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CLEANUP STANDARDS AND PATHWAYS ANALYSIS METHODS*

by

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ABSTRACT

This paper discusses cleanup standards for radioactively contaminated sites and describes the use of pathways analysis methods for deriving site-specific residual radioactivity guidelines. An example is provided in which a pathways analysis code (RESRAD) was used to establish such guidelines.

INTRODUCTION

Remediation of a radioactively contaminated site requires that certain regulatory criteria be met before the site can be released for unrestricted future use. Since the ultimate objective of remediation is to protect the public health and safety, residual radioactivity levels remaining at a site after cleanup must be below certain preset limits or meet acceptable dose or risk criteria.

Release of a decontaminated site requires proof that the radiological data obtained from the site meet the regulatory criteria for such a release. Typically release criteria consist of a composite of acceptance limits that depend on the radionuclides, the media in which they are present, and federal and local regulations. In recent years, the U.S. Department of Energy (DOE) has developed a pathways analysis model to determine site-specific soil activity concentration guidelines for radionuclides that do not have established generic acceptance limits. The DOE pathways analysis computer code (developed by Argonne National Laboratory for the DOE) is called RESRAD (Gilbert et al. 1989). Similar efforts have been initiated by the U.S. Nuclear Regulatory Commission (NRC) to develop and use dose-related criteria based on generic pathways analyses rather than simplistic numerical limits on residual radioactivity.

The focus of this paper is radionuclide contaminated soil. Cleanup standards are reviewed, pathways analysis methods are described, and an example is presented in which RESRAD was used to derive cleanup guidelines.

RELEASE CRITERIA

The primary objective of establishing release criteria is protection of the health and safety of the public and occupational workers. Residual contaminant levels at remediated sites should, therefore, be below certain limits so that a future occupant may use the site without restrictions. However, specifying the types of acceptance limits and establishing values depends on numerous scientific, regulatory, and sociopolitical factors.

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The U.S. Environmental Protection Agency (EPA) applies several criteria to cleanup actions such as maximum contaminants levels (MCLs), maximum permissible concentration (MPC), national ambient air quality standards (NAAQS), and ambient water quality criteria (WQC). The calculated human intake results can be compared to acceptable intake for subchronic exposure (AIS), acceptable intake for chronic exposure (AIC), reference level (RL), reference dose (RfD), and risk in terms of carcinogenicity. The EPA levels are defined for many chemical contaminants as well as for some radionuclides.

According to Title 40, Part 141 of the *Code of Federal Regulations* (40 CFR 141), in drinking water, the proposed MCLs for radionuclides are 20 pCi/L for Ra-226, 20 pCi/L for Ra-228, 20 μg/L for Uranium, 300 pCi/L for Rn-222, 15 pCi/L for gross α emitters (not including Rn, U or Ra-228), and 4 mrem/y (based on dose committed over 50 years assuming 2 L/day drinking water intake) for β and photon emitters. While the MCLs are enforceable standards, under the Safe Drinking Water Act, the EPA is required to publish the maximum contaminant level goals (MCLGs). The MCLG for radionuclides is zero.

Directly applicable numerical limits have been used in certain cases. For example, some cleanup actions completed under NRC jurisdiction have been based on limits specified in NRC Regulatory Guide 1.86. The DOE uses cleanup limits for some radioisotopes, such as Ra-226, Ra-228, Th-230, and Th-232 (5 pCi/g averaged over 15 cm of soil below the surface and 15 pCi/g averaged over 15 cm thick layers of soil below the first layer; "as low as reasonably achievable" (ALARA) evaluation is also performed to determine if it is reasonably possible to cleanup to levels below these standards). For other radionuclides, such as U-238, U-234, U-235, and Cs-137, site-specific cleanup guidelines are derived. Site-specific cleanup levels are derived through pathways analysis methods and are based on radiation dose limits. The DOE uses the RESRAD computer code and an individual dose limit of 100 mrem/y (for future land use scenarios) for determining residual radioactive material concentrations that can be potentially left in the ground. However, actual cleanup levels are generally lower than the derived values because DOE applies the ALARA process for radiation protection and cleanup actions. The EPA has employed the PRESTO and PATHRAE (EPA 1987) codes for pathways analysis of radioactive sites.

A major problem in cleanup standards is the lack of consistency in the dose limits recommended by various agencies. For an individual member of the public, the International Commission on Radiological Protection (ICRP) recommends an effective dose limit of 1 mSv/y (100 mrem/y). Implications of the recent ICRP recommendations for risk assessments for radioactive waste disposal and cleanup have been discussed by Devgun (1992). The DOE, through its Order 5400.5 (DOE 1990), implements an effective dose limit of 1mSv/y (100 mrem/y) as the primary standard for members of the public. However, it is the policy of DOE to apply the ALARA process. The DOE also uses a guideline of 20 μR/h above background for gamma exposure rates; values (in dpm per 100 cm^2) are also available for surficial α and β contamination. DOE's positions and views are available in a recently issued Radiological Control Manual (DOE 1992). In applying the provisions of 10 CFR 20 for the possession or use of radioactive materials, the NRC specifies an annual effective dose limit of 4 mSv for unrestricted areas. However, for uranium fuel cycle operations, the NRC also specifies that provisions of 40 CFR 190 apply, which provides an annual effective dose limit of 0.25 mSv (25 mrem). Under 10 CFR 20, the reference annual dose level for a member of the public is 100 mrem (1 mSv); however, a licensee may apply (subject to certain conditions) for prior authorization of operation that may result in exposure to an individual member of the public up to 500 mrem (5 mSv) per year. The EPA uses a limiting criterion of 0.1 mSv/y (10 mrem/y) for emission of radionuclides to ambient air (40 CFR 61).

The NRC licensing requirements for land disposal of radioactive wastes (10 CFR 61) specify an effective whole body dose limit of 0.25 mSv/y (25 mrem/y) and a dose limit of 0.75 mSv/y (75 mrem/y) to the thyroid. The EPA's standards for radioactive waste disposal include a groundwater protection requirement (40 CFR 191) that specifies an effective dose limit of 0.04 mSv/y (4 mrem/y).

Inspection-oriented numerical limits, such as those specified for residual surface contamination in NRC Regulatory Guide 1.86, are the most directly applicable. Such numerical limits facilitate confirmation for the decontamination operation, but they may not necessarily be cost-effective. A draft manual for conducting radiological surveys in support of license termination has recently been issued. The manual was prepared by Oak Ridge Associated Universities (ORAU 1992) for the NRC and describes methodologies for conducting surveys, comparing data with guidelines, and applying statistical techniques.

Dose-based cleanup limits used by the DOE (along with probabilistic risk assessments for health effects) and those currently being investigated by the NRC, inherently involve pathways analysis methodologies and the use of dose conversion factors to derive cleanup criteria. These limits can allow greater flexibility in decontamination while achieving protection goals because they are based on site-specific analyses and considerations. Such limits also lead to application of different cleanup criteria to different sites, which may have regulatory and sociopolitical implications with respect to their public acceptability.

Generally three types of surveys are necessary for releasing a site for unrestricted use after radiological decontamination:

- Surface contamination (dpm/100 cm^2),
- Soil radionuclide concentration (pCi/g), and
- Exposure rate (μR/h).

In addition, for habitable structures at the site, measurements of radon daughter concentrations are also required.

PATHWAYS ANALYSIS METHODS

Pathways analysis methods provide a means of assessing risk from a radiologically contaminated site as well as any residual risk from a decontaminated site. In simple terms, risk assessment in this case is the qualitative or quantitative characterization of potential adverse impacts of specific contaminants on individuals, populations, or the environment. Risk is a function of hazard and exposure. Thus, assessment of risk from a contaminated site involves quantification of potential hazards and an assessment of potential exposure pathways for contaminants to reach a receptor.

In general terms, risk assessment methodology involves four elements:

1. Characterization of the contaminated site and identification of contaminants of concern and their toxicity profiles;

2. Development of a conceptual site model and completion of the exposure pathways analysis;

3. Quantification of exposure assessment results in terms of risk to human health or adverse environmental effects; and

4. Interpretation of results in terms of risk limits, dose limits, or regulatory standards for contaminants of concern in the environmental media.

In its guidance on risk assessment for superfund sites, the EPA (EPA 1989) defines exposure assessment as a three step process: characterization of the exposure setting, identification of the exposure pathways, and quantification of the exposure. The methodology provided includes equations for various pathways as well as the necessary conversion and absorption factors. Baseline risk assessment conducted through the above procedure defines the hazard potential of the site and sets the course of the remediation process. For hazardous waste sites where numerous contaminants may be present, "indicator" contaminants, which present the greatest potential risk to human health, can be identified and used in the risk analysis.

A generic model of potential exposure pathways to an individual may involve several routes. Contaminants can leach into surface water or groundwater or be resuspended in air. The contaminated medium itself may be soil, surface water, or groundwater at the site. Exposure of an individual can occur through a number of pathways including ingestion of contaminated surface water, fish raised in contaminated surface water, contaminated groundwater, plant foods irrigated with contaminated surface water or groundwater, and meat from livestock raised on contaminated plant foods and/or drinking contaminated surface water or groundwater. From contaminants resuspended in air, exposure can occur as the result of inhalation, as well as ingestion of meat from exposed livestock or plants on which the contaminants may have deposited. Direct contact with contaminants can cause dermal exposure. In the case of radioactive contaminants, direct radiation exposure is another important pathway. A generic model of the pathways is shown in Figure 1. In addition to such exposure pathways, several routes of contaminant transport within a medium or between media (e.g., infiltration of surface water into groundwater) are possible.

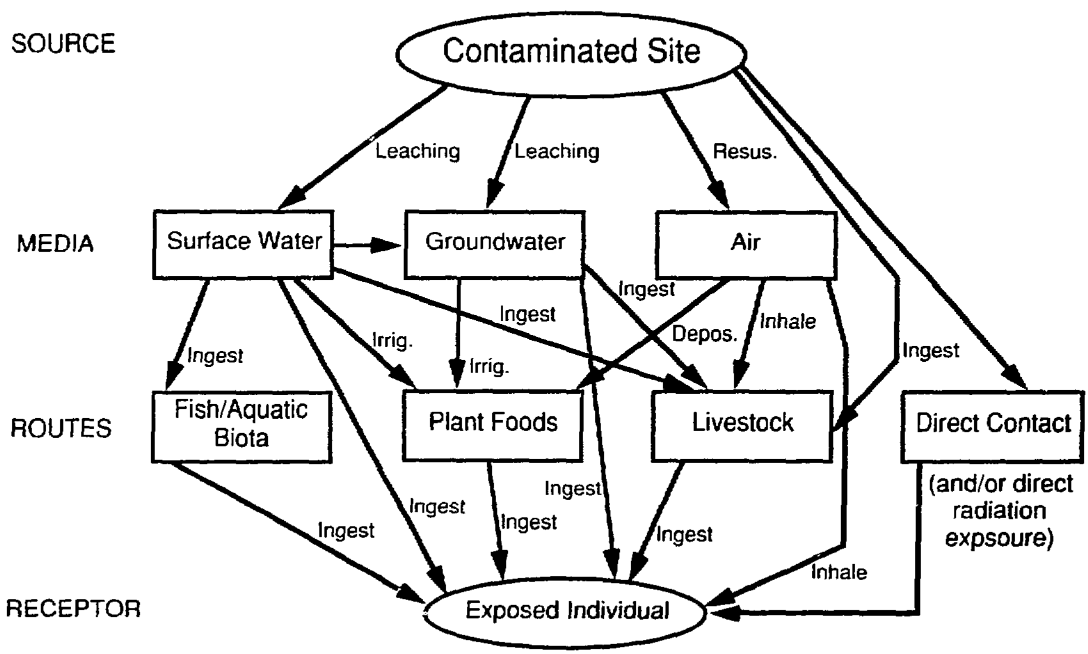

Figure 1. A Generic Model for Exposure Pathways from a Contaminated Site

The extent and sophistication of the exposure pathways analysis depend on the site- and contamination-specific characteristics, along with the potential scenarios that are relevant to the site or the remedial action. For example, the exposure scenario may involve an industrial worker (if the contaminated site is an industrial site), a resident/farmer (if the current or future projected land use at the site is for habitation or farming), a recreational user, an intruder (into the contaminated area), or a remedial action worker. Identification of the receptor or the exposed individual or population is one of the most important elements of risk assessment. For simplified models, risk calculations can be derived for individual pathways by using generic equations; overall risk to an individual can then be ascertained. Pathway-specific calculations require data on a number of variables, such as leach rate or contaminant concentrations in the specific environmental medium, ingestion or intake rates, exposure frequency, and exposure duration. However, often the analysis starts with available site information, and iterative assessments are necessary as further site-specific information becomes available. Also, it is often necessary to project calculations into the future to cover scenarios involving long-term migration of contaminants from the site and subsequent exposure of individuals via various pathways. Thus, computer models are being used increasingly in risk assessment for chemical and radioactive contamination. A number of computer codes are available [e.g., DECHEM (DOE 1989), PRESTO codes (EPA 1987), PATHRAE (EPA 1987), AIRDOS (Moore et al. 1979), SWIFT (Reeves and Cranwell 1981), and RESRAD (Gilbert et al. 1989)].

In deriving cleanup guidelines, a pathways analysis is performed for relevant current and future land use scenarios by using site-specific geologic/hydrogeologic/climatic, as well as, relevant food and water consumption parameters. Generally, the analysis is performed for a number of scenarios, and limits are chosen on the basis of the analysis results, along with other considerations such as ALARA, interagency agreements, precedent-setting limits from prior remedial actions, or other factors. The overriding philosophy in risk assessment is to not underestimate the human risk. Thus, it is common to make conservative assumptions (which will overestimate rather than underestimate the risk), and conservative values of parameters are used if site-specific data are unavailable. From a practical perspective, the cost/benefit issue of setting cleanup limits much lower than they need to be is very significant. Remedial costs may increase proportionally as the cleanup limits (e.g., soil concentration of a radionuclide in pCi/g) are lowered to a certain point, but may increase sharply if cleanup limits are lowered further. This situation arises from limitations of the cleanup and detection technologies and the extensive additional efforts required to achieve small gains in cleanup values. Cleanup of a site below background levels is subject to debate and may not make sense unless risk assessment demonstrates a clear human health risk from such levels. Under such circumstances, it is no longer a case of the contaminated site alone but the total environment at the site, that is, the site and its surroundings.

Dose-limited pathways analysis with generic or site-specific parameters may be used, for example, as in RESRAD. This type of analysis yields the most direct measure of success of the decontamination operation and can be customized to the site by using applicable hydrogeological variables, future use scenarios, and other relevant parameters. The following factors are considered in selecting guidelines and acceptance criteria for the release of decontaminated radioactive sites:

1. External Exposure. Limits on exposure rates from decontaminated sites are specified by agencies such as the NRC or DOE.

2. Affected Media. Maximum release limits may be specified for water, air, and/or soil. For soil contamination, generic limits are available only for certain radionuclides; site-specific activity concentrations must be derived.

3. Federal and Local Regulations. Variations exist in numerical limits established by regulatory authorities. These limits also change from time to time.

4. Local Acceptance. It is important that local public acceptance be gained for applications of cleanup criteria and guidelines to a specific site.

PATHWAYS ANALYSIS USING RESRAD

Pathways analysis methods have been used extensively in DOE remediation projects for radiologically contaminated sites. For example, these methods have been applied through the RESRAD code to conduct radiological risk assessment for contaminated sites (Devgun 1990a,b), as well as to establish risk-based cleanup criteria (Devgun, Hyatt, and Yu 1990, Cheng, Yu, and Devgun 1991).

For establishing cleanup guidelines, which is the focus of this paper, the pathway analysis method provides a means of calculating site-specific guidelines subject to certain use conditions (scenarios). Accurate, site-specific input parameter values should be used to the maximum extent practical. The RESRAD code incorporates considerations of the external radiation, inhalation, and ingestion pathways to exposure from sources in the ground, air, water, and food chain. Generic values for some input parameters, such as food, water, and air consumption are also available in the code. A library of radionuclides is included in the program. In the code version used for analyses described in the above-referenced reports, there were above 80 input parameters in RESRAD.

On the basis of the site- and scenario-specific inputs established, RESRAD can provide the activity concentration in soil for radionuclides identified for the site that would result in an exposure corresponding to the basic dose limit of 100 mrem/y. Here, the basic dose limit is the input, and the resulting RESRAD-calculated concentration is the limit. In this context, RESRAD is used as a preremediation planning tool by which limits on radionuclide concentrations in soil are developed. It should be recognized, however, that the ALARA process is applied in setting the actual cleanup levels. Other factors such as state and local acceptance, cost/benefit of lowering limits further, prior cleanup precedents must also be considered.

The application of the pathways analysis methods (through the RESRAD code) is illustrated here through an example. The case presented is a site located in eastern Tennessee where radionuclide contamination included uranium, thorium and radium. This 7-hectare (17-acre) area is being developed for industrial use. The site became contaminated when predecessors of the DOE, the Manhattan Engineer District, and subsequently the Atomic Energy Commission, stored uranium ore and ore processing residues there. The site was radiologically surveyed, decontaminated, and released for unrestricted use in 1972. However, after a 1987 survey by the state and a subsequent survey by DOE in 1988, the site was added to a DOE remedial action program for further cleanup. An evaluation of the cleanup alternatives was completed in 1991 (DOE 1991). Residual radioactive material guidelines for the site were derived for total uranium and uranium isotopes by using RESRAD and by using site-specific parameters and different scenarios as the input (Cheng, Yu, and Devgun 1991).

The guidelines were derived on the basis of meeting the 100 mrem/y basic dose limit and assuming that U-238, U-234, and U-235 were present in their natural activity ratio of 1:1:0.046. It was also assumed that uranium was the only radionuclide present at an above-background concentration. In all scenarios, it was assumed that at some time within 1,000 years following decontamination, the site will be released for use without radiological restrictions. The detailed assumptions, input data, and results are available in Argonne National Laboratory report (Cheng, Yu, and Devgun 1991). Summary information relevant to this paper is presented here.

Four potential exposure scenarios were considered: industrial use, recreational use, residential farm use with an adjacent and downgradient pond providing the water supply, and residential farm use with groundwater drawn from a well as the useable water. The industrial scenario assumed use of the site by a hypothetical worker who spends 2,000 hours annually (75% outdoors and 25% indoors) at the decontaminated site and does not ingest any water or food from the site. In the recreation use scenario it was assumed that the person spends 750 hours annually, all outdoors, and does not ingest any water or foods from the site. The residential farm scenario assumed that a hypothetical occupant sets up residence in the immediate vicinity of the site, uses pond or well water as the water supply source and eats plant foods grown there, and consumes meat and milk from livestock raised at the decontaminated site. Fish consumption from the pond was also included. The resident farmer was assumed to spend 50% of his or her time indoors, 25% outdoors in the decontaminated area, and 25% outdoors away from the site. For the time spent indoors the structure was assumed to reduce the external exposure by 30% and the dust level by 40%. Of the scenarios described above, industrial use is the most realistic given the current use of the site; the recreational scenario provides a plausible alternative use. The residential farm scenario, while hypothetically possible, is unlikely, but it provided the most restrictive activity concentration guideline corresponding to the basic dose limit.

Potential radiation doses resulting from eight exposure pathways were considered: direct exposure to external radiation from the decontaminated soil; internal radiation from inhalation of dust; internal radiation from inhalation of emanating Rn-222; internal radiation from ingestion of plant foods grown at the decontaminated site and irrigated with water drawn from the downgradient pond or the well; internal radiation from consumption of meat from livestock fed with fodder grown at the decontaminated site and water drawn from the pond or the well; internal radiation from ingestion of milk from similarly raised livestock; internal radiation from ingestion of fish from the pond; and internal radiation from drinking water drawn from the pond or the well.

For a specific scenario, only the relevant pathways were considered and others were suppressed in the RESRAD calculations. For example, for the industrial and recreational use scenarios, only the first three pathways were applicable, whereas for the residential farm scenario, all pathways were relevant.

Details of the calculation are available in Cheng, Yu, and Devgun 1991; the calculated residual radioactivity guidelines for this site are quoted here in Table I. Remedial action at the site was completed in 1992 and involved removal of contaminated soils to the Oak Ridge Reservation of DOE. A cleanup level of 35 pCi/g for U-238 was used and a verification survey confirmed that the residual concentrations after the cleanup were well below this level (Vitkus and Bright 1992).

Table I. Residual Radioactive Material Guidelines for the Case Study Site

Radionuclide	Guideline (pCi/g) Industrial Scenario	Recreation Scenario	Farmer/Pond Scenario	Farmer/Well Scenario
Uranium-234	2,400	5,400	590	120
Uranium-235	450	1,100	150	47
Uranium-238	1,600	3,600	430	120
Total Uranium	1,800	4,000	470	120

(Source: Cheng, Yu, and Devgun 1991)

CONCLUSIONS

Directly applicable cleanup limits are available for certain radionuclides. For others, site-specific guidelines must be derived for residual material that can be left in the soil. Pathways analysis methods provide a tool for deriving such cleanup guidelines. In this paper, the derivation of dose-based cleanup guidelines has been illustrated through the use of DOE's RESRAD computer code and a dose limit of 100 mrem/y. Application of derived guidelines, however, depends on several other factors, including regulatory and sociopolitical considerations. The actual cleanup limits may be much lower because of the application of the ALARA process as well as the consideration of other factors.

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