

FINAL REPORT

Task 6: Sampling Protocols

Radionuclide Soil Action Level Oversight Panel

December 1999

*Submitted to the Radionuclide Soil Action Level Oversight Panel
in Partial Fulfillment of Contract between RAC and the Rocky Flats Citizen's Advisory Board*



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EXECUTIVE SUMMARY

The primary goal of Task 6 is to develop recommendations for a soil sampling protocol for use at the Rocky Flats Environmental Technology Site (RFETS) to obtain soil concentration data for comparison to the soil action levels. Sampling protocols are written descriptions of the detailed procedures to be followed in collecting, packaging, labeling, preserving, transporting, and documenting the samples.

Sampling protocols are developed using the iterative data quality objective (DQO) process and require the U.S. Department of Energy (DOE) and its contractor to evaluate several important considerations. These considerations include evaluating sampling and analytical costs in relation to available resources and accepting potential decision errors that may result in remediating sites that are judged contaminated when they are actually below the soil action levels. Conversely, developing a sampling protocol must also incorporate the concerns of the general public and other stakeholders, which are represented by the Radionuclide Soil Action Level Oversight Panel (RSALOP) and the soil action level study. Because of the complexity of developing sampling protocols, with the inherent need to balance the concerns of DOE and the RSALOP, developing a comprehensive sampling protocol was not considered possible. In this report, *Risk Assessment Corporation (RAC)* presents recommendations for those elements of a soil sampling protocol considered essential to ensure representative soil samples are collected for comparison to the soil action levels. These recommendations are provided to the RSALOP for presentation to DOE and its contractor, Kaiser-Hill Company, for incorporation into the soil sampling protocol and procedures to be used for the soil action level study.

RAC conducted a review of the current sampling program used at the RFETS and found that a specific sampling protocol for the soil action levels study had not been developed. However, during this review, several procedures were identified that are available in the Rocky Flats program for incorporation into a sampling protocol. Current procedures for packaging, labeling, preserving, transporting, and documenting the samples were considered appropriate for use in a soil sampling protocol for the soil action levels study. Therefore, the main emphasis of the sampling protocol recommendations is directed toward sample collection and sampling designs. This report reviews several methods currently in use at the RFETS for collecting soil samples. The main concern with sample collection is to ensure that representative samples of the surface soil (i.e., 0 to 20-cm depth based on the conceptual model for the soil action levels) are collected. Soil sample collection protocols must be based upon the conceptual model used to derive the soil action levels to ensure that representative soil samples are collected.

This report presents recommendations for a soil sampling protocol to support the final status survey. The final status survey determines the final condition of the site and is performed after decontamination activities are complete. This survey provides the data to demonstrate that radionuclide concentrations in soil satisfy the established soil action levels.

Recommendations for a sampling protocol in support of remedial action were not developed for the Task 6 report. Soil sampling in support of remedial action is an important concept, however, a large number of soil samples have already been collected for use in evaluating the nature and extent of contamination in the surface soil at the RFETS (see the

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Task 3 report). Several studies detailing the nature and extent of contamination in the surface soil at the RFETS have also been conducted and are available for use in evaluating remedial requirements.

The U.S. Environmental Protection Agency, U.S. Nuclear Regulatory Commission, DOE, and U.S. Department of Defense developed *the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM)* (NRC 1997), which provides detailed guidance for planning, implementing, and evaluating environmental and facility radiological surveys conducted to demonstrate compliance with dose-based soil action levels. The NRC (1997) guidance focuses on demonstrating compliance based on the final status survey results. NRC (1997) is the most comprehensive guidance document currently available for developing radiological surveys. In this report, RAC uses the general principles of the MARSSIM guidance for developing recommendations for a sampling protocol and emphasizes problems with applying the MARSSIM guidance to the soil action levels at Rocky Flats and potential solutions to these problems.

RAC provides several recommendations in this report in support of developing a surface-soil sampling protocol for the final status survey. The following is a general summary of the recommendations with references to the report sections that provide additional detail. RAC recommends that

1. The DQO process be used to develop the soil sampling protocol for the final status survey (Section 3.1).
2. DOE appoint representatives from the RSALOP for inclusion on the DQO planning team (Section 3.1).
3. The RSALOP select discrete values from the soil action level distributions, for each radionuclide, and use these discrete values for comparison to the soil concentration data (Section 3.2).
4. Soil samples be collected using profile sampling (Section 4.1).
5. Profile sampling be conducted in soil depth increments of 0–3 cm to be consistent with the resuspension model parameters used to develop the soil action levels (Section 4.1.4).
6. Soil samples should not be composited, rather, individual soil samples should be analyzed for radionuclide contaminants (Section 4.1.6).
7. Soil samples be collected using a systematic grid sampling design, with a random starting point (Section 4.2).
8. A statistician familiar with the RFETS and environmental statistical designs be included on the DQO planning team (Section 4.3).

9. The arithmetic mean of the soil concentration data and its associated uncertainty at the upper 95% confidence interval be used for comparison to the soil action levels (Section 4.3).
10. The MARSSIM (NRC 1997) non-parametric statistical tests not be used for the soil action level study because these tests compare the median value of the sample distribution to the soil action levels. When the distribution is not symmetrical (i.e., skewed), the median is not equal to the mean (Section 4.3.3).
11. Parametric statistical tests, bootstrapping, or geostatistical techniques be investigated for use in comparing the 95% upper confidence limit of the mean against the soil action levels. The statistical tests should be investigated during the DQO process and chosen according to knowledge of the areas to be sampled (Section 4.3.3).
12. Spatial correlations be investigated to determine their presence in the survey unit of interest and to determine if methods are required to improve the estimate of the mean based on the systematic grid sampling method (Section 4.3.4).
13. The null (H_0) and alternative (H_a) hypothesis are stated as $H_0: \mu \leq \text{SAL}$ and $H_a: \mu > \text{SAL}$ (Section 4.3.5) where μ is the mean soil concentration and SAL is the soil action level.
14. The survey units be classified according to the NRC (1997) scheme and that the size of the survey units be limited accordingly (Section 4.6).
15. In situ gamma spectroscopy measurement be performed to identify potential hot spot locations (Section 4.7).
16. Hot spots identified by soil samples or in situ gamma spectroscopy measurements be investigated further to delineate the size of the hot spot and to determine the upper 95% confidence interval of the mean radionuclide concentrations contained in the hot spot (Section 4.7).
17. That hot spots greater than 100 m² with arithmetic mean soil concentrations at the upper 95% confidence interval that exceed the soil action levels be remediated (Section 4.7).
18. That hot spots less than 100 m² be area averaged with soil concentrations in a 100 m² area and area weighted to determine if the upper 95% confidence interval of the mean soil concentration exceeds the soil action levels and, thus, requires remediation (Section 4.7).

19. The use of surrogate measurements in the final status survey. The ratio of ^{241}Am to $^{239,240}\text{Pu}$ provides a mechanism for the in situ gamma spectroscopy measurement of ^{241}Am to be used to predict the soil concentrations of $^{239,240}\text{Pu}$ (Section 4.8).
20. DOE implement an independent verification survey for the radionuclide soil action level project. This survey should be performed by an independent third party (Section 4.9).

CONTENTS

| | |
|--|----|
| EXECUTIVE SUMMARY..... | i |
| ACRONYMS..... | xi |
| 1. INTRODUCTION..... | 1 |
| 2. REVIEW OF THE ROCKY FLATS SOIL SAMPLING PROGRAM..... | 3 |
| 2.1 Quality Assurance Project Plan..... | 3 |
| 2.2 RFETS Standard Operating Procedures..... | 4 |
| 2.2.1 Environmental Management Administrative Procedures..... | 4 |
| 2.2.2 Environmental Management Operating Procedures..... | 5 |
| 2.3 RFETS Analytical Laboratory Requirements..... | 5 |
| 2.4 RFETS Data Validation Requirements..... | 6 |
| 2.5 RFETS Soil Sample Collection Protocols..... | 6 |
| 2.5.1 Colorado Department of Public Health and Environment Method..... | 7 |
| 2.5.2 Rocky Flats Method..... | 8 |
| 2.5.3 Grab Sampling Method (Spade and Scoop)..... | 9 |
| 2.5.4 Vertical Soil Profile Method..... | 9 |
| 2.6 Summary of RFETS Soil Sampling Program Review..... | 10 |
| 3. SAMPLING DESIGN DEVELOPMENT PROCESS..... | 11 |
| 3.1 Data Quality Objectives Process..... | 11 |
| 3.1.1 Data Quality Objectives Process Description..... | 11 |
| 3.1.2 Recommendations..... | 15 |
| 3.2 Soil Action Levels..... | 15 |
| 3.2.1 Development Method..... | 15 |
| 3.2.2 Recommendations..... | 17 |
| 4. SAMPLING PROTOCOL RECOMMENDATIONS..... | 18 |
| 4.1 Soil Sample Collection Protocol..... | 18 |
| 4.1.1 Protocol Comparison Studies..... | 18 |
| 4.1.2 Depth Distribution of Plutonium and Americium..... | 19 |
| 4.1.3 Depth Distribution of the Uranium Isotopes..... | 21 |
| 4.1.4 Conceptual Model Considerations..... | 23 |
| 4.1.5 Dominant Exposure Pathways..... | 23 |
| 4.1.6 Sample Compositing..... | 24 |
| 4.1.7 Recommendations..... | 24 |
| 4.2 Sampling Design Selection..... | 25 |
| 4.2.1 Random Sampling..... | 25 |
| 4.2.2 Stratified Random Sampling..... | 26 |
| 4.2.3 Systematic Sampling..... | 27 |
| 4.2.4 Recommendations..... | 30 |

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*“Setting the standard in environmental
health”*

| | |
|--|----|
| 4.3 STATISTICAL CONCEPTS..... | 30 |
| 4.3.1 Descriptive Measures for Comparison to the Soil Action Levels..... | 30 |
| 4.3.2 Parametric Statistical Analyses..... | 31 |
| 4.3.3 Non-parametric Statistical Analyses..... | 32 |
| 4.3.4 Correlated Data..... | 34 |
| 4.3.5 Null Hypothesis..... | 34 |
| 4.3.6 False Positive and Negative Rates..... | 35 |
| 4.3.7 Recommendations..... | 37 |
| 4.4 Radionuclide Background Soil Concentrations..... | 38 |
| 4.4.1 Background Studies..... | 38 |
| 4.4.2 Recommendations..... | 40 |
| 4.5 Determining the Required Number of Samples..... | 40 |
| 4.5.1 Methods..... | 41 |
| 4.5.2 Variability of the Radionuclide Concentrations..... | 42 |
| 4.6 Classification and Identification of Survey Units..... | 42 |
| 4.6.1 Class 1 Area..... | 43 |
| 4.6.2 Class 2 Area..... | 43 |
| 4.6.3 Class 3 Area..... | 43 |
| 4.6.4 Identification of Survey Units..... | 43 |
| 4.6.5 Recommendations..... | 44 |
| 4.7 Small Areas of Elevated Activity (Hot Spots)..... | 44 |
| 4.7.1 Definitions and Detection Methods..... | 45 |
| 4.7.2 Recommendations..... | 46 |
| 4.8 Surrogate Measurements..... | 47 |
| 4.8.1 Site-specific Studies..... | 47 |
| 4.8.2 Recommendations..... | 48 |
| 4.9 Independent Confirmatory Investigations..... | 49 |
| 4.9.1 Recommendations..... | 49 |
| 5. SOIL SAMPLING QUALITY ASSURANCE..... | 50 |
| 5.1 Data Quality Objectives and Data Quality Indicators..... | 50 |
| 5.1.1 Precision..... | 52 |
| 5.1.2 Bias..... | 53 |
| 5.1.3 Accuracy..... | 54 |
| 5.1.4 Representativeness..... | 55 |
| 5.1.5 Comparability..... | 56 |
| 5.1.6 Completeness..... | 56 |
| 5.1.7 Detection Limits..... | 57 |
| 5.2 Data Validation..... | 57 |
| 5.3 Documentation..... | 58 |
| 5.3.1 Field Operation Records..... | 58 |
| 5.3.2 Laboratory Records..... | 58 |
| 5.3.3 Data Handling Records..... | 59 |
| 5.4 Data Validation Qualifiers..... | 59 |

6. CONCLUSIONS.....60

7. REFERENCES.....63

FIGURES

| | |
|---|----|
| 2-1. CDPHE soil sampling device..... | 7 |
| 2-2. Rocky Flats soil sampling device..... | 8 |
| 2-3. Rocky Flats soil sampling location spacing..... | 9 |
| 3-1. The data quality objectives process..... | 11 |
| 3-2. Example of a stochastic soil action level that shows the probability of exceeding the 15 mrem dose limit as a function of ^{239}Pu concentration in soil..... | 17 |
| 4-1. The ^{239}Pu concentrations in soil at all depths..... | 20 |
| 4-2. Example of a simple random sampling pattern with 25 sampling points..... | 26 |
| 4-3. Example of a Stratified random sampling pattern—1 sampling point per stratum with 12 sampling points..... | 26 |
| 4-4. Systematic designs for sampling in space..... | 29 |
| 5-1. Measurement bias and random measurement uncertainty..... | 53 |

TABLES

| | |
|--|----|
| 2-1. Quality Control Sample Minimum Collection Frequency at the RFETS..... | 4 |
| 2-2. RFETS Administrative Standard Operating Procedures Related to Soil Sampling..... | 5 |
| 2-3. RFETS Standard Operating Procedures Related to Soil Sampling..... | 5 |
| 4-1. False Positive and Negative Conclusions based on the Recommended Null Hypothesis.. | 36 |
| 4-2. Background Soil Concentrations of $^{239+240}\text{Pu}$ in Soils of Colorado in Bq kg^{-1} (pCi g^{-1}) | 39 |
| 4-3. Median and 95% Sign Confidence Intervals for Concentrations and Depositions of ^{241}Am in Soil from 20 Regional Background Locations along Colorado's Front Range.. | 39 |
| 4-4. Background Activities of Uranium Isotopes in Soil..... | 40 |
| 4-5. Survey Unit Suggested Areas | 44 |
| 5-1. Upper Confidence Limits for the True Variance as a Function of the Number of Quality Control Measurements Used to Determine the Estimated Variance..... | 52 |

ACRONYMS

| | |
|------------|--|
| ASME | American Society of Mechanical Engineers |
| CDPHE | Colorado Department of Public Health and Environment |
| DOD | U.S. Department of Defense |
| DOE | U.S. Department of Energy |
| DQI | data quality indicator |
| DQO | data quality objective |
| EPA | U.S. Environmental Protection Agency |
| GRRASP | (EG&G Rocky Flats) General Radiochemistry and Routine Analytical Services Protocol |
| HPGe | hyper-pure germanium |
| MARSSIM | Multi-Agency Radiation Survey and Site Investigation Manual |
| NRC | U.S. Nuclear Regulatory Commission |
| QA | quality assurance |
| QAPjP | quality assurance project plan |
| QC | quality control |
| <i>RAC</i> | <i>Risk Assessment Corporation</i> |
| RFETS | Rocky Flats Environmental Technology Site |
| RSALOP | Radionuclide Soil Action Level Oversight Panel |
| SOP | standard operating procedure |

TASK 6: SAMPLING PROTOCOLS

1. INTRODUCTION

Soil action levels are calculated to identify the concentration of one or more radionuclides in the soil above which remedial action should be considered to prevent people from receiving unacceptable radiation doses. The soil action levels for radionuclides calculated for the Rocky Flats Environmental Technology Site (RFETS) by the U.S. Department of Energy (DOE), U.S. Environmental Protection Agency (EPA), and the Colorado Department of Public Health and Environment (CDPHE) are being reevaluated because of public concern and interest in the methods previously used and recommended soil action levels proposed. A Radionuclide Soil Action Level Oversight Panel (RSALOP) was established and a contractor hired to conduct an independent assessment and calculate soil action levels for the Rocky Flats site. *Risk Assessment Corporation (RAC)* was hired to perform the study. The Rocky Flats Citizen's Advisory Board is administering a grant provided by DOE for the review.

The primary goal of Task 6 is to develop recommendations for a soil sampling protocol for use at RFETS in support of the soil action level study. Sampling protocols are written descriptions of the detailed procedures to be followed in collecting, packaging, labeling, preserving, transporting, storing, and documenting the samples (Keith 1991). The reader is referred to the EPA's guidance document on "Preparation of Soil Sampling Protocol: Techniques and Strategies" for an elementary discussion on sampling protocols (EPA 1983).

Sampling protocols are developed using the iterative data quality objective (DQO) process and require the U.S. Department of Energy (DOE) and its contractor to evaluate several important considerations. These considerations include evaluating sampling and analytical costs in relation to available resources and accepting potential decision errors that may result in remediating sites that are judged contaminated when they are actually below the soil action levels. Conversely, developing a sampling protocol must also incorporate the concerns of the general public and other stakeholders, which are represented by the RSALOP and the soil action level study. Because of the complexity of developing sampling protocols, with the inherent need to balance the concerns of DOE and the RSALOP, developing a comprehensive sampling protocol was not considered possible. In this report, *RAC* presents recommendations for those elements of a soil sampling protocol considered essential to ensure representative soil samples are collected for comparison to the soil action levels. These recommendations are provided to the RSALOP for presentation to DOE and its contractor, Kaiser-Hill Company, for incorporation into the soil sampling protocol and procedures to be used for the soil action level study.

The primary concern of this report is to develop recommendations for the design of site-specific surface soil sampling procedures that ensure representative samples are collected to determine soil action levels. Soil sampling protocols must be based upon the conceptual model used to derive the soil action levels to ensure that representative soil samples are collected. For example, the depth to which a sample is taken may affect the measured concentration if the radionuclide is deposited in the top few centimeters. Under some circumstances, averaging over the top 15 cm (approximately 6 in.) is appropriate if the exposure pathway of concern is ingestion of food raised in the area. However, averaging may underestimate the potential dose if the exposure pathway of concern is soil ingestion or inhalation of resuspended dust (SAB 1997).

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This report presents recommendations for a soil sampling protocol to support the final status survey. The final status survey determines the final condition of the site and is performed after decontamination activities (if required) are complete. This survey provides the data to demonstrate that radionuclide concentrations in soil satisfy the established soil action levels. This survey is also referred to as a termination survey, post remedial-action survey, and final survey.

Recommendations for a sampling protocol in support of remedial action were not developed for the Task 6 report. Soil sampling in support of remedial action is an important concept; however, a large number of soil samples have already been collected for use in evaluating the nature and extent of contamination in the surface soil at the RFETS (see the Task 3 report). Several studies detailing the nature and extent of contamination in the surface soil at the RFETS have also been conducted and are available for use in evaluating remedial requirements. This is not to imply that no further surface soil studies should be conducted at RFETS in support of remedial design. In fact, as noted in this report, additional studies should be conducted for uranium contamination in the surface soil at RFETS. However, RAC and RSALOP concurred that the emphasis of this report should be placed on the final status survey.

The soil sampling protocol recommendations presented in this report are based on the conceptual model, parameterization, and assumptions used to develop the soil action levels presented in the Task 5 report. The conceptual model for the soil action levels (Task 5) report is based on the surface soil (0 to 20 cm) layer. Therefore, the recommendations presented in this report are not applicable to subsurface soil layers that may be of concern for evaluating source inventories for use in groundwater transport models. Future work by the Actinide Migration Panel may indicate that groundwater transport and seeps to surface water are important processes at the RFETS. This finding would require that a sampling protocol be developed for application to subsurface soil layers (i.e., > 20 cm).

The EPA, NRC, DOE, and DOD have developed *the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM)* (NRC 1997), which provides detailed guidance for planning, implementing, and evaluating environmental and facility radiological surveys conducted to demonstrate compliance with dose-based soil action levels. The MARSSIM guidance focuses on demonstrating compliance during the final status survey following scoping, characterization, and any necessary remedial actions. The MARSSIM (NRC 1997) is the most comprehensive guidance document currently available for the development of radiological surveys. DOE and their site contractor, Kaiser-Hill Company, have used the MARSSIM guidance for use in final status surveys of building contamination. Therefore, RAC recommends that the final status surveys conducted at RFETS follow the general principles provided in the MARSSIM guidance for the soil action level project. In this report, RAC has used the general principles of the MARSSIM (NRC 1997) guidance to develop recommendations for a sampling protocol. The MARSSIM guidance and methods should not be used blindly as a recipe for final status surveys. RAC provides an emphasis in this report on problems identified with the MARSSIM guidance in terms of application to the soil action levels at Rocky Flats and potential solutions.

2. REVIEW OF THE ROCKY FLATS SOIL SAMPLING PROGRAM

RAC conducted a review of the current sampling program and procedures used at the RFETS to determine elements of the current program available for use and incorporation into the final status sampling protocol. We reviewed quality assurance (QA) requirements, standard operating procedures (SOPs), and individual site sampling and analysis plans. This section summarizes the sampling program and procedures currently used at Rocky Flats.

A soil sampling protocol for specific application to the soil action levels was not identified during this review. Because a soil sampling protocol directly addressing the sampling requirements and associated data needs for comparison to the soil action levels has not been developed for use at Rocky Flats, this review makes no attempt to evaluate the quality of such a sampling program. The review was conducted to identify the program elements and procedures available for use in developing a soil sampling protocol for the soil action levels.

RAC identified several procedures currently used at the RFETS that can be used to develop a soil sampling protocol. Overall, the administrative and field procedures were considered to be technically sound and based on standard industry guidelines. However, soil sampling methods available at Rocky Flats were not considered adequate for ensuring that representative samples are collected for the soil action level project.

The documented sampling program at the RFETS is based on the present guidelines and requirements of the American Society of Mechanical Engineers (ASME) NQA-1 (ASME 1989); DOE Order 5700.6c (DOE 1991); DOE Order 5400.1 (DOE 1989); EPA guidance for Comprehensive Environmental Response, Compensation, and Liability Act sites (including the DQO process) (EPA 1994a); and EPA, NRC, DOE, and DOD's *Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM)* methodologies (NRC 1997). Soil sampling programs at the RFETS are largely based upon EPA's Comprehensive Environmental Response, Compensation, and Liability Act guidance, which has been adopted, in part, by NRC (1997).

2.1 Quality Assurance Project Plan

The overall soil sampling QA program document for the RFETS is contained in the quality assurance project plan (QAPjP), which is titled *Rocky Flats Plant Environmental Management Site-Wide QA Project Plan* (EG&G Rocky Flats 1994a). This QAPjP meets the requirements set forth in the following guidance and regulatory documents:

- EPA QAMS/005/80, *Interim Guidelines and Specifications for Preparing Quality Assurance Project Plans* (EPA 1980a)
- DOE Order 5700.6c, "Quality Assurance" (DOE 1991)
- DOE Order 5400.1, "General Environmental Protection Program" (DOE 1989)
- ASME NQA-1, *Quality Assurance Program Requirements for Nuclear Facilities* (ASME 1989).

The QAPjP describes the policy, organization, functional responsibilities, and QA requirements for programs at RFETS. In addition to the QAPjP, SOPs at Rocky Flats describe the field techniques to be used during soil sampling field investigations at the RFETS. The SOPs, together with the QAPjP, form the RFETS sampling and analysis plan.

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In addition to the QAPjP and SOPs, the QAPjP requires developing operable unit-specific work plans or field sampling plans. Operable unit is a term used by the EPA to identify specific contamination areas to be considered as one unit for an assessment. Work plans describe how each operable unit will be characterized and include specific operable unit background information, sampling objectives, sample locations, and minimum frequency for each task or operation.

The QAPjP also requires that each work plan be accompanied by a QA addendum. The QA addendum outlines the site- or operable unit-specific measures to be taken to meet the QA requirements in the QAPjP and references the SOPs to be followed during the investigation of a specific operable unit. In addition, specific SOPs may be developed for desired variations in the standard SOPs that are necessary for specific tasks in a particular operable unit.

The QAPjP also provides guidance on QA for soil sample data quality indicators (DQIs). Table 2-1 gives the minimum frequency requirements set forth in the QAPjP.

Table 2-1. Quality Control Sample Minimum Collection Frequency at the RFETS

| Activity | Frequency |
|----------------------------------|---|
| Field duplicate | 1 in 20 |
| Field blanks ^a | As specified in work plan/QA addendum |
| Trip blanks | As specified in work plan/QA addendum |
| Equipment rinse blanks | 1 in 20 or once per day, whichever is more frequent |
| Other quality control activities | As specified in work plan/QA addendum |

^a According to the QAPjP, the use of field blanks for soil and sediment sampling at the RFETS is not appropriate because of the lack of commercially available blank soil and solid materials that adequately reflect the various soil types encountered. Developing blank soil types within the Rocky Flats Plant region is not practical because of the subjectivity of characterizing background soil conditions and variability of soil types.

2.2 RFETS Standard Operating Procedures

The SOPs at the RFETS for soil sampling are contained in two main procedural documents: (1) *Environmental Management Administrative Procedures*, Manual No. 3-21000-ADM and (2) *Environmental Management Division (EMD) Operating Procedures*, Manual No. 5-21000-OPS.

2.2.1 Environmental Management Administrative Procedures

The EG&G Rocky Flats *Environmental Management Administrative Procedures*, Manual No. 3-21000-ADM, contains administrative-level procedures. These procedures provide the requirements for developing QA addenda, procedures, forms, and records management. Table 2-2 identifies procedures that are directly related to the topic of soil sampling at the RFETS.

Table 2-2. RFETS Administrative Standard Operating Procedures Related to Soil Sampling

| Procedure number | Procedure title |
|------------------|---|
| 3.04 | Control of Quality Assurance Addenda (QAA) Development |
| 5.01 | Procedure Development |
| 5.03 | RCRA Facility Investigation (RFI)/Remedial Investigation (RI) Work Plan Development |
| 5.08 | Forms Control |
| 6.01 | Document Control |
| 8.01 | Control and Identification of Samples and Data |
| 17.01 | Quality Assurance Records Management |

2.2.2 Environmental Management Operating Procedures

Environmental Management Division (EMD) Operating Procedures, Manual No. 5-21000-OPS, consists of several volumes of SOPs. Volumes that are pertinent to soil sampling include *Volume I: Field Operations*, Manual No. 5-21000-OPS-FO, and *Volume III: Geotechnical*, Manual No. 5-21000-OPS-GT. These SOPs are provided to the field personnel and describe the procedural steps required to complete a specific task. Table 2-3 lists the procedures in these volumes that are pertinent to soil sampling at the RFETS.

Table 2-3. RFETS Standard Operating Procedures Related to Soil Sampling

| Procedure number | Procedure title |
|------------------|---|
| FO.03 | General Equipment Decontamination |
| FO.10 | Receiving, Labeling, and Handling Environmental Materials Containers |
| FO.13 | Containerization, Preserving, Handling and Shipping of Soil and Water Samples |
| FO.18 | Environmental Sample Radioactivity Content Screening |
| GT.08 | Surface Soil Sampling |
| GT.17 | Land Surveying |

2.3 RFETS Analytical Laboratory Requirements

The *EG&G Rocky Flats General Radiochemistry and Routine Analytical Services Protocol (GRRASP)* provides the procedures for analytical laboratory work at Rocky Flats (EG&G Rocky Flats 1994b). Technical requirements in the GRRASP specify the methods to be used, required detection limits, and deliverables necessary.

The analyses of radionuclides at the RFETS are conducted in accordance with the standards and guidance set forth in the following documents:

- *Test Methods for Evaluating Solid Waste* (EPA 1986)

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- *Radiochemical Analytical Procedures for Analysis of Environmental Samples* (EPA 1979)
- *Interim Radiochemical Methodology for Drinking Water* (EPA 1976)
- *Prescribed Procedures for Measurement of Radioactivity in Drinking Water* (EPA 1980b)
- *Standard Methods for the Examination of Water and Wastewater* (APHA 1989)
- *Eastern Environmental Radiation Facility Radiochemistry Procedures Manual* (EPA 1984)
- *Methods for Chemical Analysis of Water and Wastes* (EPA 1974)
- *Procedures for Radiochemical Analysis of Nuclear Reactor Aqueous Solutions* (EPA 1973)
- *The Procedures Manual of the Environmental Measurements Laboratory* (DOE 1997)
- U.S. Nuclear Regulatory Commission Regulatory Guides.

Laboratory method detection limits and DQOs are provided in Appendix B of the QAPjP.

2.4 RFETS Data Validation Requirements

Functional guidelines for validating most radiochemistry data have not been published by EPA; however, data validation functional guidelines, applied directly from EPA Contract Laboratory Program, have been established for the RFETS. The functional guidelines used to validate analytical data at the RFETS include

- *Laboratory Data Validation Functional Guidelines for Evaluating Organic Analyses* (EPA 1988a)
- *Laboratory Data Validation Functional Guidelines for Evaluating Inorganic Analyses* (EPA 1988b)
- *Water Quality Parametric Data Validation Guidelines* (EG&G Rocky Flats 1990a)
- *Radiochemical Data Validation Guidelines—Tritium Analysis by Liquid Scintillation* (EG&G Rocky Flats 1990b)
- *Radiochemical Data Validation Guidelines—Isotopic Analyses by Gamma Spectroscopy* (EG&G Rocky Flats 1991)
- *Radiochemical Data Validation Guidelines—Gross Alpha/Beta by Gas Proportional Counters* (EG&G Rocky Flats 1990c)
- *Radiochemical Data Validation Guidelines—Isotopic Analyses by Alpha Spectroscopy* (EG&G Rocky Flats 1990d).

Laboratory quality control (QC) procedures are in place for radiochemistry. The laboratory QC procedures and samples used are described in detail in the analytical methods cited in the GRRASP.

2.5 RFETS Soil Sample Collection Protocols

Soil sample collection protocols used at the RFETS are described in EMD Operating Procedure GT.08, Manual No. 5-21000-OPS-GT, Volume III: Geotechnical. This operating procedure describes the surface (near-surface) soil sampling procedures in use at the RFETS.

Near-surface soil is defined in the SOP as those soils between the ground surface and 1 m (3.3 ft) in depth. The procedure states that

The purpose of surface soil sampling at the RFETS can be related to one or more specific objectives. These are as follows: 1) resuspension availability, which determines if radionuclides are present in the top-soil that could become resuspended in the air and thus pose a migration pathway by inhalation; 2) deposit inventories, which determine the amount of accumulated radionuclides deposited on the ground; 3) distribution of contaminants, which defines the areal distribution of contaminants; and 4) deposition increment, which defines the areal distribution with depth of radionuclides in the top 15 cm (6 inches) of soil to verify the results of the Hyper-pure germanium (HPGe) surveys.

To meet these objectives, four radionuclide, surface soil sampling methods are used at the RFETS: (1) CDPHE method, designed to sample for resuspension availability, (2) Rocky Flats method, designed to sample for deposited inventories, (3) grab sampling (spade and scoop) method for under asphalt or concrete or where contamination may have occurred from a given point source, designed to sample for contaminant distribution, and (4) vertical soil profile method, designed to sample for deposition increment.

2.5.1 Colorado Department of Public Health and Environment Method

The CDPHE sampler is one method for collecting soil samples for radionuclide analyses at the RFETS. The CDPHE sampler was designed to sample radionuclides in the topsoil that could become resuspended in the air and, thus, pose a migration pathway and exposure via inhalation. The sampler was designed to obtain a sample from the upper $\frac{1}{4}$ in. deep, from an area 2 in. wide and $2\frac{3}{8}$ in. long. Figure 2-1 is a drawing of the CDPHE soil sampling device.

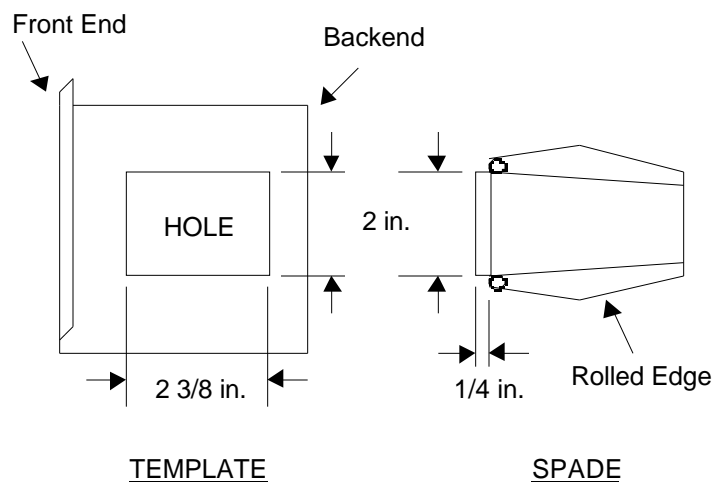


Figure 2-1. CDPHE soil sampling device. The sampler is designed to sample from the upper $\frac{1}{4}$ in. of soil.

Vegetation and any undesirable top layer of surficial material are removed. The CDPHE template is placed on the ground so that the soil surface is even with the upper surface of the sampling template. The sampling spade is placed at the backend of the template, and the front edge of the spade is placed into the soil $\frac{1}{2}$ in. deep. The sampling spade is drawn toward the front of the template to obtain the surface soil sample. The spade is again placed in the sampling hole and drawn forward to ensure that the $\frac{1}{2}$ in. soil sample has been obtained. The soil sample is then placed into a sample container.

The total number of samples and their locations are specified in the site-specific field sampling plans for each project. A specific number of samples are collected and composited from within each sample plot. The sample locations are described by an evenly spaced grid.

2.5.2 Rocky Flats Method

The Rocky Flats method is used to determine the amount of accumulated plutonium that has been deposited on the ground. This determination is accomplished by collecting a sample volume of 5000 cm^3 of soil in situ. The Rocky Flats jig outlines a square area with 10-cm sides and is driven 5 cm into the soil to cut three sides of the sampler (see Figure 2-2). At the fourth side, soil is removed from outside the jig's perimeter. The scoop is used to finish the cut on the fourth side (open face) of the sample and the bottom surface.

Ten samples are collected at each location and composited. These 10 samples are collected at the center and corners of two 1-m squares that are spaced 1 m apart. Figure 2-3 illustrates this sample collection spacing. The soil samples are passed through a 10-mesh metal sieve to remove large particles (such as cobbles and stone) that do not pass through the sieve. After sieving, the 10 soil samples are composited, mixed, and quartered to obtain a sample for laboratory analysis.

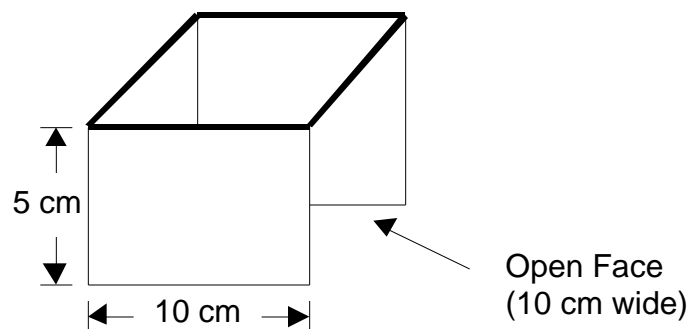


Figure 2-2. Rocky Flats soil sampling device. The device is designed to sample a 5 cm depth.

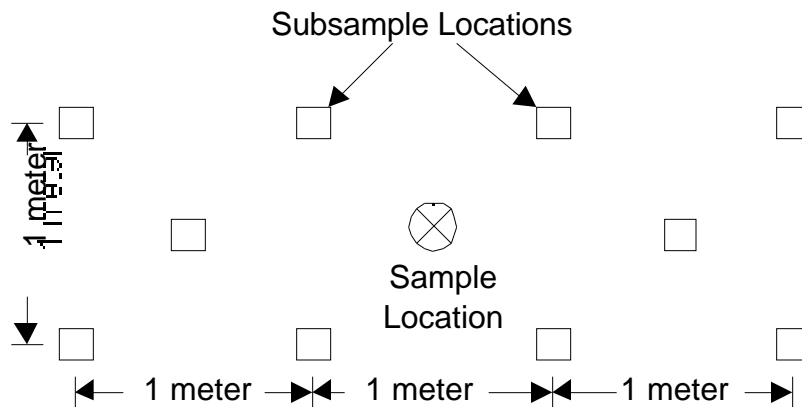


Figure 2-3. Rocky Flats soil sampling location spacing.

2.5.3 Grab Sampling Method (Spade and Scoop)

Surface soil samples can be collected for radionuclide analyses using grab sampling (spade and scoop) methods at RFETS. The vegetation and any undesired surficial material are removed from the area to be sampled. The soil sample is then collected to the desired depth using a stainless steel spoon or scoop.

The total number of samples and sample locations are specified in the site-specific field sampling plans.

2.5.4 Vertical Soil Profile Method

The vertical soil profile sampling at the RFETS defines the distribution of radionuclides in the top 6 in. of soil to verify the results of the HPGe surveys. This sampling is accomplished by collecting discrete soil samples at 2-in. intervals corresponding to depths from 0–2 in., 2–4 in., and 4–6 in. Sampling of 2 in. is required to define the extent of radiological contamination within discrete layers of the surface soil. Four RFETS procedures are used to obtain these samples: (1) collection from the surface downward, (2) collection from the side wall of a small excavation, (3) collection by coring, and (4) collection from beneath concrete and asphalt pavement. The total number of samples to be collected and their locations are specified in site-specific field sampling plans. A sample of approximately 500 g is obtained for each soil profile interval.

Soil sampling is required for in situ gamma spectroscopy surveys using HPGe detectors. In general, the depth distribution needs to be investigated to ensure that the correct parameters are used in the conversion from instrument count rate to soil concentration. As an example (from NUREG-1506, *Measurement Methods for Radiological Surveys in Support*

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of *New Decommissioning Criteria, Draft Report for Comment* [NRC 1995a]), for undisturbed soils a negative exponential profile with depth has frequently been found to be an adequate model for deposited radionuclides, that is

$$S = S_0 e^{-z/\lambda} \quad (2-1)$$

where S is the activity per unit volume of soil (pCi cm^{-3}) at depth z (cm), S_0 is the activity per unit volume at the soil surface (pCi cm^{-3}), λ is the reciprocal of the relaxation length of the exponential distribution (cm^{-1}), and ρ is the bulk density (g cm^{-3}). This expresses the profile in terms of the soil mass per unit area, ρz (g cm^{-2}), with the degree of penetration into the soil represented by the depth parameter $\rho z / \lambda$ ($\text{cm}^2 \text{g}^{-1}$). This type of profile has the maximum concentration at the soil surface (S_0) and decreases with depth. If the value of $\rho z / \lambda$ approaches infinity, the source distribution approaches a plane atop the ground, and if $\rho z / \lambda$ equals 0, the source distribution is uniform with depth. As an example, assume a soil density of 1.5 g cm^{-3} and an $\rho z / \lambda$ value of $0.2 \text{ cm}^2 \text{g}^{-1}$ (which is a typical value for an aged fallout deposit). The corresponding relaxation depth for the exponential profile would be 3.33 cm, meaning that the concentration would be reduced to $1/e$ or 37% of the surface value at this depth. For in situ measurements, the value of $\rho z / \lambda$ can be determined from the analysis of soil samples from different depth increments. The fraction of the total activity below a given depth (log value) can be plotted versus the mass depth, ρz . The slope of the line is then the value of ρ / λ .

2.6 Summary of RFETS Soil Sampling Program Review

RAC conducted a review of the current sampling program and procedures used at the RFETS to determine elements of the current program available for use and incorporation into the final status soil sampling protocol. RAC identified several procedures currently used at the RFETS that can be used to develop a soil sampling protocol. Overall, the administrative and field procedures were considered to be technically sound for their intended purpose and based on standard industry guidelines.

Field procedures have been developed for use at the RFETS that provide field personnel with the required documentation for performing assigned field duties and studies. Standard procedures are in place that document the steps necessary to containerize, label, and ship samples to the laboratory. In addition, the procedures address such issues as equipment decontamination, sample location determination using standard survey and global positioning systems, and documentation of the survey process, including sample chain of custody. These procedures are considered to be technically sound and acceptable for use in a soil sampling protocol for the soil action levels.

Four protocols for surface soil sample collection used at Rocky Flats are discussed in Section 2.5. RAC does not consider these collection protocols adequate for ensuring the collection of representative samples for comparison to the soil action levels. Therefore, this report places a major emphasis on recommendations for collecting representative samples for comparison to the soil action levels.

3. SAMPLING DESIGN DEVELOPMENT PROCESS

This section discusses the process to develop soil sampling studies and associated protocols. The MARSSIM recommends the use of the DQO process for the planning, development, and implementation of radiological surveys (NRC 1997).

3.1 Data Quality Objective Process

The process of planning a soil sampling study, implementing the sampling plan, and assessing the sampling results before making a decision is called the data life cycle. Soil sample survey planning uses the DQO process to ensure that the sampling results are of sufficient quality and quantity to support the final decision.

3.1.1 Data Quality Objective Process Description

The DQO process is described in detail in EPA (1994a) and NRC (1997). The DQO process consists of seven steps as shown in Figure 3-1.

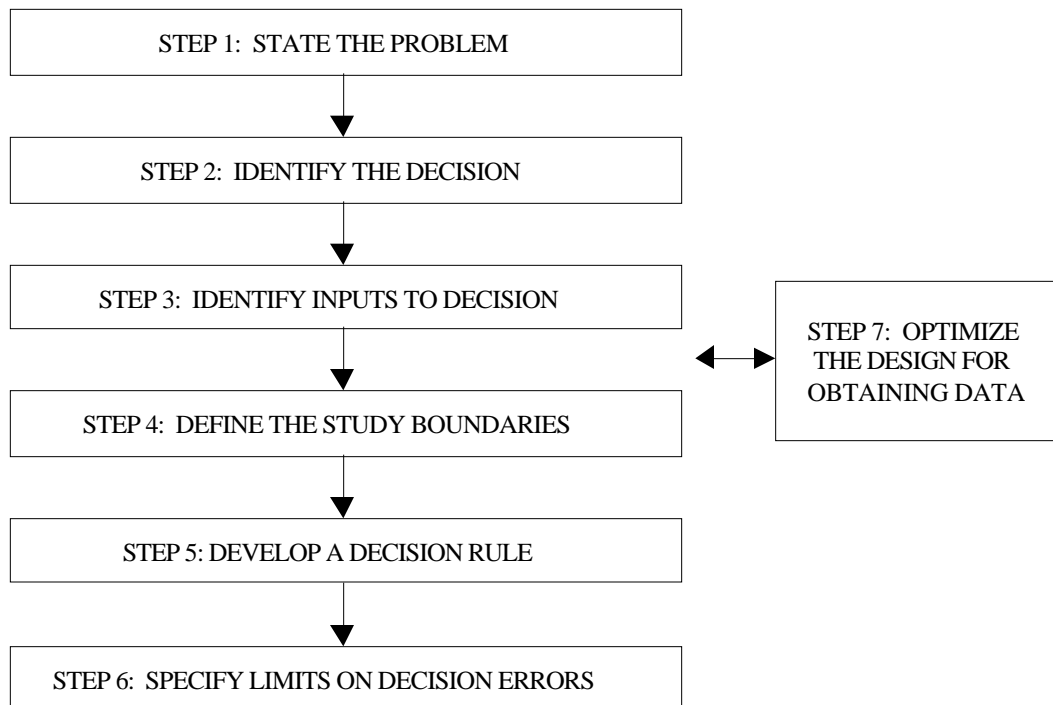


Figure 3-1. The data quality objectives process (from EPA 1994a).

Although the DQO process appears to be linear, the actual process is iterative. During decisions in the process, DQOs in previous steps may need to be reconsidered or redefined.

This iteration is encouraged because it ultimately leads to a more efficient survey design (NRC 1997).

Planning radiological surveys using the DQO process can improve the survey effectiveness and efficiency and, thereby, the defensibility of decisions. It also can minimize data collection expenditures by eliminating unnecessary, duplicative, or overly precise data. The use of the DQO process assures that the type, quantity, and quality of environmental data used in decision-making will be appropriate for the intended application. It provides systematic procedures for defining the criteria that the survey design should satisfy, including when and where to perform measurements, the level of decision errors for the survey, and how many measurements to perform (NRC 1997).

The following discussion provides a brief introduction to the DQO process. The reader is referred to the MARSSIM (NRC 1997) for a full discussion on the DQO process and its use in survey planning.

Step 1. State the Problem

The first step in any decision-making process is to define the problem so that the focus of the survey will be unambiguous. Four activities are associated with this step:

- Identifying members of the planning team and stakeholders
- Identifying the primary decision maker or decision-making method
- Developing a concise description of the problem
- Specifying available resources and relevant deadlines for the study.

The expected outputs of this step are

- A list of the planning team members and identification of the decision-maker
- A concise description of the problem
- A summary of available resources and relevant deadlines for the survey.

The planning team clarifies and defines the DQOs for a site-specific survey. This multidisciplinary team of technical experts offers the greatest potential for solving problems when identifying important aspects of a survey. Including representatives from stakeholder groups is an important consideration when assembling this team. Once formed, the team can also consider the role of public participation for this assessment and the possible survey to follow.

A concise description of the problem must be specified during this step. A description of the problem would typically involve the release of all or some portion of the RFETS that demonstrates compliance with the soil action levels. The resources and deadlines for the surveys would need to be addressed by DOE and their site contractor, Kaiser-Hill Company.

Step 2. Identify the Decision

This step defines the question that the survey will attempt to resolve and identify alternative actions that may be taken based on the outcome of the survey. The combination of these two elements is called the decision statement.

Four activities are associated with this step in the DQO process:

- Identifying the principal study question

- Defining the alternative actions that could result from resolution of the principal study question
- Combining the principal study question and the alternative actions into a decision statement
- Organizing multiple decisions.

The expected output from this step is a decision statement that links the principal study question to possible solutions to the problem.

Step 3. Identify the Inputs to the Decision

This step of the DQO process determines and specifies the information needed for the study and data to support a decision concerning the survey unit. Collecting data or information is necessary to resolve the decision statement. For the final status survey, the list of information inputs will involve measurements of the radioactive contaminants of concern in the soil.

The activities included in this step of the DQO process include

- Identifying the information required to resolve the decision statement
- Determining the sources for each item of information
- Confirming that appropriate measurement methods exist to provide the necessary data.

Step 4. Define the Boundaries of the Study

During this step, the DQO planning team develops a conceptual model of the site based on existing information. The conceptual model for the soil action levels has been developed and documented by RAC in the Task 2, 3, and 5 reports. The conceptual model is used by the DQO planning team to define the spatial and temporal boundaries of the sampling study. It is very important that the conceptual model and assumptions used to develop the soil action levels are considered during the DQO process.

During this step, the spatial boundaries that will be covered by the decision statement are defined. These considerations include spatial boundaries

- That define the physical area under consideration for release
- That define the physical area to be studied and locations where measurements could be performed
- Developed from modeling used to calculate the soil action levels.

For the final status survey, the smallest, most appropriate subsets of the site for which decisions will be made are defined as survey units (see Section 4.6.4).

Step 5. Develop a Decision Rule

This step defines the statistical parameter of interest, specifies the soil action levels, and integrates previous DQO outputs into a single statement that describes a logical basis for choosing among alternative actions (NRC 1997).

Three activities are associated with this step:

- Specifying the statistical parameter that characterizes the parameter of interest

- Specifying the soil action levels for the study
- Combining the outputs of the previous DQO steps into an “if, then” decision rule that defines the conditions that would cause the decision-maker to choose among alternative actions.

The expected outputs of this step are

- Specification of the parameter of interest that characterizes the level of residual radioactivity
- Specifications of the soil action levels
- An “if, then” statement that defines the conditions that would cause the decision maker to choose among alternative actions.

The parameter of interest is a descriptive measure (such as a mean or median) that specifies the characteristic or attribute of the residual contamination in the survey unit. The specification of the parameter of interest that characterizes the level of residual radioactivity is very important for the soil action level project. The technical basis and recommendation for the statistical parameter of interest is discussed in detail in Section 4.3.

Step 6. Specify Limits on Decision Errors

The following steps are involved in setting acceptable probabilities for decision errors (EPA 1992):

- Defining false positive and false negative errors for the decision and describing the consequences of each type of error
- Evaluating these consequences according to the relative level of concern they would cause, with emphasis on the environment, public health, economics, and social consequences
- Determining if false positive or false negative errors are of greater concern
- Establishing, with the assistance of a statistician, an acceptable probability for the occurrence of each of these errors
- Combining the probability statement into a formal statement of the levels of uncertainty that can be tolerated in the results
- Reviewing the decision rule.

During this step of the DQO process, the null and alternative hypothesis must be chosen along with acceptable probabilities of decision errors. Hypotheses and decision errors are address in Sections 4.3.4 and 4.3.5 of this report.

Step 7. Optimize the Design for Obtaining Data

This step is designed to produce the most resource-effective survey design that is expected to meet the DQOs. It will be necessary for the DQO planning team to work through this step more than once after revisiting previous steps in the DQO process.

Six activities are included in this step:

- Reviewing the DQO outputs and existing data to ensure they are internally consistent
- Developing general data collection design alternatives

- Formulating the mathematical expressions needed to solve the design problem for each data collection design alternative
- Selecting the optimal design that satisfies the DQOs for each data collection design alternative. If the recommended design will not meet the limits on decision errors within the budget or other constraints, then the planning team will need to relax one or more constraints, such as
 - Increasing the budget for sampling and analysis
 - Increasing the decision error rates
 - Changing the boundaries
 - Evaluating alternative measurement techniques with lower detection limits or lower survey costs
 - Considering the use of passive controls when releasing a survey unit rather than unrestricted release
- Selecting the most resource-effective survey design that satisfies all the DQOs
- Documenting the operational details and theoretical assumptions of the selected design in the QAPjP, the field sampling plan, and sampling and analysis plan.

Section 4.2 presents recommendations on the sampling design and mathematical expressions.

3.1.2 Recommendations

- *RAC* recommends that Kaiser-Hill Company, current operating contractor of the RFETS for DOE, use the DQO process as identified in their sampling program and QAPjP to develop the final status soil sampling protocol.
- *RAC* recommends that DOE appoint representatives from the RSALOP for inclusion on the Kaiser-Hill Company DQO planning team. This is an important recommendation to ensure that stakeholder concerns are addressed in developing the sampling protocol and an acceptable sampling protocol for the final status survey is developed. The appointment of RSALOP representatives on the DQO planning team will also ensure that Kaiser-Hill Company has the input of representatives that were directly involved in developing the soil action levels. The inclusion of RSALOP representatives on the DQO planning team is necessary to ensure that decisions that may result in deviations from the recommendations provided in this report include the technical input and acceptance of stakeholders and the general public, represented by the RSALOP.

3.2 Soil Action Levels

3.2.1 Development Method

RAC is conducting the pathway modeling for the radionuclide soil action levels considering parametric uncertainties. In other words, uncertainty in the values of specific model parameters are considered by assigning a probability distribution to each parameter that is treated as uncertain. Parametric uncertainty is concerned with propagating uncertainty in parameter values through the simulations to the resulting estimates of concentrations in

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exposure media or to dose, typically using Monte Carlo techniques. The procedure produces an uncertainty distribution for each soil action level.

As discussed in the Task 2 report, the soil action levels must be combined with measured soil concentrations of the respective radionuclides, forming ratios (soil concentration divided by soil action level). If the concentration of radionuclide i is denoted by C_i , and its radionuclide soil action level is denoted by SAL_i , then the sum of ratio rule for n radionuclides states that

$$\frac{C_1}{SAL_1} + \frac{C_2}{SAL_2} + \frac{C_3}{SAL_3} + \dots + \frac{C_n}{SAL_n} = 1 \quad (3-1)$$

There are two methods for evaluating the sum of ratios quantity: (1) deterministically and (2) stochastically. In the deterministic case, a discrete soil action level is chosen by the RSALOP for each radionuclide, then the ratios of the soil sample data and the soil action levels are summed in the deterministic case. The sum must be less than or equal to 1 to meet the dose standard (i.e., release standard of 15 or 85 mrem y^{-1}). In the stochastic case, RAC has calculated the probability of exceeding the dose limit as a function of the ^{239}Pu soil concentration (Figure 3-2). The doses calculated in the stochastic case include not only the dose from ^{239}Pu , but all other plutonium isotopes and americium. The activity levels of the other plutonium isotopes and americium are estimated using site-specific isotopic ratios described in the Task 5 report (Killough et al. 1999). For example, using Figure 3-2 and a ^{239}Pu soil concentration of 100 pCi g^{-1} , the probability of exceeding the 15 mrem dose limit is about 0.7 or about 70%. We note here that action levels for uranium isotopes have been reported separately because ratios of uranium to plutonium are not consistent across the site. In addition, uranium contamination does not appear to be as widespread as that of plutonium and is mostly restricted to source areas such as old disposal areas and burn pits. (see Section 4.1.3 for further discussion on the distribution of uranium in soil). Soil action levels for ^{238}U are reported in the same manner as those for plutonium isotopes, but they only include doses from the other uranium isotopes and do not include doses from plutonium isotopes. Specifying an action level for a site with both uranium and plutonium contamination will need to be considered case by case.

The RAC Task 2 report also provides a discussion about remedial strategies (see Section 3.1.2 and equations in Section 2.1). The Task 2 report notes that programs, such as RESRAD, proceed on the assumption of a uniformly contaminated area (subject to variation within a factor of 3). For some scenarios it could be desirable to subdivide the site area into some number P of plots, each of which can be treated as having a uniform concentration of each radionuclide but with concentrations varying from one plot to another. Such subdivision might be of assistance in the planning for remediation because the effects of reducing the most contaminated plots by various amounts can be studied explicitly. DOE and the Kaiser-Hill Company team are encouraged to consider the use of the subdivision method for evaluating remedial strategies.

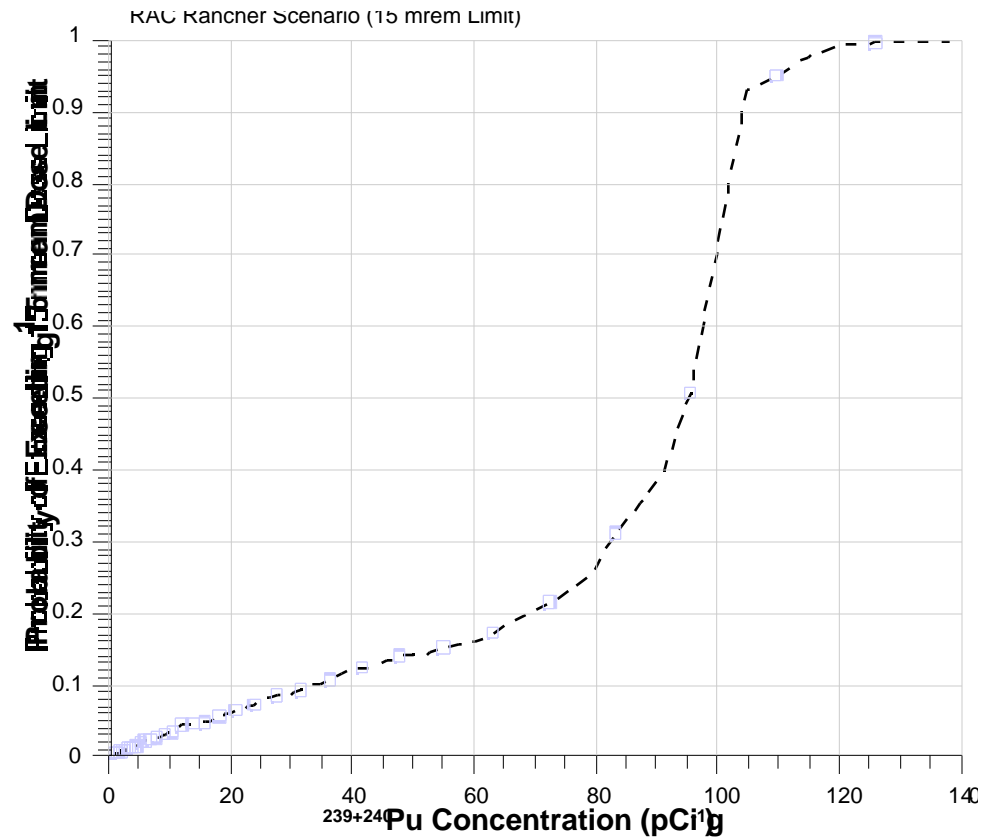


Figure 3-2. Example of a stochastic soil action level that shows the probability of exceeding the 15 mrem dose limit as a function of $^{239+240}\text{Pu}$ concentration in soil. Doses include all plutonium isotopes and americium. Activity levels of the other plutonium isotopes and americium are based on site-specific isotopic ratios.

3.2.2 Recommendations

- *RAC* recommends that the RSALOP select discrete values for the soil action levels of each radionuclide for use in the final status survey. Selecting discrete values from the soil action level distributions provides the mechanism for the RSALOP to determine the level of risk considered acceptable for use in the soil action level project.
- *RAC* has provided a potential method for evaluating remedial strategies in the Task 2 report, as described above. The RSALOP, DOE, and the site contractor (Kaiser-Hill Company) are encouraged to consider the use of these methods for evaluating remedial strategies.

4. SAMPLING PROTOCOL RECOMMENDATIONS

The discussion in Section 3 provides the reader with an understanding of the process required for developing a sampling design and explains the iterative nature of the process. The following discussion provides the recommendations considered by *RAC* to be essential to ensuring that representative surface soil samples are collected for comparison to the soil action levels.

4.1 Soil Sample Collection Protocol

The soil sample collection protocol must ensure that representative samples are collected. A representative sample is a sample collected to appropriately reflect the media and contamination being measured. The samples must also be collected such that the resulting data can be compared to the modeled soil action levels. This requires that the media be sampled in a manner that is consistent with the conceptual model and associated assumptions used in developing the soil action levels. For example, resuspension models are based on contamination contained in the upper soil surface. Sample collection protocols that collect a continuous soil sample from the surface to several centimeters in depth may not be representative of the resuspension model used to derive the soil action levels.

Several soil sample collection protocols (see Section 2.5) are currently used in studies at the RFETS. Several studies on the nature and extent of radionuclide contamination at the RFETS have used either the CDPHE or the Rocky Flats sampling methods, and in some instances, both methods were employed. These two methods, involving different sample collection depths, have created problems for researchers attempting to compare data sets. In addition to these methods, Colorado State University has developed a soil sample collection protocol for use in their radionuclide studies at the RFETS (Webb et al. 1997; Webb 1992), which involves collecting soil samples in 3-cm increments (see Section 4.1.2).

4.1.1 Protocol Comparison Studies

Bernhardt et al. (1983) documents a field soil sampling study conducted around the RFETS in May 1977. The cooperative plutonium soil sampling project was conducted by Rockwell International (contractor at the time for the DOE RFETS); CDPHE; Jefferson County Department of Health; and the EPA Office of Radiation Programs, Las Vegas. Each of the agencies collected five replicate samples from four distinctly different pedological and morphological settings around the RFETS. The following soil sampling methods were used in the study: Rocky Flats method (100 cm² area), CDPHE method (750 cm² area), Jefferson County technique (sizing of dust swept from a 4 m² area of the ground surface), and two EPA methods for samples of 0 to 1 cm (450 cm²) depth and 0 to 5 cm (500 cm²) depth.

Bernhardt et al. (1983) found statistically significant differences in average concentration between the sampling techniques. They found that for concentrations expressed on a per gram basis, those sampling methods that sample to greater depths tend to have lower average concentrations. This was attributed to the residence of most of the plutonium in the surface 1 or 2 cm, and sampling to a greater depth resulted in dilution of the

surface soil plutonium with soil from a greater depth that had a lower concentration of plutonium.

Bernhardt et al. (1983) noted that the differences in concentrations observed in the study emphasize the importance of matching sampling objectives with sampling techniques. If the objective is to assess the amount of the total deposited material, samples should be taken to a depth sufficient to collect all the material. If the objective is to assess the health significance of deposited plutonium or to obtain estimates of potential resuspension, samples of only the surface soil should be taken.

Litaor et al. (1995) used both the CDPHE and Rocky Flats methods for sampling of $^{239,240}\text{Pu}$ in the Rocky Flats environment. This study noted that because the CDPHE sampler collects only the top 0.64 cm of the soil, the sampler exhibited a serious problem in locating the boundary between the soil surface and the plant litter layer accumulated above. The study concluded that there was no significant difference in $^{239,240}\text{Pu}$ activity in soil collected with the CDPHE sampler versus the Rocky Flats sampler for use in developing surface contour maps of estimated plutonium concentrations. Twenty-five samples were composited for the CDPHE method, with 10 samples composited for the Rocky Flats method using survey unit sizes of 1.01 ha near the 903 Pad and 4.05 ha further from the 903 Pad. The Rocky Flats sampling design consisted of taking five subsamples collected from the corners and the center of two 1-m squares, which were spaced 1 m apart in the middle of each survey unit. The CDPHE sampling design required 25 equally spaced subsamples to be composited within each survey unit.

The Litaor et al. (1995) study sampling protocols required composited samples, which introduced an additional source of uncertainty in the results because of the sample homogenization process. Composite sampling generally provides a very good estimate of the mean. However, information for the variance of the concentration needed for the final status surveys was not provided. Composited samples are also unable to detect individual areas of elevated activity (see Section 4.2 for a discussion on sampling design). The sampling methods used in the Litaor et al. (1995) study also required different numbers of samples from completely different soil horizons. Nonetheless, the two sampling methods may be comparable for the purposes of the Litaor et al. (1995) study. However, the two methods are not considered equivalent for the purpose of determining if the soil action levels have been attained at the site.

4.1.2 Depth Distribution of Plutonium and Americium

The importance of sampling depth for the sample collection protocol was identified by Bernhardt et al. (1983) as described in Section 4.1.1. The following summary of studies at the RFETS for plutonium and americium provides insight into the depth distribution of these radionuclides.

Webb et al. (1997) conducted a study using a sampling protocol developed at Colorado State University, with additional surface samples collected using the CDPHE method. This study selected thirteen 100-m² macroplots, which were spaced at exponentially increasing distances from the 903 Pad along each of four transects. Samples were replicated at four randomly located, 1-m² microplots within each macroplot. The general sampling procedure

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was to (1) clip the standing vegetation at ground level inside a 1250 cm² frame, then scrape ~3 mm of surface soil using the CDPHE scoop and template method and (2) clip the standing vegetation at ground level inside a 625 cm² frame, then excavate a 25 cm long _ 10 cm wide area of soil in 3-cm layers to a depth of 21 cm using a trench technique developed earlier in Little (1976) and later modified in Webb (1992).

Webb et al. (1997) investigated the depth distribution of ²³⁹Pu in the soil at Rocky Flats (Figure 4-1). To develop the generalized depth distribution for the study area, ²³⁹Pu concentrations in each layer (including the 0–3-mm scrape) were normalized to the 0–3-cm concentration for each soil profile. The medians of the normalized values at each depth were then fit to mathematical functions by trial and error, but the final regression parameters were determined with commercial fitting software. In the final regression, a total of 643 individual ²³⁹Pu values were used to derive the following depth relationship:

$$\left[{}^{239}\text{Pu} \right]_d = \left[{}^{239}\text{Pu} \right]_{0-3\text{cm}} \left[1 - \left(-1.41e^{-0.71d} - 0.16e^{-0.19d} \right)^4 \right] \quad (4-1)$$

where

$\left[{}^{239}\text{Pu} \right]_d$ = ²³⁹Pu concentration at depth d (cm)
 $\left[{}^{239}\text{Pu} \right]_{0-3\text{cm}}$ = ²³⁹Pu concentration (pCi g⁻¹) in 0–3 cm soil increment.

Webb et al. (1997) noted that the relatively uniform concentration of plutonium over the first 2 cm is suggestive of some ongoing natural mixing process.

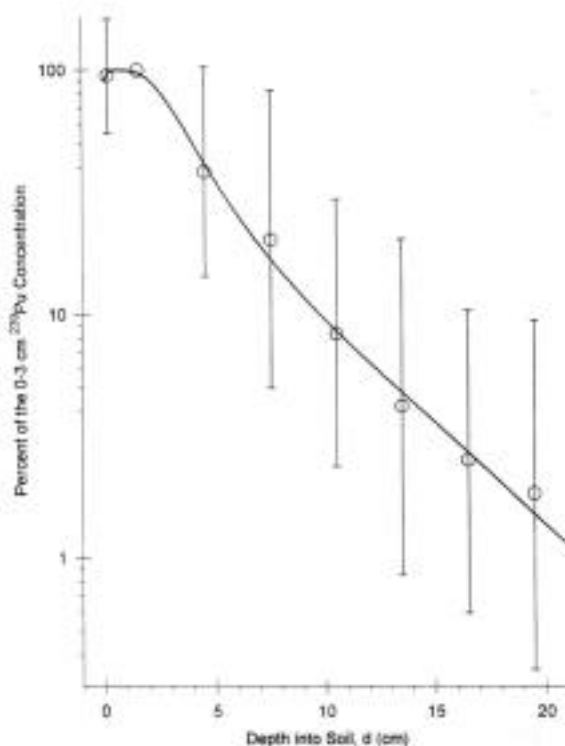


Figure 4-1. The ²³⁹Pu concentrations in soil at all depths (Webb et al. 1997).

Shierman (1994) provides information on the americium and plutonium concentration soil profiles at the Rocky Flats site. Shierman found that the concentrations of americium and plutonium decreased exponentially as a function of depth for all the locations sampled. Approximately 90% of the total inventories of the two radionuclides resided in the top 9 cm of soil, with approximately 50% of the total inventory for the two contaminants residing in the top 3 cm of soil. No difference was observed in ^{241}Am and ^{239}Pu movement vertically in the soil column.

Litaor (1999) examined the plutonium contamination in soils in open space and residential areas near Rocky Flats. Litaor included an analysis of the vertical distribution of plutonium activity from 11 soil pits collected outside RFP. The study indicated that the top layer (0–3 cm) was the most contaminated layer, with over 96% of $^{239,240}\text{Pu}$ activity accounted for in the top 12 cm of the soil. Below the 12-cm depth, plutonium activity decreased to background levels (see Section 4.4 for a discussion on background soil concentrations).

The studies discussed above indicate that the plutonium and americium appear to behave similarly in the soil. Each radionuclide is expected to be bound within the upper region of the soil profile, with an exponential decrease in activity with depth.

4.1.3 Depth Distribution of the Uranium Isotopes

Information on the depth distribution of uranium appears to be less extensive than that for plutonium and americium. RAC did not find the activity distribution of uranium with depth in the soil during a literature search. However, Litaor (1995) conducted a study of the spatial distribution of uranium isotopes in soils at the RFETS. The goal of the Litaor (1995) study was to provide information on the distribution of uranium isotopes in soils east of the RFETS. A spatial analysis of three uranium isotopes was conducted to determine the concentration and distribution pattern of uranium contamination. Geostatistical techniques were used to model the spatial dependency and construct isopleth maps showing uranium isotope distributions.

The Litaor (1995) sampling protocol required 25 equally spaced subsamples to be composited within 4.05 or 1.01-ha plots for uranium isotopes analysis. The soil at each individual location was sampled with a CDPHE sampler. Eighty-four 4.05-ha plots and thirty-four 1.01-ha plots were sampled for a total of 118 plots.

Litaor (1995) identified ^{234}U activity in soils around the RFETS that ranged from 25.9 to 92.8 Bq kg⁻¹ (0.7 to 2.5 pCi g⁻¹), with a median activity of 44.4 Bq kg⁻¹ (1.2 pCi g⁻¹). A spatial structure was not observed with the ^{234}U data. Litaor (1995) indicated that the lack of spatial structure suggested that ^{234}U was randomly distributed in the soil environment east of the RFETS. The randomness was also interpreted to reflect inherent irregular variation of ^{234}U dispersion in the soil that could not be predicted by the sampling method. As an alternative interpretation, Litaor (1995) also indicated that it may represent variability between sampling plots at distances less than that used in the study or samples collected from different populations (natural versus impacted because of Rocky Flats activity). On the basis of the available information, Litaor (1995) concluded that the contribution of Rocky Flats to the activity of ^{234}U in the soils was negligible.

Litaor (1995) identified ^{235}U activity in soils around the RFETS that ranged from 0.1 to 25.1 Bq kg⁻¹ (0.003 to 0.68 pCi g⁻¹), with a median activity of 1.8 Bq kg⁻¹ (0.05 pCi g⁻¹). The resulting ^{235}U geostatistical contour map did not indicate the same clear west-east pattern of dispersion as those identified for $^{239+240}\text{Pu}$ and ^{241}Am (Litaor et al. 1995). The activity of ^{235}U was localized, approximately 500 m east of the industrial section of the RFETS. No clear relationship between the various sources of ^{235}U contamination considered in the study (i.e., burial trenches and the 903 Pad) and the spatial pattern of the ^{235}U isopleths was found.

Litaor (1995) states that the two soil plots with the highest ^{235}U activity probably resulted from surface flow and interflow from the east spray field. It was hypothesized that the small amounts of ^{235}U in pond water were reconstituted on the soil surface of these two plots through the continuous irrigation and subsequent evapotranspiration.

Litaor (1995) identifies ^{238}U activity in soils around the RFETS that ranged from 30.7 to 286 Bq kg⁻¹ (0.83 to 7.7 pCi g⁻¹), with a median activity of 44.4 Bq kg⁻¹ (1.2 pCi g⁻¹). The ^{238}U geostatistical contour map showed a clear west-east dispersion pattern like those of $^{239+240}\text{Pu}$ and ^{241}Am (Litaor et al. 1995). The highest observed activities of ^{238}U were found around the 903 Pad; however, these values did not extend beyond the immediate vicinity of that site.

Litaor (1995) notes that the complete lack of similarity in the spatial distribution across the soilscape near the 903 Pad could be explained by fundamental differences in solubility characteristics of plutonium and uranium that, in turn, affects their mode of dispersion in the environment. Plutonium is largely insoluble in the soil environment at RFETS; hence, upon removal of the drums during the cleanup operations, the impacted area became susceptible to wind and surface erosion. Consequently, plutonium particles entrapped in the fine fraction of the topsoil were airlifted by winds and subsequently deposited across the soilscape east of the 903 Pad. In contrast, under the pH and alkalinity conditions of the soil adjacent to the 903 Pad, uranium(VI) may form complex ions with carbonates and migrate downward in the soil column. Litaor (1995) notes that Seed et al. (1971) identified four hot spots below the asphalt cap that had been placed over the entire area of the 903 Pad. They retrieved >31 kg of uranium below this cap. Most of the uranium was concentrated in the B soil horizon located 15 to 40 cm below the original ground level. Increased clay content with depth decreased the hydraulic conductivity in the soils. These flow conditions facilitated the transport of uranium through the surface horizons but greatly restricted the transport to greater depths (i.e., >1 m). Litaor (1995) notes that the mobility of uranium with depth was probably further restricted because of sorption of uranium by the sesquioxides and CaCO₃ minerals. Because of the solubility and migratory behavior of uranium in the soil system, little uranium was entrapped in the fine particles of the topsoil. Hence, wind-dispersal mechanisms did not influence the spatial distribution of uranium isotopes across the soilscape east of the 903 Pad (Litaor 1995).

The uranium study conducted by Litaor (1995) indicates that isotopes of uranium do not behave similar to plutonium and americium in the soil environment. At the present time, the development of a soil sample collection protocol must rely on the Litaor (1995) uranium study and the assumption that the uranium was leached near the source into the soil profile and not transported by resuspension. Thus, uranium is assumed to be locally concentrated near the original source areas and present throughout the surface soil profile. RAC recommends that additional studies be performed similar to the Webb (1997) study on plutonium and the Shierman (1994) study for americium to determine the depth profile of the uranium contamination in the RFETS soil environment. The study by Litaor (1995)

relied on samples collected from the upper 0.6 cm of the soil profile using the CDPHE sampling method and limited pit samples collected to a depth of 9 cm. If the uranium is mobile in the soil environment, most of the uranium activity may already reside in the soil profile below that modeled for the soil action levels (i.e., 20 cm).

4.1.4 Conceptual Model Considerations

The conceptual model and assumptions for developing soil action levels are provided in Task 3, "Input and Assumptions." Two parameters used in the RESRAD model for calculating the soil action levels relate to considering sampling depth. These parameters are the depth of the soil mixing layer and the thickness of the contaminated zone.

The depth of the soil mixing layer is the depth of surface soil available for resuspension. This value is used to calculate the depth factor, or the fraction of total resuspendible soil that is contaminated. The use of this parameter in RESRAD to calculate the depth factor requires that it represent the depth over which contamination is uniformly distributed in the resuspendible layer. In the previous soil action level calculations (DOE/EPA/CDPHE 1996), the values for soil mixing layer and thickness of the contaminated zone are equal, which is not consistent with the definition of either term. RAC selected a value of 0.03 m to maintain consistency with the definition. This value has been used in the literature to define the surface or resuspendible soils and is the value defined by Webb et al. (1997) as representative of surface soils at Rocky Flats. We note here that the resuspension model in RESRAD was bypassed in our calculations. Instead, we used a site-specific model that accounted for the spatial distribution of plutonium in soil and annual average meteorological conditions at the site. The model was calibrated to measurements of plutonium isotopes in air at samplers located in the buffer zone and along the perimeter of the site.

The thickness of the contaminated zone in RESRAD represents the vertical distance over which radionuclide contamination levels are clearly above background. The research presented in Webb et al. (1997) indicates that plutonium contamination is distributed over the top 20 cm of soil, with very little movement of that soil within the column over the past 20 years. For this reason, RAC used a deterministic value of 0.2 m (20 cm) in developing the soil action levels.

4.1.5 Dominant Exposure Pathways

Soil action levels for plutonium and uranium isotopes that are reported in the Task 5 report (Killough et al. 1999) vary depending on the scenario assumed. The most limiting scenarios (those with the lowest soil action level) were the RAC-designated scenarios of the rancher, child of the rancher, and infant of the rancher. Soil action levels for ^{239}Pu were from 80 to 110 pCi g⁻¹ for these scenarios assuming a 15 mrem dose limit and when the current vegetation was present. Under a special case where a fire removes most of the vegetative cover, soil action levels for ^{239}Pu dropped to between 10 to 25 pCi g⁻¹ mainly because resuspension was substantially higher for bare soil compared to vegetated soil. Consequently, inhalation was the dominant pathway for the fire case, while the soil ingestion was the dominant pathway for the no-fire case.

The dominant exposure pathway is an important consideration for the soil collection protocol. Because surface exposure pathways such as soil ingestion and inhalation of resuspended soil dominate the dose, the upper 3 cm of soil should be sampled. While the

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RESRAD model provides soil action levels for the entire modeled contaminated zone (i.e., 20 cm), only the surface soil concentrations (0–3 cm) influence the dose. If the soil were sampled over the entire contaminated zone of 20 cm, then dilution of the upper 3 cm soil profile may provide a soil concentration that is less than the action level for radionuclides despite the fact that the 0–3 cm concentration may exceed the action level. Therefore, based on the dominant exposure pathways being soil ingestion and inhalation of resuspended soil as reported in the Task 5 report (Killough et al. 1999), soil sampling should include the 0–3 cm layer.

4.1.6 Sample Compositing

The CDPHE and Rocky Flats soil sampling methods used at Rocky Flats are based on the concept of composite sampling from a systematic grid. Composite sampling consists of collecting several grab samples, from equally spaced intervals, that are thoroughly mixed into one composite sample. Then, either the entire composite is measured or one or more random subsamples from the composite are withdrawn and measured. If the mixing process is thorough, a physical averaging takes place so the subsamples represent the average concentration of the original grab samples.

Compositing is useful if (a) the cost of analyzing individual grab samples for contaminants is high, (b) the mixing process is thorough, (c) information on the variability or extreme concentrations for grab samples is not needed, and (d) the total amount of pollutant present in the composite is equal to or greater than any single grab sample making up the composite. Therefore, if the entire composite or large subsamples are analyzed, the pollutant may be more easily detected (Gilbert 1987). A major question about compositing soil samples is whether or not they can be adequately mixed. The basic idea is that the composite sample will provide an accurate average value for the individual samples used to make up the composite. If the entire composite is used for analysis, there should be no problem with the concept. For many transuranic analyses, however, only a relatively small mass is used; therefore, a composite itself may be subsampled (aliquoted) at the chemical-analysis stage. Whether compositing is worthwhile, then, depends on how well the sample is, or can be, mixed. The hot-particle problem in plutonium analysis, suggests that compositing may not be very effective (Eberhart and Gilbert 1980).

Composite sampling presents a problem for the radionuclide soil action level soil sampling protocol. Composite samples do not indicate the variance or information on extreme sample values. Therefore, small areas of elevated contaminants would not be identified by this method. Additionally, composite samples only provide an estimate of the mean soil concentration; therefore, the data do not allow a comparison to other percentiles of a distribution. For example, the mean concentration for each radionuclide may be within the action levels; however, no information is available to determine the fraction of the contaminant distribution that is above the action levels and potential hot spots are not identified.

4.1.7 Recommendations

- The recommended sample collection protocol for use in the soil action level project involves using profile sampling. The sample depth protocol recommendation assumes that soil ingestion and inhalation of resuspended contaminated soil are the dominant exposure pathways for the RFETS. The use of a profile sampling methods allows for

assessing the surface soil layer for comparison to the radionuclide soil action levels for these dominant exposure pathways. In addition, information from all soil profile layers may be combined to provide average soil concentrations if additional pathways are determined to be important. The profile method also provides valuable information that may apply to future actinide migration studies. Using the profile sampling method in areas that have been remediated also provides information for evaluating the effectiveness of the remedial process and could be used to evaluate surrogate radionuclide ratios as described in Section 4.8.

The profile sampling method provides valuable information to determine if the sampling method is required for other remediated sites. For example, the profile data from a particular remedial area may indicate that the remediation process results in a mixed residual contamination zone in the soil so that the profile method is no longer required. In this instance, sampling for remediated areas may revert to sampling the entire upper 20 cm of soil.

- *RAC* recommends that profile sampling be conducted in soil depth intervals of 0–3-cm to be consistent with the resuspension model parameters used to develop the soil action levels.
- *RAC* recommends that soil samples not be composited for the soil action levels study. Compositing soil samples eliminates information necessary to determine if small areas of elevated contamination are present in the survey unit. Therefore, the individual soil samples should be analyzed for their radionuclide contaminants.

4.2 Sampling Design Selection

Several sampling designs are available that could be applied to the sampling protocol. These methods include random sampling, stratified random sampling, and systematic sampling using either composited samples or individual samples. These methods are discussed in detail in standard statistical texts, such as Gilbert (1987), and in EPA guidance (EPA 1991). The aim of sampling is usually to characterize a defined area, to identify unsuspected hot spots, and to demonstrate, as far as practicable, that those parts of the site believed to be less than the soil action levels are in fact less than the action levels. Sampling designs are summarized in the following sections.

4.2.1 Random Sampling

Simple random sampling is the arbitrary collection of samples within defined boundaries of the survey unit. Random sample locations are chosen using a random selection process. The arbitrary selection of sampling points requires each sampling point to be selected independent of the location of all other points, and results in all locations within the area of concern have an equal chance of being selected. Randomization is necessary to make probability or confidence statements about the sampling results. Random sampling tends to

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produce uneven sampling, such that large areas may not contain a sampling point. Thus, random sampling does not ensure adequate coverage of the unit to be sampled and also is not efficient in identifying hot spot areas. Figure 4-2 provides an example of a simple random sampling pattern.

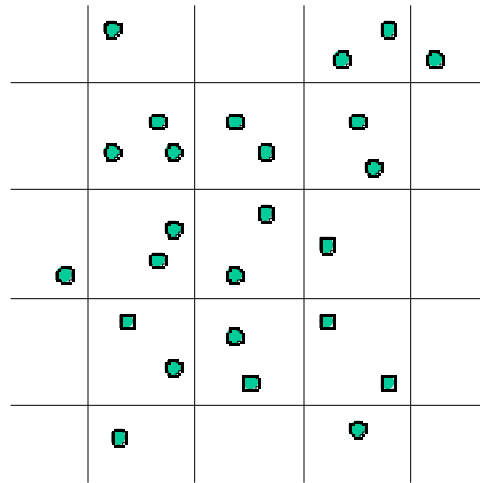


Figure 4-2. Example of a simple random sampling pattern with 25 sampling points.

4.2.2. Stratified Random Sampling

Stratified random sampling relies on prior analytical data to divide the sampling unit into smaller areas called strata. Each sampling strata is more homogeneous than the sample as a whole. Strata can be defined based on various factors, including sampling depth, contaminant concentration levels, and contaminant source areas. Stratified random sampling is a useful and flexible design for estimating the pollutant concentration within each depth interval or area of concern. The use of a stratified design for the area of concern is not needed because the use of the survey unit concept (Section 4.6) delineates the contamination area so that heterogeneity is controlled. Figure 4-3 provides an example of a stratified random sampling pattern.

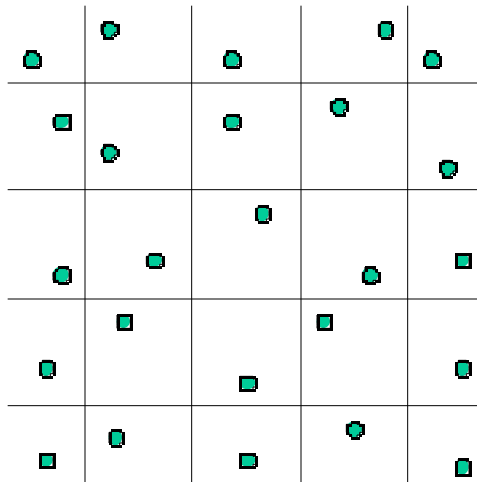


Figure 4-3. Example of a stratified random sampling pattern—1 sampling point per stratum (point located randomly within each stratum) with 12 sample points.

4.2.3 Systematic Sampling

Systematic grid sampling involves subdividing the area of concern by using a square or triangular grid and collecting samples from the nodes (intersections of the grid lines). The origin and direction for placing the grid is selected using an initial random point. From that point, the coordinate axis and grid are constructed over the whole survey unit. The distance between sampling locations in the systematic grid is determined by the size of the area to be sampled and the number of samples to be collected.

The recommended method of sampling for comparison to the soil action levels involves a systematic grid, with a random starting point as described in NRC (1997). This sampling process involves determining a uniform grid pattern over the survey unit. This sampling scheme is recommended to detect small areas of elevated contamination (i.e., hot spots—see discussion in Section 4.7), and the random starting point of the grid provides an unbiased method of obtaining measurement locations to be used in statistical tests.

By using a systematic grid, the size of a small area of elevated contamination (i.e., hot spots) that could potentially be missed by soil sampling can be easily calculated. Section 4.7 provides further discussion on identifying small areas of elevated activity. Systematic grid sampling, with a random starting point, is the final status survey method for soil sampling recommended by the MARSSIM. Therefore, use of this grid system would allow for the MARSSIM approach to be applied to the radionuclide soil action level study. However, using this grid does not confine the analyst to the use of the MARSSIM statistical analyses; other statistical techniques may be used to assess the soil action levels based upon the data obtained by this method.

Systematic sampling is usually easier to implement under field conditions than are simple random or stratified random sampling plans. Systematic sampling provides for a uniform

coverage of the target population that, in many cases, will yield more accurate estimates of mean concentrations. However, if the process being measured follows unsuspected periodicities over time and/or space, systematic sampling can give misleading and biased estimates of the population mean. Another problem with systematic sampling is the difficulty of obtaining an accurate estimate of the sampling error of the estimated mean unless the population is in random order (Gilbert 1987). A further discussion on spatially correlated data is provided in Section 4.3.3.

Gilbert (1987) provides a discussion on systematic grid sampling and the design of sampling grids. Gilbert (1987) provides methods for designing the sampling grids of aligned square grids, central aligned square grids, unaligned grid patterns, and the triangular systematic grid. These systematic sampling grid patterns are shown in Figure 4-4.

The simplest systematic designs for sampling an area are the aligned and central aligned square grids. To determine the population units to be sampled for the aligned grid, first the distance between grid lines is chosen. Then two random coordinate numbers are drawn to fix the location of point A. The remaining grid points are then fixed by the prespecified grid spacing. Deliberately placing point A at the center of the square results in the central aligned square grid (Gilbert 1987).

The unaligned grid pattern can be used to guard against bias in the estimated mean because of unsuspected periodicities over space. Gilbert (1987) notes that studies documented in Quenouille (1949) and Das (1950) suggest that this design is superior to both the square grid and stratified random sampling at least for some simple spatial correlation functions. The importance of spatial correlation is discussed in Section 4.3.

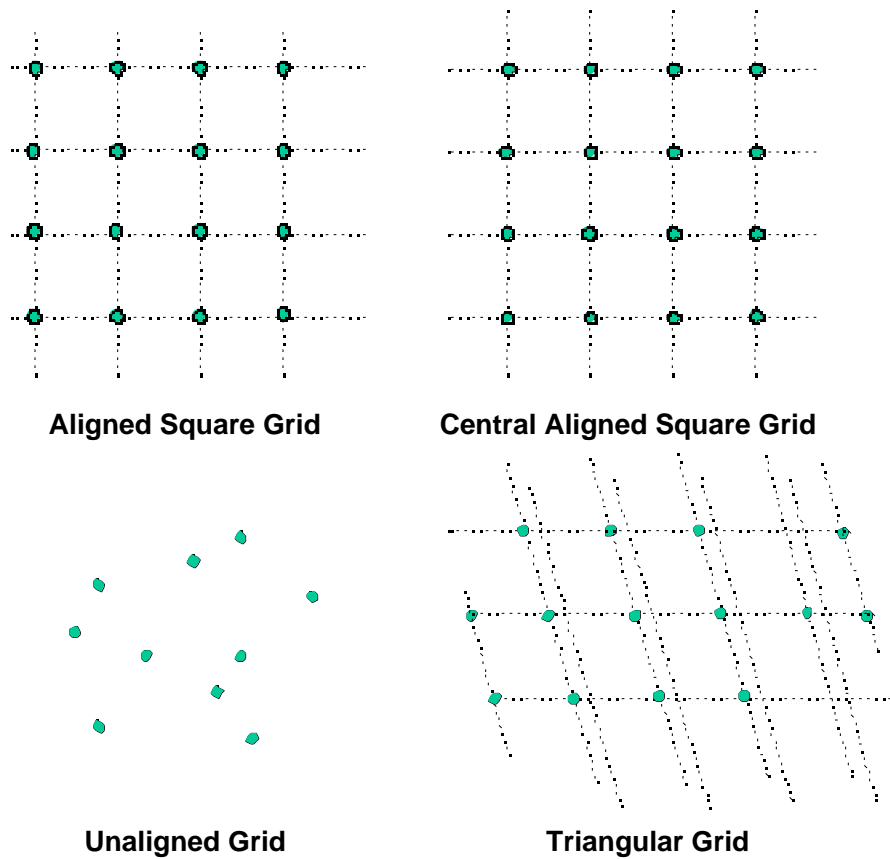


Figure 4-4. Systematic designs for sampling in space (Gilbert 1987).

The sampling locations for the unaligned grid are determined first by selecting a point A at random. The x-coordinate at point A is then used with three new random y-coordinates to determine points B, C, and D. The y-coordinate of A is then used with two new random x-coordinates to locate points E and I. The x-coordinate of E and the y-coordinate of B are then used as the coordinates of point F. Similarly, the x- and y-coordinates of E and C, respectively, are the x- and y-coordinates of point G, and so on, for the remaining cells.

The triangular grid system is a variation on the aligned square grid. After a random point A is chosen, the other sampling points are fixed by the imposed triangular arrangement. Gilbert (1987) notes that studies documented in McBratney et al. (1981) suggest the triangular grid is slightly superior to the square grid if the spatial correlation structure varies with direction.

The number of samples (n) required for the assessment (discussed in Section 4.5) is used to determine the spacing, L, of a systematic grid pattern by

$$L = \sqrt{\frac{A}{0.866 n}} \quad \text{for a triangular grid} \quad (4-2)$$

$$L = \sqrt{\frac{A}{n}} \quad \text{for a square grid} \quad (4-3)$$

where A is the area of the survey unit.

4.2.4 Recommendations

- RAC recommends that a systematic grid sampling design with a random starting point be implemented for the final status survey. A systematic grid is recommended in NRC (1997) for use in the final status survey. In addition, this sampling design ensures full coverage of the survey unit, whereas a random design can result in very uneven sampling and large areas of the survey unit may not be sampled. The choice of systematic grid designs should be evaluated during the DQO process to determine if spatial correlations exist in the survey area. When spatial correlations exist, triangular or unaligned grid systems should be considered to reduce bias in the estimate of the mean.

4.3 STATISTICAL CONCEPTS

This section discusses statistical methods and concepts for use in the soil action levels project. Statistical analyses (or tests) are typically classified into two major categories: (1) parametric statistics and (2) non-parametric statistics. These categories of statistical analyses are described in the following sections. Specific emphasis is placed on problems that have been identified by the Radiation Advisory Committee of the Science Advisory Board (SAB 1997) with the NRC (1997) statistical approach and selecting a statistical parameter for comparison to the soil action levels.

4.3.1 Descriptive Measures for Comparison to the Soil Action Levels

The parameter of interest is a descriptive measure that specifies the characteristic or attribute of the residual contamination in the survey unit. The following descriptive measures can be used for comparison to the soil action levels:

- Maximum observed concentration
- 95 percentile (i.e., the value below which 95% of actual values lie) or some other percentile
- Median of the observed concentrations
- Mean of the observed concentrations
- 95% upper confidence limit of the arithmetic mean.

The use of the maximum observed concentration or the 95 percentile for comparison to the soil action levels is not considered necessary. These descriptive measures are typically used when limited data are available and the sample size is small.

The median as a descriptive measure for comparison to the soil action levels is also not considered an appropriate parameter. The MARSSIM (NRC 1997) recommends non-parametric tests that are based on the median of the sample data. The median is equal to the

mean when the distribution is symmetrical (i.e., not skewed). However, environmental data, such as soil samples, often present data distributions that are skewed to higher concentration values; thus, the median is less than the mean.

The use of the mean as a descriptive measure for comparison to the soil action levels cannot be justified because it is not possible from any site investigation to know the true mean, which is the appropriate parameter.

SAB (1997) notes that the target statistic for any exposure assessment should be the arithmetic mean concentration for a defined area, together with the uncertainty associated with the estimate of the mean. SAB (1997) also states that “model assumptions that use a uniformly distributed source term do so as a surrogate for the arithmetic mean of a heterogeneously distributed contaminant. If one hypothetically homogenized a heterogeneously contaminated area to produce a uniform contamination, the value of the uniform contamination would be equal to the arithmetic mean of the heterogeneously contaminated system.” The SAB emphasizes that for the purposes of limiting exposure as well as estimating exposure from a defined area, the target statistic should always be the arithmetic mean, regardless of whether the underlying distribution is symmetrical or skewed. The 95% upper confidence limit is employed rather than the observed mean to allow for uncertainties in the estimated mean.

4.3.2 Parametric Statistical Analyses

Parametric statistical analyses require information on the distribution of the contaminant (e.g., normal or lognormal distribution). Generally, the tests are based upon normally distributed contaminants, or the data may be transformed to approximate a normal distribution. Environmental data typically are not normally distributed and often are lognormally distributed. The lognormality of the data requires transformation of the data to approximate a normal distribution.

Most of the common standard statistical methods are based upon the assumption that the data are normally distributed. If it can be demonstrated that the data are normally distributed and there are a sufficient number of results to support a decision concerning the survey unit, parametric tests will generally require fewer measurements to support a decision concerning the survey unit. However, tests that demonstrate the data are normally distributed generally require more measurements than non-parametric tests. The EPA provides guidance on selecting and performing statistical tests to demonstrate that data are normally distributed (EPA 1996). Guidance is also available for performing parametric statistical tests (EPA 1989, 1994b, 1996).

Common parametric statistical tests include the Students t Test (EPA 1996), t Test applied to logarithms (EPA 1996), minimum variance unbiased estimator for lognormal mean (Gilbert 1987), and the 2-Sample Quantile Test (EPA 1994b). EPA (1989) provides a complete discussion of the use of parametric statistics for testing if a survey unit soil concentration is less than the action levels using the mean or percentiles for random, stratified, or systematic sampling.

4.3.3 Non-parametric Statistical Analyses

Non-parametric, or distribution-free, statistical analyses can be used when the underlying distribution is either unknown or nonnormal. The validity of non-parametric techniques does not depend on the data being drawn from any particular distribution. The MARSSIM recommends the use of non-parametric statistical tests for evaluating environmental data. There are two reasons for this recommendation: (1) environmental data are usually not normally distributed and (2) there are often a significant number of qualitative survey results (e.g., less than the minimum detectable concentration). Either one of these reasons means that parametric statistical tests may not be appropriate. However, the MARSSIM also recommends the use of parametric statistical tests when the underlying assumptions required for these tests can be verified.

The non-parametric statistical tests presented in the MARSSIM guidance consist of the Wilcoxon Rank Sum test and the Sign test. The Wilcoxon Rank Sum and Sign tests are designed to determine whether the level of residual radioactivity uniformly distributed throughout the survey unit exceeds the soil action levels. Because these methods are based on ranks, the results are generally expressed in terms of the median. When the underlying distribution is symmetric, the mean is equal to the median. When the underlying distribution is not symmetric, these tests are still true tests of the median but only approximate tests of the mean. If the measurement distribution is skewed to the right, the average will generally be greater than the median. In severe cases, the average may exceed the soil action levels while the median does not. The reader is referred to the MARSSIM guidance for detailed discussions on the Wilcoxon Rank Sum and Sign tests. A brief description of these non-parametric tests follows.

The Wilcoxon Rank Sum test is a two-sample test that compares the distribution of a set of measurements in a survey unit to that of a set of measurements in a background reference area. The test is performed by first adding the value of the radionuclide soil action level to each measurement in the reference area (i.e., background soil concentrations). The combined set of survey unit data and adjusted reference area data are listed, or ranked, in increasing numerical order. If the ranks of the adjusted reference site measurements are significantly higher than the ranks in the survey unit measurements, the survey unit demonstrates compliance with the action levels.

The Sign test is a one-sample test that compares the distribution of a set of measurements in a survey unit to a fixed value, namely the radionuclide soil action level. The Sign test is used if the radionuclide contaminants of interest do not occur in background or the background levels are known to be a small fraction of the radionuclide soil action levels (e.g., <10%). A discussion on background radionuclide soil concentrations for the Rocky Flats environment is provided in Section 4.4. First, the value of each measurement in the survey unit is subtracted from the radionuclide soil action level. The resulting distribution is tested to determine if the center of the distribution is greater than zero. If the adjusted distribution is significantly greater than zero, the survey unit demonstrates compliance with the action level.

The Radiation Advisory Committee of the Science Advisory Board performed a review of the MARSSIM methodology (SAB 1997). The Science Advisory Board noted in their review that the recommended non-parametric statistical methods in the MARSSIM (NRC

1997) would not perform well with markedly asymmetric (skewed) distributions. The Science Advisory Board also noted that the target statistic for any exposure assessment should be the arithmetic mean concentration for a defined area, together with the uncertainty associated with the estimate of the mean. For a normally distributed population, the mean and the median are identical in value. However, when the distribution of sample evidence is moderately to highly skewed, then the non-parametric statistical techniques recommended by the MARSSIM cannot be used to determine the uncertainty associated with the estimate of the arithmetic mean, and the median of such a sample set will underestimate the true arithmetic mean of the surface soil contamination.

SAB (1997) provides two techniques that allow for estimating the mean and an estimate of the uncertainty about the mean. The Science Advisory Board suggests that the Sign test used in NRC (1997) be replaced by using a “resampling” or “bootstrap” estimator for the distribution of the arithmetic mean (Efron and Tibshirani 1998). Bootstrapping is a process that generates a series of estimates for the mean of a distribution by repeatedly resampling from the actual set of measured values and then analyzes those means with standard statistical techniques. Such an approach is straightforward; we simply perform a large number (e.g., $K = 1000$) of iterations in which we resample, with replacement, from the original N sample values (sample values can occur more than once in each resampling) and calculate the mean of each iteration. For example, if $N = 10$, the original sample might be 2, 5, 3, 2, 6, 4, 6, 3, 4, 4, which has a mean of 3.9. The first resampling might yield 5, 4, 6, 3, 3, 5, 2, 2, 2, 4 for a mean of 3.6. Additional resamplings would yield other means, both above and below 3.9. Depending on the skew of the original data and the number of iterations, the grand mean of the resampling might be higher or lower than 3.9, and researchers could also obtain an estimate of the uncertainty about that grand mean. The 50th largest mean value from the 1000 alternative realizations (of a sample of size $N = 10$) would be equal to the upper 95th percentile of the true but unknown arithmetic mean. This 95th percentile value on the arithmetic mean must be less than the soil action level in order to declare the site safe for release.

SAB (1997) also recommends a Bayesian analysis. The Science Advisory Board states that in some cases, the contamination data will not represent a truly random sample of the environment (e.g., data for hot spot samples). Such information can still be useful, but prior information about the sample’s properties is needed, leading to a Bayesian view of hypothesis testing. When data are only partially representative of a remediated site because they are not taken from a randomized design or they do not conform precisely to the same spatial and temporal scales as those upon which the soil action levels are based, then classical statistical techniques are of limited use in determining the uncertainty about the true but unknown arithmetic mean concentration for that site. SAB (1997) notes that under these circumstances, approaches based on Bayesian statistics may be advantageous. Bayesian statistics permit the explicit use of expert judgment to account for the inherent possibility of flaws and biases in the data. The result is that a credibility (or subjective confidence) interval can be obtained about the arithmetic mean (or any desired quantile) of the true but unknown distribution of soil concentration for both the remediated site and any reference site (i.e.,

background area). These credibility intervals form the basis upon which subsequent decisions are made.

The selection of statistical methods for comparing the mean soil concentrations and associated upper confidence interval to the soil action levels involves many decisions, and knowledge of the contaminant distributions and spatial correlations. Therefore, selecting a statistical method is not recommended in this report. Selecting a statistical test must be performed during the DQO process. Statistical techniques reviewed in this section can be used for the soil action level project; however, caution must be employed to ensure that the test will compare the mean soil concentrations at the 95% upper confidence interval to the soil action levels.

4.3.4 Correlated Data

Special consideration must be given to spatially correlated data when using systematic sampling. In practice, a spatial correlation may be present so that part of the information contained in one measurement is also in other measurements taken close by in space. One disadvantage of a systematic grid, with a random starting point, is that any pronounced trend in the population will result in a substantial reduction in the accuracy of the estimated population mean.

The periodic features of a population should be known before systematic sampling is adopted. In general, if a contaminant comes from a point source, concentrations will usually decrease with distance from the source. This relationship has been identified for the 903 Pad contamination, with plutonium concentrations decreasing with increasing distances from the 903 Pad. However, it is difficult to predict how the correlation structure will change after remediation of a given area and with different remedial methods. For example, contamination that is spatially correlated before remediation may effectively be randomly mixed after cleanup of contaminated soil from high contamination areas has been removed. In addition, depending on the selected size of a survey unit (Section 4.6), the correlation may not be statistically significant for small survey units. The radionuclide contamination varies considerably across the site, such that some survey units may not show a correlation between measurements and may present a random pattern. This is difficult to predict without prior knowledge of the survey unit sizes, orientation of the survey unit on the site, and knowledge of remedial methods used in the survey unit. Methods are available for improving the estimate of the mean using a systematic sample when spatial correlations are present. The reader is referred to Gilbert (1987) for a discussion of these methods.

4.3.5 Null Hypothesis

The probability of making a decision error, such as releasing a site that is actually above the soil action levels, can be controlled by adopting an approach called hypothesis testing. In hypothesis testing we begin by making a tentative assumption about a population parameter. This tentative assumption is called the null hypothesis and is denoted by H_0 . We then define another hypothesis, called the alternative hypothesis, which is the opposite of what is stated in the null hypothesis. This alternative hypothesis is denoted by H_a . The hypothesis testing

procedure involves using data from a sample to test the two competing statements indicated by H_0 and H_a .

Let μ_0 denote the specific numerical value of the population mean being considered in the null and alternative hypotheses. In general, a hypothesis test concerning the values of the population mean μ must take one of the following three forms:

$$\begin{array}{lll} H_0: \mu \geq \mu_0 & H_0: \mu = \mu_0 & H_0: \mu \leq \mu_0 \\ H_a: \mu < \mu_0 & H_a: \mu \neq \mu_0 & H_a: \mu > \mu_0 \end{array}$$

In many situations, the choice of H_0 and H_a is not obvious. In such cases, judgment on the part of the user is needed to select the proper form of H_0 and H_a . However, as the above example forms show, the equality part of the expression (either $=$, \geq , or \leq) always appears in the null hypothesis. In selecting the proper form of H_0 and H_a , we must keep in mind that the alternative hypothesis is what the sampling study is attempting to establish. Thus, asking whether the user is looking for evidence to support $\mu < \mu_0$, $\mu = \mu_0$, or $\mu > \mu_0$ will help determine H_a .

RAC recommends the following null and alternative hypotheses for the soil action level project:

$$H_0: \mu \geq \text{SAL} \text{ and } H_a: \mu < \text{SAL}$$

The null hypothesis as stated above assumes that the survey unit is contaminated above (or equal) to the action level; thus, the statistical analysis must prove that this assumption is not true. If the statistical analysis provides evidence that the null hypothesis is not true, then we accept the alternative hypothesis that the survey unit soil concentrations are less than the soil action levels.

The statement of the null hypothesis above is designed to be protective of human health and the environment as well as consistent with current methods used for demonstrating compliance with regulations. The EPA, NRC, and DOE have adopted the use of this null hypothesis in the MARSSIM for statistical analysis of contaminated sites. The EPA has also provided null hypotheses stated in this manner in their soil cleanup standards document (EPA 1989). It is acknowledged that site contamination conditions, such as lack of measurement techniques with appropriate detection sensitivities, may preclude the use of the null hypothesis that the survey unit is assumed to be contaminated. Another problem arises when there is a high variability in background concentrations of the radionuclides. When the background concentrations (see Section 4.4) are near the proposed soil action levels, it becomes difficult to distinguish between background and contamination concentrations. This does not appear to be the case at the RFETS. However, NUREG-1505 (NRC 1995b) provides guidance if this situation is encountered.

4.3.6 False Positive and Negative Rates

A Type I decision error occurs when the null hypothesis is rejected when it is true; it is referred to as a false positive error. A false positive error would result in the release of a

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survey unit containing residual radioactivity above the action levels. The probability of making a Type I decision error, or the level of significance, is denoted by alpha (α). Alpha reflects the amount of evidence the decision-maker would like to see before abandoning the null hypothesis; it is also referred to as the size of the test.

A Type II decision error occurs when the null hypothesis is accepted when it is false; it is referred to as a false negative error. A false negative error would result in either unnecessary cost because of remediation of survey units that are below the release criterion or additional survey activities to demonstrate compliance. The probability of making a Type II decision error is denoted by beta (β). The term ($1 - \beta$) is the probability of rejecting the null hypothesis when it is false; it is also referred to as the power of the test.

Table 4-1 provides an explanation of false positive and false negative conclusions based on our proposed null and alternative hypotheses:

Table 4-1. False Positive and Negative Conclusions based on the Recommended Null Hypothesis

| Decision based on the sample data are: | The true condition is: | |
|---|--|--|
| | Clean | Contaminated |
| Clean | Correct decision | False positive (Type I error) released as clean when it is really contaminated |
| Contaminated | False negative (Type II error) unnecessary remediation | Correct decision |

Recommendations on acceptable Type I and II errors cannot be provided. These error rates are developed during the DQO process and are adjusted according to considerations of the cost of sampling versus the cost of remediation. The values of α and β that are selected during the DQO process should reflect the risk involved in making a decision error. In setting the values for α , the following are important considerations (NRC 1997):

- In radiation protection practice, risk estimates are derived based on a linear function of dose for a variety of solid cancers (National Research Council 1990). Therefore, a 10% change in dose results in a 10% change in risk. This situation is quite different from one in which there is a threshold. In the latter case, the risk associated with a decision error can be quite high, and low values of α should be selected. When the risk is linear and without a threshold, much higher values of α at the action levels might be considered adequately protective when the survey design results in smaller decision error rates at doses greater than the action levels. False positives will tend to be balanced by false negatives across survey units, resulting in approximately equal human health risks.

- The radionuclide soil action levels cannot be determined with absolute precision. The dose cannot be measured directly, and many assumptions are made in converting dose to action levels. This is an additional consideration for setting the value of that could support the use of larger values in some situations. In this case, the magnitude, significance, and potential consequences of decision errors at values above the action levels would be prospectively addressed as part of the DQO process.
- The risk of making the second type of decision error, β , is the risk of requiring additional remediation when a survey unit already meets the release criterion. Unlike the health risk, the cost associated with this type of error may be highly nonlinear. There may be a threshold below which the remediation cost rises rapidly. This is primarily a concern for survey units that are near the action levels. For survey units that are very lightly contaminated or have been thoroughly remediated so that any residual contamination is expected to be far below the soil action levels, larger values of β may be appropriate, especially if final status survey sampling costs are a concern.

4.3.7 Recommendations

- As the previous discussion indicates, selecting the statistical method to analyze the data in comparison to the soil action levels involves many decisions. *RAC* recommends that the DQO planning team include a statistician familiar with the RFETS site and environmental statistical designs.
- *RAC* recommends that the arithmetic mean and its associated uncertainty at the upper 95% confidence interval be used for comparison to the soil action levels.
- *RAC* agrees with the findings of SAB (1997) that the Wilcoxon Rank Sum and Sign tests recommended in NRC (1997) are tests of the median. The MARSSIM (NRC 1997) non-parametric statistical tests should not be used for the soil action level study because these test compare the median value of the sample distribution to the soil action levels. When the distribution is not symmetrical (i.e., skewed) the median will not be equal to the mean.
- *RAC* does not recommend a specific statistical test for use in comparing the soil sample data to the soil action levels. *RAC* believes that the DQO process is the best mechanism for this selection process. Initial sampling results, or those obtained during remediation, should be evaluated to determine the potential population distribution and attributes to ensure that the most appropriate statistical techniques are employed. It is difficult to predict the affect that remediation will have in terms of changes in the present contamination attributes, including depth distribution. Various remediation strategies result in different changes in the contamination, including mixing with depth, removal of the higher contamination areas, and typically a reduction in the variance of the contamination distribution.

Risk Assessment Corporation

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- *RAC* cautions that spatial correlations could impact the accuracy of the estimated mean using a systematic grid with a random starting point. Methods are available for improving the estimate of the mean using a systematic sample, and the reader is referred to Gilbert (1987) for a discussion of these methods.

4.4 Radionuclide Background Soil Concentrations

Background concentrations for the radionuclides considered in the soil action levels must be evaluated to determine their significance. As discussed in Section 4.3.2, if the background radionuclide soil concentrations are less than 10% of the soil action levels and the soil sample data distribution is symmetric (i.e., not skewed), then the non-parametric bootstrapping can be used to test the upper 95% confidence interval of the mean from the sample distribution against the soil action levels. Appendix C of the Task 5 report (Killough et al. 1999) contains a detailed discussion on background plutonium in the environment. These data are summarized in the following section along with background data for other radionuclides. The following discussion is provided to the RSALOP to develop an understanding of background soil concentrations of important radionuclides in the Rocky Flats environment.

4.4.1 Background Studies

Background concentrations of plutonium isotopes are a result of weapons testing fallout and burnup of a Transit Navigational Satellite containing a ^{238}Pu power generator. Background studies in surface soil (Table 4-2) indicate that concentrations of $^{239,240}\text{Pu}$ in soil range from about 0.7 to 2.1 Bq kg⁻¹ along the Front Range of Colorado. Krey (1976) calculated the global-fallout plutonium in the Denver area using ^{240}Pu : ^{239}Pu ratios. The CDPHE has identified several remote locations thought to represent background levels of $^{239+240}\text{Pu}$. The $^{239+240}\text{Pu}$ activity for soils collected from these areas in 1989 using the Rocky Flats sampler ranged from 0.51 to 2.84 Bq kg⁻¹ (0.014 to 0.077 pCi g⁻¹), with a mean of 1.23 Bq kg⁻¹ (0.033 pCi g⁻¹). Purtymun et al. (1990) studied the impact of global-fallout plutonium on remote areas in Southern Colorado and Northern New Mexico. They showed that $^{239+240}\text{Pu}$ activity collected from soils ranged from 0.04 to 2.99 Bq kg⁻¹ (0.001 to 0.08 pCi g⁻¹), with a mean of 1.13 Bq kg⁻¹ (0.031 pCi g⁻¹). Litaor et al. (1995) also presented background soil concentrations for the Colorado Front Range and the Eastern Plains referred to as the Whiting-1994 and EG&G-1989 data sets. These reports were indicated to be unpublished by Litaor et al. (1995) and are reproduced in Table 4-2, as provided in the published paper by Litaor et al. (1995). Webb (1996) also estimated background $^{239,240}\text{Pu}$ in soil along the Front Range using 10 sampling sites and three sampling depths: 0–0.3 cm, 0–3 cm, and 0–21 cm. Plutonium-239,240 concentrations measured by Webb ranged from 1.2 to 3.3 Bq kg⁻¹.

Table 4-2. Background Soil Concentrations of $^{239+240}\text{Pu}$ in Soils of Colorado in Bq kg^{-1} (pCi g^{-1})

| Statistics | Whiting-1994 ^a | EG&G-1989 ^a | Krey (1976) | Webb (1996) ^c | Purtymun et al. (1990) |
|--------------------|----------------------------|----------------------------|--------------------------|--------------------------|---------------------------|
| No. of samples | 50 | 9 | 11 | 10 | 5 |
| Mean | 1.4 (0.038) | 1.23 (0.033) | 0.66 (0.018) | 2.14 (0.058) | 1.13 (0.031) |
| Standard deviation | 0.5 (0.014) | 0.73 (0.020) | 0.14 (0.0038) | 0.76 (0.021) | 1.06 (0.029) |
| Range | 0.62–2.66 (0.017–0.072) | 0.51–2.84 (0.014–0.077) | 0.40–0.92 (0.01–0.02) | 1.2–3.3 (0.032–0.089) | 0.04–2.99 (0.001–0.08) |

^a As reported in Litaor et al. (1995).

^b Unpublished data. Rocky Flats, Golden, CO 80402-0464.

^c 0–3 cm layer.

Background concentrations of ^{241}Am are also a result of weapons testing fallout. Hulse et al. (1999) evaluated background activity concentrations of ^{241}Am in 26 soil samples from depths of 0 to 3 cm (Table 4-3). The resulting data were approximately lognormally distributed with a mean of 1.3 Bq kg^{-1} (0.035 pCi g^{-1}) and 95% sign confidence interval of 1.1 to 1.6 Bq kg^{-1} (0.03 to 0.04 pCi g^{-1}). Depositions of ^{241}Am in 0 to 21 cm soil columns at 20 background locations were also approximately lognormally distributed with a median of 0.20 Bq kg^{-1} (0.005 pCi g^{-1}) and 95% sign confidence interval of 0.15 to 0.30 Bq kg^{-1} (0.004 to 0.008 pCi g^{-1}).

DOE (1995) reported that ^{241}Am concentrations in 50 soil samples from depths of 0 to 5 cm along Colorado's Front Range ranged from 0.04 to 0.9 Bq kg^{-1} (0.001 to 0.024 pCi g^{-1}), and that concentrations in similar samples from Rock Creek, north-northeast of Rocky Flats, ranged from 0.04 to 1.3 Bq kg^{-1} (0.001 to 0.035 pCi g^{-1}).

Table 4-3. Median and 95% Sign Confidence Intervals for Concentrations and Depositions of ^{241}Am in Soil from 20 Regional Background Locations along Colorado's Front Range (Hulse et al. 1999)

| Measurement | Median | 95% confidence interval. |
|---|-------------|--------------------------|
| 0 to 3 mm soil fraction, Bq kg^{-1} (pCi g^{-1}) | 1.1 (0.030) | 0.8–1.7 (0.02–0.046) |
| 0 to 3 cm soil sample, Bq kg^{-1} (pCi g^{-1}) | 1.3 (0.035) | 1.1–1.6 (0.030–0.043) |
| 21 cm depositions, kBq m^{-2} | 0.20 | 0.15–0.30 |

Concentrations of uranium isotopes are mostly from natural sources and vary depending on local geologic and geochemical conditions. Litaor (1995) presented data compiled on the

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background soil concentrations of the uranium isotopes. Table 4-4 presents the background data compiled by Litaor (1995) for the United States average, Colorado, and Rock Creek near Rocky Flats.

Table 4-4. Background Activities of Uranium Isotopes in Soil (Litaor 1995)

| Uranium isotopes | Number of samples | Range Bq kg ⁻¹ (pCi g ⁻¹) | Arithmetic mean and standard deviation |
|--------------------------------|-------------------|---|---|
| <i>U.S average^a</i> | | | |
| Uranium-238 | 355 | 4.4–140 (0.12–3.8) | 37.0 _ 30.7 (1.0 _ 0.83) |
| <i>Colorado^a</i> | | | |
| Uranium 238 | 32 | 17.3–111 (0.47–3.0) | 44.4 _ 33.6 (1.2 _ 0.91) |
| <i>Rock Creek^b</i> | | | |
| Uranium-238 | 21 | 29.6–56 (0.8–1.5) | 41.6 _ 7.3 (1.1 _ 2.0) |
| Uranium-235 | 21 | 0.4–5 (0.011–0.124) | 2.0 _ 1.3 (0.054 _ 0.035) |
| Uranium-234 | 21 | 28.5–54 (0.77–1.4) | 41.9 _ 6.4 (1.13 _ 0.17) |

^a Statistics were taken from Myrick et al. (1983).

^b Statistics compiled from a Rock Creek study west of the Rocky Flats Plant considered unaffected by Rocky Flats Plant activity.

4.4.2 Recommendations

- These radionuclide background concentrations indicate that background could be excluded from the statistical analyses. The radionuclide background concentrations are near or below 1 pCi g⁻¹. This would indicate that soil action levels for each radionuclide at 10 pCi g⁻¹ or higher could be statistically evaluated without consideration of background. However, it must be noted that DOE and the site contractor must determine whether they can accept the potential consequences of additional remediation by not considering background radionuclide concentrations in the statistical analyses.

4.5 Determining the Required Number of Samples

There are several equations available for calculating the number of samples required to obtain a specified level of confidence in the estimated value of the mean. These equations range from simple to more complex formulations. Equations are presented in the MARSSIM (NRC 1997) guidance for determining the number of required samples based on the Wilcoxon Rank Sum and Sign tests, and the reader is referred to this document for further information on these methods. Additional approaches to determine the required number of soil samples are provided in this section.

4.5.1 Methods

SAB (1997) presents an equation for the number of required samples for ensuring that sampling of a percentile, α of the distribution with some probability P (e.g., $P = 0.95$), the relevant equation for the required sample size N is

$$N = \frac{\text{Ln}(1 - P)}{\text{Ln}(\alpha)} \quad (4-4)$$

The result of Equation (4-4) with P and $\alpha = 0.95$ is 59 if we round up to the nearest integer. Therefore, SAB (1997) states that a sample size of about 60 will nearly always be sufficient to characterize a survey unit. However, SAB (1997) does not provide a reference for this particular sample size equation, so further investigation into its validity was not possible.

EPA (1989) presents a method for estimating the required number of samples for a systematic sampling design where the statistical parameter of interest is the mean. The sampling precision of an estimated mean from a systematic sample depends on the pattern of contamination at the site and how the systematic sample is constructed. However, EPA (1989) notes that the standard error of a mean based on a systematic sample will usually be comparable to or less than the standard error of a mean based on a random sample of the same size. Therefore, using the sample size formulas for a random sample when the sample was collected systematically will generally error on the side of conservatism. The following equation is presented by EPA (1989):

$$N = \frac{(z_{1-\alpha} + z_{1-\beta})^2 \sigma^2}{SAL - \mu_1} \quad (4-5)$$

where

N = number of soil samples

σ^2 = standard deviation

$z_{1-\alpha}$ = critical value for the normal distribution with probability $1-\alpha$

$z_{1-\beta}$ = critical value for the normal distribution with probability $1-\beta$

SAL = soil action level

μ_1 = the value of μ (i.e., the “true” but unknown mean contaminant concentration across the sample area, or population mean) under the alternative hypothesis for which a specified false negative rate is to be controlled ($\mu_1 < \mu_0$).

White and Hakonson (1979) investigated the statistical aspects of the use of the coefficient of variation, c , in the design of environmental plutonium studies. The number of observations, n , required for acceptable results is dependent on (1) the desired precision of the estimate (confidence interval length) or the power of the test to be obtained and (2) the variance of the data. Deming (1950) discusses how c can be used to determine the necessary

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sample size, n , to estimate a mean, \bar{X} with some standard error $\sigma_{\bar{X}}$, when the standard error is expressed as a percent of the mean. Deming denotes c as the coefficient of variation of the estimate, that is

$$c = \frac{SE(\bar{X})}{\bar{X}} \quad (4-6)$$

Without any knowledge of the mean, the sample size required to estimate the mean with standard error $\sigma_{\bar{X}}$ is

$$n = \frac{c^2}{\sigma_{\bar{X}}^2} \quad (4-7)$$

From this equation, we can see that n increases as the square of c , or that as c doubles, n quadruples. White and Hakonson (1979) evaluated the variability of plutonium concentrations in soil and reported coefficients of variation for Microplot 1 and 2 from the Little (1976) study of 2.7 and 8.4, respectively.

4.5.2 Variability of the Radionuclide Concentrations

In general, most equations for the determination of sample size require an initial estimate of the variability for the soil measurements within the survey unit. Therefore, an estimate of the standard deviation, σ , variance, σ^2 , or coefficient of variation, $c = \sigma / \bar{X}$, of the contamination distribution must be obtained. Unfortunately, the standard deviation is usually unknown and must be estimated for the purpose of determining the sample size. In practice, the estimate of σ is either obtained from past sampling data or by conducting a small preliminary investigation in the survey unit of interest.

4.6 Classification and Identification of Survey Units

All areas of the RFETS site will not have the same potential for contamination and, therefore, will not need the same level of investigation to achieve the soil action levels. The final verification surveys will be more efficient if the surveys are designed so that areas with a higher potential for contamination receive a higher degree of investigation as recommended in NRC (1997).

The MARSSIM (NRC 1997) provides classifications for areas based upon their potential for contamination exceeding the respective soil action levels. Contamination areas must be classified to determine the appropriate survey unit size. Survey unit sizes are discussed in the Section 4.6.4. The MARSSIM classifications are described in Sections 4.6.1 through 4.6.3.

4.6.1 Class 1 Area

Class 1 areas have, or had before remediation, a potential for radioactive contamination (based on site operating history) or known contamination (based on previous radiological surveys). Areas containing contamination in excess of the action levels before remediation are classified as Class 1 areas. Examples of Class 1 areas include (a) site areas previously subjected to remedial actions, (b) locations where leaks or spills are known to have occurred, (c) former burial or disposal sites, and (d) waste storage areas.

4.6.2 Class 2 Area

Class 2 areas have, or had before remediation, a potential for radioactive contamination or known contamination but are not expected to exceed the action levels. To justify changing an area's classification from Class 1 to Class 2, the existing data from scoping or characterization surveys should provide a high degree of confidence that no individual measurement would exceed the action levels. Examples of areas that might be classified as Class 2 for the final status surveys include (a) potentially contaminated transport routes, (b) areas downwind from stack release points, and (c) areas on the perimeter of former contamination control areas.

4.6.3 Class 3 Area

Class 3 areas are any impacted areas that are not expected to contain any residual radioactivity or are expected to contain levels of residual radioactivity at a small fraction of the action levels based on site operating history and previous radiological surveys. Examples of areas that might be classified as Class 3 include buffer zones around Class 1 or Class 2 areas and areas with very low potential for residual contamination but insufficient information to justify a nonimpacted classification.

4.6.4 Identification of Survey Units

A survey unit is a physical area consisting of land areas of specified size and shape for which a separate decision will be made as to whether that area exceeds the soil action levels. The survey unit is the primary entity for demonstrating compliance with the release criterion (NRC 1997).

To facilitate survey design and ensure the number of survey data points for a site are relatively uniformly distributed among areas of similar contamination potential, the site is divided into survey units. The survey units share a common history and/or other characteristics or are naturally distinguishable from other portions of the site. Dividing the site into survey units is critical only for the final status (verification) survey; scoping, characterization, and remedial action support surveys may be performed without dividing the site into survey units.

Based upon NRC (1997), survey units should be limited in size based on classification, exposure pathways modeling assumptions, and site-specific conditions. Table 4-5 contains the suggested areas for survey units given in the MARSSIM (NRC 1997) guidance.

Table 4-5. Survey Unit Suggested Areas (NRC 1997)

| Classification | MARSSIM-suggested area |
|--------------------|--------------------------------|
| Class 1—land areas | Up to 2,000 m ² |
| Class 2—land areas | 2,000 to 10,000 m ² |
| Class 3—land areas | No limit |

The equations for the number of samples needed for a survey unit are typically based on the variance of the contamination within a survey unit and do not consider the size (i.e., area) of the survey unit. Therefore, the limitation on survey unit size for Class 1 and 2 areas ensures that each area is assigned an adequate number of sample points. The limitations on survey unit size provided by NRC (1997) are only suggested values. The DQO process should consider additional factors for delineating survey units, including areas of remediation as survey unit boundaries.

Several considerations are associated with defining survey units, for example:

1. Multiple survey units should be specified for the RFETS. These areas should be defined so that they are as homogeneous as possible with respect to radionuclide concentrations.
2. Survey units should also be defined according to potential contamination events. For example, areas identified by Litaor (1995) that contain uranium contamination, such as from the east spray fields (see Section 4.1.3), should be identified as separate survey units from areas that were not contaminated by uranium.
3. The RFETS will contain areas that require remediation and others areas that do not require remediation. Areas that have been remediated should not be combined with areas that have not been remediated in the same survey unit. In addition, areas that have undergone different remediation techniques, for example removal versus mixing, should also not be combined in the same survey unit.

4.6.5 Recommendations

- RAC recommends that the MARSSIM classification scheme and limitations on survey unit size be used for the final status survey.

4.7 Small Areas of Elevated Activity (Hot Spots)

The use of systematic grid, with a random starting point, for soil sampling provides data to determine the upper 95% confidence interval of the mean for comparison to the soil action levels. Soil sampling with a systematic sampling grid may not successfully identify

small areas of elevated contamination. Instead, soil sampling on a systematic grid, along with radiation detection instrumentation, are recommended to obtain adequate assurance that small areas of elevated radioactivity (i.e., hot spots) are identified during the final status survey.

4.7.1 Definitions and Detection Methods

The RSALOP and RAC have been involved in discussions dealing with hot spot definitions and methods to ensure that acceptable radionuclide soil concentrations are attained at the RFETS. The RSALOP has developed the following hot spot definition:

A hot spot is the location where any sample (or combination of samples) taken when following a prescribed sampling protocol that results in a radionuclide soil concentration exceeding the soil action level.

The RSALOP has also developed the following supporting statements in regard to hot spots:

- If a hot spot is found to exist, it should be evaluated to determine if action is required.
 1. Hot spots with areas equal to or greater than 100 m² must be remediated if the 95% upper confidence limit of the arithmetic mean soil concentrations, calculated from sample data taken in the hot spot, exceeds the soil action levels. The arithmetic mean is calculated by simple averaging of the soil concentrations in the hot spot because the hot spot encompasses the entire averaging area of 100 m².
 2. Remediation is required for hot spots with areas less than 100 m² when the area weighted arithmetic mean soil concentrations at the 95% upper confidence interval for the hot spot, when summed with the area weighted mean (95% confidence interval) of the soil concentration in the remaining 100 m² area (or as an alternative, the averaging area for the residential scenario), exceeds the soil action levels. The area weighted arithmetic means are used for this case (i.e., hot spot area < 100 m²) because the hot spot area does not encompass the entire averaging area of 100 m².
- It is reasonable to assume that a hot spot(s) can exist within a defined area and the dose criteria for the area will still be met.
- The RSALOP may wish to establish soil action levels that if exceeded, must be remediated *regardless* of the results from averaging discussed above. The alternative action levels will ensure that small hot spots (i.e., < 25 m²) do not contain unacceptable radionuclide soil concentrations regardless of the results of area averaging.

Identification of a hot spot, according to the above definition, requires that DOE and their site contractor, Kaiser-Hill Company, conduct additional investigations of hot spots. This can be accomplished by additional soil sampling or in situ gamma spectroscopy techniques (see the following section on surrogate measurements). The size of the hot spot must be delineated and the average radionuclide soil concentrations determined. Averaging of the hot spot over a 100 m² area was considered by the RSALOP to be consistent with future residential use of the land and to be protective of children that may be exposed to these hot spot in residential backyards.

The RSALOP has conducted a workshop on instrumentation for the detection of radionuclides in the Rocky Flats environment and is aware of methods and techniques available for use in the soil action level study. At Rocky Flats, the use of in situ gamma-ray spectroscopy measurements, which employ high-energy resolution germanium gamma-ray detectors, can be used to identify areas of elevated soil contamination. The use of in situ gamma-ray measurements is recommended by RAC for identifying hot spots that may potentially be located between the soil sample locations. It should be noted that instrumentation for radiation detection is a vast field of study and new developments are continuously being announced. Therefore, RAC does not want to discourage the use of more recent techniques or the development of better detection instrumentation. RAC encourages DOE and Kaiser-Hill Company to investigate the use of these emerging technologies and techniques for application to the final status survey during the DQO process.

Recent studies have been conducted on the identifying hot spots using in situ gamma spectroscopy techniques. Reginatto et al. (1998, 1997) developed a computer code, ISD97, to analyze data from a series of in situ gamma spectroscopy measurements on a grid. The code was designed to be used as a tool when evaluating compliance with regulations that set limits on the size and magnitude of hot spots. The code calculates the location and magnitude of potential elevated activity areas consistent with the data; for each potential elevated area, it generates a corresponding distribution of radionuclides in soil. In practical applications, if any of these potential distributions appear to exceed the soil action levels, further field work (such as soil sampling, collimated measurements, or additional measurements on a closer grid spacing) would be used to determine whether such an elevated area was present. The algorithm in the code uses a maximum entropy deconvolution of the data. In deconvolution, the data and a set of additional conditions are used to find a distribution of activity in the soil that fits the data and satisfies the given set of conditions.

4.7.2 Recommendations

- RAC recommends the use of in situ gamma spectroscopy measurements to identify hot spot areas that may be located between the soil sampling points located on a systematic grid.
- RAC recommends that hot spot locations identified by soil samples or in situ gamma spectroscopy measurement be investigated further to delineate the size of the hot spot and to determine the upper 95% confidence interval of the mean concentration for radionuclides contained in the hot spot.

- RAC recommends that DOE and their site contractor, Kaiser-Hill Company, investigate the use of emerging techniques such as the ISD97 computer model (Reginatto et al. 1998, 1997) for use in locating hot spots during the final status survey.

4.8 Surrogate Measurements

With multiple radionuclides in the soil at Rocky Flats, it may be possible to use surrogate measurements. A surrogate radionuclide is easily measured and implies the concentration of other radionuclides.

4.8.1 Site-specific Studies

The application of surrogates has been investigated by Shierman (1994) and Hulse et al. (1999). Shierman (1994) investigated the use of ^{241}Am as a surrogate to determine $^{239,240}\text{Pu}$. The ^{241}Am concentrations obtained during the study along the C transect of the 903 Pad were used to describe the $^{239,240}\text{Pu}$ concentrations, and the following relationship was found:

$$^{239,240}\text{Pu} (\text{pCi g}^{-1}) = ^{241}\text{Am} (\text{pCi g}^{-1}) \cdot 0.189^{-1} \quad (4-8)$$

Shierman (1994) indicated that this relationship can be used to estimate the ^{239}Pu concentration indirectly by counting the ^{241}Am via gamma spectroscopy. This would provide a quick method to quantify ^{239}Pu without expensive and labor-intensive radiochemical techniques. However, Shierman indicates that this technique may not be useful in low-level environmental samples (less than 0.27 pCi g^{-1}) because of the difficulty in quantifying ^{241}Am using gamma spectroscopy at such levels. In these cases, radiochemical techniques would be required. The ratio of ^{241}Am to $^{239,240}\text{Pu}$ for the RFETS soil analyzed in the Shierman (1994) study remained constant ($r = 0.9946$) as depth increased, but it was highly variable. Because no relationship between the ratio and depth was found by Shierman (1994), all the samples from the C transect were pooled and a median value (the distribution of the ratio data was skewed) of 0.166 was determined. The mean and standard deviation were 0.187 ± 0.94 . The ratio of ^{241}Am and $^{239,240}\text{Pu}$ for the sampling locations on the A transect had a median of 0.189. The mean and standard deviation of the ratio for the A transect was 0.210 ± 0.85 .

Hulse et al. (1999) presented a similar relationship between ^{241}Am and $^{239,240}\text{Pu}$ using their data and data from splits of samples reported by Webb (1996) and Webb et al. (1994, 1997). The data indicated a strong relationship between ^{241}Am and $^{239,240}\text{Pu}$ in soil from depths of 0–3 cm. A log-log regression yielded

$$[^{239,240}\text{Pu}] = 5.5 [^{241}\text{Am}]^{1.1} \quad (4-9)$$

with an r-squared value of 0.87 for locations where soil concentrations exceeded decision levels. They also found that the ratio between ^{241}Am and $^{239,240}\text{Pu}$ in soil samples from depth of 0–3 cm, which was estimated by regression, was 0.18 for all onsite locations. The ratio for offsite locations was 0.36. The ratio between concentrations of ^{241}Am and $^{239,240}\text{Pu}$ in soil

from depths of 0 to 3 mm for all onsite locations was 0.22. This ratio was found by Hulse et al. (1999) to be the same as that reported by Mongan et al. (1996) in airborne effluents from Rocky Flats and almost the same as the ratio (0.21) reported by Litaor and Allen (1996) in soil from depths of 0 to 6 mm at their onsite sample locations. The results were also consistent with ratios calculated by Litaor et al. (1998) for concentrations in soil from depths of 0 to 20 cm and 20 to 40 cm at locations close to the 903 Pad.

Hulse et al. (1999) also used nonlinear regression to estimate parameters for the Hill four-parameter sigmoidal model that predicts the ratio of ^{241}Am to $^{239,240}\text{Pu}$ in soil from depths of 0 to 3 cm (Y) at distance (X in km) from the 903 Pad. The model is given by equation 4-10.

$$Y = Y_o + (aX^b)(C^b + X^b)^{-1} \quad (4-10)$$

The parameters, Y_o , a , b , C were determined to have values of 0.21, 0.21, 5.16, and 1.16, respectively. The model predicted that the ratio (95% confidence interval) should be 0.21 (0.15 to 0.29) close to the 903 Pad and 0.41 (0.35 to 0.51) at distances of more than 50 km. The predicted ratio of 0.41 at background locations was higher than the mean ratio of 0.36 estimated directly from soil concentrations in samples from offsite locations. The predicted ratio was higher than the ratio of 0.32 obtained by adjusting the ratio estimated by Krey et al. (1976) for additions of ^{241}Am from the decay of ^{241}Pu in global fallout since 1974. It was also higher than the ratio of 0.29 – 0.17 that Hulse et al. (1999) estimated using concentrations in background soil samples reported by DOE (1995). Litaor and Allen (1996) reported that the mean ratio between ^{241}Am and $^{239,240}\text{Pu}$ in soil from depths of 0 to 6 mm at offsite locations around Rocky Flats was 0.56. Hulse et al. state that these ratios should be used judiciously when predicting concentrations of $^{239,240}\text{Pu}$ from soil concentrations of ^{241}Am to delineate the extent of contamination from Rocky Flats.

The benefit of using the surrogate approach is the reduced cost of not having to perform wet chemistry analyses on each sample. This benefit must also consider the relative difficulty in establishing the surrogate ratio, as well as the potential consequences of unnecessary investigations that result from the error in using a conservative surrogate ratio.

NRC (1997) recommends that when the ratio is established before remediation, additional post-remediation samples should be collected to ensure that the data used to establish the ratio are still appropriate and representative of the existing site condition. If these additional post-remediation samples are not consistent with the pre-remediation data, surrogate ratios should be reestablished.

4.8.2 Recommendations

- RAC recommends the use of surrogate measurements in the final status survey at the RFETS. The ratio of ^{241}Am to $^{239,240}\text{Pu}$ would provide a mechanism for the in situ gamma spectroscopy measurements of ^{241}Am to be used to predict the soil concentrations of $^{239,240}\text{Pu}$.
- RAC recommends that post-remediation samples be collected to ensure that the data used to establish the ratio are still appropriate, as specified in the MARSSIM guidance (NRC 1997).

4.9 Independent Confirmatory Investigations

An independent verification survey is performed by an independent third party, contracted by the DOE, to provide data to substantiate results of the final status survey. The independent verification survey would be limited in scope to spot checking conditions at selected locations, comparing findings with those of the final status survey, and performing independent statistical evaluations of the data developed from the final status survey. This task would burden the DOE with additional costs for the soil action level project; however, *RAC* considers the benefit of such an independent confirmatory investigation to outweigh the cost. The independent confirmatory investigation would provide the public with assurances that DOE and their site contractor, Kaiser-Hill Company, have conducted the final status surveys in a technically defensible manner and that decisions to release specific survey units of the site are the correct decision.

4.9.1 Recommendations

- *RAC* recommends that the RSALOP request DOE to implement a confirmatory survey (also known as an independent verification survey) for the radionuclide soil action level project. This survey should be performed by an independent third party.

5. SOIL SAMPLING QUALITY ASSURANCE

The goal of QA/QC is to identify and implement sampling and analytical methodologies that will limit the introduction of errors into analytical data. The required QA/QC program elements are typically developed and documented in QAPjPs or in similar documents, including, but not limited to, decommissioning plans, sampling and analysis plans, and field sampling plans. Section 2 discusses the RFETS sampling program and concludes that QA program elements are documented and available for use in the final status survey soil sampling protocol. However, the DQO process must be used to define the specific elements of the QA program for use in the soil action level study.

In general, the following progression of events leads to developing a QA/QC program for soil sampling programs:

1. Statement of the study objectives
2. Evaluation of the impact of mistakes
3. Definition of the DQOs
4. Design of the soil sampling study to achieve DQOs
5. Design of the QA/QC program to confirm achievements of DQOs.

Present guidance for the development of DQOs identifies the following factors for consideration in the sampling program design (NRC 1997):

- Precision
- Accuracy
- Bias
- Completeness
- Representativeness
- Comparability.

Developing a QA program for soil sampling is beyond the scope of this report. Typically, developing a soil sampling program and associated QA/QC requirements is an iterative process that the RSALOP, DOE, and Kaiser-Hill Company will need to perform during the DQO process.

In this report, *RAC* outlines the available QA/QC guidance for soil sampling programs and suggested elements for consideration by the RSALOP. Elements of a QA/QC program for soil sampling include DQOs, documentation, chain-of-custody, laboratory requirements, data validation, and the assessment of DQIs.

5.1 Data Quality Objectives and Data Quality Indicators

This section specifically targets the QA/QC aspects (or DQOs) of the soil sampling program and the method for identifying potential errors from the point of sample collection to the final analytical result. DQOs are qualitative and quantitative statements developed by data users to specify the quality of data needed from a particular data collection activity (EPA 1987).

DQOs must address five DQIs: precision, accuracy, representativeness, completeness, and comparability. Bias is also considered by the MARSSIM methodology manual (NRC

1997). Another data characteristic, level of detection, should also be addressed because it is closely related to the other elements. Using DQIs in the QA sampling program allows researchers to determine if the data are of necessary quality to make a particular decision.

DQIs are not all quantitative (numerical) measurements; some DQIs are subject to qualitative (relative) analysis. Of the six principal DQIs, precision and bias are quantitative measures, representativeness and comparability are qualitative, completeness is a combination of both qualitative and quantitative measures, and accuracy is a combination of precision and bias.

The number of required QC measurements is determined by the available resources and the degree to which assurance is needed that a measurement process is adequately controlled. The number of QC measurements may also be driven upward as the action level approaches an instrument's detection limit.

A widely used standard practice is to collect a set percentage, such as 5% (EPA 1987), of the samples for QA purposes. However, this method has disadvantages. Depending on the number of samples to be collected, small numbers will result in insufficient QC samples, whereas large sample numbers may require too many QC samples and waste resources. A performance-based alternative is also available (EPA 1990).

The precision of an estimate of the "true" variance for precision and bias within a survey design depends on the number of QC measurements performed to provide the estimate. The MARSSIM provides one-sided upper confidence limits for selected numbers of QC measurements assuming the results of the measurements are normally distributed (see Table 5-1). At the stated level of confidence, the true variance of the estimate of precision or bias for a specified number of QC measurements will be between zero and the multiple of the estimated variance listed in the table. For example, for five field replicate samples, you would be 90% confident that the true variance for precision falls between 0 and 3.10 times the estimated variance based on the results of the five samples.

When planning surveys, the number of each type of QC measurement can be obtained from Table 5-1. For example, if the survey objective is to estimate the variance in the bias for a specific measurement system between 0 and 2 times the estimated variance at the 95% confidence level, 15 measurements of a material with known concentration (e.g., performance evaluation samples) would be indicated.

The MARSSIM recommends that the survey objective be set so that the true variance falls between 0 and 2 times the estimated variance. The level of confidence is then determined on a survey unit-specific basis to adjust the number of each type of QC measurements to the appropriate level.

Table 5-1. Upper Confidence Limits for the True Variance as a Function of the Number of Quality Control Measurements Used to Determine the Estimated Variance^a

| Number of quality control measurements | Level of confidence (%) | | | |
|--|-------------------------|-------|-------|-------|
| | 90 | 95 | 97.5 | 99 |
| 2 | 9.49 | 19.49 | 39.21 | 99.50 |
| 5 | 3.10 | 6.01 | 6.02 | 9.02 |
| 10 | 2.05 | 2.54 | 3.08 | 3.91 |
| 15 | 1.76 | 2.07 | 2.40 | 2.87 |
| 20 | 1.61 | 1.84 | 2.08 | 2.42 |
| 25 | 1.52 | 1.71 | 1.91 | 2.17 |
| 30 | 1.46 | 1.62 | 1.78 | 2.01 |
| 40 | 1.38 | 1.51 | 1.64 | 1.80 |
| 50 | 1.33 | 1.44 | 1.61 | 1.68 |
| 100 | 1.21 | 1.28 | 1.35 | 1.43 |

^a. The variance lies between 0 and x factor of the true variance at a given confidence level. For example, if 20 QC samples are taken, we are 95% confident that the true value lies between 0 and 1.84 times the estimated variance.

The DQIs are described in the following sections.

5.1.1 Precision

Precision is a quantitative measure of agreement among replicate measurements of the same property under prescribed similar conditions (ASQC 1995). Several types of replicate analyses are available to determine the level of precision. These replicates are typically distinguished by the point in the sample collection and analysis process where the sample is divided. The types of QA samples that may be used for determining precision include

- *Collocated Samples.* Collocated samples are collected adjacent to the routine field sample to determine local variability of the radionuclide concentration. Analytical results from collocated samples can be used to assess site variation but only in the immediate sampling area. They are not recommended for assessing error (EPA 1991).
- *Field Replicates.* Field replicate samples are obtained from one location, homogenized, divided into separate containers, and treated as separate samples throughout the remaining handling and analytical process. These samples are used to assess error associated with sample heterogeneity, sample methodology, and analytical procedures. Field replicates are used when determining total error for critical samples with contamination concentrations near the action level. For statistical analysis to be valid in such a case, a minimum of eight replicate samples would be required (EPA 1991).

- *Analytical Laboratory Replicate.* An analytical laboratory replicate is a subsample of a routine sample that is homogenized, divided into separate containers, and analyzed using the same analytical method. It is used to determine method precision; however, because it is a nonblind sample (i.e., known to the analyst), it can only be used by the analyst as an internal control tool and not as an unbiased estimate of analytical precision (EPA 1990).
- *Laboratory Instrument Replicate.* A laboratory instrument replicate is a repeated measurement of a sample that has been prepared for counting (i.e., laboratory sample preparation and radiochemical procedures have been completed). It is used to determine precision for the instrument (repeated measurements using same instrument) and the instrument calibration (repeated measurements using different instruments, such as two different germanium detectors with multichannel analyzers). A laboratory instrument replicate is generally performed as part of the laboratory QC program and is a nonblind sample. It is typically used as an internal control tool and not as an unbiased estimate of the analytical precision.

When collocated measurements are performed, an estimate of total precision is obtained. When collocated samples are not available for laboratory analysis, a sample subdivided in the field and preserved separately can be used to assess the variability of sample handling, preservation, and storage along with the variability in the analytical process, but variability in sample acquisition is not included. When only variability in the analytical process is desired, a sample can be subdivided in the laboratory before analysis.

5.1.2 Bias

Bias is the systematic or persistent distortion of a measurement process that causes errors in one direction (ASQC 1995). Bias is determined quantitatively based on the analysis of samples with a known concentration. There are several types of samples with known concentrations:

- *Reference Material.* Reference material is a material or substance, one or more of whose property values are sufficiently homogeneous and well established to be used for the calibration of an apparatus, the assessment of a measurement method, or for assigning values to materials (ISO 1993). A certified reference material contains certified values that are accompanied by an uncertainty at a stated level of confidence. When appropriate reference materials are available (i.e., proper matrix, proper radionuclide, and proper concentration range), they are recommended for use in determining the overall bias for a measurement system.
- *Performance Evaluation Samples.* Performance evaluation samples evaluate the overall bias of an analytical laboratory and detect any error in the analytical method used. These samples are usually prepared by a third party, using a quantity of radionuclides that is known to the preparer but unknown to the laboratory and

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always undergoes certification analysis. Laboratory procedural error is evaluated by the percentage of the radionuclide identified in the performance evaluation sample (EPA 1991).

- *Matrix Spike Samples.* Matrix spike samples are environmental samples that are spiked in the laboratory with a known concentration of a target radionuclide to verify percent recoveries. They are primarily used to check sample matrix interferences, but they can also be used to monitor laboratory performance. However, a data set of at least three or more results is necessary to distinguish between laboratory performance and matrix interference (EPA 1991).

Several types of QA/QC samples are also used to detect bias caused by contamination, which include

- *Field Blanks.* Field blanks are samples prepared in the field using certified clean sand or soil and then submitted to the laboratory for analysis (EPA 1991). A field blank is used to evaluate contamination error associated with sampling methodology and laboratory procedures. It also provides information about contaminants that may be introduced during sample collection, storage, and transport (NRC 1997).
- *Method Blanks.* A method blank is an analytical control sample used to demonstrate that reported analytical results are not the result of laboratory contamination (ATSDR 1992). It contains distilled or deionized water and reagents and is carried through the entire analytical process (laboratory sample preparation, digestion, and analysis). The method blank is also referred to as a reagent blank. The method blank is generally used as an internal control tool by the laboratory because it is a nonblind sample (NRC 1997).

5.1.3 Accuracy

Accuracy is a measure of the closeness of an individual measurement or the average of a number of measurements to the true value (EPA 1997). Accuracy includes a combination of random error (precision) and systematic error (bias) components that result from performing measurements.

Accuracy is determined by analyzing a reference material of known concentration or by reanalyzing material to which a known concentration of contaminant has been added. To be accurate, data must be both precise and unbiased. As an example, consider a target. To be accurate, the shots at the target must land close together and, on average, at the spot where they are aimed. In other words, the shots must all land near the bull's eye. Figure 5-1 shows this analogy.

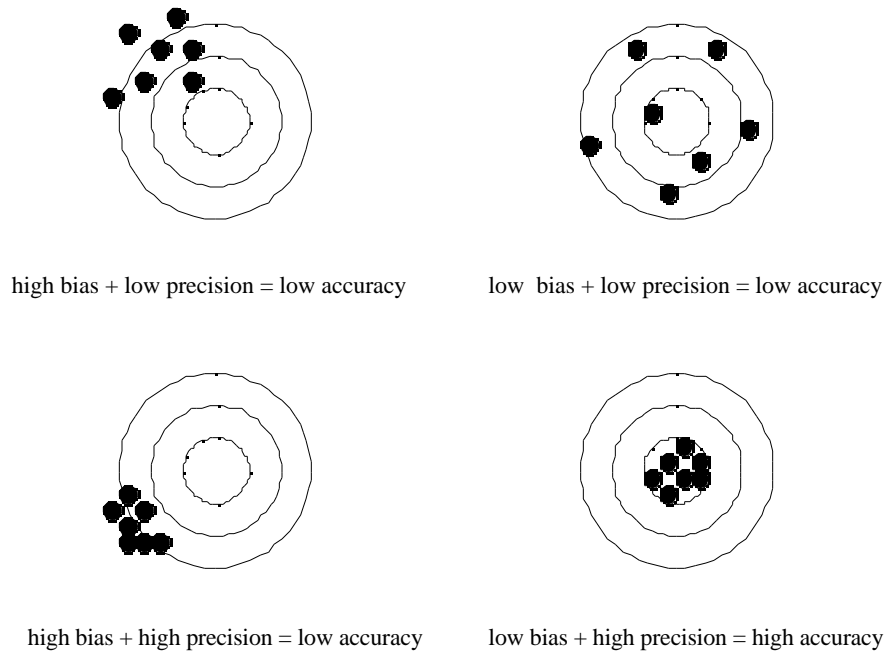


Figure 5-1. Measurement bias and random measurement uncertainty (NRC 1997).

Accuracy is usually expressed either as a percent recovery or as a percent bias. Determination of accuracy always includes the effects of variability (precision); therefore, accuracy is a combination of bias and precision.

5.1.4 Representativeness

Representativeness is a measure of the degree to which data accurately and precisely represent a characteristic of a population parameter at a sampling point (ASQC 1995). Representativeness is a qualitative term that should be evaluated to determine whether physical samples were collected in a manner that the resulting data appropriately reflect the media and contamination measured.

When soil sampling is required as part of a survey design, it is critical that the sample collection procedures consider representativeness. Sample collection procedures also need to consider the modeling basis for the radionuclide soil action levels when determining the representativeness of the samples.

Representativeness is primarily a planning concern. The solution to enhancing representativeness is in the design of the sampling plan. Analytical data quality also affects representativeness because data of low quality may be rejected for use in the analysis.

5.1.5 Comparability

Comparability is a qualitative term that expresses the confidence that two data sets can contribute to a common analysis and interpolation. Generally, comparability is provided by using the same measurement system for all analyses of a specific radionuclide. Comparability is usually not an issue except in cases where historical data have been collected and are being compared to current analysis results or when multiple laboratories are used to provide results as part of a single sampling design (NRC 1997).

The comparability objective provides the needed control over the total measurement process to ensure that different studies can be compared. Comparability provides a basis for comparing trends over time or space, evaluating the relationship between sampling programs, or ensuring that phased sampling efforts produce data of a consistent quality.

When sampling is to occur over an extended period of time or when the investigator desires to compare several sites, it is necessary to ensure that the samples be collected in a comparable manner, from comparable fraction of the soil mass, and with comparable methods. For example, samples collected by coring should not be compared with bucket auger samples.

5.1.6 Completeness

Completeness is a measure of the amount of valid data obtained from the measurement system, expressed as a percentage of the number of valid measurements to total number of measurements collected.

Completeness for measurements is calculated by the following formula (NRC 1997):

$$\% \text{ completeness} = \frac{(\text{number of valid measurements})}{\text{total number of measurements planned}} \cdot 100 \quad (5-1)$$

Samples collected on a grid to locate areas of elevated activity are also a concern for completeness. If one sample analysis is not valid, the entire sample design for locating areas of elevated activity may be invalidated. If a sufficient amount of sample was originally collected, the analysis can be repeated using archived sample material.

Completeness is not intended to be a measure of representativeness, that is, it does not describe how closely the measured results reflect the actual concentration or distribution of the contaminant in the media being measured. A project could produce 100% data completeness, but the results may not be representative of the actual contaminant concentration. Alternatively, there could be only 70% data completeness (30% lost or found invalid), but because of the nature of the sample design, the results could still be representative of the target population and yield valid results.

For most final status surveys, the issue of completeness only arises when the survey unit demonstrates compliance with the release criterion and less than 100% of the measurements are determined to be acceptable. The question then becomes whether the number of measurements is sufficient to support the decision to release the survey unit.

An alternative method to ensure completeness is to take samples in addition to those determined appropriate for the sample design. The planning stages of any study must take into consideration the fact that not all samples will make it intact through the entire measurement process. For example, sample containers will be broken, instruments will fall out of control, data will be lost, and sample tags will be lost. Many factors can lead to a sample result being invalidated. These invalid samples can be compensated for by oversampling or by using a phased sampling effort that allows areas where samples were lost to be resampled in subsequent phases. This latter approach ensures that the desired number of samples will be collected. For example, if 20% more measurements were taken in a survey design than required, then a sampling project with 80% completeness may still have sufficient power to support a decision to release the survey unit.

The design of a particular sampling effort provides a minimum number of samples needed to yield a desired level of precision for the final results. The probabilities of false positive and false negative answers are specified at the outset. Obviously, any loss from the required number of samples will impact the final results. DOE has set a completeness objective for environmental survey programs at 90% for both field sampling and laboratory analyses (DOE 1987).

5.1.7 Detection Limits

Selecting analytical methods based on detection limits is an important process. The detection limit of the method directly affects the usability of the data because results near the detection limit have increased measurement uncertainty.

5.2 Data Validation

Validation of the analytical data is the process by which the quality of the data is assessed by using the specified DQIs and QA/QC sample results. Analytical data validation, including field and laboratory data review, is defined as the systematic process, performed external from the data generator, that applies a defined set of performance-based criteria to a body of data and may result in qualification of the data. Data validation provides a level of assurance, based on a technical evaluation, that an analyte is present or absent; if present, it provides the level of uncertainty associated with the measurement. Data validation must occur before drawing a conclusion from the data.

Analytical data validation for radiochemistry includes a technical review of the laboratory data package covering the evaluation of DQI samples, the identification and quantitation of analytes, and the effect of deficiencies in QC on analytical sample data.

Although the EPA has developed numerous guidance documents relating to data validation of organic and inorganic constituents, no national standard currently covers data validation of radiochemistry concepts adequately. The need for a document of this type has been recognized by most of the DOE complex. There is reference in the MARSSIM (NRC 1997) to the development of such guidance, the *Multi-Agency Radiation Laboratory Analytical Protocols (MARLAP) Manual*; however, currently, no such document exists.

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Because of the lack of specific guidance, currently each DOE site has developed site-specific data validation procedures for radiochemistry data.

During data validation, the reviewer examines the data, documentation, and reports to determine if the sampling program was conducted within the limits specified by the DQO process.

5.3 Documentation

Three types of documentation are available for review during the data validation process: (1) field operation records, (2) laboratory records, and (3) data handling records (EPA 1997).

5.3.1 Field Operation Records

The information contained in these records documents the field operations and consists of the following:

- *Field Measurement Records.* Field measurement records provide documentation that the proper measurement protocol was performed during the sampling project. This documentation includes the names of the persons conducting the sampling, sample location and identification, maps and diagrams, equipment and SOP used during sampling, and unusual observations. Bound field notebooks are generally used to record raw data; however, data recording forms may also be used for this documentation.
- *Sample Tracking Records.* Sample tracking records, also referred to as chain-of-custody records, document the progression of samples as they travel from the original sampling location to the laboratory and finally to disposal.
- *General Field Procedures.* General field procedures, also referred to as SOPs, record the procedures used in the field for collecting soil samples.

5.3.2 Laboratory Records

The following list describes some of the laboratory-specific records that should be reviewed if available and appropriate:

- *Laboratory Measurement Results and Sample Data.* Laboratory measurement results and sample data contain information on the sample analysis used to verify the analytical methods that were followed. The overall number of samples, sample identification, sample measurement results, any deviations from the SOPs, time of day, and date should be included. Sample location information may also be provided.
- *Sample Management Records.* Sample management records should document sample receipt, handling, and storage and scheduling of analyses. The records will verify that sample tracking requirements were maintained; reflect any anomalies in the samples,

such as receipt of damaged samples; and note proper log-in of samples into the laboratory.

- *QC Measurement Records.* QC measurement records include general QC records, such as initial demonstration of capability, instrument calibration, routine monitoring of analytical performance, and calibration verification. Project-specific information from the QC checks, such as blanks, spikes, calibration check samples, replicates, and splits, should be included in these reports to facilitate data quality analysis.

5.3.3 Data Handling Records

Data handling records document protocols used in data reduction, verification, and validation. Data reduction addresses data transformation operations, such as converting raw data into reportable quantities and units, using significant figures, and calculating measurement uncertainties. The records document procedures for handling data corrections.

5.4 Data Validation Qualifiers

Data validation begins with an assessment of the quality of analytical results and is performed by a professional with knowledge of the analytical process. Depending on the survey objectives, the level and depth of review varies. The level and depth of the data validation may be determined during the planning process and should include an examination of laboratory and method performance for the measurements and radionuclides involved. This review includes

- Evaluating data completeness
- Verifying instrument calibration
- Measuring precision using replicates or split samples
- Measuring bias using reference material or spikes
- Examining blanks for contamination
- Assessing adherence to method specifications and QC limits
- Evaluating method performance in sample matrix
- Assessing applicability and validation of analytical procedures or site-specific measurements
- Assessing external QC measurement results and QA assessments.

Following the data validation process, data are assigned validation qualifiers. The person conducting the data review assigns coded qualifiers to the data when QC requirements or other evaluation criteria are not met. An explanation of the data qualifiers should be included in the data validation report, along with a summary of the quality of the data package.

6. CONCLUSIONS

The primary goal of Task 6 is to develop recommendations for a soil sampling protocol for use at RFETS to obtain soil concentration data for comparison to the soil action levels. Sampling protocols are written descriptions of the detailed procedures to be followed in collecting, packaging, labeling, preserving, transporting, and documenting the samples.

Sampling protocols are developed using the iterative DQO process and require DOE and its contractor to evaluate several important considerations. These considerations include evaluating sampling and analytical costs in relation to available resources and accepting potential decision errors that may result in remediating sites that are judged contaminated when they are actually below the soil action levels. Conversely, developing a sampling protocol must also incorporate the concerns of the general public and other stakeholders, which are represented by the RSALOP and the soil action level study. Because of the complexity of developing sampling protocols, with the inherent need to balance the concerns of DOE and the RSALOP, developing a comprehensive sampling protocol was not considered possible. In this report, *RAC* presents recommendations for those elements of a soil sampling protocol considered essential to ensure representative soil samples are collected for comparison to the soil action levels. These recommendations are provided to the RSALOP for presentation to DOE and its contractor, Kaiser-Hill Company, for incorporation into the soil sampling protocol and procedures to be used for the soil action level study.

The sampling protocol recommendations were developed for the surface-soil sampling (i.e., 0 to 20 cm) during final status survey. In this report, *RAC* used the general principles of the MARSSIM (NRC 1997) guidance to develop recommendations for a sampling protocol. *RAC* has provided an emphasis in this report on problems identified with the MARSSIM guidance in terms of its application to the soil action levels at Rocky Flats and potential solutions.

The following is a summary of the recommendations for a soil sampling protocol in support of the final status survey. *RAC* recommends that

1. The DQO process be used to develop the soil sampling protocol for the final status survey.
2. DOE appoint representatives from the RSALOP for inclusion on the DQO planning team.
3. The RSALOP select discrete values from the soil action level distributions for each radionuclide and use these discrete values for comparison to the soil concentration data.
4. Soil samples be collected using profile sampling.
5. Profile sampling be conducted in soil depth increments of 0–3 cm to be consistent with the resuspension model parameters used to develop the soil action levels.

6. Soil samples should not be composited; rather, individual soil samples should be analyzed for radionuclide contaminants.
7. Soil samples be collected using a systematic grid sampling design, with a random starting point.
8. A statistician familiar with the RFETS and environmental statistical designs be included on the DQO planning team.
9. The arithmetic mean of the soil concentration data and its associated uncertainty at the upper 95% confidence interval be used for comparison to the soil action levels.
10. The MARSSIM (NRC 1997) non-parametric statistical tests not be used for the soil action level study since these test compare the median value of the sample distribution to the soil action levels. When the distribution is not symmetrical (i.e., skewed), the median is not equal to the mean.
11. Parametric statistical tests, bootstrapping, or geostatistical techniques be investigated for use in comparing the 95% upper confidence limit of the mean against the soil action levels. The statistical tests should be investigated during the DQO process and chosen according to knowledge of the areas to be sampled.
12. Spatial correlations be investigated to determine their presence in the survey unit of interest and to determine if methods are required to improve the estimate of the mean based on the systematic grid sampling method.
13. The null (H_0) and alternative (H_a) hypothesis are stated as $H_0: \bar{x} \leq SAL$ and $H_a: \bar{x} > SAL$, where \bar{x} is the mean soil concentration and SAL is the soil action level.
14. The survey units be classified according to the NRC (1997) scheme and that the size of the survey units be limited accordingly.
15. In situ gamma spectroscopy measurement be performed to identify potential hot spot locations.
16. Hot spots identified by soil samples or in situ gamma spectroscopy measurements be investigated further to delineate the size of the hot spot and to determine the upper 95% confidence interval of the mean radionuclide concentrations contained in the hot spot.
17. That hot spots greater than 100 m^2 with arithmetic mean soil concentrations at the upper 95% confidence interval that exceed the soil action levels be remediated.

18. That hot spots less than 100 m² be area averaged with soil concentrations in a 100 m² area and area weighted to determine if the upper 95% confidence interval of the mean soil concentration exceeds the soil action levels and, thus, requires remediation.
19. The use of surrogate measurements in the final status survey. The ratio of ²⁴¹Am to ^{239,240}Pu provides a mechanism for the in situ gamma spectroscopy measurement of ²⁴¹Am to be used to predict the soil concentrations of ^{239,240}Pu.
20. DOE implement an independent verification survey for the radionuclide soil action level project. This survey should be performed by an independent third party.

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