AIRBORNE PARTICLE RESUSPENSION AND INHALATION RADIOLOGICAL DOSE ESTIMATION FOLLOWING VOLCANIC EVENTS

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ABSTRACT

This report presents concepts and factors that influence airborne radionuclide contamination in the biosphere and the calculation of inhalation radiological doses so that analysts can consider these concepts and factors to develop independent models or evaluate models developed by others for volcanic disruption consequences of a geologic repository for high-level radioactive waste and spent nuclear fuel. For extrusive volcanism in long-term performance assessments, the inhalation of resuspended radionuclides in ash is expected to dominate over other potential exposure pathways. Concepts and factors are presented for the resuspension of particles on the ground into air, inhalation of airborne particles, and inhalation dose estimation. The discussion on airborne concentration of resuspended radionuclides focuses on ash deposit characteristics, waste concentration in resuspended particles, and aerosol characteristics. For airborne particle inhalation, highlights are provided for the chemical form of inhaled material, airborne particle size, reference values for human and physiological aspects, and age dependences as they pertain to standard inhalation dosimetric models. General formulations for inhalation intake of radionuclides and conversion of radionuclide intakes to radiological dose are presented along with a discussion of additional case-specific factors that could be considered further.

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QUALITY OF DATA, ANALYSES, AND CODE DEVELOPMENT

DATA: All CNWRA-generated original data contained in this report meet the quality assurance requirements described in the Geosciences and Engineering Division Quality Assurance Manual. Sources for other data should be consulted for determining the level of quality for those data.

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Reference

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1 INTRODUCTION

Long-term assessments of geologic disposal of high-level radioactive waste and spent nuclear fuel are predicated on preventing or minimizing the release of radioactive material (i.e., radionuclides) into the biosphere. Because the intended timeframes for repositories to contain waste and protect individuals from exposure are very long (e.g., 10,000 years or longer), the occurrence of unlikely events with small annual recurrence rates becomes more likely as the period of interest or performance increases. For example, an event with a recurrence rate of 10^{-7} yr^{-1} has a very small probability (0.00001) of occurring within 100 years. For a 100,000-year period of interest, the probability of at least one event occurring is 0.01. As a result, long-term performance assessments consider disruptive events to avoid underestimating the aggregated risk from disposal.

The purpose of this knowledge capture report is to describe, in a single document, the concepts and factors that influence a radiological dose calculation for the inhalation of resuspended particles contaminated with high-level radioactive waste. This report focuses on extrusive volcanism, which is an igneous disruptive event. Extrusive volcanism is of primary interest because its consequences can be dominated by different processes and, therefore, can require different models than those for intrusive igneous events or other scenarios whose consequences are linked directly to potential groundwater contamination. Although volcanic hazard assessment and event probability determinations also factor into the long-term performance assessment, they are not addressed in this report.

Extrusive volcanism at the disposal site for high-level radioactive waste and spent nuclear fuel, hereafter referred to simply as high-level waste, is conditioned on an igneous event. Although the igneous event may be unlikely, it could make important contributions to the total system risk if a subsurface volcanic conduit directly intersected the potential repository and resulted in the atmospheric release of high-level waste incorporated in volcanic ash. In this report, the plain-language term *ash* is used in place of the more precise term *tephra*. Although ash refers to tephra with particle diameters less than 2 mm [0.08 in], usage of ash in this report does not categorically exclude particles with diameters greater than 2 mm [0.08 in]. Depending on eruption characteristics and atmospheric conditions, contaminated volcanic ash could be deposited in locations where future populations may be exposed as a result of inhalation of contaminated resuspended material. In addition to any initial ash deposited at the receptor location (an area occupied or inhabited by an individual), the long-term ash transport by wind and surficial water drainage, referred to as eolian and fluvial redistribution, respectively, could remobilize volcanic ash toward the receptor location and contribute to a persistent exposure to contaminated material.

Respiratory health effects from volcanic ash have been studied. Horwell and Baxter (2006) wrote a comprehensive review of published clinical, epidemiological, and toxicological studies. Blong (1996) remarked that ashfall covers a wide area and, thus, can affect a large number of people relative to other volcanic hazards (e.g., lava flows). These studies consider nonradiological health effects. This report, however, addresses radiological dose estimation due to high-level radioactive waste contamination in ash from volcanic events that may disrupt a potential geologic repository. Nonradiological health effects are not covered in this report. Although airborne particle concentrations can reach maximums during heavy ashfall events, eruptions are often short-lived (e.g., with active ashfall episodes representing a small fraction of time compared to 1 year). Ash deposits, on the other hand, can remain in the local environment for extended periods of time, where they can be resuspended and remobilized by natural processes or human activities, and contribute to long-term exposure. For extrusive volcanism, long-term performance assessments are likely to be more sensitive to long-term exposure and conditions rather than short-term eruption conditions. Separate considerations of short-term exposures can determine if such aspects warrant inclusion in the performance assessment model. For the previously described reasons, this report focuses on extrusive volcanic event consequences by addressing the resuspension of particles on the ground into air, inhalation of airborne particles, and inhalation dose estimation.

2 RESUSPENSION OF PARTICLES ON THE GROUND SURFACE INTO AIR

During a volcanic disruption event, high-level waste could be incorporated into ash (or lava) and result in radionuclide deposition on the ground surface. Compared to lava flows, the airborne transport of contaminated ash is more effective at distributing radionuclides in the biosphere, away from the volcanic vent and to locations where individuals can be exposed. Radiological doses from a volcanic disruption event are expected to be dominated by radionuclide contamination in volcanic ash, and therefore, radionuclide contamination in lava is not considered further. As described in the Introduction, the assessment of long-term inhalation exposure following a volcanic eruption focuses on the resuspension of ash contaminated with high-level waste into air. In addition to the previously mentioned exposure time considerations, ashfall episodes include ash (or tephra) particles of various sizes, some of which are too large to be inhaled.

Inhalation dose depends on the airborne concentration of radionuclides in the breathing zone of exposed individuals. The airborne concentration of resuspended radionuclides depends on (i) ash deposit characteristics, (ii) waste concentration in resuspended particles, and (iii) aerosol characteristics. Each of these factors is described in this section.

2.1 Relationship to Deposit Characteristics

Contaminated ash deposits on the ground surface represent a source for airborne resuspension and inhalation dose. Deposit characteristics can influence the high-level waste concentration in air. Anspaugh, et al. (2002) described three types of resuspension models—time-dependent resuspension factor, resuspension rate, and airborne mass loading—and indicated that the mass loading model has been preferred for long times after deposition. Analysts should recognize differences associated with radiological contamination events and consider them in the model development process. For a volcanic event, the resuspension of contaminated ash is of primary interest. In comparison, the airborne radionuclide concentration following the deposition of atmospheric fallout from weapons testing is determined from the resuspension of soil. Unlike fresh atmospheric fallout deposits, ash deposits can have appreciable mass and thickness. The thickness of the ash deposit relative to the resuspendible thickness can be important and may be included in performance assessment models.

Resuspendible thicknesses for soil and sediment tend to be on the order of millimeters [approximately tenths of inches] without intentional ground alteration or soil movement or on the order of centimeters [about a couple of inches] for localized mechanical surface disturbances (Linsley, 1978; Sehmel, 1980). Differences in soil and ash deposits may be pertinent to resuspendible thickness considerations (Leslie, et al., 2007, Chapter 15). For ash deposits thinner than the resuspendible thickness, the total airborne mass load of resuspended particles can be modeled as a mixture of contaminated ash particles and noncontaminated particles from underlying soil. For ash deposits thicker than the resuspendible thickness, resuspended material can be assumed to be entirely contaminated (i.e., no dilution with noncontaminated soil particles). In selecting or evaluating parameter values for resuspensible thicknesses, it is important to note that certain resuspendible thicknesses may dilute the airborne concentration of radionuclides and reduce estimated doses. Specific effects depend on modeling details, but one example for thin ash deposit cases is that greater resuspendible thicknesses could increase resuspended contributions from underlying, noncontaminated soil to the total airborne particle concentration.

2.2 Waste Concentration in Resuspended Particles

The concentration of high-level waste in resuspended particles is another factor in the inhalation dose calculation. The concentration of high-level waste in resuspended particles can be derived from the high-level waste concentration in ash and resuspended ash concentration. In some cases, noncontaminated particles or multiple contaminated sources (e.g., with different concentrations of high-level waste) could contribute to the total airborne particle concentration. Because the mass of ash generated by volcanoes is much greater than the mass of high-level waste in geologic repository concepts, mixing of high-level waste in magma and ash is expected to result in small mass fractions of high-level waste in ash (Codell, 2004). Downward migration of radionuclides into soil due to natural weathering processes may also factor into model calculations for the concentration of radionuclides at the surface that can be resuspended into air.

2.3 Aerosol Characteristics

Aerosol relates to a collection of airborne particles. In this report, an aerosol comprises ash or soil that has been resuspended into the air. Aerosol characteristics can be divided into physical characteristics and chemical form. Physical characteristics and chemical form of a radionuclide-bearing aerosol both factor into the inhalation dose calculation. Physical characteristics of particle size, density, shape, and aerodynamic behavior are addressed in this section. Because chemical form is manifested in the inhalation dose coefficient, the chemical form of airborne particles is addressed in Section 3.

During inhalation, physical characteristics of airborne particles influence their deposition in different regions and compartments of the human respiratory tract. Aerosols can consist of a range of particle sizes (polydisperse) or a single particle size (monodisperse). For most environmental exposure scenarios including potential exposure to resuspended waste following a disruptive event (i.e., the scenario of primary interest to this report), aerosols are polydisperse. A lognormal particle size distribution is assigned as standard convention. The two parameters that define the lognormal particle size distribution are the median diameter and geometric standard deviation. As described in ICRP (2003, Section 4.1.2), the activity median aerodynamic diameter (AMAD) is used when particle deposition depends mainly on sedimentation and inertial impaction (greater than about 0.5 µm). Half of the radioactivity in an aerosol is associated with particles with aerodynamic diameters greater than and less than AMAD. Activity median thermodynamic diameter (AMTD) is used when particle deposition depends mainly on diffusion (less than about 0.5 µm). Because irregular shapes are commonplace for airborne particles, equivalent diameters are used to describe airborne particle diameters. The aerodynamic equivalent diameter is the diameter of a unit density spherical particle that exhibits the same terminal settling velocity as the irregularly shaped particle in air. ICRP (1994, Equation D.5) indicates the following relationship for the aerodynamic equivalent diameter

$$D_{ae} = d_e\sqrt{\frac{\rho C(d_e)}{\chi \rho_0 C(d_{ae})}} \tag{2-1}$$

where

d_{ae} — aerodynamic equivalent diameter (µm)

d_e — diameter of a spherical particle with the same volume as the particle considered, referred to as the equivalent volume diameter (μm)

ρ — particle mass density (g cm^{-3})

ρ_0 — unit density (1 g cm^{-3})

C — slip correction factor, shown as dependent on d_e and d_{ae} (unitless)

χ — particle shape factor (unitless)

From Eq. (2-1) for a given particle (i.e., equivalent volume diameter, d_e), the aerodynamic equivalent diameter is proportional to the square root of ρ/χ. ICRP (1994, Section B.1.2) states that $C(d_e)$ and $C(d_{ae})$ are approximately one for larger particles. Information regarding resuspended particle size can be used to determine if slip correction factors are important in the determination of the aerodynamic diameter. For example, the sizes of resuspended airborne particles from basaltic deposits, as measured by Benke, et al. (2009) at the Sunset Crater Volcano, Arizona, are large enough such that the unity approximation could be applied to the quotient $C(d_e)/C(d_{ae}) \approx 1$.

For cases in which the unity approximation is appropriate for the slip correction quotient, the relationship between aerodynamic equivalent diameter and equivalent diameter simplifies to

$$d_{ae} = d_e\sqrt{\frac{\rho}{\chi}} \tag{2-2}$$

where

d_{ae} — aerodynamic equivalent diameter (μm)

d_e — equivalent volume diameter (μm)

ρ — particle mass density (g cm^{-3})

χ — particle shape factor (unitless)

ICRP (1994, Section 4.2) defines a density of 3 g cm^{-3} [187 lb ft^{-3}] and a particle shape factor of 1.5 as default values for typical compact, irregularly shaped airborne particles. Typical values for the shape factor range from 1 to 2 (ICRP, 1994, Section 4.1.2). Analog deposits can yield insights on resuspended particle sizes and aerodynamic diameters. In some cases, aerodynamic diameter can be measured directly. In other cases, Eq. (2-1) or (2-2) can be used to account for the effect of airborne particle density and particle shape on the aerodynamic diameter and particle behavior in the human respiratory tract. These physical characteristics of airborne particles determine where ash and radionuclide deposition occurs within the respiratory tract following inhalation. The distribution of radionuclide deposition in specific regions or compartments serves as input to biokinetic modeling of radionuclide absorption in, transfer within, and clearance from the human body. These aspects of the inhalation dose calculation are described further in the next two sections.

3 INHALATION OF AIRBORNE PARTICLES

This section focuses on the human and physiological aspects that factor into the calculation of radiological doses from inhalation. An inhalation internal dose occurs following the intake of airborne radioactive material. Airborne radioactive material can be suspended particulates, vapors, or gases, but must be present in the breathing zone of an individual to be inhaled. Inhaled radionuclides are either exhaled or deposited in various regions and subregions (or compartments) of the human respiratory tract (e.g., ICRP, 1994; 1979, Part 1, Section 5). The compartmental distribution of deposited radionuclides depends on the characteristics of the airborne radioactive material. Systemic uptake of radionuclides into blood and body fluids results in the biological transfer of radionuclides to individual organs as well as excretion from the body. Because radioactive decay within internal organs can continue for several years or decades following the original intake of radioactive material, internal dose calculations are performed for a commitment period. The accumulated internal dose due to an intake of radioactive material (e.g., single intake or annual intake) is typically applied as the receipt of a single dose in the year of intake that equals the accumulated dose over the commitment period. ICRP (1991, 1979) applies a 50-year commitment period for adults. A 70-year commitment period is used for children (ICRP, 2001, Section 2.3). Dose coefficients for intakes are published as committed dose quantities. In other words, the summation of accumulated doses over the commitment period has already been performed and included in the computation of internal dose coefficients.

For a particular radionuclide, the inhalation dose coefficient depends on the chemical form of the inhaled material and its airborne particle size distribution (i.e., AMAD or AMTD, as described in Section 2). Different dose coefficient values are specified for lung absorption type (ICRP, 2001) or lung inhalation class (U.S. Environmental Protection Agency, 1988), which relate to the solubility of inhaled material in lung fluid and clearance rate from the pulmonary region of the lung, respectively. Both lung absorption type and inhalation class associations are determined from the chemical forms of the compounds [e.g., as stated in ICRP (1996, Table 2) and 10 CFR Part 20, Appendix B]. Default airborne particle size distributions (e.g., AMAD values) are noted. Dose coefficients are based on reference values for human and physiological aspects. In ICRP (2001), differences in these aspects are responsible for differences in dose coefficients for workers and members of the public. Dose coefficients are also specified for different age groups in ICRP (2001). Because of the many factors considered, published dose coefficients can be applied to a wide range of exposure situations. When exposure situations differ markedly from reference values or default conditions, specific information regarding the actual conditions for retrospective assessments or anticipated conditions for prospective assessments should be factored into the dose estimation. For example, Benke, et al. (2009) indicated that most of the airborne resuspended mass measured during heavy surface-disturbing activity was associated with large particle sizes. Similar information can provide a technical basis for selecting inhalation dose coefficients corresponding to an AMAD value that is more appropriate for the exposure conditions than the one generally recommended in the absence of specific information.

4 DOSE ESTIMATION

Inhalation dose estimation relies on the characterization of the airborne radioactive material and human body response to inhaled radionuclides. Details of those aspects have been discussed in the prior two sections. A general formulation of the inhalation dose calculation is presented. For some applications, a simplified approach may be sufficient. For other applications, greater realism and complexity may be needed. For this reason, the general formulation is accompanied by a discussion of additional case-specific factors that can be considered further, as needed. This section is separated into two subsections. The inhalation intake of radionuclides is addressed in Section 4.1. The conversion of radionuclide intakes to radiological dose is presented in Section 4.2.

4.1 Intake of Radionuclides

Determination of the inhalation intake of radionuclides is an important intermediate quantity in the dose calculation. The inhalation intake on radionuclides is computed as

$$Intake = Airborne\ Radionuclide\ Concentration \times Breathing\ Rate \times Exposure\ Time \qquad (4\text{-}1)$$

Individual radionuclides, multiple sources of contamination, and different physical activity levels are not explicitly expressed in Eq. (4-1) but can be accounted for by performing computations for separate contributions followed by summation. Intake can be specified as either mass or activity for each radionuclide. Specific activity is used to convert radionuclide mass to an activity.

Although the exposed individual may or may not be involved in the generation of airborne radioactive material, the activities of the individual during exposure affect the inhalation dose calculation. The amount of air breathed is directly proportional to the breathing rate and exposure time. Breathing rate also influences the compartmental distribution of deposited radionuclides (ICRP, 2003, Table 3.2). ICRP has established reference values for workers (ICRP, 1994, Table 6) and members of the general population (ICRP, 2003, Table A4). Reference values are widely accepted for prospective dose calculations. When anticipated conditions in a prospective dose calculation differ significantly from reference values, reference values may be replaced by values that are more representative of anticipated conditions. For actual exposures, use of individual values and specific information is advised to improve the dose assessment. ICRP (2003, 1994) provides relevant information on differences among individuals including the effects of different parameter values on dose assessments.

When the individual’s local environment is contaminated, the activities of the exposed individual can influence the resuspension of and airborne concentration of radionuclides. Accounting for the time spent performing different activities and the airborne concentration of radionuclides during those activities can provide a more realistic estimate for the inhalation intake of radioactive material compared to using a single, conservative value for the airborne concentration of radionuclides, breathing rate, and exposure time during the year (or occupancy fraction). For example, different levels of surface-disturbing activity can be accounted for separately. In addition, different characteristics could be propagated for multiple sources that contribute to airborne contamination at the receptor location. This treatment could include resuspended radionuclide-bearing particles from ash deposits at the receptor location, from fluvially redistributed deposits, and from eolian redistributed deposits (Leslie, et al., 2007, Chapter 15). Such modeling flexibility can be beneficial in accounting for realizations without primary ashfall at the receptor location due to winds that direct the ash plume away from the receptor location. In those cases, inhalation dose would arise from redistributed deposit contributions.

4.2 Conversion of Radionuclide Intake to Dose

The last step of the calculation converts radionuclide intakes into radionuclide doses. Contributions from individual radionuclides are summed to yield a total inhalation dose

$$Inhalation\ Dose\ = \sum_i Intake_i \times D_i \qquad (4\text{-}2)$$

where

$Intake_i$	—	Inhalation intake of radionuclide i (Bq)
D_i	—	Inhalation dose coefficient for radionuclide i (Sv Bq^{-1})

Inhalation dose coefficients have been tabulated for individual radionuclides in ICRP (2001) and U.S. Environmental Protection Agency (1988). Each set of dose coefficients corresponds to a specific dosimetric model. Dosimetric models include biokinetic transfer of radionuclides in the body, energy deposited in individual organs due to radioactive transformations and radiation emitted within the body, radiation weighting factors for converting energy deposited to an equivalent dose, and organ weighting factors for computing the effective or whole body dose equivalent.

Dose coefficients in ICRP (2001) are consistent with the dosimetric model and organ weighting factors of ICRP (1991) and ICRP (1996), often referred to as ICRP–60 dosimetry and ICRP–72 dose coefficients. Dose coefficients in U.S. Environmental Protection Agency (1988) are consistent with older dosimetric modeling and dose coefficients in ICRP (1979) and adopted in the current 10 CFR Part 20. Although recommendations and organ weighting factors have been updated by ICRP (2007), a revised set of dose coefficients had not been published at the time this report was written.

5 SUMMARY

Concepts and factors that influence the radiological dose calculation were presented for the inhalation of resuspended particles contaminated with high-level waste. For extrusive volcanism in long-term performance assessments, the inhalation of resuspended radionuclides in ash is expected to dominate over other potential exposure pathways. Resuspension of particles on the ground into air, inhalation of airborne particles, and inhalation dose estimation were described. The discussion on particle resuspension addressed ash deposit characteristics, waste concentration in resuspended particles, and aerosol characteristics. The airborne particle size distribution, relationship of chemical form of inhaled material to lung absorption type or inhalation class, reference values for human and physiological aspects, and age dependences were highlighted as they pertain to standard inhalation dosimetric models. Inhalation intake of radionuclides and conversion of radionuclide intakes to radiological dose were presented as steps in the inhalation dose calculation. Individual concepts and factors as well as their interrelationships were introduced and explained so that they may be considered further by analysts in the development of independent models or in the evaluation of models developed by others.

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