Modeling the Inhalation Enhancement Factor in Prospective Radiological Risk Assessment

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INTRODUCTION

Inhalation of suspended particulates originating from contaminated soil is an important exposure pathway that may result from disposal of long-lived solid radioactive waste. In radiological assessments, it is often assumed that inhaled particulates originating from contaminated soil have the same radionuclide concentration as the bulk soil. One of the factors that can affect the dose from inhalation of suspended soil particulates is the radionuclide enhancement (enrichment) in airborne soil particles relative to that of the underlying soil, which results from the particle size-dependent distribution of contaminants among soil grains and the particle size-dependent distribution of atmospheric aerosols of soil origin. The enhancement factor (enrichment factor) is defined as a ratio of airborne particle activity concentration (Bq kg\(^{-1}\)) to surface soil activity concentration (Bq kg\(^{-1}\)). Contaminant interaction with soil involves processes such as surface adsorption, which favor small, clay-size particles, because of their high surface to mass ratio. Therefore, small particles will carry the bulk of adsorbed contaminants, especially for the highly-sorbing elements, such as Pu, Th and U (Sheppard and Evenden, 1992; Sheppard and Evenden, 1994; EPA, 1999). The present work evaluated some aspects of particle size-selective processes involved in resuspension and subsequent inhalation of resuspended soil.

METHODS

The particle size-dependent enhancement arises because the concentration of the contaminant sorbed onto the soil grains is proportional to the surface area available for sorption. Small particles have a larger surface-to-volume ratio than large particles and thus have a greater potential for contaminant adsorption per unit mass. Using a simple geometric approach, the enhancement factor for radionuclides adsorbed to surfaces of soil grains can be expressed as a ratio of the available surface area per unit mass of suspended particulates to the available surface area per unit mass of underlying soil. Assuming that the ratio of the particle surface area to its mass (volume) is proportional to the inverse of the particle size, and that this relationship is appropriate for the soil and for suspended particulates, it is possible to calculate the surface-to-mass ratios for particles characterized by such distributions as well as the resulting enhancement factor. The assumptions stated above reduce the process of particle size-dependent enrichment to a first-order approximation based on simple, geometric considerations. Such an approach ignores many other factors that control the distribution of a contaminant between the particle sizes. However, it can provide insight into the importance of several parameters that influence the level of airborne contaminants originating from contaminated soil. The surface-to-mass ratios for the resuspended soil particles were calculated using the single-mode and bimodal distributions of suspended soil particles (represented by lognormal distributions) and for the function describing the particle size distribution of the soils based on the method developed by Skaggs \textit{et al.} (2001).
RESULTS AND DISCUSSION

For the environments with low aerosol concentrations in air (on the order of a few tens to a few hundreds of µg m$^{-3}$), the measured particle size distributions of resuspended soil can be approximated by a single-mode, log-normal distribution with a mass median aerodynamic diameter (MMAD) in the range from 2 to 6 µm and a geometric standard deviation (GSD) of about 2 (NCRP, 1999; Patterson and Gillette, 1977; Dorrion, 1997; Pinnick et al., 1993). Using the method described above, the enhancement factor for resuspended soil particles was calculated for the three example soil types that can be characterized as sandy soil, loam soil and clay soil and for the distributions of resuspended particulates with MMAD of 2 µm, 4 µm and 6 µm, and GSD of 2. The characteristics of the particle size distributions, along with the calculated enhancement factors, are presented in Table 1.

Table 1. Characteristics of Single-Mode Particle Size Distributions of Resuspended Soil for Low Atmospheric Aerosol Loading Conditions and Estimated Enhancement Factors

<table>
<thead>
<tr>
<th>MMAD (µm)</th>
<th>GSD</th>
<th>Sandy soil</th>
<th>Loam soil</th>
<th>Clay soil</th>
</tr>
</thead>
<tbody>
<tr>
<td>6</td>
<td>2</td>
<td>3.7</td>
<td>1.4</td>
<td>0.7</td>
</tr>
<tr>
<td>4</td>
<td>2</td>
<td>5.5</td>
<td>2.1</td>
<td>1.0</td>
</tr>
<tr>
<td>2</td>
<td>2</td>
<td>11.0</td>
<td>4.2</td>
<td>2.0</td>
</tr>
</tbody>
</table>

As expected, the enhancement is the largest for the sandy soils and for the low MMADs of the airborne particulates, i.e., the enhancement factor increases as the distribution of the airborne aerosols is becomes more biased towards small particles, relative to the particle size distribution of the bulk soil. This is a consequence of the fact that clay-size particles tend to concentrate strongly-sorbing contaminants. If these particles are then selectively resuspended and/or the clay-size particles contribute only a small fraction of the soil mass (while carrying the bulk of activity), as in sandy soils, the resulting enhancement factor can be significant.

For the conditions of heavy atmospheric aerosol loading, e.g., during mechanical soil disturbances or very strong winds, the particle size distribution tends to be bimodal, with a transient large particle component (NCRP, 1999; EPA 1996; Johnston et al., 1992; Pinnick et al., 1985). The parameters describing the bimodal particle size distributions used in the calculations of the enhancement factor and the experimental conditions, along with the calculated enhancement factors for the high aerosol concentrations, are summarized in Table 2.

For the experiments summarized in Table 2, the calculated enhancement factor was less than 1, which would indicate lower contaminant concentration of resuspended soil compared to that of bulk soil and thus depletion of resuspended soil in small, clay-size particles compared to that of bulk soil. However, this effect is likely artificial, because the enhancement factor calculation does not take into account soil aggregates, and the fact that small clay particles may adhere to larger, resuspended particles. More likely, for the conditions of high and very high dust loading, the concentration of contaminants in resuspended soil is comparable to that of bulk soil.
Table 2. Characteristics of Bimodal Particle Size Distributions of Resuspended Soil for High Atmospheric Aerosol Loading Conditions and Estimated Enhancement Factors

<table>
<thead>
<tr>
<th>Experimental Conditions</th>
<th>Small Aerosol Fraction</th>
<th>Large Aerosol Fraction</th>
<th>Enhancement Factor</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>MMAD (µm)</td>
<td>GSD</td>
<td>Mass loading (g m⁻³)</td>
</tr>
<tr>
<td>Sandy soil a</td>
<td>9.5</td>
<td>2.1</td>
<td>0.18</td>
</tr>
<tr>
<td>Silty soil b</td>
<td>19.4</td>
<td>2.0</td>
<td>1.63</td>
</tr>
<tr>
<td>Sandy soil c</td>
<td>9.6</td>
<td>2.1</td>
<td>0.019</td>
</tr>
<tr>
<td>Loamy soil c</td>
<td>6.2</td>
<td>2.2</td>
<td>0.025</td>
</tr>
</tbody>
</table>

a Fresh dust generated by heavy military equipment (Pinnick et al., 1985)
b Blowing dust (Pinnick et al., 1985)
c Blowing dust (Patterson and Gillette, 1977; Pinnick et al., 1985); measurements taken in rural area of west Texas. Based on the prevalent soil types in that region it is assumed here that the soils were loamy.

CONCLUSIONS

In summary, when the radionuclide concentration in air is calculated for the scenarios involving soluble contaminant in the soil and the subsequent resuspension of contaminated soil, the enhancement of radionuclide concentration in the airborne soil-derived aerosols, compared to that of bulk soil, may need to be taken into account. The enhancement factor is strongly dependent on the characteristics of the soil and on the presence of soil-disturbing activities. The values of the enhancement factor, calculated using a simple geometric approach, indicate that the radionuclide concentration of the airborne aerosols is likely to be significantly greater than that for the underlying soil only for the sandy soils under the conditions of low (typical) atmospheric mass loading and, to a lesser degree, for the silty soils. High atmospheric aerosol loadings do not result in the enhancement of resuspended activity concentration, but such conditions are transient and not typical for most inhalation exposure circumstances. Because the enhancement depends strongly on the characteristics of the soil and the atmospheric mass loading, the site-specific conditions should be considered in radiological assessments, especially when the conditions are known to be favorable for this effect to occur.

ACKNOWLEDGEMENT

This manuscript has been authored by Sandia National Laboratories under Contract DE-AC04-94AL85000 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

REFERENCES


