Modeling the Inhalation Exposure Pathway in Performance Assessment of Geologic Radioactive Waste Repository at Yucca Mountain

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ABSTRACT

This paper describes the inhalation model for estimating radiation exposure to resuspended particles of contaminated soil. The source of radionuclides is from the use of contaminated water for crop irrigation. The choice of conceptual model and input parameter values strongly depends on site-specific conditions. This paper explains how the site-specific conditions influenced the inhalation exposure model for the Yucca Mountain performance assessment. The model parameters that were developed with consideration of the local conditions represent characteristics of the environment as well as the characteristic of the receptor occupying that environment. The model is based on the mass loading approach and the key parameters are mass loading levels that the receptor might be exposed to. Also important is the degree to which human soil disturbance results in higher mass loading levels, the degree of enhancement (enrichment) in radionuclide content of the resuspended dust, and the occupancy periods for exposure to various levels of particulate concentration in air. The significance of dosimetric parameters is also discussed.

INTRODUCTION

Inhalation of suspended particulates that originate from contaminated soil is an important exposure pathway, particularly for exposure to alpha-emitters, such as isotopes of uranium, neptunium, and plutonium. In this paper, we describe the inhalation exposure model developed to support the performance assessment for a proposed repository at Yucca Mountain, Nevada.

The Yucca Mountain inhalation exposure model was based upon a specific land use scenario developed to comply with federal regulations for the performance assessment. Those regulations required that the reference biosphere be consistent with current conditions in the region surrounding Yucca Mountain and that the receptor have a diet and living style representative of the people that live in that region. The hypothetical land use scenario involves long-term irrigation of agricultural land with contaminated water and ensuing accumulation of radionuclides in surface soil. Radiation exposure would arise from inhalation of contaminated particles originating from such soils. Resuspension of contaminated soil particles would occur due to the action of wind, but more importantly, because of soil disturbance by people. In this scenario, arising from contaminated water use, inhalation exposure also would result from the inhalation of radon decay products and from inhalation of particulates emitted from evaporative coolers. The models for radon and evaporative coolers, as well as for other pathways included in the biosphere model for the Yucca Mountain repository [1] are not discussed here. This paper only describes the modeling of exposure due to inhalation of soil particles and the term inhalation model used throughout refers only to that inhalation pathway.

Inhalation exposure pathway modeling has recently been investigated as one of the tasks of the BIOPROTA Project [2]. BIOPROTA was set up to identify and address key uncertainties in long term assessments of contaminant releases into the environment from geological radioactive waste disposal. Participants of this international Project include national authorities and agencies, both regulators and operators, with responsibility for achieving safe and acceptable radioactive waste management. The objective of the BIOPROTA inhalation task was to investigate the calculation of doses arising from inhalation of particles suspended from soils containing long-lived radionuclides that accumulated in soil from irrigation with contaminated water. An important conclusion of that investigation was that site-specific conditions influence the choice of conceptual model and input parameter values. Thus, we describe below the site-specific conditions that influenced the inhalation exposure model for the Yucca Mountain performance assessment.

CHARACTERISTICS OF THE YUCCA MOUNTIAN REGION

The region surrounding Yucca Mountain has low precipitation, hot summers, cool winters, low relative humidity, and a high rate of evaporation. Average annual precipitation in the Amargosa Farms region south of Yucca Mountain is about 115 mm and average monthly maximum temperatures range from 16°C in January to 40°C in July. Valley soils in the region are very gravelly fine sands to sandy loams and are generally deep and well to excessively drained.

The region surrounding Yucca Mountain is sparsely populated. In 2000, it was estimated that about 1,175 people resided in the 1,300-km^2 Amargosa Valley census division south of Yucca Mountain. Many of the people living there are retired or otherwise unemployed and many others travel outside of the area for employment.

There is a small agricultural industry in Amargosa Valley. About 2,000 acres there are commercially farmed, of which over 90 percent is planted in alfalfa and other hay, and there is a dairy with more than 5,000 cows. Many residences have gardens with vegetable plots and some have a few cattle and other farm animals.

There are no naturally occurring surface waters in the area. All water used for domestic, municipal, and agricultural purposes in Amargosa Valley, the populated region south of Yucca Mountain, comes from local groundwater wells.

MODEL DESCRIPTION

Most inhalation exposure models reviewed during the BIOPROTA investigation [2] were conceptually similar. The commonly used method for the assessment of exposure to resuspend particles uses an atmospheric mass loading approach, which is based on the mass of airborne particulates per unit volume of air inhaled by the receptor. This method was selected for the Yucca Mountain model because it best accounts for uncertainty and variability in site-specific conditions and because data are more readily available to develop parameter values than for other modeling methods.

The mass loading model is conceptually straightforward. It is based on the predicted or measured mass of airborne particulates per unit volume of air that is inhaled by the receptor and the inhalation exposure conditions for the receptor. The inhalation dose is calculated as

$$D_{inh} = Ca\ B\ t\ DC \qquad \text{(Eq. 1)}$$

where:

D_{inh}	=	inhalation dose (Sv)
Ca	=	radionuclide concentration in air from soil resuspension (Bq/m^3)
B	=	breathing rate (m^3 h^{-1})
t	=	inhalation exposure time (h)
DC	=	inhalation dose coefficient (Sv Bq^{-1})

In a simple case, radionuclide concentration in air per unit mass can be assumed to be the same as radionuclide concentration in soil and radionuclide concentration per unit volume of air is simply calculated as a product of radionuclide concentration in soil and mass loading:

$$Ca = Cs\ S \qquad \text{(Eq. 2)}$$

where:

Cs	=	radionuclide concentration in the surface soil per unit of mass (Bq/kg)
S	=	concentration of suspended particulates in air (mass loading) (kg/m^3)

The differences in the results that this simple produces arise from the model implementation for specific assessment contexts, and particularly from the values of model parameters used. Parameter values are selected by considering the different radionuclide processes governing environmental transport of radionuclides and conditions of human exposure, which vary depending on exposure scenarios and site-specific conditions. In the remainder of this paper the methods used to develop site-specific values for the individual parameters appearing in the above equations are discussed.

Evaluation of radionuclide concentration in soil

The radionuclide concentration is soil is calculated by considering long-term irrigation of agricultural soil with contaminated water and simultaneous removal of radionuclides from the surface soil by radioactive decay, leaching to the deep soil, and soil erosion. These processes are also included in many other models [2]. A key site-specific parameter used in these calculations is the rate of irrigation with contaminated water, which is the source of radionuclides in the soil. This parameter is particularly important for the arid climate of the Yucca Mountain region. With the annual average precipitation of only about 115 mm, frequent irrigation is essential for the sustainability of agriculture. Irrigation rates for the Yucca Mountain model were developed by considering the watering requirements for 26 locally grown commercial and garden crops [3]. The average irrigation rate of those crops is 0.95 m per year for the present-day climate.

Three processes that result in a reduction of radionuclides in soil were included in the inhalation model, leaching of radionuclides beneath the root zone of plants, soil erosion, and radioactive decay. Rates of leaching and soil erosion were calculated based on site-specific or regional soil characteristics, farming and gardening practices, irrigation rates, and climate. These processes are important because the radionuclide concentration in soil depends on both the rates of radionuclide addition and removal. Assuming constant rates of radionuclide addition and removal, radionuclide concentrations in soil will increase for as long as the soil continues to be irrigated or until a concentration that is at equilibrium between radionuclide additions and losses is reached.

The number of years that fields and gardens would be irrigated is an important parameter in the calculation of soil concentrations of radionuclides such as plutonium that take hundreds to a few thousands of years to reach equilibrium concentrations. There is considerable uncertainty in this parameter because modern agricultural and irrigation methods have been used in Amargosa Valley only since the 1960's. Duration of long-term irrigation ranging from 100 to 1,000 years for commercial fields and 25 to 250 years for gardens was used for the calculation of radionuclide concentration in surface soil. These values were developed by considering local land management practices and sustainability of agriculture, as well as social factors such as land tenure and mobility of the of the local population [4]. Radionuclide concentration in soil that was used in assessment of inhalation exposure was the greater of the equilibrium radionuclide concentration in the resuspendable layer of soil and the radionuclide concentration in the tillage soil depth, both calculated using the crop-specific irrigation rates and durations.

Evaluation of radionuclide concentration in air inhaled by the receptor

The activity per unit mass of resuspended particles may not be identical to the activity per unit mass of underlying soil that is the source of suspended particulates, in part because of the non-uniform distribution of activity with particle size and the preferential resuspension of smaller particles. Most inhalation models do not consider radionuclide enrichment in airborne soil particles relative to that of the underlying soil, although the importance of this process has been recognized [2]. Omission of this process may lead to results that are not cautious. Some modelers who included enrichment in their models obtained results that were significantly higher than those obtained using conventional methods [2].

The inhalation model for the proposed repository at Yucca Mountain includes an enhancement factor (enrichment factor) to account for possible differences in radionuclide concentrations between the soil and air. This factor is expressed as a ratio of airborne particle activity concentration (Bq kg^{-1}) to surface soil activity concentration (Bq kg^{-1}). Radionuclide concentration in air, *Ca*, are related to radionuclide concentration in surface soil and mass loading by

$$Ca = Cs\ S\ EF \qquad \text{(Eq. 3)}$$

where:

Cs	=	radionuclide concentration in the surface soil per unit of mass (Bq/kg)
S	=	concentration of suspended particulates in air (mass loading) (kg/m^3)
EF	=	enhancement factor (dimensionless)

Small particles have a larger surface to volume ratio than large particles and this ratio increases in inverse proportion to the particle size. As a result, small particles have a greater sorption capacity per unit mass or volume than larger particles. The size distribution of suspended particulates generally includes a greater fraction of smaller particles than that of the originating soil and thus, on average, suspend particles have larger available surface area, and consequently activity, per unit mass than that of soil. This enhancement has a particularly important effect on the inhalation dose for elements such as plutonium, which have high sorption coefficients, build up in surface soil, and have a relatively high contribution from inhalation to the all-pathway dose.

The enhancement factors used in the Yucca Mountain model were developed by considering the particle size distributions of local soils as well as the size distributions of suspended soil particles for different receptor exposure conditions. The exposure conditions included very dusty environments with elevated mass loading resulting from active disturbance of the soil surface and the conditions when there are few or no soil disturbing conditions and mass loading therefore is lower. Because larger particles are more likely to be resuspended during soil disturbing activities, the particle size distribution in dusty environments includes larger particles sizes (and therefore less or no enhancement) than those characteristic of low mass loading conditions. Thus, the range of enhancement factors selected for conditions of high mass loading (0.4 to 1.5) was lower that that selected for less dusty conditions (range of 2 to 7)[4]. The environments where the receptor can be exposed to different levels of radionuclides in the air are further discussed in the next section.

Evaluation of receptor exposure conditions and intakes of resuspended soil

Once the radionuclide concentration of suspended particulates in air is calculated, one needs to know the mass of inhaled particles to evaluate intake of those radionuclides by inhalation. The mass in inhaled particles can be calculated from mass loading (mass concentration of particulates in air), time exposed to this mass loading (occupancy), and breathing rate. Each of these parameters may be represented by a single average value representing the entire receptor environment, but usually modelers partition the environment into compartment to account for uncertainty and variation in those parameters at different locations where the receptor can receive inhalation exposure. Inhalation models typically use two compartments that correspond to high and low dust levels, and use corresponding occupancy and breathing rates. The model used for the Yucca Mountain repository is unique in that the receptor environment is divided into five mutually exclusive microenvironments [5, 6]. These microenvironments are associated with activities that result in differences in mass loading, exposure times, breathing rates, and enhancement factors. In the active outdoor environment, people are outdoors involved in activities that generate high levels of dust such as farming, gardening, and walking on unconsolidated soil. The model includes two indoor microenvironments that represent conditions when people are active and inactive (asleep) indoors. A fifth microenvironment is included to account for time receptors would spend traveling or conducting other activities outside of contaminated areas.

Mass loading can differ among these microenvironments by three orders of magnitude or more. To develop mass loading values for the inhalation model appropriate for the conditions in the Yucca Mountain region, site specific and analogue experimental data were used. Consideration was given to the representativeness of these data with respect to local meteorological conditions, soil morphology, site setting, and human activities [6]. The mass of resuspended particles measured in Amargosa Valley during typical soil disturbing activities ranged from about 0.1 to more than 3 mg m^{-3} [6], and concentrations of 10 mg m^{-3} or higher have been measured elsewhere under similar conditions [7]. The mass loading in the active indoor environment is much lower, with a typical value of 0.1 mg m^{-3}. The lowest mass loading occurs when people are asleep (0.03 mg m^{-3}) [6].

To calculate exposure (occupancy) times in each microenvironment, the receptor population was divided into four mutually exclusive population groups based on location and type of work: nonworkers, commuters, local outdoor workers, and local indoor workers. These groups were selected because variation among individual in exposure rates from inhalation of resuspended particles would be influenced primarily by whether people spent their working hours indoors, outdoors, or away from contaminated areas. The proportion of the receptor population in each of these groups was calculated based on information from the U.S. Bureau of Census for the Amargosa Valley census division. From that data, it was estimated that 39% of adult population did not work, 39% commuted to work outside the area, 16% worked locally indoors, and only about 6% were employed locally in occupations that involved outdoor work [5]. The amount of time each group spent in the five microenvironments was estimated from data reported by the U.S. Bureau of Census and from studies of exposure times summarized by the U.S. Environmental Protection Agency [8]. The average exposure time per environment was calculated as the average of exposure times per population group weighted by the proportion of the population in each group. These average exposure times are 0.45 hours per day in the active outdoor environment, 1.45 hours inactive outdoors, 9.45 hours active indoors, 8.3 hours asleep indoors, and 4.35 hours away from contaminated areas [5].

When implemented in the model, the microenvironments and population groups represent behavioral and environmental combinations for which the receptor could receive a substantially different inhalation exposure over a period of one year. Together, they represent variability and uncertainty in the inhalation exposure. All distributions used in the model that characterize receptor's behavior are distributions of annual averages, consistent with the endpoint of the assessment, i.e., the annual dose.

Breathing rates used in the biosphere model were based on the biometric results for adults for different levels of physical activity used in the respiratory track model developed by the International Commission on Radiological Protection [9] for the nominal mix of activity levels in various environments. Breathing rates of adults used in the calculations of inhalation exposure were 1.6 m^3/hr for time spent active outdoors, 1.1 m^3/hr while inactive outdoors and indoors, and 0.4 m^3/hr while sleeping.

Selection of Inhalation Dose Coefficients

Deposition of radioactive aerosols in the respiratory tract, and consequently the inhalation dose, depends, among other factors, upon particle sizes, chemical form of a contaminant, and age of the receptor. Federal requirements for the performance assessment for the Yucca Mountain repository define the receptor as an adult, so the following discussion does not address age effects. Environmental aerosols consisting of resuspended soil particles are polydisperse, i.e., the constituent particles within an aerosol have a range of sizes. The inhalation dose calculations in the model for the Yucca Mountain repository are based on experimentally measured concentrations of total suspended particulates in air (TSP) (mass loading). TSP represents, for the purpose of dose assessment, the mass distribution over the range of particle sizes present in resuspended soil.

Ideally, inhalation dose would be calculated using the actual particle size distribution of radionuclide concentrations in air coupled with the particle size-dependent distribution of inhalation dose coefficients, and by taking into consideration conditions of the receptor's exposure. In practice, the actual distributions of particle sizes in air are rarely available for a scenario under assessment and the dose coefficients are not expressed as particle size-dependent functions. Instead, they usually are expressed as single values for an assumed distribution of activity over particle sizes (usually lognormal) with a given activity median aerodynamic diameter (AMAD) and a geometric standard deviation. In addition, selection of mass loading measurements for use in a model may introduce additional bias because sampling of ambient aerosols is always particle size-dependent and dependent on a particular sampling technique used. It is therefore important to understand limitations of the method and approximations involved.

To minimize the impact of an unknown particle size distribution on estimates of inhalation exposure, data from or representative of an inhalable sampler should be chosen [10]. An inhalable sampler collects particles with a size spectrum corresponding to that of particles that a person can inhale, i.e., all particles that enter the respiratory tract. In this respect, TSP is an appropriate parameter for evaluating inhalation exposure when the specific size distribution of ambient aerosols is not known. The next step in the decision making process is the selection of the appropriate inhalation dose coefficients for the expected distribution of particle sizes. It is generally recommended that for evaluation of doses to the public, dose coefficients for 1-μm

AMAD be used [11]. However, when the exposure is known to have resulted from inhalation of resuspended radioactive aerosols, an AMAD of 5 μm may be more realistic for estimating the doses [12, 10]. The inhalation exposure model for the Yucca Mountain repository currently uses AMAD of 1 μm, which is a common practice, although some participants of the BIOPROTA inhalation task considered particle size distributions with different AMADs (1, 5 and 10 μm).

Dose coefficients for inhalation also depend on the chemical form of a contaminant. Because of the uncertainty in the chemical form of inhaled or ingested material, a conservative assumption was made in the inhalation model for the Yucca Mountain repository regarding the absorption rates for the radionuclides of interest, such that the dose coefficients selected for the use in the model were the highest. For actinides, the highest inhalation dose coefficients are predominantly for the compounds with absorption rates either fast or slow. For radionuclides with a fast absorption rate, measurements of TSP are a good approximation for dose estimates because they are comparable with the particle-size dependent sampling efficiency of an inhalable sampler [10]. For radionuclides with slow absorption rates, measurements of TSP and 1μm AMAD dose coefficients will generally overestimate the inhalation dose.

SUMMARY

In summary, it is important to recognize that site-specific conditions play an important role in constructing conceptual and mathematical models of inhalation exposure. In the model for inhalation exposure to suspended particles of contaminated soil from radionuclide releases from the proposed Yucca Mountain repository, site-specific conditions are reflected in the section of model parameters. Among the models used in the BIOPROTA inhalation task, differences in model results were found to relate significantly to the degree to which human soil disturbance results in high dust levels; the degree of enhancement (enrichment) in radionuclide content of the resuspended dust; and the occupancy period for which high dust levels were assumed to persist. These factors were given a detailed consideration in developing the inhalation model for the proposed Yucca Mountain repository.

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