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A NATURAL ANALOGUE APPROACH FOR ESTIMATING THE HEALTH RISKS FROM RELEASE
AND MIGRATION OF RADIONUCLIDES FROM RADIOACTIVE WASTE.

THOMAS L. GILBERT*

Argonne National Laboratory, Argonne, IL 60439 U.S.A.

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Argonne National Laboratory, Argonne, IL 60439 U.S.A.

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ABSTRACT

The health risks from radioactive waste may be expressed as a sum of products of transfer factors that characterize the causal chain of events between disposal of radionuclides in a waste field and the consequent health effects. "Model" estimates for the transfer factors are commonly obtained by modeling transport and other mechanisms in the subsystems that form the links in the causal chain. "Natural" estimates of some conversion factors for naturally occurring radionuclides can be obtained from data on the concentrations of naturally occurring radionuclides in soil, food, and the human body. These "model" and "natural" estimates can be used with scaling procedures to estimate the uncertainties and to obtain better estimates of the values. The scaling procedures take into account the differences in the source characteristics for radionuclides in a waste field of limited size and for radionuclides generally distributed in the natural environment. The ratios of the natural estimates to the model estimates for several transfer factors and several radionuclides belonging to the U-238 decay series have been determined. These ratios range from 1/8 to 4/1 for food-concentration/source-concentration transfer factors for the food pathways and from 1 to 77 for dose-rate/source-concentration transfer factors for the internal radiation dose pathways to various organs.

INTRODUCTION

Nuclear waste disposal is intended to isolate nuclear waste from the environment in order to minimize the risks of cancer and other effects of exposure to radiation from radionuclides in the waste. Estimates of the effectiveness of this isolation--measured by individual radiation doses from radionuclide releases or direct radiation from the waste--are needed in order to select a site and facility design that will meet performance objectives and provide a reasonable balance between societal risk and cost. Such estimates are based on pathway analyses, which consist of models and procedures for calculating the dose to an individual as a function of time and location due to radionuclides in the waste.

Different models apply to different pathway segments. Some are very simplified and approximate descriptions of complex physical processes. The cumulative error can be quite large--several orders of magnitude--and difficult to estimate. A means for validating a dose estimate (the sum of the partial dose estimates from different pathways) is, therefore, needed. Circumstances for which validation is possible are rare because the doses are very low and indistinguishable from larger contributions from other sources, and the times of interest can be as large as several millenia. Estimates of the dose from disposal of ore-processing residues are one such circumstance, which occurred recently in a pathway analysis carried out for

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the Formerly Utilized Sites Remedial Action Program (FUSRAP) of the U.S. Department of Energy [1]. By scaling the results of a pathway analysis for a typical FUSRAP disposal site to a hypothetical disposal site of global extent, it was possible to obtain model estimates that could be compared directly with natural analogue estimates based on measurements of the concentrations of naturally occurring radionuclides in food and in the human body. The general approach and results are summarized below.

PATHWAY ANALYSIS

Pathway Sums

The dose to any individual in an exposed population may be expressed as a sum of products of transfer factors in the form

$$D(x,y,t) = \sum_{\substack{\text{pathways}(i) \\ \text{radionuclides}(j)}} (D/E)_{ij} \times (E/C)_{ij} \times (C/R)_{ij} \times (R/S)_{ij} \times S_j \quad (1)$$

where S_j are the radionuclide source terms (radionuclide concentrations); R is the rate of release of a radionuclide from the waste field (the region defined by the boundaries of the structure in which the waste is placed) or the intensity of the emitted gamma radiation at the waste field boundary; C is the concentration of a radionuclide in food, water, air, or ground at a point of exposure; E is the exposure rate (pCi/yr of inhaled or ingested radionuclides for internal emitters or the exposure in mR/yr for external radiation); and $D(x,y,t)$ is the dose rate to the whole body or organ of an exposed individual at a location x,y relative to the site at time t .

The ratios D/E , E/C , C/R , and R/S are "transfer factors" that give the relation between the above-defined parameters for different radionuclides and pathways. They are determined by the various mechanisms that control migration and transport of the radionuclides through the different pathways from source to organ, and are functions of the parameters that characterize the facility, environment, and biological processes.

The transfer factors may be decomposed into products of factors that provide a more detailed description of the mechanisms involved (e.g., the R/S factor may be decomposed into factors that correspond to retention by waste-encapsulating material, waste containers, and engineered structures in which the containers are placed), or combined to give overall transfer factors involving several transfer mechanisms. In the application at hand, there are no man-made barriers and releases are by leaching or resuspension of radioactive residues mixed with soil. Under these circumstances, the analysis leads directly to C/S factors, where $C/S = (C/R) \times (R/S)$. The quantity of primary interest, which summarizes the overall results of the pathway analysis, are the source-to-dose transfer factors: $D/S = (D/E) \times (E/C) \times (C/S)$. The E/C factors depend on quantities that can be determined fairly easily and accurately (inhalation and ingestion rates). There will be a separate D/S factor for each pathway and for each radionuclide. Radionuclides with half-lives that are short compared to the transit time in a pathway may be omitted (apart from the contribution to the D/S factor from ingrowth of short-lived decay products).

Pathway Diagrams

The terrestrial and aquatic environmental pathways form a complex, interconnected network through which radiation from the waste and radionuclides released from the waste can reach an individual. Each term

(product of transfer factors) in the pathway sum in Equation 1 corresponds to a pathway from the source (waste field) to the body or an organ of an individual at risk from the waste. Each transfer factor corresponds to a segment in the pathway. Some pathways may have common segments and, therefore, common transfer factors in the corresponding terms in the sum. This structuring of the pathway sum may be represented most clearly by a pathway diagram.

A schematic representation of the major onsite pathways is shown in the pathway diagram in Figure 1. This diagram is a simplification of a more general diagram that includes offsite pathways [1]. The natural analogue approach is limited to onsite exposure because scaling from a typical to a global waste field is not feasible for offsite doses. This is not a severe limitation because the maximum individual dose, which is the primary quantity of regulatory concern, will come from onsite exposure.

FIG. 1. Major Pathways to the Maximally Exposed Individual. Source: Reference 1 (Figure 2.4).

A critical part of a pathway analysis is the identification of the "scenarios" by which exposure to radionuclides or radiation from the waste field is likely to occur. These are idealized patterns of human use of a disposal site for industrial, commercial, residential, or agricultural purposes. During operation of a disposal facility, the primary risk will be from occupational exposure. After closure, there will be a period of institutional control during which exposure will be small and controlled.

The primary concern is exposure following the period of institutional control (assumed to be 100 years), especially in the long term for which it is likely that awareness and records of prior use will be lost.

Scenarios are based on known patterns of human activity and are chosen to correspond to credible circumstances that can be expected to lead to the largest individual dose. Such considerations lead to identification of the "intruder-agriculture" (IA) scenario as the appropriate scenario on which to base the pathway analysis (Reference 1--Section 2.2). In the IA scenario, it is assumed that a family, unaware of the presence of the radioactive waste, builds a home and raises most of its food on the site. This scenario is well adapted to the use of natural analogue methods of analysis because the parameters that define the scenario (Reference 1--Table 2.2) can easily be scaled to correspond to normal circumstances of everyday life to which the data used for natural analogue estimates for the average individual dose are directly applicable.

The pathways identified in Figure 1 that were considered in calculating transfer factors for the IA scenario are listed in Table 1.

TABLE 1. Major pathways to the maximally exposed individual.

Pathway	Description
<u>External Radiation</u>	
Ground radiation	External gamma radiation from radionuclides in the undispersed waste
Air radiation	External radiation from the immersion cloud of suspended radioactive soil particles and radon and its decay products
<u>Internal Radiation</u>	
Inhalation	Inhalation of air containing suspended radioactive soil particles and radon and its decay products
Ingestion:	
Water	Ingestion of drinking water from an onsite or nearsite well
Plant	Ingestion of plant crops raised onsite
Meat	Ingestion of meat from livestock raised onsite
Milk	Ingestion of milk produced onsite
Fish	Ingestion of fish from onsite pond
Soil	Ingestion of contaminated soil by a child

Source: Reference 1 (Table 2.3).

Source Terms

The natural analogue approach is applicable to any naturally occurring radionuclide. This study was limited to dose estimates from U-238 and its decay products. Estimates of the D/S factors were made for U-238 and its longer-lived decay products: i.e., U-234, Th-230, Ra-226, and Pb-210. The

short-lived decay products play an important role in some pathway mechanisms--e.g., the inhalation dose due to Ra-226 is controlled by the transport mechanisms of Rn-222 and its short-lived decay products--and the contribution from ingrowth of short-lived decay products must be taken into account in calculating dose rates. However, these contributions can be included in the estimates of the transfer factors for the long-lived radionuclides, so that the short-lived radionuclides do not appear explicitly in the sum over radionuclides in Equation 1.

The release of radionuclides from the contaminated soil material at a FUSRAP site is controlled by plant uptake, radon emanation, leach rates, and resuspension rates. Details regarding the mechanisms and the models used to describe them are given in the report on which this summary article is based [1]. The rate of transfer through the hydrology pathway, which is of especial interest for materials research because it is dependent on properties of the geological media, is controlled by the leach rate. Two leaching mechanisms were considered: (1) a diffusion mechanism based on data of Nathwani and Phillips [2] for radium-contaminated soils, and (2) an ion-exchange mechanism analogous to that by which radionuclides are carried by groundwater through saturated media. The former mechanism is more realistic, but the data underestimate release rates because they are based on short-term measurements that do not take into account the effects of breakup of ore grains by weathering and other soil-formation processes. The latter mechanism overestimates the release rates. (The release rates for the two mechanisms differ by a factor of about 10^4 .) The ion-exchange mechanism was used for estimating the contribution from the water pathway in order to obtain a conservative estimate.

COMPARISON OF MODEL AND NATURAL-ANALOGUE ESTIMATES OF TRANSFER FACTORS

The model estimates for the transfer factors are based on detailed analyses of the processes involved in the migration of radionuclides and radiation exposure along the different pathways, and make extensive use of models that have been developed for the analysis of release and transport of radionuclides from nuclear power plants and from mill tailings [1, 3-5].

The natural analogue estimates are based on two different kinds of data. One is the average concentration of naturally occurring radionuclides in the soil [6-8] and in food [9-11]. These data lead to natural-analogue estimates of the C/S factors for the food pathways. A comparison of the model and natural-analogue estimates of these factors is given in Table 2. The difference between the estimates for a typical site (assumed to be 2 ha in area with an exposed waste field extending to 1.5 m below the surface) and a global site (assumed to be of global extent) is primarily from the difference in the amount of animal feed grown onsite. For a typical FUSRAP site, half of the feed was assumed to be grown onsite (and contaminated with radionuclides); the other half was assumed to be uncontaminated feed from offsite. For a global FUSRAP site, all of the feed was raised "onsite". In both cases, water for livestock came from a contaminated farm pond.

The soil and food concentration data provide independent estimates only for part of the food ingestion pathways. Some of the data used in the model estimates may have utilized some of the food concentration data; hence, it is not certain that the two estimates are completely independent. However, the analyses and at least some of the data are independent, so that the comparison provides a partial check. There was not sufficient basis to justify use of the natural analogue C/S values as the more accurate generic estimates; hence, a geometric mean of the two estimates was used in subsequent estimates of the D/S factors. Model estimates of the exposure from the remaining pathways are significantly less than model estimates of the exposure from the food ingestion pathways (Reference 1--Tables 5.6-5.8).

TABLE 2. Model and natural C/S transfer coefficients for food.

Radionuclide	C_i/S_i ^{a)}			
	Model Estimates		Natural estimates	Natural/ model ^{b)}
	Typical site	Global site		
Plant foods:				
U-238	2.8×10^{-3}	2.8×10^{-3}	3.7×10^{-4}	0.13
Th-230	4.5×10^{-3}	4.5×10^{-3}	--	
Ra-226	7×10^{-3}	7×10^{-3}	1.3×10^{-3}	0.2
Pb-210	5.3×10^{-3}	5.3×10^{-3}	1.0×10^{-3}	0.2
Meat:				
U-238	3.1×10^{-5}	5.8×10^{-5}	2.5×10^{-4}	4
Th-230	4.2×10^{-5}	8.3×10^{-5}	--	
Ra-226	1.7×10^{-4}	8.8×10^{-4}	1.2×10^{-3}	1.4
Pb-210	2.5×10^{-4}	6.0×10^{-4}	4.6×10^{-4}	0.8
Milk:				
U-238	4.1×10^{-5}	7.1×10^{-5}	2.7×10^{-5}	0.4
Th-230	4.6×10^{-5}	9.1×10^{-5}	--	
Ra-226	2.1×10^{-4}	9.7×10^{-4}	2.5×10^{-4}	0.3
Pb-210	2.7×10^{-4}	6.5×10^{-4}	2.9×10^{-4}	0.5

a) C_i is the average concentration of the i th radionuclide in the food and S_i is the average concentration of the same radionuclide in the soil, both expressed in the same units.

b) Ratio of natural estimates to model estimates for global site.

Source: Reference 1 (Tables 3.19, 3.23, and 3.26).

Apart from the dose to the bronchial epithelium, which is due to inhalation of radon decay products, most of the non-food contributions to the radiation doses are from external radiation from waste in the ground, for which the error in the D/S transfer factors is much smaller than for the other pathways, and from ingestion of soil by a child. Hence, natural-analogue estimates based on data for the concentrations of naturally occurring radionuclides in the soil and in food provide a check on model estimates for most of the dose.

The other kind of data used for obtaining natural analogue estimates is the concentrations of radionuclides in various parts of the body--the so-called "body burden" [6-7, 12-13]. These data can be used to obtain a natural analogue estimate of the internal radiation dose, which is the sum of the dose contributions from all pathways that contribute to ingestion and inhalation of radionuclides. A comparison of the natural estimates with the model estimates is given in Table 3. (The model estimates in Table 3 are based on geometric means of the C/S factors in Table 2.)

TABLE 3. Comparison of model and natural estimates of the D/S factors for internal radiation doses at a global site.

Radionuclide and organ	D/S in (mrem/yr)/(pCi/g)		Natural/ model ratio
	Natural estimate	Model estimate	
U-238 + U-234:			
Whole body	2.8	0.037	77
Bone	12.8	0.63	20
Whole lung	1.5	0.47	3.1
Th-230:			
Whole body	1.9	0.046	41
Bone	9.7	1.7	5.7
Ra-226 and decay products ^{a)} :			
Whole body	15	5.7	2.6
Bone	66	65	1.0
Whole lung	130	97	1.3
Bronchial epithelium	560	515	1.1

a) Including the contribution from Pb-210 and its decay products and $S_{\text{Ra-226}} = 0.80 \text{ pCi/g}$ [8].

Source: Reference 1 (Table 5.5).

The cause of the difference between the model and natural-analogue estimates, which is almost two orders of magnitude for the whole-body dose from U-238, has not been determined. It is probably due to differences in the methods used to estimate the dose from the body burden. A code for calculating dose from intake, based on the ICRP model, was used for the model calculations [4]. Dose estimates for naturally occurring sources were obtained from references in which the burden-to-dose models were not specified [6, 12-13]. A recalculation of the burden-to-dose estimates using the same procedure for both cases will be needed in order to check this speculation.

The comparison in Table 3 does not permit an assessment of the relative errors from different pathways. The model estimates indicate that the dominant contribution for most pathways is from ingestion of plant foods (vegetables, fruits, and grains). The exception is U-238, where half of the whole-body dose and three-fourths of the bone dose are estimated to be from drinking water (Reference 1--Table 5.6). However, it should be noted that the drinking water estimate is based on a leach rate obtained using an ion-exchange model that probably overestimates the leach rate by an order of magnitude or more.

The error in state-of-the-art radiation dose estimates based on analyses of radionuclide transport via terrestrial and aquatic pathways is generally expected to be one to two orders of magnitude, with an overestimate more likely than an underestimate [14]. The natural-analogue/model comparison in Table 3 is consistent with this judgmental estimate. The comparisons in Tables 2 and 3 are based on a generic analysis for which one

must assign average values to environmental parameters that may differ by an order of magnitude or more from one region or location to another. Site-specific pathway analyses, for which parameters can be determined more accurately and more sophisticated models can be used, are usually more reliable than generic analyses. However, site-specific pathway analyses do not lend themselves to a natural analogue approach.

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