EFFECT OF RADON DOSE ON CLEANUP CRITERIA AND USING RESRAD FOR CHEMICAL RISK ASSESSMENT

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ABSTRACT

The U.S. Department of Energy has used RESRAD, a pathway analysis program developed at Argonne National Laboratory, in conjunction with the as low as reasonably achievable (ALARA) principle to develop site-specific residual radioactive material guidelines (cleanup criteria) for many sites. This study examines the effects of the radon pathway, recently added to the RESRAD program, on the calculation of uranium, radium, and thorium cleanup criteria. The results show that the derived uranium guidelines will not be affected by radon ingrowth considerations. The effect of radon on radium and thorium generic guidelines is more significant, but the model does indicate that at the generic soil limits used for radium and thorium the indoor radon decay product concentrations would be below the 0.02 working level standard.

This study also examines the feasibility of applying RESRAD to chemical risk assessment. The results show that RESRAD can perform risk assessment of toxic chemicals after simple modifications. Expansion of the RESRAD database to include chemical compounds will increase its capability to handle chemical risk assessments.


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INTRODUCTION

Argonne National Laboratory has developed a pathway analysis computer program, RESRAD, for the U.S. Department of Energy (DOE) (1). Since its release in June 1989, RESRAD has been used to derive soil cleanup criteria for uranium and other radionuclides and to estimate dose due to residual radioactive material in soil. Recently, a radon pathway was added to RESRAD. Radon-222 is a decay product in the uranium-238 and uranium-234 chain and therefore including ingrowth of radon-222 in the calculations could affect the derivation of uranium criteria.

The RESRAD code was designed for the analysis of sites contaminated with radioactive materials, but many of these sites also are contaminated with hazardous chemicals. Currently, there is no other computer code equivalent to RESRAD that can analyze risks to DOE-related environmental restoration activities resulting from disposal of hazardous chemicals. Therefore, it is desirable that a similar approach applicable to nonradioactive hazardous chemicals be developed.

The purpose of this paper is (1) to assess the possible effects of radon on the development of cleanup criteria for uranium, radium, and thorium; (2) to study and demonstrate the applicability of RESRAD to chemical risk assessment; and (3) to identify additional pathways and databases that are needed for chemical risk assessment.

EFFECT OF RADON DOSE ON CLEANUP CRITERIA

DOE has developed site-specific residual radioactive material guidelines (cleanup criteria) for sites identified by the Formerly Utilized Sites Remedial Action Program (FUSRAP) and the Surplus Facilities Management Program (SFMP). In the derivation of uranium guidelines, seven pathways in RESRAD were considered: (1) external radiation from contaminated soil; (2) internal radiation from inhalation of contaminated dust particles; (3) internal radiation from ingestion of plant foods grown on site and irrigated with water drawn from an on-site well or pond; (4) internal radiation from ingestion of meat from livestock fed with fodder grown on site and water drawn from an on-site well or pond; (5) internal radiation from ingestion of milk from livestock fed with fodder grown on site and water drawn from an on-site well or pond; (6) internal radiation from ingestion of fish from a nearby pond; and (7) internal radiation from drinking water from an on-site well or pond.

Adding radon-222, a decay product in the uranium-238 and uranium-234 chain, as a pathway could affect the derivation of uranium cleanup criteria. Under current guidelines, indoor radon is regulated independently of other exposure pathways. Therefore, it is not considered a direct component of the dose assessment that is conducted to derive radionuclide soil cleanup guidelines. Rather, specific concentration limits are provided for protection from exposure through this pathway (5 pCi/g surface and 15 pCi/g subsurface for radium and thorium in soil, and 0.02 working level [WL] for radon-222 decay products in indoor air) (2). The DOE guidelines require that properties be decontaminated to levels that are as low as reasonably
achievable and that are lower than these concentration limits. In this study, the possible effects of radon on the development of cleanup criteria for uranium, radium, and thorium were assessed.

Approach

RESRAD has been used to derive uranium guidelines for many FUSRAP sites (3, 4, 5, 6). Several site-specific RESRAD data files that were used to derive such guidelines were rerun with the addition of the radon pathway. A dose limit of 100 mrem/yr was used in the derivation (7), and both indoor and outdoor radon dose contributions were considered. The time that the affected individual spent indoors or outdoors depended on the exposure scenario (industrial or residential) and was the same as previously assumed for the external radiation and inhalation pathways. A time frame of 1,000 years was considered. The lowest allowable concentration that will give a dose of 100 mrem/yr within 1,000 years was considered the reference cleanup criterion (100 mrem in a year represents the greatest allowable dose from the nonradon radionuclides). The reference cleanup criteria derived with and without the radon pathway then were compared. It should be noted that the actual cleanup guidelines set by DOE usually are much lower than the reference cleanup criteria because potential exposure scenarios, land use, and the as low as reasonably achievable (ALARA) principle are considered in the decision process. RESRAD also was run for radium-226 and thorium-230 to examine the effect of radon on generic criteria (5 pCi/g surface and 15 pCi/g subsurface).

Results

The RESRAD rerun results with the radon pathway included were compared to results that were obtained without the radon pathway. Table I contains the results of the analysis for a FUSRAP site. The range of derived radionuclide guidelines, with and without radon dose, for different exposure scenarios (industrial or residential) are reported in Table I. The percent reduction of the guidelines derived with radon dose also are listed in the table. Within the 1,000-year time frame, the guidelines for uranium were reduced 0-2.8% when the radon pathway was considered in the dose limiting assessment. The reduction depended on the exposure scenario and varied from one site to another. Adding the dose contributions of radon-222 to the other contributing pathways and radionuclides used to derive uranium-238 guidelines would produce negligible effects because the quantity of radon-222 ingrowth from uranium-238 is small.

Similar procedures were used for the radium and thorium isotopes. The results also are listed in Table I. The radon dose produced more significant effects in the derived guidelines for radium and thorium for the site studied. The reduction of guidelines for radium and thorium ranged from 2.5% to 18% over the 1,000-year period, depending on the exposure scenario. It should be noted that, although the derived guideline for radium-226 can be 60% lower than the generic limits, the model indicates that the generic limits still should ensure that the site meets the limit, adopted from Environmental Protection Agency (EPA) standards, of 0.02 WL indoor radon decay product (2). It is noted that the radon pathway is not directly applicable to the dose limits derived under current DOE directives. It is necessary to assess the impacts of cleanup levels lower than the generic soil limits under ALARA. The addition of this pathway to the code provides a tool to aid that analysis.
Using ResRAD for Chemical Risk Assessment

The RESRAD computer code is designed to perform risk analysis of a radioactively contaminated site. On many contaminated sites, survey results show the coexistence of toxic chemicals in the soil. To perform a parallel risk analysis for the coexistent toxic chemicals, software that adopts the consistent methodology used in RESRAD is desired. Therefore, the feasibility of applying RESRAD to the risk analysis of toxic chemicals was studied.

In RESRAD, the calculated results for radioactive materials are in terms of total dose (mrem/yr) and dose/source ratio [(mrem/yr)/(pCi/g)] -- the ratio between total dose and the original soil concentration -- during different years after decontamination for each radionuclide. The intermediate results during the calculations also are printed in the output data files. For chemical risk assessment, following EPA instructions (8), the average daily intake rate (mg/kg/day) during the exposure period and the lifetime average daily intake rate (mg/kg/day) should be predicted to facilitate the calculation of the hazard index and the excessive cancer risk for chronic toxic chemicals or carcinogens. Intake routes through inhalation or ingestion are considered separately. Therefore, when using RESRAD for chemical risk prediction, it is necessary to convert the results from total dose and dose/source ratio to average daily intake during exposure and lifetime average daily intake rate.

Pathways Consideration

There are eight environmental transport pathways considered in the RESRAD code: external exposure, inhalation of dust, ingestion of plants, ingestion of meat, ingestion of milk, ingestion of aquatic foods, ingestion of water, and inhalation of radon gas. Of these pathways, inhalation of contaminated dust particles and ingestion of plant, meat, milk, fish, and water also are applicable to chemical risk assessment. Therefore, intake rates for inhalation or ingestion can be derived from RESRAD output for these pathways. External ground radiation and inhalation of radon gas are unique to radionuclides; results from these pathways are irrelevant to chemical risk assessment.

Besides the inhalation and ingestion pathways mentioned above, there are other pathways characteristic of toxic chemicals that are not considered in RESRAD. These pathways include absorption through dermal contact while taking showers or swimming and inhalation of volatile vapors from chemicals in the soil or in water drawn from a nearby pond or well. These two pathways may be important for some toxic chemicals and should be included in chemical risk assessment. Using existent methodology, intake rate predictions for these two pathways can be developed and incorporated into RESRAD’s calculational structure.

Selection of Chemicals

RESRAD’s current database consists of 50 radionuclides in their elemental forms. In order to perform risk assessment for chemical compounds as well as for radionuclides, the database needed to be expanded. To demonstrate the applicability of RESRAD to chemical risk
assessment, elements that the EPA, after laboratory experiments and observation, has declared to cause adverse health effects or cancers in the human body were selected. These materials were manganese, nickel, lead, and antimony.

Manganese was found to cause respiratory symptoms and psychomotor disturbances in occupational workers when ingested in daily diet. The reference dose for manganese set by the EPA for inhalation is $1.1 \times 10^{-4}$ mg/kg/day; for oral ingestion, it is $1.0 \times 10^{-1}$ mg/kg/day (9). The reference dose is the limiting amount above which some sort of adverse health effect might develop and is a conservative number set by the EPA with consideration of uncertainty factors. Nickel was found to cause cancer and to reduce body and organ weights in experimental rats fed nickel sulfate constantly for two years. The EPA oral reference dose for nickel is $2 \times 10^{-2}$ mg/kg/day; the inhalation slope factor is 0.84 (mg/kg/day)$^{-1}$ (9). The slope factor can be interpreted as the possibility of cancer development per unit intake rate -- 1 mg/kg/day. Lead is known to cause liver and neuronal damage from laboratory experiments with rats. The reference doses for lead set by the EPA for oral ingestion and inhalation are $1.4 \times 10^{-3}$ mg/kg/day (10) and $4.3 \times 10^{-3}$ mg/kg/day (9), respectively. From experiments in putting antimony potassium tartrate in rats’ drinking water, antimony was found to cause cancer development, life span decrease, and altered blood chemistry. The reference dose for oral ingestion of antimony is $4.0 \times 10^{-4}$ mg/kg/day (10). However, no slope factor was recommended by the EPA for antimony.

Calculation of Intake Rates

RESRAD calculates the effective dose of a radionuclide with the following equation:

$$(Dose)_{i,p}(t) = DCF_{i,p} \times ETF_{i,p}(t) \times SF_{i}(t) \times S_{i}(0)$$

where $DCF_{i,p}$ (mrem/pCi) is the dose conversion factor; $ETF_{i,p}(t)$ (g/yr) is the environmental transport factor, defined as the ratio between the annual intake rate (pCi/yr) and the soil concentration (pCi/g) of radionuclide $i$ at time $t$ (yr); $SF_{i}(t)$ is the source factor, defined as the ratio of the soil concentration of radionuclide $i$ at time $t$ to the concentration at time 0; and $S_{i}(0)$ is the soil concentration at time 0. There are two factors affecting the soil concentrations of radionuclides. One is the ingrowth and decay process unique to radioactive materials, and the other is the leaching process caused by infiltration of rain and irrigation water. Therefore, the source factor $SF_{i}(t)$ is written in Eq. (2) as the multiplication of two factors -- the ingrowth and decay factor, $ID_{i}(t)$, and the leaching factor, $LF_{i}(t)$:

$$SF_{i}(t) = ID_{i}(t) \times LF_{i}(t)$$

When considering the toxicity of hazardous materials instead of the radiation intensity of radioisotopes, the ingrowth and decay process is not applicable. Instead, chemical and biological degradation may take place. Because information is absent about the degradation half-lives in soil phase for manganese, nickel, lead, and antimony, the influence of degradation on soil concentration is neglected in the analysis. The average daily intake rate of chemical species $i$ in terms of mg/kg/day can be calculated from RESRAD output results as:
\[ \text{Intake}_i(t) = \sum_p \text{ETF}_{i,p}(t) \times [\text{SF}_i(t)/\text{ID}_i(t)] \times S_i(0) / (70 \times 365) \quad (3) \]

The division of source factor \( \text{SF}_i(t) \) by ingrowth and decay factor \( \text{ID}_i(t) \) results in leaching factor \( \text{LF}_i(t) \), which when multiplied by \( S_i(0) \) (the initial soil concentration) gives the value of the soil concentration at time \( t \). The number 70 is used based on the assumption that the average body weight is 70 kg, and 365 is used to convert the annual intake rate to the daily intake rate. If the soil concentration \( S_i(0) \) is expressed in mg/g for chemicals, then the unit of the intake rate will be mg/kg/day in Eq.(3). For carcinogens, the lifetime average daily intake rate can be derived by multiplying the intake rate in Eq.(3) by \( 9/70 \); nine years is the average time for a person to live on site, i.e. the exposure period, and 70 years is the average life span, according to EPA publication (11). The oral intake rate of chemical species \( i \) should take into consideration the intake rate through ingestion of plant, meat, milk, water, and fish. These multiple considerations explain the summation sign in Eq.(3). For inhalation intake rate, with the current version of RESRAD code, only the pathway for inhalation of contaminated dust particles should be considered.

The hazard index and excessive cancer risk can be calculated with the use of reference dose (RfD) and slope factor:

\[ \text{(Hazard Index)}_{i,p} = \frac{\text{(Intake)}_{i,p}}{\text{(RfD)}_{i,p}} \quad (4) \]

\[ \text{(Cancer Risk)}_{i,p} = \frac{\text{(Intake)}_{i,p}}{\text{(slope factor)}_{i,p}} \quad (5) \]

where \( i \) stands for chemical species \( i \), and \( p \) is the contamination pathway of oral ingestion or inhalation.

Example and Results

A hypothetical site with an area of 10,000 m\(^2\) is considered contaminated by the disposal of manganese, nickel, lead, and antimony. The thickness of the contaminated zone is 2 m with a homogeneous concentration of 1 mg/g for each chemical. No cover material exists above the contaminated zone, and the groundwater table is 6 m below the ground surface. A pond is located at the edge of the contaminated site, and the pump intake depth below the groundwater table is 10 m for a well located at the down gradient edge of the site.

It is assumed that a farm family lives on the contaminated site; they raise vegetables and livestock for their needs and consume fish caught from a nearby pond. Of the plant food consumed by the farm family, 50% is grown on site. The adjacent pond provides 50% of their aquatic food, and an on-site well provides 100% of their drinking water and irrigation water. The farmer spends 50% of his or her time indoors on site, 25% outdoors on site, and 25% off site. The indoor dust level is assumed to be 40% of the outdoor dust level.

Based on these assumptions, RESRAD calculated the environmental transport factors \( \text{ETF}(t) \) for each pathway at different times. The source factors \( \text{SF}(t) \) and ingrowth and decay
factors ID(t) at different times also were provided in the RESRAD output file. The average daily intake rate through oral ingestion or inhalation then was calculated using Eq.(3). According to RESRAD calculation results, the breakthrough times of groundwater contamination were 1,601, 800, 801, and 1,097 years for manganese, nickel, lead, and antimony, respectively. Both the oral and inhalation intake rates had maximum values at time 0 for these chemicals. Intake rate from the plant ingestion pathway contributed most to the total oral intake rate throughout the 1,000-year time frame. Details of the calculated intake rates are listed in Tables II and III.

The maximum hazard indexes as calculated with the use of Eq.(4) are: 1.4 (inhalation and oral ingestion) for manganese, 3.6 (oral ingestion) for nickel, 170 (inhalation and oral ingestion) for lead, and 100 (oral ingestion) for antimony. When the soil concentrations are 1 mg/g for each chemical at the hypothetical site, the exposed resident farmer might develop adverse health effects in this scenario. The maximum excessive cancer risk caused by inhalation of dust contaminated with nickel, as calculated with the use of Eq.(5), is $3.1 \times 10^6$.

CONCLUSIONS

The effects of including indoor radon with other pathways in deriving guidelines for uranium-238 and uranium-234 would be minimal within the 1,000-year time period studied for the FUSRAP and SFMP sites. DOE, using RESRAD and ALARA principles, has established uranium limits for cleanup that are much lower than those calculated on the basis of a 100 mrem/year limit. Therefore, sites for which uranium guidelines already have been established will not be affected by radon ingrowth considerations.

The effects of radon dose on radium and thorium, however, are more significant. Radon ingrowth, applied to the dose limit, could result in guidelines that are 60% lower than the generic limits. However, as noted, the model also indicates that the generic guidelines still should ensure that sites meet the 0.02 WL indoor radon decay product limit adopted from EPA standards (2). The new pathway is a useful tool to aid ALARA analyses for selection of radium and thorium limits.

This study shows that RESRAD can be used in risk analysis of hazardous chemicals. RESRAD’s current calculational structure makes it easy and feasible to modify the program to produce the desired results for chemical risk assessment. Expansion of the RESRAD database to include chemical compounds will expand its capability to handle risk assessments of toxic chemicals. The addition of pathways applicable to chemical wastes, such as absorption of chemicals through dermal contact while taking showers or swimming and inhalation of volatile chemical vapors from contaminated soil or water, will provide users with the option of performing a complete chemical risk analysis. Including chemical or biological degradation models specific to hazardous chemicals would improve prediction accuracy.

The modified RESRAD code provides a consistent methodology for risk analysis of toxic chemicals comparable to that for analysis of radioactive materials in a site contaminated with mixed waste. The results thus obtained also will provide a clearer picture of the total cost/risk of cleaning up a chemical-radiological comingled site.
REFERENCES


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<td>U-238</td>
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<td>Ra-226</td>
<td>2.2-32</td>
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<td>Th-230</td>
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<td>5.7-79</td>
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<td>Th-232</td>
<td>6.4-21</td>
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<td>2.5-4.9%</td>
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TABLE II  Average Oral Daily Intake Rate (mg/kg/d)\(^a\) for Soil Concentration of 1 mg/g

<table>
<thead>
<tr>
<th>Time (yr)</th>
<th>Manganese</th>
<th>Nickel</th>
<th>Lead</th>
<th>Antimony</th>
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<td>(2.4 \times 10^{1})</td>
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\(^a\)All values are reported to two significant figures.

\(^b\)Data from Integrated Risk Information System (IRIS).

\(^c\)Data from Public Health Risk Evaluation Database (PHRED).
TABLE III  Average Inhalation Daily Intake Rate (mg/kg/d)\(^a\) for Soil Concentration of 1 mg/g

<table>
<thead>
<tr>
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<th>Lead</th>
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\(^a\)All values are reported to two significant figures.

\(^b\)Values were obtained by multiplying the reference concentration (mg/m\(^3\)) listed in IRIS by an inhalation rate of 20 m\(^3\)/d and dividing the product by a body weight of 70 kg.

\(^c\)Data from IRIS.

\(^d\)ND = no data available for inhalation RfD.

\(^e\)Data from PHRED.