CLEANUP STANDARDS FOR RADIUM CONTAMINATED SOILS

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ABSTRACT

In 1983 EPA promulgated standards for cleanup of uranium and thorium mill tailings at 40 CFR 192. These standards address a specific example of the cleanup of radium contamination. They have been used for the cleanup of radium-contaminated soils at other sites, primarily because they are the only related standards that exist. However, EPA advised caution at the time these standards were issued: "It should be noted that these standards in no way are intended to establish precedents for other situations or regulations involving similar environmental objectives, but with different economic and/or technological circumstances."(1) This paper assesses the suitability of these standards for use in the cleanup of contaminated soil at sites other than uranium or thorium mill tailings sites.

THE 40 CFR 192 CLEANUP STANDARDS

The 40 CFR 192 rules specify two types of standards. The first addresses the disposal of uranium and thorium mill tailings, and are not discussed in this paper. The second addresses cleanup, and are the subject of this paper. They include limits for indoor radon concentrations and indoor gamma exposure rates for cleanup of buildings, as well as limits on radium concentrations in soil for cleanup of land. The former, those for indoor radon and gamma exposure, are health-based standards; while the latter, radium concentrations in soil, are technology-based standards, keyed to the sensitivity of radiation monitoring systems. For uranium tailings, the increased indoor radon concentrations and indoor gamma exposure rates were caused by placing tailings around buildings and houses. Radon is a decay product of radium and, since it is an inert gas, can move through soil and enter buildings above soil that is contaminated with radium. It was assumed at the time the 40 CFR 192 standards were promulgated that the indoor standards would be achieved by removing such tailings and replacing them with clean soils.

The 40 CFR 192 standards for soil specify a concentration limit of 5 pCi/g radium in the top 15 cm of soil and 15 pCi/g radium in any 15 cm thickness below the top 15 cm. The limit for the top layer was based on limiting external exposure rates to persons who may spend time on the land. Its purpose is to indicate when cleanup of thin surface layers of windblown tailings is necessary to provide adequate public health protection. Although this criterion provides adequate health protection for the situations it was developed to address, the value was selected with the limitations of field measurement equipment in mind, and the transient nature of windblown contamination situations.

The 15 pCi/g soil concentration limit is a technology-based standard. It is a practical measurement criterion for use in locating discrete quantities of tailings that were deposited or placed in subsurface locations at mill sites. These tailings deposits are generally limited in area and volume, with little or no mixing with adjacent soils, and have activities exceeding 100 pCi/g. Convenient measurement techniques for assaying radium activity in boreholes can not readily achieve a sensitivity better than 15 pCi/g in 15 cm layers. Since this is adequate for locating the edge of subsurface deposits of uranium mill tailings, it was selected as an appropriate standard for use at the tailings sites. Cleaning up deposits of tailings using this standard will leave at most only very small deposits that would not produce sufficient radon to cause a significant increase in indoor levels in a structure built over them.

THE RELATIONSHIP BETWEEN INDOOR RADON AND RADIUM IN SOIL

In this paper it is assumed that the goal of land cleanup around houses should be to meet the health protection standards of 40 CFR Part 192. These require limiting the average indoor radon concentration to 0.02 WL (4 pCi/l) including background, and restricting the indoor gamma exposure rate to 20 microR per hour above background at any location in a permanently occupiable structure. The technical objective, therefore, is to achieve those conditions in the soil around present (and potential) occupiable structures that will satisfy these indoor requirements.

The characteristics of the soil, the pressure differential between indoor air and the atmosphere, and the air exchange rate of the building itself are major factors that determine the buildup of indoor radon. For this paper, a model called RAETRAN (2) was used to examine the relationship between radium concentrations in soil and indoor radon concentrations in a house constructed over land contaminated with radium. In the RAETRAN estimates, the soil characteristics were varied, as were the radium concentrations in soil. The pressure differential and air exchange rates were held constant at values representing a new house constructed to meet current energy conservation guidelines.

RAETRAN predicts the movement of radon in soil by both diffusion and advection. Radon moves through soils along the path of least resistance. In many cases, particularly when subsoil is in its undisturbed natural state, cracks or volumes containing porous material create "channels" through which radon can move rapidly to entrance places in buildings. This movement of radon is caused by a difference in pressure and is called advective movement, or advection. The most advantageous situation (from the radon control perspective) is that of soils which have characteristics which force radon to move primarily through diffusion. Diffusion theory is based on the thermodynamic principle that radon will move to regions of lower radon concentration in its attempt to achieve equilibrium. Diffusion movement is advantageous for radon control since it takes longer for radon to move by diffusion than by advection. More radon will decay during this longer time period. While radon control is based primarily on replacing contaminated soil with clean soil, soil characteristics which assure that primary movement of radon through the clean soil layer is by diffusion provide additional protection.

RAETRAN estimates radon movement into houses directly through basement floors and through cracks in basement floors, e.g., the crack between the basement floor slab and the basement wall.(3,4) The source of the radon is the radium in the soil under and adjacent to the basement. In the model radon 1) diffuses through the basement slab into the house and 2) flows advectively from the soil through cracks and into the house. When the house parameters are held constant, the rate at which the radon flows is determined primarily by the characteristics of the soil. If soils retain significant fractions of water, radon will flow principally by diffusion. It is noted, however, that modeling radon movement through soils is a developing field. As more sophisticated models become available, detailed specifications of models may change.

The rate at which radon moves through the soil depends on the concentration of radon (from radium) in the soil and the soil characteristics: permeability, moisture content, and radon diffusion coefficient. A slow movement of radon is desirable, i.e., a low diffusion coefficient, to assure decay of most of the radon in the soil. The scientific basis for predicting the movement of radon through soil has been developed by Rogers, et.al. (5) and is widely used (6). This work forms the basis for RAETRAN (2).

In using the model it is important to recognize that there are two sources of radium: the radium in the clean fill (sandy clay loam) that replaces the contaminated soil in the excavated zone and the radium in the soil below or outside the excavated volume, i.e., the initially contaminated soil that has not been excavated. In making estimates of excavation depths the characteristics of both the replacement soil (the clean fill or backfill) and the existing, contaminated soil are used. An indoor radon concentration can be selected and, using RAETRAN, the concentration of radium in the underlying soil (which can be different for the replaced soil and the unexcavated soil) can be calculated for soils with various properties. The excavation depth can then be increased to assure that the radon moving through various soils and entering the house builds up to less than the indoor radon limit.

Our RAETRAN estimates (7) are based on:

- an indoor radon concentration of 2 pCi/l, to allow for other sources of radon,
- a house air exchange rate of 0.35/hour,
- an indoor pressure of -2.4 Pa,
- three different soils (clay loam, sandy clay loam, and loamy sand); however, the backfill soil is sandy clay loam for all cases,
- three different moisture contents in the soils (wet to dry: -0.1, -0.3, and -15.0 matric potentials),
- three different radium concentrations in the backfill (i.e., 1 pCi/g, 3 pCi/g, and 5 pCi/g),and
- other factors (as described in the accompanying paper by Hull and Nielson)(7).

A total of 81 different cases were modelled.

The major conclusion from the RAETRAN analysis is that for most soils, if not all, radon attenuation properties are insufficient to provide adequate protection from indoor radon that is produced by a homogenous mixture of radium at a concentration of 15 pCi/g radium in soil underlying a house. The zero excavation values for all cases fell between 5.8 and 10 pCi/g radium in soil, i.e., a concentration greater than 5.8 to 10 pCi/g would require excavation of contaminated soil and replacement with cleaner (lower radium concentration) backfill.

From the model results it is evident that the properties of the underlying soil are most important in determining excavation depths (or in selecting radium concentration limits at a given depth). This is observed by comparing the allowable radium concentration in the underlying soil for an excavation depth of 2 meters. At long-term average matric potentials of -0.3 for both backfill and underlying soil, with backfill at 1.0 pCi/g radium, the difference in radium concentrations is more than an order of magnitude: 140 pCi/g for clay loam vs. 13 pCi/g for loamy sand. The difference is even greater if highly saturated soils are considered, again for a 2 meters backfill depth. At matric potentials of -0.1 for both backfill and underlying soil, with backfill at 1.0 pCi/g radium, the difference in radium concentrations limits in the underlying soil is 380 pCi/g for clay loam vs. 11 pCi/g for loamy sand. In both of these cases, the concentration limits for an underlying soil of sandy clay loam falls between the clay loam and the loamy sand.

The model results also indicate that, with sandy clay loam as the backfill for all cases, the radium concentration in the backfill greatly effects the excavation depth (or the acceptable radium concentration for a given depth). For example, the excavation depth increases from 2 meters to 3 meters as the radium concentration of the backfill is increased from 1 pCi/g to 5 pCi/g. Conversely, for a fixed excavation depth of 2 meters, the limiting radium concentration in the underlying soil decreases from 20 pCi/g to 10 pCi/g as the radium concentration in the backfill increases from 1 pCi/g to 5 pCi/g.

For cases where there is no contaminated material left beneath the excavated zone, the source of radon is primarily from the clean fill. Any of the soils having a radium concentration of less than about 5 pCi/g can be used to achieve the indoor radon limit. Therefore, the radium concentration of the backfill should not exceed 5 pCi/g which will assure that indoor radon will not exceed health protection limits (40 CFR 192).

Additional assessment is needed for cases where contaminated material is left under the clean fill. However, preliminary results indicate that radium concentrations as low as 10 pCi/g, combined with permeable backfill soils, may cause indoor radon levels that exceed the limit. Based on these preliminary results, removal of all radium contaminated soil is the preferred course. However, further assessment may lead to other options, especially if less permeable soils are used for backfill.

Based on the above estimates, with backfill soil directly beneath the floor slab, soil with a radium concentration of 5 pCi/g is consistent with the indoor radon limit. However, in all cases, the indoor radon limit will be exceeded if the soil concentration of radium is at or near the 15 pCi/g limit.

THE RELATIONSHIP BETWEEN INDOOR GAMMA RADIATION EXPOSURES AND RADIUM IN SOIL

Examination of external exposure estimates indicates that indoor gamma radiation exposure may be very close to the limit specified in 40 CFR 192 when soil concentrations approach the 15 pCi/g limit for radium-226 and/or radium-228.

Further work is needed to estimate external exposure levels in basement areas when a house is constructed in soil containing up to 15 pCi/g radium in soil.

The limit specified in 40 CFR Part 192 for gamma radiation levels is a structure applies at any location in the structure, whether occupied or not, and without limitation on the size of the area over which it applies. The standard is specified in terms of an ionization rate in air, and thus is not equivalent to dose to humans. This is to encourage direct measurement of radiation levels and ease implementation by avoiding the necessity of making numerous adjustments for occupancy, location of people within the building, and other factors.

CONCLUSIONS

The 40 CFR 192 soil concentration limits are technology-based standards that were developed specifically for application to the uranium and thorium tailings sites, and will generally not be suitable for application at other sites. However, since the 40 CFR 192 indoor radon and gamma limits are health-based standards, they can provide the basis for deciding what cleanup levels must be achieved in remedial actions involving radium-contaminated soil on which occupiable structures exist or may be constructed in the future.

The results of the model tests indicate that:

1. Based on estimates from the RAETRAN model for backfill soil directly beneath the floor slab, most soils with a radium concentration of 5 pCi/g or less will satisfy the indoor radon limit. However, in all cases, the indoor radon limit will be exceeded if the soil concentration of radium is at or near the 15 pCi/g limit.
2. For the case where essentially all soil contamination at a site is from a material that has elevated levels of radium, e.g., mill tailings, and if this contaminated soil has not been significantly mixed into adjacent uncontaminated soil, the 15 pCi/g radium limit will assure that only small quantities of contaminated soil will be left following remediation. For this case, the soil concentration limits in 40 CFR 192 provide a safe level of health protection.
3. For the case where soil contamination is from a large volume of low level radium-contaminated material, e.g., phosphogypsum wastes, or where higher concentration radium-contaminated soil has been significantly mixed into uncontaminated soils, the 15 pCi/g radium limit provides inadequate assurance that a safe level of health protection will be met.

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