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Release Criteria and Pathway Analysis for Radiological Remediation

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ABSTRACT

Site-specific activity concentrations were derived for soils contaminated with mixed fission products (MFP), or uranium-processing residues, using the Department of Energy (DOE) pathway analysis computer code RESRAD at four different sites. The concentrations and other radiological parameters, such as limits on background-subtracted gamma exposure rate, were used as the basis to arrive at release criteria for two of the sites. Valid statistical parameters, calculated for the distribution of radiological data obtained from site surveys, were then compared with the criteria to determine releasability or need for further decontamination. For the other two sites, RESRAD has been used as a preremediation planning tool to derive residual material guidelines for uranium.

INTRODUCTION

Release of radioactively contaminated sites for safe and unrestricted future use requires proof that radiological data obtained from the site meet regulatory criteria for such a release. The release criteria are typically a composite of acceptance limits that depend on the radionuclides, the media in which they are present, and on federal and local regulations. In recent years, the DOE has established a pathway analysis model to determine site-specific soil activity concentration guidelines for those radionuclides that do not have generic acceptance limits (1). The pathway analysis computer code developed by the DOE is called RESRAD. Similar efforts have been initiated by the U. S. Nuclear Regulatory Commission (NRC) to develop and use dose-related criteria based on generic pathway analyses (2) instead of simplistic numerical limits on residual radioactivity. This NRC effort is directed toward justifying release of structures at decontaminated facilities while the DOE analysis considers the potential effects of residual radioactivity in soil.

Radiological surveys performed at four sites are reported in this paper. Of the four, two were investigated for old mixed fission products (MFP) (primarily Cs-137 and Sr-90) and the other two for uranium residues. The RESRAD code was used in all cases to determine the soil activity concentration guidelines for these radionuclides. Additional site-specific release criteria were also established and generic regulatory limits were used as appropriate. In what follows, we briefly describe the case histories, the criteria, and use of RESRAD.

BACKGROUND

MFP Cases. Formerly used and adjoining areas of a nuclear test facility in an arid region in Southern California were radiologically surveyed for residual radioactivity. Both structures (buildings) and open sites within the 117-hectare (290-acre) facility were surveyed. The two cases reported here include an isolated storage yard and a side yard adjacent to a building. In both cases, slight contamination of the soil with Cs-137 was measured and contamination with the associated fission product nuclide, Sr-90, was assumed. Following the remedial actions and analysis of data from radiological surveys reported here, residual activity at the two locations was determined to be well below the acceptance limits for release without radiological restrictions.

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Uranium Cases. The two cases presented are sites under the DOE's Formerly Utilized Sites Remedial Action Program (FUSRAP), both contaminated with uranium. One site (in western New York) is a 55-hectare (135-acre) currently operating industrial facility where residual contamination remains from uranium ore processing operations carried out between 1942 and 1948 for the Manhattan Engineer District (MED). While processing residues were disposed off-site, uranium contamination currently exists at the site in several discrete areas, including buildings, a parking lot, and both surface and subsurface soils. The other uranium site, located in eastern Tennessee, is an 8-hectare (20-acre) area that is now being developed for industrial use. The site became contaminated when during the 1940s the MED, 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 subsequent surveys by the Tennessee Department of Health and Environment and with the emergence of stricter release criteria, the site was added to FUSRAP and is scheduled for further cleanup under the program. Residual radioactive material guidelines for uranium for planning and decontamination of both these sites were derived using RESRAD.

RELEASE CRITERIA

General. Remedial actions are undertaken with the ultimate objective of protecting public health and safety. Residual radioactivity levels at remediated sites should, therefore, be below certain limits so that a future occupant may use the site without radiological restrictions. Specifying the types of acceptance limits and establishing values, however, depends on numerous factors, some of which are discussed below.

Inspection-oriented numerical limits exist, such as those specified for residual surface contamination and in the NRC Regulatory Guide 1.86. These limits are the most directly applicable, and they facilitate confirmation of the decontamination operation, but they are not necessarily cost effective.

Dose-based limits, using dose conversion factors, such as those being currently investigated by the NRC (2), may be used. These allow greater flexibility in decontamination while achieving protection goals more directly than numerical limits.

Dose-limited pathways analysis, with generic or site-specific parameters, may be used, as provided by RESRAD. This 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.

Further considerations in selecting guidelines and acceptance criteria for remediation are:

1. External Exposure. Limits on dose rates from decontaminated or remediated buildings or sites are specified by agencies such as the NRC or the DOE (3).

2. Affected Media. Maximum release limits may be specified for water, air, and/or soil. For the cases of soil contamination, generic limits are available only for Ra-226 by the DOE and others, and for uranium and thorium by the NRC (4). Site-specific activity concentrations must be established for other radionuclides. For the present work, these were established using RESRAD for the MFP as well as uranium [as required by Ref. (3) for FUSRAP sites] and are the principal subject of discussion in this paper.

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

Release Criteria for the MFP Locations. Although the DOE guidelines (3) recommend a value of 20 μR/h (at 1 m) above background for gamma exposure rates, a lower value of 5 μR/h above background was chosen for these surveys and was based on a previous NRC stipulation for the unrestricted release of a dismantled test reactor facility in the complex. Also, the 5 μR/h above background corresponds to the recently issued NRC limit of 10 mrem/yr (2000-h occupancy) under NRC's "Below Regulatory Concern" policy (5).

Criteria for residual Cs-137 activity concentration in soils were established for the above cases using the RESRAD code. Combined presence, in equal activity concentrations, of both Cs-137 and Sr-90 was assumed in both cases because of a conservative determination that the incidents leading to the contamination of these sites involved the presence of MFP. Selection of parameters and future-use scenarios employed in the calculations of the activity concentrations are discussed in the next section.

Measured gamma exposure rate and soil activity concentration data were also statistically analyzed with respect to the State of California guidelines (6) to demonstrate compliance. The statistical analysis procedure, known as sampling inspection by variables (7), is widely used in industry and the military. Its application to radiological surveys is described in Ref. (8). It suffices to state here that analysis of the measured data in this manner results in a parameter known as the test statistic (TS). To satisfy the State of California guideline for the MFP sites, it was necessary to demonstrate that the TS values for the gamma exposure rates and the soil activity concentrations were less than the 5 μR/h and the RESRAD-calculated limit, respectively.

Release Criteria for the Uranium Residue Locations. The above-referenced DOE guidelines apply to these FUSRAP sites, including the 100 mrem/yr "basic dose limit" for an individual member of the general public. Similarly, activity concentration guidelines for uranium isotopes and total uranium were calculated for these sites using RESRAD. The generic guideline specified for surface contamination and airborne radon decay product will also be applicable as remediation efforts progress at these sites.

PATHWAY ANALYSIS USING RESRAD

The pathway analysis method provides an estimate of a site-specific or generic radiation dose, subject to certain use conditions (scenarios), depending on whether the calculational parameters are accurate values for the site or are more general (representative) values, respectively. Accurate, site-specific values should be used to the maximum extent practical, for this not only improves the accuracy of the resulting dose but also enhances the acceptability of both the method and the results. As estimated doses approach limiting levels, it is important to use accurate values to ensure credibility of the result and avoid bias.

The RESRAD code is designed for interpretation of situations involving distributed radioactivity in soil and can aid investigations of long-term changes in the site. As described in Ref. (1), RESRAD incorporates considerations of the external radiation, inhalation, and ingestion pathways to exposure from sources in the ground, air, water, and in the food chain. Generic values for the hydrogeological parameters, food, water, and air consumption are provided as "default" inputs for the case of a family farm scenario. In all, there are about 80 input parameters. A library of radionuclides is included in the program.

The default scenario and associated parameters can be used for performing screening evaluation. Based on site characteristics and plausible scenarios for site use, the parameters are then modified. Sensitivity analyses are also performed to identify those parameters that significantly affect the end results, allowing the user to focus on obtaining accurate site-specific values for these parameters.

Based on the site- and scenario-specific inputs thus 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/yr. Here, the basic dose limit is the input, and the resulting RESRAD-calculated concentration is the limit. In this context, RESRAD can be used as a preremediation planning tool (the uranium residue locations discussed here) where limits on soil concentration are developed and variations in cleanup decisions are tested for "as low as reasonably achievable" (ALARA) considerations. For postremediation cases (the MFP locations), RESRAD provides a means to not only compare the measured soil concentrations with the limit but also to estimate the dose corresponding to the measured concentration (used as input) to demonstrate the effectiveness of the cleanup accomplished by the remediation. If the measured concentration and its extent are less than the limit, the resulting RESRAD-calculated dose will be less than the basic dose limit, satisfying this acceptance criterion. As discussed below, RESRAD was used in both these modes.

RESRAD PARAMETERS AND RESULTS

MFP Cases. The hydrogeological parameters employed in the MFP cases are nearly identical because they are part of the same complex, located in the arid southwestern United States. These parameters were determined for the complex and used as inputs to RESRAD. As for use scenarios, the sites are integral to ongoing industrial activities, and, thus, the industrial use scenario would currently be appropriate. Other credible future-use scenarios include residential or wilderness, given their proximity to both suburban and recreational locations. For these reasons, the family farm scenario, although used with the default RESRAD dietary and occupancy parameters for screening calculations, is not considered credible.

With site-specific hydrogeological data and modified dietary and occupancy parameters, RESRAD calculations were performed to determine the soil activity concentrations for the combined presence of Cs-137 and Sr-90 for the three credible scenarios. The screening and sensitivity calculations showed that conservative values should be used with regard to the extent of contamination (area and depth), the cover depth, and occupancy/inhalation shielding factors. For this arid region and for these credible scenarios, the results showed the majority of dose contributions are from external exposure pathways, which are determined by these factors. Thus, although the areas and extent of the contamination were relatively small, an "infinite" contamination area and depth were chosen for determining the limits. An area of 100,000 m^2 and a depth of 1 m, as inputs to RESRAD, correspond to the infinite case: that is, the calculated dose remains the same at larger values of these two parameters.

Of the two MFP cases, the storage yard area is smaller than that of the side yard. Therefore, different values were used for the shielding factors associated with occupancy and inhalation. Because of the relatively larger area of the side yard, the residential or industrial occupant would be exposed to more direct external exposures when he or she is outside of the building. For the smaller storage yard, a uniform slab shielding was a more accurate approximation. For the wilderness scenario, however, no shielding was appropriate.

Table I shows the RESRAD-calculated Cs-137 concentration limits for the three credible scenarios, using the infinite extent contamination and the applicable occupancy- and inhalation-shielding factors for the two areas. At both sites, all other variables were nearly identical except for the shielding factors. Reductions in external exposure rates by the uniform slab, assumed for the case of the storage yard, is the cause of the increased values for the limits for the residential and industrial scenarios. Because of the absence of any shielding in the wilderness scenario, the corresponding limits remained nearly the same for both locations.

The lowest value for either site corresponds to the residential scenario. Thus, if the measured concentrations were below this, then the site can be used for all the credible uses: that is, it can be released for unrestricted use. The lowest value is then the limit corresponding to the "credible and bounding" residential scenario.

Data shown in Table I are for the presence of Cs-137 alone. Because of the MFP-related assumption that an equal activity concentration of Sr-90 is also present, corresponding values for this

Table I. Cs-137 Soil Activity Concentration Limits (pCi/g)

Scenario	Limit (pCi/g)	
	Storage Yard	Side Yard
Industrial	2,520	239
Residential	984	71
Wilderness	3,840	3,830

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radionuclide were also calculated using RESRAD. For the combined presence of both nuclides, in equal activity concentrations, the resulting acceptance limits for the credible bounding residential scenario were 314 and 60 pCi/g for the storage yard and side yard, respectively.

Results of activity concentrations measured from the soil samples collected from the two yards after their remediation can now be compared with the above limits. These are shown in Figs. 1 and 2 for the storage yard and side yard, respectively. Both figures have the measured Cs-137 soil activity concentration values (in pCi/g) on the y-axis and the Gaussian cumulative probability calculated for that distribution of data on the x-axis.

For acceptance, the TS calculated for the data must be below the acceptance limit. In Fig. 1, the TS for the storage yard data is shown at 29.5 pCi/g, which is well below the acceptance limit of 314 pCi/g. Similarly, for the side yard, Fig. 2 shows a TS of 11.7 pCi/g, much lower than the

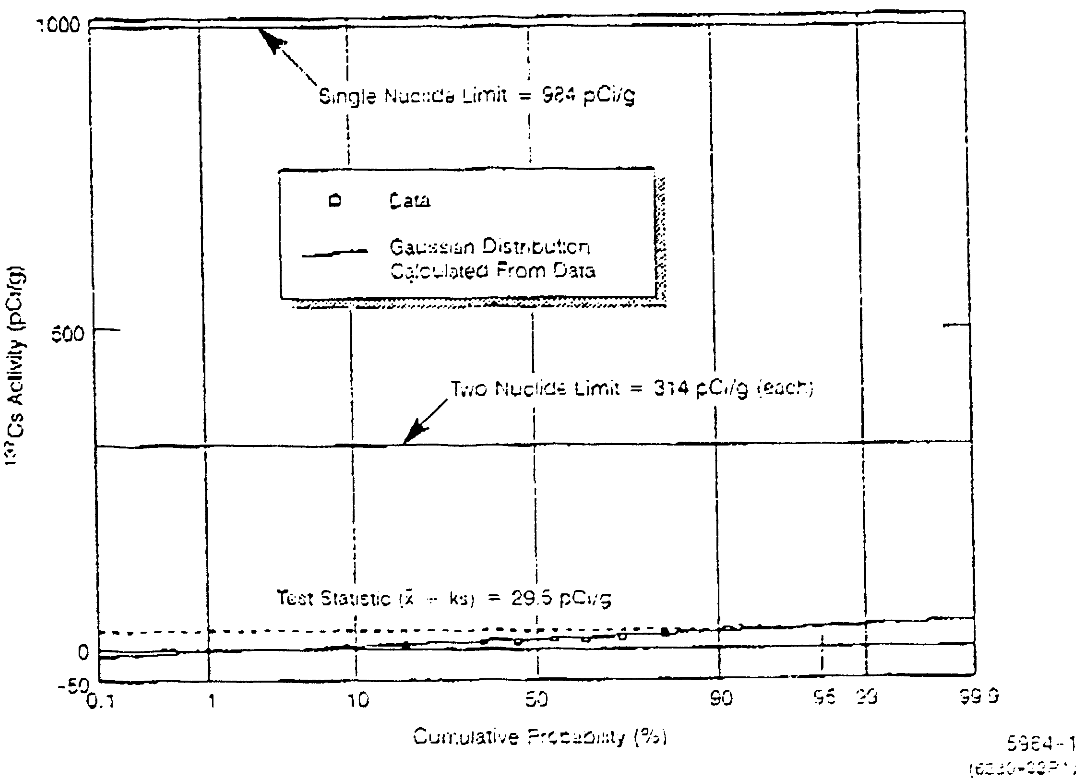

Fig. 1. Measured Cs-137 Activity in the Storage Yard Compared with RESRAD-Calculated Limits

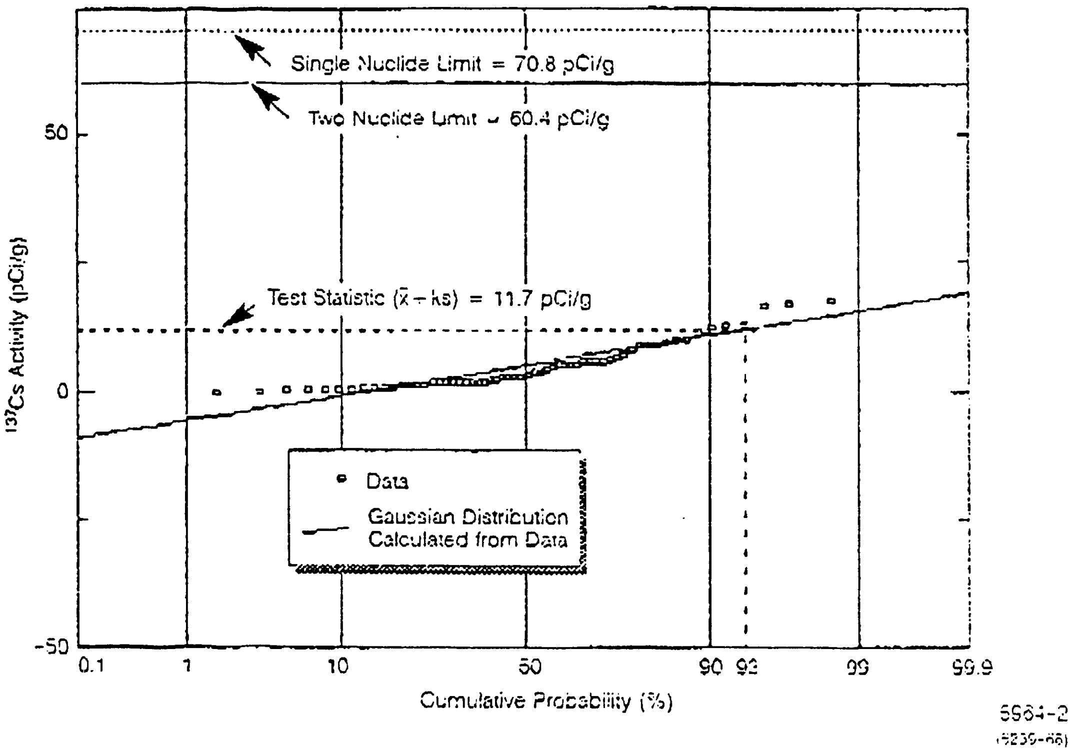

Fig. 2. Measured Cs-137 Activity in the Side Yard Compared With RESRAD-Calculated Limits

acceptance limit of 60 pCi/g. The data are also within the calculated Gaussian line, showing no significant deviations.

The average of the data shown for the two yards was also used to estimate, using RESRAD, the annual dose for a hypothetical current resident. In each case, actual areas and conservative estimates of the depth of the soil contamination were used. Results showed annual doses of 0.39 and 5.2 mrem, respectively, for the storage yard and side yard, both well below the 100 mrem basic dose limit.

Recalling that regulatory compliance also required that gamma exposure rates for the two yards be less than 5 μR/h above background, the survey data on these exposure rates were also examined. Statistical plots of the data, collected on gridded locations 1 m above ground, are shown in Figs. 3 and 4 for the side yard before and after remediation, respectively. The ambient exposure rates shown in Fig. 3 are for a larger area covering the side yard before remediation. The average of background data obtained from the nearby unaffected areas is 15.6 μR/h. About a dozen grid points are well above this background and also above the 5 μR/h above-background limit. The number of grid locations with such elevated exposure rates and their magnitudes resulted in the significant departure from the Gaussian line shown for the data. It was indeed from these data, and from preliminary analysis of soils collected from selected locations which showed Cs-137 contamination, that a determination was made to proceed with soil cleanup of the side yard. Data shown in Fig. 4 are from grid locations in the side yard after the cleanup and after subtracting the 15.6 μR/h background. The TS value for this data, 3.6 μR/h, is within the limit. The data also closely follow the Gaussian line in this figure. Similar compliance was achieved with respect to the storage yard.

Uranium Residue Cases. At the two FUSRAP sites, residual radioactive material guidelines were derived for total uranium and uranium isotopes using site-specific parameters and different scenarios as RESRAD inputs. The guidelines were derived on the basis of meeting the 100

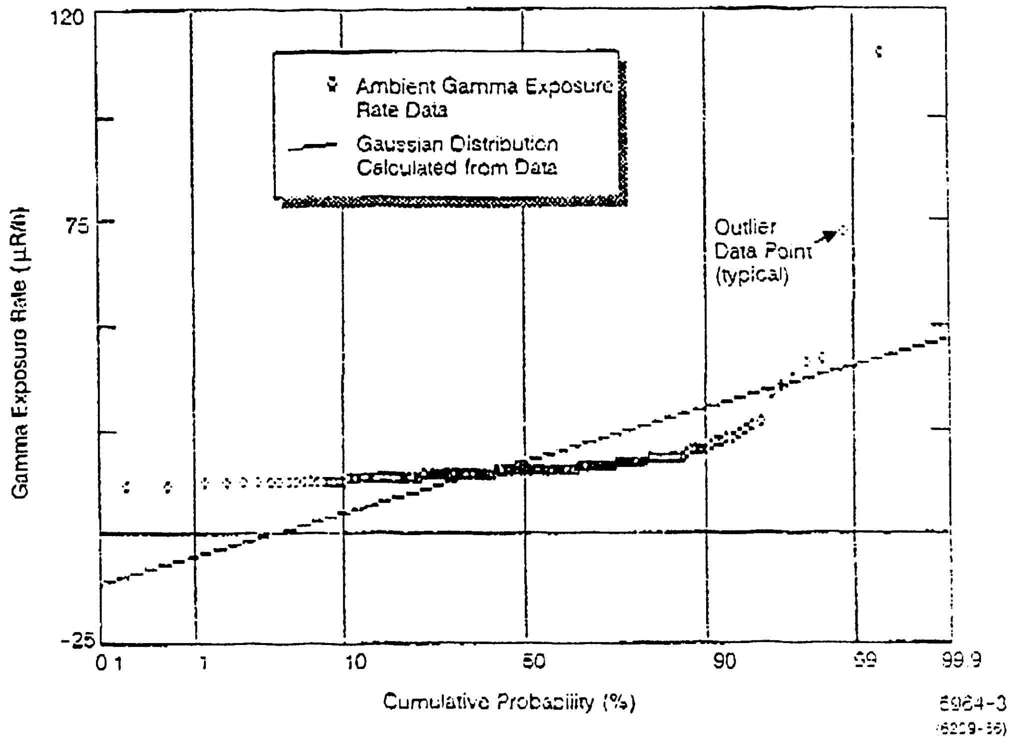

Fig. 3. Ambient Gamma Radiation in Locations In and Around the Side Yard Before Remediation

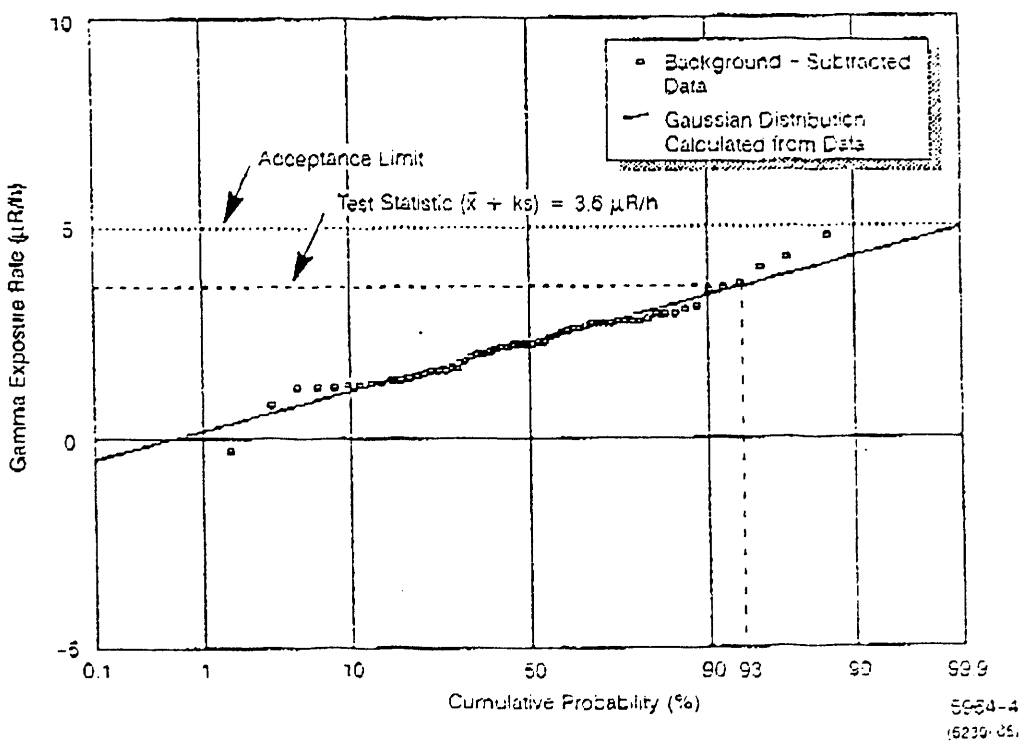

Fig. 4. Background-Subtracted Gamma Exposure Rates in the Side Yard (After Decontamination Remediation) Compared with Acceptance Limit

mrem/yr basic dose limit and assuming that U-238, U-234, and U-235 are present in their natural activity ratio of 1:1:0.046. It was also assumed that uranium is the only radionuclide present at an above-background concentration. All scenarios assumed that at some time within 1000 years following decontamination the area is released for use without radiological restrictions. The detailed assumptions and input data are available in Argonne National Laboratory reports (9,10), the results of which are presented here.

Three potential scenarios were considered for the New York site: industrial, residential farm, and recreational. Pond water is assumed to be the only usable water, based on site geological and other considerations (9). The industrial scenario assumes use of the site by a hypothetical worker who spends 2000 h annually (75% outdoor and 25% indoor) at the site and does not ingest any water or food from the site. The residential farm scenario assumes that a hypothetical occupant sets up residence in the immediate vicinity of the site, drinks water from a natural pond downstream of the site, eats plant foods grown there, and consumes meat and milk from livestock raised there. Fish consumption from the pond is also included. The recreational scenario (use as a public park) assumes the user spends 750 h annually at the site but does not consume water or food from the decontaminated site. Of the three scenarios, the industrial use is the most realistic, given current use of the site, and the recreational scenario provides a plausible alternative use. The residential farm scenario, while hypothetically possible, is unlikely but does provide a conservative upper bound dose estimate.

Potential radiation doses resulting from the RESRAD-identified exposure pathways were considered. The seven pathways are:

1. Direct exposure to external radiation from the decontaminated soil
2. Internal radiation from inhalation of dust
3. Internal radiation from ingestion of plant foods grown at the site and irrigated with water drawn from the down-gradient side of the pond
4. Internal radiation from consumption of meat from livestock fed with fodder grown at the site and water drawn from the pond
5. Internal radiation from consumption of milk from similarly raised livestock
6. Internal radiation from consumption of fish from the pond
7. Internal radiation from drinking water drawn from the pond.

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 pathways 1 and 2 are applicable, whereas for the residential farm scenario, all pathways, 1 through 7, are relevant.

Table II provides the guideline values [in (pCi/g)] for residual radioactivities in the soil for the New York site, calculated by RESRAD that would result in a 100 mrem/yr exposure from the uranium isotopes and total natural uranium.

For the Tennessee site, the industrial and recreational scenarios are similar to those discussed above. The residential farmer scenario has two variations: an adjacent pond provides the source of usable water in one, while the other uses groundwater drawn from a well at the down-gradient edge of the site. The exposure pathways are similar to those described earlier; however, an additional pathway of internal radiation from inhalation of emanating Radon-222 was also included. Again, pathways not relevant to a specific scenario were suppressed in the RESRAD calculations. Table III shows the resulting residual radioactive material guidelines for this site.

Table II. Residual Radioactive Material Guidelines for New York Site (9)

Radionuclide	Guideline (pCi/g)		
	Industrial Use	Residential Use	Recreational
Uranium-234	2,400	77	5,300
Uranium-235	480	27	1,200
Uranium-238	1,600	83	3,700
Total Uranium	1,800	77	4,200

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When implementing the above derived radionuclide guidelines for decontamination of the site when other nuclides are present, the law of the sum of fractions applies, as was the case for the combined presence of Cs-137 and Sr-90 for the MFP sites discussed earlier. As such, the data shown in Table II for the residential farmer and in Table III for the farmer/well scenarios provide the most restrictive guidelines on residual concentrations of uranium. The decision making on residual concentrations is, however, a complex process where other factors must be considered. These may include interagency agreements and past lower cleanup precedents, as was the case, for example, for the low limit of 5 μR/h above-background gamma exposure rate applied in the MFP sites. Similarly, relevant release criteria on exposure rates, surface contamination levels, and radon levels also need to be satisfied for the two uranium residue sites. For example, reference (11) provides such data and comparisons for the Tennessee site.

Table III. Residual Radioactive Material Guidelines for Tennessee Site (10)

Radionuclide	Guideline (pCi/g)			
	Industrial	Recreation	Farmer/Pond	Farmer/Well
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

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SUMMARY

1. The RESRAD pathway analysis code was used to determine soil activity concentration limits for four sites. Of the four, two are sites remediated for the presence of MFP (Cs-137 and Sr-90) and the other two are sites with uranium ore residues.

2. Site-specific hydrogeological variables were developed and used as inputs to RESRAD in all cases. A variety of credible use scenarios were considered and soil activity concentrations were derived for these scenarios. The lowest of the concentrations determined the credible and bounding scenario.

3. In the remediated MFP sites, the soil activity concentration for the credible and bounding scenario (residential) was used as the acceptance limit for MFP

residual radioactivity. The test statistic calculated from the measured soil activity concentration data at the two sites were well below the limits. Similarly, data on the gamma exposure rates were also found to satisfy the corresponding acceptance limits for the two sites. Therefore, the sites can be released for use without radiological restrictions.

4. In the uranium residue cases, RESRAD was used as a preremediation planning tool. The soil activity concentrations established for the residential farm scenario provide the most restrictive guidelines for the decision making in regard to future remediation of the sites, along with other release criteria.

REFERENCES

1. T. L. Gilbert, et al., "A Manual for Implementing Residual Radioactive Material Guidelines," ANL/ES-160, DOE/CH/8901, Argonne National Laboratory (for U.S. Department of Energy), June 1989.

2. W. E. Kennedy, Jr., and R. A. Peloquin, "Residual Radioactive Contamination from Decommissioning - A Technical Basis for Translating Contamination Levels to Annual Dose," NUREG/CR-5512 (also PNL-7212), (Draft) January 1990.

3. U.S. Department of Energy, "Radiation Protection of the Public and the Environment," DOE Order 5400.5, February 1990.

4. U.S. Nuclear Regulatory Commission, "Disposal or Onsite Storage of Thorium or Uranium Wastes From Past Operations," Federal Register, vol. 40, no. 205, October 23, 1981.

5. "Below Regulatory Concern; Policy Statement," U.S. Nuclear Regulatory Commission, July 1990, and "Below Regulatory Concern - A Guide to the Nuclear Regulatory Commission's Policy on the Exemption of Very Low-Level Radioactive Material, Wastes and Practices," NUREG/BR-0157, July 1990.

6. "State of California Guidelines for Decontaminating Facilities and Equipment Prior to Release for Unrestricted Use," DECON-1, June 1977.

7. "Sampling Procedures and Tables for Inspection by Variables for Percent Defective," MIL-STD-414, June 1957.

8. G. Subbaraman, R. J. Tuttle, and B. M. Oliver, "Radiological Surveys: Methods, Criteria, and Their Implementation," Proceedings of Waste Management '91, Tucson, Arizona, vol. 2, p. 121-127, 1991.

9. J. S. Devgun, G. Hyatt, and C. Yu, "Derivation of Uranium Residual Radioactive Material Guidelines for the Linde Air Products Site," Argonne National Laboratory report, October 1990.

10. J. J. Cheng, C. Yu, and J. S. Devgun, "Derivation of Uranium Residual Radioactive Material Guidelines for the Elza Gate Site," Argonne National Laboratory report, February 1991.

11. J. S. Devgun, "Radiological Risk Assessment of a Radioactively Contaminated Site," Proceedings of Waste Management '90, vol. I, p. 513-518, Tucson, Arizona 1990.

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