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ALLOWABLE RESIDUAL CONTAMINATION LEVELS OF RADIONUCLIDES IN SOIL FROM PATHWAY ANALYSIS

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STATEMENT OF THE PROBLEM

Remedial actions at Oak Ridge National Laboratory (ORNL) will include well drilling, facility upgrades, and other waste management operations involving soils contaminated with radionuclides. Planners and program managers in ORNL's Remedial Action Program (RAP) will need a way to determine what health risks remain after a proposed cleanup. How much soil will have to be removed? Will 50 or 100 years of institutional control eliminate most of the risk through radioactive decay? Which radionuclides pose the greatest risk and by what pathways might they reach man?

For most radionuclides released into the environment, current regulatory standards (in some instances no applicable standards exist) are based on the concept of limiting the dose from ionizing radiation to humans. Doses to potentially exposed individuals or populations are estimated using models that predict exposures to radionuclides via multiple pathways (e.g., external exposure, inhalation of radionuclides in air, and ingestion of radionuclides in food and water) and calculated doses from those exposures.

We need a method which incorporates models and approaches consistent with those taken by the regulatory agencies [e.g., the Environmental Protection Agency (EPA)] and with the current recommendations of the International Commission on Radiological Protection (ICRP) models. But to be useful, the models must also be flexible enough to encompass the diversity of situations and applications expected to be encountered in the ORNL RAP, without being too complicated for use by planners and program managers.

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GENERAL APPROACH

The uncertainty regarding radionuclide distributions among RAP sites and long-term decommissioning and closure options for these sites requires a flexible approach capable of handling different levels of contamination, dose limits, and closure scenarios. This requirement led to the identification of a commercially available pathway analysis model, DECOM (Till and Moore 1986), which had been used previously in support of remedial activities involving contaminated soil at the Savannah River Plant. The DECOM computer code, which estimates concentrations of radionuclides uniformly distributed in soil that correspond to an annual effective dose equivalent (EDE), is written in BASIC and runs on an IBM PC or compatible microcomputer.

We obtained the latest version of DECOM and modified it to make it more user friendly and applicable to the ORNL RAP. Some modifications involved changes in default parameters (Table 1) or changes in models based on approaches (or anticipated approaches) used by the EPA in regulating remedial actions for hazardous substances under the Comprehensive Environmental Response, Compensation, and Liability Act of 1980 (CERCLA or "Superfund"), as amended by the Superfund Amendments and Reauthorization Act of 1986 (SARA), or under the Resource Conservation and Recovery Act of 1976 (RCRA), as amended by the Hazardous and Solid Waste Amendments of 1984 (HSWA). In addition, we created a version of DECOM as a LOTUS spreadsheet, using the same models as the BASIC version of DECOM.

In the following sections we discuss the specific modeling approaches taken, the regulatory framework that guided our efforts, the strengths and limitations of each approach, and areas for improvement. We also demonstrate how the LOTUS version of DECOM can be applied to specific problems that may be encountered during ORNL RAP activities.

DECOM (BASIC)

In the BASIC version of DECOM, two modes are available to the user. In Mode 1, activities are calculated under the assumption that no data are available regarding the concentration of radionuclides as a function of depth. The user is prompted for the radionuclides to be considered, their relative fractions, pathways to be considered, the EDE standard, and various site-specific parameter values. At each point, the user is given the opportunity to change various default values and, before final calculations, to reconsider initial responses. DECOM then prints on the screen the uniform soil concentrations that would meet the standard and the percentage contributions from each pathway for each radionuclide considered. In Mode 2, the concentration of each radionuclide in soil is entered for depth increments of 15 cm. These values are

Table 1. Default Input Values for DECOM
(Consumption values are dry weights)

Soil bulk density (food pathway)	1.4 g/cm^3
Ground bulk density (external exposure pathway)	1.7 g/cm^3
Thickness of contaminated layer	100 cm
Area of contaminated region	∞
Depth of plowed layer	15 cm
Vegetation consumption by milk cattle	18 kg/d
Vegetation consumption by beef cattle	8 kg/d
Soil consumption by cattle	500 g/d
Consumption of produce	160 kg/y
Consumption of leafy vegetables	15 kg/y
Consumption of soil	40 g/y
Consumption of milk	100 L/y
Consumption of meat	35 kg/y
Consumption of drinking water	500 L/y
Atmospheric soil loading by suspension	.000016 g/m^3
Breathing rate	8000 m^3/y

used to calculate a total EDE from all 15-cm soil layers down to a maximum of 20 layers. If the calculated EDE exceeds the limit specified by the user, DECOM reports how many successive 15-cm layers of soil must be removed for the calculated dose to be less than the standard.

In all calculations, the DECOM methodology assumes that following remediation, land is released for unrestricted use at a user-specified time after cleanup, and possible pathways are: (1) external radiation from the radionuclides in soil, (2) inhalation of suspended soil, (3) ingestion of contaminated food, and (4) ingestion of contaminated drinking water (Figure 1). DECOM assumes that removal of radionuclides from contaminated soils occurs via radiological decay and leaching. The decay constants used in DECOM are based on radiological half-lives compiled by Kocher (1981). Leaching removal constants are calculated according to a model proposed by Baes and Sharp (1983) that is appropriate only for tilled agricultural surface soils. Because the model overestimates the leaching losses from untilled soils, the original version of DECOM was modified to turn off the leaching removal calculation. We suggest that a user of DECOM assume that remedial options will include provisions to significantly reduce or eliminate leaching of radionuclides from RAP sites (e.g., fixation, capping, etc.) and use the code with leaching removal turned off. The following paragraphs briefly describe the pathway models incorporated into DECOM.

Direct Exposure Pathway

In the BASIC version of DECOM, the external exposure calculation assumes that an individual is standing on an infinite slab of soil uniformly contaminated with the user-specified mixture of radionuclides. There are no other pathways of external exposure (e.g., contaminated structures). The code employs a "look-up" table of precalculated values that, in effect, correct the estimated external dose rate at 1 m above a contaminated ground surface for attenuation of gamma radiation by soil. The values in the table are based on numerous runs of a computer code that estimates dose rates for a target 1 m aboveground from radionuclides in infinite slabs of soil at different depths (Kocher and Sjoreen 1985). The basic methodology is documented in the MLSOIL and DFSOIL computer codes (Sjoreen et al. 1984). DECOM allows the user to specify the bulk density of the ground containing the radionuclides and makes a correction from the bulk density of 1.4 g cm^{-3} assumed in MLSOIL and DFSOIL. The user may also specify the fraction of time the exposed person spends on-site (default of 1.0). The annual EDE is calculated by multiplying dose rate by an "external dose conversion factor," which is based on recommendations by the ICRP (1977), but modified to account for contributions from decay products (see below).

A very important consideration for potential users of the DECOM methodology is that DECOM cannot handle radionuclide decay chains explicitly, and as a consequence, the authors of the code attempted to incorporate the effects of daughters into the external exposure calculations. That is, a radionuclide that emits no gamma radiation may, nevertheless, have an external dose calculated for it by DECOM if the radionuclide has decay daughters which emit high energy gamma radiation. As a case in point, ^{234}U, which has a half-life of 244,500 years, decays by emission of alpha particles and very weak gamma radiation. However, DECOM attributes 99% of the dose from ^{234}U to external exposure. This paradox is explained by the fact that it is the gamma radiation from daughters down the decay chain from ^{234}U (not entered separately by the user) that account for the external exposure attributed to ^{234}U. Generally, DECOM assumes that all short-lived daughters of longer-lived parents are in secular equilibrium with the parent, and the authors of the code have used some judgment in modifying accepted external dose conversion factors (ICRP 1977) to reflect such contributions in DECOM. In situations where secular equilibrium should not be assumed (e.g., contamination from processed fuels) or where buildup of daughters through time may be important, the user is cautioned to make appropriate adjustments to the original source term or take

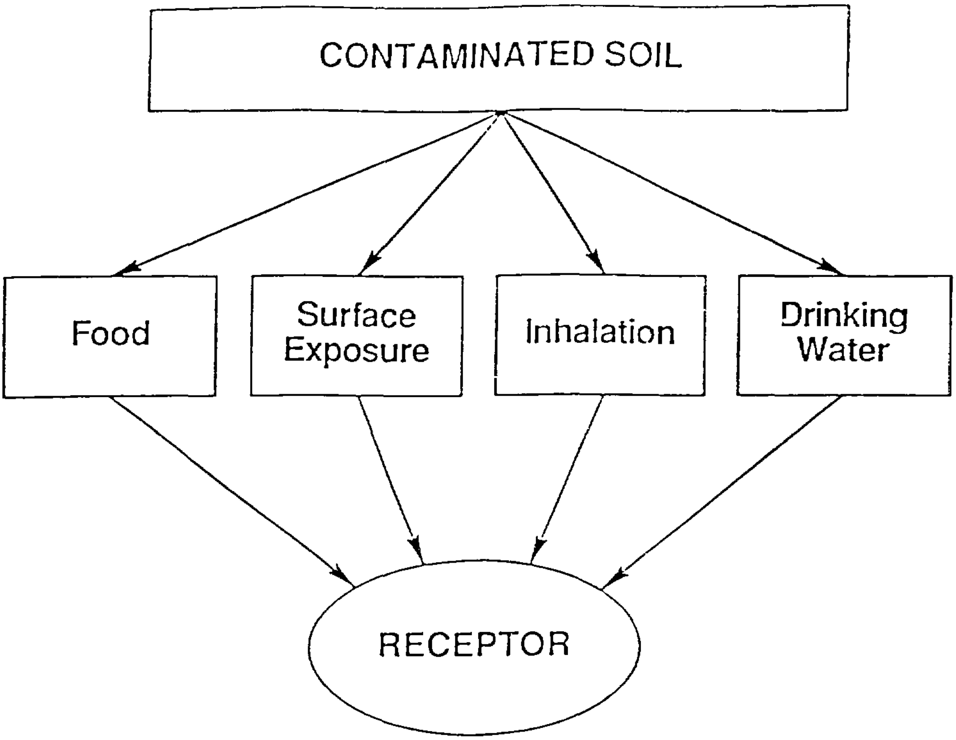


Figure 1. Pathways in DECOM

such considerations into account when interpreting DECOM's results.

Inhalation Pathway

The annual EDE (mrem) via the inhalation pathway is calculated by multiplying the following parameters: (1) the concentration of radionuclides in soil (pCi/g), (2) an atmospheric mass loading factor (g/m^3), (3) the inhalation rate (m^3/y), (4) the dose conversion factor for inhalation (mrem/pCi), and (5) the fraction of time spent on-site (unitless). That is, doses are assumed to occur by inhaling soil suspended into the atmosphere (specific activity is assumed to be the same for all particle sizes). The critical parameter in this calculation is the mass loading factor, which in turn depends on type of soil, vegetation cover, and average wind velocity. That is, mass loading would be expected to be greater in arid, windblown areas than in lush, wet areas. The default mass loading factor in DECOM is 1.6×10^{-5} g/m^3 (Baes et al. 1984a). This value is the geometric mean of values (range 5×10^{-5} to 3×10^{-4} g/m^3) for the 2.5 to 15 μm diameter particle fraction collected with high volume samplers by the EPA (Suggs et al. 1981).

Food Ingestion Pathway

The annual EDE (mrem) via ingestion is calculated for the following food categories: (1) leafy vegetables, (2) produce, (3) milk, and (4) beef. Milk and beef are assumed to be from cows and cattle grazing pastures and fed grains produced on-site. For leafy vegetables, produce, forage, and cattle feeds, the concentration of radionuclides in soil (pCi/g) is multiplied by a soil/plant concentration ratio (unitless) for either vegetative portions of plants (B_v) or reproductive portions of plants (B_r). This approach is basically the approach taken in the computer code TERRA (Baes et al. 1984b), and the concentration ratios used as default are based on a review and analysis of many studies of plant uptake of radionuclides and trace metals from soil (Baes et al. 1984a).

Concentrations of radionuclides in beef and milk are estimated via a modification of the approach adopted by the U.S. Nuclear Regulatory Commission (NRC 1977); i.e. cattle are also assumed to ingest soil while grazing pastures. The daily ingestion rate of radionuclides by milk cows and beef cattle (pCi/d) is calculated by multiplying the concentration of radionuclides in forage and feed (pCi/kg) by feed ingestion rates (kg/d) for milk cows and beef cattle and adding the product of the soil concentration of radionuclides (pCi/g) and a soil ingestion rate (g/d). The soil ingestion rate used in DECOM is based on a review of the subject by Zach and Mayoh (1984). The forage and feed ingestion rates used in DECOM are based on an analysis performed by Shor et al. (1982). The estimated daily ingestion rate of radionuclides by milk cows and beef cattle is then multiplied by either a milk-beef transfer factor (d/kg) that relates the ingestion rate of a radionuclide to its equilibrium concentration in milk or beef (Ng 1982).

Once concentrations of radionuclides in the food items have been calculated (pCi/kg), the consumption rate of the food item (kg/y) is multiplied by the dose conversion factor for ingestion (mrem/pCi) and the fraction of the food item consumed that is grown on-site (unitless). Consumption rates for leafy vegetables, produce, milk, and beef used in DECOM are based on a review of the 1977-1978 U.S. Department of Agriculture nationwide food consumption survey (Nelson and Yang 1984).

Drinking Water Pathway

The basic calculation for estimating the annual EDE for the drinking water pathway in DECOM is to multiply: (1) the concentration of radionuclides in drinking water (pCi/L), (2) the consumption rate of drinking water (L/y), (3) the dose conversion factor for ingestion (mrem/pCi), and (4) the fraction of drinking water from on-site (unitless). The critical parameter in this calculation is the estimation of radionuclide concentration in drinking water. In all models that make such calculations, a soil-water distribution coefficient, K_d (mL/g), is used in the calculation. The default K_d values in DECOM are based on a review of measured K_ds for radionuclides and trace metals in agricultural soils (Baes and Sharp 1983). The review, however, indicated large uncertainties (several orders of magnitude) associated with K_d values owing to effects of soil type, organic matter content, chemical form of the radionuclide, soil pH, and other variables. Additionally, considerations of transport of radionuclides to aquifers and dilution effects make selection of a generic drinking water model extremely difficult.

The original BASIC version of DECOM contained a single drinking water pathway model based on a spill-dilution transport model (Codell and Duguid 1983). This model essentially moves a pulse of radionuclides in leachate from the contaminated soil to an aquifer and then dilutes the pulse in the total volume of the aquifer in order to calculate the concentration of radionuclides in drinking water. A second, and much more conservative model was added to the BASIC version of DECOM based on the approach that the EPA uses in regulating final closure of hazardous waste land disposal units under RCRA (EPA 1987). Although RCRA does not regulate radionuclides, the EPA continues to adopt strategies that make the various regulations dealing with hazardous substances consistent with each other. In other words, remedial action strategies adopted under CERCLA and SARA for radionuclides are very likely to be based on strategies for demonstrating cleanup of hazardous wastes under RCRA.

Under the RCRA closure strategy, demonstration of no harm to human health and the environment requires very conservative models if wastes and residues remain on site. When the closed unit is to be released for unrestricted use, the model that the EPA uses for demonstration of no harm assumes that exposure occurs within the unit boundary, takes no consideration for control measures (e.g., fencing or capping), and allows no consideration for attenuation during transport. Therefore, we added a second, highly conservative model that estimates the concentration of radionuclides in pore water by dividing the concentration of radionuclides in soil (pCi/g) by the soil-water distribution coefficient, K_d (mL/g). This pore water is assumed to be the drinking water consumed by the on-site inhabitant.

For most radionuclides, the drinking water pathway often makes only a small contribution to the total annual EDE to a person living on a decommissioned site. In situations where the drinking water pathway makes a significant contribution, the large uncertainties associated with estimating leachate concentrations, transport, and dilution in a generic model should be considered. If the second drinking water option is selected, the extremely conservative nature of the model should be taken into account. A user of DECOM might consider omitting the drinking water pathway and using a more sophisticated model.

DECOM (LOTUS)

The BASIC version of DECOM has several strengths: (1) it is interactive, (2) it runs on an IBM PC or compatible with as little as 64K memory, and (3) it has been tested with satisfactory results at the Savannah River Plant (Till and Moore 1986). Unfortunately, the original code is cumbersome to use because a separate run (beginning with the first screen and continuing through each option offered) is required each time a single parameter is changed. Additionally, DECOM back-calculates to an initial soil concentration rather than the reverse, calculating a dose from an observed soil concentration – a more useful application for the RAP. Typically, a user would know radionuclide concentrations in soils based on field and laboratory analysis and would be interested in the effectiveness of various remedial options in limiting the potential dose to persons who might inhabit the area after decommissioning the site.

To remedy these problems, we entered the DECOM models into a LOTUS spreadsheet. The spreadsheet version of DECOM has the advantages that: (1) calculations are made in the forward direction, with the user entering radionuclide concentrations and the program automatically recalculating the dose each time a parameter is changed, (2) the effect of varying a single parameter is easily tested, allowing the user to conduct a sensitivity analysis, and (3) the graphics package built into LOTUS can be used to display results. We also added an option in the spreadsheet not available in DECOM BASIC. In the LOTUS version, the user may identify depths to the top and bottom of the contaminated layer of soil with the implicit assumption that soil above the top of contamination is a cap of clean soil. This option allows the user to examine the effects of cap thickness on external doses. We also included in the LOTUS version an option that allows direct ingestion of contaminated soil in the food ingestion pathway (poorly washed vegetables).

We are convinced that both the spreadsheet and BASIC versions of DECOM provide an easy-to-use dose assessment model. However, the user must keep in mind the limitations of the models incorporated into DECOM. Dose assessment models are limited by the data used, and many of the parameters built into the model are approximations, mathematical measures of central tendency of a distribution of measured values (often derived from different sources, each more or less representative of the parameter as defined by the model), or best guesses. The user is encouraged to examine the effect of varying parameter values on the results, and, of course, to replace default values with site-specific measurements.

EXAMPLE APPLICATIONS OF DECOM

The examples discussed in this section were designed to illustrate the ease and flexibility of DECOM. They are not intended to represent most probable exposure scenarios for the Oak Ridge Reservation (ORR).

The Jogger

Suppose that a jogging enthusiast jogs one half hour per day along the Oak Ridge Reservation's perimeter 365 days a year. What is annual effective dose equivalent that DECOM predicts? The food and drinking water pathways are turned off, and the total exposure time is about 80 h/y. We will assume that the jogger's breathing rate is doubled (8000 m^3/y is the default) and that he kicks up a lot of dust (atmospheric mass loading is 1.0×10^{-4} g/m^3).

Eighteen soil samples collected around the ORR perimeter (Williams et al. 1987) were analyzed by gamma spectroscopy (Table 2).

For this simple example DECOM calculates a total annual EDE of less than 1 mrem. This small dose from the soil is mainly by external radiation. We conclude that the jogger need not alter his route.

The Farmer

Now let us consider a slightly more complicated example. Assume that after 100 years of institutional control the Oak Ridge Reservation is released for unconditional use. Further assume that a homesteader settles at the present

Table 2. Jogger Scenario

Radionuclide	Activity(pCi/g)	Annual EDE (mrem)
^{137}Cs	0.77	0.06
^{40}K	10.0	0.04
^{232}Th	1.9	0.8
^{238}U	1.0	0.03

location of a storage pad near building 3503 (selected only because we have data for the soil there), and that there has been no remediation at this site. The farmer lives there year round, and this is where he grows all his crops, and raises all his livestock.

A gamma survey (Williams et al., 1987) found that most of the contamination at this site is concentrated in a few "hotspots". Sixty-nine soil samples where taken at the site and analyzed by gamma spectroscopy and neutron activation. The area-weighted average radionuclide activities in soil are presented in Table 3. Radionuclides present in negligible amounts have been omitted.

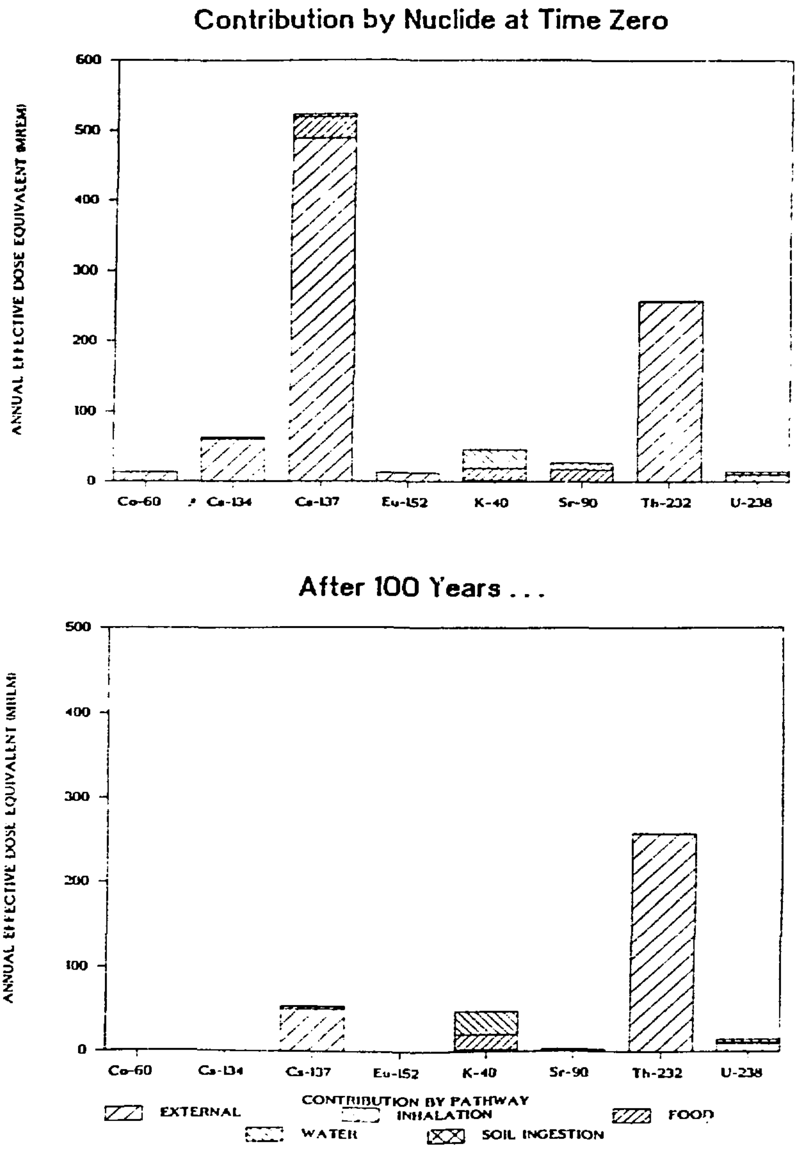
Table 3. Farmer Scenario

		Annual EDE	
Radionuclide	Activity (pCi/gm)	t=0 y (mrem)	t=100 y (mrem)
^{60}Co	0.86	13	0.0
^{134}Cs	6.8	63	0.0
^{137}Cs	140	520	53
^{152}Eu	1.7	12	0.1
^{40}K	16	47	47
^{232}Th	13	260	260
^{238}U	13	14	14

DECOM's predictions (Figure 2) show that both at the present and in 100 years; the primary pathway is from external radiation. While ^{137}Cs is the most important contributor at present, it is relatively short lived, and in 100 years the daughters of ^{232}Th predominate (this assumes secular equilibrium, see earlier discussion). The natually occurring ^{40}K and and residual ^{137}Cs are of lesser importance but still contribute significantly to the annual EDE. Given these results, remediation is advised before the farmer is allowed to settle.

CONCLUSIONS

BASIC DECOM and LOTUS DECOM are potentially useful tools for the RAP planners and program managers. They are easy to use and run quickly on an IBM PC or compatible. By varying input values such as the radionuclide concentrations, thickness of cap layer, period of institutional control, etc., the user can explore a series of "what ifs" to gain an appreciation of the relative importance of the model parameters.



Contribution by Nuclide at Time Zero

After 100 Years . . .

Figure 2. Contribution by radionuclide to the annual effective dose equivalent after 0 and 100 years (Farmer Scenario).

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