/g)	Th-232 (pCi/g)	Th-228 (pCi/g)	U-235 (pCi/g)
WT-04-SL-063	0.306	0.361	0.307	< 0.205	76.2	< 0.134
WT-25-SL-028	< 0.068	< 0.084	< 0.050	< 0.041	9.87	< 0.084
WT-22-SL-079	0.225	0.281	0.431	< 0.198	8.07	< 0.031
WT-19-SL-041	0.683	0.830	0.502	0.401	8.55	0.163
WT-01-SL-084	< 0.265	< 0.266	< 0.686	< 0.685	1.81	< 0.403
WT-08-SL-047	0.922	0.910	0.525	0.428	7.18	< 0.116
WT-06-SL-046	0.708	0.746	0.473	0.157	8.76	< 0.079
WT-04-SL-050	< 0.246	< 0.248	< 0.237	< 0.145	6.03	< 0.250
WT-09-SL-054	< 0.064	< 0.053	< 0.160	< 0.159	48.3	< 0.065
WT-23-SL-055	0.268	0.291	< 0.173	< 0.111	5.52	< 0.052
Average	0.343	0.374	0.289	0.176	18.0	0.077
St. Dev	0.314	0.334	0.195	0.154	24.3	0.064
Median	0.247	0.286	0.325	0.101	8.31	0.050
Minimum	0.032	0.027	0.025	0.021	1.81	0.016
Maximum	0.922	0.910	0.525	0.428	76.2	0.202

5.0 LANDFILLS

Leachate samples were collected at 51 PA landfills. Nine of the 51 landfills were selected to be surveyed and sampled in more detail due to the volume of waste accepted from the O&G industry. Surveys at the nine selected landfills included scans of gamma radiation and measurements of total and removable α/β surface radioactivity. Ambient air at the fence line of these landfills was sampled for Rn analysis, and filter cake was sampled from three of these landfills.

5.1 Leachate

Samples of leachate were collected from 51 landfills and analyzed using gamma spectroscopy for Ra-226 and Ra-228. The gamma spectroscopy results are presented in **Table 5-1** for the 42 landfills not selected based on volume of O&G waste accepted and **Table 5-2** for the nine landfills selected based on the volume O&G waste accepted. Radium was detected above the MDC value in 38 of 51 samples. Sample results from the 42 unselected landfills showed Ra-226 results that ranged from 36.5 to 416 pCi/L with an average of 116 pCi/L. Radium-226 results from the nine selected landfills ranged from 67.0 pCi/L to 378 pCi/L with an average of 125 pCi/L. Radium-228 results ranged from 2.50 to 55.0 pCi/L with an average of 11.9 pCi/L in the 42 unselected landfills. Radium-228 results from the nine selected landfills ranged from 3.00 pCi/L to 84.0 pCi/L with an average of 18.0 pCi/L.

Due to high solids content, the samples were not filtered in the field or at the laboratory. The aqueous portion was decanted from 10 of the 51 samples after they had been allowed to settle. The aqueous portion was analyzed for Ra-226 and Ra-228. These results are presented in **Table 5-3** along with the original gamma spectroscopy results for the entire sample. The entire sample results include dissolved and undissolved Ra-226 and Ra-228 and are generally one to two orders of magnitude higher than analyses of only the aqueous phase, indicating that the Ra-226 and Ra-228 in these samples were mostly in the form of undissolved solids.

5.2 Nine Selected Landfills

5.2.1 Influent and Effluent Leachate

Nine influent and seven effluent leachate samples were collected at the nine selected landfills. All nine landfills treat leachate onsite. The samples were analyzed using gamma spectroscopy. The results of the Ra-226, Ra-228, K-40, as well as gross α and gross β activity levels are presented in **Table 5-4** for effluent samples and in **Table 5-5** for influent samples. Radium was detected in all but 3 of the leachate samples. Radium-226 results ranged from 67.0 to 378 pCi/L with an average of 142 pCi/L for effluent samples. Radium-228 results ranged from 3.00 to 1,100 pCi/L with an average of 178 pCi/L for effluent samples. Radium-226 results ranged from 48.5 to 116 pCi/L with an average of 83.4 pCi/L for influent samples. Radium-228 results ranged from 4.00 to 15.0 pCi/L with an average of 7.94 pCi/L for influent samples. The influent and effluent samples from the same facility do not represent the same leachate at different times in treatment.

5.2.2 Leachate Filter Cake

Filter cake from three of the nine landfills was sampled and analyzed using gamma spectroscopy. The results of the Ra-226 and Ra-228 analyses are presented in **Table 5-6**. Radium was detected

in all of the filter cake samples. Radium-226 results ranged from 8.73 to 53.0 pCi/g, with an average of 24.3 pCi/g. Radium-228 results ranged from 1.53 to 5.03 pCi/g, with an average of 3.85 pCi/g.

5.2.3 Effluent Discharge Sediment-Impacted Soil

At three landfills that discharged effluent water to the environment, a sediment-impacted soil sample was collected at each of the three effluent outfalls. The gamma spectroscopy results are presented in **Table 5-7**. Radium was detected in all of the samples. Radium-226 results ranged from 2.82 to 4.46 pCi/g with an average of 3.57 pCi/g. Radium-228 results ranged from 0.979 to 2.53 pCi/g with an average of 1.65 pCi/g.

5.2.4 Ambient Air

Ambient air was sampled at the fence line of each of the nine selected landfills and analyzed for Rn concentration. A combination of EIC and ATD monitors were used. Because it was impractical to place monitors on the actual working face of the landfill, monitors were deployed at the fence line around the landfill in roughly the four cardinal directions. The exact locations of the monitors are depicted in **Appendix E**. Duplicate monitors were placed at each location, inside a single Tyvek® bag. The Tyvek® bag is permeable to Rn gas, but impermeable to particulate matter. The monitors were hung on the fence line approximately 5 ft above grade. Deployment of the Rn monitors ranged from 74 to 103 days. Monitor device selection was based upon availability at the time of deployment. The results are presented in **Table 5-8**. Radon activity ranged from 0.200 to 0.900 pCi/L. The Rn monitor analytical reports are presented in **Appendix H**.

5.2.5 Surveys

Radiological surveys were conducted at each of the nine selected landfills, resulting in four data sets:

- *Removable* α/β *surface radioactivity* measurements recorded in units of dpm/100 cm^2
- *Total* α/β *surface radioactivity* measurements recorded in units of dpm/100 cm^2
- *Gross Gamma Radiation Scan* measurements recorded in units of cpm
- *Gamma Radiation Exposure Rate* measurements recorded in units of μR/hr

5.2.5.1 Removable Alpha/Beta Surface Radioactivity Measurement Results

Measurements of removable α/β surface radioactivity were performed to assess potential internal radiation exposures to workers through ingestion and/or inhalation. The results were evaluated using the RG 1.86 guidelines, Table 1. RG 1.86 requires that α and β radioactivity levels be evaluated separately. The primary α emitter of concern is Ra-226, with a removable criterion of 20 dpm α/100 cm^2. The primary β emitter of concern is Ra-228 of the natural Th decay series, with a removable criterion of 200 dpm β/100 cm^2. The average removable α and β levels at each landfill were below the RG 1.86 criteria. The maximum removable α and β levels were also below the RG 1.86 criteria. The results of removable α and β surface radioactivity for the subject landfills surveyed are presented in **Table 5-9.** Individual removable α and β surface radioactivity measurement results are presented in **Appendix D**.

5.2.5.2 Total Alpha/Beta Surface Radioactivity Measurement Results

Measurements of total α/β surface radioactivity were performed to assess potential internal radiation exposures to workers through ingestion and/or inhalation. The results were evaluated using the RG 1.86 guidelines, Table 1. RG 1.86 requires that α and β levels be evaluated separately. The primary α emitter of concern is Ra-226, with a total criterion of 100 dpm α/ 100 cm^2. The primary β emitter of concern is Ra-228 of the natural Th decay series, with a total criterion of 1,000 dpm β/100 cm^2. All average total α and β surface radioactivity levels were below the RG 1.86 criteria. The maximum total α and β concentrations were 84.6 dpm/100 cm^2 and 3,630 dpm/100 cm^2. The summary results of total α and β surface radioactivity for the nine selected landfills surveyed are presented in **Table 5-10**. Individual total α and β surface radioactivity measurement results are presented in **Appendix D**.

5.2.5.3 Gross Gamma Radiation Scan Results

Gross gamma radiation scans, recorded in cpm, were performed on open land areas and accessible areas of the nine selected landfills to identify areas with gamma radiation levels above local background. Summary results for the selected landfills are presented in **Table 5-11**. The highest average count rate at any of the nine selected landfills was 10,816 cpm, and the maximum count rate recorded at any of the nine selected landfills was 74,928 cpm. A graphic display of the gamma scan results at each facility was prepared using GIS software and is presented in **Appendix E**.

5.2.5.4 Gamma Exposure Rate Results Summary

Gross gamma scan results in units of cpm presented in **Table 5-11** were converted to μR/hr by using 800 cpm per μR/hr, a conversion factor appropriate for Ra-226 gamma energy as detected with 2-inch by 2-inch NaI detectors, rounded to one significant figure (Table 6.4, NaI Scintillation Detector Scan MDCs for Common Radiological Contaminants, NUREG-1507, Minimum Detectable Concentrations With Typical Radiation Survey Instruments for Various Contaminants and Field Conditions, USNRC June 1998). **Table 5-12** presents statistical results for each of the nine selected landfills. The highest average exposure rate was 13.5 μR/hr, and the maximum gamma exposure rate measured was 93.7 μR/hr.

5.3 Radon Ingrowth Within Filter Cake From WWTP to Landfills

Radon in filter cake is the result of the decay of Ra, which is referred to as ingrowth. Radium-226 from the U series and Ra-228 from the Th series are present in flowback and produced water. Radioactive precursors to Ra (U-238 and Th-232) are not present due to their relative insolubility. When these wastewaters are processed at WWTPs, the Ra is removed and concentrated in the resulting filter cake or sludge.

During handling and/or transport, the sludge or filter cake may be disturbed and some of the Rn gas may escape, greatly reducing the gamma-emitting progeny that follow Rn-222 in the natural decay series. Using the software program MicroShield®, the following source terms were evaluated to determine the resulting gamma exposure rate measured 6 inches from the outside of a standard roll-off container filled with sludge at a concentration of 13.4 pCi/g of Ra-226. The source terms assume that all of the Rn and progeny are removed at day zero. Ingrowth of Rn and

progeny was calculated for each time period in accordance with half-lives to determine the subsequent source terms, as follows:

a. 0-day ingrowth (13.4 pCi/g of Ra-226 only)
b. 1-day ingrowth (13.4 pCi/g of Ra-226 + 16 percent progeny)
c. 3-day ingrowth (13.4 pCi/g of Ra-226 + 41 percent progeny)
d. 10-day ingrowth (13.4 pCi/g of Ra-226 + 86 percent progeny)
e. 21-day ingrowth (13.4 pCi/g of Ra-226 + 100 percent progeny)

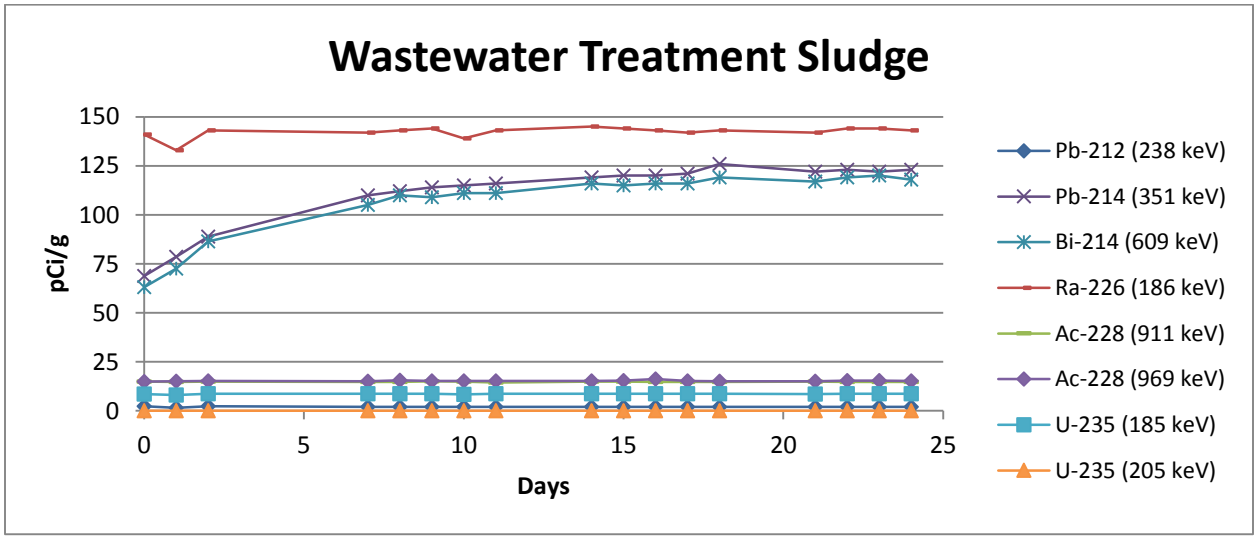
The results of the MicroShield® modeling are presented in **Figure 5-1**. The exposure rate increased rapidly to approximately 21 days post ingrowth, at which time the maximum exposure rate was achieved. Starting from zero Rn progeny to full equilibrium after 21 days, the exposure rate measured 6 inches from the outside of the roll-off container increased six-fold. Based on the MicroShield® modeling results, there may be an increase of six times the gamma exposure rate measured 6 inches from the surface of the roll-off container during the first 21 days after a wastewater treatment sludge is generated. This is a theoretical curve and assumes all of the Rn is removed when the sludge is formed at time zero.

Figure 5-1. Ra-226 Progeny Ingrowth (Days Post Removal) versus Exposure Rate from 13.34 pCi/g Ra-226

To further evaluate the Rn and short-lived progeny ingrowth in wastewater sludge, a series of recently generated sludge samples were collected at six WWTPs and analyzed using gamma spectroscopy. The samples were analyzed when received and then 15 additional times over the next 24 days. The activity results versus time, post sample, were plotted. Radon ingrowth is demonstrated in each set of sample results. **Figure 5-2** and **Table 5-13** present the data from one of the sludge samples. The following was observed:

- The Pb-214 and Bi-214, short-lived progeny of Rn-222, increased from approximately 50 percent of the Ra-226 activity in the sample to 85 percent of the Ra-226 activity. Radium-226 was identified directly from the 186 keV gamma line. The average of the Pb-214 and Bi-214 results was 69.6 pCi/g at day zero and 120 pCi/g at day 24 compared to the Ra-226 activity of 142 pCi/g each day.
- Radon gas progeny were present at 50 percent of the Ra-226 activity in the recently generated sludge. Only 50 percent of the Rn gas escapes the sludge during processing.
- The Rn gas only increased to 85 percent of the Ra-226 parent activity in three weeks. This could be due to leakage of Rn through the sample container seal.
- The reported U-235 activity (185.7 keV gamma line) was consistently measured at 8.64 pCi/g, matching the theoretical overestimation of 8.7 pCi/g of U-235 based on 142 pCi/g of Ra-226. See Section 2.3.2 and Table 2-1 for a detailed discussion of Ra-226 and U-235 identification and potential overestimation using gamma spectroscopy. The U-235 identified by the 205 keV line was consistently 0 pCi/g.

Figure 5-2. Ra-226 Progeny Ingrowth versus Days (Days Post Removal)



5.4 Landfill Worker Exposure Assessment

5.4.1 Landfill External Radiation Exposure

The maximum average gamma radiation exposure rate measured at any of the nine selected landfills was 13.5 µR/hr. The minimum, limiting local background was 5 µR/hr. Assuming the duration of exposure is a full occupational year of 2,000 hours, the external gamma radiation exposure at the landfill was estimated as follows:

Maximum Average Landfill External Gamma Exposure Estimate

(13.5 – 5) µR/hr x 2,000 hr/yr x (1 mrem/1,000 µR gamma) = 17 mrem/yr

This is an estimate of the maximum average exposure based on 2,000 hours in one year. The result is less than the 100 mrem/yr dose equivalent limit for a member of the public. Actual exposure is dependent upon the actual exposure rates and occupancy time for individual workers.

The maximum exposure rate measured at any of the nine selected landfills was 93.7 μR/hr. Work in this area would result in an exposure of 100 mrem in 1,130 hours of annual exposure of an employee's 2,000-hour occupational year. Actual annual exposure for a landfill worker is dependent upon actual exposure rates and actual time worked in the proximity of the tank.

5.4.1.1 Landfill Worker Potential Internal Alpha/Beta Radioactivity Exposure

The total and removable α/β survey results are presented in Sections 5.2.5.1 and 5.2.5.2. None of the 195 α measurements and 17 of the 195 β measurements of total surface radioactivity exceeded the RG 1.86 criteria. None of the 205 removable α or β surface radioactivity measurements exceeded the RG 1.86 criteria. The average values for total and removable α and β surface radioactivity are below the RG 1.86 criteria, indicating that there is little potential for internal α and β exposure to landfill workers.

5.4.1.2 Landfill Worker Internal Radon Exposure

The results of the landfill ambient air Rn samples are presented in Section 5.2.4. The Rn in ambient air at the fence line of the landfills ranged from 0.200 to 0.900 pCi/L consistent with U.S. background levels of 0.00 – 1.11 pCi/L in outdoor ambient air. Consequently, the potential for internal Rn exposure is low.

Table 5-1. Landfill Leachate – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/l)	Ra-228 (pCi/l)	K-40 (pCi/l)	Gross Alpha (pCi/l)	Gross Beta (pCi/l)
LF-10-LQ-024	322	< 20.0	201	< 140	< 192
LF-11-LQ-025	109	13.0	485	< 145	491
LF-12-LQ-026	102	< 6.00	558	< 129	440
LF-13-LQ-027	81.0	< 11.0	369	< 155	284
LF-14-LQ-028	101	19.0	1,110	< 167	1,110
LF-15-LQ-029	121	< 10.0	1,060	< 163	1,020
LF-16-LQ-030	114	< 7.00	122	< 136	< 191
LF-17-LQ-031	342	< 21.0	524	< 126	489
LF-18-LQ-032	120	< 25.0	764	< 161	703
LF-19-LQ-033	159	< 105	1,040	< 193	1,200
LF-20-LQ-034	< 130	< 110	615	182	806
LF-21-LQ-035	< 87.0	< 10.0	670	< 162	850
LF-22-LQ-036	< 77.0	< 13.0	332	< 156	531
LF-23-LQ-037	< 148	< 26.0	268	< 306	489
LF-24-LQ-038	145	< 15.0	477	< 134	489
LF-25-LQ-039	79.0	< 12.0	175	< 118	< 199
LF-26-LQ-040	< 146	< 31.0	268	< 134	< 190
LF-27-LQ-041	< 108	< 22.0	148	< 205	< 203
LF-28-LQ-042	< 89.0	< 16.0	64.0	< 277	< 221
LF-29-LQ-043	416	< 19.0	181	< 119	< 200
LF-30-LQ-044	84.0	< 6.00	551	< 342	412
LF-31-LQ-045	150	< 9.00	282	< 206	< 203
LF-32-LQ-046	112	< 21.0	127	< 125	< 189
LF-33-LQ-047	< 153	< 37.0	573	< 146	667
LF-34-LQ-048	< 111	< 21.0	423	< 157	401
LF-35-LQ-049	136	< 19.0	758	< 254	728
LF-36-LQ-050	106	22.0	471	< 353	466
LF-37-LQ-051	73.0	19.0	503	< 341	845
LF-38-LQ-052	54.0	< 5.00	249	< 152	550
LF-39-LQ-053	< 82.0	< 18.0	222	< 149	< 194
LF-40-LQ-054	91.0	35.0	505	< 143	239
LF-41-LQ-055	65.0	9.00	383	< 164	286
LF-42-LQ-056	148	< 16.0	< 54.0	< 137	384
LF-43-LQ-057	371	< 8.00	110	< 128	< 199
LF-44-LQ-058	101	< 12.0	629	< 206	365
LF-45-LQ-059	< 73.0	< 14.0	480	< 111	< 208
LF-46-LQ-060	140	15.0	354	< 486	< 416
LF-47-LQ-061	70.0	13.0	131	< 121	< 202
LF-48-LQ-062	57.0	< 5.00	354	< 181	284
LF-49-LQ-063	126	< 9.00	209	< 316	< 232
LF-50-LQ-064	85.0	< 10.0	128	< 112	< 201
LF-51-LQ-065	106	9.00	49.0	< 113	< 202

Table 5-1. Landfill Leachate – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/l)	Ra-228 (pCi/l)	K-40 (pCi/l)	Gross Alpha (pCi/l)	Gross Beta (pCi/l)
Average	116	11.9	404	94.4	389
Std. Dev.	88.0	11.4	272	43.6	311
Median	96.0	9.00	362	77.8	326
Minimum	36.5	2.50	27.0	112	94.5
Maximum	416	55.0	1,110	243	1,200

< – indicates a value less than the reported number which is the MDC.

Table 5-2. Selected Landfill Leachate – Gamma Spectroscopy and Miscellaneous Results

Study ID	Ra-226 (pCi/l)	Ra-228 (pCi/l)	K-40 (pCi/l)	Gross Alpha (pCi/l)	Gross Beta (pCi/l)
LF-01-LQ-002	378	< 20.0	< 72.0	< 3.46	< 2.07
LF-02-LQ-003	136	84.0	637	< 110	295
LF-03-LQ-008	140	16.0	221	< 275	< 202
LF-04-LQ-009	118	< 6.00	64.0	< 253	< 395
LF-05-LQ-023	115	< 20.0	182	< 323	< 233
LF-06-LQ-010	85.0	< 8.00	351	< 160	259
LF-07-LQ-004	< 134	< 35.0	353	< 121	221
LF-08-LQ-017	70.0	9.00	743	< 357	280
LF-09-LQ-005	105	< 8.00	155	< 314	< 233
Average	125	18.0	305	106	176
Std. Dev.	98.1	25.0	245	59.8	98.5
Median	85.0	10.0	221	127	198
Minimum	67.0	3.00	36.0	1.73	1.04
Maximum	378	84.0	743	357	395

< – indicates a value less than the reported number which is the MDC.

Table 5-3. Landfill Leachate Original and Aqueous Sample Analysis Results

Study ID	Original Gamma Spec – Unfiltered Sample			Re-Analysis Using EPA 903.1/904.0 Technique – Aqueous Phase Sample Only					
	Ra226 Result (pCi/L)	Ra226 Error (pCi/L)	Ra226 MDC (pCi/L)	Ra226 Result (pCi/L)	Ra226 Error (pCi/L)	Ra226 MDC (pCi/L)	Ra228 Result (pCi/L)	Ra228 Error (pCi/L)	Ra228 MDC (pCi/L)
LF-17-LQ-031	342	92.0	131	10.3	0.294	0.063	7.82	1.02	0.956
LF-24-LQ-038	145	60.0	91.0	1.91	0.107	0.032	4.27	1.06	1.33
LF-13-LQ-027	81.0	33.0	51.0	1.70	0.103	0.021	2.20	0.806	1.08
LF-45-LQ-059	47.0	45.0	73.0	0.472	0.085	0.090	0.896	0.662	0.998
LF-18-LQ-032	120	73.0	115	6.01	0.218	0.073	5.77	0.946	0.966
LF-10-LQ-024	322	85.0	121	1.22	0.089	0.057	1.41	0.770	1.13
LF-08-LQ-017	70.0	29.0	47.0	0.414	0.067	0.068	1.06	0.732	1.09
LF-12-LQ-026	102	40.0	62.0	0.842	0.086	0.069	2.55	0.771	1.00
LF-01-LQ-002	378	96.0	132	0.066	0.027	0.030	0.643	0.664	1.04
LF-04-LQ-009	118	35.0	53.0	0.124	0.031	0.017	0.976	0.717	1.08

Table 5-4. Selected Landfill Effluent Leachate – Gamma Spectroscopy and Miscellaneous Results

Source of Sample	Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha[a] (pCi/L)	Gross Beta[a] (pCi/L)
Effluent	LF-01-LQ-002	378	< 20.0	< 72.0	< 3.46	< 2.07
Effluent	LF-02-LQ-003	136	84.0	637	< 110	304
Effluent	LF-03-LQ-008	< 140	16.0	221	< 275	< 202
Effluent	LF-04-LQ-009	118	< 6.00	64.0	< 253	< 395
Effluent	LF-07-LQ-004	< 134	< 35.0	353	< 121	221
Effluent	LF-09-LQ-005	105	1,100	18,100	< 314	< 233
Effluent	LF-09-LQ-021	117	15.0	165	ND	ND
	Average	142	178	2,800	89.7	157
	Std. Dev.	107	408	6,750	59.9	106
	Median	117	16.0	221	93.5	157
	Minimum	67.0	3.00	36.0	1.73	1.04
	Maximum	378	1,100	18,100	157	304

[a]ND – Sample Matrix was not suitable for analysis.
< – indicates a value less than the reported number which is the MDC.

Table 5-5. Selected Landfill Influent Leachate – Gamma Spectroscopy and Miscellaneous Results

Source of Sample	Study ID	Ra-226 (pCi/L)	Ra-228 (pCi/L)	K-40 (pCi/L)	Gross Alpha (pCi/L)	Gross Beta (pCi/L)
Influent	LF-01-LQ-019	< 139	< 21.0	236	< 18.3	117
Influent	LF-02-LQ-020	< 120	15.0	755	< 201	524
Influent	LF-03-LQ-015	116	< 14.0	246	< 168	< 203
Influent	LF-04-LQ-016	92.0	< 15.0	571	< 134	416
Influent	LF-05-LQ-023	115	< 20.0	182	< 323	< 233
Influent	LF-06-LQ-010	85.0	< 8.00	351	< 160	259
Influent	LF-07-LQ-011	< 97	< 8.00	278	< 200	< 200
Influent	LF-08-LQ-017	70	9.00	743	< 357	280
Influent	LF-09-LQ-012	95	< 9.00	242	< 195	< 200
	Average	83.4	7.94	400	97.6	224
	Std. Dev.	23.5	3.64	227	49.9	158
	Median	85.0	7.50	278	97.5	117
	Minimum	48.5	4.00	182	9.15	100
	Maximum	116	15.0	755	179	524

< – indicates a value less than the reported number which is the MDC.

Table 5-6. Selected Landfill Solids, Filter Cake – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)
LF-02-SL-002	8.73	4.98	4.83
LF-03-SL-004	53.0	5.03	2.72
LF-04-SL-005	11.1	1.53	2.73
Average	24.3	3.85	3.43
Std. Dev.	24.9	2.01	1.22
Median	11.1	4.98	2.73
Minimum	8.73	1.53	2.72
Maximum	53.0	5.03	4.83

Table 5-7. Selected Landfill Solids, Sediment – Gamma Spectroscopy Results

Study ID	Ra-226 (pCi/g)	Ra-228 (pCi/g)	K-40 (pCi/g)	U-238 (pCi/g)	U-235 (pCi/g)	Th-232 (pCi/g)
LF-01-SL-001	4.46	2.53	15.2	< 2.51	0.177	2.48
LF-02-SL-003	2.82	1.44	12.8	< 0.671	< 0.069	1.41
LF-04-SL-006	3.44	0.979	10.0	< 0.868	< 0.128	0.960
Average	3.57	1.65	12.7	0.675	0.092	1.62
Std. Dev.	0.828	0.796	2.60	0.505	0.075	0.781
Median	3.44	1.44	12.8	0.434	0.064	1.41
Minimum	2.82	0.979	10.0	0.336	0.035	0.960
Maximum	4.46	2.53	15.2	1.26	0.177	2.48

< – indicates a value less than the reported number which is the MDC.

Table 5-8. Selected Landfill Radon Concentrations

Study ID	County	Location	Exp. End Date	Radon Concentration S.D. (pCi/L)	Error (+/- 2 Std. Dev.) (pCi/L)[b]	MDC (pCi/L)
LF-01-RA	McKean	01	1/2014	0.200	0.200	0.200
		02	1/2014	0.400	0.200	0.200
		03	1/2014	0.300	0.200	0.200
		04	1/2014	0.400	0.200	0.200
LF-02-RA	Elk	01	6/2014	0.200	0.200	0.200
		02	6/2014	0.300	0.200	0.200
		03	6/2014	Missing		
		04	6/2014	0.400	0.200	0.200
LF-03-RA	Butler	01	6/2014	0.300	0.200	0.200
		02	6/2014	0.500	0.200	0.200
		03	6/2014	0.900	0.200	0.200
		04	6/2014	0.400	0.200	0.200
LF-04-RA	Butler	01	6/2014	0.300	0.200	0.200
		02	6/2014	0.700	0.200	0.200
		03	6/2014	0.500	0.200	0.200
		04	6/2014	0.400	0.200	0.200
LF-05-RA	Fayette[a]	01	7/2014	< 0.400	NA	0.400
		02	7/2014	< 0.400	NA	0.400
		03	7/2014	< 0.400	NA	0.400
		04	7/2014	< 0.400	NA	0.400
LF-06-RA	Fayette[a]	01	7/2014	< 0.400	NA	0.400
		02	7/2014	< 0.400	NA	0.400
		03	7/2014	< 0.400	NA	0.400
		04	7/2014	< 0.400	NA	0.400
LF-07-RA	Washington[a]	01	7/2014	< 0.400	NA	0.400
		02	7/2014	< 0.400	NA	0.400
		03	7/2014	< 0.400	NA	0.400
		04	7/2014	< 0.400	NA	0.400
LF-08-RA	Somerset[a]	01	7/2014	< 0.400	NA	0.400
		02	7/2014	< 0.400	NA	0.400
		03	7/2014	< 0.400	NA	0.400
		04	7/2014	< 0.400	NA	0.400
LF-09-RA	Cambria[a]	01	7/2014	< 0.400	NA	0.400
		02	7/2014	< 0.400	NA	0.400
		03	7/2014	< 0.400	NA	0.400
		04	7/2014	< 0.400	NA	0.400

The ATD laboratory does not report an error term on devices with results below their MDC.
[a] Represents landfills with ATDs deployed.
[b] An error presented as NA represents a result that was less than the reported MDC.

Table 5-9. Selected Landfill Removable Alpha and Beta Surface Radioactivity Results Summary

Study ID	No. of Data Points	Removable Alpha (dpm/100 cm^2)				Removable Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
LF-01-FS-073	31	4.25	11.3	1.27	4.48	65.0	65.0	0.000	65.0
LF-03-FS-076	27	5.80	5.80	0.000	5.80	63.0	63.0	0.000	63.0
LF-05-FS-050	27	8.30	8.30	0.000	8.30	64.0	64.0	0.000	64.0
LF-08-FS-070	19	5.80	5.80	0.000	5.80	63.0	63.0	0.000	63.0
LF-02-FS-135	30	4.25	4.25	0.000	4.25	65.0	65.0	0.000	65.0
LF-04-FS-132	23	7.30	7.30	0.000	7.30	63.0	63.0	0.000	63.0
LF-06-FS-131	10	5.80	5.80	0.000	5.80	63.0	63.0	0.000	63.0
LF-09-FS-133	30	7.30	7.30	0.000	7.30	63.0	63.0	0.000	63.0
LF-07-FS-134	10	7.30	7.30	0.000	7.30	63.0	63.0	0.000	63.0

Note: During the calculations to convert from raw counts to dpm, the calculated value was compared to half of the MDC. If the value was below this number, half of the MDC was inserted into the tables. Where the standard deviation is zero and the minimum, maximum, and average are the same, then all measurements were below half of the MDC.

Table 5-10. Selected Landfill Total Alpha and Beta Surface Radioactivity Results Summary

Study ID	No. of Data Points	Total Alpha (dpm/100 cm^2)				Total Beta (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average	Minimum	Maximum	Standard Deviation	Average
LF-01-FS-073	26	7.45	39.8	9.42	13.0	301	779	112	332
LF-03-FS-076	28	7.45	84.6	21.7	18.1	288	3,630	642	682
LF-05-FS-050	27	7.45	29.8	6.84	11.4	285	942	221	410
LF-08-FS-070	19	7.45	24.9	5.08	9.81	268	1,900	524	580
LF-02-FS-135	22	18.6	38.9	4.61	20.3	288	1,270	356	692
LF-04-FS-132	22	7.50	69.6	17.5	13.3	274	1,560	371	646
LF-06-FS-131	10	7.45	49.7	13.5	14.9	289	766	194	381
LF-09-FS-133	30	7.45	19.9	5.00	10.8	272	1,360	250	401
LF-07-FS-134	11	7.45	19.9	4.45	9.94	468	1,960	578	730

Note: During the calculations to convert from raw counts to dpm, the calculated value was compared to half of the MDC. If the value was below this number, half of the MDC was inserted into the tables. Where the standard deviation is zero and the minimum, maximum, and average are the same, then all measurements were below half of the MDC.

Table 5-11. Selected Landfill Gross Gamma Radiation Scan Results Summary

Site	GWS Max[a] (cpm)	GWS Min[a] (cpm)	GWS Average[a] (cpm)	GWS Std Dev (cpm)	No. Data Points
LF-01	74,928	3,837	9,250	1,656	9,210
LF-02	16,737	3,299	9,097	2,954	13,977
LF-03	13,900	5,141	8,022	1,713	11,484
LF-04	16,545	5,272	10,742	2,807	8,691
LF-05	14,730	3,783	8,190	2,658	8,942
LF-06	10,994	5,118	7,649	902	9,129
LF-07	11,620	4,530	7,190	1,260	5,432
LF-08	18,894	3,466	6,573	1,909	10,977
LF-09	27,144	4,304	10,816	2,914	9,779

[a]Convert count rate data to exposure rate by dividing count rate by 800 to yield µR/hr.

Table 5-12. Results Summary of NaI Count Rate Data Converted to Exposure Rates

Site	GWS Max (µrem/hr)	GWS Min (µrem/hr)	GWS Average (µrem/hr)	GWS Std Dev (µrem/hr)	No. Data Points
LF-01	93.7	4.80	11.6	2.07	9,210
LF-02	20.9	4.12	11.4	3.69	13,977
LF-03	17.4	6.43	10.0	2.14	11,484
LF-04	20.7	6.59	13.4	3.51	8,691
LF-05	18.4	4.73	10.2	3.32	8,942
LF-06	13.7	6.40	9.56	1.13	9,129
LF-07	14.5	5.66	8.99	1.58	5,432
LF-08	23.6	4.33	8.22	2.39	10,977
LF-09	33.9	5.38	13.5	3.64	9,779

Table 5-13. Gamma Spectroscopy Results (pCi/g) of Sealed Wastewater Treatment Sludge Sample Over 24 Days

Time (days)	Pb-212 238 keV	Pb-212 300 keV	Pb-214 295 keV	Pb-214 351 keV	Bi-214 609 keV	Bi-214 1,120 keV	Bi-214 1,764 keV	Ra-226 186 keV	Ac-228 911 keV	Ac-228 969 KeV	U-235 184 KeV
T00	2.40	2.11	68.1	68.8	63.0	70.4	77.8	141	14.9	15.0	8.58
T01	1.55	#NA	77.3	78.6	72.5	81.0	90.1	133	14.8	15.1	8.09
T02	2.38	2.52	87.7	88.8	86.5	91.7	78.3	143	14.9	15.3	8.67
T07	2.07	2.21	108	110	105	114	95.5	142	14.8	15.1	8.64
T08	2.01	1.96	110	112	110	117	98.5	143	14.8	15.6	8.71
T09	2.04	2.38	112	114	109	119	100	144	15.1	15.3	8.73
T10	1.99	1.99	113	115	111	120	102	139	14.9	15.3	8.44
T11	1.99	2.21	114	116	111	120	102	143	14.5	15.3	8.68
T14	2.00	2.01	117	119	116	124	105	145	15.0	15.3	8.78
T15	1.98	NA	119	120	115	125	106	144	14.9	15.4	8.72
T16	2.01	1.58	118	120	116	124	106	143	14.8	16.3	8.68
T17	1.98	2.45	119	121	116	125	106	142	14.8	15.2	8.64
T18	1.98	2.03	119	126	119	126	108	143	14.7	15.1	8.69
T21	1.98	1.82	120	122	117	126	108	142	15.0	15.1	8.60
T22	2.05	1.59	120	123	119	128	108	144	14.8	15.4	8.75
T23	2.04	2.02	120	122	120	128	108	144	14.7	15.4	8.72
T24	2.07	1.76	121	123	118	127	109	143	14.8	15.3	8.71

#NA – indicates the analyte was not requested and subsequently not reported by the laboratory.

6.0 GAS DISTRIBUTION AND END USE

Uranium-238 is distributed throughout the crust of the earth, typically at concentrations of 0.33 to 1.0 pCi/g. However, concentrations can be much higher in certain rock types or formations. The U-238 decay series consists of 18 decay progeny, including Rn. Radon is the only member of the decay series that is a gas at typical ambient conditions. All of the other decay series members are solids. Because Rn is a gas, it is highly mobile within the soil and rock matrix and it easily enters into structures. There are two additional potential pathways for Rn entry into structures: well water and natural gas combustion, e.g., cooking and unvented heating. Natural gas samples were collected at underground storage sites, natural gas-fired power plants, gas compression and transmission facilities, and natural gas processing plants.

6.1 Natural Gas in Underground Storage

Natural gas samples were collected at four underground storage sites in Pennsylvania. Duplicate samples were collected at each site during injection into the storage formation and during withdrawal from the storage formation. Sampling during injection was conducted during the period of May to August 2013. Sampling during withdrawal was conducted during the period of January to early February 2014. At three of the sites the samples were obtained from the exhaust of the gas chromatograph, which continuously analyzes the natural gas. At the fourth site, the sample was collected from the injection flow dehydration unit. The results for injection sampling are presented in **Table 6-1**. The results for withdrawal sampling are presented in **Table 6-2**. The results indicate Rn concentrations are lower after underground storage. The Rn analytical reports are presented in **Appendix H**.

6.2 Natural Gas-Fired Power Plants

Two natural gas-fired power plants (PP-01 and PP-02) were surveyed for gamma radiation exposure rates. Natural gas samples were collected at both plants, and ambient Rn measurements were performed at the PP-02 fence line. The natural gas Rn concentration results are presented in **Table 6-3**, and the ambient Rn concentrations measured at the plant fence line are presented in **Table 6-4**. All of the Rn analytical reports are presented in **Appendix H**.

The gamma radiation exposure rate survey at the PP-02 power plant was conducted using a Ludlum Model 19 Micro-R Meter. With the exception of one area, the range of measurement results observed were 5-10 µR/hr, which is within the range of natural background of gamma radiation for Pennsylvania. The exception occurred on the external surface of a pipe elbow where the range of measurement results observed were 15-17 µR/hr. During a subsequent survey event, the measurement results observed at the surface of that pipe elbow were 5-10 µR/hr, which is within the range of natural background of gamma radiation levels.

Ambient air was sampled at the PP-02 power plant site fence line. Eight EIC passive Rn monitors were used. The monitors were deployed at the fence line around the power plant in roughly the four cardinal directions. See figures in **Appendix E** for exact locations. The monitors were placed, in duplicate, inside a single Tyvek® bag. The Tyvek® bag is permeable to Rn gas but impermeable to particulate matter. The monitors were hung on the fence line approximately 5 ft above grade. Deployment of the Rn monitors was for 64 days. The fence line Rn monitor results were all at or

below the MDC value for the analysis. The results are presented in **Table 6-4**. The Rn analytical reports are presented in **Appendix H**.

6.3 Compressor Stations

Duplicate natural gas samples were collected at intake flow lines of both facility CS-01 and CS-03. Duplicate samples were collected at the compressor station discharge at facility CS-04. The CS-04 compressor station is associated with the natural gas processing plant (CP-01) discussed below. Because of high pressure in the intake flow lines, duplicate natural gas samples were collected at the continuous natural gas quality analyzer at CS-02. This sample point is a small line off of a main exhaust for CS-02. All compressor stations were receiving predominately Marcellus Shale unconventional natural gas at the time of sample collection. Radon-measured concentrations are presented in **Table 6-5**. The compressor station natural gas Rn results are consistent with the production site Rn sample results. The Rn analytical reports are presented in **Appendix H**.

Ambient air was sampled at the CS-01 compressor station fence line for the measurement of Rn concentrations. Eight EIC passive Rn monitors were used. The monitors were deployed at the fence line around the power plant in roughly the four cardinal directions. See figures in **Appendix E** for exact locations. The monitors were placed, in duplicate, inside a single Tyvek® bag. The Tyvek® bag is permeable to Rn gas but impermeable to particulate matter. The monitors were hung on the fence line approximately 5 ft above grade. Deployment of the Rn monitors was for 62 days. The fence line Rn monitor results ranged from 0.100 to 0.800 pCi/L. The average concentration at each fence line location was within the range of typical ambient background Rn concentrations in outdoor ambient air in the U.S., i.e., 0.00 to 1.11 pCi/L. The results are presented in **Table 6-6**. The Rn analytical reports are presented in **Appendix H**.

6.4 Natural Gas Processing Plant

Two natural gas samples were collected at the processing plant (CP-01) on two separate occasions: March 12, 2014 and September 11, 2014. The results are presented in **Table 6-7**. The Rn analytical reports are presented in **Appendix H**.

Gamma radiation exposure rate surveys were performed during the two site visits. The exposure rate surveys were performed using a Ludlum Model 19 Micro-R Meter. The first survey was performed on a rainy, windy day, limiting the outdoor areas surveyed. The results include:

- Background in areas not impacted by the plant – 5-10 μR/hr.
- General areas of the plant – 5-10 μR/hr.
- Filter housings (exposure rate measured on the outside surface):
  - Contact readings measured on contact with filter housings ranged from background to 75 μR/hr, with two exceptions; one measured 350 μR/hr and the other measured 900 μR/hr.
- Propane processing – radiation exposure rates measured up to 380 μR/hr on contact with heat exchangers, reboilers, pipelines, and pumps.
- Propane storage area:
  - Pipeline exposure rates measured from local background to 400 μR/hr on contact.
  - Ladder to decking area measured 80 μR/hr general area.
  - Decking above ladder measured 50 μR/hr general area.

  - Propane storage tank measured 210 μR/hr on contact.
- Propane tank trailer being filled – 100 μR/hr on contact with the tank.
- Rail yard:
  - Tank filling area – local background to 20 μR/hr general area.
  - Racks of filling pipes – local background to 100 μR/hr on contact.
  - Propane rail car tank – 30 μR/hr on contact.

Radon in natural gas sample results are presented in **Table 6-7**. The highest concentration of Rn, 71.1 pCi/L, was measured in natural gas entering the processing plant. The lowest concentration of Rn, 8.60 pCi/L, was measured in natural gas at the processing plant outflow. The Rn analytical reports are presented in **Appendix H**.

A second visit to the facility was made to survey and sample filter media. The filter housing with the highest exposure rate measured was selected for sampling and gamma spectroscopy analysis. The outside of the filter housing measured 50 μR/hr. The general radiation exposure rate in the area of the filters was 15 μR/hr. The filter housing on the facility propanizer equipment was opened during a filter change-out and a sample of the cardboard filter media was collected. The filter media sample was smeared for removable α and β surface radioactivity. Smear samples of removable α and β surface radioactivity were taken on each of the individual filter cases housing the filter media within the filter bank. The gross α and β removable surface radioactivity results summary statistics of the 11 smear sample counts from the filter case are presented in **Table 6-8**. The average α and β surface radioactivity levels are below the RG 1.86 α and β removable surface radioactivity criterion.

The results of the filter gamma spectrometry analysis are presented in **Table 6-9**. A Pb-210 activity result of 3,580 pCi/g was identified, but no other gamma-emitting NORM radionuclide results were above 1 pCi/g. The gross α and β removable surface radioactivity results for the filter media sample are presented in **Table 6-10**. The results are elevated relative to the RG 1.86 gross α and β removable surface radioactivity criterion.

6.5 Potential Exposure from Gas Scale Inside Pipes and Equipment

Materials deposited on interior surfaces of natural gas plant pipes and equipment are different from conventional oil industry Ra-based pipe scale. Natural gas plant scale typically consists of Rn decay progeny that accumulate on the interior surfaces of plant pipes and equipment without the long-lived Ra parent.

As a result, the only radionuclides that remain and adhere to the interior surfaces of machinery/pipes are the Rn decay progeny Po-210 and Pb-210. These longer-lived decay progeny are not readily detected on the outside of pipes. However, Pb-210 and Po-210 emit α and β radioactive particles that may be a potential inhalation or ingestion hazard when pipes and machinery are opened for maintenance and/or cleaning.

Access to the internal surfaces of pipes and equipment for surveys of surface α and β activity was not available. However, the facility propanizer equipment opened and sampled during filter change-out is representative of interior conditions and was described in Section 6.4. The results are presented in **Table 6-9**. A Pb-210 activity result of 3,580 pCi/g was identified. No other

gamma-emitting NORM radionuclides above 1 pCi/g were identified. The results confirm the build-up of the longer-lived Rn decay progeny in equipment and pipes. The concentration of Pb-210 identified may present a potential inhalation or ingestion hazard during routine system maintenance.

6.6 Radon Dosimetry

Radon exposure in homes due to the use of natural gas appliances is presented in this section. Radon is transported with natural gas into structures (homes, apartments, and buildings) that use natural gas for purposes such as heating and cooking.

The incremental increase of Rn-222 for a typical home was estimated using the values and assumptions presented in **Table 6-11** and as follows:

1. Well Site Rn-222 Concentration in Natural Gas – For the Rn gas concentration, only production site samples from Marcellus Shale well sites were used (n=16). The median value was 43.6 pCi/L, and the maximum value was 148 pCi/L. Both of these values are used in the estimations of potential Rn exposure.

2. Natural Gas/Rn-222 Transit Time and Decay – Assumed there is no Rn decay during transit.

3. Radon-222 Influx Rate – The American Gas Association average natural gas use per day value of 5,465 L/day was used. The value does not consider the types of appliances used. The amount of Rn liberated into the home per hour is calculated using the estimated natural gas use per day (5,465 L/day) and the Rn concentration in that natural gas (43.6 and 148 pCi/L). The resulting values are 238,274 pCi/day for the median concentration and 808,820 pCi/day for the maximum concentration. Dividing each value by 24 hours per day results in 9,928 pCi/hr and 33,700 pCi/hr, respectively. These estimates assume that none of the appliances are vented. Consequently, all of the Rn in the natural gas is assumed to be liberated into the residence.

   Rn-222 Influx Rate = (5,465 L/day x 43.64 pCi/L) / 24 hrs/day = 9,928 pCi/hr

   Rn-222 Influx Rate = (5,465 L/day x 148 pCi/L)/ 24 hrs/day = 33,700 pCi/hr

4. Air Exchange Rate – Using a residence volume of 385,152 L and an air exchange rate of 0.68 air changes per hour, 261,903 L/hr of home air is exchanged with outdoor air.

5. Consistent with EPA Rn assessments, an equilibrium factor of 40 percent is assumed.

6. Indoor Rn-222 Activity Concentration – The Rn-222 influx per hour divided by the home air exchange rate per hour, 9,928 pCi/hr / 261,903 L/hr = **0.04 pCi/L for the median value.** The Rn-222 influx per hour divided by the home air exchange rate per hour, 33,700 pCi/hr / 261,903 L/hr = **0.13 pCi/L for the maximum value.** This is the increase in Rn-222 in the home resulting from natural gas use containing both a median value of 43.6 pCi/L and a maximum value of 148 pCi/L of Rn-222.

The increase in Rn concentration of 0.04 and 0.13 pCi/L along with the standard values presented in **Table 6-11** are used to estimate potential additional annual radiation dose to an exposed individual.

Therefore,

$$\frac{0.04\,pCi/L * 0.4}{100} = 0.00016\,WL$$

The cumulative exposure is then WL multiplied by the number of hours exposed divided by 170 hrs/working month.

$$\frac{0.00016\,WL * 6{,}136\,hrs/yr}{170\,hrs/working\ month} = .006\,WLM/yr$$

This value was converted to a radiation dose by multiplying by the dose conversion factor, the tissue weighting factor, and the radiation weighting factor:

$$0.08 * \frac{0.006\,WLM}{yr} * \frac{0.54\,rad}{WLM} * \frac{20\,rem}{rad} * \frac{1000\,mrem}{rem} = 5.2\,mrem/yr$$

The result is **5.2 mrem/yr for the median dose and 17.8 mrem/yr for the maximum whole body effective dose**.

Based on the Rn and natural gas data collected as part of this study and the conservative assumptions made, the incremental Rn increase in a home using natural gas appliances is estimated to be very small and would not be detectable by commercially available Rn testing devices. The radiation dose received by home residents is a small fraction of the allowable general public dose limit of 100 mrem/yr.

Table 6-1. Natural Gas Underground Storage Radon Concentrations, Injection

Site	County	Formation Geology	Sample Results, pCi/L	Injection Average Concentration (pCi/L)	Error (± 2 Std. Dev.) (pCi/L)	MDC (pCi/L)
US 01	Potter	Oriskany Sandstone	32.6 and 26.7	29.6	8.20	0.200
US 02	Tioga	Oriskany Sandstone	25.7 and 21.2	23.5	6.40	0.200
US 03	Armstrong	Sandstone	20.4 and 20.4	20.4	0.000	0.200
US 04	Fayette	Limestone	20.3 and 21.2	20.8	1.20	0.200

Scintillation Cells
Note: All results adjusted to ambient air by dividing by 1.054, according to Jenkins et. al., Health Physics, Vol. 106, No. 3, March 2014.

Table 6-2. Natural Gas Underground Storage Radon Concentrations, Withdrawal

Site	County	Formation Geology	Sample Results, pCi/L	Withdrawal Average Concentration (pCi/L)	Error (± 2 Std. Dev.) (pCi/L)	MDC (pCi/L)
US 01	Potter	Oriskany Sandstone	4.90 and 5.30	5.10	0.600	0.300
US 02	Tioga	Oriskany Sandstone	10.9 and 9.30	10.1	2.20	0.200
US 03	Armstrong	Sandstone	5.60 and 5.90	5.80	0.400	0.200
US 04	Fayette	Limestone	10.8 and 11.7	11.3	1.20	0.400

Scintillation Cells
Note: All results adjusted to ambient air by dividing by 1.054, according to Jenkins et. al., Health Physics, Vol. 106, No. 3, March 2014.

Table 6-3. Natural Gas-Fired Power Plant Samples Analyzed for Radon Content

Site	County	Gas Source	Radon Concentration (pCi/L)	Error (± 2 Std. Dev.) (pCi/L)	MDC (pCi/L)
PP 01	Fayette	Marcellus Shale	33.7	1.80	1.50
PP 02	Berks	Marcellus Shale	35.7	110	0.200

Table 6-4. Natural Gas-Fired Power Plants Ambient Fence Line Radon Monitors (PP 02)

Location	Radon Concentration (pCi/L)	Error (± 2 Std. Dev.) (pCi/L)	MDC (pCi/L)
West Fence	0.300	0.200	0.200
	0.400	0.200	0.200
North Fence	0.100	0.200	0.200
	0.100	0.200	0.200
East Fence	0.000	0.200	0.200
	0.200	0.200	0.200
South Fence	0.200	0.200	0.200
	0.200	0.200	0.200

Table 6-5. Compressor Station Radon Samples

Site	County	Gas Source	Radon Concentration (pCi/L)	Error (± 2 Std. Dev.) (pCi/L)	MDC (pCi/L)
CS-01-RG	Berks	Marcellus Shale	28.8	1.40	0.200
CS-02-RG	Fayette	Mostly Marcellus Shale	39.8	4.40	0.200
CS-03-RG	Clinton	98% Marcellus Shale	34.0	0.200	0.200
CS-04-RG	Washington	Marcellus Shale	58.1	1.10	0.200

Table 6-6. Compressor Station Ambient Fence Line Radon Monitors (CS 01)

Location	Radon Concentration (pCi/L)	Error (± 2 Std. Dev.) (pCi/L)	MDC (pCi/L)
Northeast Fence	0.500	0.200	0.200
	0.800	0.200	0.200
Southeast Fence	0.300	0.200	0.200
	0.300	0.200	0.200
Northwest Fence	0.300	0.200	0.200
	0.100	0.200	0.200
Southwest Fence	0.300	0.200	0.200
	0.200	0.200	0.200

Table 6-7. Natural Gas Processing Plant Radon Samples

Site	County	Gas Source	Radon Concentration (pCi/L)	Error (± 2 Std. Dev.) (pCi/L)	MDC (pCi/L)
CP-01	Washington	Processing Plant Inflow 1	67.7	1.50	0.200
CP-01	Washington	Processing Plant Inflow 2	71.1	1.60	1.60
CP-01	Washington	Processing Plant Outflow to Transmission Line 1	8.60	0.400	0.300
CP-01	Washington	Processing Plant Outflow to Transmission Line 1	9.30	0.400	0.300

Table 6-8. Compressor Station and Natural Gas Processing Plant Filter Case Removable Radioactivity Results

Study ID	No. of Data Points	Removable Alpha (dpm/100 cm^2)			
		Minimum	Maximum	Standard Deviation	Average
CP-01-FS-136	11	4.70	29.6	8.78	15.5
Study ID	**No. of Data Points**	**Removable Beta (dpm/100 cm^2)**			
		Minimum	**Maximum**	**Standard Deviation**	**Average**
CP-01-FS-136	11	8.25	96.0	23.9	32.2

Table 6-9. Compressor and Natural Gas Processing Plant Filter Media, Gamma Spectroscopy

Nuclide	Result (pCi/g)	Error (pCi/g)	MDC (pCi/g)
Ac-228	0.141	0.053	0.077
Bi-212	0.287	0.000	0.373
Bi-214	0.564	0.082	0.054
K-40	1.30	0.216	0.225
Pb-210	3,580	552	14.2
Pb-212	0.066	0.044	0.071
Pb-214	0.629	0.070	0.076
Ra-226	0.585	0.566	0.926
Ra-228	0.141	0.053	0.077
Th-232	0.125	0.047	0.077
U-235	-0.105	0.000	0.382
U-238	-14.7	0.000	3.15

Table 6-10. Natural Gas Processing Plant Filter Media, Gross Alpha/Gross Beta

Sample	Gross Alpha	Gross Beta
Filter Media	708 ± 15.2 dpm/cm^2	1,910 ± 11.9 dpm/cm^2

Table 6-11. Radon Dosimetry Values for a Typical Home

Parameter	Value	Reference
Median Sq. Feet of House	1,700 ft^2	1
Ceiling Height	8 ft	NA
Air Change Rate	0.68	2
Home Occupancy Factor	70% (6,136 hrs/yr)	3
Average Daily Nat. Gas Use	193 ft^3/day (5,465 L/day)	4
Pipeline Distance	260 miles	5
Avg. pipeline speed (gas)	5 mph	6
Dose Conversion Factor	0.54 rad/WLM	7
Tissue Weighting Factor (Bronchial region)	0.08	7
Rad. Weighting Factor, alpha	20 rem/rad	7
Equilibrium Factor	0.4	3
Lung Cancer Risk per Unit Exposure	5.38E-4 per WLM	3

Table References:

1. U.S. Census, American Housing Survey, 2011, Table C-02-AH.
2. Nazaroff, W.W. and Nero, A.V. Radon and its Decay Products in Indoor Air. John Wiley & Sons, 1988.
3. Pawal, D.J. and Puskin, J.S. EPA Assessment of Risks from Radon in Homes. U.S. EPA, June 2003.
4. American Gas Association, Washington, D.C.
5. National Pipeline Mapping System, User Guide, U.S. DOT, 2011.
6. Spectra Energy Transmission, Personal Communication, May 2014.
7. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), Annex E, 2006.

7.0 OIL AND GAS BRINE-TREATED ROADS

Brine produced from O&G wells and other sources such as brine treatment plants and brine wells is used as a dust suppressant and road stabilizer on unpaved secondary roads in Pennsylvania. The O&G brine used is from conventional formations only. DEP has developed a fact sheet, *Roadspreading of Brine for Dust Control and Road Stabilization*, for use as a guide when utilizing brine on unpaved roads. The fact sheet was developed under the authority of the Clean Streams Law, the Solid Waste Management Act, and Chapters 78 and 101 of DEP's Rules and Regulations (DEP 2013).

For this study, roads in the southwest, northwest, and north-central regions were surveyed and sampled. Most O&G operations occur in these regions. The surveys and sampling included:

- Thirty-two O&G brine-treated roads were surveyed. Thirty-one biased surface samples were collected from the O&G brine-treated roads. The biased locations were selected based on increased instrument audio response monitored by the technician during scan surveys.

- Eighteen reference background roads were surveyed, consisting of roads geographically close to an O&G brine-treated road that had not been identified as O&G brine-treated. Fourteen surface samples were collected from reference background roads.

7.1 Gamma Radioactivity Survey Results

The surveys included gross gamma radiation scans performed using 2-inch x 2-inch NaI detectors and a Ludlum Model 2221 scaler/ratemeter instrument. Two detectors were attached to the hitch of a standard sport utility vehicle (SUV) approximately 3 ft apart. This detector array was offset to provide as much edge/shoulder coverage as possible. Each detector was mounted approximately 6 inches above the road surface. Every road had a complete scan on both sides. A total of four detector passes on each road were conducted. The instrument data were recorded along with the location information using a pair of Trimble™ ProXT global positioning system (GPS) units.

7.1.1 Gross Gamma Radiation Scan Results

Gross gamma radiation scans, recorded in cpm, were performed on 32 road surfaces treated with O&G brine for dust suppression and road stabilization. The gamma radiation count rate data and GPS data were downloaded and placed on maps using the most recent aerial maps available from Pennsylvania Spatial Data Access (PASDA). GIS software was used to develop a graphic display of the gamma scan results. The resulting gamma radiation count rate intensity images are presented in **Appendix E**. The minimum, maximum, median, mean, and standard deviations for each data set are presented in **Table 7-1**. In addition to calculating the file statistics, a two-sample student t-test was performed.

The two-sample student t-test was used to compare the subject road (O&G brine-treated) results with a reference background road. ProUCL version 5.0 was used to perform the student t-test on the data. The Null Hypothesis tested is that the mean value of the treated road gamma radiation count rate data is statistically different from the mean value of the reference background road gamma radiation count rate data at the 95 percent confidence level. The results of the t-test for each pair of road results are included in **Table 7-1**. Fourteen of 29 comparisons of O&G brine-

treated and reference background roads are statistically different at the 95 percent confidence level. The t-test output files are included in **Appendix G**.

7.1.2 Gamma Radiation Exposure Rate Results Summary

Gross gamma radiation scan results in units of cpm were converted to μR/hr using 800 cpm per μR/hr, a conversion factor appropriate for Ra-226 gamma energy as detected with 2-inch by 2-inch NaI detectors rounded to one significant figure (Table 6.4, NaI Scintillation Detector Scan MDCs for Common Radiological Contaminants, NUREG-1507, Minimum Detectable Concentrations With Typical Radiation Survey Instruments for Various Contaminants and Field Conditions, USNRC June 1998). **Table 7-2** presents the results for each road.

7.2 Soil Sample Results

Biased surface soil samples were collected based on the audio response of the gamma scan survey instrument ratemeter on 31 of the 32 O&G brine-treated roads. When an area with elevated radioactivity was detected, surface soil samples were collected at that area.

7.2.1 Road Surface Soils Biased Sample Results

The gamma spectroscopy results are presented in **Tables 7-3** through **7-5** for the U, Th, and Ac series radionuclides. A review of the U series radionuclides indicates excess Ra-226 activity in 19 of 33 surface soil samples. For the purposes of this study, excess Ra-226 activity is defined as Ra-226 activity greater than the natural background U decay series activity in surface soil. The excess Ra-226 activity was determined as follows:

- The O&G brine applied to road surfaces contains Ra-226 and its progeny. It does not contain U, which is insoluble. Therefore, the U-238 activity identified in the gamma spectroscopy analysis results represents the natural background U series activity in surface soil for the area. The average U-238 activity of the 31 samples is 0.882 pCi/g.

- U-235 makes up 0.7 percent by weight of natural U, which equates to 1/22 of the U-238 activity. Therefore, 0.040 pCi/g of U-235 is present in the surface soil samples.

- Radium-226 is measured directly by detection of its 186.2 keV energy line (3.28 percent yield). However, the presence of U-235 can cause interference with direct Ra-226 detection because it has a gamma line of similar energy (185.7 keV at 54 percent yield). In solid samples where natural U including U-238 and Ra-226 are at equal activity and U-235 is at 1/22 the activity of U-238, overestimation of Ra-226 is quantified by multiplying the U-235 activity by the ratio of the yields of the similar gamma radiation emissions, i.e., 54/3.28. Therefore, the Ra-226 overestimation in the surface soil samples is equal to 0.659 pCi/g [0.040 pCi/g x (54/3.28) = 0.659 pCi/g].

- After correcting the reported Ra-226 activity by 0.882 pCi/g of natural background activity and 0.659 pCi/g of U-235 bias, 19 of 31 samples have excess Ra ranging from 0.109 to 5.42 pCi/g above natural background.

See Section 2.3 for a complete discussion of the identification of NORM radionuclides by gamma spectroscopy.

The gamma spectroscopy results for the Th series radionuclides indicate the Th series is in secular equilibrium. The Th-232 mean and median values are essentially equal and the standard deviation is a fraction of the mean value, indicating the data is normally distributed. A normal distribution of radioactivity measurements is indicative of natural background radioactivity, which is more homogeneous than contaminated soil. The mean Ra-228 activity of the 31 surface soil samples is 0.977 $\pm$ 0.351 pCi/g. The range of the results is from 0.455 to 1.85 pCi/g.

7.2.2 Road Surface Soils – Reference Background Roads Soils

As a point of reference and for comparison, 18 roads in the geographic vicinity of the subject roads that have not been identified as O&G brine-treated were selected for surveying, and 14 biased soil samples were collected. The gamma spectroscopy results of the background samples are presented in **Tables 7-6** through **7-8** for the U, Th, and Ac series radionuclides. A review of the U series radionuclides indicates excess Ra-226 activity in 14 surface soil samples. Excess Ra-226 activity is greater than the natural background U decay series activity in surface soil. The excess Ra-226 activity was determined as follows:

- The O&G brine applied to road surfaces contains Ra-226 and its progeny. It does not contain U, which is insoluble. Therefore, the U-238 activity identified in the gamma spectroscopy analysis results represents the natural background U series activity in surface soil for the area. The average U-238 activity of the 14 samples is 0.819 pCi/g.

- U-235 makes up 0.7 percent by weight of natural U, which equates to 1/22 of the U-238 activity. Therefore, there is 0.037 pCi/g of U-235 present in the surface soil samples.

- Radium-226 is measured directly by detection of its 186.2 keV energy line (3.28 percent yield). However, the presence of U-235 can cause interference with direct Ra-226 detection since it has a gamma line of similar energy (185.7 keV at 54 percent yield). In solid samples where natural U including U-238 and Ra-226 at equal activity and U-235 at 1/22 the activity of U-238, overestimation of Ra-226 is quantified by multiplying the U-235 activity by the ratio of the yields of the similar gamma emissions, i.e., 54/3.28. Therefore, the Ra-226 overestimation in the surface soil samples is equal to 0.037 pCi/g x (54/3.28) = 0.61 pCi/g.

- After correcting the reported Ra-226 activity by 0.819 pCi/g of natural background activity and 0.609 pCi/g of U-235 bias, 11 of 14 samples have excess Ra ranging from 0.0210 to 61.6 pCi/g above natural background.

See Section 2.3 for a complete discussion of the identification of NORM radionuclides by gamma spectroscopy.

The gamma spectroscopy results for the Th decay series are not normally distributed nor indicative of natural Th background radioactivity. Thorium-232 mean and median values are not equal and the standard deviation is large relative to the mean value, indicating the data are not normally distributed and heterogeneous. A normal distribution of radioactivity measurements is indicative of natural background radioactivity, which is more homogeneous than contaminated soil. The

mean Ra-228 activity of the 14 surface soil samples is 1.93 ± 2.81 pCi/g. The range of the results is from 0.396 to 11.2 pCi/g.

The background reference road soil sample results are positive for excess Ra-226 at 11 of 14 roads sampled. Three of the Ra-228 results are greater than 2.98 pCi/g, which is approximately three times natural background for the Th series. The excess Ra is higher than for the identified O&G brine-treated roads. The average excess Ra-226 for roads identified as having been O&G brine-treated is 1.13 pCi/g compared to an average of 8.23 pCi/g on the background reference roads. One possible explanation is that all of the roads have been treated with O&G brine. After the 32 roads had been identified as O&G brine-treated, the reference background roads were selected by proximity to the 32 roads. Nothing precluded the selected background roads from having been treated with O&G brine.

7.3 Public Exposure to Oil and Gas Brine-Treated Roads

A total of 31 samples were collected from roads treated with O&G brine. An additional 14 surface soil samples were taken in reference background areas not expected to be impacted by O&G brine treatment. Both the treated and the reference background roads were positive for excess Ra. To evaluate potential exposure to the public from the O&G brine-treated roads, a source term of 1 pCi/g of Ra-226 and 0.5 pCi/g of Ra-228 was assumed within a 6-inch layer of surface material (treated road surface).

The Argonne National Laboratory RESidual RADioactivity (RESRAD) Version 7.0 code for modeling exposure from residual radioactivity was used to evaluate potential exposure from the O&G brine-treated roads. RESRAD is a computer model designed to estimate radiation doses and risks from residual radioactive materials. RESRAD has been used widely by DOE, its operations and area offices, and its contractors for deriving limits for radionuclides in soil. RESRAD has also been used by EPA, U.S. Army Corps of Engineers (USACE), NRC, industrial firms, universities, and foreign government agencies and institutions. The recreationist is an appropriate exposure scenario based on the remote location of the roads. A recreationist, such as a jogger or hunter, usually spends less time on the impacted area, e.g., two hours a day, three days a week, than a resident. However, a recreationist may have a higher inhalation rate than a resident. Recreational land use addresses exposure to people who spend a limited amount of time at or near a site while playing, fishing, hunting, hiking, or engaging in other outdoor activities. Environmental exposure pathways included in the recreationist scenario include ground external gamma, inhalation, Rn, plant consumption, meat consumption, milk consumption, and soil ingestion.

The estimated total dose from 1 pCi/g of Ra-226 and 0.5 pCi/g of Ra-228 above natural background in surface soil, to a recreationist, in the year of maximum exposure (year 1) is 0.441 mrem/yr, which is below the 100 mrem/yr public exposure criteria based on assumed activity concentrations. The results of the environmental pathways for year 1, the year of maximum dose, are presented in **Table 7-9**. The actual dose received is dependent upon both the excess Ra radioactivity in surface soil and the time spent exposed to the soil surface.

Table 7-1. Gamma Scan Survey Summary

Study ID	GWS Max (cpm)	GWS Min (cpm)	GWS Median (cpm)	GWS Mean (cpm)	GWS Std Dev (cpm)	No. Data Points	NaI BKG (cpm)	T-Test Results (Sample to BKG)
BR-04-SL-011	16,512	7,892	13,022	12,655	1,588	2,906	12,511	Reject
BR-04-SL-010	Part of same road as BR-04-SL-011							
BR-05-SL-009	16,067	10,936	13,431	13,411	732	1,387	12,511	Reject
BR-06-SL-004	15,757	9,875	13,430	13,363	799	1,452	12,511	Reject
BR-07-SL-008	**15,641**	**7,975**	**12,843**	**12,511**	**1,449**	**2,389**	**NA**	**NA**
BR-01-SL-001	17,778	4,106	11,456	11,759	1,564	11,536	11,135	Reject
BR-02-SL-002	**13,268**	**9,766**	**11,050**	**11,135**	**615**	**850**	**NA**	**NA**
BR-08-SL-003	14,234	9,771	11,988	11,990	693	5,590	11,960	Accept
BR-09	**13,565**	**10,313**	**11,998**	**11,960**	**736**	**222**	**NA**	**NA**
BR-10-SL-012	15,179	5,888	11,977	11,968	996	9,253	10,898	Reject
BR-11	**12,762**	**9,449**	**10,882**	**10,898**	**591**	**596**	**NA**	**NA**
BR-13-SL-006	**13,180**	**9,526**	**11,311**	**11,273**	**646**	**961**	**NA**	**NA**
BR-12-SL-005	12,050	6,114	9,121	9,136	895	4,644	11,273	Accept
BR-15-SL-014	**14,509**	**7,695**	**10,816**	**10,873**	**1,128**	**1,359**	**NA**	**NA**
BR-14-SL-013	14,053	2,032	10,861	10,759	1,053	5,395	10,873	Accept
BR-16-SL-015	**12,360**	**9,470**	**10,587**	**10,614**	**461**	**592**	**NA**	**NA**
BR-17-SL-016	13,870	9,100	11,586	11,555	761	4,388	10,614	Reject
BR-18-SL-017	**9,949**	**6,066**	**7,479**	**7,524**	**616**	**727**	**NA**	**NA**
BR-19-SL-018	16,990	6,821	9,395	9,510	921	5,231	7,524	Reject
BR-20-SL-019	**13,511**	**5,404**	**8,747**	**8,825**	**1,317**	**3,944**	**NA**	**NA**
BR-21-SL-020	12,463	6,232	8,560	8,611	899	877	8,825	Reject
BR-22-SL-021	**13,126**	**5,947**	**9,019**	**9,317**	**1,646**	**704**	**NA**	**NA**
BR-23-SL-022	13,740	5,491	9,335	9,376	1,352	3,605	9,317	Accept
BR-24-SL-023	13,217	5,349	8,498	8,590	1,182	3,375	9,317	Accept
BR-25-SL-024	13,248	5,069	7,436	7,781	1,487	1,984	8,226	Accept
BR-26-SL-025	**11,208**	**5,882**	**8,254**	**8,226**	**893**	**343**	**NA**	**NA**
BR-27-SL-026	**11,333**	**5,708**	**8,281**	**8,267**	**955**	**579**	**NA**	**NA**
BR-28-SL-027	12,475	4,597	7,678	7,785	1,234	3,376	8,267	Accept
BR-29-SL-028	14,465	5,309	9,041	9,490	1,924	2,556	7,925	Reject
BR-30-SL-029	**10,360**	**5,687**	**7,965**	**7,925**	**703**	**759**	**NA**	**NA**
BR-31-SL-030	14,415	6,200	9,744	9,801	1,172	7,245	10,093	Accept
BR-32-SL-031	**14,117**	**6,527**	**10,057**	**10,093**	**1,118**	**1,958**	**NA**	**NA**
BR-33-SL-032	10,975	6,030	8,442	8,406	658	2,603	10,093	Accept
BR-34-SL-033	11,448	5,340	8,276	8,211	790	3,347	10,093	Accept
BR-35-SL-034	12,056	5,972	9,036	9,076	925	2,186	10,093	Accept

Table 7-1. Gamma Scan Survey Summary

Study ID	GWS Max (cpm)	GWS Min (cpm)	GWS Median (cpm)	GWS Mean (cpm)	GWS Std Dev (cpm)	No. Data Points	NaI BKG (cpm)	T-Test Results (Sample to BKG)
BR-36-SL-035	**10,981**	**5,693**	**8,566**	**8,502**	**748**	**975**	**NA**	**NA**
BR-37-SL-036	11,617	5,591	8,069	8,059	699	10,257	8,502	Accept
BR-38-SL-037	**10,668**	**6,105**	**8,006**	**7,979**	**662**	**406**	**NA**	**NA**
BR-39-SL-038	10,535	6,124	7,942	7,920	649	1,124	7,979	Accept
BR-40-SL-039	11,617	5,684	7,883	7,866	653	3,712	7,974	Accept
BR-41	**10,227**	**5,868**	**8,001**	**7,974**	**679**	**510**	**NA**	**NA**
BR-42-SL-040	10,859	5,774	7,951	7,950	722	1,560	NA	NA
BR-43-SL-041	12,789	5,048	7,978	7,954	1,036	3,399	NA	NA
BR-44-SL-042	15,498	5,710	9,911	9,995	1,759	5,223	6,260	Reject
BR-45-SL-043	15,390	6,376	11,268	11,015	1,531	1,399	6,260	Reject
BR-46-SL-044	**8,437**	**5,017**	**6,195**	**6,260**	**578**	**917**	**NA**	**NA**
BR-47-SL-045	10,560	5,177	7,252	7,258	822	3,434	6,260	Reject
BR-48-SL-046	12,338	5,208	7,868	7,991	1,239	3,152	6,260	Reject
BR-49-SL-047	14,314	5,523	8,906	9,124	1,418	2,928	6,260	Reject
BR-50-SL-048	12,933	6,066	9,315	9,292	1,067	2,293	6,260	Reject

Notes:
1. **Each group** of O&G brine-treated and associated background road(s) are shaded the same.
2. **Bold** – represents the background population for each shaded or unshaded group, respectively.
3. NA – indicates reference background road.
4. Accept (the Null Hypothesis) indicates there is a statistical difference in the data at the 95 percent confidence level. Reject (the Null Hypothesis) indicates the resulting surveys are statistically the same at the 95 percent confidence level.

Table 7-2. Summary of NaI Gamma Count Rate Data Converted to Exposure Rate

Study ID	GWS Max (µR/hr)	GWS Min (µR/hr)	GWS Median (µR/hr)	GWS Mean (µR/hr)	GWS Std. Dev. (µR/hr)	No. Data Points
BR-04-SL-011	20.6	9.90	16.3	15.8	2.00	2,906
BR-04-SL-010	Part of same road as BR-04-SL-011 – file statistics are same.					
BR-05-SL-009	20.1	13.7	16.8	16.8	0.90	1,387
BR-06-SL-004	19.7	12.3	16.8	16.7	1.00	1,452
BR-07-SL-008	19.6	10.0	16.1	15.6	1.80	2,389
BR-01-SL-001	22.2	5.10	14.3	14.7	2.00	11,536
BR-02-SL-002	16.6	12.2	13.8	13.9	0.800	850
BR-08-SL-003	17.8	12.2	15.0	15.0	0.900	5,590
BR-09	17.0	12.9	15.0	15.0	0.900	222
BR-10-SL-012	19.0	7.40	15.0	15.0	1.20	9,253
BR-11	16.0	11.8	13.6	13.6	0.700	596
BR-13-SL-006	16.5	11.9	14.1	14.1	0.800	961
BR-12-SL-005	15.1	7.60	11.4	11.4	1.10	4,644
BR-15-SL-014	18.1	9.60	13.5	13.6	1.40	1,359
BR-14-SL-013	17.6	2.50	13.6	13.4	1.30	5,395
BR-16-SL-015	15.5	11.8	13.2	13.3	0.600	592
BR-17-SL-016	17.3	11.4	14.5	14.4	1.00	4,388
BR-18-SL-017	12.4	7.60	9.30	9.40	0.800	727
BR-19-SL-018	21.2	8.50	11.7	11.9	1.20	5,231
BR-20-SL-019	16.9	6.80	10.9	11.0	1.60	3,944
BR-21-SL-020	15.6	7.80	10.7	10.8	1.10	877
BR-22-SL-021	16.4	7.40	11.3	11.6	2.10	704
BR-23-SL-022	17.2	6.90	11.7	11.7	1.70	3,605
BR-24-SL-023	16.5	6.70	10.6	10.7	1.50	3,375
BR-25-SL-024	16.6	6.30	9.30	9.70	1.90	1,984
BR-26-SL-025	14.0	7.40	10.3	10.3	1.10	343
BR-27-SL-026	14.2	7.10	10.4	10.3	1.20	579
BR-28-SL-027	15.6	5.70	9.60	9.70	1.50	3,376
BR-29-SL-028	18.1	6.60	11.3	11.9	2.40	2,556
BR-30-SL-029	13.0	7.10	10.0	9.90	0.900	759
BR-31-SL-030	18.0	7.80	12.2	12.3	1.50	7,245
BR-32-SL-031	17.6	8.20	12.6	12.6	1.40	1,958
BR-33-SL-032	13.7	7.50	10.6	10.5	0.800	2,603
BR-34-SL-033	14.3	6.70	10.3	10.3	1.00	3,347
BR-35-SL-034	15.1	7.50	11.3	11.3	1.20	2,186
BR-36-SL-035	13.7	7.10	10.7	10.6	0.900	975
BR-37-SL-036	14.5	7.00	10.1	10.1	0.900	10,257
BR-38-SL-037	13.3	7.60	10.0	10.0	0.800	406

Table 7-2. Summary of NaI Gamma Count Rate Data Converted to Exposure Rate

Study ID	GWS Max (µR/hr)	GWS Min (µR/hr)	GWS Median (µR/hr)	GWS Mean (µR/hr)	GWS Std. Dev. (µR/hr)	No. Data Points
BR-39-SL-038	13.2	7.70	9.90	9.90	0.800	1,124
BR-40-SL-039	14.5	7.10	9.90	9.80	0.800	3,712
BR-41	12.8	7.30	10.0	10.0	0.800	510
BR-42-SL-040	13.6	7.20	9.90	9.90	0.900	1,560
BR-43-SL-041	16.0	6.30	10.0	9.90	1.30	3,399
BR-44-SL-042	19.4	7.14	12.4	12.5	2.20	5,223
BR-45-SL-043	19.2	7.97	14.1	13.8	1.91	1,399
BR-46-SL-044	10.5	6.27	7.74	7.82	0.722	917
BR-47-SL-045	13.2	6.47	9.06	9.07	1.03	3,434
BR-48-SL-046	15.4	6.51	9.84	9.99	1.55	3,152
BR-49-SL-047	17.9	6.90	11.1	11.4	1.77	2,928
BR-50-SL-048	16.2	7.58	11.6	11.6	1.33	2,293

Table 7-3. Road-Biased Soil – Uranium Series Gamma Spectroscopy Results

Study ID	U-238 (pCi/g)	Ra-226 (pCi/g)	Pb-214 (pCi/g)	Bi-214 (pCi/g)
BR-01-SL-001	0.905	2.57	1.36	1.30
BR-04-SL-010	1.08	2.03	0.959	0.872
BR-04-SL-011	< 2.75	1.51	0.991	0.985
BR-05-SL-009	0.792	2.12	1.03	0.932
BR-06-SL-004	< 1.54	2.05	0.891	0.858
BR-12-SL-005	< 1.96	1.81	1.02	1.03
BR-14-SL-013	< 1.45	2.98	1.90	1.82
BR-15-SL-014	1.63	2.55	1.31	1.22
BR-17-SL-016	< 0.901	2.22	1.17	1.07
BR-19-SL-018	< 1.19	1.44	0.598	0.587
BR-21-SL-020	1.27	4.57	2.86	2.69
BR-23-SL-022	1.81	4.38	2.32	2.18
BR-24-SL-023	< 1.03	4.22	2.85	2.67
BR-25-SL-024	1.19	6.96	4.89	4.48
BR-28-SL-027	1.50	3.07	2.02	1.74
BR-29-SL-028	1.52	2.50	1.20	1.15
BR-31-SL-030	< 0.599	1.93	0.840	0.822
BR-33-SL-032	0.624	1.53	0.820	0.751
BR-34-SL-033	0.605	1.22	0.648	0.564
BR-35-SL-034	0.949	1.65	0.867	0.811
BR-37-SL-036	0.790	1.75	0.842	0.771
BR-39-SL-038	< 0.912	1.14	0.638	0.625
BR-40-SL-039	0.930	< 0.057	0.458	0.507
BR-42-SL-040	0.562	1.35	0.626	0.561
BR-43-SL-041	< 0.563	1.18	0.635	0.613
BR-44-SL-042	0.931	1.95	0.909	0.830
BR-45-SL-043	< 0.720	< 0.070	0.590	0.763
BR-47-SL-045	1.39	0.970	0.481	0.443
BR-48-SL-046	< 1.02	1.45	0.716	0.725
BR-49-SL-047	0.696	1.30	0.595	0.547
BR-50-SL-048	0.865	1.99	1.02	0.949
Average	0.882	2.14	1.23	1.16
Std. Dev.	0.410	1.38	0.932	0.852
Median	0.792	1.93	0.909	0.858
Minimum	0.282	0.029	0.458	0.443
Maximum	1.81	6.96	4.89	4.48

< – indicates a value less than the reported number which is the MDC.

Table 7-4. Road-Biased Soil – Thorium Series Gamma Spectroscopy Results

Study ID	Th-232 (pCi/g)	Ra-228 (pCi/g)	Ac-228 (pCi/g)	Pb-212 (pCi/g)	Bi-212 (pCi/g)
BR-01-SL-001	1.08	1.09	1.13	1.40	0.626
BR-04-SL-010	1.31	1.33	1.37	1.62	0.809
BR-04-SL-011	1.49	1.51	1.56	1.56	0.912
BR-05-SL-009	1.43	1.43	1.50	1.73	0.857
BR-06-SL-004	1.16	1.18	1.22	1.22	0.720
BR-12-SL-005	1.14	1.16	1.19	0.987	0.605
BR-14-SL-013	1.15	1.17	1.21	1.57	0.708
BR-15-SL-014	1.16	1.18	1.22	1.51	0.651
BR-17-SL-016	1.29	1.45	1.35	1.59	0.763
BR-19-SL-018	0.746	0.760	0.781	0.926	0.565
BR-21-SL-020	0.882	0.901	0.923	1.16	0.463
BR-23-SL-022	1.26	1.29	1.32	1.60	0.737
BR-24-SL-023	1.48	1.51	1.55	1.79	0.748
BR-25-SL-024	1.81	1.85	1.89	2.07	0.760
BR-28-SL-027	0.711	0.727	0.744	0.675	0.426
BR-29-SL-028	1.04	1.06	1.08	1.37	0.762
BR-31-SL-030	0.771	0.789	0.807	0.971	0.492
BR-33-SL-032	0.701	0.717	0.734	0.846	0.412
BR-34-SL-033	0.581	0.595	0.609	0.764	0.405
BR-35-SL-034	0.798	0.817	0.835	0.909	0.484
BR-37-SL-036	0.768	0.787	0.804	0.917	0.471
BR-39-SL-038	0.670	0.687	0.701	0.704	0.370
BR-40-SL-039	0.616	0.632	0.645	0.213	0.386
BR-42-SL-040	0.664	0.681	0.695	0.782	0.386
BR-43-SL-041	0.684	0.702	0.717	0.875	0.423
BR-44-SL-042	1.11	1.12	1.16	1.38	0.714
BR-45-SL-043	0.863	0.872	0.904	0.210	0.586
BR-47-SL-045	0.450	0.455	0.471	0.559	0.277
BR-48-SL-046	0.773	0.780	0.809	0.864	0.479
BR-49-SL-047	0.577	0.582	0.604	0.685	0.376
BR-50-SL-048	0.515	0.520	0.539	0.688	0.259
Average	0.972	0.979	1.00	1.10	0.569
Std. Dev.	0.334	0.349	0.355	0.465	0.179
Median	0.873	0.872	0.904	0.971	0.565
Minimum	0.450	0.455	0.471	0.210	0.259
Maximum	1.81	1.85	1.89	2.07	0.912

Table 7-5. Road-Biased Soil – Actinium Series and Miscellaneous Gamma Spectroscopy Results

Study ID	U-235 (pCi/g)	K-40 (pCi/g)
BR-01-SL-001	< 0.075	10.6
BR-04-SL-010	< 0.107	21.4
BR-04-SL-011	< 0.212	29.4
BR-05-SL-009	0.117	24.8
BR-06-SL-004	< 0.152	21.7
BR-12-SL-005	< 0.157	7.01
BR-14-SL-013	< 0.183	13.2
BR-15-SL-014	< 0.150	12.5
BR-17-SL-016	< 0.083	17.6
BR-19-SL-018	< 0.114	10.9
BR-21-SL-020	< 0.127	5.61
BR-23-SL-022	< 0.110	13.0
BR-24-SL-023	< 0.103	16.9
BR-25-SL-024	< 0.093	16.3
BR-28-SL-027	0.074	11.4
BR-29-SL-028	< 0.209	20.1
BR-31-SL-030	0.094	8.84
BR-33-SL-032	< 0.045	7.35
BR-34-SL-033	< 0.051	11.3
BR-35-SL-034	0.071	7.21
BR-37-SL-036	< 0.048	8.92
BR-39-SL-038	< 0.007	6.85
BR-40-SL-039	< 0.044	7.22
BR-42-SL-040	< 0.042	7.49
BR-43-SL-041	0.100	8.39
BR-44-SL-042	< 0.055	19.1
BR-45-SL-043	< 0.051	15.0
BR-47-SL-045	< 0.035	6.10
BR-48-SL-046	< 0.071	12.3
BR-49-SL-047	0.102	7.96
BR-50-SL-048	< 0.091	5.40
Average	0.056	12.6
Std. Dev.	0.029	6.19
Median	0.052	11.3
Minimum	0.018	5.40
Maximum	< 0.091	29.4

< – indicates a value less than the reported number which is the MDC.

Table 7-6. Reference Background Road – Uranium Series Gamma Spectroscopy Results

Study ID	U-238 (pCi/g)	Ra-226 (pCi/g)	Pb-214 (pCi/g)	Bi-214 (pCi/g)
BR-02-SL-002	< 1.64	3.07	1.69	1.69
BR-07-SL-008	< 1.58	2.38	1.05	0.965
BR-13-SL-006	< 1.08	6.09	3.81	3.59
BR-16-SL-015	< 1.55	2.24	1.09	0.967
BR-18-SL-017	< 0.753	0.828	0.479	0.445
BR-20-SL-019	< 3.14	63.0	51.0	48.4
BR-22-SL-021	< 1.99	16.1	14.2	12.7
BR-26-SL-025	< 0.919	4.25	3.01	2.85
BR-27-SL-026	0.643	4.10	2.83	2.70
BR-30-SL-029	1.61	2.86	1.55	1.45
BR-32-SL-031	< 0.854	1.69	1.11	0.940
BR-36-SL-035	0.825	1.41	0.640	0.609
BR-38-SL-037	12.7	1.55	0.784	0.711
BR-46-SL-044	8.04	1.13	0.523	0.468
Average	2.18	7.91	5.98	5.61
Std. Dev.	3.61	16.3	13.4	12.7
Median	0.805	2.62	1.33	1.21
Minimum	0.377	0.828	0.479	0.445
Maximum	12.7	63.0	51.0	48.4

< – indicates a value less than the reported number which is the MDC.

Table 7-7. Reference Background Road – Thorium Series Gamma Spectroscopy Results

Study ID	Th-232 (pCi/g)	Ra-228 (pCi/g)	Ac-228 (pCi/g)	Pb-212 (pCi/g)	Bi-212 (pCi/g)
BR-02-SL-002	1.38	1.41	1.45	1.70	0.826
BR-07-SL-008	1.28	1.30	1.34	1.66	0.874
BR-13-SL-006	3.26	3.32	3.43	2.03	0.885
BR-16-SL-015	1.28	1.30	1.34	1.58	0.778
BR-18-SL-017	0.392	0.399	0.410	0.509	0.244
BR-20-SL-019	11.0	11.2	11.5	10.5	1.53
BR-22-SL-021	2.93	2.99	3.06	3.47	0.765
BR-26-SL-025	1.05	1.08	1.10	1.12	0.414
BR-27-SL-026	0.838	0.857	0.877	0.982	0.331
BR-30-SL-029	0.543	0.556	0.568	0.778	0.307
BR-32-SL-031	0.709	0.725	0.742	1.07	0.433
BR-36-SL-035	0.637	0.653	0.667	0.788	0.376
BR-38-SL-037	0.752	0.772	0.788	0.890	0.441
BR-46-SL-044	0.392	0.396	0.410	0.513	0.249
Average	1.89	1.93	1.98	1.97	0.604
Std. Dev.	2.76	2.81	2.89	2.57	0.359
Median	0.944	0.969	0.989	1.10	0.437
Minimum	0.752	0.396	0.410	0.509	0.244
Maximum	11.0	11.2	11.5	10.5	1.53

Table 7-8. Reference Background Road – Actinium Series and Miscellaneous Gamma Spectroscopy Results

Study ID	U-235 (pCi/g)	K-40 (pCi/g)
BR-02-SL-002	< 0.223	13.6
BR-07-SL-008	< 0.149	23.1
BR-13-SL-006	< 0.165	18.1
BR-16-SL-015	< 0.161	12.0
BR-18-SL-017	< 0.131	6.14
BR-20-SL-019	< 0.322	9.32
BR-22-SL-021	< 0.197	20.7
BR-26-SL-025	< 0.085	6.07
BR-27-SL-026	< 0.069	4.87
BR-30-SL-029	< 0.058	6.68
BR-32-SL-031	< 0.050	13.0
BR-36-SL-035	< 0.050	7.18
BR-38-SL-037	< 0.044	8.73
BR-46-SL-044	0.077	4.44
Average	0.066	11.0
Std. Dev.	0.040	6.03
Median	0.071	9.03
Minimum	0.022	4.44
Maximum	0.161	23.1

< – indicates a value less than the reported number which is the MDC.

Table 7-9. Dose Assessment Results for Oil and Gas Brine-Treated Roads

Nuclide	Ground (mrem)	Inhalation (mrem)	Radon (mrem)	Plant (mrem)	Meat (mrem)	Milk (mrem)	Soil (mrem)
Ra-226	5.46E-02	1.25E-05	1.22E-05	0.000E+00	8.30E-02	0.000E+00	3.09E-04
Pb-210	3.40E-05	3.21E-05	0.000E+00	0.000E+00	2.20E-01	0.000E+00	1.55E-03
Ra-228	1.77E-02	4.10E-05	6.17E-05	0.000E+00	4.12E-02	0.000E+00	1.60E-04
Th-228	2.02E-02	2.06E-04	3.38E-04	0.000E+00	1.09E-03	0.000E+00	7.43E-05
Total	9.26E-02	2.92E-04	4.12E-04	0.000E+00	3.45E-01	0.000E+00	2.10E-03

8.0 QUALITY ASSURANCE AND QUALITY CONTROL

The quality assurance (QA) and QC objectives and criteria for this study were established in the study-specific Quality Assurance Project Plan (QAPP) which, along with the FSP, is available on the DEP website.

The purpose of the QAPP is to provide procedures and metrics for evaluating and ensuring that all data are technically sound and legally defensible. This is accomplished by establishing sample collection and preservation procedures, data collection procedures, analytical requirements and data evaluation processes, which result in accurate, precise, representative and complete data.

All sampling and analyses performed for this study were conducted in accordance with the QAPP standards.

8.1 Data Quality Levels (DQLs)

The requirements for this study were based on DQL I for field screening methods and DQL III for Non-Contract Laboratory Program (non-CLP) laboratory methods.

8.2 Quality Control Parameters

The established QC parameters for evaluating data in this study were precision [duplicates, matrix spikes (MS), matrix spike duplicates (MSD)], accuracy (spiked samples, laboratory control samples), and completeness (percentage of valid data).

Precision and accuracy obtained during this study met QC parameters unless otherwise noted.

Completeness is determined by calculating the percentage of valid data. Approximately eight percent of the gross α/β analyses were invalidated due to excessive concentrations of total dissolved solids (TDS). The TDS remaining after the water was evaporated were in excess of the allowable mass. Attempts to dilute the samples to allow valid analyses to be performed were unsuccessful.

8.3 Field Screening

Field surveys were performed by Perma-Fix personnel trained in the use of the survey instrumentation required. DQL I criteria were used to collect the following types of data:

- Gamma radiation exposure rate measurements
- Gross gamma radiation measurements
- Total α and β surface radioactivity
- Removable α and β surface radioactivity
- Background gamma radiation exposure rate and gross gamma radioactivity measurements (outside the influence of sampling areas)
- Liquid and solid samples for off-site analysis

8.4 Sample Identification

Field samples were assigned a unique number to identify information such as the sampling technician, the sequential number corresponding to the sample type, and the order in which it was collected in accordance with the FSP.

8.5 Sample Custody

A field chain-of-custody form or sample submission form was used to record the custody of all samples collected. This chain-of-custody form documented the transfer of the custody from the sampling personnel to another person, to the laboratory, or another party, such as a courier delivery service.

Field samples were packaged and shipped to the laboratory on the day of collection in accordance with chain-of-custody protocols. All samples were transported to the laboratory by the quick courier service or hand delivered to the laboratory. The original chain-of-custody form was sent with the samples. The remaining copy was stored in the field team files.

Further details pertaining to chain-of-custody may be found in the FSP.

8.6 Analytical Procedures

Analytical methods and procedures were established before the study began based on preliminary assumptions and are listed in **Table 8-1**. Additional analytical methods were subsequently added and/or modified when preliminary assumptions were found to be different due to the amount of TDS in the samples. Additional analytical method selection was based on the following:

- Original specified methodologies for radiochemistry failed due to elevated dissolved solids and Barium (Ba) concentrations.
- Alternate EPA methods, which were used as necessary.

All procedures for environmental sample handling, storage, and documentation while in the laboratory's custody and deliverable requirements upon delivery of the data to the user are documented in the laboratory's quality assurance manual (QAM).

8.7 Instrument Calibrations

All field and laboratory equipment were calibrated to NIST traceable standards before use to ensure proper operating accuracy. Laboratory instrument calibration procedures are presented in the laboratory QAMs. Field calibrations were performed in accordance with specified procedures. Prior to the use of field equipment, daily operational QC checks were completed. All daily QC instrumentation checks are presented in **Appendix B.**

8.8 Data Evaluation and Validation

The following subsections describe the field and laboratory data validation processes used for the study.

8.8.1 Validation of Field Data

During the field operations, field measurements were validated at the time of collection by the field sampler through the use of standard operating procedures (SOPs) and field QC checks. Field-obtained data, as well as ongoing QA/QC checks of environmental samples collected, were validated by trained Perma-Fix and DEP field technicians. All field data were reviewed at the time of sample collection.

8.8.2 Validation of Laboratory Data

Prior to reporting laboratory data, the analyst validated the sample results based on the QC criteria specified in the analytical methods. The data validation process included verification of the following steps:

- Ensure the standard regression coefficient is within the acceptable range.
- Ensure standard reference materials were analyzed at the proper frequencies and acceptable results were obtained.
- Ensure the reagent blanks were analyzed at the proper frequency.
- Ensure precision requirements of the plan were met.
- Ensure accuracy requirements of the plan were met.
- Ensure completeness requirements of the plan were met.
- Ensure samples were analyzed within the proper sample holding times.
- Verify all calculations were correct.
- Ensure proper units were reported.
- Ensure the proper methodologies were used.

In addition to the review of analytical results and project-specific precision, accuracy, and completeness requirements, the laboratory department manager or senior chemist performed internal audits of report forms and other data sheets as well as regular reviews of instrument logs, performance test results, and analysts' performance. Where review of analytical results or internal QA/QC checks indicated discrepancies, immediate corrective actions were taken and all data results collected since the previous approved QC audits were reviewed for validity. Specific laboratory procedures for validation of the analytical data generated are described in the laboratory QAMs.

8.9 Data Reporting – Analytical Laboratory

After the data were validated internally by the laboratory, the results were entered into the laboratory's data management system. The laboratory data management system contains the final data results. When data entries were completed, the laboratory director (or his/her designee) issued a final data report. The director then issued the final data report to the data user.

The data reports prepared for this project contain all pertinent information for the data user in determining the applicability and validity of the data. A specified and uniform data reporting format was implemented to facilitate this effort. For this project, DQL III data packages were reported as a DQL IV (CLP-like) deliverable to facilitate data validation and are presented in

Appendix K. The following criteria and information were supplied, as a minimum, for data reports generated for this project:

- A descriptive case narrative describing the internal data validation.
- Completed and legible chains-of-custody for all analyses contained within each submitted data package.
- A laboratory sample record documenting which analyses were performed for the samples contained in the data package is presented in **Table 8-1**.
- All of the laboratory sample identifications and the correlating field sample identifications.
- All applicable analytical results, counting errors, and MDCs reported in the correct number of significant figures and reporting units.
- Included in the individual sample reporting results are the complete sample identifications, the sample dilutions (if necessary), and the individual sample analysis dates.

8.9.1 DQL III Reporting

The following summary forms and raw data deliverable requirements apply for DQL III.

The following forms are required for all analyses using gamma spectroscopy; isotopic U and Th; and gross α, gross β and Ra methods, and were provided by the DEP Laboratory in various forms:

- Narrative and sample identification cross reference
- Copies of chain-of-custody documentation
- Laboratory chronicle
- Method summaries and references
- MS/MSD summary or any laboratory duplicate
- Method blank summary and results
- Instrument performance check summary
- Initial calibration summary for all constituents of interest

8.10 Quality Control Procedures

QC procedures and checks ensure the accuracy of the data.

For any laboratory QC result that was outside of the acceptance criteria, the samples were reanalyzed and/or the results were qualified in the final report.

8.10.1 Field QC Checks

Duplicate samples were collected and analyzed to assess the quality of field sampling techniques. These samples were treated as separate and discrete samples and analyzed by the selected offsite laboratory. The results are provided in Section 8.16.

8.10.2 Internal Laboratory QC Checks

The laboratory followed the internal QC checks specified in the QAPP for each analysis type employed. In addition, these QC checks have met the requirements specified in the respective EPA analytical methods.

8.10.2.1 Initial and Continuing Calibration

Each instrument and measurement system was calibrated prior to use to verify the instrument met performance criteria throughout the course of the analytical cycle. Continuing calibration checks were performed at a minimum frequency in accordance with the DEP Laboratory QAM. For instruments used for radiological analysis, performance checks are conducted each day samples are analyzed. For instruments used for non-radiological analysis, performance checks are conducted for each batch of 20 samples or less.

8.10.2.2 Reagent Blanks

A reagent blank was analyzed with each set of samples received for analysis. No responses above the reportable detection limit were observed in any of the blanks, indicating no possible laboratory contamination. The exact frequency and method of use is presented in the laboratory QAM.

8.10.2.3 Matrix Spike and Duplicate (Matrix Spike Duplicate) Analysis

One in 20 samples were analyzed as MSs and MSDs or one per day, whichever was greater. MS/MSD QC is not required for gamma spectroscopy analysis because no sample preparation is involved. The MS/MSD QC measures the effects of the sample matrix on method performance. The percent recovery for spiked samples was calculated using the equations documented in Section 11.0 of the QAPP and compared to the accuracy criteria specified in the QAM for the associated analytical method. The relative percent difference (RPD) of replicate spikes or replicate analytical results was calculated using the equations documented in Section 11.0 of the QAPP and compared to the precision criteria for the associated analytical method.

8.10.2.4 Calibration Standards

Calibration standards were analyzed as required in the reference methods throughout the course of the analysis. The exact frequencies and methods of use are presented in the laboratory QAM.

8.11 Laboratory Performance Audits

Laboratory performance audits are conducted by the DEP Laboratory QA officer three times per year. Each laboratory analyst is provided a performance evaluation or proficiency test sample containing analytes for the parameters which he/she usually performs. These proficiency test sample results are used to identify issues in sample preparation, analysis techniques, or methodologies. Any issues are identified, investigated, documented on the proper form, resolved with a corrective action plan to eliminate the issues and prevent reoccurrence, and then shared with the accreditation bodies.

The DEP Laboratory internal audits include verification of each analyst's record keeping, proper use and understanding of procedures, and performance documentation. Deficiencies/findings are

discussed with the analyst, documented, and resolved through the implementation of a corrective action.

8.12 Laboratory System Audits

Laboratory system audits are conducted by an external third-party assessor once every two years. These audits are used to ensure that all aspects of the DEP Laboratory's QAM are operative and within compliance. This involves a thorough review of all laboratory methods performed and documentation to confirm that all analytical procedures are performed according to the DEP Laboratory's QAM. An external third-party assessment was not conducted during the time period that samples from the TENORM study were received, processed, analyzed, and reported.

8.13 Assessment Procedures for Data Acceptability

The following subsections describe the data validation procedures that were used to evaluate the precision, accuracy, and completeness of the data generated.

8.13.1 Precision

Precision is the evaluation of agreement among individual measurements of the same property under prescribed similar conditions. Precision is assessed by calculating the RPD of replicate spike samples or replicate sample analyses according to the following equation:

Relative Percent Difference: $RPD = \frac{R_1 - R_2}{(R_1 + R_2)/2} \times 100$

Where: R_1 = result 1
R_2 = result 2

8.13.2 Accuracy

Accuracy is the evaluation of closeness of an individual measurement to the true value. Accuracy is measured by calculating the percent recovery (%R) of known levels of spike compounds as follows:

Percent Recovery:

$$\%R = \frac{[spike\ sample] - [unspiked\ sample]}{[spike\ added]} \times 100$$

8.13.3 Completeness

Completeness is the quantification of the amount of valid data obtained from a measurement system, expressed as a percentage of the number of valid measurements that could have been accomplished. More than one completeness check can be evaluated. It is calculated as follows:

$$Completeness\ (\%) = \frac{number\ of\ valid\ samples\ reported}{total\ number\ of\ samples\ analyzed} \times 100$$

8.13.4 Quality Control Charts

Valid QC charts can be prepared after the initial 20 analytical determinations to graphically evaluate precision and accuracy criteria. The charts are prepared by calculating the mean value of the determinations and setting control limits at $\pm$ 3 standard deviations from that mean. The following equations are used:

Mean:

$$\bar{X} = \frac{1}{N}\sum_{i=1}^{N} x_i$$

Where: N = number of samples
X_i = sample value

Standard Deviation:

$$\sigma = \sqrt{\frac{\sum_{i=1}^{N}(x_i - \bar{X})^2}{N-1}}$$

The control limits must be within acceptance limits or ranges presented in the DEP Laboratory's SOPs. If the values are found to be outside these limits or ranges, the measurement system is examined to determine if possible problems exist. Most of the values were found inside the limits; however, those values which exceeded the control limits were reported with an appropriate data qualifier.

8.14 Preventative Maintenance

Performance of preventative maintenance was completed on equipment to ensure operability. Instrument manuals are kept on file and used for reference whenever equipment required repair or maintenance.

8.14.1 Field Equipment

Field sampling personnel were responsible for preventative maintenance of all field instruments. The field sampling personnel ensured field instrumentation was protected from extreme weather conditions as well as physical hazards.

8.14.2 Laboratory Instruments

Preventative maintenance schedules and/or procedures for laboratory equipment are presented in the DEP Laboratory QAM. No major preventative maintenance was performed on the DEP Laboratory equipment during the time period that samples from the TENORM study were received, processed, analyzed and reported.

8.15 QA Reports to Management

Audit reports have been provided by the DEP Laboratory director (or his/her designee) as a means of tracking program performance. An annual method internal audit was performed covering the

period of January 1, 2013, to present. In addition, the state of New Jersey Department of Environmental Protection (NJDEP) performed an audit of the DEP Laboratory management system, QA program, and analytical testing procedures performed by the DEP Laboratory. The NJDEP submitted a February 11, 2013, report to the DEP Laboratory that concluded no findings for the Radiation Measurement Section.

Field QA reports were not necessary due to the size and length of individual sample collection activities. Any problems noted during sampling were immediately communicated to the project certified health physicist.

8.16 Third-Party Quality Control

QC samples were collected as follows:

- Solid Samples – five percent (field replicate/split) QC samples, i.e., one every 20 samples collected to verify results of onsite laboratory per total samples in a calendar year.
- Aqueous Samples – five percent (field replicate/split) QC samples, i.e., one every 20 samples collected to verify results of onsite laboratory per total samples in a calendar year.

The samples were sent offsite to an independent, third-party, accredited laboratory for gamma spectroscopy analysis and compared to the DEP Laboratory gamma spectroscopy analysis of the split sample using NRC Inspection Manual Procedure 84750:

- Divide each offsite laboratory result by its associated uncertainty to obtain the resolution. For purposes of this procedure, the uncertainty is defined as the relative standard deviation, one sigma, of the offsite laboratory results as calculated from counting statistics, i.e., the 95 percent confidence level reported error divided by 1.96.
- Divide each DEP Laboratory result by the corresponding offsite laboratory result to obtain the ratio (DEP Laboratory/offsite laboratory).
- The DEP Laboratory's measurement is in agreement if the value of the ratio falls within the limits shown in the following table for the corresponding resolution:

Criteria for Accepting the DEP Laboratory's Measurements

Resolution	Ratio
<4	Statistics are too poor for comparison
4-7	0.5-2.0
8-15	0.6-1.66
16-50	0.75-1.33

Resolution	Ratio
51-200	0.80-1.25
>200	0.85-1.18

The results of the comparison for solid samples are presented in **Tables 8-2** through **8-5** for four of the radionuclides identified using gamma spectroscopy. If either the DEP Laboratory or the third-party laboratory (GEL) result was < MDC value reported, the comparison was not made.

There were 28 comparisons of split solid samples made; 14 passed and 14 failed. The pass/fail rate of 50 percent is likely due to the difficulty with splitting solid samples in regards to the total radioactivity concentration. The performance has been determined to be acceptable based on the following criteria: split sampling of solid samples, especially at low-activity concentrations, rarely results in equal activity for both resulting samples. Radioactive particulate contamination within solids is usually not homogenous, making split sampling improbable to split activity evenly between the two aliquots.

- Mixing or blending of the solid sample prior to splitting into equal aliquots does not ensure the radioactivity is evenly divided.
- Duplicate analysis of the same solid sample is more appropriate as a third-party QC comparison, eliminating the large variability of split samples, but was not possible for this study.
- Liquid samples are much easier to mix prior to splitting and are a more appropriate measure of the agreement between the two laboratories.

The results of the comparison for liquid samples are presented in **Tables 8-6** through **8-9** for four of the radionuclides identified using gamma spectroscopy. If either the DEP Laboratory or the independent laboratory (GEL) result was < MDC value reported, the comparison was not made.

Of the 35 comparisons made on split liquid sample gamma spectroscopy analysis results, 30 met acceptance criteria. The agreement between the DEP Laboratory and the independent laboratory (GEL) gamma spectroscopy results is acceptable.

The following actions and/or conclusions were made based on the split solid sample analytical results:

1. Split sampling of radioactive solid samples does not result in equal radioactivity in the two resulting samples. Solid samples were mixed in the field prior to filling two sample containers (splitting the sample). Low-activity solid sample media does not readily split into equal radioactivity concentration.

2. All of the split solid samples failing the comparison acceptance criteria were reviewed by asking the question: “Would the result of one of the two splits result in a different conclusion?” For example, would the result of one split pass a compliance test that may be applicable to the media and the result of the other split fail? Only one sample, with results of 363 versus 6.02 pCi/g, would result in a different action based on the result.

3. Duplicate analysis of the same sample (no splitting) is a much better comparison of laboratory performance and is recommended for any future sample and analysis study.

In addition, the 5% of the total solid samples selected for QC were re-analyzed by the DEP Laboratory and then forwarded to an offsite laboratory for duplicate analysis. The samples were sent offsite to an independent, third-party, accredited laboratory for gamma spectroscopy analysis and compared to the DEP Laboratory gamma spectroscopy analysis of the same sample using two methods: the duplicate error ratio (DER) in the equation below and RPD equation from Section 8.13.1.

Duplicate Error Ratio: $DER = \frac{|S-D|}{\sqrt{TPU_S{}^2 + TPU_D{}^2}}$

Where:
S = Sample result
D = Duplicate result
TPU_S = Total propagated uncertainty of the sample
TPU_D = Total propagated uncertainty of the duplicate

A DER result less than 1.42 means the sample results may be identical, while a RPD of 35% means that the sample results may be identical. A total of 40 evaluations were made between the DEP Laboratory re-analysis results and the duplicates sent to the third-party laboratory. **Table 8-10** through **Table 8-13** provide the analytical results and the results of the DER and RPD calculations. Evaluating the results with the DER demonstrated the two laboratories produced statistically different results 49% of the time, while the RPD demonstrated a difference 32% of the time. Overall, duplicate analysis provided only slightly better agreement between the two laboratories as did split sample analyses.

The following actions and/or conclusions were made based on the duplicate solid sample analytical results:

1. The activity reported for Bi-214 and Pb-214 were generally higher for the third-party laboratory. This supports the conclusion of improperly sealed containers and the loss of some activity below Rn-222 in the uranium series.
2. A majority of the time the Ra-226 activity was reported higher by the DEP Laboratory. A difference in analytical technique may provide a bias. The DEP Laboratory counts Ra-226 directly while the third-party laboratory reports the Bi-214.
3. The activity reported for Pb-212 was generally higher for the DEP Laboratory than the third-party laboratory, although most of the difference can be attributed to the counting statistics of low activity samples.

Table 8-1. Summary of Analytical Procedures

Sample Type	Media/ Sample Type	Analytical Parameters	Analytical[(a)] Methods	Frequency[(b)]
Cuttings as produced on a drilling rig including cuttings stored temporarily on site in lined pits or containers Solid phase from flowback and produced water Solids accumulated in vessels or on equipment Scale from drilling rigs and associated equipment Soil/salt samples from beneficial reuse areas (Off-site Lab)	Soil/soil-like	Gamma spectroscopy to identify TENORM radionuclides Alpha spectroscopy to identify isotopic U (233/234, 235, and 238) and isotopic Th (228, 230, and 232)	USEPA 901.1 Health and Safety Laboratory (HASL) 300	Once per site
WWTP sludge WWTP discharge sediments (Off-site Lab)	Soil/soil-like	Gamma spectroscopy to identify TENORM radionuclides Alpha spectroscopy to identify isotopic U (U-233/234, 235, and 238) and isotopic Th (Th-228, 230, and 232)	USEPA 901.1 HASL 300	Three times per facility
Flowback and produced waters Accumulated liquids from production equipment (Off-site Lab)	Aqueous (Grab)	Gross α and β Gamma spectroscopy to identify TENORM radionuclides	USEPA 900.0 USEPA 901.1	Once per site
Influent Marcellus Shale industry water (as is and filtered) WWTP effluent discharge water (as is and filtered) (Off-site Lab)	Aqueous (Grab)	Gross α and β Gamma spectroscopy to identify TENORM radionuclides	USEPA 900.0 USEPA 901.1	Quarterly x3
Landfill Leachate	Aqueous (Grab)	Gross α and β Gamma spectroscopy analysis Radium (Ra-226 and Ra-228)	USEPA 900.0 USEPA 901.1 EPA 903.1 and EPA 904.0 equivalent	Once per landfill

Table 8-1. Summary of Analytical Procedures

Sample Type	Media/ Sample Type	Analytical Parameters	Analytical[(a)] Methods	Frequency[(b)]
Gas sampling as necessary (Off-site Lab) Ambient Radon	Gaseous (Grab) Charcoal canister	Radon Radon		As determined by DEP

(a) Analytical methods are as follows:
- Up to 10 percent of the samples, based on the gross α and β and gamma spectroscopy results, are also analyzed by α spectroscopy for U (U-238, U-235, and U-234), Th-232, Ra (Ra-226 and Ra-228), and for any unsupported decay chain radionuclides.
- Analytical method as specified or an equivalent method where appropriate.

(b) QC samples were collected as follows:
- Solid Samples – five percent (field replicate/split) QC samples, i.e., one every 20 samples collected to verify results of onsite laboratory per total samples in a calendar year.
- Aqueous Samples – five percent (field replicate/split) QC samples, i.e., one every 20 samples collected to verify results of on-site laboratory per total samples in a calendar year.

Table 8-2. Bi-214 Split Solid Sample Comparison Results

Study ID	Bi-214 Result (pCi/g)	Bi-214 Err (pCi/g)	Bi-214 MDC (pCi/g)	Bi-214 Resolution / Ratio	Bi-214 Criteria / Pass-Fail
5942116	0.001	0.000	0.016	7.00	NA
5942116GEL	0.556	0.158	0.120	0.002	NA
5942130	26.5	4.16	0.217	5.00	0.5-2.0
5942130GEL	12.1	4.77	0.857	2.19	Fail
5942134	0.638	0.106	0.057	5.00	0.5-2.0
5942134GEL	4.19	1.58	0.461	0.152	Fail
5942145	0.000	0.269	1.05	12.0	0.6-1.66
5942145GEL	1.14	0.185	0.109	0.000	Fail
5942155	3.77	0.317	0.056	21.0	0.75-1.33
5942155GEL	2.63	0.250	0.079	1.43	Fail
5942180	0.780	0.073	0.048	14.0	0.6-1.66
5942180GEL	0.969	0.133	0.074	0.805	Pass
5942189	370	25.3	1.11	377	0.85-1.18
5942189GEL	589	3.06	0.973	0.628	Fail
5942188	24.0	1.97	0.156	58.0	0.80-1.25
5942188GEL	21.6	0.726	0.241	1.11	Pass

NA = one or both results were less than the reported MDC; no comparison performed.

Table 8-3. Pb-212 Split Solid Sample Comparison Results

Study ID	Pb-212 Result (pCi/g)	Pb-212 Err (pCi/g)	Pb-212 MDC (pCi/g)	Pb-212 Resolution / Ratio	Pb-212 Criteria / Pass-Fail
5942116	-0.008	0.000	0.014	NA	NA
5942116GEL	0.533	0.093	0.099	NA	NA
5942130	6.31	0.377	0.484	7.00	0.5-2.0
5942130GEL	11.4	3.02	0.545	0.554	Pass
5942134	1.19	0.137	0.089	3.00	NA
5942134GEL	1.54	1.05	0.318	0.773	NA
5942145	0.909	0.129	0.062	27.0	0.75-1.33
5942145GEL	1.57	0.115	0.085	0.579	Fail
5942155	1.47	0.104	0.036	23.0	0.75-1.33
5942155GEL	1.51	0.131	0.066	0.974	Pass
5942180	0.832	0.072	0.059	21.0	0.75-1.33
5942180GEL	0.898	0.083	0.059	0.927	Pass
5942189	154	20.7	0.998	256	0.85-1.18
5942189GEL	146	1.12	0.743	1.06	Pass
5942188	8.40	0.589	0.178	19.0	0.85-1.18
5942188GEL	2.29	0.238	0.179	3.67	Fail

NA = one or both results were less than the reported MDC; no comparison performed.

Table 8-4. Pb-214 Split Soil Sample Comparison Results

Study ID	Pb-214 Result (pCi/g)	Pb-214 Err (pCi/g)	Pb-214 MDC (pCi/g)	Pb-214 Resolution / Ratio	Pb-214 Criteria / Pass-Fail
5942116	0.289	0.033	0.034	10.0	0.6-1.66
5942116GEL	0.689	0.132	0.120	0.419	Fail
5942130	26.4	1.93	0.217	8.00	0.6-1.66
5942130GEL	17.1	4.43	0.812	1.54	Pass
5942134	6.05	0.527	0.061	5.00	0.5-2.0
5942134GEL	3.89	1.39	0.418	1.56	Pass
5942145	1.21	0.213	0.066	19.0	0.75-1.33
5942145GEL	1.34	0.140	0.104	0.903	Pass
5942155	4.18	0.283	0.054	23.0	0.75-1.33
5942155GEL	3.18	0.271	0.086	1.31	Pass
5942180	0.822	0.072	0.059	16.0	0.6-1.66
5942180GEL	1.25	0.155	0.082	0.658	Pass
5942189	373	62.5	1.03	4.00	0.5-2.0
5942189GEL	6.02	3.13	4.47	62.0	Fail
5942188	26.3	1.73	0.152	66.0	0.80-1.25
5942188GEL	24.4	0.724	0.240	1.08	Pass

Table 8-5. Ra-226 Split Soil Sample Comparison Results

Study ID	Ra-226 Result (pCi/g)	Ra-226 Error (pCi/g)	Ra-226 MDC (pCi/g)	Ra-226 Resolution / Ratio	Ra-226 Criteria / Pass-Fail
5942116	-0.060	0.000	0.183	NA	NA
5942116GEL	0.556	0.158	0.120	NA	NA
5942130	31.7	2.66	2.49	5.00	0.5-2.0
5942130GEL	12.1	4.77	0.857	2.62	Fail
5942134	7.73	0.957	0.756	5.00	0.5-2.0
5942134GEL	4.19	1.58	0.461	1.85	Pass
5942145	1.99	0.418	0.595	12.0	0.6-1.66
5942145GEL	1.14	0.185	0.109	1.75	Fail
5942155	6.14	0.609	0.650	21.0	0.75-1.33
5942155GEL	2.63	0.250	0.079	2.34	Fail
5942180	1.50	0.382	0.579	14.0	0.6-1.66
5942180GEL	0.969	0.133	0.074	1.55	Pass
5942189	421	38.5	8.80	377	0.85-1.18
5942189GEL	589	3.06	0.973	0.715	Fail
5942188	35.1	2.67	1.75	58.0	0.80-1.25
5942188GEL	21.6	0.726	0.241	1.63	Fail

NA = one or both results were less than the reported MDC; no comparison performed.

Table 8-6. Bi-214 Split Liquid Sample Comparison Results

Study ID	Bi-214 Result (pCi/L)	Bi-214 Error (pCi/L)	Bi-214 MDC (pCi/L)	Bi-214 Resolution / Ratio	Bi-214 Criteria / Pass-Fail
5942389	41.0	7.00	7.00	6.00	0.5-2.0
5942389GEL	32.8	11.5	10.6	1.25	Pass
5942390	57.0	6.00	5.00	6.00	0.5-2.0
5942390GEL	29.3	9.13	9.10	1.95	Pass
5942391	181	24.0	22.0	15.0	0.6-1.66
5942391GEL	187	24.6	20.4	0.968	Pass
5942392	229	19.0	8.00	20.0	0.75-1.33
5942392GEL	251	25.0	13.6	0.912	Pass
5942228	458	35.0	8.00	30.0	0.75-1.33
5942228GEL	669	43.5	22.4	0.685	Fail
5942275	4,660	377	37.0	94.0	0.80-1.25
5942275GEL	4,450	92.9	38.8	1.05	Pass
5942276	4,320	38.0	11.0	105	0.80-1.25
5942276GEL	4,860	90.8	34.7	0.889	Pass
5942277	2,020	245	14.0	75.0	0.80-1.25
5942277GEL	2,370	62.2	26.0	0.852	Pass
5942278	2,150	33.0	22.0	71.0	0.80-1.25
5942278GEL	2,230	61.2	26.0	0.964	Pass
5942291	15,300	1,340	44.0	195	0.80-1.25
5942291GEL	16,400	165	62.2	0.933	Pass

Table 8-7. Pb-214 Split Liquid Sample Comparison Results

Study ID	Pb-214 Result (pCi/L)	Pb-214 Error (pCi/L)	Pb-214 MDC (pCi/L)	Pb-214 Resolution / Ratio	Pb-214 Criteria / Pass-Fail
5942389	45.0	8.00	9.00	8.00	0.6-1.66
5942389GEL	52.1	13.1	10.4	0.864	Pass
5942390	64.0	5.00	5.00	3.00	NA
5942390GEL	18.2	10.8	18.2	3.52	NA
5942391	178	23.0	23.0	17.0	0.75-1.33
5942391GEL	201	23.1	17.9	0.886	Pass
5942392	255	18.0	8.00	4.00	0.5-2.0
5942392GEL	43.4	23.9	43.4	5.88	Fail
5942228	510	33.0	9.00	33.0	0.8-1.25
5942228GEL	790	47.2	28.1	0.646	Fail
5942275	4,710	655	30.0	97.0	0.8-1.25
5942275GEL	4,770	96.2	200	0.987	Pass
5942276	4,320	373	20.0	106	0.80-1.25
5942276GEL	5,350	99.3	46.7	0.807	Pass
5942277	2,180	243	16.0	81.0	0.80-1.25
5942277GEL	2,570	61.9	135	0.848	Pass
5942278	2,160	249	28.0	72.0	0.80-1.25
5942278GEL	2,500	67.8	32.7	0.864	Pass
5942291	15,300	1,340	56.0	205	0.85-1.18
5942291GEL	18,100	173	84.4	0.845	Fail

NA = one or both results were less than the reported MDC; no comparison performed.

Table 8-8. Ra-226 Split Liquid Sample Comparison Results

Study ID	Ra-226 Result (pCi/L)	Ra-226 Error (pCi/L)	Ra-226 MDC (pCi/L)	Ra-226 Resolution / Ratio	Ra-226 Criteria / Pass-Fail
5942389	104	60.0	95.0	2.00	NA
5942389GEL	119	127	119	0.874	NA
5942390	117	40.0	63.0	2.00	NA
5942390GEL	135	117	135	0.867	NA
5942391	445	190	300	3.00	NA
5942391GEL	218	137	218	2.04	NA
5942392	453	70.0	98.0	2.00	NA
5942392GEL	221	190	221	2.05	NA
5942228	2,000	158	118	7.00	0.5-2.0
5942228GEL	1,200	324	312	1.67	Pass
5942275	8,360	1,490	533	20.0	0.75-1.33
5942275GEL	5,690	559	564	1.47	Pass
5942276	7,950	835	257	24.0	0.75-1.33
5942276GEL	6,740	560	511	1.18	Pass
5942277	3,910	698	220	18.0	0.75-1.33
5942277GEL	3,120	338	336	1.25	Pass
5942278	4,300	801	362	15.0	0.6-1.66

Table 8-8. Ra-226 Split Liquid Sample Comparison Results

Study ID	Ra-226 Result (pCi/L)	Ra-226 Error (pCi/L)	Ra-226 MDC (pCi/L)	Ra-226 Resolution / Ratio	Ra-226 Criteria / Pass-Fail
5942278GEL	3,100	410	374	1.39	Pass
5942291	25,500	3,270	713	59.0	0.8-1.18
5942291GEL	22,000	731	924	1.16	Pass

NA = one or both results were less than the reported MDC; no comparison performed.

Table 8-9. Ra-228 Split Liquid Sample Comparison Results

Study ID	Ra-228 Result (pCi/L)	Ra-228 Error (pCi/L)	Ra-228 MDC (pCi/L)	Ra-228 Resolution / Ratio	Ra-228 Criteria / Pass-Fail
5942389	94.0	15.0	14.0	8.00	0.6-1.66
5942389GEL	88.4	21.8	19.8	1.06	Pass
5942390	112	12.0	12.0	3.00	Poor Stats
5942390GEL	41.4	28.7	19.4	2.71	Fail
5942391	392	46.0	32.0	17.0	0.75-1.33
5942391GEL	434	49.0	36.2	0.903	Pass
5942392	467	36.0	13.0	21.0	0.75-1.33
5942392GEL	506	47.3	26.0	0.923	Pass
5942228	442	31.0	18.0	24.6	0.75-1.33
5942228GEL	318	54.9	40.0	1.39	Pass
5942275	571	79.0	67.0	10.0	0.6-1.66
5942275GEL	439	86.3	81.7	1.30	Pass
5942276	523	39.0	21.0	11.0	0.6-1.66
5942276GEL	561	98.9	64.2	0.932	Pass
5942277	230	25.0	22.0	9.00	0.6-1.66
5942277GEL	262	57.2	49.2	0.878	Pass
5942278	250	30.0	42.0	9.00	0.6-1.66
5942278GEL	231	52.3	55.4	1.08	Pass
5942291	1,740	164	56.0	26.0	0.75-1.33
5942291GEL	1,980	151	124	0.879	Pass

Table 8-10. Bi-214 Duplicate Sample Comparison Results

Study ID	Bi-214 Result (pCi/g)	Bi-214 Error (pCi/g)	Bi-214 MDC (pCi/g)	DER	RPD
5942107	0.089	0.013	0.012	0.860	68.2
5942107GEL	0.181	0.106	0.181		
5942111	80.9	7.37	0.076	2.91	24.0
5942111GEL	103	1.81	0.799		
5942116	0.500	0.058	0.029	1.36	36.3
5942116GEL	0.722	0.153	0.123		
5942134	6.04	0.714	0.030	0.010	0.170
5942134GEL	6.05	0.396	0.171		
5942145	0.798	0.144	0.025	0.910	25.9
5942145GEL	0.615	0.140	0.120		
5942155	3.96	0.485	0.030	0.280	4.44
5942155GEL	4.14	0.412	0.246		
5942180	0.829	0.133	0.033	0.510	13.3
5942180GEL	0.947	0.191	0.148		
5942186	51.2	4.67	0.046	1.15	10.2
5942186GEL	56.7	1.06	0.420		
5942189	457	81.2	0.567	0.270	4.70
5942189GEL	479	3.76	1.51		
5942189	2.25	0.268	0.028	0.130	2.25
5942189GEL	2.20	0.287	0.176		

Table 8-11. Pb-212 Duplicate Sample Comparison Results

Study ID	Pb-212 Result (pCi/g)	Pb-212 Error (pCi/g)	Pb-212 MDC (pCi/g)	DER	RPD
5942107	0.071	0.009	0.008	0.620	37.7
5942107GEL	0.104	0.052	0.104		
5942111	52.3	9.39	0.179	1.68	35.6
5942111GEL	36.5	0.851	0.730		
5942116	0.563	0.113	0.021	0.450	12.0
5942116GEL	0.635	0.115	0.095		
5942134	1.45	0.154	0.050	4.38	101
5942134GEL	0.475	0.161	0.165		
5942145	0.784	0.112	0.030	1.02	21.9
5942145GEL	0.629	0.103	0.085		
5942155	2.52	0.182	0.039	1.66	19.6
5942155GEL	2.07	0.200	0.193		
5942180	0.865	0.063	0.034	0.170	3.29
5942180GEL	0.837	0.151	0.133		
5942186	13.2	0.862	0.115	8.97	91.6
5942186GEL	4.91	0.334	0.351		
5942189	184	25.9	0.569	2.47	42.1
5942189GEL	120	1.62	1.37		
5942189	1.71	0.180	0.042	0.720	11.1
5942189GEL	1.53	0.175	0.156		

Table 8-12. Pb-214 Duplicate Sample Comparison Results

Study ID	Pb-214 Result (pCi/g)	Pb-214 Error (pCi/g)	Pb-214 MDC (pCi/g)	DER	RPD
5942107	0.087	0.010	0.007	3.09	123
5942107GEL	0.367	0.090	0.092		
5942111	102	6.43	0.138	2.98	17.9
5942111GEL	122	1.94	0.965		
5942116	0.581	0.125	0.021	1.00	32.0
5942116GEL	0.802	0.181	0.283		
5942134	6.50	0.561	0.037	1.88	18.2
5942134GEL	7.80	0.407	0.199		
5942145	0.827	0.110	0.030	0.310	7.40
5942145GEL	0.768	0.156	0.250		
5942155	4.46	0.305	0.036	1.24	13.2
5942155GEL	5.09	0.406	0.255		
5942180	0.859	0.068	0.032	1.32	29.8
5942180GEL	1.16	0.218	0.175		
5942186	57.4	3.64	0.081	2.89	17.5
5942186GEL	68.4	1.13	0.474		
5942189	472	61.4	0.661	2.02	23.2
5942189GEL	596	4.11	7.56		
5942189	2.43	0.212	0.031	1.15	15.6
5942189GEL	2.84	0.287	0.215		

Table 8-13. Ra-226 Duplicate Sample Comparison Results

Study ID	Ra-226 Result (pCi/g)	Ra-226 Error (pCi/g)	Ra-226 MDC (pCi/g)	DER	RPD
5942107	0.250	0.047	0.061	0.600	32.0
5942107GEL	0.181	0.106	0.181		
5942111	114	7.69	1.44	1.39	10.1
5942111GEL	103	1.81	0.799		
5942116	0.820	0.178	0.152	0.420	12.7
5942116GEL	0.722	0.153	0.123		
5942134	7.27	0.804	0.078	1.36	18.3
5942134GEL	6.05	0.396	0.171		
5942145	1.49	0.250	0.235	3.05	83.1
5942145GEL	0.615	0.140	0.120		
5942155	6.14	0.609	0.650	2.72	38.9
5942155GEL	4.14	0.412	0.246		
5942180	1.56	0.178	0.217	2.35	48.9
5942180GEL	0.947	0.191	0.148		
5942186	59.2	3.98	0.585	0.610	4.31
5942186GEL	56.7	1.06	0.420		
5942189	450	60.0	4.39	0.480	6.24
5942189GEL	479	3.76	1.51		
5942189	3.92	0.458	0.290	3.18	56.2
5942189GEL	2.20	0.287	0.176		

9.0 OBSERVATIONS AND RECOMMENDATIONS

Radiological sampling and surveys were conducted at well sites, WWTPs, landfills, gas distribution facilities and facilities that use natural gas, and O&G brine-treated roads. Various samples of solids, liquids, natural gas, and ambient air were collected and analyzed for radiological constituents and in some cases additional parameters. The data and various assessments are presented in Sections 3.0, 4.0, 5.0, 6.0, and 7.0. The following observations were made based upon the data compiled from the samples collected and surveys conducted as part of this study.

9.1 Observations

9.1.1 Well Sites (Section 3.0)

- *There is little potential for internal radiation exposure to workers and members of the public from α and β surface radioactivity from natural gas well site development drilling operations.*

  Ten of the 491 α measurements and 69 of the 491 β measurements of total surface radioactivity exceeded the RG 1.86 criteria. Only 1 of 493 α removable surface activity measurements and 1 of 493 β surface radioactivity measurements exceeded RG 1.86 criteria, indicating the total α/β surface radioactivity measured is fixed to the surface and not readily available for inhalation or ingestion. (Section 3.5.2)

- *There is little potential for exceeding public dose limits from external gamma radiation during the drilling phase of natural gas wells.*

  The gamma dose rates during the drilling phase ranged from background (measured at 5 μR/hr) to a maximum of 38.5 μR/hr, and the highest average exposure rate at any of the well sites was 18.1 μR/hr. (Section 3.5.1)

- *There is little potential for additional Rn exposure to workers and members of the public during the flowback phase of unconventional natural gas wells.*

  The Rn in ambient air measurement results during the flowback phase are within the range of typical ambient background Rn concentrations (0.00 to 1.11 pCi/L in outdoor ambient air in the U.S.). (Section 3.5.3)

- *There is little potential for radiological exposure to workers and members of the public from the handling, hauling, and temporary storage of vertical drill cuttings on natural gas well sites.*

  Vertical drill cuttings contain U, average of 1.47 $\pm$ 0.881 pCi/g, and Th, average 1.64 $\pm$ 0.403, slightly above typical background in surface soil. Both the U natural decay series and the Th natural decay series are identified in equilibrium. (Table 3-6)

- *There is little potential for radiological exposure to workers and members of the public from handling, hauling, and temporary storage of horizontal drill cuttings on natural gas well sites.*

Horizontal drill cuttings contain U, average 8.40 ± 6.70 pCi/g, and Th, average 1.42 ± 0.331. The Th is slightly above typical background in surface soil. The U activity is higher than typical surface soil background U activity and statistically higher than vertical drill cuttings U activity. Both the U natural decay series and the Th natural decay series are identified in equilibrium. (Table 3-8)

- *There is little potential for radiological exposure to workers and members of the public from hydraulic fracturing proppant sand.*

  Nominal U and Th activity was identified in hydraulic fracturing proppant sand samples. The U and Th activity was less than typical background for surface soil. (Section 3.2.4)

- *There is little potential for radiological exposure to workers and members of the public from drilling mud.*

  Nominal U and Th activity was identified in liquid and solid drilling mud samples. The U and Th activity was less than typical background for surface soil. (Section 3.2.3)

- *There is little potential for radiological exposure to workers and members of the public from handling and temporary storage of hydraulic fracturing fluid on natural gas well sites.*

  However, there is a potential for radiological environmental impacts from spills of hydraulic fracturing fluid on natural gas well sites and from spills that could occur from the transportation and delivery of this fluid.

  Radium-226 was detected within the hydraulic fracturing fluid ranging from 64.0 – 21,000 pCi/L. Radium-228 was also detected ranging from 4.50 – 1,640 pCi/L. The hydraulic fracturing fluid was made up of a combination of fresh water, produced water, and reuse flowback fluid. (Section 3.3.2)

- *There is little potential for radiological exposure to workers and members of the public from handling and temporary storage of flowback fluid on natural gas well sites.*

  However, there is a potential for radiological environmental impacts from spills of flowback fluid on natural gas well sites and from spills that could occur from the transportation and delivery of this fluid.

  Radium-226 concentrations were detected within flowback fluid samples ranging from 551 – 25,500 pCi/L. Radium-228 was also detected ranging from 248 – 1,740 pCi/L. (Section 3.3.3)

- *There is little potential for radiological exposure to workers and members of the public from handling and temporary storage of produced water on natural gas well sites.*

  However, there is a potential for radiological environmental impacts from spills of produced water from unconventional natural gas well sites and from spills that could occur from the transportation and delivery of this fluid.

Radium-226 concentrations were detected in produced water samples ranging from 40.5 – 26,600 pCi/L. Radium-228 concentrations were also detected ranging from 26.0 – 1,900 pCi/L. The Ra-226 activity in unconventional well site produced water is approximately 20 times greater than that observed in conventional well site produced water. The ratio of Ra-226 to Ra-228 in unconventional well site produced water is approximately eight times greater than that found in conventional well site produced water. (Sections 3.3.4 and 3.6.3)

- *There were no statistically significant differences observed between filtered and unfiltered liquid sample analytical results.*

  Because the liquid samples were preserved by addition of acid prior to filtering, the radioactive particulates may have entered solution and were therefore not removed by filtering. (Section 3.6.2)

- *The Rn concentrations in natural gas sampled at Pennsylvania well sites during this study are consistent with the Rn concentrations in natural gas reported by the U.S. Geological Survey (USGS) for Pennsylvania, which range from 1 to 79 pCi/L with an overall median of 37 pCi/L.*

  The Rn in natural gas measured ranged from 3.00 to 148 pCi/L, with a median Rn concentration of 41.8 pCi/L. (Section 3.4.2)

- *There is little potential for additional Rn exposure to workers and members of the public on or near natural gas well sites.*

  With the exception of one outlier at 1.70 pCi/L, the Rn concentrations in ambient air sampled at well sites during this study are consistent with the typical ambient background Rn concentrations of 0.00 to 1.11 pCi/L. It should be noted that the outlier is still well below the EPA guideline for indoor Rn concentration of 4 pCi/L.

9.1.2 Wastewater Treatment Plants (Section 4.0)

9.1.2.1 Publicly Owned Treatment Works

- *There is little potential for internal radiation exposure to workers and members of the public from α and β surface radioactivity at POTWs.*

  Nine of the 566 α measurements and 68 of the 566 β measurements of total surface radioactivity exceeded the RG 1.86 criteria. One of the 286 removable α measurements and none of the 286 removable β measurements exceeded the RG 1.86 criteria. Fixed or removable α and β surface radioactivity may present a potential inhalation or ingestion hazard if disturbed in the future. (Section 4.1.6.2)
- *There is little potential for exceeding public dose limits from external gamma radiation for workers and members of the public at POTWs.*

  The highest average gamma radiation exposure rate was 36.3 μR/hr, and the maximum gamma radiation exposure rate measured was 257 μR/hr. Assuming the time period of exposure is a full occupational year of 2,000 hours, the maximum average POTW annual external gamma

radiation exposure was estimated as 62.6 mrem/yr, which is less than the maximum public dose limit of 100 mrem/yr. (Sections 4.1.2.1 and 4.1.6.1)

- *There is little potential for radiological exposure to workers and members of the public from handling and temporary storage of filter cake at POTW-I's.*

  However, there is a potential for radiological environmental impacts from spills and the long-term disposal of POTW-I filter cake.

  The filter cake analytical results for POTW-I plants show Ra-226 and Ra-228 are present above typical background concentrations in soil. The average Ra-226 result was 20.1 pCi/g with a large variance in the distribution. The maximum result was 55.6 pCi/g. The average Ra-228 result was 7.63 pCi/g, and the maximum result was 32.0 pCi/g Ra-228. (Section 4.1.2.1)

- *There is little potential for radiological exposure to workers and members of the public from handling and temporary storage of filter cake at POTW-N's.*

  There is little potential for radiological environmental impacts from spills and the long-term disposal of POTW-N filter cake.

  The radioactivity levels at **POTW-N** plants presented in **Table 4-6** were above typical background concentrations in soil with Ra-226 average and maximum results of 9.72 pCi/g and 35.4 pCi/g. The average and maximum Ra-228 results were 2.26 pCi/g and 7.26 pCi/g. (Section 4.1.2.1)

- *There is little potential for radiological exposure to workers and members of the public from sediment-impacted soil at POTW-I's.*

  However, there is a radiological environmental impact to soil from the sediments from POTW-I's.

  The analytical results for POTW-I sediment-impacted soil samples indicate Ra-226 and Ra-228 are present at concentrations above typical background in soil. The average Ra-226 result was 9.00 pCi/g, and the maximum result was 18.2 pCi/g. The average Ra-228 result was 3.52 pCi/g, and the maximum result was 6.25 pCi/g. (Section 4.1.2.2)

- *There is little potential for additional Rn exposure to workers and the members of the public inside POTW-I's.*

  Indoor Rn results from POTW-I results ranges from 0.200 to 8.70 pCi/L. One result exceeds the EPA action level of 4 pCi/L. The Rn measured in indoor air averaged 1.74 pCi/L. The average is above the average indoor level of 1.3 pCi/L in the U.S. as reported by EPA. (Section 4.1.4)

9.1.2.2 Centralized Wastewater Treatment Plants

- *There is potential for internal radiation exposure to workers and members of the public from α and β surface radioactivity at CWTs that treat O&G wastewater. Fixed α and β surface*

radioactivity may present a potential inhalation and ingestion hazard if disturbed during routine system maintenance.

One hundred eighty-six of the 777 α measurements and 461 of the 777 β measurements of total surface radioactivity exceeded the RG 1.86 criteria. Seven of the 805 removable α measurements and 6 of the 805 removable β measurements exceeded the RG 1.86 criteria. The average of the β total surface radioactivity measurements exceeded the RG 1.86 criteria in 10 of the 11 CWT facilities surveyed. The average of the total α surface radioactivity measurements exceeded the RG 1.86 criteria in four of the 11 CWT facilities surveyed. The corresponding removable radioactivity measurements are mostly less than the RG 1.86 criteria, indicating the total radioactive contamination measured is fixed to the surface and not immediately available for inhalation or ingestion. (Section 4.2.6.2)

- *There is little potential for exceeding public dose limits from external gamma radiation for workers and members of the public at CWTs that treat O&G wastewater.*

  Assuming the time period of exposure is a full occupational year of 2,000 hours, and the average maximum exposure rate of 19.1 μR/hr (24.1 μR/hr less the background rate of 5 μR/hr), the maximum average CWT annual external gamma radiation exposure was estimated at 38 mrem/yr. The maximum gamma radiation exposure rate measured was 502 μR/hr on contact with the outside of a wastewater tank. (Section 4.2.6.1)

- *There is little potential for radiological exposure to workers and members of the public from handling and temporary storage of filter cake at CWTs that treat O&G wastewater.*

  However, there is a potential for radiological environmental impacts from spills and the long-term disposal of CWT filter cake from CWTs that treat O&G wastewater.

  The analytical results indicate all the CWT filter cake samples contain elevated Ra-226 and Ra-228 above typical background levels for soil. The maximum results were 294 pCi/g of Ra-226 and 177 pCi/g of Ra-228. Five of 27 filter cake samples exceeded the DOT Ra threshold for labeling as radioactive material. (Section 4.2.2.1)

- *There is little potential for radiological exposure to workers and members of the public from sediment-impacted surface soil at CWTs that treat O&G wastewater.*

  However, there is a radiological environmental impact to soil from the sediments from CWTs that treat O&G wastewater.

  Sediment-impacted soil was collected at the accessible effluent discharge points at the CWTs. Radium above typical soil background levels to a maximum of 508 pCi/g of total Ra was identified in the sediment-impacted soil samples. (Section 4.2.7)

- *There is little potential for radiological exposure to workers and members of the public from impacted soil at CWTs that treat O&G wastewater.*

However, there is a radiological environmental impact to surface soil at CWTs that treat O&G wastewater.

Gamma radiation walkover surveys identified areas with radioactivity above local background. At three of these locations, a biased soil sample was collected to determine the amount of activity at or near the surface. Radium above soil typical background levels to a maximum of 444 pCi/g Ra-226 and 83.1 pCi/g Ra-228 was identified in biased soil samples. (Section 4.2.2.3)

- *There is little potential for additional Rn exposure to workers and the members of the public inside CWTs that treat O&G wastewater.*

  Indoor air was sampled and analyzed for Rn concentration at various CWT indoor locations such as break rooms, laboratories, offices, etc. The results ranged from 0.900 to 5.00 pCi/L. Two results exceeded the EPA action level. The Rn measured in indoor air averaged 2.0 pCi/L. The average is above the average indoor level of 1.3 pCi/L in the U.S. as reported by EPA. (Sections 4.2.4 and 4.2.6.3)

9.1.2.3 Zero Liquid Discharge Plants

- *There is potential for internal α and β surface radioactivity exposure to workers and members of the public at ZLDs that treat O&G wastewater. Fixed α and β surface radioactivity may present a potential inhalation and ingestion hazard if disturbed during future routine system maintenance.*

  One hundred fifty-nine of the 566 α measurements and 175 of the 566 β measurements of total surface radioactivity exceeded the RG 1.86 criteria. Fourteen of the 589 removable α measurements and two of the 589 removable β measurements exceeded the RG 1.86 criteria. The highest average total α and β surface radioactivity levels were 239 dpm/100 cm^2 and 4,740 dpm/100 cm^2. The maximum total α and β surface radioactivity levels were 1,410 dpm/100 cm^2 and 49,700 dpm/100 cm^2. The corresponding removable surface radioactivity measurements are mostly less than the RG 1.86 criteria, only 14 of 589 measurements exceeded the applicable criteria, indicating the total surface radioactivity measured is fixed to the surface and not immediately available for inhalation or ingestion. Fixed α and β surface radioactivity may present a potential inhalation or ingestion hazard if disturbed during routine system maintenance. (Section 4.3.6.2)

- *There is little potential for exceeding public dose limits from external gamma radiation for workers and members of the public at ZLDs that treat O&G wastewater.*

  The maximum average gamma radiation exposure rate measured at any of the ZLD plants was 43.1 μR/hr. The lowest background gamma radiation exposure rate measured at any of the sites was 5 μR/hr. Assuming the time period of exposure is a full occupational year of 2,000 hours, the maximum average ZLD annual external gamma radiation exposure was estimated as 76 mrem/yr. The maximum gamma radiation exposure rate measured was 445 μR/hr. (Sections 4.3.1.4 and 4.3.6.1)

- *There is little potential for radiological exposure to workers and members of the public from handling and temporary storage of filter cake at ZLDs that treat O&G wastewater.*

  However, there is a potential for radiological environmental impacts from spills and the long-term disposal of filter cake from ZLDs that treat O&G wastewater.

  Radium-226 and Ra-228 were measured in ZLD filter cake samples at concentrations above typical background levels for surface soils. Radium-226 concentrations ranged from 3.08 to 480 pCi/g and Ra-228 concentrations ranged from 0.580 to 67.3 pCi/g. (Section 4.3.2.1)

- *There is little potential for radiological exposure to workers and members of the public from influent and effluent water at ZLDs that treat O&G wastewater.*

  However, there is a potential for radiological environmental impacts from spills of influent and effluent water at ZLDs that treat O&G wastewater.

  Radium (Ra-226 and Ra-228) was routinely detected in all liquid influent and effluent sample types with an approximate 50 percent difference between influent and effluent, but little difference between filtered and unfiltered results. Results ranged from 29.0 to 20,900 pCi/L. (Section 4.3.5)

- *There is little potential for additional Rn exposure to workers and the members of the public at ZLDs that treat O&G wastewater.*

  Indoor air was sampled and analyzed for Rn concentration at various indoor locations such as break rooms, laboratories, offices, etc. The results ranged from 0.50 to 4.90 pCi/L. Two results exceeded the EPA action level. The Rn measured in indoor air averaged 2.29 pCi/L. The average is above the average indoor level of 1.3 pCi/L in the U.S. as reported by EPA. (Sections 4.3.4 and 4.3.6.3)

- *There is little potential for exceeding public dose limits from external gamma radiation for truck drivers from hauling O&G wastewater or sludge/filter cake from facilities that treated O&G wastewater.*

  It was assumed a truck driver hauled full containers with either wastewater or sludge/filter cake for four hours per day and made return trips with empty containers for four hours per day. The driver was assumed to work 40 hours per week for 10 weeks per year hauling O&G wastewater or sludge. The total estimated dose to the wastewater truck driver was 0.35 mrem/yr. The total estimated dose to the sludge truck driver was 52 mrem/yr. (Section 4.3.6.4)

9.1.3 Landfills (Section 5.0)

- *There is little potential for radiological exposure to workers and members of the public from leachate at landfills.*

- *There is little difference in the radium detected in the leachate from the nine landfills selected based on the volume of O&G industry waste accepted and from the 42 other landfills.*

Samples of leachate were collected from the nine landfills selected based on the volume of O&G industry waste received and from the 42 other landfills not selected based on the volume of O&G industry waste received and analyzed using gamma spectroscopy for Ra-226 and Ra-228. Radium was detected above the MDC value in 38 of 51 samples. Radium-226 results ranged from 36.5 to 416 pCi/L with an average of 116 pCi/L in the 42 unselected landfills and 125 pCi/L in the nine selected landfills. Radium-228 results ranged from 2.50 to 55.0 pCi/L with an average of 11.9 pCi/L in the 42 unselected landfills and 18.0 pCi/L in the nine selected landfills. (Section 5.1)

- *There is limited potential for radiological environmental impacts from spills or discharges of effluent or influent leachate at landfills that accept O&G waste for disposal.*

  Nine influent and seven effluent leachate samples were collected at the nine selected landfills. Radium was detected in all of the leachate samples. Radium-226 results ranged from 48.5 to 378 pCi/L with an average of 138 pCi/L for effluent samples and 83.4 pCi/L for influent samples. Radium-228 results ranged from 3.00 to 1,100 pCi/L with an average of 178 pCi/L for effluent samples and 7.94 pCi/L for influent samples. The influent and effluent samples from the same facility do not represent the same leachate at different times in treatment. (Section 5.2.1)

- *There is little potential for radiological exposure to workers and members of the public from handling and temporary storage of filter cake at landfills that accept O&G waste for disposal.*

  However, there is a potential for radiological environmental impacts from spills and the long-term disposal of landfill filter cake from landfills that accept O&G waste for disposal.

  Filter cake from three of the nine selected landfills was sampled and analyzed using gamma spectroscopy. Radium was detected in all of the filter cake samples. Radium-226 results ranged from 8.73 to 53.0 pCi/g, with an average of 24.3 pCi/g. Radium-228 results ranged from 1.53 to 5.03 pCi/g, with an average of 3.85 pCi/g. (Section 5.2.2)

- *There is little potential for radiological exposure to workers and members of the public from sediment-impacted soil at landfills that accepted O&G waste for disposal.*

  However, there may be a radiological environmental impact to soil from the sediments from landfill leachate treatment facilities that treat leachate from landfills that accept O&G waste for disposal.

  The three landfills that had filter cake sampled also discharged effluent water to the environment. At each of the three effluent outfalls, a sediment-impacted soil sample was collected. Radium was detected in all of the samples. Radium-226 results ranged from 2.82 to 4.46 pCi/g with an average of 3.57 pCi/g. Radium-228 results ranged from 0.979 to 2.53 pCi/g with an average of 1.65 pCi/g. (Section 5.2.3)

- *There is little potential for additional Rn exposure to workers and the members of the public at or from landfills that accept O&G waste for disposal.*

Ambient air was sampled at the fence line of each of the nine selected landfills and analyzed for Rn concentration. The Rn in ambient air at the fence line of the landfills ranged from 0.200 to 0.900 pCi/L consistent with U.S. background levels of 0.00 to 1.11 pCi/L in outdoor ambient air.

- *There is little potential for internal α and β surface radioactivity exposure to workers and members of the public at landfills that accept O&G waste for disposal.*

  None of the 195 α measurements and 17 of the 195 β measurements of total surface radioactivity exceeded the RG 1.86 criteria. All average total α and β surface radioactivity levels were below the RG 1.86 criteria. The maximum total α and β concentrations were 84.6 dpm/100 cm^2 and 3,630 dpm/100 cm^2. The average removable α and β levels at each landfill were below the RG 1.86 criteria. The maximum removable α and β levels were also below the RG 1.86 criteria. None of the 205 removable α or β surface radioactivity measurements exceeded the RG 1.86 criteria. (Section 5.4.1.1)

- *There is little potential for exceeding public dose limits from external gamma radiation for workers and members of the public at landfills that accept O&G waste for disposal.*

  The highest average exposure rate was 13.5 μR/hr, and the maximum gamma exposure rate measured was 93.7 μR/hr. The minimum, limiting local background measured was 5 μR/hr. Assuming the duration of exposure is a full occupational year of 2,000 hours, the external gamma radiation exposure at the landfill was estimated as 17 mrem/yr, which is much less than the 100 mrem/yr dose equivalent limit for a member of the public. (Sections 5.3 and 5.4.1)

9.1.4 Gas Distribution and End Use (Section 6.0)

9.1.4.1 Natural Gas in Underground Storage

- *Radon concentrations in natural gas are lower after underground storage.*

  Natural gas samples were collected at four underground storage sites in Pennsylvania. Duplicate samples were collected at each site during injection into the storage formation, and also during withdrawal from the storage formation. (Section 6.1)

9.1.4.2 Natural Gas-Fired Power Plants

- *Radon concentrations in the natural gas sampled entering power plants are consistent with the Rn in natural gas concentrations in samples collected at well sites.*

  The two natural gas sample results from natural gas-fired power plants were 33.7 ± 1.80 pCi/L and 35.7 ± 11.0 pCi/L. (Section 6.2 and Table 6.3)

- *There is little potential for exceeding public dose limits from external gamma radiation for workers and members of the public at natural gas-fired power plants.*

The gamma radiation exposure rate survey results at the PP-02 power plant were within the range of natural background of gamma radiation for PA. The exception occurred on the external surface of a pipe elbow where the range of measurement results observed was 15 to 17 μR/hr. (Section 6.2)

- *There is little potential for additional Rn exposure to workers and the members of the public at or from natural gas-fired power plants.*

  Ambient air was sampled at the PP-02 power plant site fence line. The fence line Rn monitor results were all at or below the MDC value for the analysis. (Section 6.2)

9.1.4.3 Compressor Stations

- *Radon concentrations in the natural gas sampled at compressor stations are consistent with the Rn in natural gas concentrations in samples collected at well sites.*

  All compressor stations were receiving predominately Marcellus Shale unconventional natural gas at the time of sample collection. The range of compressor station natural gas Rn results is 28.8 ± 1.40 to 58.1 ± 1.10 pCi/L, which is consistent with the production site Rn sample results. (Section 6.3 and Table 6.5)

- *There is little potential for additional Rn exposure to workers and the members of the public at or from natural gas compressor stations.*

  Ambient air was sampled at the CS-01 compressor station fence line for the measurement of Rn concentrations. The fence line Rn monitors results ranged from 0.100 to 0.800 pCi/L. The average concentration at each fence line location was within the range of typical ambient background Rn concentrations in outdoor ambient air in the U.S. (Section 6.3)

9.1.4.4 Natural Gas Processing Plant

- *Radon concentrations in natural gas entering the natural gas processing plant are consistent with levels measured at well sites.*

  Radon in natural gas sampled entering the plant measured 67.7 pCi/L. The Rn in natural gas sampled at the processing plant outflow measured 9.30 pCi/L. (Section 6.4 and Table 6.7)

- *There is potential for exceeding public dose limits from external gamma radiation for workers at the natural gas processing plant.*

  Contact readings measured with filter housings ranged from background to 75 μR/hr, with two exceptions; one measured 350 μR/hr and the other measured 900 μR/hr. Radiation exposure rates with values ranging from 20 to 400 μR/hr were measured on additional system components. (Section 6.4)

- *There is potential for internal α and β surface radioactivity exposure to workers at the natural gas processing plant when a filter housing is opened.*

The filter housing on the facility propanizer equipment was opened during a filter change-out and a sample of the cardboard filter media was collected. The filter media sample was smeared for removable α and β surface radioactivity. The average α and β surface radioactivity levels are below the RG 1.86 α and β removable surface radioactivity criterion. The results of samples collected from the facility propanizer equipment filter had a Pb-210 activity result of 3,580 pCi/g, but no other gamma-emitting NORM radionuclide results were above 1 pCi/g. The gross α and β removable surface radioactivity results for the filter media sample are elevated relative to the RG 1.86 gross α and β removable surface radioactivity criterion. (Section 6.5)

9.1.4.5 Radon Dosimetry

- *There is little potential for additional Rn exposure to members of the public in homes using natural gas from Marcellus Shale wells.*

- *The potential radiation dose received by home residents is a small fraction of the allowable general public dose limit of 100 mrem/yr.*

  Radon is transported with natural gas into structures (homes, apartments, and buildings) that use natural gas for purposes such as heating and cooking. The incremental increase of potential dose from Rn-222 to occupants of a typical home from use of natural gas was conservatively estimated as 5.2 mrem/yr for the median dose and 17.8 mrem/yr for the maximum dose. Based on the Rn and natural gas data collected as part of this study and the conservative assumptions made, the incremental Rn increase in a home using natural gas appliances is estimated to be very small, and would not be detectable by commercially available Rn testing devices. The average and maximum calculated Rn concentration increase in homes were 0.04 and 0.13 pCi/L. (Section 6.6)

9.1.5 Oil and Gas Brine-Treated Roads (Section 7.0)

- *Radium activity measured in O&G brine-treated road samples is greater than typical surface soil concentrations.*

  Biased surface soil samples were collected based on the audio response of the gamma scan survey instrument ratemeter on 31 of the 32 O&G brine-treated roads. When an area with elevated radioactivity was detected, surface soil samples were collected at that area. After correcting the reported Ra-226 activity by 0.882 pCi/g of natural background activity and 0.659 pCi/g of U-235 bias, 19 of 31 samples have excess Ra ranging from 0.109 to 5.42 pCi/g above natural background. (Sections 7.0 and 7.2.1)

- *Radium activity measured in reference background road samples is greater than typical surface soil concentrations. The reference background roads were selected by geographical location to O&G brine-treated roads selected for the study.*

  As a point of reference and for comparison, 18 roads in the geographic vicinity of the subject roads that have not been identified as O&G brine-treated were selected for surveying, and 14 biased soil samples were collected. After correcting the reported Ra-226 activity by

0.819 pCi/g of natural background activity and 0.710 pCi/g of U-235 bias, 11 of 14 samples have excess Ra ranging from 0.0210 to 61.5 pCi/g above natural background. Three of the Ra-228 results are greater than 2.98 pCi/g, which is approximately three times natural background for the Th series. (Section 7.2.2)

- *The excess Ra measured in reference background samples is higher than for the identified O&G brine-treated roads.*

  The average excess Ra-226 for roads identified as having been brine-treated is 1.13 pCi/g compared to an average of 8.23 pCi/g on the background reference roads. One possible explanation is that all of the roads have been treated with brine. After the 32 roads had been identified as brine-treated, the reference background roads were selected by proximity to the 32 roads. (Section 7.2.2)

- *There is little potential for members of the public exceeding the public dose limit from exposure to Ra in O&G brine-treated roads.*

  To evaluate potential exposure to the public from the brine-treated roads, a source term of 1 pCi/g of Ra-226 and 0.5 pCi/g of Ra-228 was assumed within a 6-inch layer of surface material (treated road surface). The estimated total dose from 1 pCi/g of Ra-226 and 0.5 pCi/g of Ra-228 above natural background in surface soil, to a recreationist, in the year of maximum exposure (year 1) is 0.441 mrem/yr, which is below the 100 mrem/yr public exposure criteria based on assumed activity concentrations. The actual dose received is dependent upon both the excess Ra radioactivity in surface soil and the time spent exposed to the soil surface. (Section 7.3)

9.2 Recommendations for Future Actions

9.2.1 Well Sites

- Conduct research and investigation of vertical and horizontal drill cuttings for beneficial use, onsite disposal, and future landfill disposal protocols.

- Add sampling and analyses for Ra-226, Ra-228, and additional man-made radionuclides such as tracers used in the O&G industry to Pennsylvania spill response protocol for spills of flowback fluid, hydraulic fracturing fluid, or produced water. Field survey instrumentation should also be available for surveys of areas impacted by the spill.

9.2.2 Wastewater Treatment Plants

- Perform routine survey and assessment of areas impacted with surface radioactivity to determine personnel protective equipment (PPE) use and monitoring during future activity that may cause surface α and β radioactivity to become airborne.

- Conduct additional radiological sampling and analyses and radiological surveys at all WWTPs accepting wastewater from O&G operations to determine if there are areas of contamination that require remediation; if it is necessary to establish radiological effluent discharge limitations; and if the development and implementation of a spill policy is necessary.

9.2.3 Landfills

- Evaluate and, if necessary, modify the landfill disposal protocol for sludges/filter cakes and other solid waste-containing TENORM.

- Conduct additional radiological sampling and analyses and radiological surveys at all facilities that treat leachate from landfills that accept waste from O&G operations to determine if there are areas of contamination that require remediation; if it is necessary to establish radiological effluent discharge limitations; and if the development and implementation of a spill policy is necessary.

- Add total Ra (Ra-226 and Ra-228) to the annual suite of contaminants of concern in leachate sample analyses.

9.2.4 Gas Distribution and End Use

- Survey and sample internal surfaces of natural gas plant piping and filter housings for radiological contamination. This effort should include evaluation of worker exposure and buildup of radioactivity in systems from natural gas processing and transmission. Evaluate monitoring and recommendation of PPE and other controls to be used during pipe clean-out and other activities when internal surfaces are exposed.

9.2.5 Oil and Gas Brine-Treated Roads

- Perform further study of O&G brine-treated roads. This study should evaluate produced water radionuclide concentrations prior to treatment, resultant surface activity and radionuclide concentration of road surfaces and future Ra migration.

10.0 REFERENCES

Section 1.0:

1. PSU Marcellus Center for Outreach and Research (MCOR) 2014. www.marcellus.psu.edu, August.
2. U.S. EPA 2014. > Climate Change > Natural Gas Star Program > Basic Info, www.epa.gov/methane/gasstar/basi-information/index.html, August.
3. U.S. EPA 2003. "EPA Assessment of Risks from Radon in Homes," June.

Section 2.0:

1. ANSI/HPS 2009. N13.53-2009, Control and Release of Technologically Enhanced NORM (TENORM).
2. DOE 1990. DOE Procedure Gamma 4.5.2.3, EML Procedures Manual, 27th Edition, Vol. I, DOE Report HASL-300-ED. 27-VOL. 1.
3. DOT, 49 CFR 173.436, Radioactive Material.
4. EPA 2012. U.S. EPA/OAR/IED (6609J), EPA 402-K-12-002, A Citizen's Guide to Radon, May. Also: http://www.epa.gov/radon/pubs/citguide.html
5. EPA 2000. Directive No. 9200.4-35, Remediation Goals for Radioactively Contaminated CERCLA Sites.
6. EPA 1980. EPA 900.0, "Prescribed Procedures for Measurement of Radioactivity in Drinking Water," U.S. EPA-6000/4-80-032, August.
7. EPA, Drinking Water Standard, 40 CFR 141.66.
8. Jenkins, P.H., Burkhart, J.F., and Camley, R.E. 2014. "Errors in Measurement of Radon-222 in Methane and Carbon Dioxide using Scintillation Cells Calibrated for Radon-222 in Air." Health Physics, Vol. 106, No. 3, March.
9. IAEA 2010. Radiation Protection and the Management of Radioactive Waste in the Oil and Gas Industry, Training Course Series No. 40, Vienna, May.
10. NRC, 10 CFR 20 Appendix B, Table 1, Col 3.
11. NRC, 10 CFR 20 Appendix B, Table 2, Liquid Effluent.
12. NRC, 10 CFR 20.1201, Occupational Dose Limits for Adults.
13. NRC, 10 CFR 20.1301, Radiation Dose Limits for Members of the Public.
14. NRC, 10 CFR 20.1402-20.1403, Radiological Criteria for Unrestricted Use.
15. NRC 1974a. Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors (1974)—Criteria for Natural Thorium Including Ra-228.
16. NRC 1974b. Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors (1974)—Criteria for Ra-226.
17. OSHA, 29 CFR 1910.1096.
18. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) 2000. Sources and Effects of Ionizing Radiation.

Section 3.0:

1. Jenkins et al., 2014. Health Physics, Vol. 106, No. 3, March.
2. NRC 1998a. Table 6.4, NaI Scintillation Detector Scan MDCs for Common Radiological Contaminants, NUREG-1507, Minimum Detectable Concentrations With Typical Radiation Survey Instruments for Various Contaminants and Field Conditions, June.
3. NRC 1998b. Table 6.3, NaI Scintillation Detector Count Rate Versus Exposure Rate (cpm/μR/hr), NUREG-1507, Minimum Detectable Concentrations With Typical Radiation Survey Instruments for Various Contaminants and Field Conditions, June.
4. NRC 1974. Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors (1974)—Criteria.

Section 4.0:

1. EPA 2012. U.S. EPA/OAR/IED (6609J), EPA 402-K-12-002, A Citizen's Guide to Radon, May. Also: http://www.epa.gov/radon/pubs/citguide.html
2. NRC 1998a. Table 6.4, NaI Scintillation Detector Scan MDCs for Common Radiological Contaminants, NUREG-1507, Minimum Detectable Concentrations With Typical Radiation Survey Instruments for Various Contaminants and Field Conditions, June.
3. NRC 1974. Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors (1974)—Criteria.

Section 5.0:

1. NRC 1998a. Table 6.4, NaI Scintillation Detector Scan MDCs for Common Radiological Contaminants, NUREG-1507, Minimum Detectable Concentrations With Typical Radiation Survey Instruments for Various Contaminants and Field Conditions, June.
2. NRC 1974. Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors (1974)—Criteria.

Section 6.0:

1. American Gas Association, Washington, D.C.
2. DOT 2011. National Pipeline Mapping System, User Guide.
3. EPA 2003. Pawal, D.J. and Puskin, J.S. EPA Assessment of Risks from Radon in Homes, June.
4. Jenkins et al., 2014. Health Physics, Vol. 106, No. 3, March.
5. Nazaroff, W.W. and Nero, A.V., 1988. Radon and its Decay Products in Indoor Air. John Wiley & Sons.
6. NRC 1974. Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors (1974)—Criteria.
7. Rowan, E.L. and Kraemer, T. F. U.S. Geological Survey. Radon-222 Content of Natural Gas Samples from Upper and Middle Devonian Sandstone and Shale Reservoirs in Pennsylvania: Preliminary Data, Open-file Report Series 2012-1159.
8. Spectra Energy Transmission 2014, Personal Communication, May.

9. UNSCEAR 2006. Annex E.
10. U.S. Census, American Housing Survey, 2011, Table C-02-AH.

Section 7.0:

1. Pennsylvania Department of Environmental Protection (DEP) 2013. Fact sheet, *Roadspreading of Brine for Dust Control and Road Stabilization*, developed under the authority of the Clean Streams Law, the Solid Waste Management Act, and Chapters 78 and 101 of DEP's Rules and Regulations.
2. NRC 1998a. Table 6.4, NaI Scintillation Detector Scan MDCs for Common Radiological Contaminants, NUREG-1507, Minimum Detectable Concentrations With Typical Radiation Survey Instruments for Various Contaminants and Field Conditions, June.

Section 8.0:

1. DEP 2013. Sample and Analysis Plan Part I, Field Sampling Plan (FSP), April.
2. DEP 2013. Sample and Analysis Plan Part II, Quality Assurance Project Plan (QAPP), April.
3. DEP. Laboratory QAM.
4. NRC 1994. NRC Inspection Manual Procedure 84750, March.

Section 9.0:

1. EPA 2012. U.S. EPA/OAR/IED (6609J), EPA 402-K-12-002, A Citizen's Guide to Radon, May. Also: http://www.epa.gov/radon/pubs/citguide.html
2. NRC 1974. Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors (1974)—Criteria.

EXHIBIT A-30

Environmentally Sustainable Solutions to Recycle Oil Cuttings

Re: Comments to the Railroad Commission of Texas on the August 2024 Proposed Rulemaking on Rule 8 and Subchapter B.

THE UNIVERSITY OF TEXAS AT AUSTIN
CENTER FOR TRANSPORTATION RESEARCH

Environmentally Sustainable Solutions to Recycle Oil Cuttings

Prof. Robert Gilbert

Prof. Amit Bhasin

Prof. Lynn Katz

Prof. Maria Juenger

Prof. Chadi El Mohtar

Darren Hazlett

Chih-Yu Tung

Jae Kyeong Jang

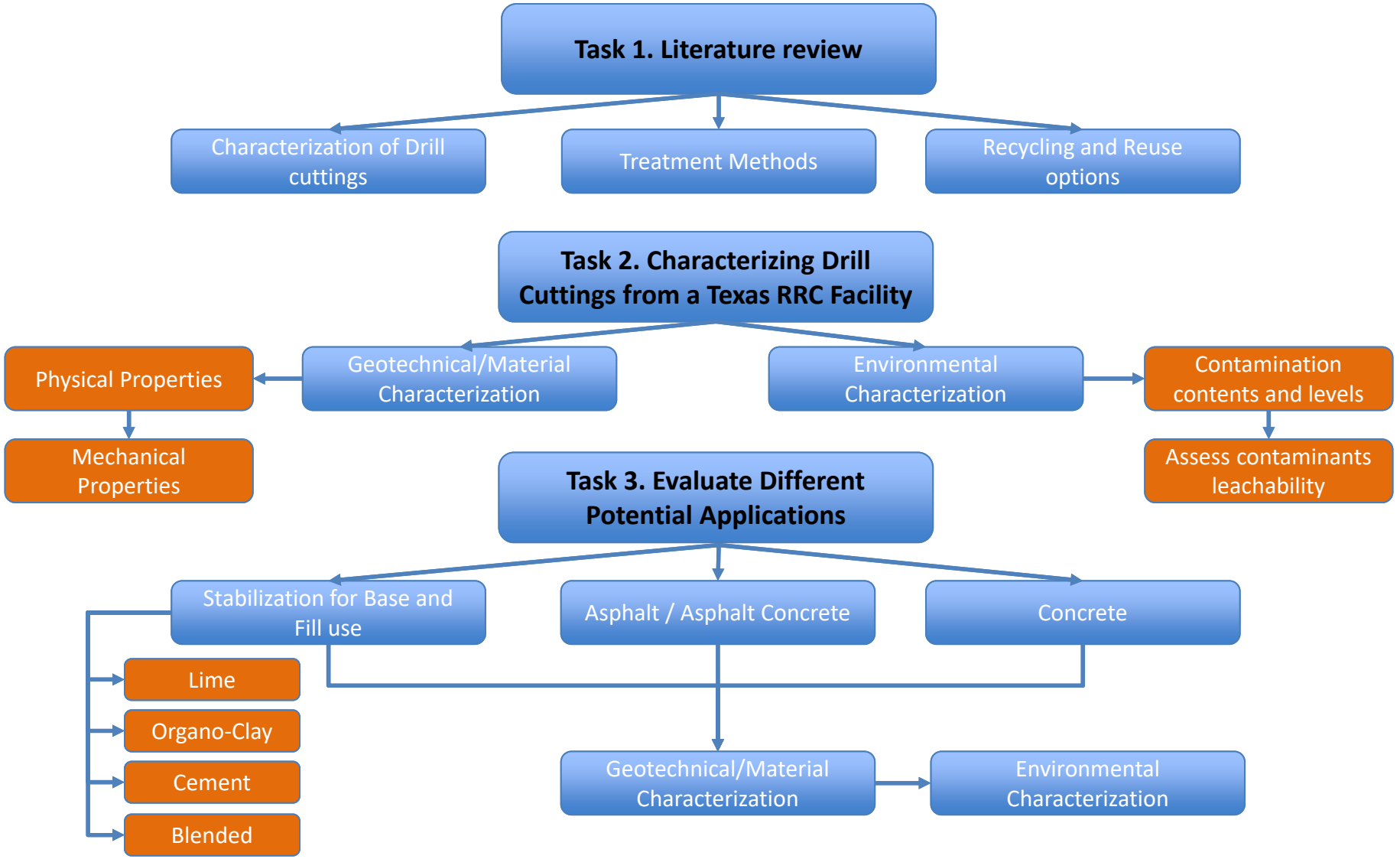
Karen Mena Arango

Introduction

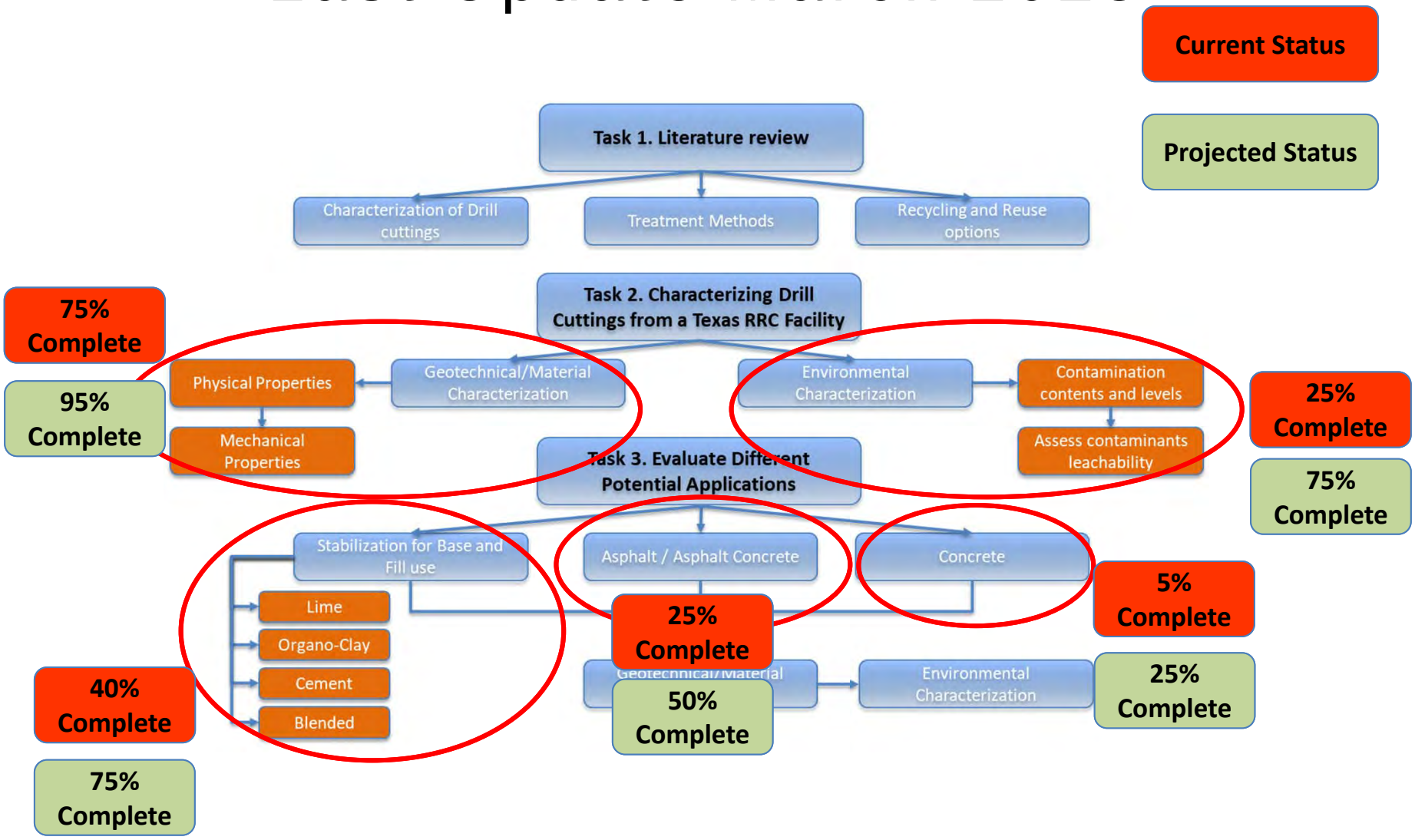
- Drill cuttings and spent drilling fluids are the major drilling wastes generated in greatest volumes during well installation
- In Texas, oil field drill cuttings are stockpiled at multiple locations throughout the state.
- These drill cuttings have amassed to millions of cubic yards and pose environmental risks due to presence of various potential contaminants:
  - Metals
  - Benzene, toluene, ethyl benzene, and xylene (BTEX)
  - Polycyclic aromatic hydrocarbons (PAHs)
  - Naphthalene, phenanthrene, and dibenzothiophene (NPD)
  - Naturally occurring radioactive materials (NORM)
  - Potential carcinogens and mutagens

Water surrounding drill cutting pile

Work Plan


Task 1. Literature review
Characterization of Drill cuttings
Treatment Methods
Recycling and Reuse options
Task 2. Characterizing Drill Cuttings from a Texas RRC Facility
Physical Properties
Geotechnical/Material Characterization
Environmental Characterization
Contamination contents and levels
Mechanical Properties
Assess contaminants leachability
Task 3. Evaluate Different Potential Applications
Stabilization for Base and Fill use
Asphalt / Asphalt Concrete
Concrete
Lime
Organo-Clay
Cement
Blended
Geotechnical/Material Characterization
Environmental Characterization

Last Update March 2023

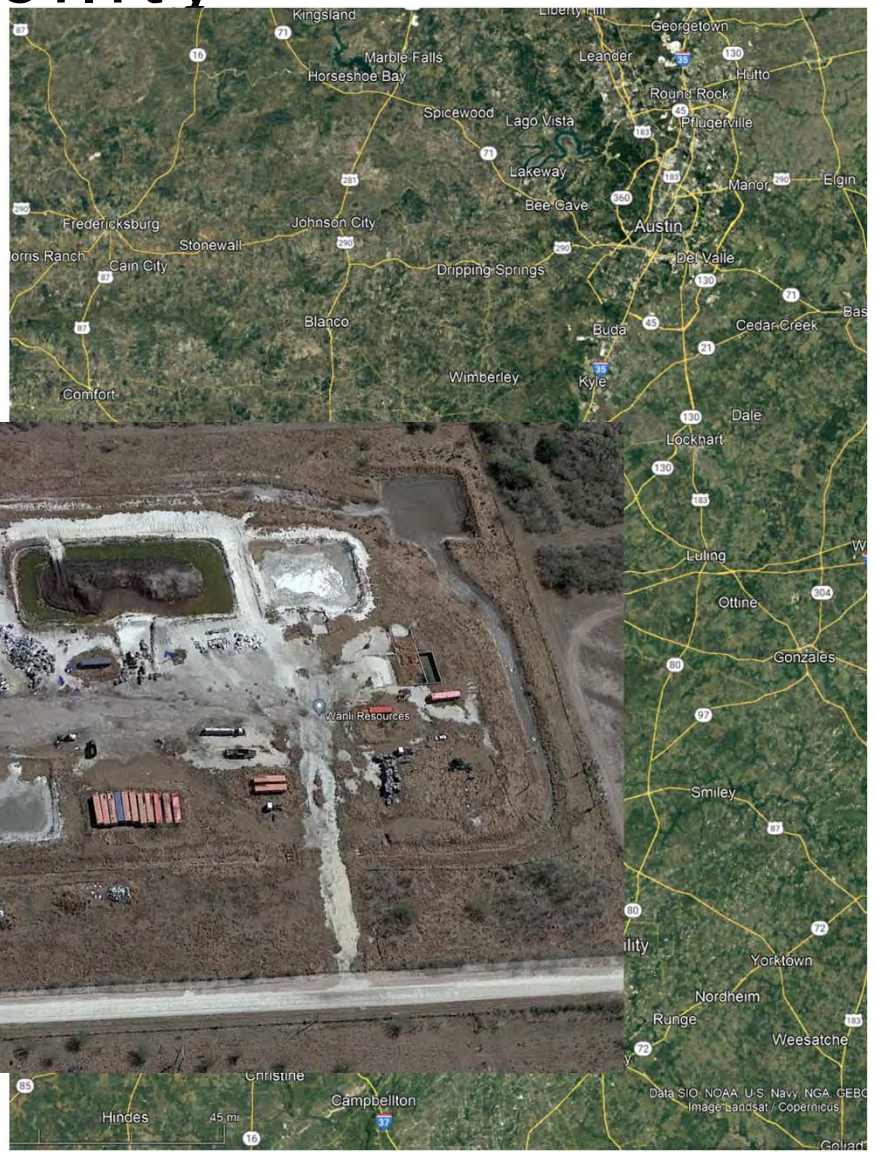


Material Characterization

POLK FACILITY

Polk Facility

- Polk site (Near Falls City)
  - Mostly untreated cuttings
  - Small treated stockpile!!

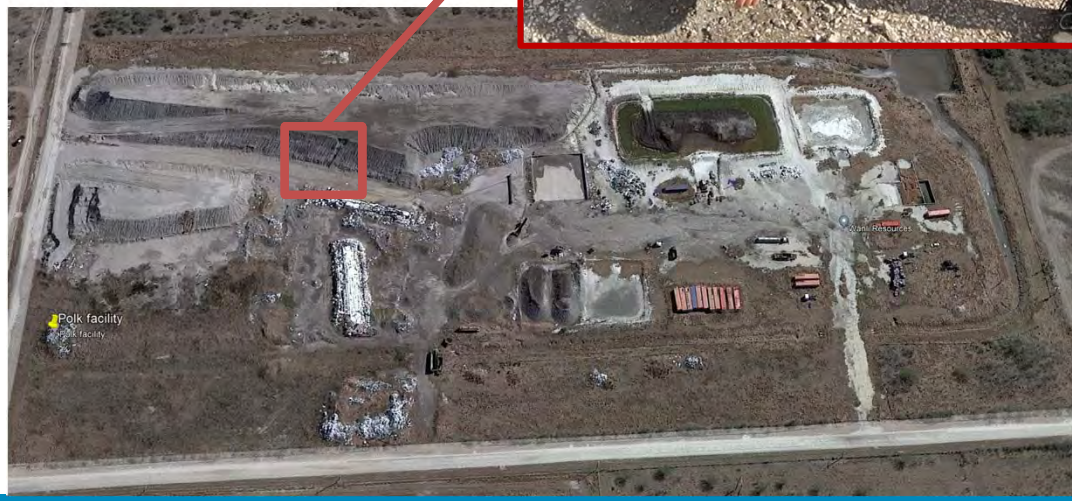


Polk Facility

- Initial site visit (June 29th, 2022)
  - Walked facility
  - Obtained small samples for initial characterization

Polk **Facility**

- Second site visit (July 22nd, 2022)
  - Obtained 30 5-gallon buckets
  - Homogenized in the lab



Polk Facility

RAW MATERIAL CHARACTERIZATION

Raw Material Characterization

Geotechnical Testing	Environmental Testing
Water Content	Ignition Testing
pH	X-ray Fluorescence
Specific Gravity	X-ray Diffraction
Atterberg Limits	Thermogravimetric Analysis
Particle Size Distribution	Leaching Testing
Compaction	Total Organic Content
Unconfined Compression Strength	ICP-MS
	SVOCs / VOCs
	Chloride Concentration
	Total Petroleum Hydrocarbons

Geotechnical/Material Characterization

- Dark gray with a strong smell of petroleum
  - Larger clumps crumble with some pressure
- Initial water content: 21.2%

- Specific gravity: 2.63

Trial	1	2	3	Average
Specific Gravity	2.62	2.62	2.64	2.63

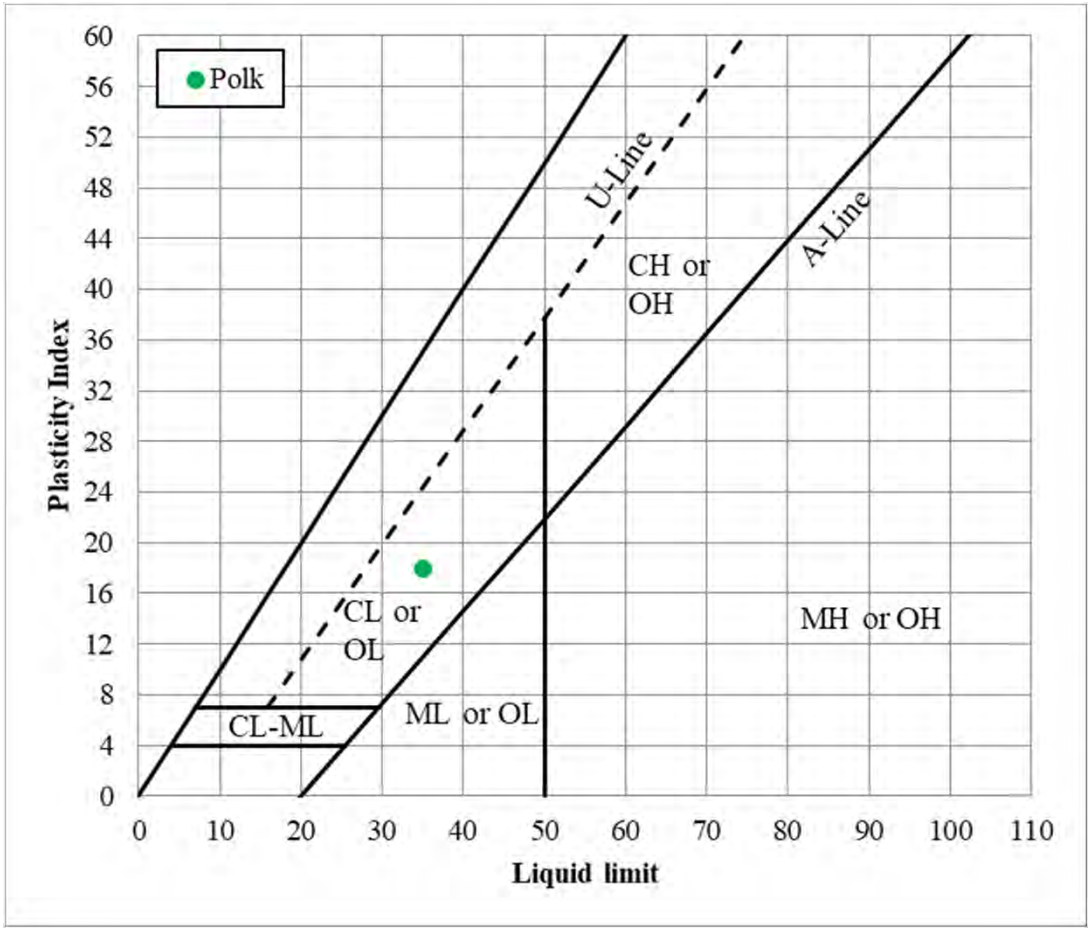
- Soil pH: 7.6

Trial	1	2	Average
pH	7.57	7.69	7.63
Temperature (°C)	24.4	24.6	24.5

Geotechnical/Material Characterization

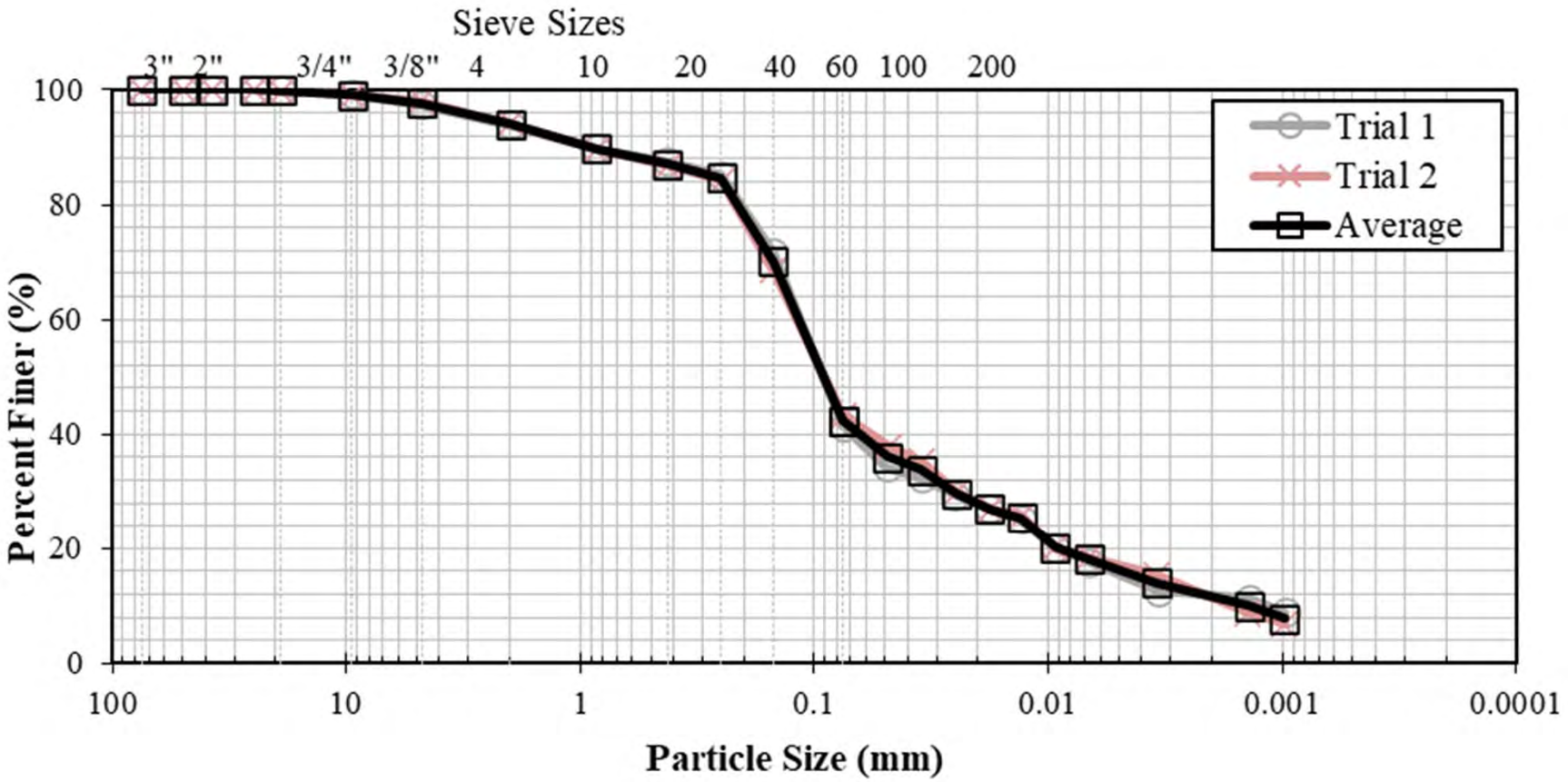
- Atterberg limits:

Trial	1	2	Average
LL(%)	34	35	35
PL(%)	16	17	17
PI(%)	18	18	18



Geotechnical/Material Characterization

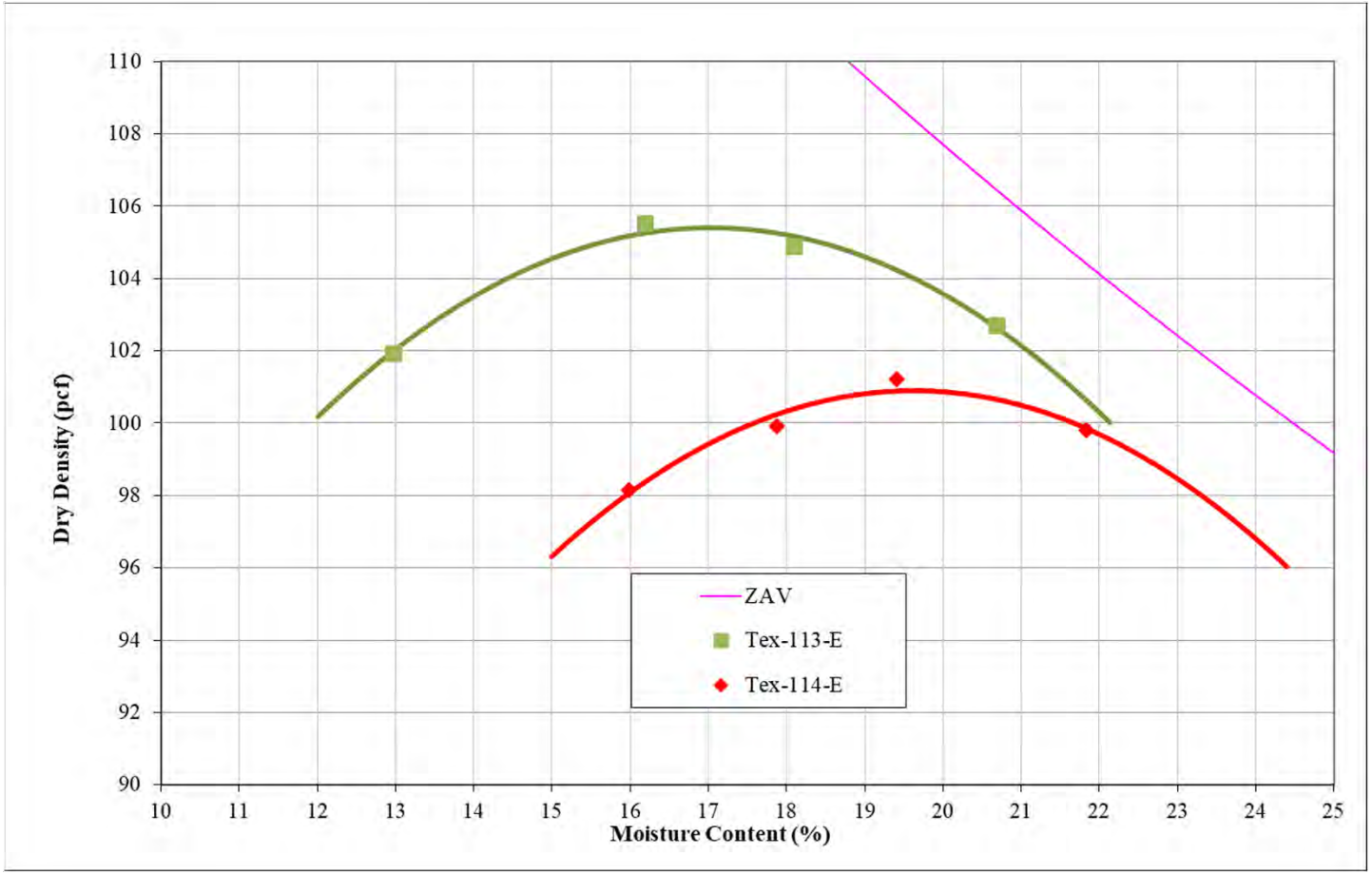
- Soil classification:
  - Clayey sand (SC)



Geotechnical/Material Characterization

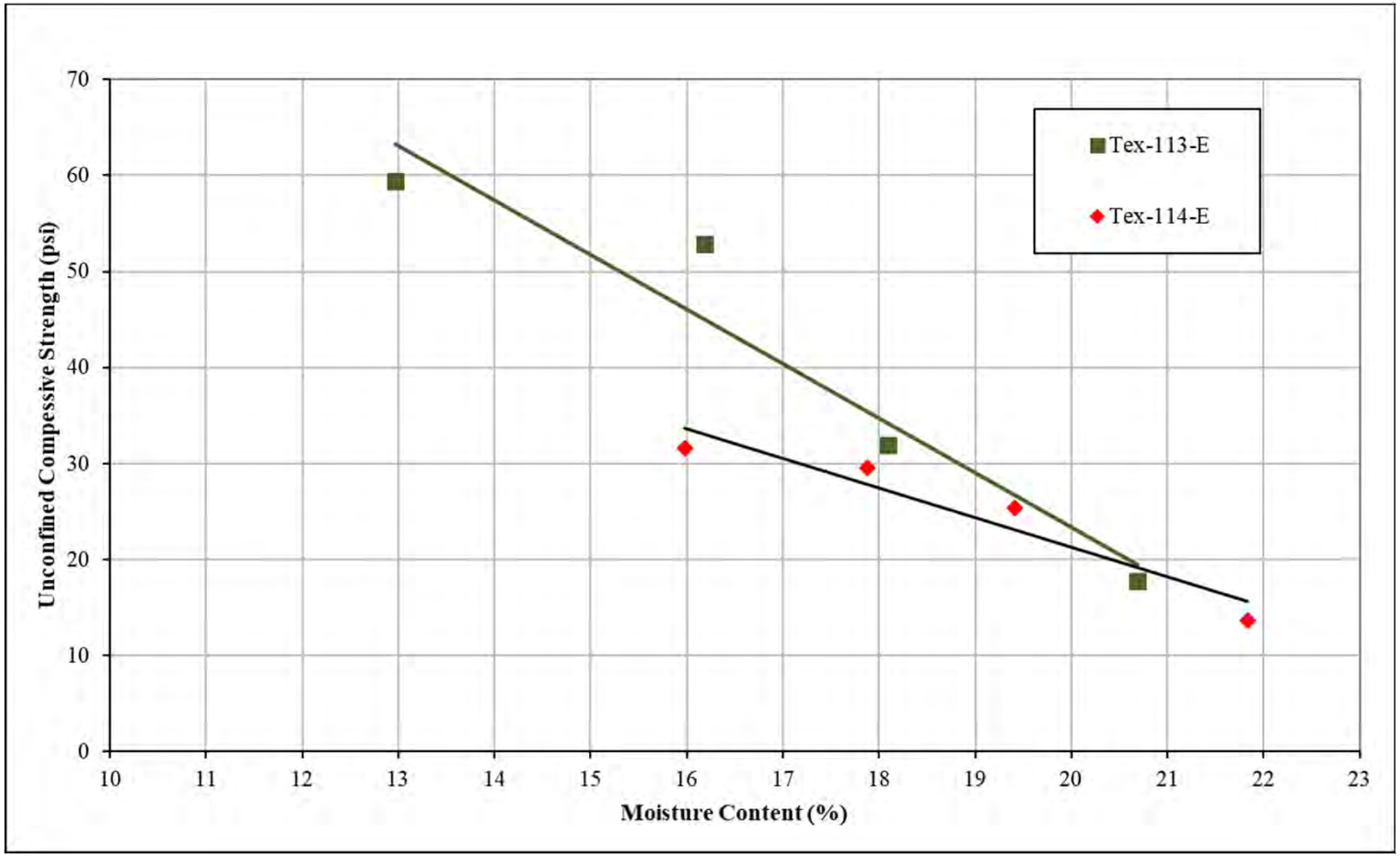
- Proctor Results

	Tex-113-E	Tex-114-E
Maximum Dry Density (pcf)	105.4	100.9
Optimum Water Content (%)	17.0	19.6



Geotechnical/Material Characterization

- Unconfined Compression for Tex-114-E and Tex-113-E



Environmental Characterization

- Moisture Content and Loss of Ignition

Material	Moisture Content (%)	LOI (%)
AA1010	26.4 ± 0.82	13.4 ± 0.27
AA1011	30.5 ± 0.75	15.6 ± 0.18
Polk	27.3 ± 0.91	9.6 ± 0.14

- Total Organic Carbon

Material	NPOC (ppm)	pH
P-1	19.61 ± 0.090	7.82 ± 0.03
P-2	17.53 ± 0.222	7.83 ± 0.03

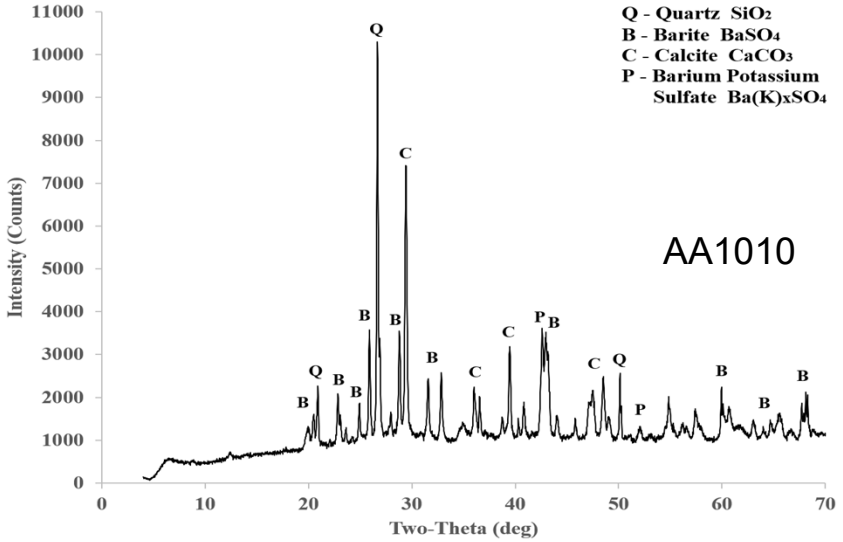
Environmental Characterization

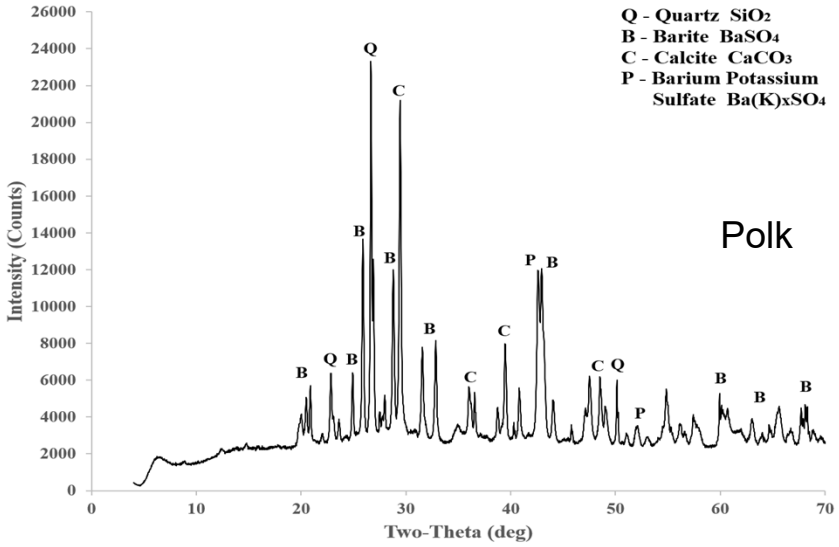
- Oxide Composition

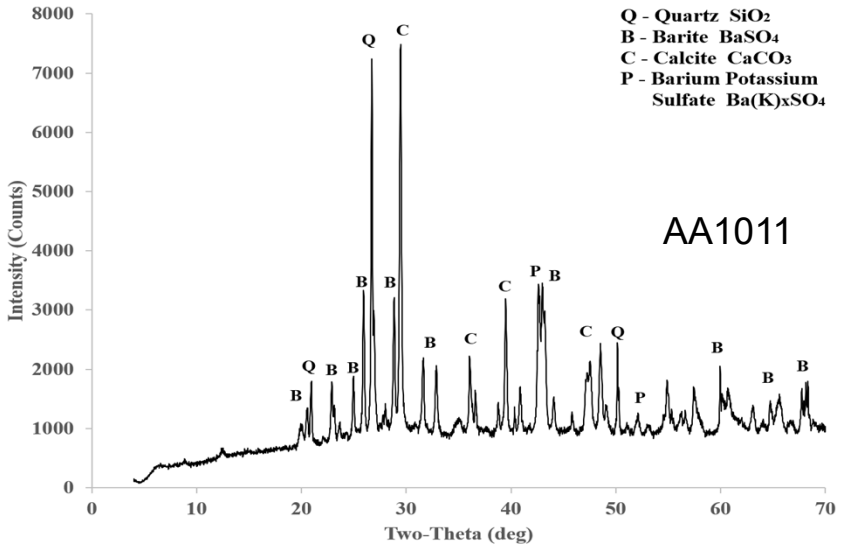
Component	Mass %		
	AA1010	AA1011	Polk
Na_2O	0.6936	0.6177	0.8398
MgO	1.6250	1.5481	1.4697
Al_2O_3	12.5229	11.5336	11.5329
SiO_2	41.4785	38.0155	42.6109
P_2O_5	0.2040	0.2123	0.1667
SO_3	4.8261	5.2476	7.9659
Cl	0.2176	0.1334	0.1922
K_2O	1.6665	0.5549	1.6243
CaO	20.8779	24.9648	17.4801
TiO_2	0.5276	0.4929	0.3997
Cr_2O_3	0.0232	0.0000	0.0187
MnO	0.0912	0.0767	0.0511
Fe_2O_3	4.8912	4.7305	3.1644
NiO	0.0070	0.0104	0.008
CuO	0.0261	0.0270	0.0167
ZnO	0.0305	0.0333	0.014
Br	0.0023	0.0000	0.0037
Rb_2O	0.0067	0.0059	0.0087
SrO	0.4301	0.5169	0.4972
Y_2O_3	0.0000	0.0000	0.0000
ZrO_2	0.0041	0.0000	0.0079
I	0.0345	0.0299	-
BaO	9.7860	10.2223	11.9156
PbO	0.0266	0.0263	0.0121

Environmental Characterization

- Qualitative Phase Analysis
  - Identified crystalline phases:
    - Quartz, SiO_2
    - Barite, $BaSO_4$
    - Calcite, $CaCO_3$
    - Barium Potassium Sulfate, $Ba(K)_xSO_4$



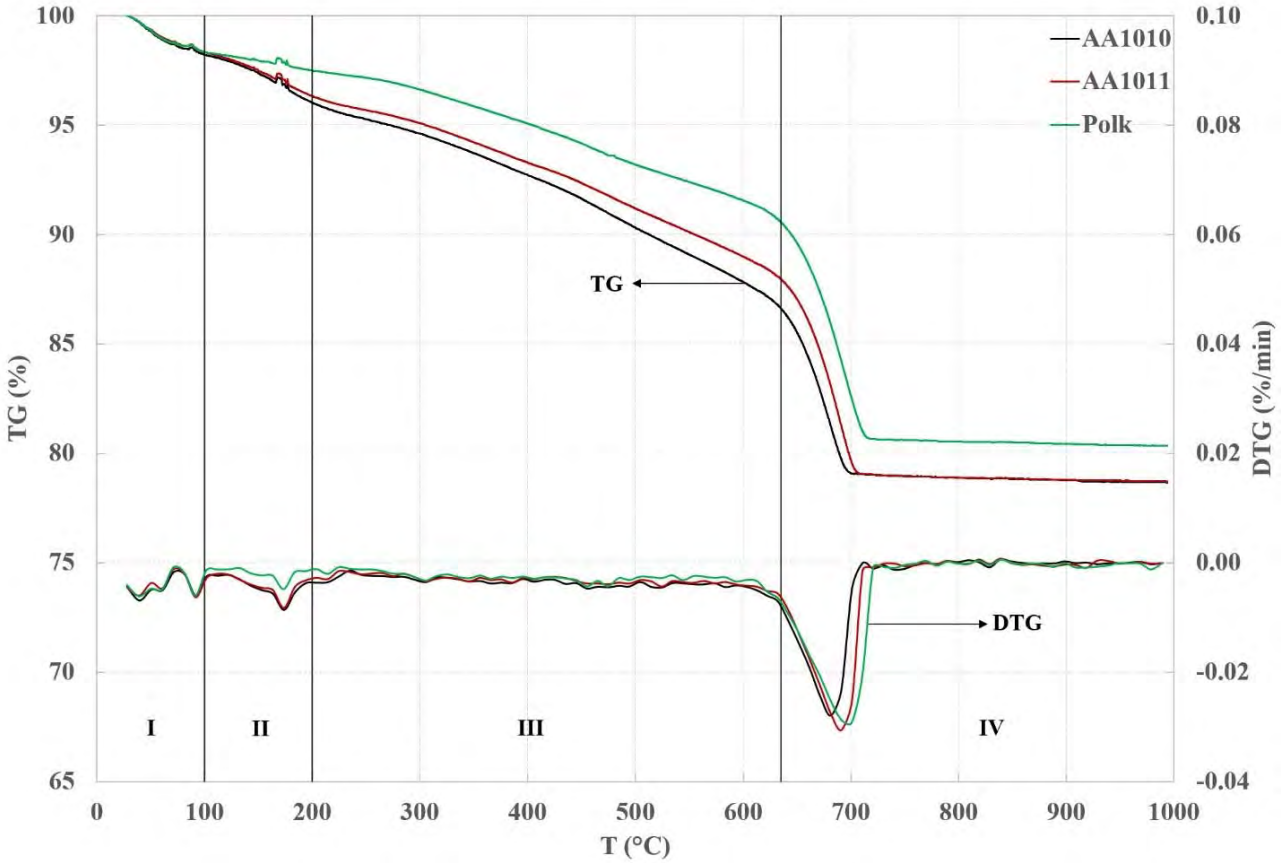




Environmental Characterization

- Thermogravimetric Analysis

- Stage I
  Loss of absorbed moisture
- Stage II
  Desorption of adsorbed water
- Stage III
  Thermal decomposition of organic matter
- Stage IV
  Decomposition of organic matte and the dehydroxylation of clay minerals, or decomposition of calcite



TGA results of drill cuttings collected using nitrogen purge gas. Data were collected using a Mettler-Toledo TGA/DSC 1, 50 mL/min of N_2 and a heating rate of 10 °C/min.

Environmental Characterization

- Trace Metal Concentrations

- Microwave digested drill cuttings (EPA Method 3050B)
- ICP-MS (Standard Method 3125)
- Trace metal grade nitric acid was used for dilution and sample preparation

Element	AA1010	AA1011	Polk
Li	382	304	345
Na	61600	27300	23600
Mg	14200	23800	19700
Al	30800	65400	51800
K	15600	17600	16100
Ca	30600	56900	44900
Cr	1150	112	403
Fe	51200	98800	96800
Co	20.1	34.1	32.4
Ni	286	150	1430
Cu	332	386	483
Zn	3750	1570	1640
As	75.7	102	124
Se	20.4	16.8	17.7
Sr	9810	9460	7790
Cd	5.81	5.94	5.81
Ba	75100	79700	85700
Pb	181	501	553

Environmental Characterization

- Semivolatile Organic Compounds
  - EPA Method 8270E
  - Gas Chromatography / Mass Spectrometry (GC-MS)
  - SVOCs included, but not limited to, the 16 priority-pollutant PAHs
  - Polk-u1- not homogenized
  - Polk-h2 and Polk-h3 - homogenized stockpile

Detection Summary

Material	Analyte	Result	Qualifier	RL	MDL	Unit	Dil Fac	Method
Polk-u1	Pyrene	0.0387	J	0.167	0.0146	ppm	1	8270E
Polk-h2	Pyrene	0.213	J	1.66	0.146	ppm	10	8270E
Polk-h3	Pyrene	0.228	J	1.66	0.146	ppm	10	8270E
LRA	No detections							
RAP	No detections							

RL: Reporting Limit, MDL: Method Detection Limit, Dil Fac: Dilution Factor
J: result is less than the RL but greater than or equal to the MDL and the concentration is an approximate value

Environmental Characterization

- Volatile Organic Compounds
  - EPA Method 8260D
  - Gas Chromatography / Mass Spectrometry (GC-MS)
  - Analytes included, but not limited to, benzene, toluene, ethylbenzene, xylene, and vinyl chloride

Detection Summary

Material	Analyte	Result	Qualifier	RL	MDL	Unit	Dil Fac	Method
Drill Cuttings-1	Xyelens, Total	0.000458	J	0.00109	0.000235	mg/kg	1	8260D
	m,p-Xylenes	0.000458	J	0.00109	0.000235	mg/kg	1	8260D
Drill Cuttings-2	Xyelens, Total	0.000298	J	0.00101	0.000219	mg/kg	1	8260D
	m,p-Xylenes	0.000298	J	0.00101	0.000219	mg/kg	1	8260D
Drill Cuttings-3	Xyelens, Total	0.000458	J	0.00104	0.000224	mg/kg	1	8260D
	m,p-Xylenes	0.000458	J	0.00104	0.000224	mg/kg	1	8260D

RL: Reporting Limit, MDL: Method Detection Limit, Dil Fac: Dilution Factor
J: result is less than the RL but greater than or equal to the MDL and the concentration is an approximate value

Material Performance after Treatment

STABILIZATION FOR FILL APPLICATIONS

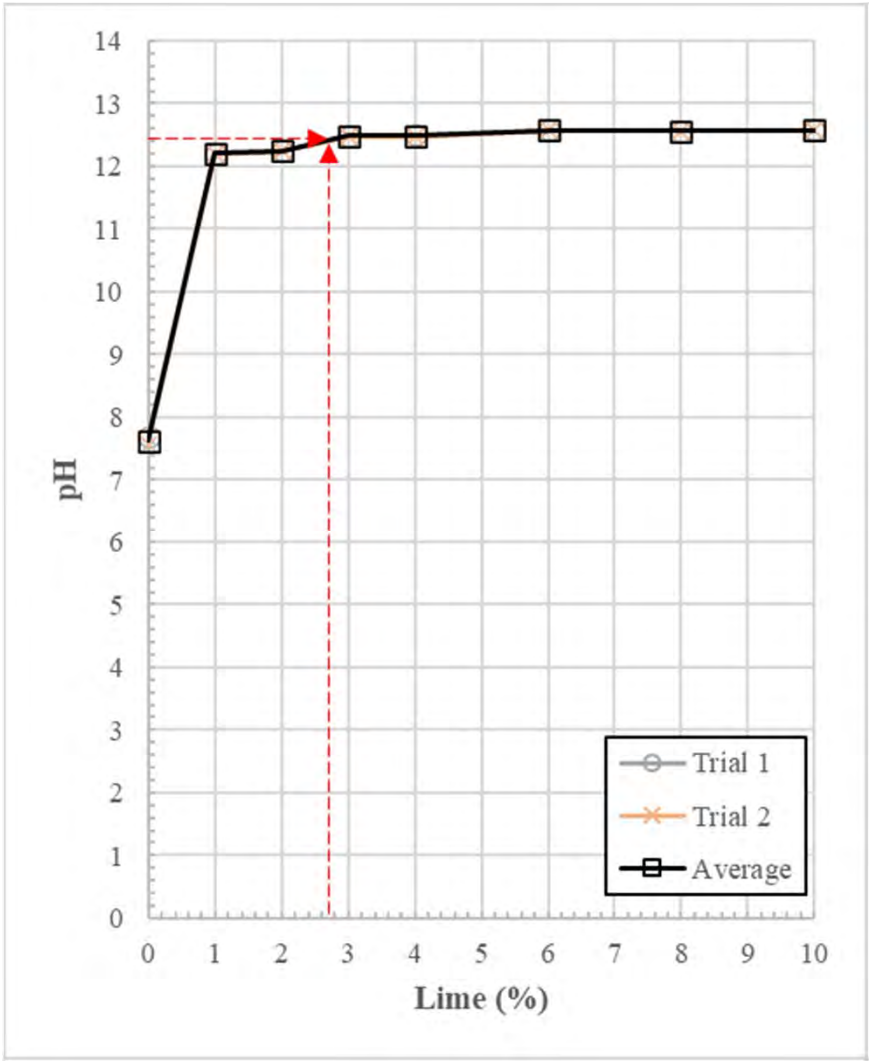
Stabilization for Fill Applications

- Potential treatments
  - Hydrated lime
  - Organoclay
  - Cement

Stabilization with Admixtures-Lime

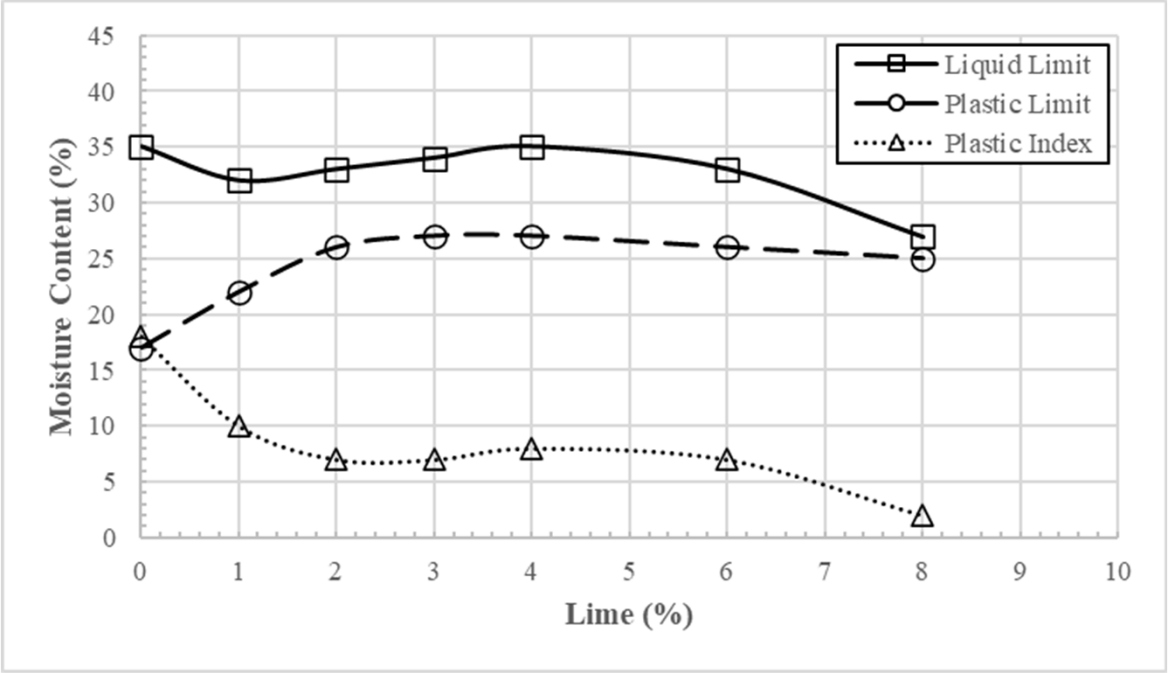
- Soil-lime pH:
  - Recommended: 2.7%

Lime (%)	pH	Temp (°C):
0	7.63	24.5
1	12.21	24.9
2	12.24	24.6
3	12.49	24.9
4	12.49	24.9
6	12.57	23.7
8	12.56	24.3
10	12.57	24.1



Stabilization with Admixtures-Lime

- Soil-lime Atterberg:

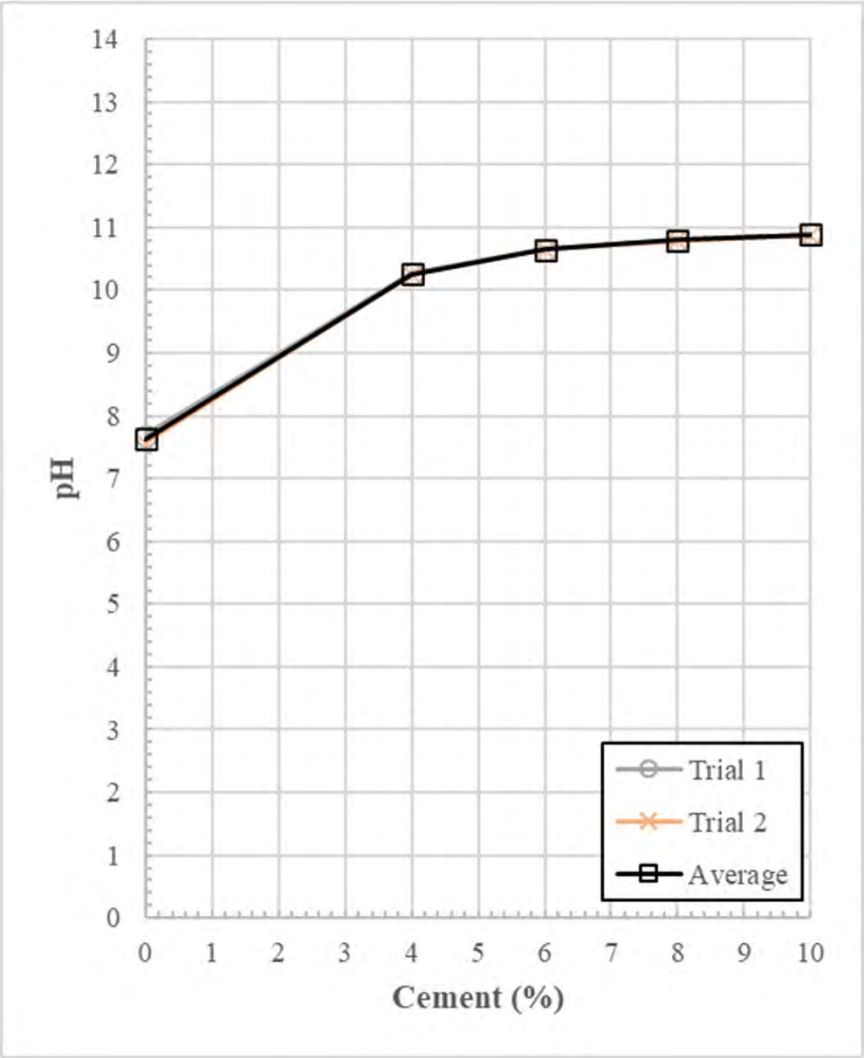


Lime (%)	0	1	2	3	4	6	8
Liquid Limit (%)	35	32	33	34	35	33	27
Plastic Limit (%)	17	22	26	27	27	26	25
Plastic Index (%)	18	10	7	7	8	7	2

Stabilization with Admixtures-Cement

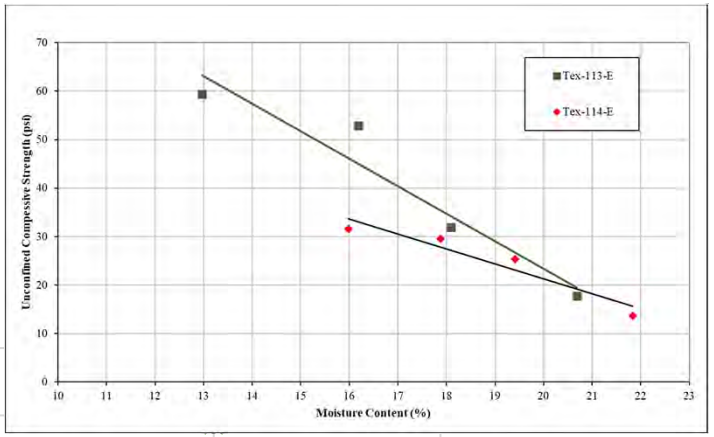
- Soil-cement pH:

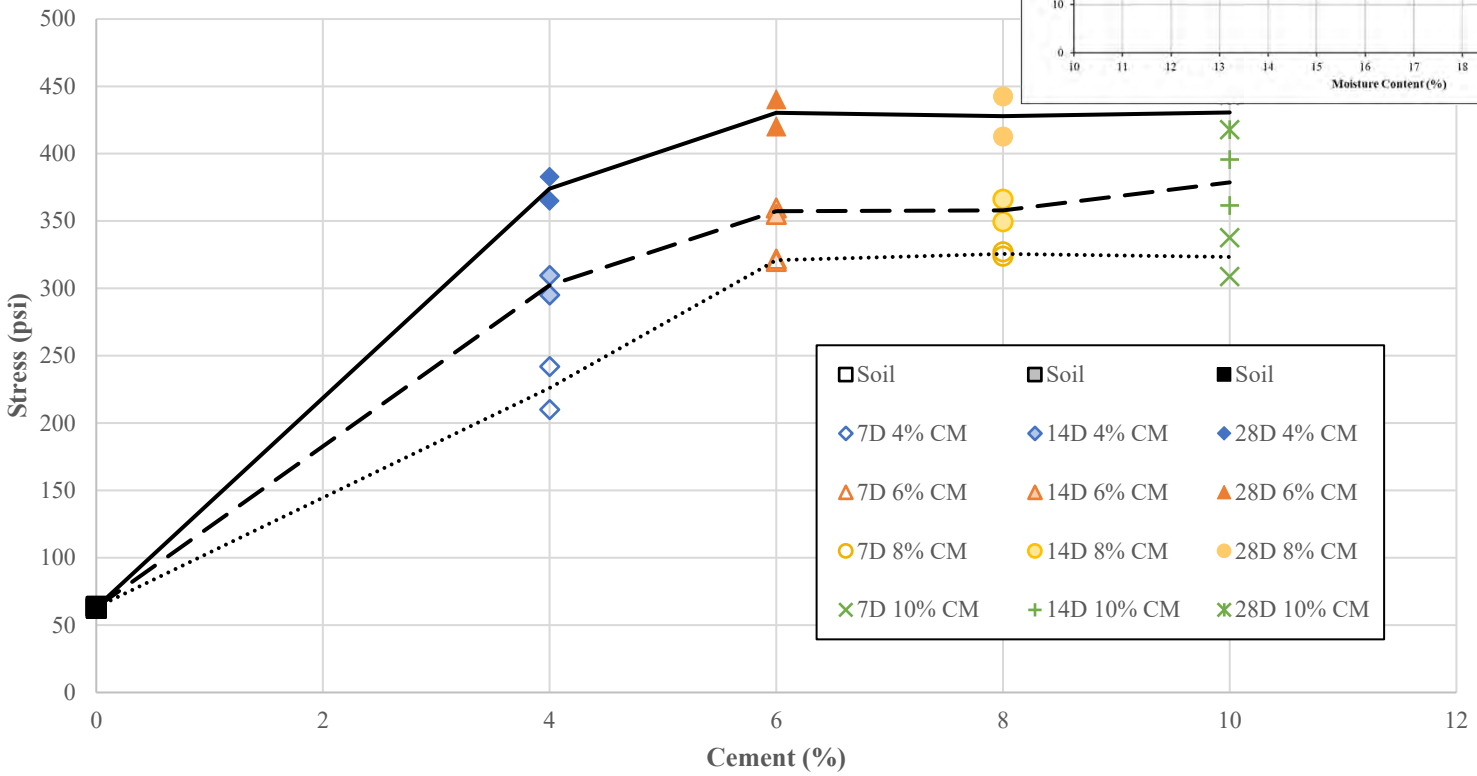
Cement (%)	pH	Temp (°C):
0	7.63	24.7
4	10.26	24.7
6	10.65	24.7
8	10.80	24.7
10	10.89	24.7



Stabilization with Admixtures-Cement

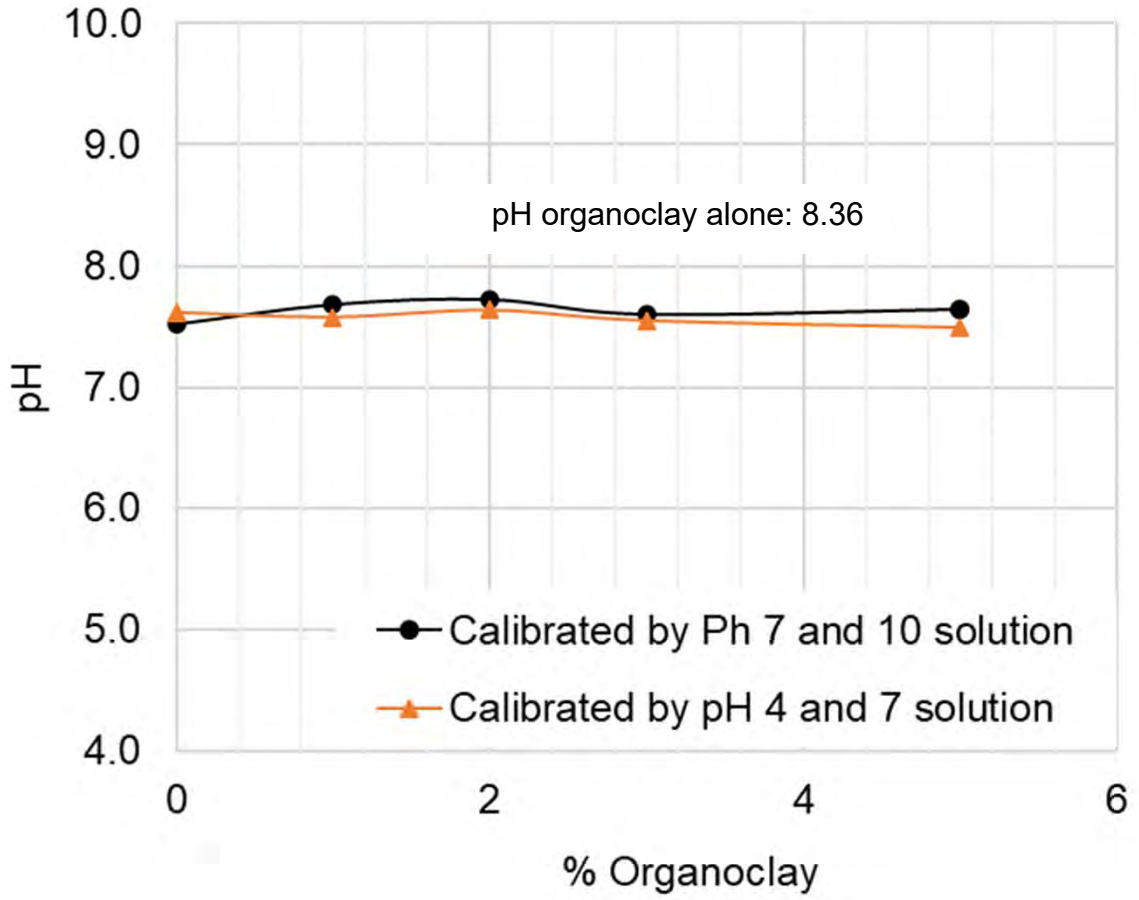
- Soil-cement Strength:





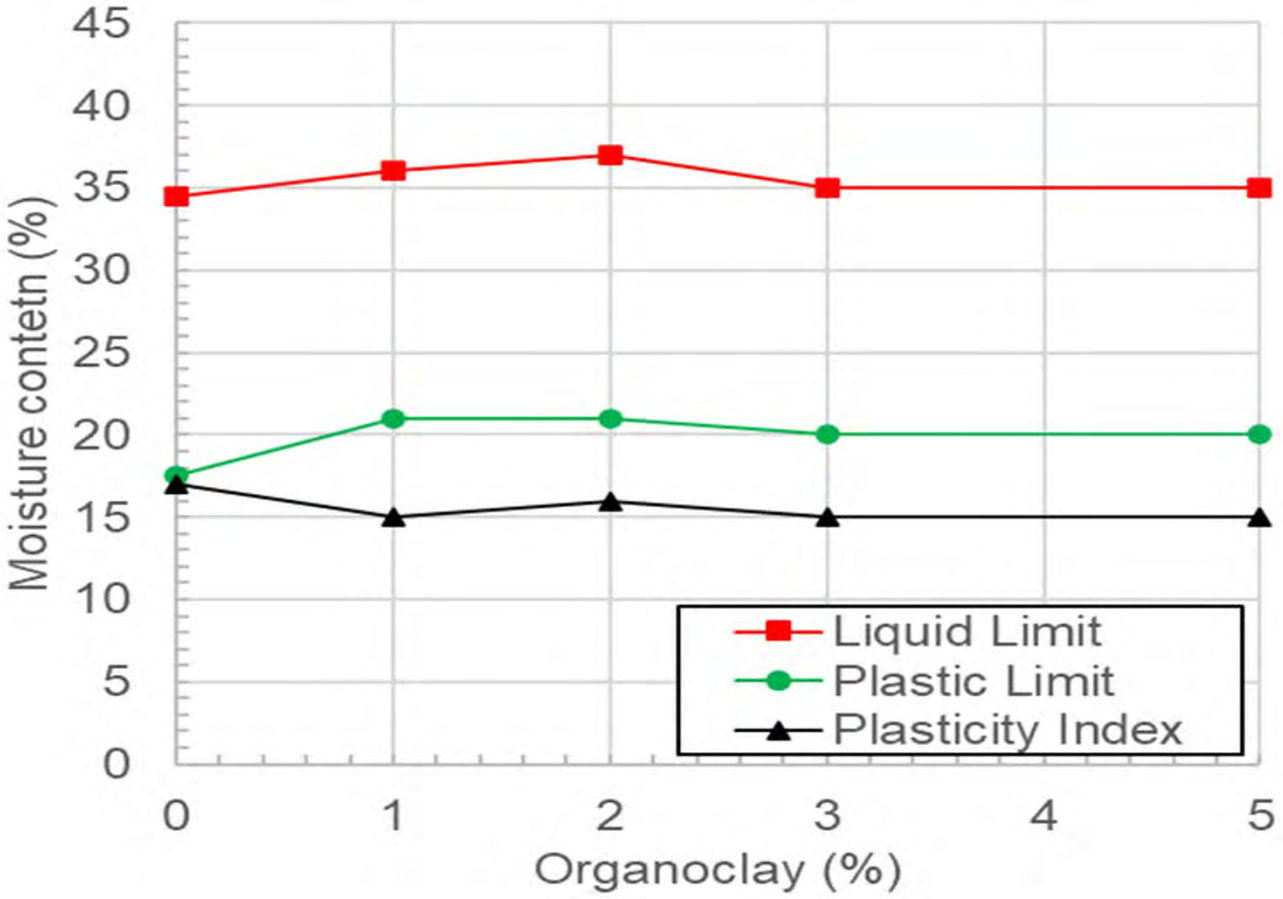
Stabilization with Admixtures- Organoclay

- Soil-OrganoClay pH:



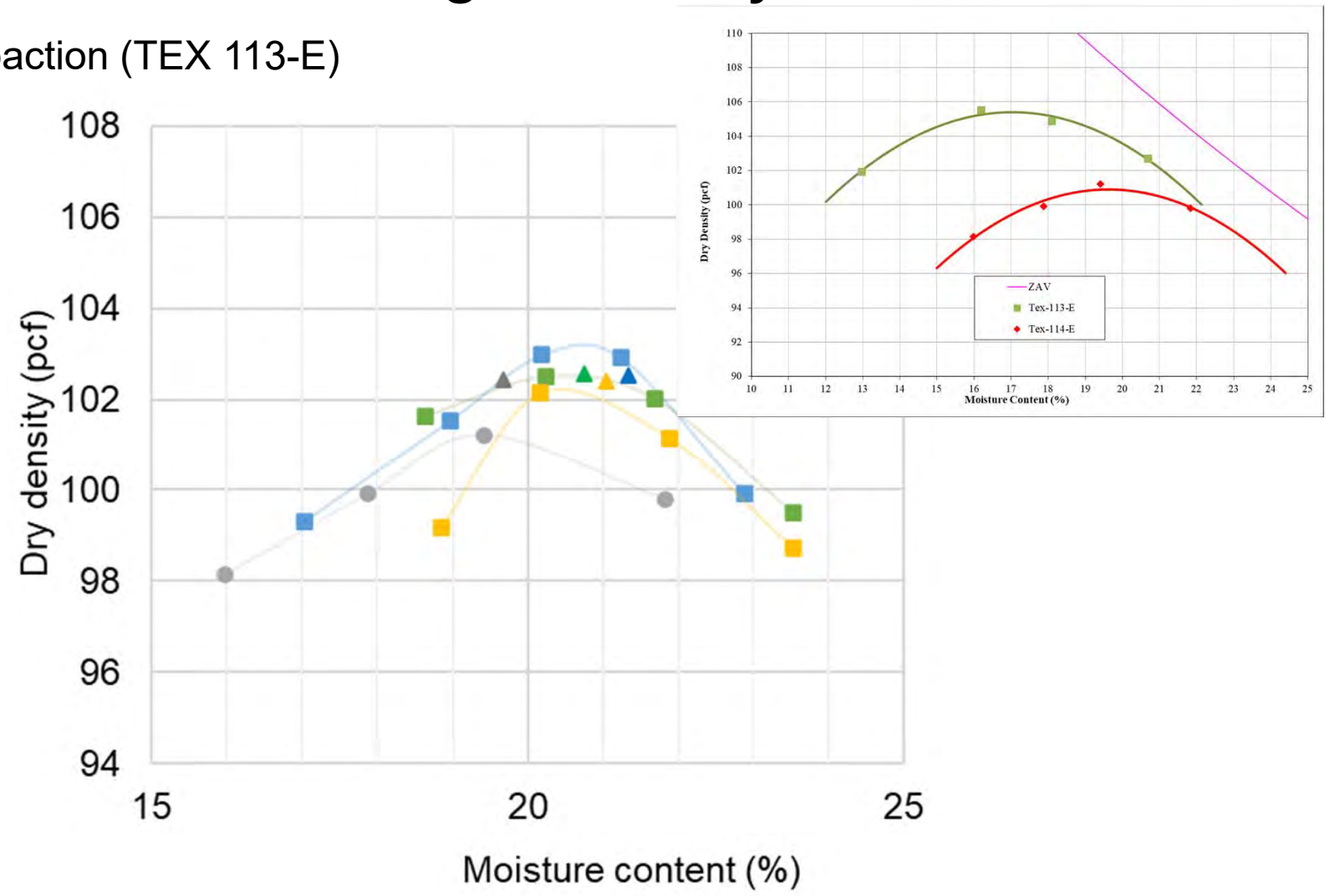
Stabilization with Admixtures- Organoclay

- Atterberg Limits



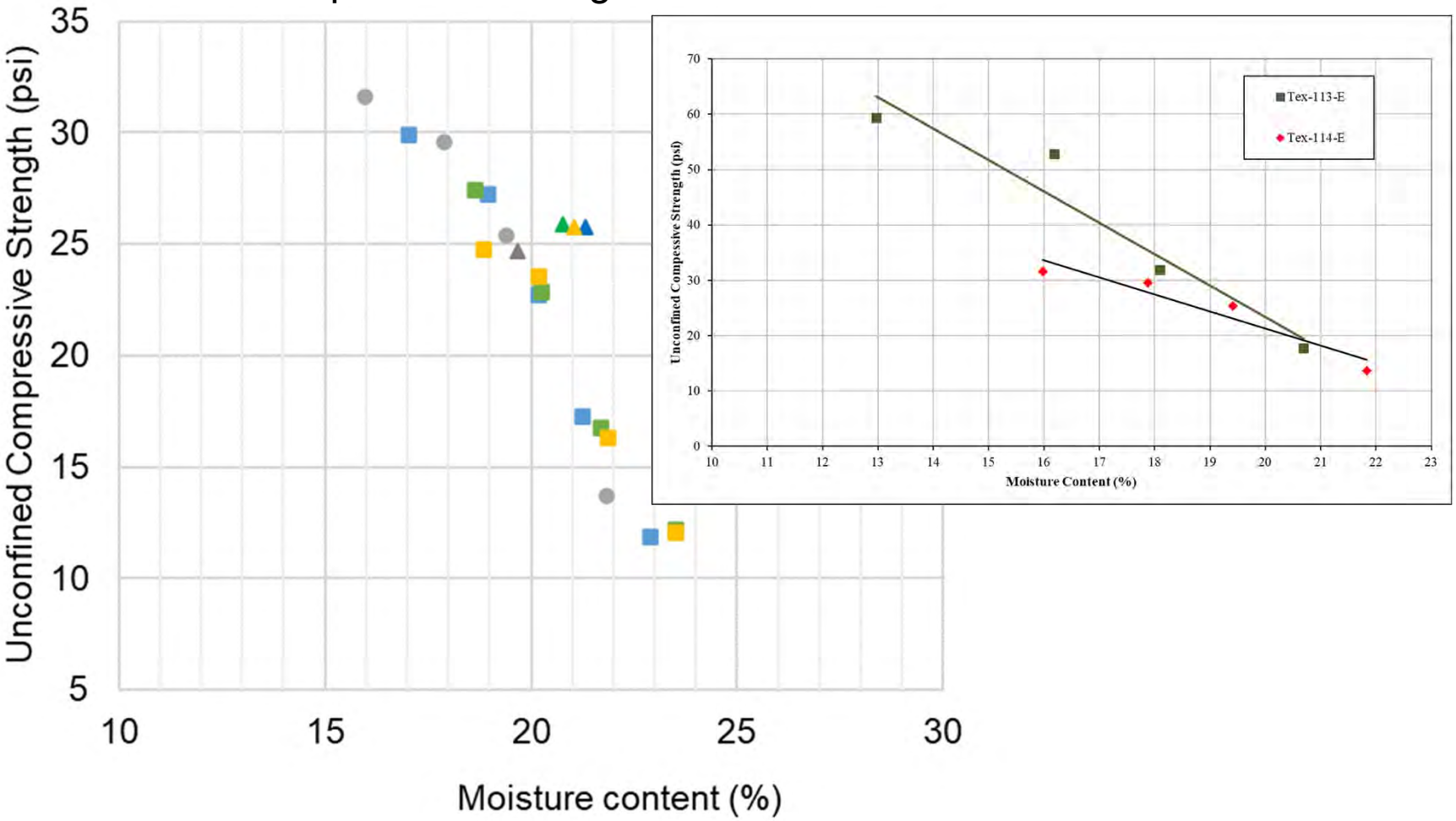
Stabilization with Admixtures- Organoclay

- Compaction (TEX 113-E)



Stabilization with Admixtures- Organoclay

- Unconfined Compressive Strength tests



Stabilization with Admixtures- Summary

- Lime:
  - Lime treatment may not work well
  - pH curve is not a gradual increase like typically seen
  - Low Plastic Index (may not enough clay minerals to provide aluminum to the reaction)

- Cement
  - Adding Cement can help increase strength of drill cuttings
  - Strength can be increased 7 times, from 60 psi to 420 psi.
  - Adding 6% cement to the drill cuttings is the optimal dosage from strength enhancement.

- OrganoClay
  - no obvious impact to the physical properties of drill cuttings

Material Performance after Treatment

STABILIZATION FOR BASE APPLICATIONS

Base Materials

- TxDOT Item 247: Flexible Base

Property	Test Method	Grade 1–2	Grade 3	Grade 4[2]	Grade 5
Sampling	Tex-400-A				
Master gradation sieve size (cumulative % retained)	Tex-110-E				
2-1/2"		0	0	As shown on the plans	0
1-3/4"		0–10	0–10		0–5
7/8"		10–35	–		10–35
3/8"		30–65	–		35–65
#4		45–75	45–75		45–75
#40		65–90	50–85		70–90
Liquid Limit, % Max	Tex-104-E	40	40	As shown on the plans	35
Plasticity Index, Max[1]	Tex-106-E	10	12	As shown on the plans	10
Plasticity index, Min[1]		As shown on the plans	As shown on the plans	As shown on the plans	As shown on the plans
Wet ball mill, % Max	Tex-116-E	40	–	As shown on the plans	40
Wet ball mill, % Max increase passing the #40 sieve		20	–	As shown on the plans	20
Min compressive strength, psi	Tex-117-E			As shown on the plans	
lateral pressure 0 psi		35	–		–
lateral pressure 3 psi		–	–		90
lateral pressure 15 psi		175	–		175

1. Determine plastic index in accordance with Tex-107-E (linear shrinkage) when liquid limit is unattainable as defined in Tex-104-E.
2. Grade 4 may be further designated as Grade 4A, Grade 4B, etc.

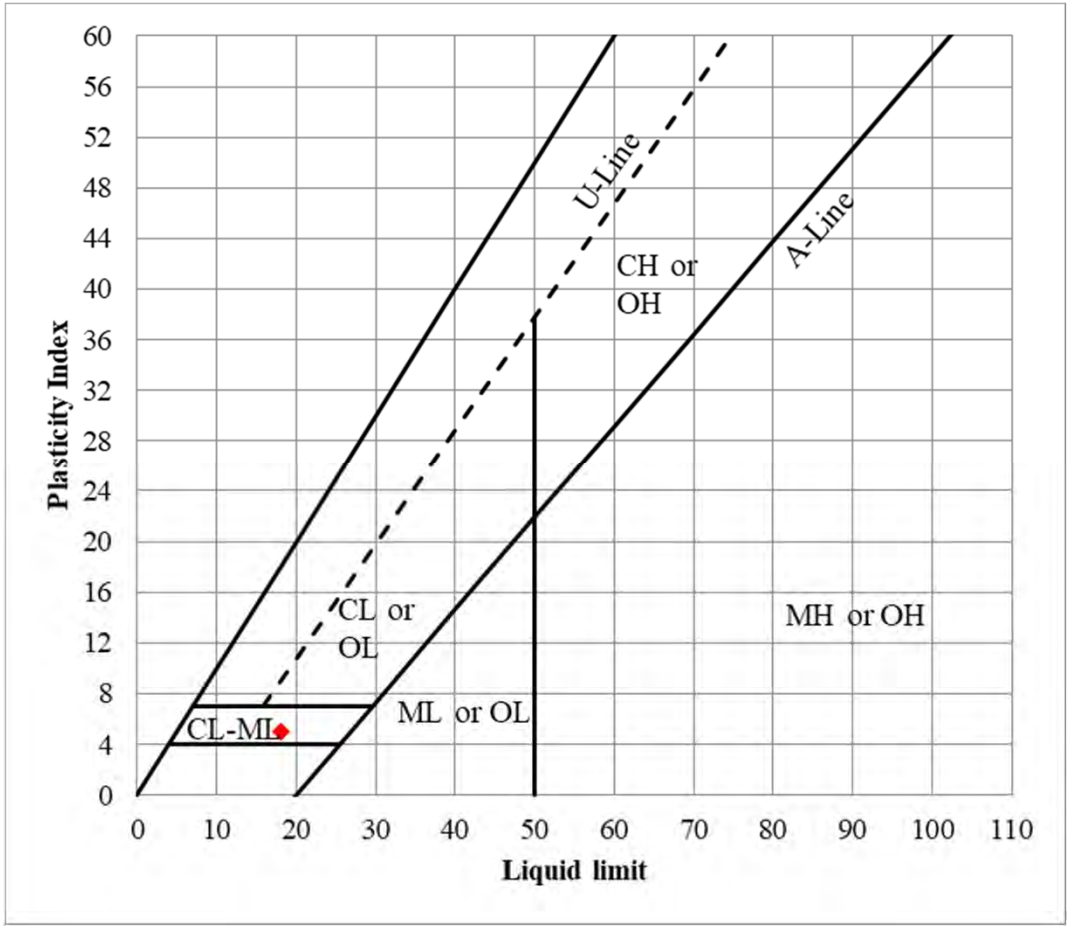
Raw Base Course Materials

- Marble Falls Site
  - Reddish tan
    - Hard clumps cant be easily broken
  - Initial water content: 3.0%
  - Base course material for roadway construction
  - Blend with drill cuttings

Raw Base Course Materials

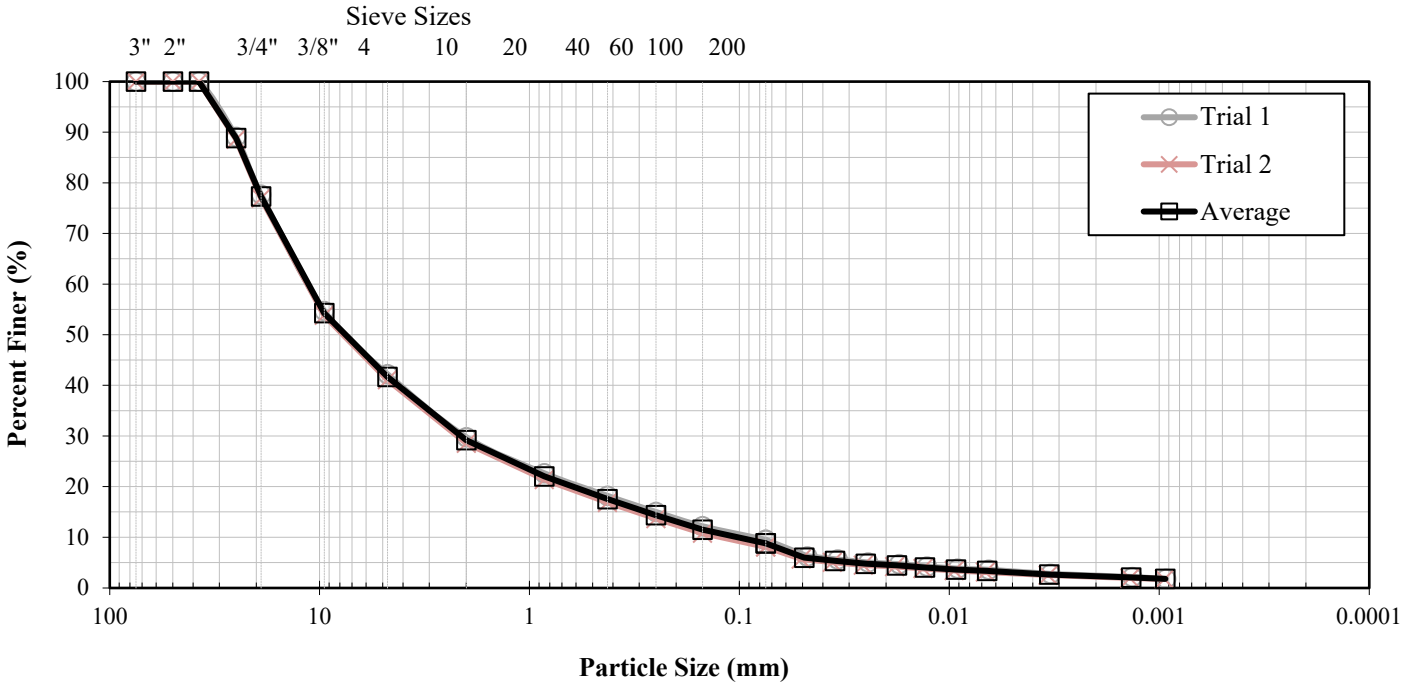
- Atterberg limits:

Liquid Limit (%)	18
Plastic Limit (%)	13
Plastic Index (%)	5



Raw Base Course Materials

- Soil classification:
  - Well-graded gravel with silt (GW-GM)

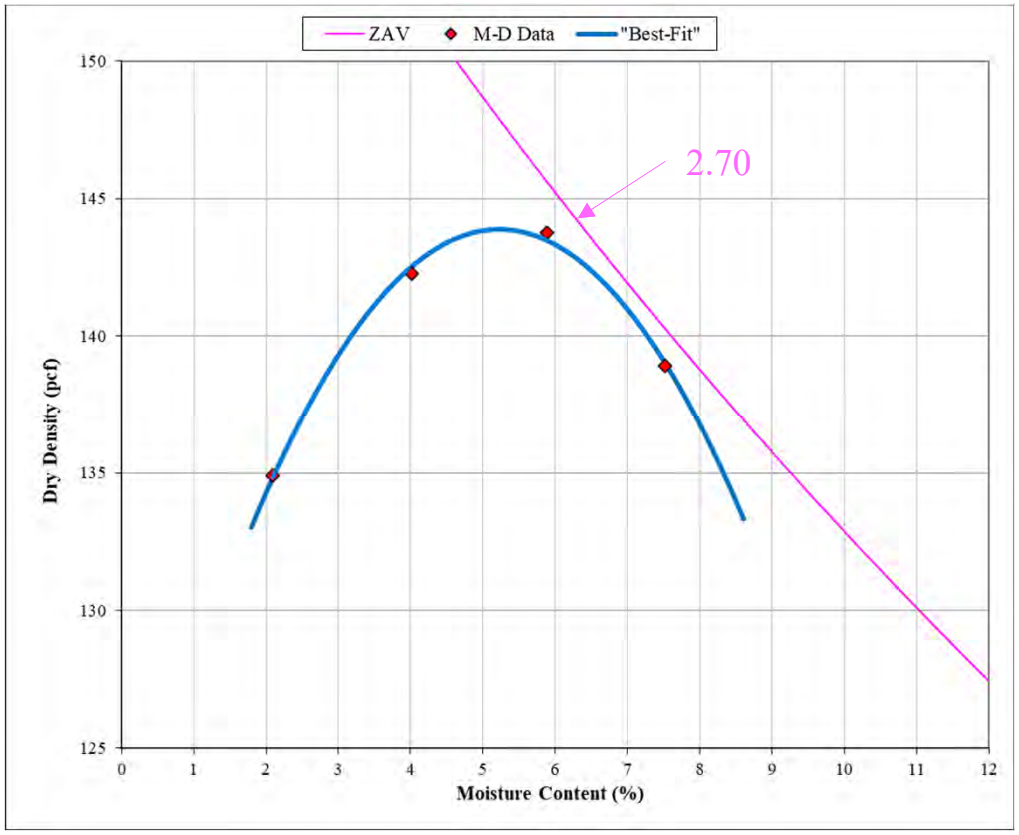


D10	0.089
D30	2.053
D60	11.700

Cu	131.4
Cc	4.0

Raw Base Course Materials

- Proctor (Tex-113-E)

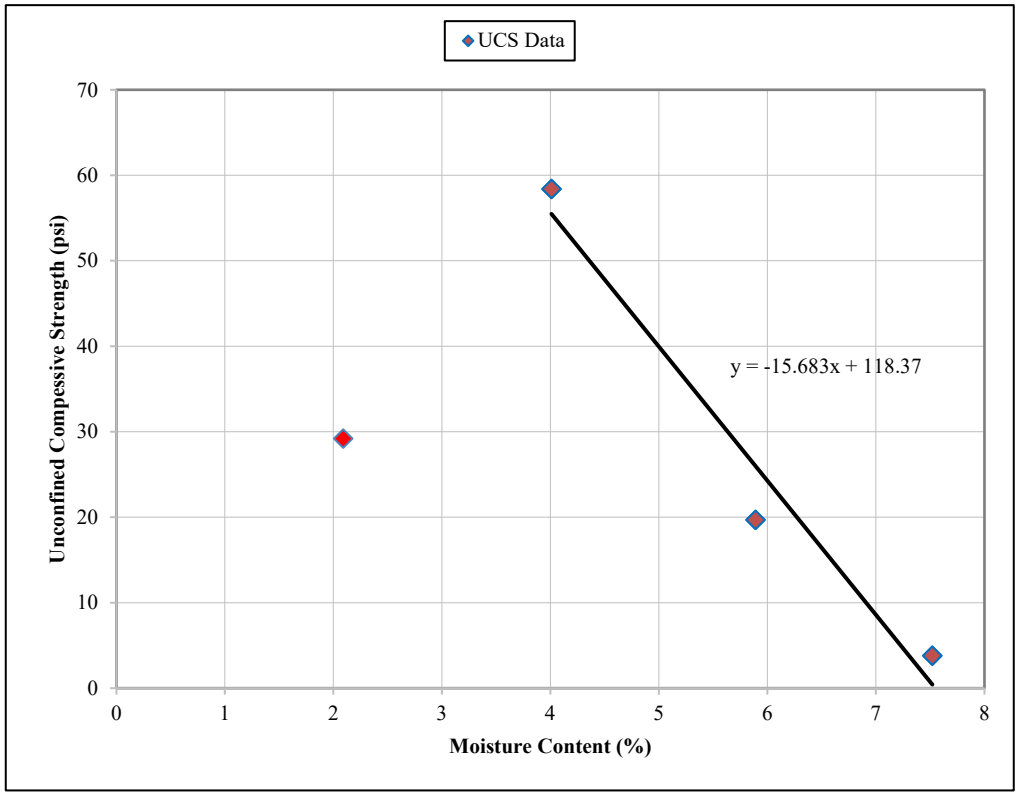


Water Content (%)	2.1	4.0	5.9	7.5
Dry Density (pcf)	134.9	142.3	143.8	138.9

Maximum Dry Density (pcf)	143.9
Optimum Water Content (%)	5.2
M-D Graph R^2	1.00

Raw Base Course Materials

- Unconfined Compression

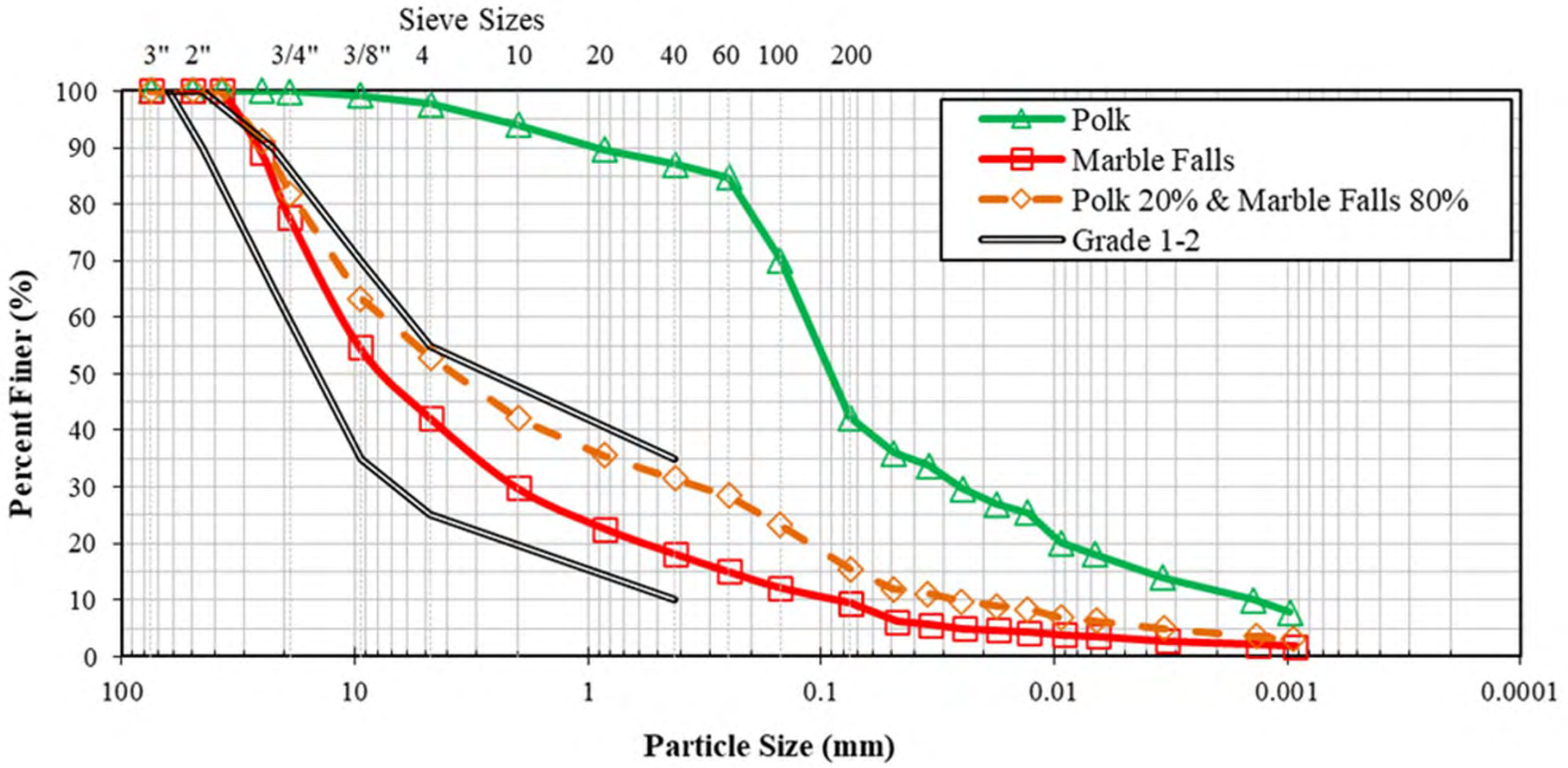


2.1% WC before test (too dry)

Water Content (%)	2.1	4.0	5.9	7.5
Unconfined Strength (psi)	29.8	59.6	20.3	3.9
Strain (%)	2.1	2.1	3.1	3.1

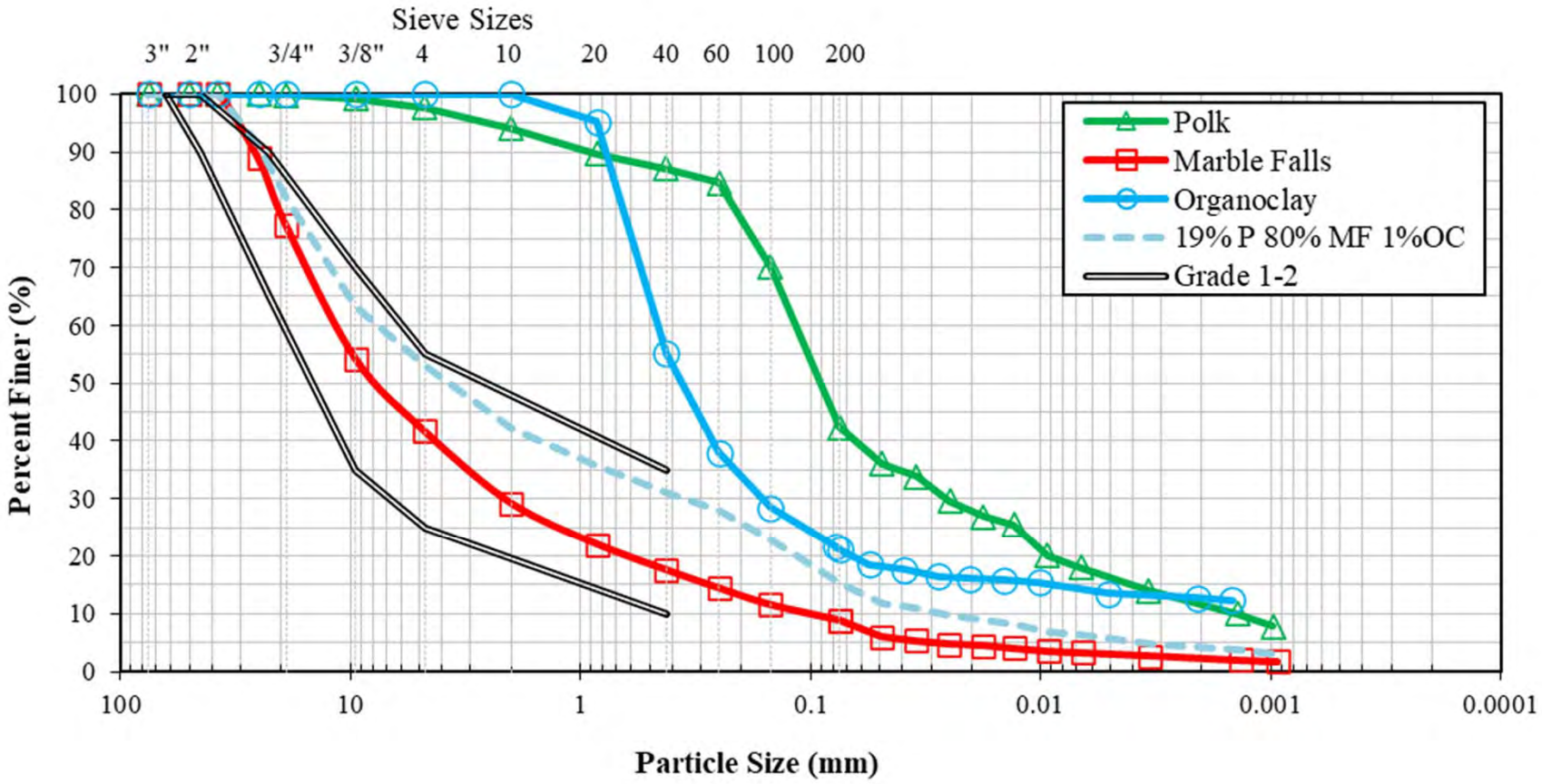
Blended Base Course Materials

- Blended gradation curve:



Blended Base Course Materials

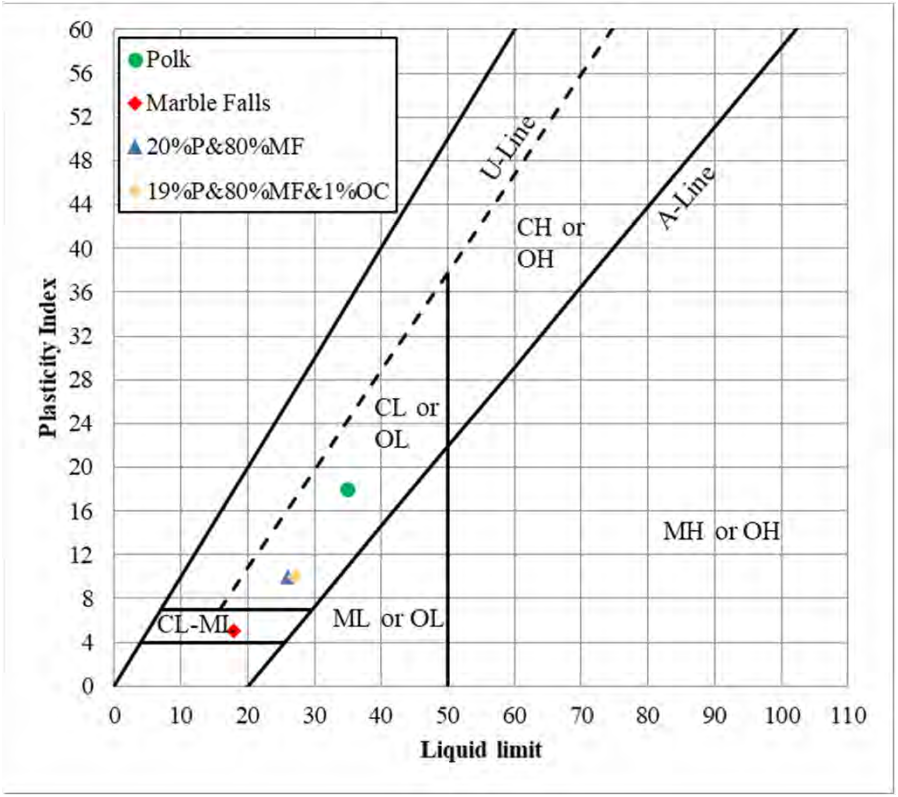
- Blended gradation curve (with Organoclay):



Blended Base Course Materials

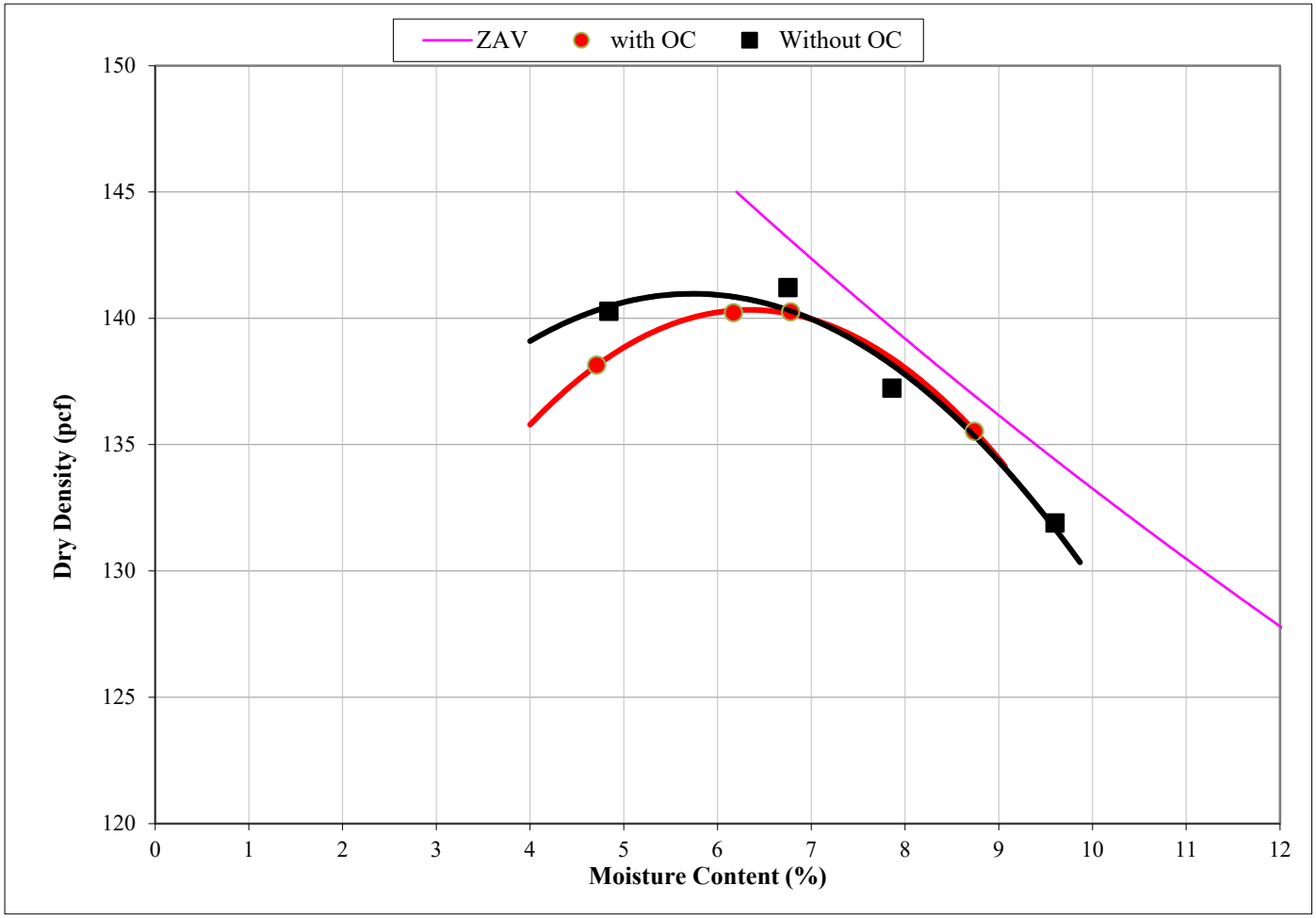
- Atterberg limits:

Trial	Polk	Marble Falls	20% P 80% MF	19% P 80% MF 1% OC
LL(%)	35	18	26	27
PL(%)	17	13	16	17
PI(%)	18	5	10	10
LS(%)	7	4	5	6
Type	CL	CL-ML	CL	CL
USCS	SC	GW-GM	GC	GC



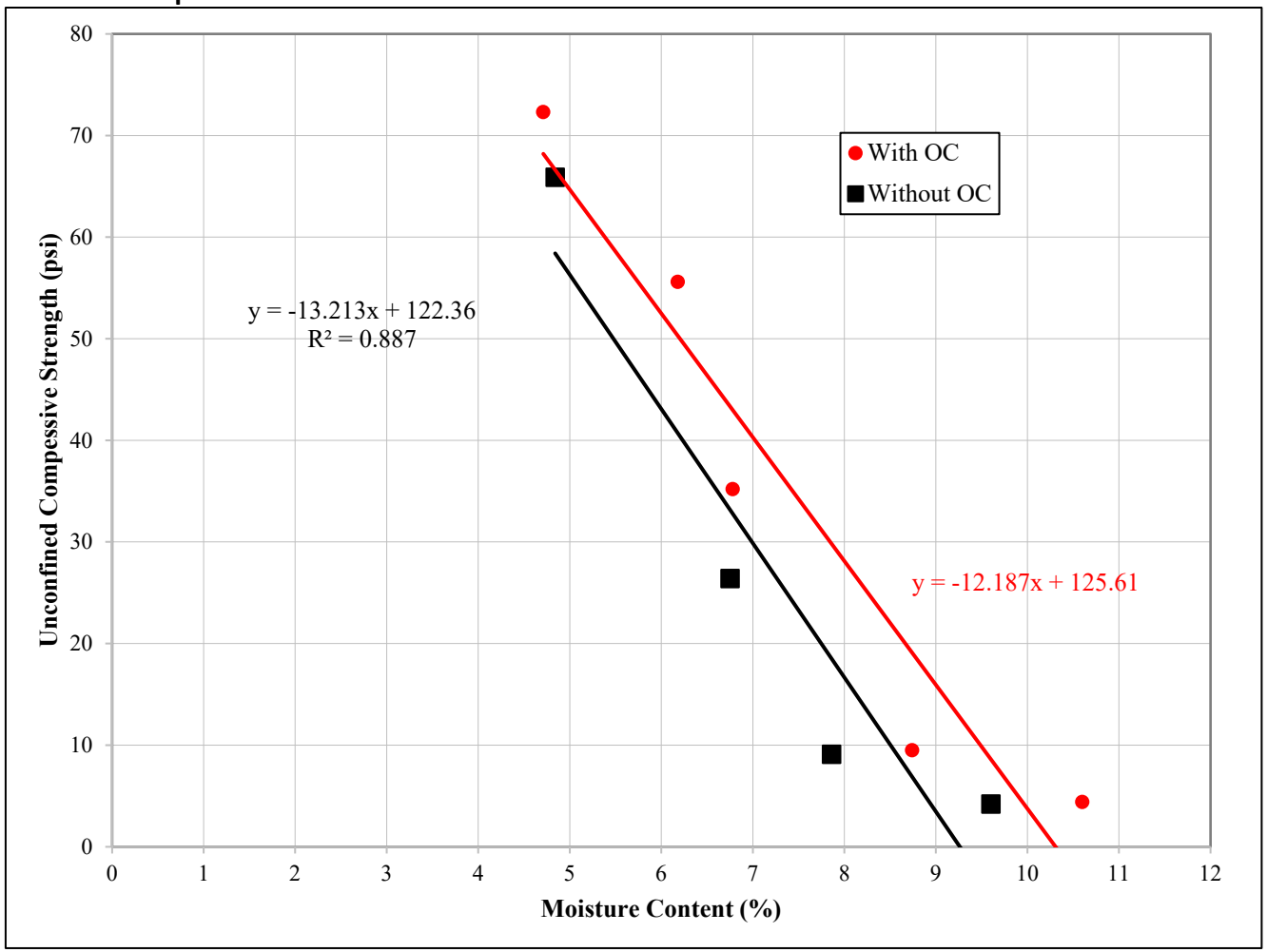
Blended Base Course Materials

- Proctor (Tex-113-E)



Blended Base Course Materials

- Unconfined Compression for Tex-113-E



Blended Base Course Materials

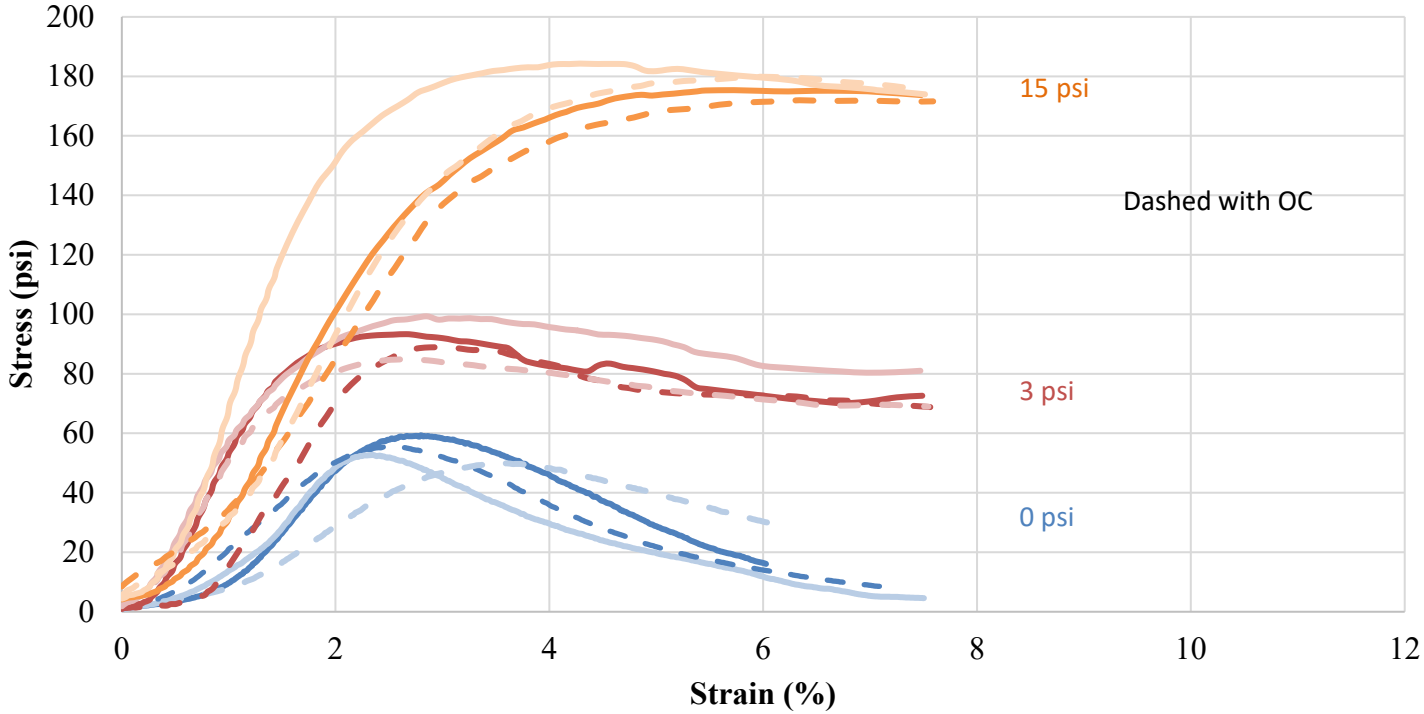
- Wet ball mill test:

Water Content (%)	Wet ball Mill		Wash Sieve Analysis	
	1	2	1	2
Wihtout OC Strain (%)	40	40	33	33
With OC Strain (%)	42	42	34	34

  - The average wet ball mill value increase from 33% in the proportion passing the No. 40 sieve.

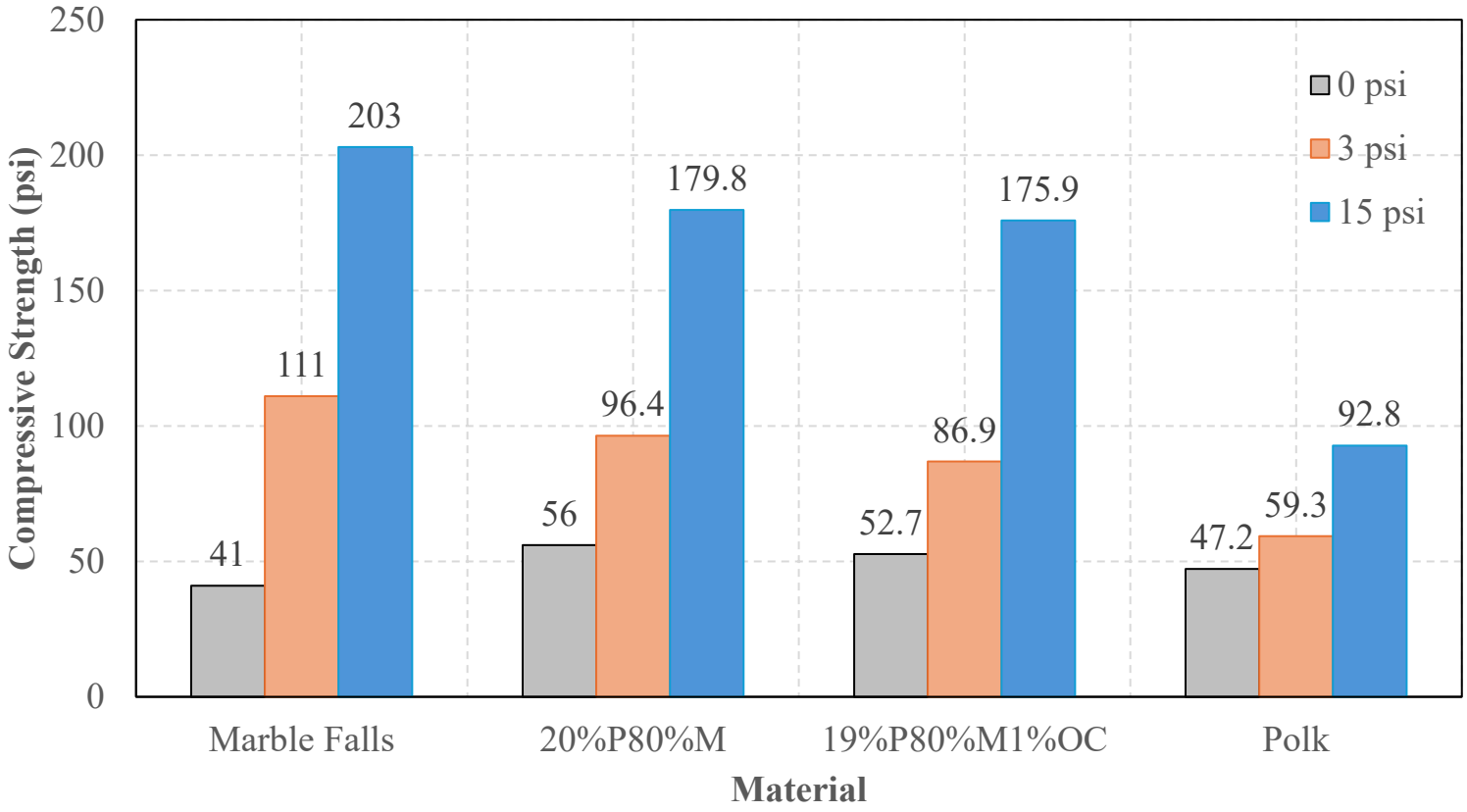
Blended Base Course Materials

- Texas triaxial test:



Blended Base Course Materials

- Texas triaxial test:



Base Materials

- TxDOT Item 247: Flexible Base

Property	Test Method	Grade 1–2	Grade 3	Grade 4[2]	Grade 5
Sampling	Tex-400-A				
Master gradation sieve size (cumulative % retained)	Tex-110-E				
2-1/2"		0	0	As shown on the plans	0
1-3/4"		0–10	0–10		0–5
7/8"		10–35	–		10–35
3/8"		30–65	–		35–65
#4		45–75	45–75		45–75
#40		65–90	50–85		70–90
Liquid Limit, % Max	Tex-104-E	40	40	As shown on the plans	35
Plasticity Index, Max[1]	Tex-106-E	10	12	As shown on the plans	10
Plasticity index, Min[1]		As shown on the plans	As shown on the plans	As shown on the plans	As shown on the plans
Wet ball mill, % Max	Tex-116-E	40	–	As shown on the plans	40
Wet ball mill, % Max increase passing the #40 sieve		20	–	As shown on the plans	20
Min compressive strength, psi	Tex-117-E			As shown on the plans	
lateral pressure 0 psi		35	–		–
lateral pressure 3 psi		–	–		90
lateral pressure 15 psi		175	–		175

1. Determine plastic index in accordance with Tex-107-E (linear shrinkage) when liquid limit is unattainable as defined in Tex-104-E.
2. Grade 4 may be further designated as Grade 4A, Grade 4B, etc.

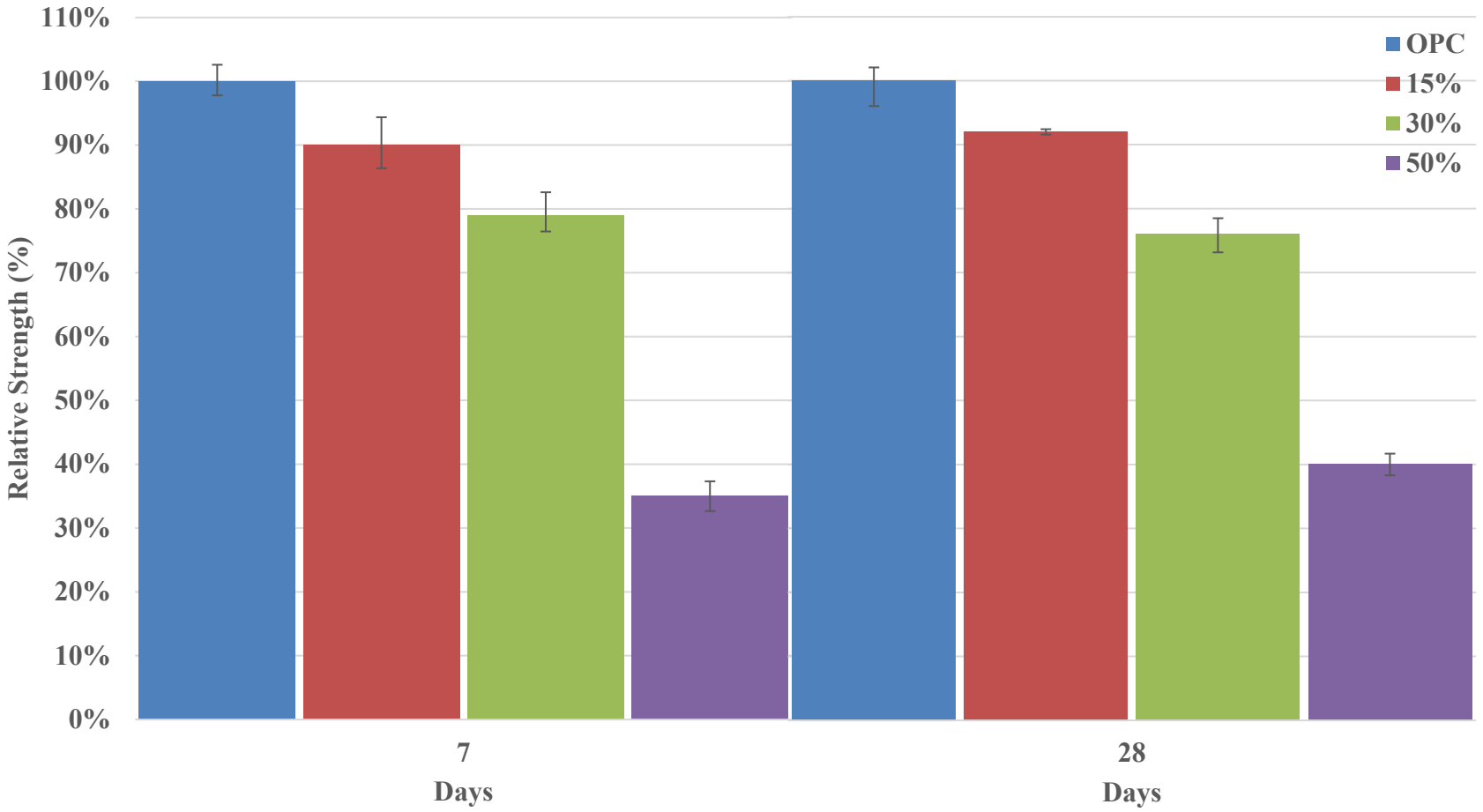
Material Performance after Treatment

USE IN CONCRETE AS FINE AGGREGATE

Concrete Testing

- Four concrete mixtures were prepared for compressive strength testing.
- OPC mixture was the control (w/c=0.6, sand-to-cement ratio 2.75, 28 days compressive strength of 3000 psi)
- The fine aggregates constitute 30% of the total weight of the concrete mix (excluding water).
- Mixtures with 15, 30, and 50% replacement levels of fine aggregates were prepared.

Concrete Testing



Concrete Testing

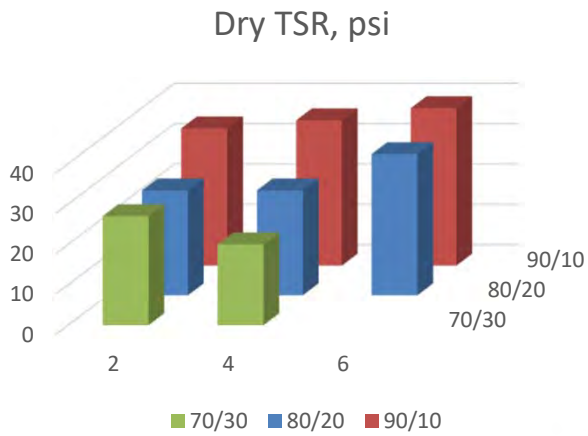
- Four concrete mixtures were prepared for compressive strength testing.
- OPC mixture was the control (w/c=0.6, sand-to-cement ratio 2.75, 28 days compressive strength of 3000 psi)
- The fine aggregates constitute 30% of the total weight of the concrete mix (excluding water).
- Mixtures with 15, 30, and 50% replacement levels of fine aggregates were prepared.
- Up to 30% replacement of fine aggregates with drill cuttings met the desired target strength of 3000 psi. However, the 50% replacement mixture required more water (w/c=0.75) and failed to met the target strength.

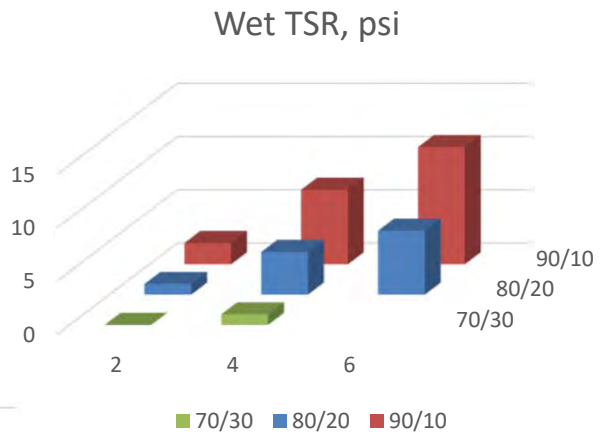
Material Performance after Treatment

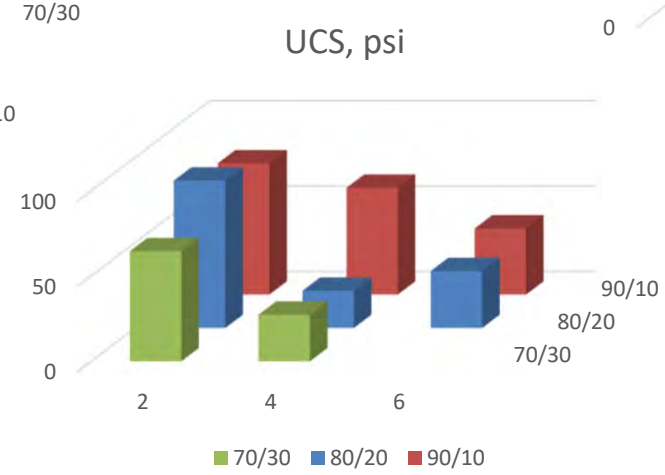
STABILIZATION FOR ASPHALT / ASPHALT CONCRETE

Stabilization with Admixtures-Asphalt

Mixture Property[1]	Test Method	Minimum Requirement
Indirect Tensile Strength (IDT) psi	Provided by MTD	50
Moisture Conditioned[2] IDT, psi		30
Moisture Conditioned[2] Unconfined Compressive Strength (UCS)[3], psi		120







Material Performance after Treatment

ENVIRONMENTAL TESTING

Environmental Characterization

- Leaching Testing
  - Leaching Environmental Assessment Framework (LEAF)
    - Toxicity Characteristic Leachate Procedure (TCLP) is the required test method for hazardous waste, but LEAF tests provide more flexibility by evaluating leaching under a wider range of environmental conditions
    - EPA encourages the use of LEAF to evaluate the potential for adverse impacts to human health or the environment
  - EPA SW-846 Test Method 1313 - Liquid-Solid Partitioning as a Function of Extract pH Using a Parallel Batch Extraction Procedure
    - Liquid-to-solid ratio of 10
    - Granular material
    - Extracts of solid material (i.e., the eluates) tested for Total Organic Carbon (non-purgeable organic carbon, NPOC)
  - EPA SW-846 Test Method 1315: Mass Transfer Rates of Constituents in Monolithic or Compacted Granular Materials Using a Semi-Dynamic Tank Leaching Procedure
    - Liquid-to-surface area ratio (L/A) of 9 ± 1 mL / cm^2
    - Monolithic material (cylindrical mortar specimens)
    - These tests were performed on the Concrete Specimens

Total Organic Carbon

- LEAF 1313 Eluate
- P-1, -2: Polk drill cuttings
- LRA: limestone rock asphalt
- RAP: reclaimed asphalt pavement
- L: lime (1.2 g)
- L3%7d, 28d: lime stabilized drill cuttings, 3% wt. lime, 7-d and 28-d
- OC: Organoclay
- OC 1%-5%: Organoclay stabilized drill cuttings, % wt.

Material	NPOC (ppm)	pH
P-1	19.61 ± 0.090	7.82 ± 0.03
P-2	17.53 ± 0.222	7.83 ± 0.03
LRA	4.48 ± 0.194	8.19 ± 0.11
RAP	5.70 ± 0.751	8.14 ± 0.26
L	0.19 ± 0.007	12.73 ± 0.02
L3%7d-1	11.62 ± 0.012	12.54 ± 0.01
L3%7d-2	11.72 ± 0.053	12.56 ± 0.00
L3%28d	12.84 ± 0.110	12.10 ± 0.03
OC	3.69 ± 0.179	8.43 ± 0.13
OC1%	13.97 ± 0.205	7.52 ± 0.06
OC2%	12.06 ± 0.380	7.86 ± 0.01
OC3%	10.77 ± 0.055	7.70 ± 0.20
OC5%	10.29 ± 0.803	7.65 ± 0.00

Total Organic Carbon

- OC 1%-5%12d: Organoclay stabilized drill cuttings, cured for 12 days, % wt.
- New OC 1%-5%: Organocaly from a different source, % wt.
- Cement 4%-10%: Cement stabilized drill cuttings, % wt.

Material	NPOC (ppm)
OC1%12d	16.61
OC2%12d	19.04
OC3%12d	19.86
OC5%12d	21.95
newOC1%	21.11
newOC2%	18.71
newOC3%	17.24
newOC5%	14.82
cement 4%	45.99
cement6%	44.39
cement8%	41.06
cement10%	39.04

Trace Metal Concentrations

- LEAF 1313 Eluate
- ICP-MS
  (Standard Method 3125)
- Trace metal grade nitric acid was used for dilution and sample preparation

- DC1, DC2: drill cuttings
- LRA1, 2: limestone rock asphalt
- RAP: reclaimed asphalt pavement
- Lime1, 2 & 3: lime 3%, 7 day cured
- L28-1 & 2: lime 3%, 28 day cured

ICP-MS Results in ppb

Sample	Li	Mg	Al	K	Ca	Cr	Fe	Co	Ni	Cu	Zn	As	Se	Sr	Cd	Ba	Pb
DMS			24000			100		1500	490	1300	7300	10	50		5	2000	15
DC1	17.7	8820		8530	12500			3.35	17.3	8.40			4.77	7520		43.4	
DC2	21.5	8550	10.1	8340	12200			4.39	9.46	6.62			21.0	7350		45.4	
LRA1	1.80	3750	68.6	2380	2470								1.49	427		27.7	
LRA2	3.57	6430	32.1	5950	3870			3.86	1.48					653		39.4	
RAP	1.19	1160	378	1270	1030	6.30	181			5.90			1.88	73.1		12.3	12.4
Lime1	14.3	84.2	94.7	67200	56400			52.1	124	94.4			38.8	50000		1550	50.3
Lime2			108	6310	53000				49.5	93.4				48600		1500	
Lime3			87.8	31900	58300				307	566				173000		6000	
L28-1			762	8910	43200			9.68	70.6	131				44100		904	
L28-2	11.9	313	1090	10300	53200			14.3	88.9	173			37.7	49700		1220	39.4

DMS 11000, leachate concentrations for traditional construction materials (ug/L). Values in the table are for Asphaltic Binders (same or lower than aggregates and cementitious materials)

Trace Metal Concentrations

- LEAF 1313 Eluate
- ICP-MS (Standard Method 3125)
- Trace metal grade nitric acid was used for dilution and sample preparation

- OC: organoclay / OC1-5%: organoclay % wt.
- o-OC1-5%: organoclay % wt., cured for 12 days
- n-OC1-5%: organoclay from a different source, % wt.

ICP-MS Results in ppb

Sample	Li	Mg	Al	K	Ca	Cr	Fe	Co	Ni	Cu	Zn	As	Se	Sr	Cd	Ba	Pb
DMS			24000			100		1500	490	1300	7300	10	50		5	2000	15
DC1	17.7	8820		8530	12500			3.35	17.3	8.40			4.77	7520		43.4	
DC2	21.5	8550	10.1	8340	12200			4.39	9.46	6.62			21.0	7350		45.4	
LRA1	1.80	3750	68.6	2380	2470								1.49	427		27.7	
LRA2	3.57	6430	32.1	5950	3870			3.86	1.48					653		39.4	
RAP	1.19	1160	378	1270	1030	6.30	181			5.90			1.88	73.1		12.3	12.4
OC	20.9	11700		2150	7890									2760			
OC1%	15.4	7660		8390	13200				7.85	5.79			2.1	8790		55.1	
OC2%	20.8	7890		8760	13200				8.21	5.97	6.63		2.4	8810		50.5	
OC3%	14.9	7890		8000	13100				8.14	5.10			2.50	8440		47.5	
OC5%	16.8	8340		8010	13300				8.40	5.13	6.31		2.94	8760		48.5	
o-OC1%	22.0	8500		7940	14000			0.965	9.58	10.4	5.04		2.25	8550		257	
o-OC2%	18.2	8570		8710	14000				8.33	8.54			2.06	9080		52.8	
o-OC3%	17.9	9420		8370	15000				8.75	8.37			2.34	8890		49.9	
o-OC5%	19.5	9950	23.5	8540	15500				8.11	7.40			2.50	9340		51.7	
n-OC1%	18.5	9360		10200	15100				8.94	6.75			2.89	8610		47.9	123
n-OC2%	17.8	8770		9710	14200				8.67	6.15			2.17	8480		48	
n-OC3%	18.3	9850		10600	16500				9.40	6.41			2.1	9400		50.9	
n-OC5%	21.4	9750		10200	16300				8.95	6.27			2.14	9060		46.1	

Semivolatile Organic Compounds

Detection Summary, LEAF 1313 Eluate

Material	Analyte	Result	Qualifier	RL	MDL	Unit	Dil Fac	Method
Drill Cuttings	2, 4, 6-Trichlorophenol	0.00409	J	0.0100	0.00189	ppm	1	8270E
	Benzoic acid	0.0535	J	0.0600	0.00430			
	Di-n-butyl phthalate	0.00235	J	0.0100	0.00226			
Drill Cuttings (LEAF 1314 Eluate)	2, 4, 6-Trichlorophenol	0.00200	J	0.00500	0.000946	ppm	1	8270E
RAP	Benzoic acid	0.0515	J	0.0600	0.00430	ppm	1	8270E
LRA	No detections							
Lime	Benzoic acid	0.0450	J	0.0300	0.0189	ppm	1	8270E
Lime 3%	2, 4, 6-Trichlorophenol	0.00566	J	0.0100	0.00189	ppm	1	8270E
	2, 4-Drichlorophenol	0.00218	J	0.0100	0.00208			
	Benzoic acid	0.0660	*-	0.0600	0.00430			
	Naphthalene	0.00162	J	0.0100	0.00150			
Cement Paste	Benzoic acid	0.0236	J	0.0300	0.0189	ppm	1	8270E
Cement 4%	2, 4, 6-Trichlorophenol	0.00209	J	0.0100	0.00189	ppm	1	8270E
	Benzoic acid	0.0968	*-	0.0600	0.00430			
Cement 10%	Benzoic acid	0.0985	*-	0.0600	0.00430	ppm	1	8270E

RL: Reporting Limit, MDL: Method Detection Limit, Dil Fac: Dilution Factor
J: result is less than the RL but greater than or equal to the MDL and the concentration is an approximate value
*-: Laboratory Control Samples and/or Sample Duplicates are outside acceptance limits, low biased

Total Petroleum Hydrocarbons

TCEQ Method 1005, LEAF 1313 Eluate

Material	Analyte	Result	Qualifier	RL	MDL	Unit	Dil Fac	Method
Drill Cuttings 1	>C12-C28 Range Hydrocarbons	1.74	J	5.45	0.941	ppm	1	TX 1005
	Total Petroleum Hydrocarbons (C6-C35)	1.74	J	5.45	0.965			
Drill Cuttings 2	>C12-C28 Range Hydrocarbons	1.72	J	5.34	0.921	ppm	1	TX 1005
	Total Petroleum Hydrocarbons (C6-C35)	1.72	J	5.34	0.945			
RAP	No detection							
LRA	>C12-C28 Range Hydrocarbons	1.32	J	4.66	0.804	ppm	1	TX 1005
	Total Petroleum Hydrocarbons (C6-C35)	1.32	J	4.66	0.825			
Cement 4%	>C12-C28 Range Hydrocarbons	2.88	J	5.58	0.962	ppm	1	TX 1005
	Total Petroleum Hydrocarbons (C6-C35)	2.88	J	5.58	0.987			
Cement 10%	>C12-C28 Range Hydrocarbons	2.21	J	5.28	0.912	ppm	1	TX 1005
	Total Petroleum Hydrocarbons (C6-C35)	2.21	J	5.28	0.935			

RL: Reporting Limit, MDL: Method Detection Limit, Dil Fac: Dilution Factor
J: result is less than the RL but greater than or equal to the MDL and the concentration is an approximate value
*-: Laboratory Control Samples and/or Sample Duplicates are outside acceptance limits, low biased

Volatile Organic Compounds

Detection Summary, LEAF 1315 Eluate

Material	Analyte	Result	Qualifier	RL	MDL	Unit	Dil Fac	Method
Control, interval 1	No detections							
Control, interval 2	No detections							
15%, interval 1	No detections							
15%, interval 2	No detections							
30%, interval 1	No detections							
30%, interval 2	No detections							

RL: Reporting Limit, MDL: Method Detection Limit, Dil Fac: Dilution Factor
J: result is less than the RL but greater than or equal to the MDL and the concentration is an approximate value
*-: Laboratory Control Samples and/or Sample Duplicates are outside acceptance limits, low biased

- EPA Method 8260C
- Gas Chromatography / Mass Spectrometry (GC-MS)

Semivolatile Organic Compounds

Detection Summary, LEAF 1315 Eluate

Material	Analyte	Result	Qualifier	RL	MDL	Unit	Dil Fac	Method
Control, interval 1	No detections							
Control, interval 2	No detections							
15%, interval 1	Di-n-butyl phthalate	0.942	J	1.14	0.765	ppb	1	8270E
15%, interval 2	No detections							
30%, interval 1	No detections							
30%, interval 2	No detections							

RL: Reporting Limit, MDL: Method Detection Limit, Dil Fac: Dilution Factor
J: result is less than the RL but greater than or equal to the MDL and the concentration is an approximate value
*-: Laboratory Control Samples and/or Sample Duplicates are outside acceptance limits, low biased

- EPA Method 8270E
- Gas Chromatography / Mass Spectrometry (GC-MS)

Total Petroleum Hydrocarbons

Detection Summary, LEAF 1315 Eluate

Material	Analyte	Result	Qualifier	RL	MDL	Unit	Dil Fac	Method
Control, interval 1	No detections							
Control, interval 2	No detections							
15%, interval 1	No detections							
15%, interval 2	No detections							
30%, interval 1	No detections							
30%, interval 2	No detections							

RL: Reporting Limit, MDL: Method Detection Limit, Dil Fac: Dilution Factor
J: result is less than the RL but greater than or equal to the MDL and the concentration is an approximate value
*-: Laboratory Control Samples and/or Sample Duplicates are outside acceptance limits, low biased

- TCEQ Method 1005
- Gas Chromatography