*White Paper*

# Applicability of the RESRAD Codes and the PRG Calculator for CERCLA Sites

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This white paper compares the modeling and assessment tools for evaluation of soils contaminated with radionuclides developed by the U.S. Department of Energy (DOE), i.e., the RESRAD-ONSITE and RESRAD-OFFSITE codes (Yu et al., 2001, Kamboj, et al., 2018, Gnanapragasam et al., 2020, Yu et al., 2020), and the U.S. Environmental Protection Agency (EPA), i.e., the Preliminary Remediation Goals (PRG) Calculator (EPA 2022). Use of these modeling tools at DOE sites are discussed in this White Paper with recommendations on when to use these tools.

RESRAD-ONSITE, RESRAD-OFFSITE and PRG Calculator are designed to derive soil cleanup criteria [e.g., EPA's PRG, DOE's soil guidelines or Authorized Limits (ALs), and Nuclear Regulatory Commission's (NRC's) Derived Concentration Guideline Levels (DCGLs)] based on the potential radiological dose and/or risk from exposure to contaminated soils. A comparison of RESRAD-ONSITE and the PRG Calculator was done in 2015 (Yu et al., 2015). Since then, the PRG Calculator has been modified to fix some issues identified in the report. The current version of the PRG Calculator is used to compare with the current version of RESRAD-ONSITE and RESRAD-OFFSITE codes. Section 1 of this white paper presents the approach and main features of RESRAD-ONSITE and RESRAD-OFFSITE. Section 2 presents the approach used by the PRG Calculator. Section 3 identifies the major issues/limitations associated with the approach of the PRG Calculator and discusses how these issues/limitations are handled in RESRAD-ONSITE and in RESRAD-OFFSITE. Based on the comparison of approach and discussion on issues and limitations, Section 4 proposes a sequence of application of these tools to sites under EPA's Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) containing radiological contamination in soils.

## 1. Features of RESRAD-ONSITE and RESRAD-OFFSITE Codes

The RESRAD-ONSITE code computes the radiological dose and the excess cancer risk to a receptor (person) who engages in various activities directly above the footprint of a contamination in soil. The code models nine exposure pathways: (1) direct external exposure from the initial contamination, (2) inhalation of dust particles (and vapor for tritium [H-3] and gaseous carbon-14 [C-14]) that become airborne from the surface of the soil, (3) inhalation of radon that diffuses out of the contamination and through any intervening layers of cover and/or any building foundation into the space (indoor and/or outdoor) occupied by the receptor, and inhalation of radon that outgasses from the water used in the home, (4) ingestion of plant food that is grown directly above the initial contamination, (5) ingestion of meat, (6) ingestion of milk from livestock raised directly above the initial contamination, (7) ingestion of water from a well or surface water body that is adjacent to the initial contamination, (8) ingestion of aquatic food from the surface water body that is adjacent to the initial contamination, and (9) incidental ingestion of soil. The RESRAD-OFFSITE code computes the radiological dose and the excess cancer risk to a receptor who engages in various activities in the vicinity of, but not necessarily directly on top of, the initial contamination. The well and the surface water body can each be located away from the initial contamination. Exposure by the same nine pathways described for RESRAD-ONSITE are modeled, accounting for the potential offsite locations of exposure. The code has a delayed and time-distributed release option and allows three different conceptualizations of the initial contamination to model the effects of engineering barriers. It also models more release, transport, and accumulation processes than RESRAD-ONSITE.

Both RESRAD-ONSITE and RESRAD-OFFSITE codes model the release of radionuclides to infiltrating water and its subsequent transport to the well and the surface water body; the transfer of radionuclides to the air above the contamination; the transport to offsite locations by air, if appropriate; the accumulation of radionuclides at the off-site locations of secondary contamination; the incorporation of radionuclides from dust, irrigation water, the initial contamination, and, if appropriate, the secondary contamination into plants via root uptake and foliar interception, and the intake of radionuclides by livestock from ingestion of water, feed, and soil. Radiological decay and ingrowth are modeled not only at the initial source contamination, but also during transport in groundwater, during accumulations at the off-site locations of secondary contamination, and during storage after harvest or extraction of the food and water. Mass balance of a radionuclide is maintained over the analysis time horizon between the radionuclides that remain in-situ and the radionuclides that are released to infiltration and to runoff while accounting for ingrowth and decay. Mass balance ensures that the sum of the quantity of a radionuclide that remains in situ at any time, the quantity of that radionuclide that is released up to that time, and the net quantity of that radionuclide that is lost due to radiological decay less the quantity produced due to radiological ingrowth is equal to its initial inventory.

The direct exposure model used by the two RESRAD codes considers the size (area and thickness) of the contamination, any soil cover on top of the contamination, the locations of the receptor in relation to the contamination and the occupancies of the receptor at those locations. It accounts for the contribution of each of the progeny, considering the dose or slope factors and the radiation energies emitted from each progeny radionuclide (except spontaneous fission) when modeling the effects of shape, area, and thickness of the contamination, and the location of the receptor relative to the contamination.

The inhalation model for onsite exposure considers the size of the contamination using an area factor and a cover-and-depth factor to account for the fraction of the particulates in air that are derived from the contamination and of the occupancy. The inhalation model for offsite exposure considers both inhalation of the respirable particles transported by air to the offsite locations and those resuspended from accumulation at the offsite locations. The latter involves all releases from the primary contamination, various forms of transport to the offsite locations, and accumulation at the exposure locations.

The radon model considers the diffusion of radon out of the contamination through any soil cover; ingrowth and decay of its progeny during transport via air to the receptor location; entry of radon and its progeny into the residence by air exchange from outdoors; the entry of radon by diffusion through the floor; the introduction of radon outgassing from the water used in the residence; the ingrowth and decay of its progeny within the residence; and the occupancies at the various exposure locations. There are also special models in the RESRAD codes for special radionuclides such as H-3 and C-14 in gaseous forms (HTO or $^{14}CO_2$). This allows users to use the same code to evaluate all potential radionuclides for any sites without the need to use different models/codes for different radionuclides.

The vegetable, meat, and milk ingestion exposure models consider root uptake at onsite and offsite locations as appropriate and foliar interception of particulates and potentially contaminated irrigation water followed by translocation. The meat and milk ingestion models include the ingestion of water, feed, and soil with the feed by the livestock. This again involves all releases from the primary contamination, various forms of transport to the offsite locations, and accumulation at the exposure locations where appropriate, and the transfer to and accumulation in the edible parts of the plant and the livestock.

The water ingestion exposure considers use of water from different water sources. The modeling of contamination of water in the water sources involves all releases from the primary contamination, various forms of transport to the offsite locations, accumulation in the surface water body, and dilution in the well as appropriate.

Almost all the parameters used by the two RESRAD codes to compute the dose and risk are transparent to the user in easily accessible forms. Users are encouraged to input site-appropriate values to replace the defaults in the code. The user can model various scenarios by activating/deactivating the exposure pathways and by specifying the appropriate inputs. The input files that are created in the code can be saved for later uses. The files can be modified and saved under different names as the scenario is refined and made more site-specific. The RESRAD codes can read the input files created in the previous version to allow reanalysis of a site, should that become necessary.

Both RESRAD-ONSITE and RESRAD-OFFSITE codes have the functionality to perform sensitivity analysis, either one input at a time or simultaneously over multiple inputs (i.e., probabilistic uncertainty analysis). Scatter plots and regression analysis are available in the probabilistic uncertainty analysis to identify the inputs that have a significant effect on the dose. These tools allow the analyst to identify and prioritize the inputs that cause a significant change in the dose from among the many inputs in a typical multi-pathway scenario, when spending the limited resources to refine site-specific values.

The codes offer a choice of standard dose coefficients file (DCF) libraries (DCFPAK3.02, International Commission on Radiological Protection [ICRP] 72/ICRP 60, ICRP 30, Federal Guidance Report [FGR] 11, FGR 12, and FGR 13 slope factors) and the DOE Standard (DOE-STD-1196-2021, *Derived Technical Concentration Standard)* per capita dose coefficients that is population and gender averaged (DOE 2021). The RESRAD dose conversion factor (DCF) Editor can be used to create and share DCF libraries. The user created libraries reside in the database files and can be used by any RESRAD-ONSITE or RESRAD-OFFSITE input file on that computer. The user created libraries can be exported and shared with other users who can then import them into their database file. This facilitates the use of user created libraries by colleagues and reviewers.

Both RESRAD-ONSITE and RESRAD-OFFSITE compute the time-integrated dose over a year and the time-integrated risk over the specified exposure duration. These are computed at specified number of time points over the time horizon. Temporal plots of the time-integrated risk and dose are available to provide many different views: by initially present radionuclide, by initially present radionuclide and progeny, summed over all radionuclides, from individual pathways, or summed over pathways. Temporal plots of concentrations in environmental media and in food and water are also available. The underlying data can be viewed and exported for further analysis, if necessary.

The computed doses, risks, and concentrations vary with time because the RESRAD codes model some or all of the following processes as appropriate: time varying releases, reactive advective dispersive transport in groundwater, atmospheric transport, and accumulation at the offsite locations.

The exposure from different initially present radionuclides typically peaks at different times not only because of the differences in transformation rates and transformation products, but also because of differences in release, transport, and accumulation characteristics and differences in dominant exposure pathways. The time integrated dose and the time integrated risk from the same initially present radionuclide can peak at different times because the exposure durations for the two are different and because the relative contributions of the parent and progeny nuclides to dose and to risk can be different, i.e., because the dose coefficients and risk factors are not strictly proportional. The temporal graphics viewers in the two RESRAD codes provide tools to easily determine the peak dose and peak risk and the time of peak dose and risk.

While both RESRAD codes were developed to determine the exposure from an initial contamination in soil, RESRAD-OFFSITE can be flagged to read and use the temporally varying concentration data in the well and the surface water. It can also be flagged to read and use temporally varying releases. These can be used to model scenarios where the temporal concentration of radionuclides in water and/or air are the initial drivers of the scenario.

All RESRAD codes are developed following DOE and Argonne quality assurance (QA) procedures and requirements and conform to the regulatory requirements of DOE Order 414.1D, *Quality Assurance* (DOE 2011) and ASME NQA-1-2008, *Quality Assurance Requirements for Nuclear Facility Applications* (ASME 2008). Many benchmarking and code comparisons have been done using real world data, including those collected from Chernobyl and Fukushima. The results are published in refereed journal articles and Technical Reports from the International Atomic Energy Agency (IAEA), as well as Argonne National Laboratory Technical Memorandum reports (See list of references in Section 5). More than 130 countries have downloaded and used RESRAD codes, and numerous applications of RESRAD codes have been published in the literature (e.g., journal articles, PhD dissertations, and reports). From the QA point of view, in addition to the features and functionality described in the preceding paragraphs, the RESRAD codes are a logical choice for performing site-specific dose and risk analysis at DOE sites.

## 2. Features of the PRG Calculator

The PRG Calculator was developed by EPA to establish preliminary remediation goals (PRGs) at radioactively contaminated CERCLA sites. The derived PRGs, expressed as medium concentrations of an individual radionuclide, are based on a target lifetime excess cancer risk with a default of $1 \times 10^{-6}$ for contaminated soil, air, water, or biota (produce, fish, game, fowl, livestock, and livestock product). For example, there are soil external exposure PRG, soil ingestion PRG, soil inhalation PRG, soil produce consumption PRG, air inhalation PRG, air submersion PRG, tap water immersion PRG, tap water ingestion PRG, tap water inhalation PRG, tap water produce consumption PRG, direct biota consumption PRG, surface water immersion PRG, and surface water ingestion PRG.

A variety of receptor options are provided for selection, including generic resident, indoor worker, outdoor worker, composite worker, construction worker, recreator, and farmer. Based on the selection of receptor, various contaminated media for which total PRG can be derived are made available for selection. The total PRG is derived by combining the applicable medium-exposure PRG that is calculated using scenario-specific exposure parameters. For example, for a resident scenario, the total PRG for soil is derived by combining soil external exposure PRG, soil inhalation PRG, soil ingestion PRG, and soil produce consumption PRG. For a farmer scenario, the total PRG for combined soil and biota is derived by combining soil external exposure PRG, soil inhalation PRG, soil ingestion PRG, and soil produce consumption PRG, as well as soil biota consumption PRG.

The total soil PRG derived for various receptors evaluate potential radiation exposure only from the water-independent pathways. They consider the amount of radioactivity in soil to change over time due only to radiological ingrowth and decay.

The PRG Calculator provides an option for evaluation of “Soil to Groundwater.” This option is listed with various receptor options and is the only option in the PRG Calculator that relates water contamination to soil contamination. When this option is selected, Soil Screening Levels (SSLs) are derived based on the “Resident” exposures. SSLs are similar to the total soil PRGs for residents. However, rather than considering radionuclides would remain attached to soil particles as in the derivation of soil PRGs, the derivation of SSLs considers radionuclides in contaminated soil would dissolve to the interstitial water, leach out, and discharge to the groundwater aquifer, resulting in radiation exposures from the use of contaminated groundwater. In short, the SSLs are derived considering radiation exposures through the water-dependent pathways. To derive the SSL for each radionuclide, the maximum concentration level (MCL) that EPA set for drinking water, if available, and the total water PRG for residents are related linearly to soil concentration, i.e., with a constant factor that does not change with time. This constant factor considers dilution in the groundwater aquifer and equilibrium partitioning of the nuclide between the solid phase and the liquid phase in soil. The time needed for radionuclides to transport through the soil column beneath the contaminated zone is not considered, nor is the decay and ingrowth that would occur during the transport. Therefore, for each radionuclide, the change of concentration in the groundwater is proportional to the change of concentration in the contaminated zone, which is due only to radiological ingrowth and decay.

For the resident scenario, both the SSLs and the soil PRGs can be derived for the same contamination in soil, but the PRG Calculator does not provide guidance on how to develop the final remediation goals combining the SSLs and the soil PRGs. From a realistic perspective, it is noteworthy that the radiation exposures associated with the water-independent pathways as considered in the derivation of the soil PRGs could occur at different times from the radiation exposures associated with the water-dependent pathways as considered in the derivation of the SSLs.

The PRG Calculator establishes radiological decay chains with the transformation data from ICRP 107 (ICRP 2008). To derive PRGs (or SSLs), the calculator first evaluates the lifetime cancer risk rate for each radionuclide in the decay chain, assuming their concentrations in the contaminated medium remain constant and are the same as the initial concentration of the parent radionuclide. These cancer risk rates of individual radionuclides in the decay chain are used to derive the PRGs or SSLs reported in the result tables based on the selected decay and output option.

Four decay and output options are provided by the PRG Calculator: (1) assumes the period of peak risk, (2) assumes secular equilibrium throughout the chain all the time, (3) does not assume secular equilibrium all the time and provides results for progeny throughout the chain, and (4) does not assume secular equilibrium all the time but provides results only for selected radionuclides. Options (3) and (4) produce medium-specific PRGs and the total PRG for the parent radionuclide without considering the cancer risks that could result from exposures to the progenies. Hence, the potential risks could be underestimated. Option (2) produces PRGs for the parent radionuclide with consideration of cancer risk from exposure to progenies, but overestimates progenies' contributions. Option (1) provides the most conservative approach to handling radiological ingrowth and decay but fails to consider that the peak risk may occur at different times for different radionuclides in the decay chain when they transport from the source to the receptor location. The PRG Calculator uses a simplistic (and unrealistic) method to adjust cancer risk rates of each radionuclide in the decay chain at the receptor location by using the relative activity concentrations as function of time (calculated with the Bateman equation) at the source location. This approach may result in erroneous peak risk when the progeny radionuclides do not travel with the parent radionuclides in the environment [i.e., radionuclides with different soil-to-water distribution coefficients ($K_d$)].

## 3. Issues of the PRG Calculator

In general, the intention of the PRG Calculator is to analyze radiation exposure and the associated health risk for several common human receptors considered in risk assessment by employing conservative conceptual models and assumptions without consideration of mass balance. For example, there is no consideration of leaching loss of radionuclides from contaminated soil when deriving soil PRGs, and no consideration of the lag in time for radionuclides transporting through deeper soils and reaching the groundwater table when deriving SSLs. However, this conservativeness in the modeling approach, when coupled with the conservative default values selected for the input parameters, could lead to the derivation of PRGs or SSLs that are impractically low and beyond achievement with existing remediation technologies. Although the default values of the input parameters can be replaced with site-specific values, the practicality of the derived PRGs and SSLs could still be impeded by the conservative nature of the conceptual models and assumptions. For some of the input parameters, site-specific values may not be readily available (or measurable) and they need to be evaluated using a more sophisticated model. One example is the soil leach rate, which is used to model the accumulation of radionuclides in soil through irrigation. In the PRG Calculator, a single input of soil leach rate is applied to all the radionuclides in the analysis. Using the same leach rate for all radionuclides in the PRG Calculator to calculate water biota consumption PRGs is a fundamentally incorrect approach because radionuclides have different leach rates. Even the same radionuclide at different sites may have different leach rates due to different site conditions and radionuclide chemical forms.

The PRG Calculator treats soil and water as independently contaminated medium when deriving soil or water PRGs. Therefore, for the resident and the farmer scenario, biota contamination is attributed to either soil or water contamination, but not both. In the derivation of SSLs, the transfer of radionuclides from soil to groundwater is considered but the quantity of transfer is not calculated and tracked over time. Hence, mass balance is not maintained when it combines the total soil PRGs and the total water PRGs for the resident scenario to derive the SSLs.

RESRAD-ONSITE and RESRAD-OFFSITE were designed to evaluate potential radiological doses and human health risks associated with radioactively contaminated soils and to derive soil cleanup criteria or remediation goals based on a radiological dose constraint or a target human health risk level. The RESRAD-ONSITE and RESRAD-OFFSITE results can be used to demonstrate compliance with both dose-based or risk-based regulations and requirements.

When evaluating soil contamination, unlike the PRG Calculator, RESRAD-ONSITE and RESRAD-OFFSITE codes consider soil as the initial source of contamination. They employ fate and transport models to project the spreading of radionuclides from soil to surrounding media (air, water, biota, and deeper soils) as time progresses. The fate and transport models used in RESRAD codes track the concentrations of radionuclides in different media over time by considering radionuclide-specific transfer rate and transport speed as well as the radiological decay and ingrowth to maintain mass balance. The concentrations in different environmental media are used to estimate the potential radiation exposures and the associated radiological doses and cancer risks from different exposure pathways. The total dose and risk from all exposure pathways at different times for the receptor of concern are also calculated. The maximum total radiation dose (or cancer risk) over the specified time frame (e.g., 1,000 years) is then used to derive soil cleanup criteria (or authorized limits or remediation goals) for the initially present radionuclide. The soil cleanup criteria derived by RESRAD-ONSITE and RESRAD-OFFSITE codes factor into account not only radiation exposures associated with the water-independent pathways and the water-dependent pathways, but also the respective occurrence in time of the corresponding dose or risks and the time of the peak total dose or risk.

The PRG Calculator does not handle radionuclides in gaseous form such as radon, tritium (HTO), and C-14 ($^{14}CO_2$). All radionuclides are treated as particulates. Users will need to use another calculator for the evaluation of radon, and currently there is no other calculator for tritium and C-14. All PRGs are derived based on risk, not dose. No dose coefficients (including the DOE Standard per capita dose coefficients) are used in the PRG Calculator.

## 4. Suggested Applications for Soil Contamination

As indicated by its name, “Preliminary” Remediation Goal, the best use of the PRG Calculator is to screen radioactively contaminated CERCLA sites to eliminate the need for further site-specific analysis for some radionuclides. Because of the limitations discussed in previous sections (i.e., lack of mass balance, hard to combine water-dependent SSL and water-independent PRG, cumbersome treatment of decay and ingrowth, etc.), it is recommended that RESRAD codes are used to double check the screening PRGs for DOE sites. For radionuclide concentrations exceeding the screening PRGs, RESRAD codes are recommended for site-specific analysis and derivation of site-specific cleanup criteria (or authorized limits).

RESRAD-ONSITE and RESRAD-OFFSITE employ a more comprehensive approach than the PRG Calculator to address the dependency of radiological contamination between soil and the surrounding environmental media over time. The dynamic fate and transport models implemented in RESRAD-

ONSITE and RESRAD-OFFSITE allow for conceptualization of a contaminated site in a more realistic way, including consideration of radionuclides leaching out from soil, transporting to deeper soils, and transporting in a groundwater aquifer to a well. At the same time, the models are capable of simulating radiological ingrowth and decay and considering different transport speeds of radionuclides in a decay chain to ensure mass balance. The RESRAD-ONSITE and RESRAD-OFFSITE codes have been applied to derive soil cleanup criteria at numerous sites and successful remediation and site closure have been achieved. For radioactively contaminated CERCLA sites that do not pass the generic screening evaluation with the PRG Calculator, RESRAD-ONSITE and RESRAD-OFFSITE are ideal tools to use for site-specific analyses, so more realistic but still conservative remediation goals can be derived for the contaminated soils.

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