

United States
Environmental Protection
Agency

Office of Emergency and
Remedial Response
Washington, DC 20460

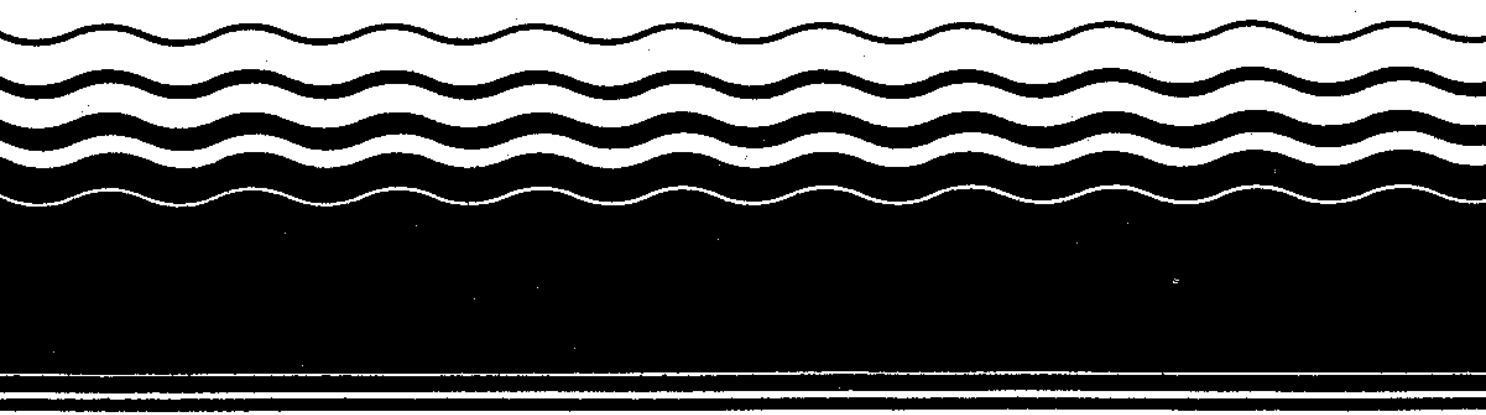
Publication 9285.7-09B
PB92 - 963362
May 1992

Superfund



Guidance for Data Useability in Risk Assessment (Part B)

Final



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Notice: This is a supplement to Guidance
for Data Useability in Risk Assessment -
Part A

Office of Emergency and Remedial Response
U.S. Environmental Protection Agency
Washington, DC 20460

NOTICE

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Tips*

- *A health physicist or radiochemist should work with the risk assessor from the beginning of the remedial investigation process. (page 1)*
- *Field measurements must be made using instruments sensitive to the type of radioactivity present. (page 13)*
- *The shipper of radioactive material is responsible for ensuring that the recipient is authorized to receive the shipped material and for compliance with all applicable shipping and labelling regulations. (page 25)*

* For further information, refer to the text. Page numbers are provided.

PREFACE

This document is the second part (Part B) of the two-part *Guidance for Data Useability in Risk Assessment*. Part A, developed by the EPA Data Useability Workgroup, provides guidance on the analytical data quality and useability requirements needed for the cleanup of hazardous waste sites under the Comprehensive Environmental Response, Compensation, and Liability Act of 1980 (CERCLA) as amended by the Superfund Amendments and Reauthorization Act of 1986 (SARA). Part B provides supplemental guidance to Part A on planning and assessing radioanalytical data needs for the baseline human health risk assessment conducted as part of the remedial investigation (RI) process at sites containing radioactive substances. Part B is not a stand-alone document, and at all times it must be used in conjunction with Part A.

This guidance is addressed primarily to the remedial project managers (RPMs) who have the principal responsibility for leading the data collection and assessment activities that support the human health risk assessment. It also should be of use to risk assessors who must effectively communicate their data needs to the RPMs and use the data provided to them. Because

of the special hazards and unique sampling and analysis considerations associated with radioactive substances, RPMs and risk assessors are strongly encouraged to consult with a health physicist, radiochemist, or both, starting at the beginning of the RI planning process. For reference, a list of the EPA Headquarters, Regional and Laboratory radiation program staff is provided in the Appendices.

Comments on the guidance should be sent to:

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ACKNOWLEDGEMENTS

This document was prepared under the direction of Ruth Bleyler and Lisa Matthews of the Toxics Integration Branch (TIB), and Anthony Wolbarst and Michael Boyd of the Office of Radiation Programs (ORP), all of EPA Headquarters. Preparation of this document benefited greatly from the technical reviews and recommendations provided by the following individuals, to whom we wish to express our sincere appreciation:

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Chapter 1

Introduction and Background

This guidance provides supplemental information regarding the useability of analytical data for performing a baseline risk assessment at sites contaminated with radioactivity. The reader should be familiar with the guidance provided in *Guidance for Data Useability in Risk Assessment - Part A* before proceeding with this document. Although Part A focuses primarily on chemical contamination, much of the information presented also applies to the risk assessment process for radioactive contamination. The guidance offered in this document is intended as an overview of the key differences between chemical and radionuclide risk assessments, and not as a comprehensive, stand-alone document to assess the risks posed by radionuclide exposures. Part A of this guidance should be used side by side with this document because of the many references to information and exhibits found in Part A.

• *A health physicist or radiochemist should work with the risk assessor from the beginning of the remedial investigation process.*

There are special hazards and problems associated with radioactivity contamination. Accordingly, it is recommended that a professional experienced in radiation protection and measurement (health physicist or radiochemist) be involved in all aspects of the risk assessment process from the beginning of the remedial investigation/feasibility study.

Additional information on important aspects of radiation protection and measurement is provided in the appendices. These appendices are included to provide greater detail on topics presented in this guidance and to

facilitate a comprehensive understanding for the interested reader. Appendix I is a glossary of terms that apply to radioactivity. Appendix II is a discussion on naturally occurring radionuclides and their presence in the environment. Appendix III provides a list of the names and addresses of the EPA Regional, Laboratory, and Headquarters Radiation Program staff for health physics and radioanalytical support.

1.1 CRITICAL DATA QUALITY ISSUES IN RISK ASSESSMENT

The five basic environmental quality issues discussed in Part A Section 1.1 also apply to radioactive contamination. Specifics for data sources, detection limits, qualified data, background samples, and consistency in sample collection will be discussed later in this guidance.

1.2 FRAMEWORK AND ORGANIZATION OF THE GUIDANCE

This document is organized the same as Part A. Part A, Exhibit 2 describes the organization of this document. The assessment of radioanalytical data as opposed to chemical data is emphasized.

This guidance discusses the data collection and evaluation issues that affect the quality and useability of radioanalytical data for baseline human health risk assessments. Part A, Exhibit 3 lists the four components of the risk assessment process and the information sought in each of the components.

Chapter 2

The Risk Assessment Process

This chapter discusses the data collection and evaluation issues that affect the quality and useability of radioanalytical data for baseline human health risk assessments. Part A, Exhibit 3 lists the four components of the risk assessment process and the information sought in each of the components.

2.1 DATA COLLECTION AND EVALUATION

Part A, Section 2.1.1 contains an overview of methods for data collection and evaluation that can be applied to sites contaminated with radioactivity as well as with chemical hazards. The development of data quality objectives as part of a carefully designed sampling and analysis program will minimize the subsequent need to qualify the analytical data during the data analysis phase. Specific radioanalytical methods are described in Section 3.0 of this guidance, along with a discussion of chemicals of concern in Section 3.2. Strategies for selecting analytical methods and designing sampling plans can be found in Section 4.0.

2.2 EXPOSURE ASSESSMENT

The approach to risk assessment for radionuclides shares the objectives stated in Part A, Section 2.1.2:

- Identify or define the source of exposure.
- Define exposure pathways (receptors) including external exposure.
- Identify potentially exposed populations.
- Measure or estimate the magnitude, duration, and frequency of exposure to site contaminants for each receptor (or receptor group).

Exposure pathways should be designated before the design of sampling procedures.

2.2.1 Identifying Exposure Pathways

This section describes a methodology for estimating the radiation dose equivalent to humans from exposure to radionuclides through all pertinent exposure pathways. These estimates of dose equivalent can be compared with radiation protection standards and criteria, with an important cautionary note. These standards have been developed for regulating occupational exposure for adults and are not completely applicable to assessing risk for the population at large. Section 2.4 describes a methodology for estimating health risk.

Part A, Section 2.1.2 describes the procedures for exposure assessment for chemical contaminants, and many aspects of this section apply directly to radionuclides. However, the term “exposure” has a specific meaning for radionuclides which is distinct from its use with chemical contamination (see Appendix I). For chemicals, exposure usually refers to the intake of the toxin (e.g., inhalation, ingestion, dermal exposure) expressed in units of mg/kg-day, the same units used for toxicity values. Unlike chemical toxins, an exposure assessment for radionuclides can include an explicit estimate of the radiation dose equivalent.

Inhalation and ingestion remain as important exposure pathways for radionuclides, although the units to express intake are in activity (i.e., Bq or Ci) rather than mass. Radionuclides entering through these pathways may become incorporated within the body where they emit alpha, beta or gamma radiation providing internal exposure to tissues or organs. Absorption is not an important exposure pathway for radionuclides. Dose equivalent is a quantity that incorporates both the energy deposited internally from ionizing radiation and the effectiveness of that radiation to cause biological damage to the organism. The dose equivalent was developed to normalize the unequal biological effects produced from equal absorbed doses of different types of radiation (i.e., alpha, beta, or gamma).

Radionuclides need not be taken into or brought in contact with the body to produce biological damage. High energy emissions of beta particles and photons from radionuclides can travel long distances with minimal attenuation, penetrate the body, and deposit their energy in human tissues. External radiation exposures can result from either exposure to radionuclides at the site area or to radionuclides that have been transported from the site to other locations in the environment. Potential external exposure pathways to be considered include immersion in contaminated air or water and direct exposure from ground surfaces contaminated with beta- and photon-emitting radionuclides. Gamma and x-rays are the most penetrating of the emitted radiations and comprise the primary contribution to the radiation dose from external

Acronyms

DCF	dose conversion factor
EPA	U.S. Environmental Protection Agency
HEAST	Health Effects Assessment Summary Tables
IRIS	Integrated Risk Information System
RPM	remedial project manager

exposures. External exposure to beta particles primarily imparts a dose to the outer layer skin cells, although high-energy beta radiation can penetrate into the human body. Alpha particles are not sufficiently energetic to penetrate the outer layer of skin and do not contribute significantly to the external dose.

The amount of energy deposited in living tissue is of concern because the potential adverse health effects of radiation are proportional to the energy deposited. The energy deposited is a function of a radionuclide's decay rate, not its mass. Therefore, as mentioned earlier, radionuclide quantities and concentrations are expressed in units of activity.

Environmentally dispersed chemicals, stable and radioactive, are subject to the same processes that affect their transfer rates and therefore their bioaccumulation potential. Radionuclides undergo radioactive decay. In some respects, this decay can be viewed as similar to the chemical or biological degradation of organic compounds. Both processes change the quantity of the hazard present in the environment and produce other substances. The products of radioactive decay may also be radioactive and can contribute significantly to the radiation exposure. These radioactive decay products must be considered for risk assessment purposes.

2.2.2 Exposure Quantification

One of the objectives stated for exposure assessment was to make a reasonable estimate of the maximum exposure to receptors or receptor groups. The equation presented in Part A, Exhibit 7 to calculate intake for chemicals can be applied to exposure assessment for radionuclides, except that the body weight and averaging time terms should be omitted from the denominator. However, exposures to radionuclides include both internal and external exposure pathways, and radiation exposure assessments take the calculation an additional step in order to estimate radiation effective dose equivalent which is directly translatable to risk.

Radionuclide intake by inhalation and ingestion is calculated in the same manner as chemical intake except that it is not divided by body weight or averaging time. For radionuclides, a reference body weight and averaging time are already included in the dose conversion factors (DCFs), and the calculated dose is an expression of energy deposited per gram of tissue.

External exposures may be determined by monitoring and sampling of the radionuclide concentrations in environmental media, by direct measurement of radiation fields using portable instrumentation, or by mathematical modeling. Portable survey instruments that have been

properly calibrated can display dose rates (e.g., Sv/hr or mrem/hr), and dose equivalents can be estimated by multiplying the dose rate by the duration of exposure to the radiation field. Alternatively, measured or predicted concentrations in environmental media may be multiplied by DCFs, which relate inhaled or ingested radionuclide quantities to effective dose equivalent. Federal Guidance Report No. 11 (EPA 1988) provides DCFs for each of over 700 radionuclides for both inhalation and ingestion exposures, as well as immersion exposures to tritium and the principle radioactive noble gases. It is important to note that these DCFs were developed for regulation of occupational exposures to radiation and may not be appropriate for the general population. The Integrated Risk Information System (IRIS) (EPA 1989) and the Health Effects Assessment Summary Tables (HEAST) (EPA 1990) provide slope factors for radionuclides of concern for each of the three major exposure pathways (inhalation, ingestion, and external exposure) that may be applied to determining the risk to the general population.

The dose equivalents associated with external and internal exposures are expressed in identical terms (i.e., Sv), so that contributions from all pathways can be summed to estimate the total effective dose equivalent value and prioritize risks from different sources. A more extensive discussion of quantifying exposure from radioactivity can be found in *Risk Assessment Guidance for Superfund: Volume I, Human Health Evaluation Manual, Part A, "Baseline Risk Assessments"* (EPA 1991).

The radiation exposure assessment should include a discussion of uncertainty. This should include, at a minimum, a tabular summary of all values used to estimate exposures and doses, and a summary of the major assumptions used in the assessment process. Special attention should be paid to the three sources of uncertainty listed below:

- Correlation of monitoring data and the actual conditions on site.
- Exposure models, assumptions, and input variables used for the exposure estimate.
- Values of variables used to estimate intakes and external exposures.

2.3 TOXICITY ASSESSMENT

The objectives of toxicity assessment are to evaluate the inherent toxicity of the compounds under investigation, and to identify and select toxicological measures for use in evaluating the significance of the exposure. Certain

fundamental differences between chemicals and radionuclides somewhat simplify toxicity assessment for radionuclides.

Theoretically, any dose of radiation, no matter how small, has the potential to produce adverse effects, and therefore, exposure to any radioactive substance is hazardous. A large body of data derived from human and experimental animal studies establishes the principal adverse biological effects of exposure to ionizing radiation to be carcinogenicity, mutagenicity, and teratogenicity. EPA's current estimates of adverse effects associated with human exposure to ionizing radiation indicate that the risk of cancer is limiting and may be used as the sole basis for assessing the radiation-related human health risks of a site contaminated with radionuclides.

The dose-response assessment for radionuclides is also more straightforward, and this relationship is relatively well characterized at high doses. Accordingly, a detailed toxicity assessment for individual radionuclides at each site is not required. In general, radiation exposure assessments need not consider acute toxicity effects because the quantities of radionuclides required to cause adverse effects from acute exposure are extremely large and such levels are not normally encountered at Superfund sites.

2.4 RISK CHARACTERIZATION

The final step in the risk assessment process is risk characterization. This is an integration step in which the risks from individual radionuclides and pathways are

summed to determine the likelihood of adverse effects in potentially exposed populations. Since the concern is for radiation dose equivalent, and since all pathway doses are calculated in comparable units, the total effective dose equivalent from all pathways is easily computed and can be translated directly to risk.

All supporting documentation provided for the exposure assessment should be compiled to ensure that it is sufficient to support the analysis, to allow an independent duplication of the results, and to ensure that all exposure pathways have been addressed. Additionally, all assumptions regarding site conditions, environmental transfer factors, etc., must be carefully reviewed to ensure that they are applicable.

Once all data are in order, the next step is to calculate the risk based on the estimated committed effective dose equivalents. As stated earlier, risk assessment for radionuclides needs to be considered only for the end point of radiation carcinogenesis.

2.5 ROLES AND RESPONSIBILITIES OF KEY RISK ASSESSMENT PERSONNEL

The key risk assessment personnel and their responsibilities are discussed in Part A, Section 2.2. It is recommended that a health physicist or radiochemist be involved in the risk assessment process to provide technical assistance to the remedial project manager (RPM) and the risk assessor. For a listing of EPA health physics and radiochemical support staff, see Appendix III.

Chapter 3

Useability Criteria for Baseline Risk Assessments

This chapter discusses data useability criteria and preliminary sampling and analysis issues. This information can be used to plan data collection efforts in order to maximize the useability of environmental radioanalytical data in baseline risk assessments.

3.1 DATA USEABILITY CRITERIA

The data useability criteria presented in Part A, Section 3.1 are generally applicable to analytical data required for baseline risk assessment, including radioanalytical data.

3.1.1 Data Sources

The data source considerations given in Part A, Section 3.1.1 also apply to radioactively contaminated sites. Since radioactive contamination can often be detected in the survey process, preliminary assessment/site inspection (PA/SI) and any other field measurements may be of particular importance. Field measurements that provide data for external exposure rates, while usually considered screening, can be used for risk assessment purposes directly, provided they meet the data useability requirements. Also of potential importance are the operating history of the site, handling and disposal manifests, and U.S. Nuclear Regulatory Commission (USNRC) licenses or state agency permits regulating the possession of radioactive materials.

3.1.2 Documentation

The four major types of documentation discussed in Part A, Section 3.1.2 apply equally to radionuclides:

- Sampling and analysis plan (SAP) and quality assurance project plan (QAPjP).
- Standard operating procedures (SOPs), particularly those for the calibration and use of all field survey instruments.
- Field and analytical records, including all survey information relating to radiation or radioactivity concentrations.
- Chain-of-custody records.

3.1.3 Analytical Methods and Detection Limits

The importance of selecting proper analytical methods based on detection limits that meet risk assessment requirements is discussed for chemical analyses in Part A, Section 3.1. A discussion of detection limits for

radiation detection instruments can be found in Section 3.2. A strategy for selecting radioanalytical methods that meet risk assessment requirements is described in Section 4.2.

3.1.4 Data Quality Indicators

Data quality indicators are the performance measurements of data quality objectives (DQOs). These objectives should be a function of the desired confidence level of the risk assessment and not based on the availability or capability of specific analytical methods. DQOs must be clearly defined for all radiation and radioactivity measurements.

Quantitative data quality indicators for radioanalytical measurements may include a lower limit of detection, minimum detectable concentration, precision, accuracy, and completeness. Qualitative data quality indicators can be expressed as goals but cannot be demonstrated quantitatively. Such qualitative data quality indicators might include representativeness and comparability.

In setting DQOs, the relationship to the decision-making process is paramount. The primary rationale for setting DQOs is to ensure that the data will be of sufficient quality to support the planned decisions and/or actions to be taken based on those data.

The DQO process involves three stages: defining the decision, reviewing the existing data to determine what new data are required, and designing the sampling and analytical program to obtain the required data. Data

Acronyms

CLP	Contract Laboratory Program
DOT	U.S. Department of Transportation
DQO	data quality objective
EPA	U.S. Environmental Protection Agency
G-M	Geiger-Muller
HP	health physics
IDL	instrument detection limit
LLD	lower limit of detection
MDC	minimum detectable concentration
PA	preliminary assessment
PC	pressurized ion chamber
QAPjP	quality assurance project plan
QC	quality control
RPM	remedial project manager
SAP	sampling and analysis plan
SI	site inspection
SOP	standard operating procedure
SQL	sample quantitation limit
TCL	Target Compound List
TIC	tentatively identified compound
USNRC	U.S. Nuclear Regulatory Commission

quality will be a function of the chemical preparation, measurement system, selection of sampling and counting parameters, and the control limits set for the data quality indicators. After the establishment of the isotope-pathway combinations of interest, the risk assessor must develop the maximum uncertainties that can be tolerated in the assessment of the activity for an isotope in each media. These parameters define the data quality indicators which in turn determine the available procedures.

3.1.5 Data Review

While the RPM or other personnel can perform many aspects of basic data review, an individual experienced in radiochemistry or health physics must perform the detailed technical review of both the field and laboratory data. Such a review should be performed on preliminary data as they are collected and should continue throughout the risk assessment process.

Special attention must be paid to all reports prepared by data reviewers to ensure that there is a narrative summary in addition to the data summary tables provided. The additional, clarifying information in the narrative summary will be of particular importance to reviewers unfamiliar with radioanalytical data.

3.2 PRELIMINARY SAMPLING AND ANALYSIS ISSUES

A discussion of issues affecting sampling and analysis for baseline risk assessment is beyond the scope of this document. A framework of key issues, tools, and guidance used in the design and assessment of environmental sampling and analysis procedures is described in Part A, Section 3.2. This section concentrates on the differences between sampling and analysis for radioactive contamination compared to sampling and analysis for chemical contamination.

3.2.1 Radionuclides of Potential Concern

EPA classifies all radioactive substances as Class A carcinogens (i.e., known human carcinogens). Any radioactive substance detected or suspected of being present at or released from a site will be considered to be of potential concern and evaluated accordingly. The risk assessor should review the list of radionuclides of concern for each migration pathway. These lists should contain the following information for each radionuclide listed (see Appendix I for a more detailed discussion of each of the factors):

Atomic number and atomic weight. The elemental identity of a radioisotope is determined by the number

of protons in its nucleus (i.e., its atomic number), and its isotopic identity is determined by the total number of protons plus neutrons (i.e., its atomic weight). For example, plutonium has an atomic number of 94. Isotopes of plutonium, such as Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242, have identical atomic numbers but different atomic weights. The origin, use, isotopic abundance, radioactive (and perhaps physical) properties, and cancer potency of each plutonium isotope are unique. Thus, it is imperative that each radionuclide be properly identified.

Radioactive half-life. The radioactive half-life of a radioisotope is the time required for the activity of that isotope to be reduced by one half. Half-life is a unique characteristic of each radioisotope and is not affected by chemical or physical processes. Knowledge of the half-life of a radioisotope is important for the following reasons:

- The half-life determines the activity and cancer potency of the isotope.
- The half-life affects holding times for analyses (radionuclides with shorter half-lives must be analyzed in a shorter time frame than longer-lived radionuclides).
- The half-life determines the degree of activity equilibrium between decay products (radionuclides in equilibrium maintain equal levels of radioactivity, if the equilibrium is disturbed the activity levels of the progeny need to be measured separately).

Principal decay modes, radiation decay modes, energies, and abundances. Radioisotopes emit radiation in the form of alpha, beta, and neutron particles, as well as gamma photons and x-rays. The type, abundance, and energies of the radiations emitted by a radioisotope are unique to that isotope. Consequently, the selection and use of sampling and analysis procedures, radiochemical methods, and radiation detection instruments must be consistent with the decay mode (i.e., alpha, beta, neutron, or photon) and radiation energies and abundances of the radionuclide of concern.

Chemical and physical forms. The mobility, bioaccumulation, metabolic behavior, and toxicity of a radioisotope are governed by its chemical and physical form, not by its radioactive properties. Radioisotopes in the environment may exist as solids, liquids, or gases in a variety of chemical forms, oxidation states, and complexes. Information should be provided in the data package describing the most likely chemical and physical form(s) of each radionuclide at the time of production, disposal, release, and measurement.

Decay products. Radioactive decay of an isotope of one element results in the formation of an isotope of a different element. This newly formed isotope, the

decay product, will possess physical and chemical properties different from the parent isotope. For example, Ra-226 may be present as a solid in the form of radium sulfate while its daughter Rn-222 is a noble gas. Often, a decay product is also radioactive and decays to form a different radioisotope. It is important to consider all radioisotopes for the following reasons:

- The total activity content (and thus, the potential hazard) of a radioactive source or sample may be underestimated if progeny are excluded.
- An isotope's progeny may be more toxic, either alone or in combination, than the parent radioisotope. For example, Ra-226 decays to Rn-222 by alpha particle emission with a half-life of 1600 years, while Rn-222 and its daughters emit three additional alpha particles and two beta particles through the principle decay modes with a combined half-life of less than four days.
- The environmental transport, fate, and bioaccumulation characteristics of the progeny may be substantially different from those of the parent isotope.

The site records, including the operating history, handling and disposal manifests, and radioactive materials licenses or permits, will be useful in determining if the initial list of radionuclides of concern derived from these records and those radionuclides identified in media samples are consistent. All omissions or inconsistencies in the expected versus the observed radioisotopes at the site should be noted, and additional information should be sought to explain these discrepancies.

At sites containing both radioactive and other hazardous substances, the list of chemicals of concern should be reviewed for each sample medium for consistency and completeness. The manner in which radioactive substances are associated with nonradioactive hazardous substances on the site should be described by the RPM or risk assessor, to the extent that such information is available. This description also should include a discussion of the possible effects that these chemicals may have on radionuclide mobility and bioaccumulation.

3.2.2 Tentatively Identified Radionuclides

Because radionuclides are not included on the Target Compound List (TCL), they may be classified as tentatively identified compounds (TICs) under Contract Laboratory Program (CLP) protocols. In reality, however, radioanalytical techniques are sufficiently sensitive that the identity and quantity of radionuclides of potential concern at a site can be determined with a high degree of confidence. In cases where a

radionuclide's identity is not sufficiently well-defined by the available data set: (1) further analyses may be performed using more sensitive methods, or (2) the tentatively identified radionuclide may be included in the risk assessment as a contaminant of potential concern with notation of the uncertainty in its identity and concentration. A health physicist or radiochemist should review the identification of any radionuclide to determine if the radionuclide is actually present or is an artifact of the sample analysis.

3.2.3 Detection and Quantitation Limits

The terms used to describe detection limits for radioanalytical data are different than the terms used for chemical data. Detection limits must be specified by the equations and confidence limits desired as well as being defined numerically. Normally, detection limits will be requested as the detection limits with a 5% chance each of Type I and Type II errors. Exhibit 1 lists typically achievable sensitivity limits for routine environmental monitoring.

In order to satisfy these purposes, two concepts are used. The first level is an estimated detection limit that is related to the characteristics of the counting instrument. This limit is not dependent on other factors in the analytical method or the sample characteristics. The limit, termed the lower limit of detection (LLD), is analogous to the instrument detection limit (IDL). The second limit corresponds to a level of activity that is practically achievable with a given instrument, analytical method, and type of sample. This level, termed the minimum detectable concentration (MDC), is analogous to the sample quantitation limit (SQL) and is the most useful for regulatory purposes.

3.2.4 The Estimated Lower Limit of Detection

The LLD may be defined on the basis of statistical hypothesis testing for the presence of activity. This approach is common to many authors and has been described extensively (Pasternack and Harley 1971, Altshuler 1963, Currie 1968, NCRP 1978).

The LLD is an *a priori* estimate of the detection capabilities of a given instrument system. This limit is based on the premise that from a knowledge of the background count and measurement of system parameters (e.g., detection efficiency), an *a priori* limit can be established for a particular measurement. The LLD considers both the α and β errors. In statistical hypothesis testing, α and β are the probabilities for what are frequently referred to as Type I (false detection) and

EXHIBIT 1. EXAMPLES OF TYPICAL MINIMUM DETECTION CONCENTRATION (MDC) VALUES FOR ENVIRONMENTAL RADIOANALYSES*

Media	Approximate Sample Size	Isotope	MDC	Reporting Units	Method ^a
Soil	200 grams	¹³⁷ Cs	1	pCi/g (dry)	1
	200 grams	⁶⁰ Co	1	pCi/g (dry)	1
	200 grams	²²⁶ Ra	0.1	pCi/g (dry)	1
	10 grams	⁹⁰ Sr	1	pCi/g (dry)	2
	10 gram	U Isotopes	0.1	pCi/g (dry)	3
	10 gram	Th Isotopes	0.1	pCi/g (dry)	3
	10 gram	Pu Isotopes	0.1	pCi/g (dry)	3
Water	50 mls	³ H	400	pCi/L	4
	4 liters	¹³⁷ Cs	1	pCi/L	1
	4 liters	⁶⁰ Co	1	pCi/L	1
	1 liter	²²⁶ Ra	0.1	pCi/L	5
	1 liter	⁹⁰ Sr	1	pCi/L	2
	1 liter	U Isotopes	0.1	pCi/L	3
	1 liter	Th Isotopes	0.1	pCi/L	3
	1 liter	Pu Isotopes	0.1	pCi/L	3
Air	300 m ³	¹³⁷ Cs	0.01	pCi/m ³	1
	300 m ³	⁶⁰ Co	0.01	pCi/m ³	1
	300 m ³	²²⁶ Ra	0.01	pCi/m ³	5
	300 m ³	⁹⁰ Sr	0.05	pCi/m ³	2
	300 m ³	U Isotopes	0.0002	pCi/m ³	3
	300 m ³	Th Isotopes	0.0002	pCi/m ³	3
	300 m ³	Pu Isotopes	0.0002	pCi/m ³	3
Biota	1000 g (ash)	¹³⁷ Cs	1	pCi/Kg (wet)	1
	1000 g (ash)	⁶⁰ Co	1	pCi/Kg (wet)	1
	1000 g (ash)	²²⁶ Ra	1	pCi/Kg (wet)	1
	1000 g (ash)	⁹⁰ Sr	1	pCi/Kg (wet)	2
	1000 g (ash)	U Isotopes	0.1	pCi/Kg (wet)	3
	1000 g (ash)	Th Isotopes	0.1	pCi/Kg (wet)	3
	1000 g (ash)	Pu Isotopes	0.1	pCi/Kg (wet)	3

* For purposes of illustration only. Actual MDCs for listed radionuclides in the media shown will vary, depending on sample specific preparation and analytical variables.

- a) Methods
- 1 = High Resolution Gamma Spectrometry
 - 2 = Chemical Separation followed by Gas Proportional Counting
 - 3 = Chemical Separation followed by Alpha Spectrometry
 - 4 = Liquid Scintillation Counting
 - 5 = Radon Emanation

Type II (false non-detection) errors, respectively. A common practice is to set both risks equal and accept a 5% chance of incorrectly detecting activity when it is absent ($\alpha = 0.05$) and a 95% confidence that activity will be detected when it is present ($1 - \beta = 0.95$). The expression for the LLD becomes:

$$LLD = K * (4.65 * s_b)$$

where:

K = the proportionality constant relating the detector response (counts) to the activity, such as $K=1/e$, where e is an overall detection efficiency or $K=1/I_\gamma e_\gamma$, where I_γ is the photon emission probability per disintegration and e_γ is the detection efficiency for the photon

s_b = the estimated standard deviation of the background count (assumed to be equal to the standard deviation of the sample count near the LLD)

3.2.5 The Estimated Minimum Detectable Concentration

The MDC is a level of activity at which detection can be achieved practically by an overall measurement method. As distinguished from the LLD, the MDC considers not only the instrument characteristics (background and efficiency), but all other factors and conditions that affect the measurement. The MDC is also an *a priori* estimate of the activity concentration that can be achieved practically under a set of typical measurement conditions. These conditions include sample size, net counting time, self-absorption and decay corrections, chemical yield, and any other factors that comprise the activity concentration determination. The MDC is useful for establishing that some minimum overall measurement conditions are met. Any of several factors, such as sample size or counting time, may be varied to meet a specific MDC value. Exhibit 1 lists typical MDCs for radionuclides in several media.

Expressions for the MDC are similar to those for the LLD. For the MDC, the proportionality constant K would include not only the factors for the LLD but also the factors that relate the detector response (counts) to the activity concentration in a sample for a typical set of measurement conditions.

3.2.6 Media Variability Versus Measurement Error

Sampling and analysis variability and measurement error are two key issues involved in planning and

assessing data collection efforts. Part A, Exhibit 31 lists field quality control (QC) samples that are used in defining variation and bias. These QC sample types have similar purposes for radioactively contaminated samples with one exception. The trip blank is not required for radioactively contaminated samples because there is less likelihood of contamination from direct exposure to air than for samples of volatile organic chemicals. Confidence level, power, and minimum detectable relative difference are defined in Part A, Section 4.1, and these definitions also apply in radionuclide sampling.

3.2.7 Sample Preparation and Sample Preservation

Proper sample preparation and preservation are essential parts of any radioactivity sampling program. The sampling requirements must be specified in the SAP before sampling activities begin. Precise records of handling are required to ensure that data obtained from different locations or time frames are correctly compared.

The appropriateness of sample preparation is a function of the required analysis. Some examples of sample treatment to be avoided or performed with great care include:

- Aliquots of samples selected for H-3 should not be dried, ashed or acidified.
- Aliquots of samples selected for C-14 should not be ashed or leached with acid.
- Aliquots of samples selected for elements with volatile oxidized forms, such as Iodine, should not be treated with oxidizing acids.
- Aliquots of samples selected for Ra-226 analysis by gamma spectrometry should be dried, crushed and/or sieved, but an appropriate post-preparation holding time must be included to allow the attainment of equilibrium with radon daughters.
- Aliquots of samples selected for elements with volatilized forms at high temperatures (e.g., I, Cs, Ru) should not be ashed, or ashed with great care. A radiochemist or health physicist should be consulted on the proper handling of the samples from a specific site.

The requirements of sample preservation are determined by the required analysis as well as the chemical characteristics of the radionuclide to be analyzed. The purpose of preserving a sample is to maintain the

sample in the condition required for analysis between the time the sample is collected and the time the sample is analyzed. Many of the radiochemical species of interest behave like trace metals, and the preservation of water samples is easily achieved by acidification. This prevents metallic species from depositing on the walls of the container. Usually, nitric acid is used to maintain a pH of less than 2.0. Water samples preserved in this manner have a holding time of six months. The exceptions to this general rule are given below:

- Samples for H-3 and C-14 analysis should be unpreserved.
- Samples for analysis of elements with volatile oxidized forms (e.g., I-129, I-131) should not be preserved with oxidizing acids.
- Certain laboratories may require samples for uranium analysis to be preserved with hydrochloric acid.

The container material for stored samples can also be a factor in sample preservation. Metals have an affinity for glass when preserved with nitric acid. Iodine and transition metals such as iron and cobalt have shown an affinity for polyethylene and polypropylene under certain conditions (Bernabee 1980). The selection of containers for different sample types should be specified in the SAP.

Soil samples are generally collected and shipped to the analytical laboratory "wet," meaning their inherent moisture has not been deliberately removed. The SAP should address the questions regarding if, how (air or oven), and when (prior to or after aliquotting) the sample will be dried. Often, a soil sample contains much extraneous matter, e.g., root matter, rocks, stones, organisms. The question arises whether these "extraneous" materials are just that, or whether they constitute part of the sample itself. These issues should be specified in the analytical program design, and the risk assessor must ensure that sample preservation has not compromised the sample's integrity.

Samples of contaminated structural samples may be collected at some sites. For structural material the data may be reported as fixed or as removable contamination. Fixed contamination refers to contamination that is incorporated in the material or is firmly bound on the surface of the material. Fixed contamination is measured by cleaning the surface of the material and using a field survey instrument to measure the activity of the material. Removable contamination is contamination that can be transferred from the surface of the material to another object. Removable contamination is measured by smearing the surface of the material with a small piece of paper or cloth and measuring the amount of activity

on the smear. Special handling and analysis procedures for these types of samples should be included in the SAP.

The presence of radioactive and hazardous chemical wastes (mixed wastes) at a site can influence the quality of the analytical data obtained for that site. Two general areas are affected by the special considerations of mixed wastes. First, the radioactive nature of the waste necessitates special plans and operations for on-site measurements and sampling. Second, the radioactivity in the samples may limit the number of laboratories that can receive the samples or the types of analyses that can be performed. The nature of such influences is not always self-evident. Data users should be aware of the potential effects on data quality resulting from the complications of mixed waste characterization.

Field work demands that the on-site staff be able to make decisions at the job site, a necessary prerequisite if the sampling and measurement teams are to be capable of reacting to unforeseen circumstances. It is also true that in those circumstances, personnel tend to make judgments based on their best, most applicable experience. The experience of a worker who has handled hazardous wastes will be biased toward the chemical handling aspects, and decisions appropriate to those types of wastes are to be expected. The opposite may be true of workers experienced with handling radioactive materials. It will be up to the data user to critically review the field records to ensure that such on-site decisions properly considered the data validity of both sample components and that data were not compromised.

The design of the sample collection program may require compromises due to the differences in sample handling and staff experience required for the principal components of the waste. Mixed waste is only a small fraction of all the low-level radioactive waste generated in the country and an infinitesimal fraction of the total hazardous waste. Therefore, staff with the appropriate experience in both areas may not be available. The requirements for special training and staff may conflict with limitations in potential resources. Any given risk assessment may be required to use staff that are very experienced in one area (e.g., radiochemical sampling) but may have only minimal training in the other mixed waste component (e.g., sampling for organics). Data recipients need to be especially alert to potential problems caused by large discrepancies in the experience of staff working such programs.

The external exposure rates or radioactivity concentration of a specific sample may limit the time that workers will be permitted to remain in intimate contact with the samples. Possibly, collection personnel

could take large samples and then split them into specific analytical aliquots in a radioactively "cold" area. This area may be "cold" with respect to radioactive contamination but may still be contaminated chemically. This process increases both the chances of nonequivalent samples being sent for different analyses and the potential for cross-contamination between samples or from the area chosen for sample splitting. Additionally, external exposure rates from individual samples may require that smaller samples be taken and special holding areas be provided. Special handling requirements may conflict with the size requirements for the analytical protocol, normal sampling procedures, or equipment. For example, sampling for hazardous waste constituents or properties may require that samples be kept refrigerated. Samples containing radioactive materials may have to be kept in a restricted area to prevent personnel radiation exposure or the spread of alpha and/or beta contamination. The shielding requirements for radioactive samples depend on their external exposure rate, and confinement is based on the potential for removable contamination. Such decisions will be made by site health physics (HP) personnel who may be unaware of temperature or holding time requirements. In some cases, samples will have to be physically surrendered to HP personnel for clearance prior to removal from the site. Again, data recipients need to be alert for potential handling errors arising from these types of situations.

Varying requirements for storage, preservation, and special shipping complicate the logistics of mixed waste programs. While most radiochemical procedures have holding times and preservation methods in common with metals analysis, they differ greatly with organic analyses. Holding times for radioactively contaminated samples are also affected by the half-life of the radionuclide to be analyzed. After seven half-lives, less than 1% of the original activity would remain in the sample. Separate samples should be taken for the analyses requiring different handling and preservation.

Less obvious is the potential for biasing sampling programs by selecting samples that can be safely handled or legally shipped to the support laboratories. There will be a human bias in the direction of handling samples with the least shipping and storage complications. This selection process can involve several assumptions about the waste distribution which may or may not be acknowledged. In an effort to ship the most convenient samples, workers may assume that the chemical contamination is not related to the radioactivity levels in any way. The assumptions may also be made that there are no qualitative differences in the radioactivity content at different concentrations and that the low activity samples can be quantitatively

analyzed and scaled to the higher activity areas by the use of a simple ratio, of external exposure rates, for example. Without documentary support, all of these assumptions may be unwarranted, and sampling and analysis schemes based on such assumptions may compromise data integrity. The risk assessor must ensure that such assumptions were not part of the sample selection process by reviewing the appropriate plans and records.

3.2.8 Fixed Laboratory Versus Field Analysis

Fixed laboratory and field analyses are compared in Part A, Section 3.2.9. A major factor to be considered in this decision for radioactively contaminated sites is the type of radiation present. Alpha-emitting radionuclides often cannot be measured in the field because of the attenuation of the alpha particles by the sample matrix. Attenuation can also cause problems for beta measurements under certain conditions. Gamma-emitting radionuclides can generally be measured in the field if the data can be confirmed by fixed laboratory measurements.

■ *Field measurements must be made using instruments sensitive to the type of radioactivity present.*

Selection of a radiometric method depends on the number of radionuclides of interest and their activities and types of radiations emitted, as well as on the level of sensitivity required and the sample size available. Exhibit 2 provides information on field survey instruments for measuring gamma radiation, including the advantages and disadvantages associated with each type of instrument. Exhibit 3 provides similar information for alpha and beta field survey instruments.

Measurements of external gamma radiation exposure rates are used to delineate areas of contamination and areas of observed contamination. Exposure rates are usually measured with hand-held radiation survey meters that utilize ion chambers, Geiger-Muller (G-M) tubes, or gamma scintillation probes.

Surface gamma readings provide data only on radiation levels at the surface, and they may miss contamination from radionuclides at a greater depth that are shielded by soil cover. In order to accurately characterize the depth distribution of the radioactive contamination, boreholes are augured or driven through key areas of the site. Detectors, generally gamma scintillators, are lowered into these boreholes, and readings of the gamma exposure rate or gamma count-rate are obtained at regular predetermined depths. Exhibit 4 shows a typical borehole apparatus. The risk assessor should consider several issues pertaining to down-hole gamma profiling.

EXHIBIT 2. FIELD SURVEY INSTRUMENTS FOR MEASURING GAMMA RADIATION

Detection	Specifications	Advantages	Disadvantages
Ion Chamber	<ul style="list-style-type: none"> Moderate to high range, approximately 0-2,000 mR/hour. Accuracy $\pm 5\%$ at the high end of the scale. 	<ul style="list-style-type: none"> Reading is directly proportional to radiation field. Suitable for use in high radiation fields. Very portable. 	<ul style="list-style-type: none"> Poor sensitivity, not adequate for near-background radiation rates.
Pressurized Ion Chamber (PIC)	<ul style="list-style-type: none"> Range 1-500 μR/hour. Accuracy $\pm 5\%$ full scale. 	<ul style="list-style-type: none"> Suitable for near-background radiation rates. Reading is directly proportional to radiation field. 	<ul style="list-style-type: none"> Not as portable as Ion Chamber, therefore, fewer measurements per day can be recorded.
"Modern" Geiger-Muller (GM) Tube	<ul style="list-style-type: none"> Moderate to high range: 0-5,000 mR/hour. Accuracy $\pm 10\%$ full scale. 	<ul style="list-style-type: none"> Very portable. Can also be used for beta radiation detection. 	<ul style="list-style-type: none"> Poor sensitivity, not adequate for near-background radiation rates. Reading is not directly proportional to radiation field unless an energy compensated tube is used.
Gamma Scintillation Detectors	<ul style="list-style-type: none"> Low range 0-5,000 μR/hour. Accuracy $\pm 10\%$ at high end to $\pm 30\%$ at low end of scale. 	<ul style="list-style-type: none"> Suitable for background radiation rates. Very portable. 	<ul style="list-style-type: none"> Reading is not directly proportional to radiation field; response varies with energy.
Organic Scintillators	<ul style="list-style-type: none"> Low range 0-25 μR/hour. Accuracy $\pm 10\%$ full scale. 	<ul style="list-style-type: none"> Suitable for background radiation rates. Very portable. 	<ul style="list-style-type: none"> Response is generally linear with energy.

EXHIBIT 3. SURVEY INSTRUMENTS FOR MEASURING ALPHA AND BETA RADIATION

Detection	Radiation Detected	Advantages	Disadvantages
Alpha Scintillation Probe*	<ul style="list-style-type: none"> alpha only 	<ul style="list-style-type: none"> High detection efficiency. Useful for many screening applications. Very portable. 	<ul style="list-style-type: none"> Delicate window may be easily broken. Measures only alpha particles.
Air Proportional Detector	<ul style="list-style-type: none"> alpha only 	<ul style="list-style-type: none"> Large surface area. High detection efficiency. 	<ul style="list-style-type: none"> Delicate window may be easily broken. Measures only alpha particles. Can be affected by moisture.
Geiger-Muller (GM) Pancake Type Probe*	<ul style="list-style-type: none"> alpha, beta and gamma 	<ul style="list-style-type: none"> Large surface area. Can be used to detect all types of radiation. Good for general screening. 	<ul style="list-style-type: none"> Sensitivity to all types of radiation decreases ability to discriminate between radiation types.
Side-Shielded GM Probe*	<ul style="list-style-type: none"> beta and gamma 	<ul style="list-style-type: none"> Discriminates between gamma and beta radiation. Good in high gamma radiation fields. 	<ul style="list-style-type: none"> Gamma reading is not directly proportional to radiation field; response varies with energy.

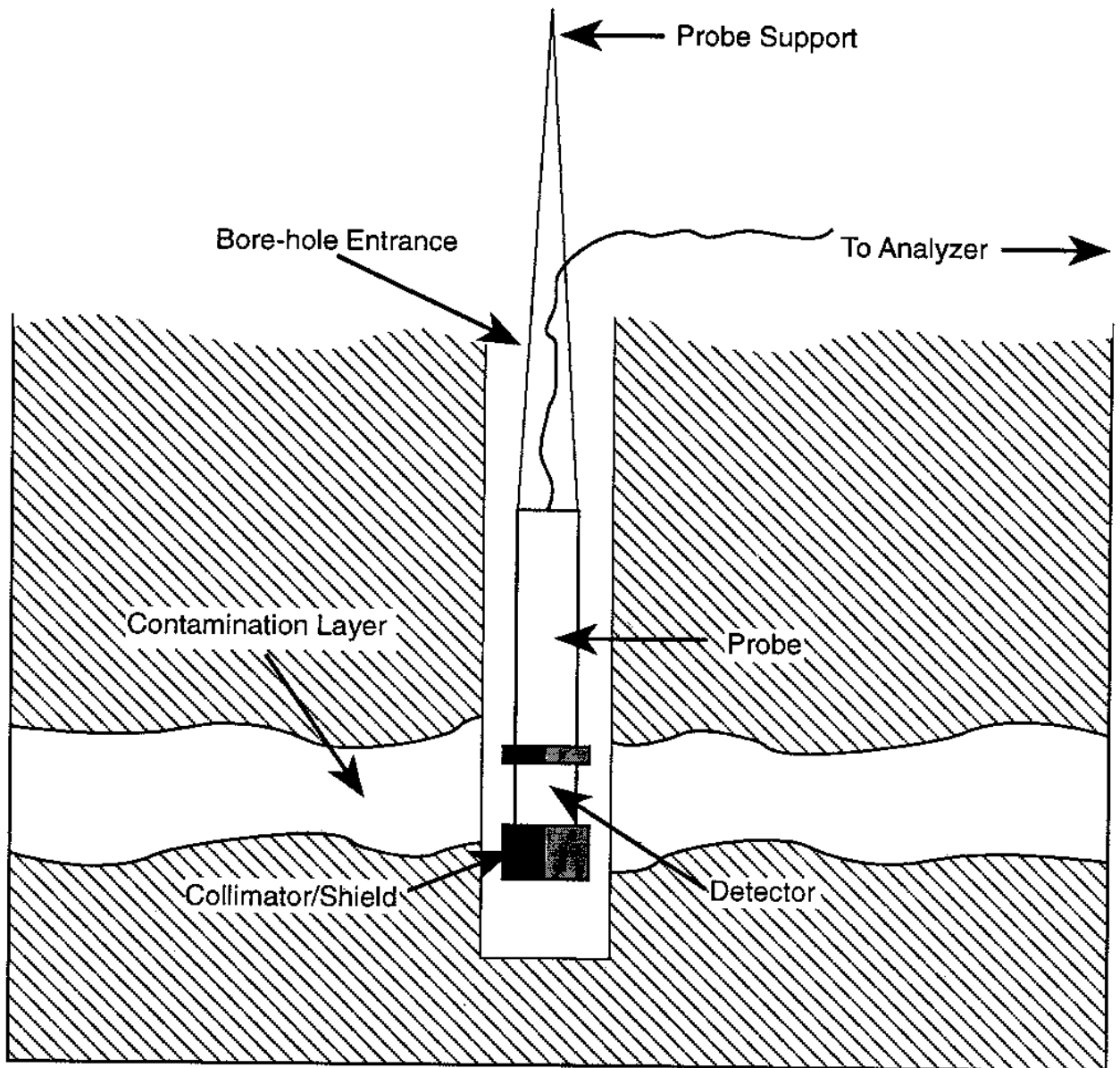
* All probes are attached to the appropriate rate meter or scaler.

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These include the calibration conditions for the detector, the energy range the instrument is set to measure, and variations in background caused by heterogeneous layers of naturally occurring radioactivity.

Alpha and beta radiations lack the penetrating ability and range of gamma radiation, making their detection in the field more difficult, but equally important, to characterize. Preliminary radiation screening of samples for alpha- or beta-emitting radionuclides must be

EXHIBIT 4. ILLUSTRATION OF BORE-HOLE GAMMA PROFILING



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performed using instruments sensitive to the type of radiation being measured and must be performed much closer to the contamination source. These results, usually referred to as screening, can be used to identify samples or areas containing radioactive contamination,

to establish that all samples leaving the site comply with applicable U.S. Department of Transportation (DOT) regulations, and to estimate the radioactivity content of samples sent off site for analysis to ensure compliance with the recipients radioactive materials license limits.

Chapter 4

Steps in Planning for the Acquisition of Useable Environmental Data

This chapter provides guidance to the RPM and the risk assessor for designing an effective sampling plan and selecting suitable analytical methods to collect environmental data for use in baseline risk assessments. Part A, Chapter 4 contains worksheets that can be used to assist the risk assessor or RPM in designing an effective sampling plan and selecting the proper analytical methods.

4.1 STRATEGIES FOR DESIGNING SAMPLING PLANS

The discussion in Part A, Section 4.1 regarding sample location, size, type, and frequency applies to radioactively contaminated sites as well. However, the resolution and sensitivity of radioanalytical techniques permit detection in the environment of most radionuclides at levels that are well below those that are considered potentially harmful, while analytical techniques for nonradioactive chemicals are usually not this sensitive. For radionuclides, continuous monitoring of the site environment is important, in addition to the sampling and monitoring programs described in Part A, Section 4.1. Many field devices that measure external gamma radiation, such as high pressure ionization chambers, provide a real time continuous record of radiation exposure levels. Such devices are useful for determining the temporal variation of radiation levels at a contaminated site and for comparing these results to the variability observed at background locations. Continuous measurements provide an added level of resolution for quantifying and characterizing radiological risk.

Additional factors that affect the frequency of sampling for radionuclides include the half-lives and the decay products of the radionuclides. Radionuclides with short half-lives, such as I-131 (half-life = 8.04 days), have to be sampled more frequently because relatively high levels of contamination can be missed between longer sampling intervals. The decay products of the radionuclides must also be considered, because their presence can interfere with the detection of the parent nuclides of interest, and because they also may be important contributors to risks.

The Sampling Design Selection Worksheet shown in Exhibit 5 may be used to assist in the design selection for the most complex environmental situation, which is usually soil sampling. This worksheet is similar to the worksheet found in Part A, Exhibit 45. Directions for filling out the worksheet can be found in Part A, Section

4.1.2. The worksheet should be completed for each medium and exposure pathway at the site. Once completed, this initial set of worksheets can be modified to assess alternative sampling strategies.

There are two details to keep in mind while filling out the worksheet:

- Providing expedited sampling and analysis when radionuclides with short half-lives are a concern.
- Increasing reliance on field survey data in all aspects of planning, since field data often provide easy identification of many radionuclides and guide sample collection.

Since field duplicates and blanks are such an important determinant of measurement error precision, careful attention must be paid to the number that are collected. Part A, Exhibit 48 provides the number of duplicate pairs of QC samples required to obtain a specific confidence level.

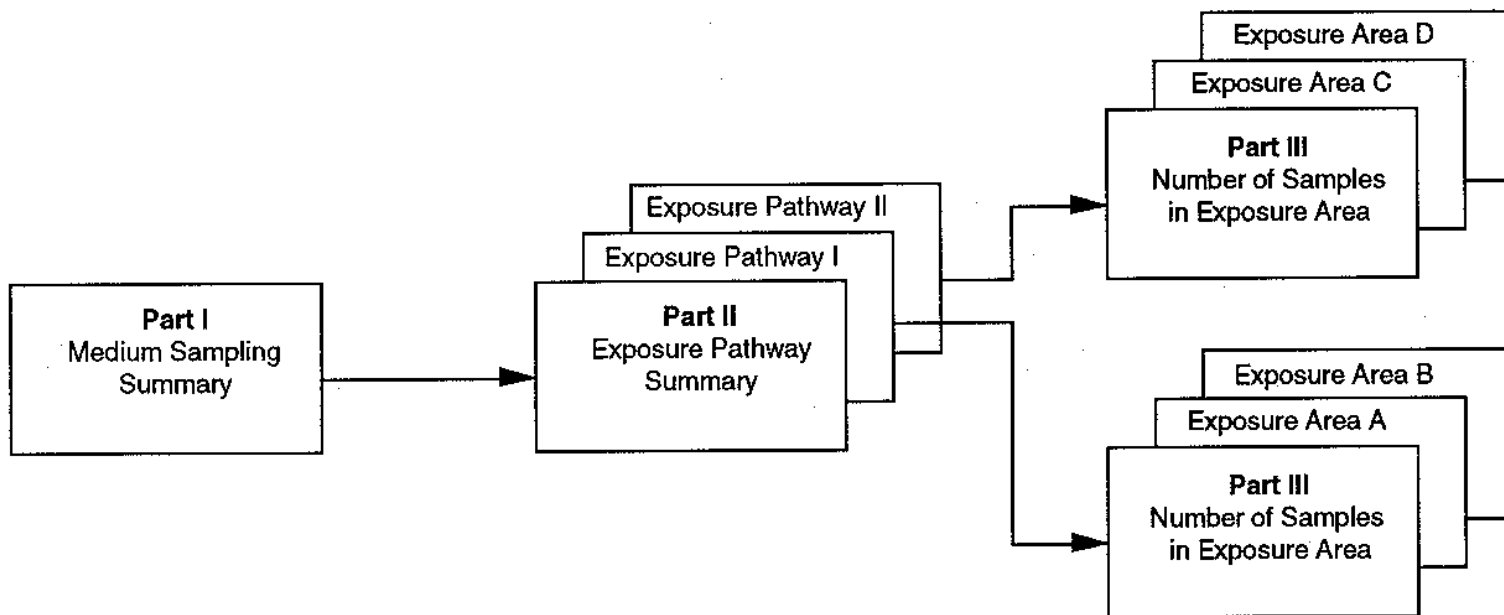
4.1.1 Determining the Number of Samples

An important aspect in designing a sampling plan is the number of samples required to fully characterize each of the three exposure pathways. Several methods for

Acronyms

CLP	Contract Laboratory Program
DQO	data quality objective
EMSL/LV	Environmental Monitoring Systems Laboratory/Las Vegas
NAREL	National Air and Radiation Environmental Laboratory
NESHAPs	National Emission Standards for Hazardous Air Pollutants
NIST	National Institute of Standards and Technology
ORP/LVF	Office of Radiation Programs/Las Vegas Facility
PRP	potentially responsible party
QA	quality assurance
QAP	Quality Assurance Program
QC	quality control
RPM	remedial project manager
SAP	sampling and analysis plan
SDWA	Safe Drinking Water Act
USNRC	U.S. Nuclear Regulatory Commission

**EXHIBIT 5. HIERARCHICAL STRUCTURE OF SAMPLING DESIGN
SELECTION WORKSHEET**



**EXHIBIT 5. PART II: EXPOSURE PATHWAY SUMMARY
SAMPLING DESIGN SELECTION WORKSHEET
(Cont'd)**

H. Radionuclide of Potential Concern and CAS Number	I. Frequency of Occurrence	J. Estimation		K. CV	L. Background
		Arithmetic Mean	Maximum		

M. Code (CAS Number) of Radionuclide of Potential Concern Selected as Proxy _____

N. Reason for Defining New Stratum or Domain (Circle one)

1. Heterogeneous Radionuclide Distribution
2. Geological Stratum Controls
3. Historical Information Indicates Difference
4. Field Screening Indicates Difference
5. Exposure Variations
6. Other (specify) _____

O. Stratum or Exposure Area		Q. Number of Samples from Part III					
Name and Code	P. Reason	Judgmental/ Purposive	Back- ground	Statistical Design	Geo- metrical or Geo- statistical Design	QC	Row Total
R. Total (Part I, Step F):							

EXHIBIT 5. PART III: EXPOSURE AREA SUMMARY SAMPLING DESIGN SELECTION WORKSHEET (Cont'd)

O. Stratum or Exposure Area _____ Domain Code _____
 E. Medium/Pathway Code _____ Pathway Code _____

S. Judgmental or Purposive Sampling
 Comments: _____

Use prior site information to place samples, or determine location and extent of contamination. Judgmental or purposive samples generally cannot be used to replace statistically located samples.

An exposure area and stratum MUST be sampled by at least TWO samples.

Number of Samples

T. Background Samples
 Background samples must be taken for each medium relevant to each stratum/area. Zero background samples are not acceptable. See the discussion on pp. 74-75 of Guidance for Data Useability in Risk Assessment Part A.

Number of Background Samples

U. Statistical Samples
 CV of proxy or radionuclide of potential concern _____
 Minimum Detectable Relative Difference (MDRD) _____ (<40% if no other information exists)
 Confidence Level _____ (>80%) Power of Test _____ (>90%)

Number of Samples
 (See formula in Appendix IV)

V. Geometrical Samples
 Hot spot radius _____ (Enter distance units) _____
 Probability of hot spot prior to investigation _____ (0 to 100%)
 Probability that NO hot spot exists after investigation _____ (enter only if >75%)
 (see formula in Appendix IV)

W. Geostatistical Samples
 Required number of samples to complete grid +
 Number of short range samples

X. Quality Control Samples
 Number of Duplicates (Minimum 1:20 environmental samples) _____
 Number of Blanks (Minimum 1 per medium per day or 1 per sampling process, whichever is greater) _____

Y. Sample Total for Stratum
 (Part II, Step U)

Judgmental/ Purposive	Back- ground	Statis- tical Design	Geo- metrical or Geo- statistical	QC	Row Total

determining the required number of samples are available, including the method discussed in Part A, Chapter 4 and Part A, Appendix IV. Alternative methods have been proposed by Schaeffer, et. al. (Schaeffer 1979) and Walpole and Meyers (Walpole 1978).

Each of the three exposure pathways from different sample media present separate problems in designing a sampling plan. A full discussion of sampling problems is beyond the scope of this guidance. A brief discussion of sampling soil, groundwater, and air pathways is included as an example for a typical 10-acre site. The number of samples and sampling locations listed are the minimum number of samples required, and these numbers will increase for most applications. The area of consideration, the time available for monitoring, the potential concentration levels of the contaminants, and the funding available all influence the number of samples to be analyzed.

Measurements of external exposure from soil are taken with portable instruments as described in Section 3.2, usually at 1 meter above ground level. The initial measurements will be performed at predetermined grid intersections, typically at intervals of 50 feet or 20 meters. This spacing produces about 20 to 25 measurements per acre. Larger spacing could be used when surveying larger areas, especially if the contamination is expected to be widespread and evenly distributed at a constant depth below the surface. Conversely, the distance between measurements would decrease if the initial readings indicate contamination that is localized or particularly elevated relative to background. The primary objective in both cases is to collect enough data to determine the locations of maximum gamma radiation and to indicate zones of equal intensity (i.e., isopleths) around these points. This results in the familiar "bullseye" drawings indicating areas of suspected maximum contamination. Gamma exposure data are essential in selecting the locations for soil sampling and borehole surveys. For a typical 10-acre site, upwards of 250 radiation measurements will be required. These data are normally superimposed on a map or figure for ease of interpretation. The data should indicate where background readings were obtained for all sides of the site. Sources of radium activity will decay to radon gas. The radon gas is more mobile and can travel under the ground to give elevated surface readings where there is no source of radioactivity. When the radium source is removed the radon sources disappear. In these situations borehole surveys and a qualified health physicist or radiochemist can be used to help interpret the data.

Borehole surveys involve the use of a gamma-sensitive probe which is lowered into drilled or driven holes as

described previously. Measurements of gamma count rate are made at predetermined depth intervals, typically every 6 inches. A site investigation may produce 100 or more borehole surveys. Depths of each hole will normally extend at least 1 foot beyond the bottom of the contaminated layer. When grade levels are approximately equal, boreholes normally terminate at the same depth. Therefore, boreholes showing no evidence of contamination should have penetrated to at least the same depth as those showing contamination. Practically speaking, borehole depths vary across a site as a function of the site characteristics and the sampling equipment used.

Exhibit 6 illustrates the need for borehole measurements. Surface surveys cannot detect contamination occurring at a great depth. Overlying soil cover which shields the radioactivity may produce a greatly reduced response at the surface. Depth profiles also provide a means for selecting soil sampling locations and are useful in prioritizing radiochemical analyses. This information can also be used to correlate data for non-gamma-emitting radionuclides to field surface radiation measurements.

Both surface soil composites and core samples from a subset of the locations selected by borehole profiling should be collected. Subsurface soil cores should be collected from 10 to 20% of the boreholes at a minimum of approximately 12 locations. The distribution of soil sample locations should be as follows:

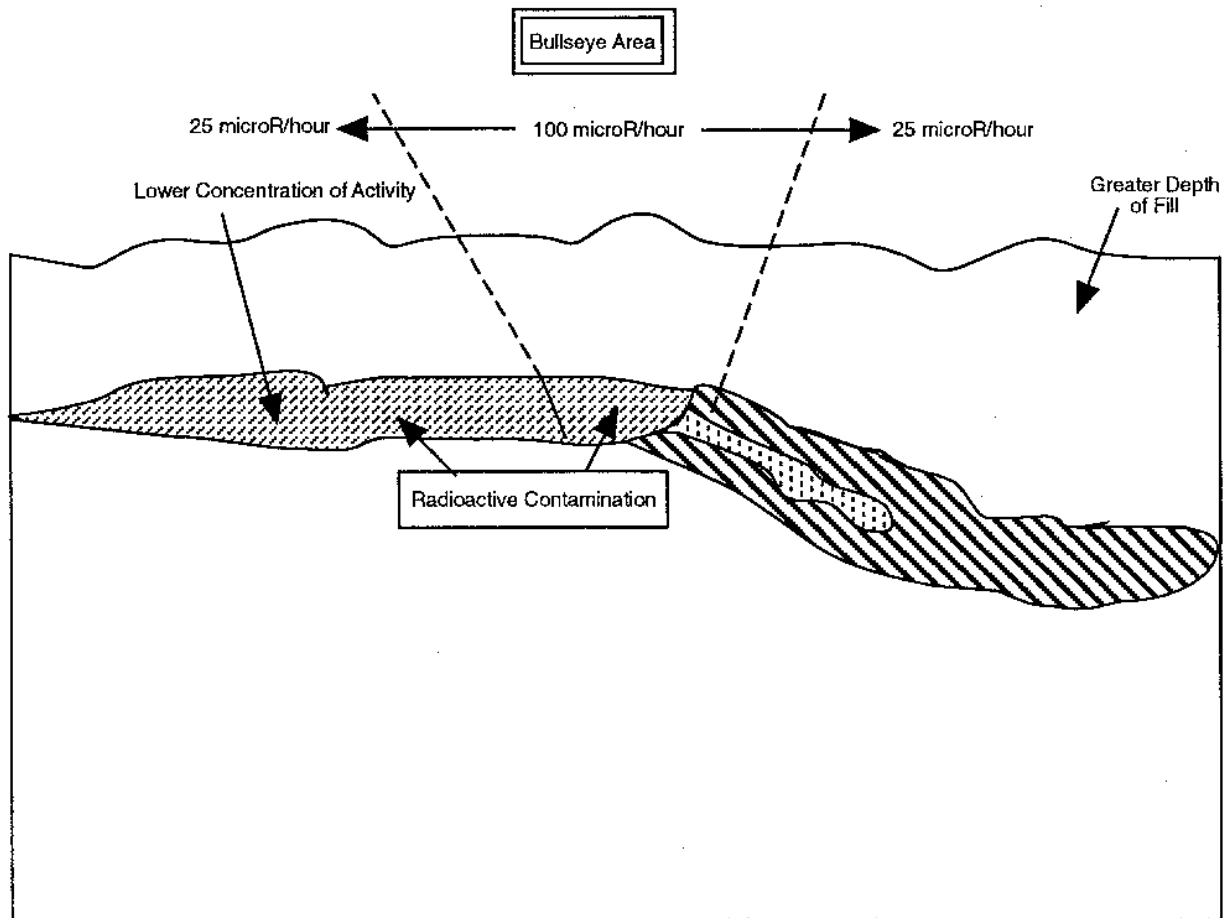
- Three from background locations.
- Three from hot spot ("bullseye") locations identified in the surface radiation survey.
- Three from locations defining the limits of the hot spots.
- Three defining the fringes or boundaries of the contaminated zone.

Soil cores are normally split into 6-inch increments. These cores can also be combined and analyzed as a composite, when resources are of critical importance. Borehole samples are taken to provide information concerning the extent of the contamination as well as the depth of the contamination.

Compositing of borehole samples can result in misinterpretation of the results when contamination varies with depth across the area being investigated.

Groundwater samples should be taken from a minimum of four locations: two background and two indicator locations. If the sampling locations were chosen in the absence of knowledge of the groundwater flow patterns,

EXHIBIT 6. EFFECT OF SOURCE DEPTH ON SURFACE GAMMA RADIATION MEASUREMENTS



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close inspection of comparative data is required to ensure that background samples are not potentially contaminated. Without knowledge of the groundwater flow, background samples may be collected on opposite sides of the site. If the groundwater flow is perpendicular to the line between these two locations, both are likely to be true backgrounds. If the flow is parallel to this line, one or the other may be contaminated. Contamination of both "background" samples may suggest local flow reversal or contamination from sources other than the site under investigation. A thorough data evaluation should indicate the true nature of the situation.

Air samples should be collected from a minimum of six locations. At least two of these should be background locations. To achieve the required sensitivity for environmental analyses, approximately 300 m³ will be required. Occasionally, a specific isotope may require

special collection efforts. For example, tritium will normally not be collected on filters but on silica gel or other absorbers, and sampling for gases usually requires special equipment and techniques. These special circumstances should be described in the sampling and analysis plan (SAP). The choice of filter material is also important; it is determined by flow rate, the size of the particulate matter being sampled, and the expected loading of the filter during the sampling time. In general, membrane filters are used for low flow rates to detect small amounts of submicron particles, while paper or glass fiber filters are used for larger flow rates and larger particles. Some filter materials contain large amounts of naturally occurring radioactivity (i.e., K-40 in glass fiber filters) and will not be applicable in certain situations.

A maximum of 10 to 12 samples per site can be expected from other sources as indicators of an ingestion pathway.

These may be surface water, sediment, benthic organisms, fish or other indicators. A minimum of two background samples per media should also be collected.

4.2 STRATEGY FOR SELECTING ANALYTICAL METHODS

Currently, there is no single, universally accepted compilation of radiochemical procedures. However, there is a preferred priority of procedures (although developed or approved for other applications) that can be applied to risk assessments.

In general, where the Agency has mandated or recommended radiochemical analytical procedures for compliance with other programs, those procedures should be considered for the same or analogous media when analyzing samples for risk assessments. A key factor in method selection is the constraints that were established during the data quality objective (DQO) process. Exhibit 7 summarizes a preferred order of method selection.

Media-specific procedures are as follows:

Water. Procedures mandated for compliance with the Safe Drinking Water Act (SDWA) should be used for analysis of both surface and groundwater samples for analytes specified in the SDWA. Procedures for analytes not specifically mentioned in the SDWA may be selected from the other compendia listed in Exhibit 8.

Air samples. The National Emission Standards for Hazardous Air Pollutants (NESHAPs): Radionuclides (40 CFR 61 Appendix B) includes methods for the analysis of radioactivity in air samples. This appendix presents both citations of procedures for specific isotopes

and general "principles of measurement." The general principles are similar to the counting methods discussed previously. Where the analyte/media combinations match those pathways under investigation at a site, the applicable individual method should be used. When a specific isotope is not mentioned, methods utilizing the appropriate principles of measurement in concert with appropriate QA/QC procedures will be acceptable.

Soil, sediment, vegetation, and benthos. A number of procedures exist that contain methods for the analysis of soil, sediment, and biological media for a variety of radionuclides. Compendia for these procedures are listed in Exhibit 8 and provide ample resources for the selection of analytical methods.

In general, whether the procedures are selected from the SDWA, NESHAPs, or one of the other suggested compilations, the procedures are subject to many limitations. Some procedures assume the presence of only the isotope of interest; some assume the absence of a specific interfering isotope. Procedures involving dissolution or leaching may assume that the element of interest is in a specific chemical form. Careful attention to the conditions and limitations is essential both in the selection of radiochemical procedures and in the interpretation of data obtained from those procedures. If the user is unsure of the applicability of a method to a candidate site or specific situation, assistance can be obtained from the Regional Radiation Representative, Office of Radiation Programs, or radiochemistry staff at the National Air and Radiation Environmental Laboratory in Montgomery, Alabama (NAREL), the Office of Radiation Programs/Las Vegas Facility (ORP/LVF), or the Office of Research and Development-Environmental Monitoring Systems Laboratory in Las Vegas, Nevada (EMSL/LV).

EXHIBIT 7. ORDER OF PRIORITY FOR SELECTION OF ANALYTICAL METHODS

- Methods Required by EPA Regulations (e.g., NESHAPs or NPDWR)
- Methods Published by EPA Laboratories (e.g., NAREL, Montgomery, AL or EMSL, Las Vegas, NV)
- National Consensus Standards (e.g., ASTM, APHA, IEEE)
- Methods Published by Other Federal Agencies (e.g., DOE, USGS)
- Methods Published in Refereed Technical Literature
- Methods Published by Other Countries or International Organizations (e.g., IAEA, NRPB)

EXHIBIT 8. REFERENCES FOR RADIOCHEMICAL PROCEDURES

- American Public Health Association, "Methods of Air Sampling", 2nd Edition, APHA, New York, NY (1977).
- American Society for Testing Materials, "1987 Annual Book of ASTM Standards", ASTM, Philadelphia, PA.
- APHA/AWNA/WPCF, "Standard Methods for the Examination of Water and Wastewater", 17th Ed., APHA, Washington, DC.
- Department of Energy, "RESL Analytical Chemistry Branch Procedures Manual", IDO-12096, VSDOE, Idaho Falls, ID.
- Department of Energy, "EML Procedures Manual", 26th Edition, Report EML-300, USDOE, New York, NY.
- Environmental Protection Agency, "Radiochemical Analytical Procedures for Analysis of Environmental Samples", EMSL-LV-0539-17, USEPA Environmental Monitoring and Support Laboratory, Las Vegas, NV.
- Environmental Protection Agency, "Radiochemistry Procedures Manual", EPA 520/15 84-006, EERF, Montgomery, AL.
- Environmental Protection Agency, "Indoor Radon and Radon Decay Product Measurement Protocols", EPA 520/1-89-009, USEPA, Washington, DC.

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4.2.1 Selecting Analytical Laboratories

• *The shipper of radioactive material is responsible for ensuring that the recipient is authorized to receive the shipped material and for compliance with all applicable shipping and labelling regulations.*

The risk assessor needs to be aware of limitations placed on the samples by regulatory or licensing considerations due to the sample's radioactivity content. Adherence to existing regulations is an obvious requirement. Radioactively contaminated sites are likely to generate samples that may be receivable only by laboratories having an appropriate license to handle radioactive materials. Such licenses may be issued by state agencies or the U.S. Nuclear Regulatory Commission (USNRC). In either case, the shipper is responsible for ensuring that the recipient is authorized to receive the shipped material and is responsible for complying with all applicable shipping and labeling regulations (DOT, etc.). Two prerequisites must be filled to permit the shipper to fulfill this obligation:

- A copy of the recipient laboratory's current valid radioactive materials license must be obtained prior to shipment of any samples and be available

to the shipper at the location of sample packaging and shipment.

- The shipper must have adequate field measurement equipment available at the site to ensure that samples are within license limits.

Laboratories may have license limits which are specified either on a per sample basis or for the facility as a whole. When facility limits are imposed, the laboratory should be requested to provide its administrative limits on individual samples or sample batch lots. While these requirements do not directly affect the data, compliance with these requirements can be complicated and time-consuming and may interfere with holding times or other analytical requirements. The risk assessor should review the procedures used to comply with these requirements to ensure that such compliance will not affect data integrity.

Many radiochemistry laboratories may not be prepared to associate individual sample data with specific analytical batches. Efficiency calibrations, backgrounds, analytical blanks, instrument performance checks, and other QC parameters all can have varying frequencies and therefore apply to different time periods and different analytical batches. The traditionally applied data qualifiers may not have direct analogues in

radiochemistry or may require alternate interpretation. When receiving data from a mixed waste laboratory which has historically developed from a radiochemistry laboratory, the risk assessor will be required to evaluate different relationships between QC and samples that are typical for non-radiochemical data.

The conventions for the use of data qualifiers are closely tied to data reporting requirements. QA/QC programs for radiochemical laboratories have developed separately with a different emphasis. The emphasis for chemical analysis has been to coordinate the QC data with batches of analyses within fairly narrow time periods. Radiochemical measurement methods emphasize QC data collection based on measurement systems, due to the stability of properly maintained systems and the count-time intensive nature of the analyses. It is not unusual for single measurements to monopolize a given instrument for several hours. It is, therefore, impractical to rerun standard curves at frequent intervals, since other methods of establishing instrument and method performance have been devised.

The probability that non-Contract Laboratory Program (CLP) data or potentially responsible party (PRP) data may have to be used for evaluation will be greater for sites that have more serious mixed waste considerations. Consideration of non-CLP data usage is discussed in Chapter 5. In addition, not all methods may be available for every sample. Availability of a specific method depends on contamination levels and types and levels of containment available at the laboratory. Not all equipment may be available for every level of containment and shielding. It is possible that different equipment or methods may be used for the same parameter in samples with different levels of radioactive contamination. Personnel protection restrictions may limit exposure rates from individual or batch analytical aliquots. Resulting limitations on sample size may be reflected in limitations on the achievable detection limits.

Laboratories performing radiochemical analyses should have an active and fully documented Quality Assurance Program (QAP) in place. There are several documents that provide guidance for the preparation of a QAP. Some of these documents include Test Methods for Evaluating Solid Wastes (SW846) (EPA 1986), United States Nuclear Regulatory Commission Regulatory Guide 4.15 (NRC 1977), United States Department of Energy Environmental Survey Manual (DOE 1988), and ANSI/ASME NQA-1 (ASME 1989). The procurer of radioanalytical services should specify the type of QAP that is required and should be prepared to evaluate programs in such formats. The following are the criteria

that are common to these documents and should be considered as the minimum requirements of an adequate QAP:

Quality Assurance Program. The QAP must be written and must state the QA policy and objectives for the laboratory. The primary function of QA/QC is the definition of procedures for the evaluation and documentation of the sampling and analytical methodologies and the reduction and reporting of data. The objective of QA/QC is to provide a uniform basis for sample handling, sample analysis, instrument and methods maintenance, performance evaluation, and analytical data gathering.

Organizational structure. The laboratory should maintain an organizational document defining the lines of authority and communication for reporting relationships. This document should include job descriptions of management and staff, including a QA officer.

Qualifications of personnel. Qualifications of personnel performing quality related tasks should be specified and documented, including resumes, education level, previous training, and satisfactory completion of proficiency testing.

Operating procedures and instructions. Written instructions and/or procedures covering the administrative, operations, and quality levels of the laboratory should be established and include, but are not limited to:

- Sample collection.
- Sample receipt and shipping.
- Analytical methods.
- Radioactive material handling.
- Radioactive waste disposal.
- Data verification.
- Software quality assurance.
- Sample preparation and storage.
- Procurement.
- Quality assessment.
- Chain-of-custody.
- Review of procedures.
- Data evaluation.
- Reporting of data.
- Records.

- Audits.
- Implementation of inter- and intralaboratory QC program.
- Calibration and operation of laboratory instruments.
- Performance checks and maintenance of laboratory instruments.
- Preparation and standardization of carrier and tracer solutions.

The following are criteria that should be considered as additional requirements for an environmental sampling program:

Design control. The laboratory should maintain a document defining the flow path of samples through the laboratory, including sample receipt, sample log-in, sample analysis and measurement, data validation and processing, reporting, and records management.

Inter- and intralaboratory analyses. Reagent blanks, matrix blanks, field (equipment) blanks, field duplicates (splits), laboratory duplicates, blind and double blind matrix spikes, and verification (reference) standards should constitute at least 10% of the samples analyzed. The actual numbers of each type of analysis should be specified in the SAP.

Appropriate QC testing should be included in the work plan for projects other than the established, routine services supplied by the analytical laboratory.

The laboratory should assure that measuring and testing devices used in activities affecting quality are of the proper range, type, and accuracy to verify conformance to established requirements. To assure accuracy, measuring and test equipment should be controlled, calibrated, adjusted, and maintained at prescribed intervals as specified by procedures. Calibrations should be performed using standards or systems that are traceable to the National Institute of Standards and Technology (NIST). If no national standards exist, the basis for calibration should be documented. The method and interval of calibration for each item should be defined. The specifications should be based on the type of equipment, stability characteristics, required accuracy, and other conditions affecting measurement control. Additional routine checks of baseline or background characteristics and performance checks should be made on frequencies appropriate for each instrument with such frequencies established in approved procedures.

Each of the above situations places a greater burden on the risk assessor to perform a careful review. Professional judgment is required to assess the final effect of varying methods, equipment, aliquot sizes, and QA/QC activities on the analytical results.

Chapter 5

Assessment of Environmental Data for Useability in Baseline Risk Assessments

This chapter provides guidance for the assessment and interpretation of environmental radioanalytical data for use in baseline human health risk assessments. Data assessment is accomplished by examining two general sets of data. One set of data consists of the data supporting the individual analysis. Questions often asked of these data include:

- Were all the correct parameters used?
- Were the specified methods used?
- Were all controlled parameters maintained within specified limits?
- Were the calculations performed correctly?
- Do the final analytical results make sense in light of the site history and results obtained for other samples?
- Are the analytical results legally defensible if enforcement activity or cost recovery activity is to be pursued by EPA?

The second set of data supports the validity of the method and proper operation and calibration of measurement equipment. This set of data comprises instrument calibration, operational checks, method demonstration and cross-check programs, and routine QC samples. Both sets of data need to be examined to judge the validity of individual analyses.

To evaluate radioanalytical data, it is necessary to understand the normal methods of calculating radiochemical values for activity concentration, error, minimum detectable concentration (MDC), and lower limit of detection (LLD). Generalized equations for these calculations are given in Exhibits 9 and 10. These equations contain the parameters used to calculate the radioactivity in a given sample. Although not all parameters will be used in every radioanalysis, these equations will serve as the basis for the following discussion of individual parameters. This discussion assumes the user has specified, received, or can obtain access to the data shown in Exhibit 11.

Activity, error, and detection limits are the parameters generally reported by radioanalytical laboratories. Activity, which is the estimate of radioactivity in a sample, may be a screening parameter (e.g., gross alpha) or isotope specific (e.g., Sr-90). Activity must always be calculated from a net count-rate because all radioactivity measurement systems are subject to background count-rates from cosmic radiation, the laboratory environment, and their own construction materials, among other sources.

Error terms are usually reported based on counting statistics only. While Equation 2 in Exhibit 9 calculates a single standard deviation, it is common practice to report radiochemical data to two standard deviations. To determine whether two analytical results are significantly different, it is important to know the number of standard deviations to which the reported errors correspond.

A standard radiochemical data report should include values for the activity concentration and the associated error, or the MDC. The data user must ensure that the MDC value is in fact sample specific, and not a generalized value. Some laboratories report the activity concentration and associated error only when the sample is above the sample-specific MDC. Others will report the activity concentration and associated error even when the results are less than zero (negative). The reporting conventions should be decided prospectively and the requirements communicated to the analytical laboratory.

The risk assessor must evaluate the radioanalytical data for completeness and appropriateness and to determine if any changes were made to the work plan or the sampling and analysis plan (SAP) during the course of the work. The risk assessor will assess the radioanalytical data for completeness, comparability, representativeness, precision, and accuracy as described in Part A, Chapter 5.

Acronyms

EPA	U.S. Environmental Protection Agency
LLD	lower limit of detection
MDC	minimum detectable concentration
QC	quality control
SAP	sampling and analysis plan

EXHIBIT 9. GENERALIZED EQUATIONS FOR RADIOACTIVITY CALCULATIONS

$$\text{ACT} = \frac{\frac{\text{SC}}{\text{ST}} - \frac{\text{BC}}{\text{BT}}}{2.22 \times 10^6 \times \text{EFF} \times \text{CY} \times \text{ALI} \times \text{RY} \times \text{DIFs}} \quad (1)$$

$$\text{ERR} = \frac{\sqrt{\frac{\text{SC}}{\text{ST}^2} + \frac{\text{BC}}{\text{BT}^2}}}{2.22 \times 10^6 \times \text{EFF} \times \text{CY} \times \text{ALI} \times \text{RY} \times \text{DIFs}} \quad (2)$$

$$\text{MDC} = \frac{4.65 \times \sqrt{\frac{\text{BC}}{\text{BT} \times \text{ST}}}}{2.22 \times 10^6 \times \text{EFF} \times \text{CY} \times \text{ALI} \times \text{RY} \times \text{DIFs}} \quad (3)$$

$$\text{LLD} = \frac{4.65 \times \sqrt{\frac{\text{BC}}{\text{BT} \times \text{ST}}}}{2.22 \times 10^6 \times \text{EFF} \times \text{RY}} \quad (4)$$

Where:

ACT	=	Activity in units of microCuries per units of ALI
ERR	=	One standard deviation counting error (Same units as ACT)
MDC	=	Minimum detectable concentration (Same units as ACT)
LLD	=	Lower limit of detection in units of microCuries at time of counting
SC	=	Total sample counts
ST	=	Elapsed time for which sample was counted (minutes)
BC	=	Total background counts
BT	=	Elapsed time for which background was counted (minutes)
2.22×10^6	=	Number of disintegrations per minute (dpm) per microCurie
EFF	=	Counting efficiency for radiation being measured (counts per minute detected for each disintegration per minute actually occurring in sample)
ALI	=	Aliquot of sample actually analyzed (units of volume or mass)
CY	=	Yield of the radiochemical separation procedure (fractional unit of recovery)
RY	=	Radiation yield (number of radiations of the type being measured which are produced per each disintegration which occurs. For gamma spectrometry this is commonly called gamma abundance.)
DIFs	=	Product of various decay and ingrowth factors. The most commonly used DIFs are shown in Exhibit 10.

**EXHIBIT 10. GENERALIZED EQUATIONS FOR RADIOACTIVITY
DECAY AND INGROWTH CORRECTION FACTORS**

$$DFA = e^{-\frac{0.693}{HLA} \times T_1} \quad (5)$$

$$DFC = \frac{\frac{0.693}{HLA} \times T_2}{1 - e^{-\frac{0.693}{HLA} \times T_2}} \quad (6)$$

$$IDF = 1 - e^{-\frac{0.693}{HLD} \times T_3} \quad (7)$$

$$DFD = e^{-\frac{0.693}{HLD} \times T_4} \quad (8)$$

Where:

- DFA = Decay correction to obtain activity at the end of the sampling period (continuous collection) or at the time of collection (grab sample)
- DFC = Corrects average count rate during acquisition to count rate at beginning of counting
- IDF = Calculates fraction of the decay product ingrowth for radiochemical methods where the decay product is the entity actually counted
- DFD = Corrects for decay of the decay product between the end of ingrowth and beginning of counting
- HLA = Half-life for isotope of interest
- HLD = Half-life of the decay product (if the decay product is isotope counted)
- T₁ = Time interval between end of sampling and beginning of counting
- T₂ = Elapsed time for acquisition of sampling counts
- T₃ = Time permitted for ingrowth of the decay product activity
- T₄ = Time interval between last separation of parent and the decay product isotopes and the beginning of counting of the decay product.

EXHIBIT 11. DATA REPORT REQUIREMENTS FOR TYPICAL RADIOCHEMICAL ANALYSIS

The following are the minimum parameters required on a radiochemical analytical report to recreate and verify the analytical report.

- Lab Sample ID
- Field Sample ID
- Start Collection Time/Date
- Stop Collection Time/Date
- Flow Rate
- Volume/Weight Adjustment Factors
- Aliquot Analyzed (Vol/Wgt)
- Chemical Yields
- Start and Stop Times and Dates for the Sample Count
- Total Sample Acquisition Time
- Start and Stop Times and Dates for the Background Count
- Total Background Acquisition Time
- Energy Regions of Interest
- Uncorrected Gross Sample Counts
- Gross Background Counts
- Gamma Abundance Values
- Counter Efficiency
- Sample Specific Correction Factors
- Start and Stop Times & Dates for Decay Product Ingrowth
- Start and Stop Times & Dates for Radioactive Decay

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Chapter 6

Application of Data to Risk Assessment

This chapter discusses the application of radioanalytical data for risk assessment. Guidance is provided for reviewing data for consistency and completeness and for evaluating observed contamination, source term quantity, and contamination levels. Because similarities exist between the evaluation and application of analytical data for radioactive and nonradioactive risk assessment, the reader is encouraged to review the discussions provided in Part A, Chapter 6.

Before radioanalytical data can be used for risk assessment, the user must determine the acceptability and usefulness of the data sets derived from the field and laboratory analyses. The data user should then review the entire data package for consistency and completeness among the data sets. At a minimum, this review should focus on the following areas:

- Radionuclides of concern.
- Discrimination of site contamination from background.
- Exposure pathways.
- Documentation of analytical procedures and results.

6.1 RADIONUCLIDES OF CONCERN

The data user should review the list of radionuclides of concern for each migration pathway for completeness with respect to the criteria listed in Section 3.2:

- Atomic number and atomic weight.
- Radioactive half-life.
- Principal decay modes, radiation decay modes, energies, and abundances.
- Chemical and physical form.
- Decay products.

6.2 DISCRIMINATION OF SITE CONTAMINATION FROM BACKGROUND

Radionuclide specific activity concentrations (and radiation exposure rates, where applicable) for background samples are required for each pathway. These data are used to characterize the naturally occurring levels of radionuclides in all pertinent media and to facilitate discrimination of site contamination from background. These data need to be of sufficient quality

for risk assessment purposes. Data quality depends on whether background levels were determined by site-specific analysis or were derived from the literature. In general, site-specific background data are recommended over values obtained from the literature because site-specific measurements can account for the local background variability, and the quality of site-specific analytical data can be directly assessed through the use of QA/QC samples.

Care must be taken to ensure that the appropriate background sample is taken for each analytical sample, and that the background sample is the equivalent of the analytical sample. It must originate in the same conditions of an uncontaminated area, e.g., the same soil classification as a borehole sample taken on site, but from an environmentally uncontaminated area.

When published data are used to establish background concentrations, the data must be determined to be representative of the site. The concentration utilized to represent the background should be in the 95% upper confidence limit of the range of literature data.

Ideally, both site-specific data and that from the literature should be available and utilized to draw comparisons between and conclusions about the quality of background concentration data. Reported background values for a specific radionuclide in a given medium that fall outside (i.e., either below or above) the concentration range expected from values in the literature, should alert the data user to the need to review the appropriateness or representativeness of the background sampling location or the performance and sensitivity of sampling and analysis techniques, radiochemical procedures, or measurement techniques.

6.3 EXPOSURE PATHWAYS

The risk assessor should review the data package to ensure that all relevant exposure pathways have been sampled and that radioanalytical data are provided for these pathways. For example, evaluation of the soil exposure pathway should include measurements of activity concentrations of radionuclides in soil, as well as external radiation exposure measurements from all

Acronyms

QA	quality assurance
QC	quality control
SAP	sampling and analysis plan
SOP	standard operating procedure

contaminated areas. The locations of all background and site sampling points should be clearly defined and marked on the site map.

6.4 DOCUMENTATION OF ANALYTICAL PROCEDURES AND RESULTS

All radioanalytical procedures used to determine site data should be documented. These procedures and resulting data sets should be reviewed to determine whether the proper procedures were used for the types, abundances, and energies of the radiations emitted by each radionuclide and should ensure that the data are presented in the appropriate activity concentration units (e.g., pCi/g dry weight or pCi/g wet weight for soil, pCi/L for water, pCi/g fresh weight or pCi/g dry weight or

pCi/g ash weight for vegetation, or pCi/m³ for air), along with their associated error. The required activity concentration units should be specified in the sampling and analysis plan (SAP).

To document radiochemical results properly, a detailed compilation of supporting documentation is required. Records of all types should be continuous. Data originally recorded in a notebook may be transferred to a form, entered into a computer, and finally printed as either input parameters or as intermediate, calculated data. In these cases, copies of all supporting logbooks and forms are required, not just the final printed copy. To support the reported analytical data, a broad range of documentation should be required of the analytical laboratories. The materials required for QA support documentation are shown in Exhibit 12.

EXHIBIT 12. RADIOCHEMICAL QUALITY ASSURANCE SUPPORT DOCUMENTATION

Sample Collection Data:

- Field survey data
- Sample collection field logs
- Field preparation data sheets
- Shipping/transmittal forms
- Chain-of-Custody forms
- Sample receipt logs
- Sample login forms/logs
- Laboratory analysis request and distribution forms
- Calibration data for sample collection equipment
- Radiation screening information
- Copy of NRC/State RAM license of party receiving samples

Analytical Data:

Preparation/Chemistry Data

- Sizes of aliquots processed
- Concentration/dilution factors
- Chemical yield data
- Evidence of preparation of counting aliquots
- Dates and times of processing and separations
- Analogous data for applicable QC samples
- Initials of the analyst(s)
- Copy of SOPs used for preparation

Counting Data

- Sample sizes and counting geometries
- Sample counts
- Background counts
- Reagent blank counts
- Acquisition times, sample & background
- Date and times of all counting
- Counter efficiencies
- Identification of analysts
- Identification of counters used
- Counter printouts, including but not limited to peak search and quantitation printouts for spectral methods
- Counter crossover and interference data (GPC)
- Analogous data for appropriate QC samples
- Calculated results, propagated errors, detection limits

**EXHIBIT 12. RADIOCHEMICAL QUALITY ASSURANCE
SUPPORT DOCUMENTATION
(Cont'd)**

Instrument Data:

Performance Data

- Instrument backgrounds
- Efficiency checks
- Check source documentation
- Energy calibration/resolution checks (spectrometry)
- Plateau checks (gas proportional counters)
- Logs and control charts of these data
- Acceptance criteria
- Corrective actions taken and the bases for same

Instrument Calibrations

- Standards preparation and traceability
- Calculation of efficiencies
- Supporting counting data
- Quench correction curves (LSC)
- Acceptance criteria
- Efficiency vs Energy curves (HRGS or NaI)
- Transmission Factor curves (GPC)
- Energy vs. Channel plots (spectrometry)
- Corrective actions taken and bases for same

Quality Control Data:

- Results and supporting raw data for scheduled blanks, replicates and reference samples
- Results and supporting raw data for blind blanks, replicates and reference samples
- Results and supporting raw data for participation in interlaboratory programs
- Control charts of above data
- Acceptance criteria
- Corrective actions taken and bases for same

The following procedures and supporting information may be submitted once, either at the project inception or prior to contract award:

- Official or controlled copies of all procedures used to acquire, preserve and ship samples; perform the above analyses; and calculate results
- Calculation and reporting conventions
- Algorithms used to calculate the submitted data
- Verification of software program results
- Qualifications for all analysts

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APPENDIX I

Glossary of Radiation Concepts, Terminology and Units

Absorbed dose (D) is the mean energy imparted by ionizing radiation per unit mass of material (e.g., biological tissue). The SI unit of absorbed dose is the joule per kilogram, also assigned the special name the gray (1 Gy = 1 joule/kg). The conventional unit of absorbed dose is the rad (1 rad = 100 ergs per gram = 0.01 Gy).

Activity refers to the average number of nuclear disintegrations of a radioisotope that occur per unit time. It is the product of the number of atoms and the radioactive decay constant, λ , of a given radioisotope, and can be defined as follows:

$$A = \lambda N$$

where A is the activity of the radioisotope in units of disintegrations per second (dps) or disintegrations per minute (dpm), N is the number of atoms present at a specified time, and λ is the decay constant in reciprocal units of time (i.e., sec^{-1} or min^{-1}), defined as:

$$\lambda = \frac{\ln(2)}{T_{1/2}} \approx \frac{0.693}{T_{1/2}}$$

where $T_{1/2}$ is the radioactive half-life of the radioisotope. Further, the activity of a radioisotope alone (i.e., unsupported by the decay of another radioisotope) can be calculated at any point in time t based on the activity present at some initial time $t = 0$ and on its decay constant, as follows:

$$A(t) = A_0 e^{-\lambda t}$$

where $A(t)$ is the activity of the radioisotope at time t and A_0 is the initial activity of the isotope at $t = 0$. Quantities of radioactive isotopes are typically expressed in terms of activity at a given time t (see the definitions for Becquerel, Curie, counts per minute, and disintegrations per minute).

Atomic number is the number of protons in the nucleus of an atom. In its stable and neutral state, an atom has the same number of electrons as it has protons. The number of the protons determines the atom's chemical properties. For example, an atom with one proton is a hydrogen atom, and an atom with 92 protons is a uranium atom. The number of neutrons of an atom may vary in number without changing its chemical properties, only its atomic weight.

Atomic weight is the total number of neutrons and protons in the nucleus of an atom.

Becquerel (Bq) is the SI unit of activity defined as the quantity of a given radioisotope in which one atom is transformed per second (i.e., one decay per second or 1 dps). One Bq is equal to 2.7E-11 Ci.

Committed dose equivalent ($H_{T,50}$) is the integral of the dose equivalent in a particular tissue for 50 years after intake (corresponding to a working lifetime) of a given radionuclide.

Cosmogenic radionuclides are those radionuclides (e.g., H-3 and C-14) continually produced by natural cosmic processes in the atmosphere and not by the decay of naturally occurring series radionuclides.

Counting efficiency is the ratio of the number of counts registered by a given radiation-detection instrument each minute (i.e., cpm) over the number of nuclear disintegrations per minute of the radioactive source (dpm) being measured. For example, given a source decaying at a rate of 1,600 dpm and an instrument that detects 400 cpm, then the counting efficiency of this detection system would be 0.25 ($400/1,600 = 1/4$) or 25%.

Counts per minute (cpm) is the unit that describes the number of disintegrations detected by a radiation-detection instrument. Because radiation is emitted isotropically (i.e., equally in all directions) from a radioactive source, the probes of most radiation-detection instruments cannot detect all radiation emitted from a source. Therefore, cpm and dpm will not be equal. However, if the response characteristics of a detector are known for a given radiation source, the relation between cpm and dpm can be determined (see Counting efficiency).

Curie (Ci) is the conventional unit of activity defined as the quantity of a given radioisotope that undergoes nuclear transformation or decay at a rate of 3.7×10^{10} (37 billion) disintegrations each second. One Ci is equal to 3.7×10^{10} Bq and approximately equal to the decay rate of one gram of Ra-226. Because the curie is a very large amount of activity, subunits of the curie are often used:

1 millicurie (mCi)	=	10^{-3} Ci
1 microcurie (μ Ci)	=	10^{-6} Ci
1 nanocurie (nCi)	=	10^{-9} Ci
1 picocurie (pCi)	=	10^{-12} Ci
1 femtocurie (fCi)	=	10^{-15} Ci

Disintegrations per minute (dpm) is the unit that describes the average number of radioactive atoms in a source disintegrating each minute. A 500 dpm source, for example, will have 500 atoms disintegrating every minute on the average. One picocurie (pCi) equals approximately 2.22 dpm.

Dose equivalent (H) considers the unequal biological effects produced from equal absorbed doses of different types of radiation and is defined as:

$$H = DQN$$

where D is the absorbed dose, Q is the quality factor that considers different biological effects, and N is the product of any modifying factors. Quality factors currently assigned by the International Commission on Radiological Protection (ICRP) include Q values of 20 for alpha particles, 10 for protons, and 1 for beta particles, gamma photons, and x-rays. Q values for neutrons depend on their energies and may range from 2 for thermal neutrons to 11 for 1 MeV neutrons. These factors may be interpreted as follows: On the average, an alpha particle will inflict approximately 20 times more damage to biological tissue than a beta particle or gamma ray, and twice as much damage as a neutron. The modifying factor is currently assigned a value of unity ($N=1$) for all types of radiation. The SI unit of the dose equivalent is the sievert (Sv), and the conventional unit is the rem ($1 \text{ rem} = 0.01 \text{ Sv}$). A commonly used subunit of the rem is the millirem (mrem).

Electron Volt (eV) is the unit used to describe the energy content of radiation, defined as the energy acquired by any charged particle carrying a unit (electronic) charge when it falls through a potential of 1 volt; it is equivalent to 1.6×10^{-12} ergs. Alpha particles range in energy from 1 to 10 million electron volts (MeV), and beta particles are emitted over a wide energy range from a few thousand electron volts (keV) to a few MeV. Gamma photons also typically range from a few keV to one to two MeV.

Effective dose equivalent (H_E) and the **committed effective dose equivalent ($H_{E,50}$)**, defined as the weighted sums of the organ-specific dose equivalents, were developed by the ICRP to account for different cancer induction rates and to normalize radiation doses and effects on a whole body basis for regulation of occupational exposure. In general, the reader need not be concerned with these concepts for HRS scoring purposes. Still, the interested reader is referred to ICRP publications (ICRP 1977 and ICRP 1979) for additional information on these topics.

Exposure (sometimes called the **exposure dose**) refers to the number of ionizations occurring in a unit mass of air due to the transfer of energy from a gamma or x radiation field. The unit of exposure is the **roentgen (R)** expressed as coulombs of charge per kilogram of air ($1 \text{ R} = 2.58 \times 10^{-4} \text{ C/kg}$). A common simplification is that 1 R of gamma or x-radiation is approximately equal to 1 rad of absorbed dose and to 1 rem of dose equivalent.

Exposure rate (or **exposure dose rate**) refers to the amount of gamma or x-ray radiation, in roentgen, transferred to air per unit time (e.g., R/hr or R/yr). Commonly used subunits of the roentgen are the milliroentgen ($1 \text{ mR} = 10^{-3} \text{ R}$) and the microroentgen ($\mu\text{R} = 10^{-6} \text{ R}$), with corresponding subunits of mR/hr or $\mu\text{R/hr}$ for exposure rates. The roentgen may be used to measure gamma or x radiation only.

External exposure refers to radiation exposure from radioactive sources located outside of the body.

Gray (Gy) is the SI unit of absorbed dose ($1 \text{ Gy} = 1 \text{ Joule kg}^{-1} = 100 \text{ rad}$).

Internal exposure refers to radiation exposure from radionuclides distributed within the body.

ICRP is the International Commission on Radiological Protection.

Ionization of an atom is the removal of one of its orbital electrons. When an electron is removed, two charged particles, or ions, result: the free electron, which is electrically negative, and the rest of the atom, which bears a net positive charge. These are called an **ion pair**. Radiation is one mechanism that produces ionization. Alpha and beta radiation cause ionization primarily through collisions, that is, moving alpha and beta particles physically "collide" with orbital electrons, transferring some or all their energy to these electrons. Multiple collisions with electrons eventually reduce the energy of the alpha or beta particle to zero. These particles are then either absorbed or stopped. De-energized beta particles become free electrons that often are absorbed by positive ions. A doubly-positive alpha particle frequently captures two free electrons to become a helium atom. Gamma radiation causes ionization by three processes: the photoelectric effect, the Compton effect, and pair production. The photoelectric effect occurs when the total energy of the gamma photon is absorbed by an electron and the incident gamma photon is annihilated. The Compton effect occurs when part of the energy of the gamma photon is transferred to an orbital electron and the initial incident gamma photon is deflected with reduced energy. In pair production, the incident gamma photon interacts with the atomic nucleus forming two electrons and the photon is annihilated. Because of their ability to remove orbital electrons from neutral atoms, alpha, beta, and gamma radiation are referred to as **ionizing radiation**.

Isotopes are atoms of the same chemical element that have the same number of protons but different numbers of neutrons. All isotopes of a given element have the same atomic number but different atomic weights.

Naturally occurring radionuclides are those radionuclides of primordial origin and terrestrial nature which possess sufficiently long half-lives to have survived in detectable quantities since the formation of the earth (about 3 billion years ago), with their radioactive decay products.

Rad is the conventional unit of absorbed dose ($1 \text{ rad} = 100 \text{ ergs/g of tissue} = 0.01 \text{ Gy}$).

Radiation (specifically, Ionizing Radiation) refers to the energy released in the form of particles (i.e., alpha, beta, or neutrons), electromagnetic waves (i.e., gamma photons and x rays), or both, during the radioactive decay of an unstable atom.

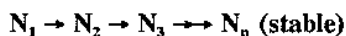
Radioactivity is the property of an unstable atom of a radioactive element whereby the atom transforms (decays) spontaneously by emission of radiation into an atom of a different element. Radioactive properties of unstable atoms are determined by nuclear considerations only and are independent of their physical or chemical states.

Radioactive contamination is commonly used to describe radioactive atoms that are unconfined or in undesirable locations.

Radioactive decay is the process whereby an unstable nucleus of a radioactive atom ejects one or more particles (i.e., alpha, beta, or neutrons) from its nucleus to establish a more stable state. These particles are sometimes accompanied by a release of electromagnetic energy (i.e., gamma or x ray radiation). Together, ejected particles and released energy are called **radiation**. Radioactive decay results in the formation of an atom of a different element called a **decay product (progeny or daughter)** which also may be radioactive. There are three principal modes of radioactive decay: alpha, beta, and neutron.

- **Alpha decay** occurs when the neutron to proton ratio is too low and, because of this instability, the unstable nucleus ejects an **alpha particle** (alpha radiation). An alpha particle has two protons and two neutrons. Emission of an alpha particle from an atom decreases its atomic weight by four and its atomic number by two. Thus, the new atom of another element has two fewer protons and two fewer neutrons and its chemical properties are different from those of its parent element. It too may be radioactive. For example, when an atom of radium-226 (with 88 protons and 138 neutrons) emits an alpha particle, it becomes an atom of radon-222 (with 86 protons and 136 neutrons), a gas. Since radon-222 is also radioactive, it too decays and forms an atom of still another element. Alpha particles are somewhat massive and carry a double positive charge. They can be completely attenuated by a sheet of paper.
- **Beta decay** occurs when an electrically neutral neutron splits into two parts, a proton and an electron. The electron is emitted as a **beta particle** (beta radiation) and the proton remains in the nucleus. The atomic number of the resulting decay product is increased by one, and the chemical properties of the progeny differ from those of its parent. Still, the atomic weight of the decay product remains the same since the total number of neutrons and protons stays the same, that is, a neutron has become a proton, but the total number of neutrons and protons combined remains the same. Beta particles will penetrate farther than alpha particles because they have less mass and only carry a single negative charge. Beta radiation can be attenuated by a sheet of aluminum.
- **Neutron decay** occurs during nuclear fission reactions, resulting in the emission of a **neutron**, two smaller nuclei, called fission fragments, and beta and gamma radiation. In general, neutron-emitting radionuclides are unlikely to be encountered or of much concern at most Superfund sites.
- **Gamma radiation** may accompany alpha, beta, or neutron decay. It is electromagnetic energy emitted from the atomic nucleus and belongs to the same wave family as light, radio waves, and x rays. X rays, which are extra-nuclear in origin, are identical in form to gamma rays, but have slightly lower energies. Gamma radiation can be attenuated by heavy material such as concrete or lead.

Radioactive Decay Series or Chains are radionuclides which decay in series. In a decay series, an unstable atom of one radioisotope (the parent isotope) decays and forms a new atom of another element. This new atom may, in turn, decay to form a new atom of another element. The series continues until a stable or very long-lived atom is formed. At that point, the decay chain ends or is stopped. The number of radionuclides in a series varies, depending upon the number of transformations required before a stable atom is achieved. This process can be illustrated as follows:



where N_1 is the number of atoms of the parent radioisotope decaying to form atoms of the first decay product, N_2 , which in turn decays to form atoms of the second decay product, N_3 , which continues to decay until a stable atom, N_n , is formed. Examples of important naturally occurring decay series include the uranium series, the thorium series, and actinium series. There are three major reasons why it is important to identify decay series and to characterize the properties of each decay product in those series:

- First, the total activity content (and the potential hazard) of a radioactive source may be substantially underestimated if the activity contributions from each of the decay products are not included. If it is assumed incorrectly that only one radionuclide of potential concern is present in a source when, in fact, one or more decay products also may be present, then the total activity of and threat posed by that source may not be considered completely;
- Second, decay products may be more toxic, either alone or in combination, than the parent nuclide. Because each radioactive isotope possesses its own unique chemical, physical, and radioactive properties, the hazard presented by decay products may be substantially greater than that posed by the parent nuclide alone.
- And third, the environmental fate, transport, and bioaccumulation characteristics of the decay products may be different from those of the parent nuclide. All relevant migration pathways for both the parent nuclide and decay products must be considered to account for site threats.

Radioactive equilibrium refers to the activity relationship between decay series members. Three types of radioactive equilibrium can be established: secular, transient, and no equilibrium. **Secular equilibrium** refers to the state of equilibrium that exists when series radioisotopes have equal and constant activity levels. This equilibrium condition is established when the half-life of the parent isotope is much greater than that of its decay product(s) (i.e., $T_{1/2}$ of the parent $\gg T_{1/2}$ of the decay product, or when expressed in decay constants, $\lambda_2 \gg \lambda_1$). **Transient equilibrium** is the state of equilibrium existing when the half-life of the parent isotope is slightly greater than that of its decay product(s) (i.e., $T_{1/2}$ of the parent $> T_{1/2}$ of the decay product, or $\lambda_2 > \lambda_1$) and the daughter activity surpasses that of the parent. **No equilibrium** is the state that exists when the half-life of the parent isotope is smaller than that of the decay product(s) (i.e., $\lambda_2 < \lambda_1$). In this latter case, the parent activity will decay quickly, leaving only the activity of the decay product(s).

Radioactive half-life ($T_{1/2}$) (sometimes referred to as the **physical half-life**) is the time required for any given radioisotope to decrease to one-half its original activity. It is a measure of the speed with which a radioisotope undergoes nuclear transformation. Each radioactive isotope has its own unique rate of decay that cannot be altered by physical or chemical operations. For example, if one starts with 1,000 atoms of iodine-131 (I-131) that has a half-life of 8 days, the number of atoms of I-131 remaining after 8 days (one half-life), 16 days (two half-lives), and 24 days (three half-lives) will be 500, 250, and 125, respectively. In fact, the fraction of the initial activity of any radioisotope remaining after n half-lives can be represented by the following relationship:

$$\frac{A}{A_0} = \frac{1}{2^n}$$

where A_0 is the initial activity and A is the activity left after n half-lives. After one half-life ($n=1$), 0.5 (or 50%) of the initial activity remains; after three half-lives ($n=3$), 13% remains; and after five half-lives ($n=5$), 3% remains. Further, the activity of any radioisotope is reduced to less than 1% after 7 half-lives. For radioisotopes with half-lives greater than six days, the change in activity in 24 hours will be less than 10%. Over 1,600 different radioisotopes have been identified to date, with half-lives ranging from fractions of a second to billions of years.

Radioactive isotopes (radioisotopes or radionuclides) are radioactive atomic variations of an element. Two radioactive isotopes of the same element have the same number of protons but different numbers of neutrons. They share common chemical properties, but exhibit different and unique radioactive, and possibly physical, properties because of the differences in their respective nuclear stabilities and decay modes.

Radionuclide slope factor is the lifetime excess cancer incidence rate per unit intake of (or per unit exposure to) a given radionuclide.

Rem is the acronym for roentgen equivalent man and is the unit of dose equivalent (1 rem = 0.01 Sv).

Roentgen (R) is a unit of external exposure which refers to the number of ionizations occurring in a unit mass of air due to the transfer of energy from a gamma or x radiation field emitted by a radioactive source. The unit is expressed as coulombs of charge per kilogram of air (1 R = 2.58×10^{-4} C/kg). Commonly used subunits of the roentgen are the milliroentgen (mR = 10^{-3} R) and the microroentgen (μ R = 10^{-6} R), with corresponding subunits of mR/hr or μ R/hr for exposure rates. The roentgen may be used to measure gamma or x radiation only. [See Exposure and Exposure Rate.]

System International (SI) is the international system of radiation measurements and units.

Sievert (Sv) is the SI unit for dose equivalent (1 Sv = 100 rem).

Specific activity (SpA) relates the number of curies per gram of a given radioisotope, as follows:

$$SpA \text{ (Ci/g)} = \frac{1.3 \times 10^8}{(\text{half-life, days}) (\text{atomic weight})}$$

For example, the SpA for the long-lived, naturally occurring uranium isotope U-238 (half-life, 4.51×10^9 years) is 3.3×10^{-7} Ci/g, whereas the SpA for the short-lived phosphorous isotope P-32 (half-life, 14.3 days) is 2.9×10^5 Ci/g. Expressed in another way, one Ci of U-238 weighs 3 megagrams (3×10^6 grams), whereas one Ci of P-32 weighs 3.4 micrograms (3.4×10^{-6} gram). From this example it is clear that the shorter the half-life (i.e., the faster the disintegration rate) of a radioisotope, the smaller the amount of material required to equal a curie quantity; conversely, the longer the half-life of a radioisotope, the larger the amount of material required to obtain a curie amount. The specific activity of a radioisotope is one major factor determining its relative hazard.

Specific ionization is the number of ion pairs produced by ionizing radiation per unit path length. The number of ion pairs produced depends on the mass and charge of the incident radiation. Because of their somewhat massive size and charge, alpha particles create more ion pairs than do beta particles, which, in turn, create more ion pairs than do gamma photons. Since it may take more than one ionizing collision to absorb a radiation particle or photon, particulate or electromagnetic radiation may produce several ion pairs.

Total ionization is the total number of ion pairs produced by ionizing radiation in a given media (e.g., air or biological material).

Ubiquitous manmade radionuclides are those radionuclides, naturally occurring or synthetic, generated by man's activities and widely distributed in the environment.

Working level (WL) is a special unit used to describe exposure to the short-lived radioactive decay products of radon (Rn-222) and is defined as any combination of radon decay products in one liter of air that will result in the ultimate emission of 1.3×10^5 MeV of alpha energy.

Working level month (WLM) is the exposure to 1 WL for 170 hours (1 working month).

APPENDIX II

Radioactive Substances in the Environment

This appendix identifies potential sources, properties, and pathways of radioisotopes in the environment to provide the reader with a useful context for discussions of measurement techniques and their application to HRS scoring. In general, radioactive sources at Superfund sites contain either naturally occurring radionuclides or manmade radionuclides, or both, in varying concentrations and physical and chemical forms.

Radionuclides present in the natural environment can be divided into three groups according to origin:

- (1) **Naturally occurring radionuclides** are those terrestrial radionuclides (and their decay products) of primordial origin with half-lives comparable to the age of the earth (about 3 billion years);
- (2) **Cosmic radiation and cosmogenic radionuclides** consist of primary charged and neutral particles that bombard the earth's atmosphere and the secondary particles generated by the primary particles in the earth's atmosphere; and
- (3) **Ubiquitous manmade radionuclides** are those radionuclides generated by man's activities and widely distributed in the environment.

Group #1: Naturally Occurring Radionuclides

Naturally occurring terrestrial radionuclides include several dozen or more radionuclides of the uranium, thorium, and actinium series that decay in series to eventually form isotopes of stable lead. Also included among the naturally occurring radionuclides are a group of "non-series" radioisotopes, e.g., H-3, K-40, and Rb-87, that decay directly to a stable isotope. Uranium-238, U-235, and Th-232 head the uranium, actinium, and thorium series, respectively. Each of these series can be further divided into several subseries based on the differences in the radioactive and physical properties of their progeny, as discussed below. When the decay members of these series are not subjected to either chemical or physical separation processes in the environment, a state of secular equilibrium may be achieved whereby the all series members decay at the same rate as the parent nuclide heading the series. More often, however, series members separate from each other in the environment to some extent due to their differing physical and chemical properties. As a result, varying degrees of activity disequilibrium can occur among series members.

Uranium Series

The members of the uranium series are shown in Exhibit 1 along with their respective radioactive half-lives and principal decay modes. Uranium-238, which heads this series, constitutes 99.28% by weight of the four isotopes of uranium with mass numbers 230, 234, 235, and 238 found in nature. By comparison, the natural abundances of U-234 and U-235 are only 0.0058% and 0.71%, respectively.

The first uranium subseries consists of the radioisotopes U-238, Th-234, Pa-234m, and U-234. In general, all four isotopes are found together in equal activity concentrations (i.e., secular equilibrium) under a wide range of environmental settings. However, less than equal activity concentrations of U-238 and U-234 have been reported by several investigators, indicating that some separation of these isotopes may occur in the environment. For example, Rosholt et al. (Ro66) reported a $^{234}\text{U}/^{238}\text{U}$ activity ratio as low as 0.58 in a soil horizon weathered to clay, and Smith and Jackson (Sm69) reported activity ratios of 0.914 to 0.985 in 16 widely distributed sources. A uranium activity ratio of 1.1 in water was determined from samples taken from the Atlantic, Pacific, and Indian-Oceans (Ro64). Because of the large variability that can exist in uranium isotope activity ratios, it is very important to determine the degree of isotopic equilibrium between U-234 and U-238 in media samples on a site-specific basis.

Exhibit 1. Uranium Decay Series*

Radioisotope (atomic #)	Historical name	Half-life**	Major radiation energies (MeV) and intensities***		
			α	β	γ
U-238 (92) ↓	Uranium I	4.51 x 10 ⁹ y	4.15 (25%) 4.20 (75%)	---	---
Th-234 (90) ↓	Uranium X ₁	24.1 d	---	0.103 (21%) 0.193 (79%)	0.063c (4%) 0.093c (4%)
Pa-234m [†] (91) ↓	Uranium X ₂	1.17 m	---	2.29 (98%)	0.765 (0.3%) 1.001 (0.6%)
U-234 (92) ↓	Uranium II	2.47 x 10 ⁵ y	4.72 (28%) 4.77 (72%)	---	0.53 (0.2%)
Th-230 (90) ↓	Thorium	8.0 x 10 ⁴ y	4.82 (24%) 4.88 (76%)	---	0.068 (0.6%) 0.142 (0.07%)
Ra-226 (88) ↓	Radium	1602 y	4.60 (6%) 4.78 (95%)	---	0.186 (4%)
Rn-222 (86) ↓	Radon (gas)	3.82 d	5.49 (100%)	---	0.510 (0.07%)
Po-218 [†] (84) ↓	Radium A	3.05 m	6.00 (~100%)	0.33 (~0.02%)	---
Pb-214 (82) ↓	Radium B	26.8 m	---	0.65 (50%) 0.71 (40%) 0.98 (6%)	0.295 (19%) 0.352 (36%)
Bi-214 [†] (83) ↓	Radium C	19.7 m	5.45 (0.012%) 5.51 (0.008%)	1.0 (23%) 1.51 (40%) 3.26 (19%)	0.609 (47%) 1.120 (17%) 1.764 (17%)
Po-214 (84) ↓	Radium C'	164 μ s	7.69 (100%)	---	0.799 (0.014%)
Pb-210 (82) ↓	Radium D	21 y	---	0.016 (85%) 0.061 (15%)	0.047 (4%)
Bi-210 (83) ↓	Radium E	5.01 d	4.85 (0.00007%) 4.89 (0.00005%)	1.161 (~100%)	---
Po-210 (84) ↓	Radium F	138.4 d	5.305 (100%)	---	0.803 (0.0011%)
Pb-206 (82)	Radium G	Stable	---	---	---

* Source: Lederer and Shirley (1978) and Shleien and Terpilak (1984).

** Half-life given in seconds (s), minutes (m), days (d), or years (y).

*** Intensities refer to percentage of disintegrations of the nuclide itself, not to the parent of the series.

† Approximately 0.13% of all Pa-234m β particle emissions form an intermediate radioisotope, Pa-234 (6.75 hrs: β -emitter), before decaying to U-234. For Po-218, 0.02% decays through At-218 (~2 sec: α -emitter) before forming Bi-214. For Bi-214, 0.02% decays through Tl-210 (1.3 m: β -emitter) to Pb-210.

The second uranium subseries, headed by U-234, includes Th-230 and Ra-226. In general, the activity concentrations of Th-230 and Ra-226 measured in most soils and rocks are comparable to those of U-238 and U-234, suggesting that Th and Ra do not tend to migrate from either of their uranium precursors under stable conditions. This may not be true in the case of ground water, surface water, or sediments. For example, Rosholt et al. (Ro66) reported that the disequilibrium between Th-230 and U-238 or U-234 may range by a factor of two in sea water and enhanced in sediments. Other evidence suggests that Ra-226 is readily mobile in natural waters, either due to recoil or breakdown of entrapping solids. A common place for accumulation of radium isotopes is in the calcium carbonate "sinter" deposited at the orifices of, and with the out-wash from, hot springs. Such locations typically show little activity from the uranium precursors. In other environmental settings, Ra-226 demonstrates a strong affinity for anions, particularly sulfate. Thus, in uranium deposits that have been subjected to strong sulfuric acid water produced by the oxidation of ferrous sulfide, low concentrations of Ra-226 are present.

The third subseries, headed by Ra-226, consists of Rn-222, a noble gas, and its short half-life progeny, Po-218, Pb-214, Bi-214, and Po-214. Due to its inert gas structure and relatively long radioactive half-life, Rn-222 is highly mobile in the environment. The short-lived radon progeny are readily ionized and are attracted to dust particles in the air or to clay minerals in soil. In general, Rn-222 and its short half-life progeny quickly establish equilibrium activity concentrations in most samples.

The final subseries consists of the longer-lived radon decay products, Pb-210, Bi-210, and Po-210, and terminates with the formation of stable Pb-206. Due primarily to the migration of Rn-222, Pb-210 concentrations in environmental media are highly variable. Variable concentrations of Po-210 are also common due to its chemical properties.

Actinium Series

Uranium-235 heads the actinium series shown in Exhibit 2. Similar to the uranium series, the actinium series also includes radionuclides with half-lives long enough to permit disequilibrium conditions. Rosholt (Ro59) considers all progeny of U-235 to be a single group headed by Pa-231, which he has shown to be out of equilibrium with U-235. The short half-life of Ra-223 (11.4 days) usually precludes any significant disequilibrium between itself and its parent Pa-231. For the case of radium deposits from ground water, a separate subgroup headed by Ra-223 and ending with stable Pb-207 is often considered. Disequilibrium due to migration of the noble gas Rn-219 is local due to its 4 second half-life.

Thorium Series

The thorium series (Exhibit 3), headed by Th-232, comprises a number of somewhat short-lived progeny. Given no migration of these progeny, the series reaches secular equilibrium in 60 years in minerals, rocks, and soils of low permeability. In highly permeable soils, waters, natural gas, petroleum, and the atmosphere, the chemical and physical properties of the progeny can cause disequilibrium.

The thorium series may be divided into three subseries. The first subseries consists of Th-232 only, the least mobile of the series radionuclides. This radioisotope exists naturally as a very stable oxide and is strongly adsorbed on silicates (Cl76). The second subseries consists of Ra-228, Ac-228, Th-228, and Ra-224. The equilibrium of this subseries is governed by radioactive recoil, adsorption, and changes in carrier compounds with which the radionuclides become associated. Thoron, Rn-220, and its progeny down to stable Pb-208 make up the third possible subseries. As with the actinium series, disequilibrium caused by migration of the noble gas Rn-220 is unlikely due to the short half-life of Rn-220 (55 second).

Non-Series Radionuclides

Exhibit 4 lists 7 of the 17 naturally occurring radionuclides that decay to stable isotopes. Of the 17, 15 have combinations of half-lives, isotopic abundances, and elemental abundances which result in their having insignificant specific activities. Only K-40, Rb-87 and H-3 occur in significant concentrations in nature. K-40 and Rb-87 are alkali metals and Rb-87 is found in nature as a replacement for potassium in minerals.

Exhibit 2. Actinium Decay Series*

Radioisotope (atomic #)	Historical name	Half-life**	Major radiation energies (MeV) and intensities***		
			α	β	γ
U-235 (92) ↓	Actinouranium	7.1 x 10 ⁸ y	4.37 (18%) 4.40 (57%) 4.58c (8%)	---	0.143 (11%) 0.185 (54%) 0.204 (5%)
Th-231 (90) ↓	Uranium Y	25.5 h	---	0.140 (45%) 0.220 (15%) 0.305 (40%)	0.026 (2%) 0.084c (10%)
Pa-231 (91) ↓	Protactinium	3.25 x 10 ⁴ y	4.95 (22%) 5.01 (24%) 5.02 (23%)	---	0.027 (6%) 0.29c (6%)
Ac-227[†] (89) ↓	Actinium	21.6 y	4.86c (0.18%) 4.95 (1.2%)	0.043 (~99%)	0.70 (0.08%)
Th-227 (90) ↓	Radioactinium	18.2 d	5.76 (21%) 5.98 (24%) 6.04 (23%)	---	0.050 (8%) 0.237c (15%) 0.31c (8%)
Ra-223 (88) ↓	Actinium X	11.43 d	5.61 (26%) 5.71 (54%) 5.75 (9%)	---	0.149c (10%) 0.270 (13%) 0.33c (6%)
Rn-219 (86) ↓	Actinon (gas)	4.0 s	6.42 (8%) 6.55 (11%) 6.82 (81%)	---	0.272 (9%) 0.401 (5%)
Po-215[†] (84) ↓	Actinium A	1.78 ms	7.38 (~100%)	0.74 (~0.0002%)	---
Pb-211 (82) ↓	Actinium B	36.1 m	---	0.29 (1.4%) 0.56 (9.4%) 1.39 (87.5%)	0.405 (3.4%) 0.427 (1.8%) 0.832 (3.4%)
Bi-211[†] (83) ↓	Actinium C	2.15 m	6.28 (16%) 6.62 (84%)	0.60 (0.28%)	0.351 (14%)
Tl-207 (81) ↓	Actinium C''	4.79 m	---	1.44 (99.8%)	0.897 (0.16%)
Pb-207 (82)	Actinium D	Stable	---	---	---

* Source: Lederer and Shirley (1978) and Shleien and Terpilak (1984).

** Half-life given in seconds (s), minutes (m), days (d), or years (y).

*** Intensities refer to percentage of disintegrations of the nuclide itself, not to the parent of the series.

† Approximately 1.4% of all Ac-227 emissions form an intermediate radioisotope, Fr-223 (22 m; β -emitter), before decaying to Ra-223. For Po-215, 0.00023% decays through At-215 (~ 0.1 msec; α -emitter), before forming Bi-211. For Bi-211, 0.28% decays through Po-211 (0.52 sec; β -emitter) to Pb-207.

Exhibit 3. Thorium Decay Series*

Radioisotope (atomic #)	Historical name	Half-life**	Major radiation energies (MeV) and intensities***		
			α	β	γ
Th-232 (90) ↓	Thorium	1.41 x 10 ¹⁰ y	3.95 (24%) 4.20 (75%)	---	---
Ra-228 (88) ↓	Mesothorium I	6.7 y	---	0.005 (100%)	---
Ac-228 (89) ↓	Mesothorium II	6.13 h	---	1.18 (35%) 1.75 (12%) 2.09 (12%)	0.34c (15%) 0.908 (25%) 0.96c (20%)
Th-228 (90) ↓	Radiothorium	1.910 y	5.34 (28%) 5.43 (71%)	---	0.084 (1.6%) 0.214 (0.3%)
Ra-224 (88) ↓	Thorium X	3.64 d	5.45 (6%) 5.68 (94%)	---	0.241 (3.7%)
Rn-220 (86) ↓	Thoron (gas)	55 s	6.29 (100%)	---	0.55 (0.07%)
Po-216 (84) ↓	Thorium A	0.15 s	6.78 (100%)	---	---
Pb-212 (82) ↓	Thorium B	10.64 h	---	0.346 (81%) 0.586 (14%)	0.239 (47%) 0.300 (3.2%)
Bi-212 [†] (83) ↓ ↓ (64%) (36%) ↓ ↓	Thorium C	60.6 m	6.05 (25%) 6.09 (10%)	1.55 (5%) 2.26 (55%) 0.98 (6%)	0.040 (2%) 0.727 (7%) 1.620 (1.8%)
Po-212 ↓ (84) ↓ ↓ ↓	Thorium C'	304 ns	8.78 (100%)	---	---
↓ Tl-208 ↓ (81) ↓ ↓	Thorium C''	3.01 m	---	1.28 (25%) 1.52 (21%) 1.80 (50%)	0.511 (23%) 0.583 (86%) 0.860 (12%) 2.614 (100%)
Pb-208 (82)	Thorium D	Stable	---	---	---

* Source: Lederer and Shirley (1978) and Shleien and Terpilak (1984).

** Half-life given in seconds (s), minutes (m), hours (h), days (d), or years (y).

*** Intensities refer to percentage of disintegrations of the nuclide itself, not to the parent of the series.

† Percentages in brackets are branching fractions.

Exhibit 4. Non-Series Naturally Occurring Radioisotopes*

Radioisotope (atomic #)	Name (elemental abundance)	Half-life**	Major radiation energies (MeV) and intensities***		
			α	β	γ
K-40 (19)	Potassium (0.0118%)	1.3×10^9 y	---	1.314 (89%)	1.46 (11%)
Rb-87 (37)	Rubidium (27.85%)	4.7×10^{10} y	---	0.274 (100%)	---
La-138 (57)	Lanthanum (0.089%)	1.1×10^{11} y	---	0.21 (100%)	0.81 (30%) 1.43 (70%)
Sm-147 (62)	Samarium (15.07%)	1.1×10^{11} y	2.2 (100%)	---	---
Lu-176 (71)	Lutetium (2.6%)	2.2×10^{10} y	---	0.43 (100%)	0.088 (15%) 0.202 (85%) 0.306 (95%)
Re-187 (75)	Rhenium (62.9%)	4.3×10^{10} y	---	0.043 (100%)	---

* Source: Lederer and Shirley (1978).

** Half-life given in years (y).

*** Intensities refer to percentage of disintegrations of the nuclide itself.

Distribution of Naturally Occurring Radionuclides:

In Rocks

The source of the primordial radionuclides is the earth's crust and underlying plastic mantle. Because of sedimentary processes sorting the products of weathering, several major types of sedimentary rock (shales, sandstones, and carbonate rocks) develop that differ significantly in radionuclide concentrations:

- Shales are composed of fine grains of clay (normally 35%), silt, or mud obtained from the breakdown of other rock. A significant fraction of shale contains potassium as a major constituent. All shale can adsorb the series radionuclides. The radionuclides also may be present in the cement that binds the shale together. Mean values for common shales are 2.7 percent potassium, 12 ppm thorium, and 3.7 ppm uranium (Cl66).
- Sandstones are composed of medium-sized grains, usually of quartz (SiO_2), that contain little in the way of radioactive impurities. Sandstone consisting of quartz grains bound with quartz cement is one of the least radioactive rocks. Such sandstone may contain less than 1 percent potassium, less than 2 ppm thorium, and less than 1 ppm uranium. Arkoses - sandstones that contain greater than 25 percent potassium-bearing feldspar - may contain upwards of two to three percent potassium. Clark et al. (Cl 66) report averages of 6.4 ppm thorium and 3.0 ppm uranium for modern beach sands. Thus, sandstone made from beach sand may be high in the series nuclides. In general, sandstones are low in both series and non-series radionuclides.
- Carbonate rocks (limestone and dolomites), derived by chemical precipitation from water or by accumulation of shells, bones, and teeth of organisms, are low in radionuclide content. Still the intergranular spaces contain a variety of elements characteristic of the sea water where most radionuclides may be deposited. Carbonate rocks are low in potassium due to the high solubility of potassium salts, and are low in thorium because it is highly depleted in sea water. Uranium becomes fixed by the reducing conditions prevailing in the decaying organic matter at the sea bottom and thus becomes incorporated in the carbonate rocks.

Exhibit 5 provides summary data on the average concentrations of K-40, Rb-87, Th-232, and U-238 in various types of rocks and sediments.

In Soil

Radionuclides in soil are derived from source rock. In most cases, soil activity concentrations are often less than source rock concentrations due to water leaching, dilution as a result of the soil's increased porosity, and the addition of organic matter and water. In addition, biochemical processes taking place during soil development also tend to reduce the radionuclide concentrations in comparison to the source rock. However, in some cases, soil radioactivity may be augmented by sorption or precipitation of radionuclides from incoming water, by redistribution of wind-blown soils, or by activities such as adding fertilizer or importing top soil to a location. Exhibit 5 provides summary data on average concentrations of K-40, Rb-87, Th-232, and U-238 in soil.

In the Hydrosphere

The concentrations of naturally occurring radionuclides in water are several orders of magnitude less than those in rocks and soils. Potassium-40 is one of the more abundant radionuclides in most water systems. For uranium and thorium series isotopes, there is a shift away from equilibrium between parent radionuclides and progeny. Concentrations of uranium and Rn-222 daughters are frequently observed to be elevated compared to Ra-226 levels.

Exhibit 5. Concentrations of Naturally Occurring Radioisotopes in Rock and Soil^a

Type of Rock or Soil	Potassium-40		Rubidium-87		Thorium-232		Uranium-238	
	Percent	pCi/g	ppm	pCi/g	ppm	pCi/g ^b	ppm	pCi/g ^c
Igneous rocks								
Basalt (crustal average)	0.8	8	40	0.8	3 - 4	0.3 - 0.4	0.5 - 1	0.2 - 0.3
Maefic ^d	0.3 - 1.1	2 - 11	10 - 50	0.03 - 1	1.6 - 2.7	0.2 - 0.3	0.5 - 0.9	0.2 - 0.3
Sialic ^d	4.5	30 - 41	170 - 200	4 - 5	16 - 20	1.6 - 2.2	3.9 - 4.7	1.4 - 1.6
Granite (crustal average)	> 4	> 30	170 - 200	4 - 5	17	1.9	3	1.1
Sedimentary rocks								
Shale sandstones	2.7	22	120 ^e	3	12	1.4	3.7	1.1
Clean quartz	< 1	< 8	< 40 ^e	< 1	< 2	< 0.2	< 1	< 0.3
Dirty quartz	27	117	907	27	3 - 67	0.3 - 0.7	2 - 37	< 1.1
Arkose	2 - 3	16 - 24	80 - 120	2	27	< 0.2	1 - 27	0.3 - 0.77
Beach sands (unconsolidated)	< 1	< 8	< 40	< 1	6	0.7	3	1.1
Carbonate rocks	0.3	2	10	0.2	2	0.2	2	0.7
Continental upper crust Average^f	2.8	23	112	3	10.7	1.2	2.8	1.0
Soils^g	1.5	11	65	1	9	1.0	1.8	1.8
Soils ^h	---	3 - 19	---	3.5	---	0.2 - 1.4	---	0.2 - 1.4

- a. References cited in text unless otherwise noted; single values are average; values estimated in the absence of reference are followed by a question mark.
- b. To obtain series equilibrium alpha, beta, or approximate gamma activity (excluding bremsstrahlung and X rays), multiply by 6, 4, or 3, respectively.
- c. To obtain series equilibrium alpha, beta, or approximate gamma activity (excluding bremsstrahlung or X rays), multiply by 8, 6, or 3, respectively.
- d. From Cl66 for potassium and rubidium, the range of values for rocks within the class is given; for thorium and uranium, the median and mean values are given, respectively.
- e. Estimated by application of crustal abundance ratio with respect to potassium.
- f. From Ta85.
- g. In-situ gamma spectral measurements at 200 locations by Lowder et al. (1964).
- h. Potassium, thorium, and uranium from Annex, 1, UN82; rubidium from NCRP (1976).

Elevated Rn-222 concentrations, ranging from several hundreds to several thousands of pCi/L, are often found in ground water samples, whereas Ra-226 concentrations in the same sample are typically a factor of 1000 lower. Radium and thorium isotopes tend to concentrate in bottom sediments.

Radionuclide concentrations of fresh water bodies and urban water supplies vary widely depending on local geology, hydrology, geochemistry, and radionuclide soil concentrations. Sea water, on the other hand, exhibits a rather narrow range of activity concentrations (Ko62, Ch86).

In the Atmosphere

The level of radioactivity in air and soil water is due primarily to Rn-222, Rn-220, Rn-219, and their decay products. Approximately 35 percent of the Rn-222 produced from Ra-226 in soil emanates into soil pore spaces, resulting in a Rn-222 concentration of about 500 pCi/L of pore fluid per ppm of U-238 in equilibrium with Ra-226 (NCRP87b). At a soil concentration of 1-2 ppm of U-238, Rn-222 levels in soil pores range 10^2 to 10^3 pCi/L, several orders of magnitude greater than typical atmospheric levels. Atmospheric radon concentrations depend on the amount of radon exhaled by the soil and on atmospheric factors that control its upward dispersion. Rn-222 measurements outdoor show that the mean concentrations can range from 100 to 1100 pCi/m³ (NCRP87b). Exhibit 6 summarizes typical concentrations of naturally occurring radionuclides in the atmosphere.

In the Biosphere

Potassium-40 is the most abundant radionuclide in the biosphere. Concentrations of other naturally occurring radionuclides in plants and animals are highly variable and are almost never in equilibrium (NCRP76). For example, Ra-226 is preferentially taken up by plants relative to U-238 or U-234. In general, activity concentrations in plants range from 1 to 50 pCi/g for ⁴⁰K, from 0.01 to 10 pCi/g for Po-210, and are about 0.1 pCi/g for Rb-87 (NCRP76), as shown in Exhibit 7.

Group #2: Cosmic Radiation and Cosmogenic Radionuclides

Cosmic radiation consists of primary charged and neutral particles that bombard the earth's atmosphere and the secondary particles (e.g., H-3 and C-14) generated by the primary particles in the earth's atmosphere. Primary cosmic radiation, produced by supernovas and solar flares, is composed of approximately 87 percent photons, 11 percent alpha particles, 1 percent heavier nuclei, and 1 percent electrons with energies up to at least 10^{20} eV (average energy is 10^8 to 10^{11} eV). Secondary cosmic particles are produced by a variety of spallation and neutron activation reactions, mostly with the nuclei of argon, nitrogen, and oxygen.

Cosmic radiation increases with altitude as the mass of the atmosphere decreases. Cosmic flux density is least near the geomagnetic equator and increases with latitude. At sea level, the flux density is about 10% lower at the equator than at high latitudes. Energetic solar flares generate large numbers of photons that can penetrate the earth's magnetic field and add to the cosmic ray flux density incident on the atmosphere. These bursts seldom produce significant effects at ground level. There is evidence for an 11-year cycle in mean solar activity that produces a modulation of the cosmic radiation reaching the earth's atmosphere. At ground altitudes, the effect is about 10 percent.

Exhibit 8 shows the typical environmental radiation field at 1 meter above sea level due to cosmic and terrestrial radionuclides.

A total of 20 radionuclides are produced by cosmic rays in the earth's atmosphere. From the point of view of radiation measurements and doses, only carbon-14 (C-14) and, to a lesser extent, tritium (H-3) are worth considering.

Exhibit 6. Radionuclides In The Atmosphere*

Radionuclide	Surface air content	
	Typical range (pCi/m ³)	Mean value (pCi/m ³)
Uranium series:		
Rn-222		
Pb-214	20 - 500	120
Bi-214	0 - 500	100
Pb-210	0 - 500	100
Po-210	0.003 - 0.03	0.01
	--	0.003
Thorium series:		
Rn-220		
Pb-212	--	100
	0.5 - 10	2
Others:		
Kr-85	--	17
Be-7	0.02 - 0.20	0.06

* Source: NCRP (1976): Table 2-8.

Exhibit 7. Total Natural Radioactivity In Plants*

Radiation	Concentration (pCi/g gross weight)	Source
Gross alpha	0.14 - 3.1	mainly as Po-210; other U + Th series nuclides
Gross beta	7.8 - 123	mainly as K-40; Pb-210; Bi-210; other U + Th series nuclides
K-40	1 - 50	--
Rb-87	~0.1	--
Po-210	0.01 - 10	--

* Source: NCRP (1976): Table 2-9b.

Exhibit 8. Typical Environmental Radiation Field (One Meter Height)*

Radiation	Energy (MeV)	Source	Absorbed dose rate in free air (microrad/hr)
alpha	1 - 9	radon (atm)	2.7
beta	0.1 - 2 0.1 - 2 2 - 200	radon (atm) K, U, Th, Sr (soil) cosmic rays	0.2 2.5 0.7
gamma	<2.4 <1.5 <2.4 <2.6 <0.8	radon (atm) K (soil) U (soil) Th (soil) Cs + other fallout (soil)	0.2 2.0 1.0 2.4 0.3
neutron proton muons	0.1 - 100 10 - 2,000 100 - 30,000	cosmic rays	0.1
			Total: 14.5

* Source: NCRP (1976): Table 2-10.

Tritium (H3)

Tritium, a radioactive isotope of hydrogen, is a beta emitter (average energy 5.69 keV) with a radioactive half-life of 12.3 years. It occurs naturally in the surface waters of the earth as a product of the atmospheric interaction of high-energy cosmic rays with nitrogen and oxygen gases (UN72, NCRP79). Its annual production rate is approximately 2 megacuries (MCi), resulting in a steady-state inventory of about 30 MCi in the biosphere. Since 1954, large amounts of manmade tritium have been released into the environment primarily from nuclear weapons testings, discharges from nuclear power plants (Exhibit 10), and some nuclear weapons production plants. Tritium is used as a radioactive luminizing material in consumer products, such as watches, clocks, and emergency signs, and as a component of nuclear weapons.

Prior to the injection into the biosphere from nuclear tests, levels of H-3 in waters of the mid-latitude regions of the earth were in the range of 6 to 24 pCi/L. The amount of tritium added to the global inventory as a result of nuclear weapons testing is discussed under the next section on manmade radionuclides. About 90% of natural H-3 resides in the hydrosphere, 10% in the stratosphere, and only 0.1% in the troposphere. The low inventory of H-3 in the troposphere is due to the fact that tritium in the form of HTO is rapidly washed out by rain, with an estimated residence time of between 20 to 40 days.

Carbon-14 (C-14)

Carbon-14 is the one of the three isotopes of carbon: C-12 (99.8%), C-13 (1.1%), and C-14 (0.1%). It is a pure beta-emitting radionuclide (average energy 50 keV) with a radioactive half-life of 5,730 years. Natural C-14 is produced in the upper atmosphere by interaction of cosmic-ray neutrons with nitrogen. Its production rate is not accurately known, but may correspond to about 0.03 MCi per year with a steady-state inventory of approximately 280 MCi (UN72). Similar to tritium, C-14 has been produced in significant quantities by nuclear weapons testing and discharges from nuclear power plants (see the section on manmade radionuclides).

As an isotope of carbon, C-14 is involved with all biological and geochemical process on earth. It is present in the atmosphere as carbon dioxide, in the terrestrial biosphere as incorporated carbon, and in surface waters as dissolved bicarbonates. The concentration of C-14 in the environment varies widely. At present, the United Nations assumes a specific activity of 6.1 pCi/g in the terrestrial biosphere (UN 72).

Group #3: Ubiquitous Manmade Radionuclides

Manmade radioisotopes that are widely distributed in the environment are due primarily to releases from nuclear weapons testing and nuclear power facilities. Exhibits 9 and 10 list some of the important radionuclides produced by these processes.

Radionuclides released during nuclear weapons testing: Since the first test of a nuclear weapon at Alamogordo, New Mexico, in 1945, approximately 450 additional nuclear weapons have been detonated in the atmosphere. These detonations resulted in the production and global dispersal of several millions of curies of radioactive fission and activation products, transuranic elements, and unfissioned uranium and plutonium isotopes.

These detonations also significantly increased natural concentrations of H-3 and C-14. Between 1,900 to 8,000 MCi of H-3 were added to the northern hemisphere by nuclear weapons testing through 1963 (Er65, Mi71). As a result, average concentrations of H-3 in surface waters in the U.S. rose from 3 to 16 pCi/L to about 4,000 pCi/L in 1963 (Be73). Today, tritium concentrations due to fallout H-3 have decreased below the level due to natural H-3 (NCRP79). By the end of 1962, nuclear testing had increased the atmospheric concentration of C-14 to about twice its pre-1950 concentration of 6 pCi/g. Because of exchange with the ocean and to a lesser extent the biosphere, C-14 concentrations in the atmosphere due to weapons testing dropped to about 3 pCi/g by the end of 1970 (NCRP87b). The increase in C-14 concentrations in the ocean has been greatest in the surface waters since C-14 has a residence time of three to eight years in the mixing layers before it is transferred below the thermocline. Because it takes a few thousand years before C-14 reaches the ocean floor, there is no increase in C-14 concentrations for deep ocean sediments.

Strontium-90 and Cs-137 are two of the most important fission products that were widely distributed in near-surface soils because of the weapons testing. Measurable concentrations of Sr-90 and Cs-137 in soil exist today. These concentrations are distributed almost exclusively in the upper 15 cm of soil and decrease roughly exponentially with depth.

Radionuclides released from nuclear power stations: Releases of radionuclides produced by nuclear fission in boiling water reactors (BWRs) and in pressurized water reactors (PWRs) occur because of periodic fuel failure, defects, or corrosion that results in transfer of some fission and activation products into the reactor coolant. In PWRs, the primary coolant is in a sealed loop that is continually purged for control of chemical composition and purification. Gaseous wastes released in the process are held in tanks for between 30 to 120 days to allow short-lived nuclides to decay prior to release. Other gaseous effluent streams originate from the condenser exhaust on the steam circuit, secondary coolant blowdown, reactor building ventilation (including containment purges), and turbine plus ancillary building ventilation (UN82). In BWRs, the main condenser air-ejector system continuously removes non-condensable gases from the steam flow. This is the main source of noble gases released with the gaseous waste stream. Secondary pathways include the purging system for the turbine gland seals, the condenser mechanical vacuum pump, and any process fluid leaks to ventilated buildings.

Radionuclides released to the atmosphere include noble gases (argon, krypton, and xenon), C-14, tritium, iodines, and particulates. Radionuclides discharged in liquid effluents include tritium, fission products, and activated corrosion products. Exhibit 10 lists the nuclide composition of typical liquid and gaseous effluents for PWRs and BWRs in the U.S. Compositions often vary depending on waste treatment methods employed, the age and condition of the plant, etc. Release rates are not listed for the nuclides since these data vary greatly from plant to plant. Environmental monitoring programs typically show that the nuclides in the effluents are not readily detectable in the environment except near the point of release.

Exhibit 9. Ubiquitous Manmade Radioisotopes*

Radioisotope (atomic #)	Name (Origin)†	Half-life**	Major radiation energies (MeV) and intensities***		
			α	β	γ
H-3 (1)	Tritium (NE, NF)	12.3 y	---	0.0186 (100%)	---
C-14 (6)	Carbon (NE, FF)	5730 y	---	0.156 (100%)	---
Mn-54 (25)	Manganese (NE)	303 d	---	---	0.835 (100%)
Fe-55 (26)	Iron (NE)	2.6 y	---	---	0.23 (0.004%)
Co-60 (27)	Cobalt (NE, NF)	5.26 y	---	1.48 (0.12%) 0.314 (99%)	1.17 (100%) 1.33 (100%)
Zn-65 (30)	Zinc (NE, NF)	245 d	---	$\beta+$: 0.327 (1.4%)	0.511 (3.4%) 1.12 (49%)
Kr-85 (36)	Krypton (NE, NF)	10.76 y	---	0.173 (0.4%) 0.687 (99.6%)	0.514 (0.4%)
Sr-90 (38)- Y-90 (39)	Strontium - Yttrium (NE, NF)	28 y (Sr) 64 h (Y)	--	0.546 (100% Sr) 2.27 (100% Y)	---
Zr-95 (40)	Zirconium (NE)	65.5 d	---	0.366 (55%) 0.398 (44%)	0.724 (49%) 0.756 (49%)
Nb-95 (41)	Niobium (NE)	35 d	---	0.160 (99.9%)	0.765 (100%)
Ru-106 (44) - Rh-106 (45)	Ruthenium - Rhenium (NE, NF)	368 d (Ru) 30 s (Rh)	---	0.039 (100% Ru) 3.54 (79% Rh)	0.512 (21%) 0.622 (11%) 1.05 (1.5%)
Sb-125 (51) - Te-125m (52)	Antimony - Tellurium (NE)	2.77 y (Sb) 58 d (Te)	---	0.61 (14% Sb)	0.153 (82% Te) 0.176 (6% Sb) 0.270 (25% Te) 0.427 (10% Sb) 0.599 (24% Sb) 0.634 (11% Sb) 0.66 (3% Sb) 0.92-1.14 (36% Te) 1.22 (67% Te) 2.09 (4% Te)
I-129 (53)	Iodine (NF)	1.7 x 10 ⁷ y	---	0.150 (100%)	0.040 (9%)
Cs-134 (55)	Cesium (NE, NF)	2.05 y	---	0.662 (100%)	0.57 (23%) 0.61 (98%) 0.796 (99%)

Exhibit 9 - Continued¹

Radioisotope (atomic #)	Name (Origin) [†]	Half-life ^{**}	Major radiation energies (MeV) and intensities ^{***}		
			α	β	γ
Cs-137 (55) - Ba-137m (56)	Cesium - Barium (NE, NF)	30 y (Cs) 2.55 m (Ba)	---	0.514 (95% Cs) 1.176 (5% Cs)	0.428 (30% Ba) 0.463 (11% Ba) 0.601 (18% Ba) 0.836 (12% Ba) 0.662 (89% Ba)
Ce-144 (58) - Pr-144 (59)	Cerium - Praseodymium (NE)	284 d (Ce) 17.3 m (Pr)	---	0.31 (76% Ce) 2.99 (98% Pr)	0.080 (2% Ce) 0.134 (11% Ce) 0.695 (1.5% Pr) 1.487 (0.3% Pr) 2.186 (0.7%)
Pu-238 (94)	Plutonium (SNAP, NE)	87 y	5.50 (72%) 5.46 (28%)	---	0.145 (2%)
Pu-239 (94)	Plutonium (NE, NF)	2.439 x 10 ⁴ y	5.155 (73%) 5.143 (15%) 5.105 (12%)	---	0.039 (0.007%) 0.052 (0.020%) 0.129 (0.005%) 0.375 (0.0012%)
Pu-240 (94)	Plutonium (NE, NF)	6580 y	5.1683 (76%) 5.1238 (24%)	---	---
Pu-241 (94) - Am-241 (95)	Plutonium - Americium (NE, NF)	13 d (Pu) 458 y (Am)	4.90 (0.002% Pu) 4.85 (0.003% Pu) 5.3884 (1.6% Am) 5.443 (12.8% Am) 5.486 (85% Am)	---	0.0264 (2.5% Am) 0.0595 (36% Am)

¹ Source: Lederer and Shirley (1978) and NCRP (1976).

^{**} Half-life given in minutes (m), hours (h), days (d), or years (y).

^{***} Intensities refer to percentage of disintegrations of the nuclide itself.

[†] "NE" = Nuclear explosions; "NF" = Nuclear facilities; "SNAP" = SNAP-9a (System for Nuclear Auxiliary Power) which was a satellite which dispersed 1 kg of Pu-238 in the earth's atmosphere when it burned up upon re-entry; and "FF" = Fossil fuel power plants and other industries.

Exhibit 10. Radioisotopes in Nuclear Reactor Effluent*

Effluent Type	Reactor Type	
	PWR	BWR
Gaseous	H-3, K-40, Ar-41, Co-57, Co-58, Co-60, Ni-63, Br-82, Kr-85, Kr-85m, Kr-87, Kr-88, Sr-90, Nb-95, Zr-95, Zr-97, Tc-99m, I-132, I-133, Xe-133m, Cs-134, I-135, Xe-135, Xe-135m, Cs-137, Xe-138, Ce-139, Ce-143, Ce-144, Bi-214, Rn-222, Ra-226, Th-228, Th-232	H-3, N-13, Na-24, Ar-41, Cr-51, Mn-54, Mn-56, Co-58, Fe-59, Co-60, Zn-65, Br-82, Kr-83m, Kr-85, Kr-85m, Kr-87, Kr-88, Kr-89, Sr-89, Kr-90, Sr-90, Sr-91, Y-91, Nb-95, Mo-99, Ru-103, Rh-106, Ag-110m, I-131, Xe-131m, I-132, Xe-133, Xe-131m, I-132, I-133, Xe-133, Xe-133m, Cs-134, I-135, Xe-135, Xe-135m, Cs-136, Cs-137, Xe-138, Ba-139, Xe-139, Ba-140, La-140, Ce-141, Ce-144, Hg-203
Liquid	H-3, Ba-7, Na-24, K-40, Ar-41, Cr-51, Mn-54, Fe-55, Mn-56, Co-57, Co-58, Fe-59, Co-60, Ni-63, Cu-64, Zn-65, Zn-69m, Se-75, As-76, Ge-77, Br-82, Kr-88, Rb-88, Y-88, Rb-89, Sr-89, Mo-90, Sr-90, Y-91, Y-91m, Sr-92, Y-92, Nb-94, Nb-95, Zr-95, Nb-97, Zr-97, Mo-99, Tc-99m, Ru-103, Ru-105, Ru-106, Ag-108m, Cd-109, Ag-110m, Sn-113, Cd-115, In-115m, Sn-117m, Sb-112, Sb-124, Sb-125, Sb-127, I-131, Xe-131m, I-132, Te-132, Ba-133, Cd-133m, I-133, Xe-133, Xe-133m, Cs-134, I-134, I-135, Xe-135, Xe-135m, Cs-136, Cs-137, Cs-138, Ba-139, Ce-139, Ba-140, La-140, Ce-141, Ce-144, Pr-144, W-187, Hg-203, Bi-214, Pb-214, Ra-226, Th-228, Np-239	F-18, Na-24, P-32, Cl-38, Cr-51, Mn-54, Fe-55, Mn-54, Mn-56, Co-57, Co-58, Fe-59, Co-60, Cu-64, Zn-65m, Zn-69m, As-76, Br-84, Kr-85, Kr-85m, Kr-88, Sr-89, Sr-90, Y-91m, Sr-90, Y-91m, Sr-92, Y-92, Nb-95, Zr-95, Nb-97, Mo-99, Tc-99m, Ru-103, Tc-104, Rh-103, Tc-104, Rh-105, Ru-105, Rh-106, Ru-106, Ag-110m, Ag-111, Sb-124, Sb-125, Te-129, Te-129m, I-131, Xe-131m, Te-132, I-133, Xe-133, Xe-133m, Cs-134, I-135, Xe-135, Xe-135m, Cs-136, Cs-137, Cs-138, Ba-139, Ba-140, La-140, Ce-141, La-142, Ce-144, Hg-203, Np-239

* Radioisotope composition of gaseous and liquid effluent from Pressurized Water Reactors (PWRs) and Boiling Water Reactors (BWRs) in the United States for 1979 (NCRP 1987a).

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