

Benchmarking the New RESRAD-OFFSITE Source Term Model with DUST-MS and GoldSim - 13377

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ABSTRACT

RESRAD-OFFSITE is a computer code developed by Argonne National Laboratory under the sponsorship of U.S. Department of Energy (DOE) and U.S. Nuclear Regulatory Commission (NRC). It is designed on the basis of RESRAD (onsite) code, a computer code designated by DOE and NRC for evaluating soil-contaminated sites for compliance with human health protection requirements pertaining to license termination or environmental remediation. RESRAD-OFFSITE has enhanced capabilities of modeling radionuclide transport to offsite locations and calculating potential radiation exposure to offsite receptors. Recently, a new source term model was incorporated into RESRAD-OFFSITE to enhance its capability further. This new source term model allows simulation of radionuclide releases from different waste forms, in addition to the soil sources originally considered in RESRAD (onsite) and RESRAD-OFFSITE codes. With this new source term model, a variety of applications can be achieved by using RESRAD-OFFSITE, including but not limited to, assessing the performance of radioactive waste disposal facilities.

This paper presents the comparison of radionuclide release rates calculated by the new source term model of RESRAD-OFFSITE versus those calculated by DUST-MS and GoldSim, respectively. The focus of comparison is on the release rates of radionuclides from the bottom of the contaminated zone that was assumed to contain radioactive source materials buried in soil. The transport of released contaminants outside of the primary contaminated zone is beyond the scope of this paper.

Overall, the agreement between the RESRAD-OFFSITE results and the DUST-MS and GoldSim results is fairly good, with all three codes predicting identical or similar radionuclide release profiles over time. Numerical dispersion in the DUST-MS and GoldSim results was identified as potentially contributing to the disagreement in the release rates. In general, greater discrepancy in the release rates was found for short-lived, fast-moving radionuclides than for long-lived, slow-moving radionuclides.

INTRODUCTION

Radioactive sources within the contaminated zone could consist of waste materials such as sludge that was dispersed directly within soils, or waste materials such as activated metals that were contained by drums or canisters and buried underground. The radionuclides contained or imbedded in waste materials have the potential to dissolve in water when water infiltrates soils or enters the waste containers through cracks or holes and makes contact with the waste materials. Upon dissolving in water, the radionuclides could be either carried directly downward through the soil column, or carried outside of the containers and then downward through the soil column, to the bottom of the contaminated zone where further transport through the underlying unsaturated zone(s) toward the groundwater aquifer would begin.

The release rates of radionuclides would depend on the interaction between the contaminants, waste materials, and water; therefore, the release rates would be different for different types of waste materials. Three release options are currently provided by the RESRAD-OFFSITE [1] new source term model to simulate the release of radionuclides to surrounding soils in the contaminated zone: (1) first-order release — the release rate of radionuclides is proportional to the inventory in waste materials,

(2) equilibrium desorption release — the release rate is controlled by a linear equilibrium partitioning between the solid and aqueous phases, and (3) uniform release — the nuclides are released from a constant fraction of the waste form over time. To evaluate the performance of the new source term model, two computer codes, DUST-MS [2] and GoldSim (the RT [radionuclide transport] module) [3], were employed to generate radionuclide release rates at the bottom of a contaminated zone for comparison with those from RESRAD-OFFSITE. The comparison involves five cases under study featuring different radionuclides, source dimensions, release mechanisms, and water infiltration rates.

The DUST-MS code has been developed by Brookhaven National Laboratory for more than a decade. Its predecessor, DUST, which stands for Disposal Unit Source Term, was first available in 1993 [4]. In subsequent years, DUST was expanded to evaluate multiple species (MS) and was issued as DUST-MS in 2001 [2]. Another version, DUSTMS-D, which allows consideration of distributed failure of waste containers, was also issued in the same year [5]. The DUST codes (DUST, DUST-MS, and DUSTMS-D) are mostly used for evaluating radionuclide release rates from disposal units buried underground; the release rates then can be input to a groundwater transport model for further evaluation of potential groundwater contamination. The DUST code that was used for comparison with RESRAD-OFFSITE results was DUST-MS.

The GoldSim code is a commercially available software designed as a general purpose Monte Carlo simulator for modeling complex systems in business, engineering, and science [6,7]. It is equipped with a graphical user interface that helps users build, link, and dynamically simulate their models. GoldSim provides several specialized extension modules that provide additional features or functionality for particular applications. One of these, the Contaminant Transport Module for Radionuclides (RT Module) [3], allows users to simulate the transport of contaminants in the environment and was used to generate source release rates for this comparison.

CONCEPTUALIZATION OF RADIOACTIVE SOURCES WITHIN THE CONTAMINATED ZONE

To model the release of radionuclides from waste materials, RESRAD-OFFSITE conceptualizes the waste materials being distributed evenly over the depth of the contaminated zone, as depicted in Figure 1. After being released from waste materials, radionuclides are assumed to be dispersed evenly in the lateral direction and to be transported vertically to the bottom of the contaminated zone. While moving through the surrounding soils, adsorption/desorption between radionuclides and soil particles would occur. As a result, radionuclide concentration in the surrounding soils would change with depth, which is depicted by the gradual darkening of the color for soil in Figure 1.

Unlike RESRAD-OFFSITE, which analytically solves the mathematical equations describing the transport of radionuclides in soils, DUST-MS implements a numerical analysis method called finite difference (FD) to solve the same radionuclide transport equations. To use the FD method, users are required to subdivide the contaminated zone with a number of grids, which can be associated with release from radioactive waste materials. The distribution of the waste materials into these grids is determined by the users, with the amount over the grids adding up to the total inventory. Upon leaving the waste materials, radionuclides are assumed to partition between solid and liquid phases in equilibrium in the surrounding soil and to transport downward from one grid to another. The conceptualization of radioactive sources within the contaminated zone by DUST-MS is also shown in Figure 1. The transition of the radionuclide concentration profile with the RESRAD-OFFSITE modeling is continuous over the depth of the contaminated zone, whereas the transition of the concentration profile with the DUST-MS modeling is step-wise.

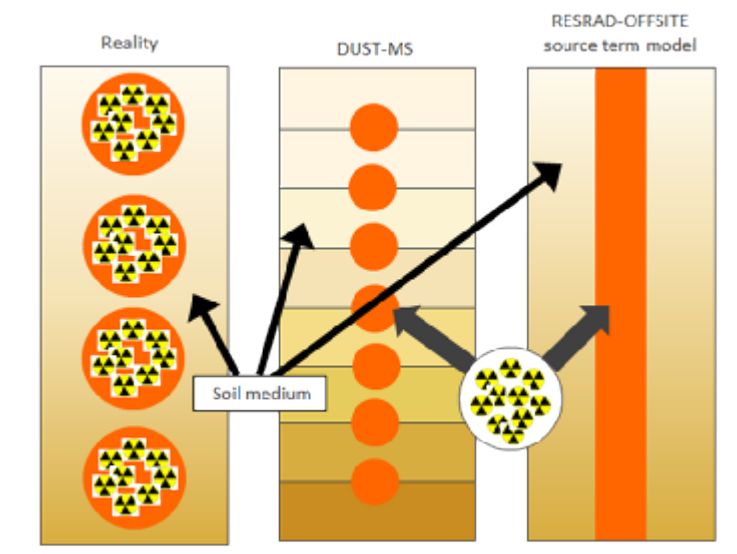


Fig. 1 Conceptualization of Radioactive Sources within the Contaminated Zone

The GoldSim RT module solves for the movement of contaminant mass through the environmental system. The user can create an environmental system by defining a network of transport pathways, which are connected through mass flux links. For this comparison, an initial mass of source-containing radionuclides was created and linked with cell, aquifer, and pipe elements to construct source release and transport models for comparison with RESRAD-OFFSITE. The Cell element is mathematically equivalent to a mixing cell and can be used to explicitly represent the partitioning of contaminants among different phases/media, with the constraint of solubility limits. The Aquifer element is intended to represent a feature that essentially behaves as a fluid conduit. It can be used to simulate vertical transport through an unsaturated zone or horizontal transport in a saturated zone. The Aquifer element internally uses a set of linked Cell elements during the simulation, which are subsequently removed at the end of the simulation. In other words, the simulation approach used by the Aquifer element is similar to the finite difference approach used by DUST-MS, which subdivides the domain of analysis into multiple grids and assumes homogeneity for the solid and liquid phases, respectively, within each grid. The intention for the Pipe element is the same as that for the Aquifer element. Mass is considered to enter at one end of the conduit, and travel through and disperse within the conduit before exiting at the other end of the conduit. However, the Pipe element uses a Laplace transform approach to provide analytical solutions to the equations governing one-dimensional transport. This simulation approach is similar to that used by RESRAD-OFFSITE, which also solves the governing equations analytically.

SETTING THE SAME RELEASE CONDITIONS

As described above, the RESRAD-OFFSITE new source term model provides three options for estimating the release rates of radionuclides from waste materials to the surrounding soils – first-order release, equilibrium desorption release, and uniform release.

The first-order release considers the release rate of each radionuclide to be proportional to the remaining inventory of that radionuclide in the waste materials. The proportionality constant is characterized by a

leach rate specified by the user. Waste materials that resemble this type of release may include dewatered sludge or resins used for ion-exchange.

The equilibrium desorption release assumes that all radionuclides would distribute between the solid and liquid phases of the surrounding soils from the beginning of simulation on the basis of equilibrium desorption. This assumption implies that radionuclides would be available for release from the waste materials and dissolve in water immediately when they are in contact with water. For this scenario to occur, radionuclides would have to attach to the waste materials loosely and, most likely, on the surface. Waste materials that fit this specification may include compacted laboratory trash, such as clothes or glove boxes, as well as small gadgets or tools.

The uniform release option considers the dissolution of a constant fraction of the initial radionuclide inventory over time. However, because of the adjustment for radiological ingrowth and decay, the actual radioactivity released may be different as time progresses. To be eligible for this option, a fixed portion of the waste materials would need to dissolve in water or disintegrate upon contact with water each year, if the distribution of radionuclides throughout the waste materials is homogeneous. This dissolution or disintegration of waste materials would then release the radionuclides contained or imbedded within them. An example of waste materials that may fit these descriptions is activated metal, which would corrode in the environment and release the imbedded radionuclides.

When calculating radionuclide release rates with DUST-MS, the dissolution mechanism was used to simulate the uniform release option considered in RESRAD-OFFSITE, while the rinse release mechanism was used to simulate the equilibrium desorption option. Because there is no provision in DUST-MS that can be used to simulate the first-order release option in RESRAD-OFFSITE, comparisons of RESRAD-OFFSITE and DUST-MS results are limited to the two preceding options.

When calculating radionuclide release rates with the GoldSim RT module, a Source element, in conjunction with a Cell and a Pipe element within the Source, was used to simulate the first-order release option, as well as the uniform release option considered in RESRAD-OFFSITE. The equilibrium desorption option in RESRAD-OFFSITE was simulated by imbedding the waste material in a Pipe or an Aquifer element to calculate the release rate of radionuclides under the condition of dispersion or no dispersion, respectively.

DEVELOPMENT OF RELEASE SOURCES

For the comparison of radionuclide release rates, five different cases involving different source dimensions and radionuclides were developed. The radionuclides could be imbedded in any specific waste forms, which are used interchangeably with “release sources” and “source materials” in this paper. The first three comparison cases, designated as Cases I, II, and III, involve radionuclides in the source material with a thickness of 0.3 m. For Case I, the source was assumed to contain Tc-99. For Case II, the source was assumed to contain Cs-137, which has a significantly shorter half-life than that of Tc-99. For the third case, Case III, the source was assumed to contain U-234, which would subsequently decay to Th-230, Ra-226, Pb-210, and Po-210. For Cases IV and V, the source was assumed to be 3-m thick; in Case IV, the source contains Tc-99, and in Case V, the source contains U-234. Because transport of radionuclides was considered only in the vertical direction, the size of the cross sectional area would not affect the concentration profile of radionuclides over the thickness of the contaminated zone. Therefore, an area of 1 m² was assumed for all of the release sources. The initial concentration of radionuclides in each source was assumed to be 3.7 Bq/g (100 pCi/g). In addition to the dimensions and initial concentration levels, the density of the waste form was assumed to be 1.5 g/cm³, resulting in an initial

total radioactivity of 1.67×10^6 Bq (4.5×10^{-5} Ci) for Cases I, II, and III, and 1.67×10^7 Bq (4.5×10^{-4} Ci) for Cases IV and V.

All of the waste forms were assumed to start releasing radionuclides at time 0 as a result of water infiltration. A water infiltration rate of 0.4 m/yr was assumed for Cases I and II, whereas a smaller water infiltration rate of 0.1 m/yr was assumed for Cases III, IV, and V. Table 1 summarizes the assumptions made for the release sources in the five comparison cases. Although in this comparison, the release rates of radionuclides to the surrounding soil are calculated starting at time 0 — meaning that immediate breaching of the waste containers occurs — a container integrity time period during which no release would occur can be considered by RESRAD-OFFSITE.

Table 1. Assumptions for the Release Sources in Different Comparison Cases

Parameter	Case I	Case II	Case III	Case IV	Case V
Area (m ²)	1	1	1	1	1
Thickness (m)	0.3	0.3	0.3	3	3
Density (g/cm ³)	1.5	1.5	1.5	1.5	1.5
Initial radionuclide	Tc-99	Cs-137	U-234	Tc-99	U-234
Initial concentration (Bq/g)/(pCi/g)	3.7/100	3.7/100	3.7/100	3.7/100	3.7/100
Water infiltration rate (m/yr)	0.4	0.4	0.1	0.1	0.1

RESULTS OF COMPARISON

Results for Case I

Case I considers releases from a 0.3-m Tc-99 source. Annual release rates of Tc-99 in terms of pCi/yr were calculated over 500 years. The release rates were calculated with no dispersion, as well as with different levels of dispersion in the soil column. However, the differences between the RESRAD-OFFSITE results and those of DUST-MS and GoldSim show similar trends with different levels of dispersion; therefore, in this paper, the comparisons are presented for only one selected level of dispersion.

Figure 2 shows the comparison between the RESRAD-OFFSITE results and DUST-MS results under the equilibrium desorption release condition with no dispersion. Figure 3 presents the comparison between the results of these two codes under the uniform release condition, also with no dispersion. For the uniform release, the waste form was assumed to disintegrate consistently with a constant fraction of 0.01/yr for 100 years. As shown in these two figures, observation can be made that, regardless of the soil K_d value of Tc-99, the DUST-MS results would approach those of RESRAD-OFFSITE as the number of grids used in the DUST-MS calculation was increased. On the basis of this observation, the discrepancy in the release rates calculated by these two codes could be caused in part by numerical dispersion associated with the DUST-MS results. This inference is made because numerical dispersion associated with the implementation of a numerical analysis method tends to decrease with increasing subdivision of the analysis domain, and DUST-MS implements the finite difference method in its modeling.

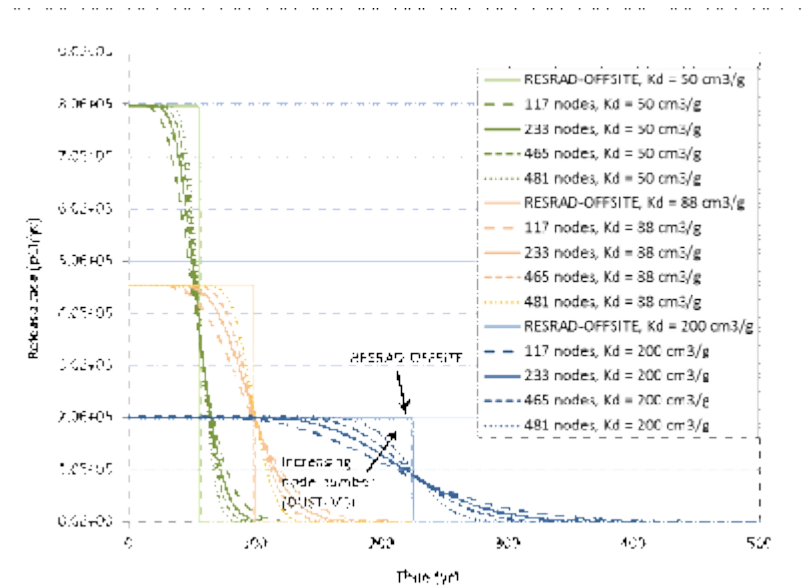


Fig. 2 Comparison of RESRAD-OFFSITE and DUST-MS Results for Case I Concerning Release from a 0.3-m Tc-99 Source under the Equilibrium Desorption Condition with No Dispersion

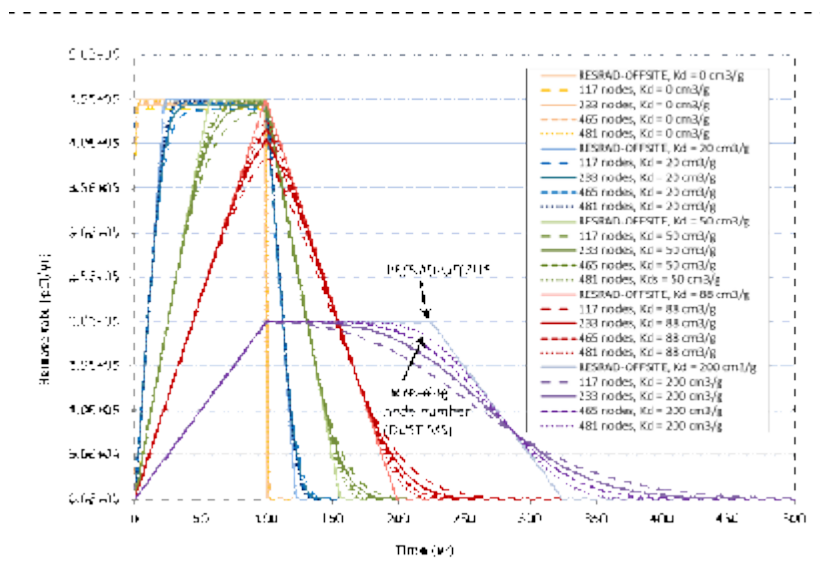


Fig. 3 Comparison of RESRAD-OFFSITE and DUST-MS Results for Case I Concerning Release from a 0.3-m Tc-99 Source under the Uniform Release Condition with No Dispersion

Figure 4 compares the RESRAD-OFFSITE, DUST-MS, and GoldSim results obtained with a soil K_d value of $50 \text{ cm}^3/\text{g}$ for Tc-99 and with no dispersion. The GoldSim results for the equilibrium desorption release were calculated with the use of an Aquifer element, which simulates the transport of dissolved species with multiple linked Cell elements and inevitably introduces numerical dispersion into its solution. For the uniform and first-order releases, the GoldSim results were obtained with the use of a Pipe element, which implements a Laplace transform approach to provide analytical solutions. However, a small dispersivity was automatically introduced to the solution for the Pipe element (as indicated by a Run Log warning message in the GoldSim run) even though the no-dispersion condition was specified to run the RT module. The discrepancy between the results of GoldSim and RESRAD-OFFSITE for the equilibrium desorption release could be contributed by the numerical dispersion associated with the GoldSim results. For the uniform and first-order releases, the discrepancy between the results of GoldSim and RESRAD-OFFSITE could be contributed by the small dispersivity automatically introduced to the GoldSim solutions. Overall, the agreement between the results of RESRAD-OFFSITE and those of DUST-MS and GoldSim is considered quite favorable.

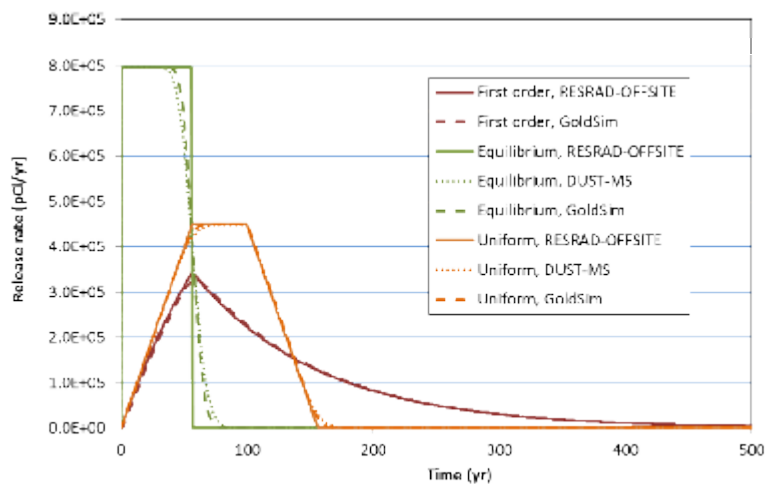


Fig. 4 Comparison of RESRAD-OFFSITE, DUST-MS, and GoldSim Results for Case I Concerning Release from a 0.3-m Tc-99 Source with No Dispersion

Figure 5 presents the comparison of the results from the three computer codes for the same study case as discussed in the preceding paragraph but with a dispersivity of 0.03 m. The RESRAD-OFFSITE results agree better with the results of DUST-MS than with the results of GoldSim. However, except for the much sharper rise and decline at the beginning of the equilibrium desorption release associated with the GoldSim results, the agreement among the results of the three codes is considered to be fairly good.

Results for Case II

Case II considers the release of Cs-137 from a 0.3-m source. Cs-137 was selected for comparison because of the shorter half-life of 30 years compared to Tc-99's half-life of 2.13×10^5 years. Consequently, the influence of half-life on the release rates would be more pronounced for Cs-137 as compared to Tc-99.

Although multiple calculations each with a different soil K_d value for Cs-137 were performed, only the results with a K_d of $200 \text{ cm}^3/\text{g}$ are represented here. The DUST-MS results were obtained with a total of 481 grids distributing in the domain of analysis.

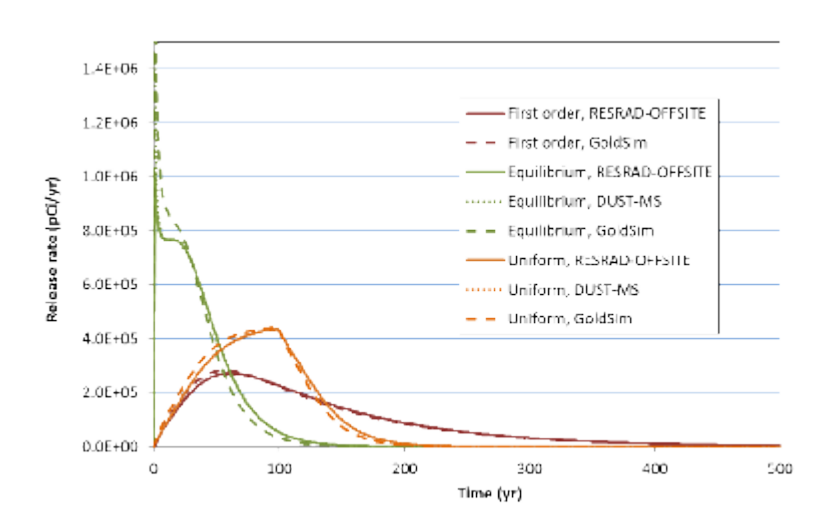


Fig. 5 Comparison of RESRAD-OFFSITE, DUST-MS, and GoldSim Results for Case I Concerning Release from a 0.3-m Tc-99 Source with a Dispersivity of 0.03 m

Figure 6 compares the calculation results from RESRAD-OFFSITE, DUST-MS, and GoldSim for the no-dispersion condition. For an equilibrium desorption release, if radioactive decay during the transport in the contaminated zone is negligible, the release rate of radionuclides leaving the bottom of the contaminated zone would stay constant for a certain period of time, that is, the time period required for the radionuclides to transport from the top of the contaminated zone to the bottom of the contaminated zone, as seen in Figures 2 and 4 for Tc-99. However, because of the short half-life of Cs-137 and hence the significant radioactive decay in the contaminated zone, the release rate at the bottom of the contaminated zone cannot be maintained at the initial level for a period of time; rather, it would decrease from the beginning until all Cs-137 radionuclides leave the contaminated zone.

For the uniform release, even though the release fraction specified for the calculations is 0.01/yr so that the constant disintegration of waste material would last for 100 years, the amount of Cs-137 dissolved in water each year would decrease noticeably because of radioactive decay. As a result, the calculated release rate of Cs-137 would start to decline before 50 years rather than at the expected 100 years, if radioactive decay is negligible. The significant radioactive decay in the contaminated zone also explains the earlier decline in the release rate of Cs-137 associated with the first-order release.

The agreement among the RESRAD-OFFSITE, DUST-MS, and GoldSim results shown in Figure 6 is excellent because the three curves almost collapse together. The same situation is found with the calculated results of the three codes with a dispersivity of 0.03 m.

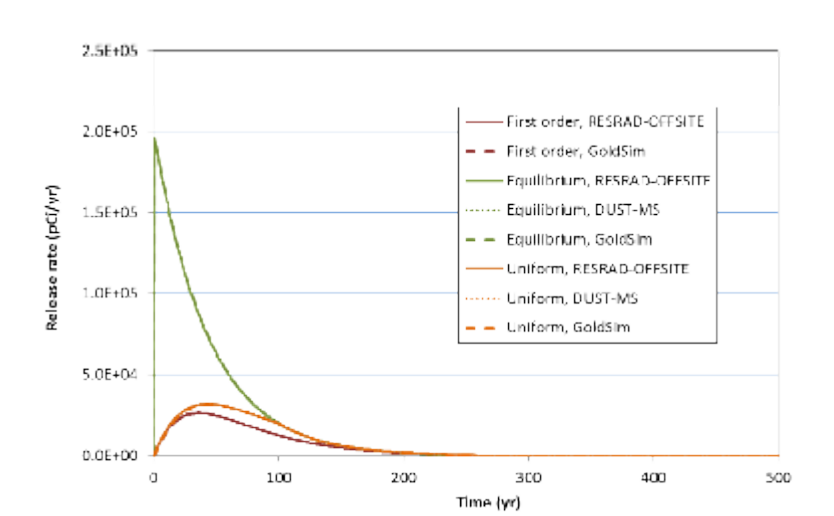


Fig. 6 Comparison of RESRAD-OFFSITE, DUST-MS, and GoldSim Results for Case II Concerning Release from a 0.3-m Cs-137 Source with No Dispersion

Results for Case III

Case III concerns a radioactive waste material containing U-234. The comparison involves not only the release rate of U-234, but also the release rates of Th-230, Ra-226, Pb-210, and Po-210. The progeny nuclides would be formed not only within the waste form, but also in the soil column during the transport of U-234. They may assume different K_d values from U-234 and thereby transport with different speeds toward the bottom of the contaminated zone. The K_d values assumed for U-234 and its progeny nuclides were 200, 6,000, 70, 100, and 10 cm^3/g , respectively.

When progeny nuclides are formed within the waste form, they are assumed to be released through the same mechanism as is their parent nuclide to the surrounding soils. To allow for more ingrowth of progeny nuclides within the waste form, the water infiltration rate was reduced from 0.4 m/yr as used in Cases I and II to 0.1 m/yr. Furthermore, when calculating radionuclide release rates under the uniform release condition, a uniform release of 0.00111/yr for 900 years was assumed. The DUST-MS results were obtained with a design of 481 grids.

Figure 7 compares the release rates of U-234 under the no-dispersion condition. Numerical dispersion is considered to contribute to the discrepancies between RESRAD-OFFSITE results and DUST-MS and GoldSim results (for the equilibrium desorption release).

Figure 8 compares the release rates of Th-230 under the no-dispersion condition. The results of the three computer codes are almost identical. The release rates increase starting at time 0 as Th-230 nuclides are formed because of the decay of U-234. The Th-230 nuclides formed would largely adsorb to soil particles and would stay in the soil column longer than would U-234 nuclides, because the K_d value assumed for Th-230 was much greater than that used for U-234. The adsorbed Th-230 nuclides feed into the continuous release of Th-230 nuclides long after all U-234 nuclides have left the source.

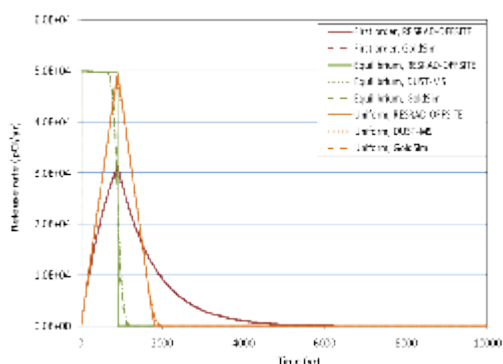


Fig. 7 Comparison of RESRAD-OFFSITE, DUST-MS, and GoldSim Results for Case III Concerning Release of U-234 from a 0.3-m U-234 Source with No Dispersion

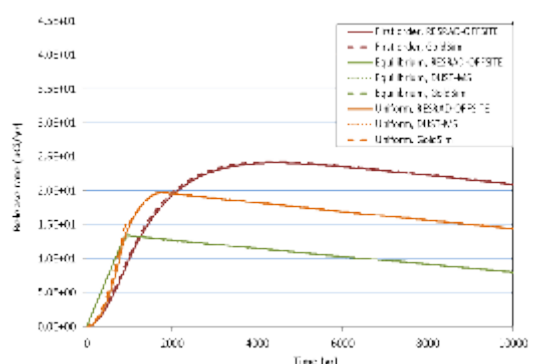


Fig. 8 Comparison of RESRAD-OFFSITE, DUST-MS, and GoldSim Results for Case III Concerning Release of Th-230 from a 0.3-m U-234 Source with No Dispersion

Ra-226 nuclides are formed as Th-230 nuclides undergo radioactive decay. Figure 9 compares the release rates of Ra-226 calculated by RESRAD-OFFSITE, DUST-MS, and GoldSim. The calculations show that the release rates of Ra-226 would increase from the beginning and reach maximum levels earlier than the release rates of Th-230 reach theirs and then decline. The release rates of Ra-226 reach maximum levels earlier than the release rates of Th-230, because the soil K_d assumed for Ra-226 is much smaller than that assumed for Th-230, resulting in that Ra-226 nuclides transport faster than Th-230 nuclides in the soil column. After reaching the maximum levels, the release rates of Ra-226 would have decreased quickly, as do the release rates of U-234, had there not been Th-230 nuclides adsorbing to the soil particles and gradually generating Ra-226 by undergoing radioactive decay.

The decay of Ra-226 generates Pb-210. Because the K_d value assumed for Pb-210, $100 \text{ cm}^3/\text{g}$, is close to the K_d value of $70 \text{ cm}^3/\text{g}$ assumed for Ra-226, the release rate profile of Pb-210 would be similar to the release rate profile of Ra-226, as shown in Figure 10.

The last radioactive radionuclide in the decay chain of U-234 is Po-210, which has a short half-life of 0.38 year. Because of the short half-life, the total radioactivity level of Po-210 at any depth of the soil column is expected to be about the same as that of Pb-210. On the other hand, because Po-210 has a K_d value ($10 \text{ cm}^3/\text{g}$) 10 times smaller than that of Pb-210, the dissolution of Po-210 in the pore water is expected to be much higher than that of Pb-210, and so is the release rate of Po-210 versus the release rate of Pb-210. Comparing the release rates of Po-210 in Figure 11 to those of Pb-210 in Figure 10 confirms this expectation.

Overall, under the no-dispersion condition, the radionuclide release rates calculated by RESRAD-OFFSITE agree very well with those calculated by GoldSim. The same can be said comparing the RESRAD-OFFSITE results with the DUST-MS results, except for the release rates of Po-210. For Po-210, the profiles of release rates calculated by RESRAD-OFFSITE and DUST-MS are the same; however, the magnitudes vary slightly. The exact reason for this discrepancy is not known; perhaps it is related to the short half-life of Po-210, which could result in more numerical dispersion in the calculation results when using the same time step and grid design as for other radionuclides in the DUST-MS calculations.

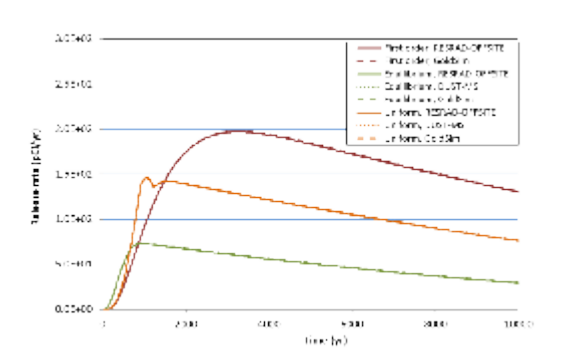


Fig. 9 Comparison of RESRAD-OFFSITE, DUST-MS, and GoldSim Results for Case III Concerning Release of Ra-226 from a 0.3-m U-234 Source with No Dispersion

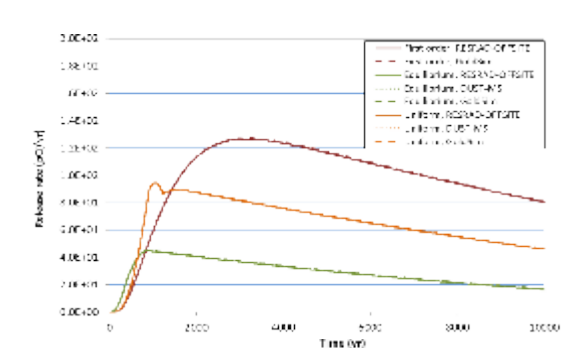


Fig. 10 Comparison of RESRAD-OFFSITE, DUST-MS, and GoldSim Results for Case III Concerning Release of Pb-210 from a 0.3-m U-234 Source with No Dispersion

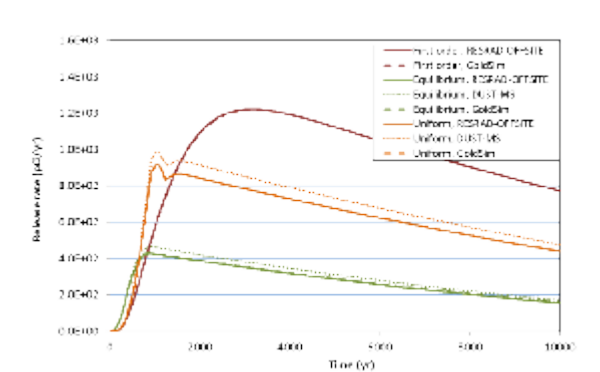


Fig. 11 Comparison of RESRAD-OFFSITE, DUST-MS, and GoldSim Results for Case III Concerning Release of Po-210 from a 0.3-m U-234 Source with No Dispersion

Figures 12–16 compare the results of RESRAD-OFFSITE, DUST-MS, and GoldSim for U-234, Th-230, Ra-226, Pb-210, and Po-210, respectively, considering a dispersivity of 0.03 m for all radionuclides in the soil column. Greater discrepancy than that associated with the results for the no-dispersion condition is observed starting with the first progeny, Th-230, and continuing for the rest of the decay chain. However, the release rate profiles are similar, and the agreement is considered acceptable, given the fact that the simulations were carried out for an extended time period of 10,000 years.

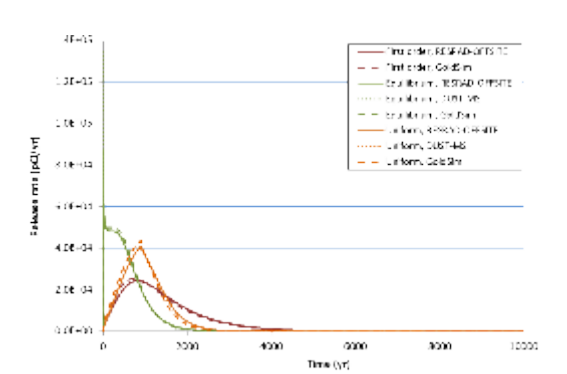


Fig. 12 Comparison of RESRAD-OFFSITE, DUST-MS, and GoldSim Results for Case III Concerning Release of U-234 from a 0.3-m U-234 Source with a Dispersivity of 0.03 m

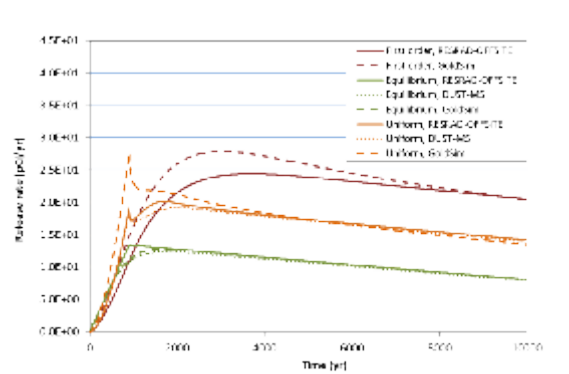


Fig. 13 Comparison of RESRAD-OFFSITE, DUST-MS, and GoldSim Results for Case III Concerning Release of Th-230 from a 0.3-m U-234 Source with a Dispersivity of 0.03 m

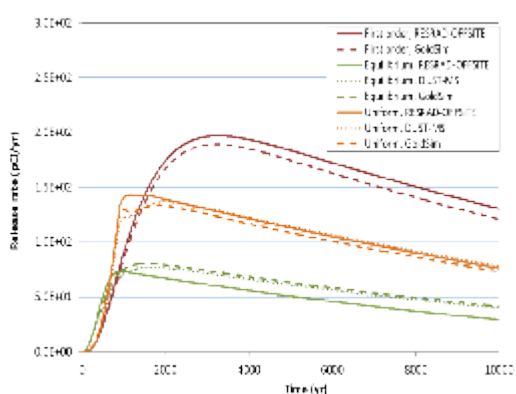


Fig. 14 Comparison of RESRAD-OFFSITE, DUST-MS, and GoldSim Results for Case III Concerning Release of Ra-226 from a 0.3-m U-234 Source with a Dispersivity of 0.03 m

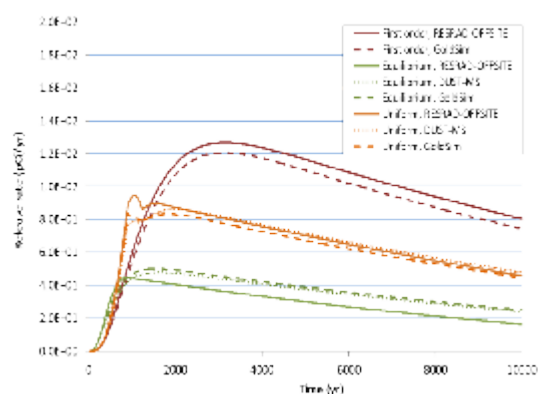


Fig. 15 Comparison of RESRAD-OFFSITE, DUST-MS, and GoldSim Results for Case III Concerning Release of Pb-210 from a 0.3-m U-234 Source with a Dispersivity of 0.03 m

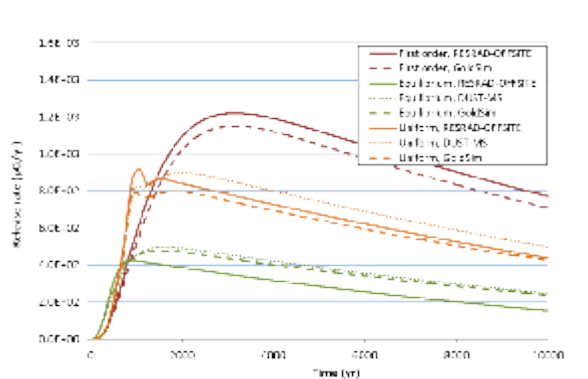


Fig. 16 Comparison of RESRAD-OFFSITE, DUST-MS, and GoldSim Results for Case III Concerning Release of Po-210 from a 0.3-m U-234 Source with a Dispersivity of 0.03 m

Results for Case IV

Cases I, II, and III consider a release source with a thickness of 0.3 m. In Case IV, a release source with 10 times that thickness was considered. Because of the greater thickness, the transport of radionuclides within the soil column would take a longer time to reach the bottom of the contaminated zone. In addition, a smaller water infiltration rate of 0.1 m/yr, less than the 0.4 m/yr assumed for Cases I and II, was used. The smaller water infiltration rate would carry fewer radionuclides leaving the contaminated zone within the same period of time. As a result, the release of radionuclides from the bottom of the contaminated zone would last for a much longer time in Case IV than in previous cases. The release source for Case IV was assumed to contain Tc-99.

Good agreement was observed between the results of RESRAD-OFFSITE and DUST-MS, as seen in Case I. Although the GoldSim simulation was not carried out for this case, judging by the results of RESRAD-OFFSITE and DUST-MS, it is expected that the GoldSim results would also agree well with those of RESRAD-OFFSITE. Because the figures showing comparison of the results have profiles similar to those of Figures 3 and 4 for Case I, they are not displayed here. On the basis of the comparison, the small discrepancy between the RESRAD-OFFSITE and DUST-MS results is considered also to be attributable to the numerical dispersion in DUST-MS results.

Results for Case V

Case V concerns a 3-m release source containing U-234. The release source was assumed to disintegrate with a rate of 0.001/yr for 1,000 years under the uniform release condition. A water infiltration rate of 0.1 m/yr was assumed. The Kd values assumed for U-234, Th-230, Ra-226, Pb-210, and Po-210 in soils were 50, 6,000, 70, 100, and 10 cm³/g, respectively, which were the same as those assumed in Case III.

As for Case IV, GoldSim simulation was not carried out for this case. The release rates of radionuclides calculated by RESRAD-OFFSITE and DUST-MS have the same profiles as those presented in Figures 7–16 for different radionuclides; therefore, additional figures displaying the comparison are not presented in this paper. The same observations made using Figures 7–16 for Case III can be made with the comparison of the calculation results for this case. Under the no-dispersion condition, the agreement for U-234

progeny release rates, except for Po-210, between RESRAD-OFFSITE and DUST-MS is very good. Some discrepancy is observed with the U-234 and Po-210 release rates. The former is thought to be attributable to numerical dispersion in the DUST-MS results. The actual reason for the discrepancy with Po-210 release rates is not yet clear. It could be related to the short decay half-life of Po-210, as well as the assumed small K_d value that allows Po-210 nuclides to transport faster in the soil column as compared to other radionuclides in the same decay chain. With a dispersivity of 0.3 m, the agreement with U-234 release rates is better than it is without dispersion. However, a greater discrepancy is observed with the release rates of progenies (Th-230, Ra-226, Pb-210, and Po-210) when dispersion (with an assumed value of 0.3 m) takes place. The discrepancy eventually decreases or stabilizes as time increases as seen in Figures 13–16. Overall, the radionuclide release rates calculated by RESRAD-OFFSITE and DUST-MS have similar profiles, and the agreement is considered acceptable.

CONCLUSIONS

Comparisons of radionuclide release rates calculated by RESRAD-OFFSITE, DUST-MS, and GoldSim were conducted for five different cases involving different source dimensions, radionuclides, water infiltration rates, and release mechanisms. Overall, the agreement among the results calculated by these three codes is fairly good, with all three codes predicting the same or very similar profiles over time.

The cause of disagreement in release rates of parent nuclides calculated by RESRAD-OFFSITE and DUST-MS under the no-dispersion condition was studied with a 0.3-m Tc-99 source (Case I). The study showed that numerical dispersion in the DUST-MS results was responsible for the disagreement. The disagreement can be reduced by increasing the grid number when subdividing the domain of analysis for the DUST-MS calculations. Because some of the GoldSim simulations were designed with the use of an Aquifer element that implements a numerical analysis method in the calculations, the above conclusion drawn from DUST-MS results would be also applicable to the GoldSim results obtained with the use of an Aquifer element.

When the release source contains U-234 (Cases III and V), which decays to multiple progenies, the comparison shows that agreement among the RESRAD-OFFSITE, DUST-MS, and GoldSim results is better for the longer-lived progenies (Th-230, Ra-226, and Pb-210) than for the shorter-lived progeny (Po-210) under the no-dispersion condition. Although the exact cause of the discrepancy in agreement is not clear, it is thought to be related to the combination of a short half-life and greater mobility in the soil column. Even with some discrepancy, the agreement with the Pb-210 release rates calculated by the three codes is still considered to be good or acceptable.

The comparisons demonstrate that the radionuclide release rates calculated by the RESRAD-OFFSITE source term model are comparable to those calculated by DUST-MS and GoldSim under the release conditions that it is equipped to consider. This new source term model can be utilized to simulate the release of radionuclides from different waste forms and hence would expand the applications of RESRAD-OFFSITE to different contamination situations, including but not limited to performance evaluations of waste disposal facilities.

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