THE ROLE OF PETROGRAPHIC AND RADIOCHEMICAL ANALYSES IN RISK ASSESSMENT AT SUPERFUND SITES CONTAMINATED WITH RADIOACTIVE MATERIALS

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ABSTRACT

For the past four years, the EPA Office of Radiation Programs has been evaluating the use of Volume Reduction/Chemical Extraction (VORCE) technologies as a means of remediating sites containing large volumes of soil contaminated with relatively low levels of enhanced sources of naturally occurring radionuclides, primarily radium and thorium (1). In the process of evaluating these technologies, data have been gathered on the chemical and physical characteristics of the radioactive contaminants (i.e., radiochemical data) and the mineral composition of the soil matrix (i.e., petrographic data) at a number of Superfund sites. The radiochemical and petrographic data are used as a first step in determining the feasibility and potential cost-effectiveness of VORCE technologies as an alternative to soil excavation (2).

Questions have recently been raised regarding whether the site-specific radiochemical and petrographic data may also have use in the performance of Baseline Risk Assessments at these sites. In order to address these questions, petrographic and radiological analyses have been performed on soil samples obtained from a radium contaminated site in Montclair, New Jersey. While radon is the primary risk at this site by approximately three orders of magnitude, this investigation has focus on radium in the soil. The results of the analyses reveal that Ra-226 contamination is associated predominantly with the silt/clay size fraction of the soil and in a highly insoluble form. These findings indicate that standard default assumptions regarding soil to air suspension factors and inhalation dose conversion factors may not be appropriate for this site. Work is continuing on determining the potential significance of these findings and extending these studies to other Superfund sites.

INTRODUCTION

EPA guidelines regarding the performance of dose and risk assessments at Superfund sites contaminated with radioactive material are currently provided in the following EPA documents:

- "Risk Assessment Guidance for Superfund, Volume 1, Human Health Evaluation Manual (Part A) (EPA/540/1-89/002)."
- "Role of the Baseline Risk Assessment in Superfund Remedy Decisions," OSWER Directive 9355.0-30
- "Human Health Evaluation Manual, Supplemental Guidance: Standard Default Exposure factors," OSWER Directive 9285.6-03

Inspection of these guidelines and their references reveals that the following parameters, which are fundamental to the performance of dose and risk assessments, are also highly dependent on the physical and chemical properties of the radionuclides and, in the case of contaminated soil, the properties of the soil matrix within which the radionuclides are contained.

- Soil to air suspension factors
- Soil to plant transfer factors
- Binding coefficients (kds)
- Inhalation and ingestion dose conversion factors
- Weathering Factor

Guidance exists regarding generally accepted default values that may be used for each of these parameters (3, 4, 5, 7). However, it is widely recognized that there is a great deal of uncertainty regarding the applicability of the default values for these parameters at specific sites. For example, for the inhalation pathway, summaries of the literature on the airborne suspension of soil contaminated with radionuclides (3,4) acknowledges the highly variable nature of the suspension process and its dependence on wind speed and the physical and chemical properties of the soil. As documented in studies by the EPA (8), at Montclair, New Jersey, it has been shown that the risk assessment of radon is approximately three orders of magnitude higher than the risk by the inhalation of radium in the soil. The risk for radon decay products ranges from 0.0 to 361 excess deaths per thousands of peoples exposed, whereas, the number for radium ranges from 0.01 to 0.5 deaths per thousands of peoples exposed. Till sites literature which shows suspension rates varying by over 4 orders of magnitude (3). Table 12.9 of Randerson lists resuspension factors varying over 8 orders of magnitude (4).

Notwithstanding this variability, pathway models often use default values for the resuspension factor of about 10^{-5} to 10^{-4} pCi/m^3 per pCi/m^2. Alternatively, some models simply assume the radionuclide concentration in the dust in the air is the same as in the soil on the ground, and apply typical values of the airborne dust loading, such as 100 to 400 μg/m^3 (5).

As applied to specific sites, both approaches could result in estimates of the concentration of airborne contaminants

that are incorrect by one or more orders of magnitude. In the case of the suspension factors, suspension is likely only if the particle size of the contaminant is less than about 50 microns (3). In the case of the dust loading approach, there is evidence that enrichment or discrimination processes could result in radionuclide concentrations in airborne dust that are many times higher (i.e., enrichment) or lower (i.e., discrimination) than in the underlying soil (6). The purpose of this study is to evaluate the degree to which site-specific petrographic analyses of contaminated soil for several size fractions can provide information that can have a significant effect on the assumptions used to calculate inhalation doses from the suspension pathways of radium. Although the risk effects of radium in the soil are three orders of magnitude less than radon inhalation, an effective remediation of radium contaminated soils will mitigate any possible health effects of the radon inhalation and its daughter by-products.

METHODOLOGY

Radiochemical and petrographic data of radium contaminated soil obtained from Monclair, New Jersey sites have been compiled.

A detailed description of dry and wet sieving procedures and radiochemical and petrographic analyses are provided in contractor reports and previously published papers (1, 2, 10). In summary, dry sieving was performed on a 50 g soil sample using a model VS Brinkman vibrating siever. Wet sieving was performed on 200 to 400 g samples using the Gilson Wet-Vac Tester, Model WV-1. Gamma ray spectrometry, using high purity germanium detectors, was used to determine the Ra-226 concentration in soil samples. Ra-226 was detected and measured using the 186 KeV photopeak or the Bi-214 photopeak for samples with low specific activity.

RESULTS

Table I presents the grain size distribution, along with the Ra-226 concentration, in each size fraction for both dry and wet sieving. The dry sieving results reveal that the Ra-226 concentration in the fraction of the soil that may be suspended by the wind and other erosion processes (i.e., soil grain sizes at approximately 50 microns or less) is 282 pCi/g. This is as compared to the overall Ra-226 concentration in the soil of 126 pCi/g. Accordingly, it appears that soil suspension processes should result in an approximate two- to three-fold enrichment of Ra-226. These results are compatible with NRC guidance, which recommends a 2.5 dust-to-ore enrichment ratio for estimating the airborne concentration of uranium and daughter products at uranium mill operations (9).

For wet sieving, it appears that a larger portion of the soil is found in the smaller size fractions. For example, following dry sieving, only about 1.5% of the mass is in the fraction less than 75 microns. However, following wet sieving, 22% of the mass is in the fraction less than 75 microns. The reason for this is believed to be the break-up of the soil particles during the wet sieving process. The results also reveal that wet sieving results in a shift in the radioactive contaminants to the smaller size fraction. For example, the Ra-226 concentration in the grain size fraction less than 75 microns is about 395 pCi/g, as compared to 282 pCi/g in the dry sieved soil. These results reveal an approximate 3-fold enrichment in the Ra-226 concentration in the soil particles in the suspendable size range.

Another parameter of interest is the degree to which the radium is associated with particles in the respirable size range; i.e., less than 10 microns. However, since dry sieving was performed only down to the 75 micron sieve, it is not possible to determine from this analysis if a significant fraction of the Ra-226 is associated with particles in the respirable range.

Wet sieving found a large portion of the activity in the respirable size range. However, these results should be reviewed with a degree of caution because the wet sieving process causes a separation of soil particles, resulting in an enhancement of the Ra-226 in the clay sized fraction. There is some question whether the actual particle size distribution

TABLE I

Ra-226 Concentration Distribution as a Function of Mass Size Fractions in Montclair Soil Samples

	Dry Sieving		Wet Sieving	
Size Fraction Fraction (microns)	Mass Fraction	Ra-226 Concentration (pCi/g)	Mass Fraction	Ra-226 Concentration (pCi/g)
0.5 - 2			0.01	1113
2 - 5			0.02	644
5 -15			0.04	430
15 - 38			0.06	360
38 - 50			0.05	240
50 - 75	0.015*	282*	0.04	304
75 - 106	0.017	242	0.05	170
106 - 150	0.036	335	0.05	138
150 - 300	0.057	200	0.14	113
300 - 1180			0.22	100
1180 - 2000	0.084	189	0.05	39
2000 - 4750	0.149	113	0.07	26
>4750	0.465	57	0.20	44

* < 75 microns

of soil and contaminants in the natural setting at Montclair is better represented by the results of dry versus wet sieving.

Finally, the results of the analyses revealed that Ra-226 in the soil samples is strongly bound to the soil matrix. Virtually none of the Ra-226 was found in the wash water. Petrographic analyses revealed that the Ra-226 is strongly bound to, or associated with, ore minerals (carnotite and uranite), acid leach products (radium barium sulfate and amorphous silica), furnace fired materials (coal ash and slag), and clay (illite, etc.). Accordingly, upon inhalation, the Ra-226 may be expected to behave as a non-transportable material, remaining in the lung for a relatively long period of time. This may indicate that the Ra-226 at the Montclair sites should be classified as a Y-Class radionuclide with a lung clearance rate on the order of years.

This finding can be significant because the standard method for calculating the inhalation dose from Ra-226, as recommended in Federal Guidance No. 11 (11), revised 10 CFR 20, and ICRP 30 (12), is based on the assumption that Ra-226 is moderately transportable (i.e., it is assumed to have a lung clearance rate on the order weeks and is treated as a W-Class radionuclide). Accordingly, there is a need to determine whether W-Class or Y-Class transportability is appropriate for Ra-226 in the form observed in the samples taken at Montclair. The use of the W classification in ICRP-30 is based on the assumption that the most common form of radium is radium sulfate. The petrographic analyses indicate that some of the Ra-226 at the Monclair sites may be in a more complex and less transportable matrix.

CONCLUSIONS

It appears that petrographic and radiological analyses of contaminated soil for several size fractions can provide information that can have a significant effect on the assumptions used to calculate inhalation doses from the suspension pathways of radium. At a minimum, this information should be helpful in defining site-specific radium suspension factors, determining whether the contaminant is in the respirable size range, and determining whether the material should be assigned to the D-, W-, or Y-Class for the purposes of calculating the doses due to inhalation of radium. In site specific cases where a gaseous component such as radon is not involved, this type of investigation could have direct application in determining risk assessment.

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