RADIONUCLIDE MIGRATION MODEL FOR BURIED WASTE AT THE SAVANNAH RIVER PLANT*

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ABSTRACT

Solid waste has been buried at the Savannah River Plant burial ground since 1953. The solid waste is contaminated with alpha-emitting transuranium (TRU) nuclides, with beta-gamma-emitting activation and fission products, and with tritium. To provide guidance for the current use and eventual permanent retirement of the burial site from active service, a radionuclide environmental transport model has been used to project the potential influence on man if the burial site were occupied after decommissioning.

The model used to simulate nuclide migration includes the various hydrological, animal, vegetative, atmospheric, and terrestrial pathways in estimating dose to man as a function of time. Specific scenarios include a four-person home farm on the 195-acre burial ground. Key input to the model includes site-specific nuclide migration rates through soil, nuclide distribution coefficients, and site topography. Coupled with literature data on plant and animal concentration factors, transfer coefficients reflecting migration routes are input to a set of linear differential equations for subsequent matrix solution. Output from the model is the nuclide-specific decayed curie intake by man. To discern principal migration routes, model-compartment inventories with time can also be displayed. Dose projections subsequently account for organ concentrations in man for the nuclide of interest.

Radionuclide migration has been examined in depth with the dose-to-man model. Movement by vegetative pathways is the primary route for potential dose to man for short-lived isotopes. Hydrological routes provide a secondary scheme for long-lived nuclides. Details of model methodology are reviewed.

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INTRODUCTION

One centrally located solid waste disposal site is used to bury all radioactive solid waste produced at the Savannah River Plant (SRP) and occasional special Department of Energy (DOE) shipments from offsite. This disposal site occupies 195 acres between the two chemical separations areas at SRP, approximately 6 miles from the nearest plant boundary. The original area of 76 acres, which began to receive waste in 1953, was filled in 1972, and operations were shifted to a 119-acre site contiguous to the original area.

The purpose of our ongoing study of radionuclides in buried waste is to address the validity of the current limits for burial of all radionuclides and to provide guidance in developing criteria for future management, surveillance, and control of the burial site in the years following the end of plant operation.

As part of our approach to providing guidance, a mathematical model to simulate the potential movement of radionuclides from buried solid waste, through the environment, to man has been formulated. Model results specify critical pathways for nuclide transport and also estimate projected dose to man from buried solid waste. The radionuclide transport model is formulated to be specific to the SRP burial ground, as governed by the input data. However, the formulation is generic and applicable to the migration analysis of nuclear wastes at other sites, as well as toxic chemical wastes.

DESCRIPTION OF THE BURIAL GROUND

The 195-acre burial ground[1] resides on the Barnwell formation of the Coastal Plain geologic province, about 30 miles southeast of the Piedmont Plateau (the other principal geologic province in South Carolina). The principal surface and near-surface soils are clayey sands or sandy clays, averaging about one-third clay, with some cation exchange capacity (1-5 meq/100 g soil). The mean water table is at a depth of 45 feet, with a fluctuation of about 2 ft/yr. The rate of downward migration of percolate water is 7 ft/yr, and the lateral flow of water in the saturated zone at the water table is 40 ft/yr. The nearest perimeter of the burial site is 0.5 miles from the closest onsite stream (Four Mile Creek). The predominant storage mode is earthen trench burial.

Radioactive waste disposed by shallow land, earthen trench burial is truly a heterogeneous mixture. Examples of materials in storage include: (1) contaminated equipment (obsolete tanks, pipes, jumpers, and other process equipment from the fuel separation plants), (2) reactor and reactor fuel hardware (fuel components and housing; spent Li-Al targets), (3) incidental lab and production wastes (cellulosic and plastic materials, analytical and decontamination residues, spent equipment), (4) chemicals (oil in drums containing absorbents; mercury in one-liter polyethylene bottles; ion exchange resins), (5) off-site materials (tritiated waste from Mound; LANL and Mound ^{238}Pu process waste; debris from two U.S. Military airplane accidents in foreign countries), (6) miscellaneous materials (animal carcasses, building rubble).

For modeling purposes, inventories of key nuclides buried in earthen trenches have been retrieved from computer records. Nuclides of half-life greater than 20 years (^{238}Pu, ^{239}Pu, ^{137}Cs, ^{90}Sr) are of greatest interest for projecting potential environmental transport since they will persist well into the future and are of greatest potential concern. Coupled with recent history of burial ground operations and projections on future waste generation rates, model inventories for these four nuclides have been estimated through the year 2000.

DESCRIPTION OF THE SRL DOSTOMAN MODEL TO PROJECT ENVIRONMENTAL TRANSPORT

The DOSTOMAN model, as developed and coded, makes a compartmentalized simulation of the transport of radionuclides in the environment. The compartments, which represent different portions of the environment, include the nuclide source, soil, vegetation, herbivores, atmosphere, groundwater, surface water, and man (Fig. 1). Movement of radionuclides between compartments is controlled by transfer coefficients, which specify the fraction of radionuclides entering or leaving a compartment during a specified period of time. Time functions account for factors such as institutional control prior to public use. Sources and sinks independent of the natural movement of radionuclides are provided.

The general form of the model equation was developed by ORNL personnel for a study to predict the uptake of selected radionuclide species by cows.[2] Refinements were made to expand the description of the numerous pathways for potential transport and to project radiation doses to man. The DOSTOMAN model employs a single equation that considers only the movement of radionuclides through the system. Such factors as water and wind velocity, erosion, intrusion, resuspension, vegetative decay, etc., are accounted for in the transfer coefficients. This equation accounts for the four factors determining the radionuclide inventory in a compartment: (1) transfer in from other compartments, (2) transfer out to other compartments, (3) source or sink terms, and (4) radioactive decay.

The DOSTOMAN model calculates the radionuclide inventory in each compartment at the end of every time step specified. Because a finite difference technique is used to solve the simultaneous equations, some possibility exists for error to accumulate during a simulation run. This error can be minimized by controlling the specification of the time steps. Subroutines for evaluating the numerical stability are provided in the program: (1) RESID determines the difference (residual) between the calculated values for the right- and left-hand sides of the set of simultaneous equations and (2) MASBAL calculates the state of mass balance of the nuclide inventory at a particular time and compares the total to the sum of initial nuclide inventory values. These data are output with each incremental time-step calculation.

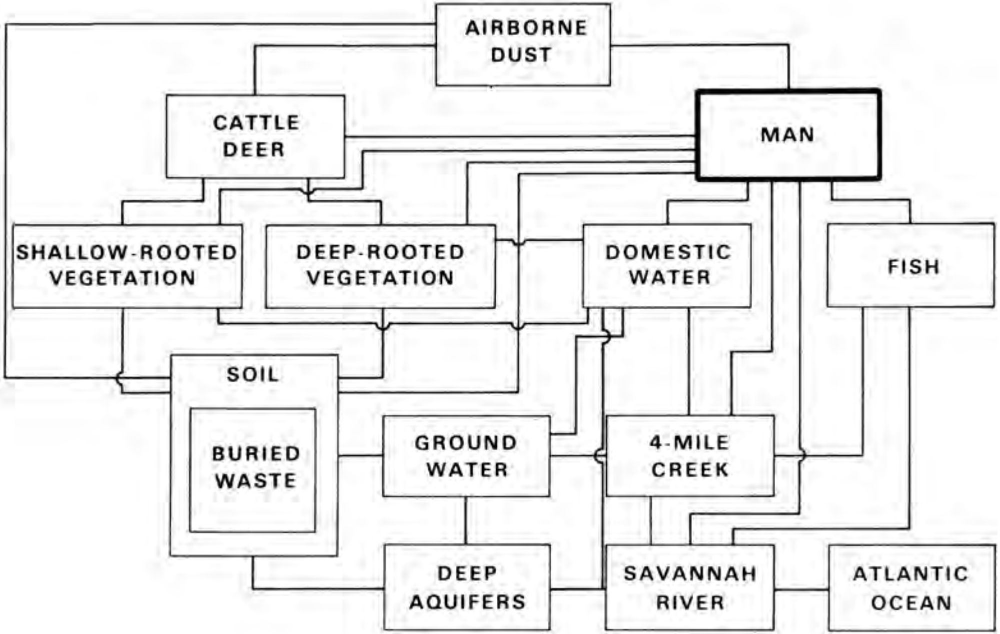

Fig. 1. DOSTOMAN Diagram

DOSTOMAN may be applied to other disposal facilities at other locations - only the initial radionuclide inventories and the transfer coefficients make it site specific. However, the model was intended for application to humid locations - those with relatively high rates of precipitation and shallow water table. The Savannah River Plant has these characteristics.

The data required to run the model include all those factors which influence the rate of movement of radionuclides in the environment. Topographical, hydrogeologic, and geochemical information such as depth to water table, distance to streams, aquifers and rivers, ion exchange coefficients, and bulk density and porosity of the disposal media must be known. The initial radionuclide inventory must be defined. Plant and animal concentration factors must be specified. Physiology of radionuclides in animals and man must be included. Although site-specific data are desirable, considerable information may be obtained from the literature.

DOSTOMAN has seven general data files: (1) initial radionuclide inventory of each compartment, (2) transfer coefficients between compartments, (3) compartment interactions, (4) radioactive decay constant, (5) time-step size, (6) time functions (to account for the delayed presence of man), and (7) sources and sinks.

DOSTOMAN is written in FORTRAN and may be used on any modern computer facility. Core requirements will vary with the number of compartments and time steps being simulated. For example, on the IBM 360/195 at SRL, a simulation involving 200 time steps, 200 transfer coefficients, and 69 separate compartments (a 69 x 69 matrix) requires about six minutes of central processing unit time. About 500 K bytes are requird for such a simulation. Hardware for plotting is desirable. Only one radionuclide can be considered in each run because the decay constant and many of the compartment inventories and transfer coefficients are specific to that radionuclide.

Development work on DOSTOMAN is complete and simulations of several simple test cases have been used to demonstrate the model. It is currently being used to simulate hypothetical post-closure land use scenarios. Reference 3 contains a more detailed description of the model.

The Significance of K_d: The Nuclide Soil/Water Distribution Coefficient

The underground disposal of radioactive wastes is a method to deplete the radiation by storing the wastes for a sufficiently long period of time, in relation to the half-life of the radionuclides, utilizing the capacity of soil to exchange and absorb nuclides. Hence, it is necessary to gain clear understanding of soil dynamics since hydrological transport in soil is the initial pathway by which radioactive substances released from disposed waste migrate. Proper predictions of the migration of radioactive substances underground are mandatory to secure the validity of underground disposal. The distribution coefficient (K_d) of a radionuclide between water and soil is often used as a parameter to reflect the rate of migration of radioactive substances in soils.[4] Using distribution coefficients and the assumption that exchange equilibrium exists and is maintained between the concentration of nuclides in groundwater and that in soil, predictions of the underground migration of nuclides can be achieved. Hence, the distribution coefficient may affect very significantly the safety evaluation of the underground disposal method.

The distribution coefficient will vary[5] depending on such factors as the method of measurement, oxidation state and physical form of the nuclides, coexisting ions in underground water, the chemical stability of coexisting ions, the pH of percolating water, and soil properties (clay and organic content, ion exchange capacity, pH). Generally, two methods of measurement are used to experimentally determine distribution coefficients.[6] The "batch" (static method) and "column" (dynamic measurement) procedures will generally provide a range in the value of K_d to reflect equilibrium or pseudo-equilibrium conditions which exist in the heterogeneous system of nuclides in contact with soil.

Prior studies [4,7] have shown that the distribution coefficient can be related to the time required for the nuclide to move through a soil column, if soil properties such as bulk density, porosity, and linear rate of percolation are known. Hence, for our site specific case of SRP burial ground migration, time requirements for nuclide movement to the groundwater and Four-Mile Creek can be estimated and are, in fact, the basis for some of the time function input to the model. Put in perspective to nuclide decay patterns, this information is useful for a qualitative assessment of the significance of K_d. Figure 2 illustrates such a comparison for the long half-life nuclides of interest to the transport model (^{239}Pu, ^{238}Pu, ^{137}Cs, ^{90}Sr).

If K_d is large (>100: Pu isotopes, ^{137}Cs), movement to the primary bodies of water takes a long time relative to ^{238}Pu or ^{137}Cs half-lives, and radioactive decay can be nearly quantitative before hydrological transport to the groundwater occurs. Qualitatively, groundwater/drinking water pathways for dose to man should be modest and other transport mechanisms have to be operative for significant dose to a population. For nuclides of long half-life (^{239}Pu), hydrological transport to drinking water supplies, as well as other routes, may be competitive with the rate of radioactive decay, leading to multiple pathways for potential dose to individuals. This will be dictated by the value of the distribution coefficient.

If K_d is low (<50), downward movement to underlying groundwater systems may occur at significant rates, and be competitive with the spontaneous decay of nuclides of half-lives >20 years. Under this circumstance, deep soil and groundwater compartments

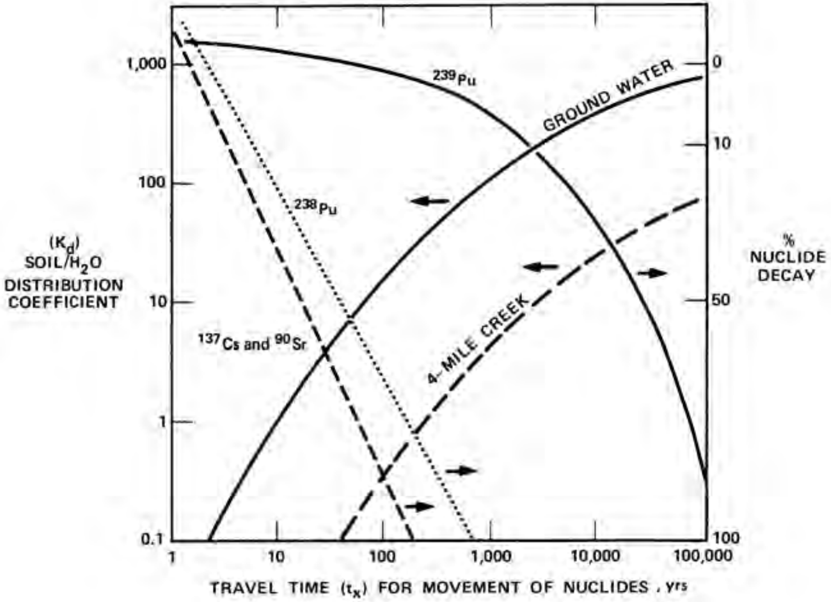

Fig. 2. Nuclide Migration in SRP Soil

should accumulate radionuclides, and drinking water pathways for dose to man may dominate.

K_d values for Cs and Pu isotopes are consistently >500, from a variety of measurements.[4,7] Values for ^{90}Sr are somewhat more uncertain. For example, measurements of ^{90}Sr K_d for SRP soil[7] vary by a factor of 10^2 (5 to 500) depending upon pH, cation content of percolate and soil, the clay content of a given layer of soil, and Sr concentration. More recent measurements[6] for well-characterized SRP soil and well water suggest K_d values for ^{90}Sr at $\leq$50. Japanese workers[4] show an interesting correlation of ^{90}Sr K_d with the cation exchange capacity of the soil. Coupled with data on SRP burial soil exchange capacity,[7] ^{90}Sr K_d values $\leq$50 are implied.

In any case, it should be clear that, in the assessment of risk of potential movement of buried radionuclides, results will be very sensitive to the soil/water exchange equilibria as reflected by the values of the distribution coefficient. For the purposes of this paper to illustrate model projections, the following K_d values have been assumed: 1600 for Pu isotopes, 1000 for ^{137}Cs, and 50 for ^{90}Sr.

LAND USE SCENARIO

The model has been used to project the dose to man from buried inventories of nuclides for the postulated future use of the burial ground as a small home-farm 100 years after burial ground decommissioning. Localized utilization of farm produce (vegetables, fruit trees), cattle (milk, meat), and wild deer (meat) to provide the total sustenance for a family of four residing continuously on the 195-acre burial site is assumed. The family obtains its water supply from a well drilled in the Barnwell formation, which is also used for irrigation. Land utilization is partitioned to a vegetable plot (2 acres), fruit trees (48 acres), and pasture (145 acres), with the pasture supporting 40 cattle. Deer (60) intrude on the farm to forage. The population will occasionally intrude into the buried waste and contaminated soil and routinely breathe the air above the site. Recreation such as fishing or swimming in surface waters, which may be receiving contaminants from the burial ground, is also included. Although this is a limited population scenario, this land use is entirely consistent with historical documents which trace the agricultural community which once existed on the site of the Savannah River Plant.[8]

MODEL PROJECTIONS FOR LONG-LIVED RADIONUCLIDES

Pu Isotopes

Model results for Pu isotopes have been reported previously[9] and are illustrated in Fig. 3 for ^{238}Pu (2600 Ci) and ^{239}Pu (500 Ci) for population curie intake to the bone skeleton (critical organ for Pu) as a function of time. Multiple transport pathways are illustrated here.

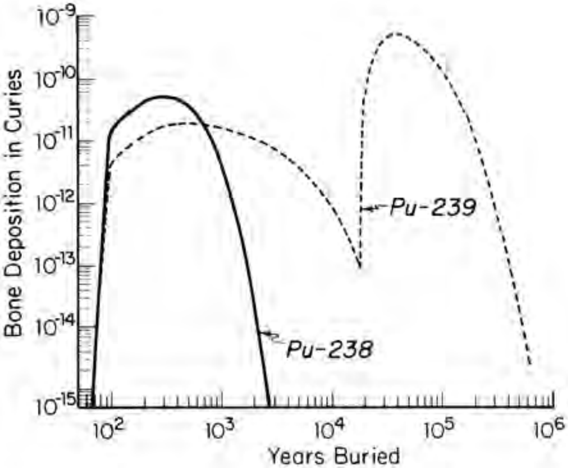

Fig. 3. **Model Projections for Pu Isotopes (Example)**

^{238}Pu shows a peak intake to bone of 60 pCi, corresponding to a maximum dose of 4 mrem/person/year, at approximately 300 years after cessation of burial ground operations. Analysis of model compartment inventories indicates the primary mode of movement is uptake by deep-rooted vegetation with primary dose from fruit consumption and a smaller dose from ingestion of animal stock. Direct intrusion into waste by man makes a minor contribution to dose via ingestion and inhalation of soil containing Pu from waste-to-soil migration and vegetative decay. ^{238}Pu is retained in soil, due to a high distribution coefficient, but decays before hydrological transport becomes significant. Prior studies[10] have shown that, with time, the major risk of ^{238}Pu in the environment is associated with daughter products (i.e., ^{226}Ra) due to decay:

$$^{238}_{94}\mathrm{Pu} \xrightarrow{\alpha} {}^{234}_{92}\mathrm{U} \xrightarrow{\alpha} {}^{230}_{90}\mathrm{Th} \xrightarrow{\alpha} {}^{226}_{88}\mathrm{Ra},$$

since the radiological hazard of ^{226}Ra is significantly greater.[11] By use of the Bateman equation for successive decay,[12] a ^{226}Ra waste inventory of no more than 30 Ci would accumulate with time. Worst-case assumptions on movement through the environment to man result in a dose estimate of <0.1 mRem/person/yr.

^{239}Pu shows a bimodal distribution for bone deposition with time, illustrative of two critical pathways for migration of this radionuclide. A maximum intake of 20 pCi (2 mRem/person/yr) is observed at 400 years after burial operations ceased, due to the deep-rooted vegetation pathway. Eventually, long-lived ^{239}Pu is transported downward into the groundwater and is ingested via the well water supply. This contributes to a peak intake of 600 pCi (130 mRem/person/yr) at 38,000 years.

Fission Byproducts

Model projections for annual intake to the gastrointestinal tract via ingestion, as a function of period of residence, are shown in Fig. 4 for ^{137}Cs and ^{90}Sr. Results are shown graphically for individual intake. Fission byproducts are transported from buried waste primarily by four pathways: hydrologic transport to deep soil, uptake by vegetation, human intrusion, and erosion of the ground surface. Of these, only vegetative uptake leads to a significant dose. Movement by hydrologic transport is slow (particularly for Cs) due to the soil ion exchange characteristics and the low flow velocities in the subsurface. The short half-lives of both nuclides ($\leq$30 years) result in near-quantitative decay in the time frame of reaching domestic water supplies. Erosion is so slow and human intrusion so infrequent that neither contribute significantly to dose.

^{137}Cs is relatively soluble and essentially 100% is absorbed through the gastrointestinal tract[11] and widely distributed by the blood - leading to a whole-body dose from assimilation. Annual dose rises from 18 mRem per person for the first year of residence (101 years since cessation of burial operations) to a peak of 190 mRem per person in the 33rd year of residence. The maximum dose

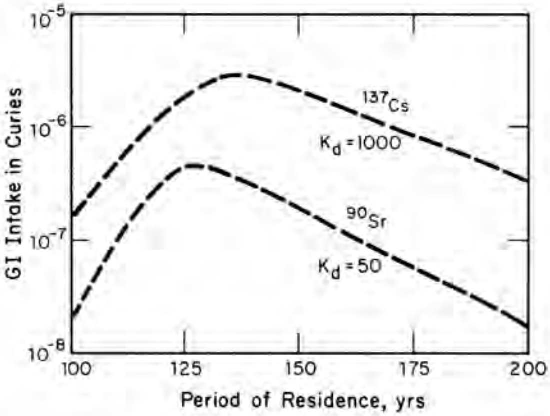

Fig. 4. **DOSTOMAN Projections for Cs/Sr (Example)**

is comparable with average annual dose due to background radiation.[13]

Although the curie intake projections for ^{90}Sr (Fig. 4) are about an order of magnitude less than ^{137}Cs, the dose results are similar. Only a small portion of ^{90}Sr enters the body by absorption through the gastrointestinal tract (2.25%)[11]. That which is absorbed concentrates in bone, the critical organ for ^{90}Sr. Model projections for annual bone dose increase from 13 mRem per person (1st year of residence) to 170 mRem per person (year of maximum intake), again comparable to annual background radiation dose.

Plackett-Burman statistical analysis[14] has indicated the key model parameters, (1) period of institutional control and (2) soil/water distribution coefficients, are two of the more sensitive variables dictating results. For ^{90}Sr, this parametric sensitivity is illustrated in Fig. 5, using a 50-year dose commitment calculation. Based on the assumed K_d values, the model projects that control over the site may be mandatory for as long as 200 years after burial has ceased, to keep the risk of exposure below public standards. The model has also been used to project that viable options to such long surveillance periods include an earthen overburden to the existing site or deeper burial in a new disposal facility. For critical pathways such as vegetative uptake, greater distance between the root zone and waste will diminish the radiological hazard as long as the waste inventory relationship to groundwater is maintained ($\geq$20 ft).

The importance of the distribution coefficient is well illustrated in Fig. 5. Low values of K_d also provide an interesting result. For K_d's <10, rate of downward movement is rapid with ^{90}Sr potentially reaching the groundwater in <100 years. The net effect is accumulation of ^{90}Sr in deep soil (>20 ft) and groundwater. Four Mile Creek would not be subject to ^{90}Sr flows in <400 years, during which time decay is near-quantitative. Burial soil retains less Sr, exposing much less inventory to the vegetative pathway of migration. The dose pathway is the same - vegetative uptake and ingestion - but the available upper soil inventories are reduced.

ANALYSIS OF THE RADIOLOGICAL HAZARD

The lifetime effect of chronic one-year intake of a nuclide, expressed as a 50-year dose commitment, is a more valid assessment of the radiological hazard. In this form, the influence of biological retention in the critical organ and radioactive decay are expressed over the average lifetime of an adult. DOSTOMAN model projections are summarized in Table I with dose commitment results presented in relation to useful standards. For ^{239}Pu, the peak individual dose of 130 mRem/person/yr increases to a lifetime effect of 6 Rem/person due to the long bone retention (72,000 days)[11] and radioactive decay half-life (2.4 x 10^4 years)[11] of that isotope. The ^{137}Cs result (0.2 Rem/ person) is nearly equal to the annual dose since the isotope is rapidly excreted with a biological half-life of 115 days.[11] For ^{90}Sr, the maximum annual

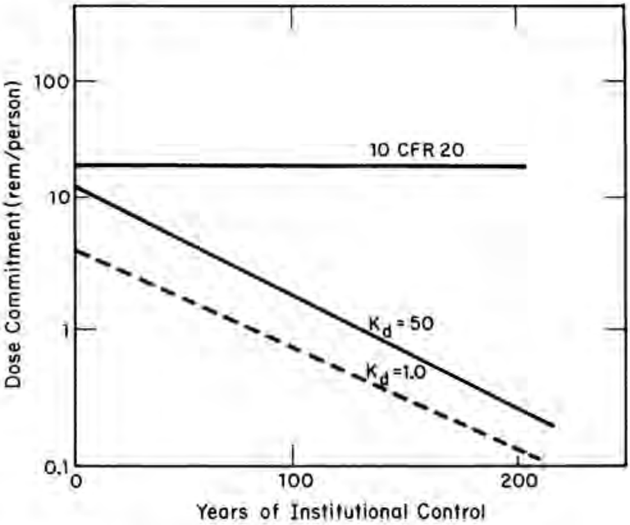

Fig. 5. **^{90}Sr Results as a Function of Model Parameters (Example)**

TABLE I

DOSTOMAN Model Projections for SRP Burial Ground Nuclides
(Assumed K_d 1600 for Pu, 1000 for ^{137}Cs, and 50 for ^{90}Sr)

Nuclide	Maximum Intake to GI Tract (μCi/Person/Year)	Maximum Annual Dose (mRem/Person/Year)	50 Year Dose Commitment (Rem/Person)
^{238}Pu	0.23	4	0.2
^{239}Pu	0.12	2	0.1
	7.6	130	6.0
^{137}Cs	3.0	190	0.2
^{90}Sr	0.50	170	3.8
Background Radiation		100	5
10 CFR 20 Public		500	25
DOE Occupational		5000	250

dose of 0.17 Rem/person/yr increases to a 50-year dose commitment of 3.8 Rem/person due to the long biological half-life of ^{90}Sr in bone (18,000 days),[11] coupled with the influence of an energetic decay daughter (^{90}Y).[11] Although the projected maximum intake by ingestion is about an order of magnitude less for ^{90}Sr, relative to ^{137}Cs and ^{239}Pu, the radiological effect over a lifetime is more significant. The projected lifetime dose commitment results are still comparable to the cumulative effect of background radiation.

SUMMARY

Nuclide transport modeling can be useful as an analytical approach to addressing the question of the validity of burial ground practices. In fact, migration modeling continues to receive industry-wide attention as a method of evaluating currently active commercial sites[15] as well as future locations for institutional wastes,[16,17] to address the nationwide need for regional burial repositories.

The DOSTOMAN code, developed to treat the site specific case of SRP buried waste, will continue to be useful as one of the tools to provide guidance. Subject to choice of exposure scenario, critical pathways for nuclide movement can be defined. In cases where the results are more uncertain due to the nature of the input data, the model has been used to discern which alternatives to burial ground management should receive active consideration (i.e., intermediate depth burial, overburden). Continuing refinements of the model, pathway descriptions, population scenarios, and input data will improve the validity of projections.

The predominance of deep-rooted vegetative uptake as a recurring critical pathway for dose to man clearly suggests immediate site management practices to control such species. This type of control has, in fact, been implemented into the low-level waste management operating procedures on a routine basis.

PROGRAM

The accuracy of DOSTOMAN migration projections receives constant scrutiny. Several programs will be pursued to test this uncertainty. The EPA PRESTO code, developed by ORNL,[15] will be used to project migration with SRP burial ground input data. In addition, DOSTOMAN will be used to project the transport trends for the forage-cow-milk-man scenario[2] for which experimental data are available.

The validity of site specific input data is being examined in field and lab studies. An extensive lysimeter program (mini-burial ground) has been active since 1978 to measure nuclide movement in burial ground soil as a function of waste form. This will provide updated data on waste-to-soil migration rates and distribution coefficients. Direct measurement of deep-rooted vegetative uptake by trees and vegetable crops growing in place on a small portion of the SRP burial ground has been in progress since 1978.

Along with refinements of the nuclides examined to date, future programs will address the risk of burying larger inventories of other fission products. For example, ^{99}Tc will be a saltcrete component from the proposed Defense Waste Processing Facility at SRP. DOSTOMAN projections will be used to address the question of the safe rate of disposal of ^{99}Tc.

Expanded population scenarios are needed to supplement the home-farm treatment. Cases now being planned include a commercial farm, with typical regional crops (corn, soybeans), and a commercial forest with widespread distribution of products.

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