United States
Environmental Protection
Agency

Office of Radiation and Indoor Air
National Air and Radiation
Environmental Laboratory

EPA 402-R-12-006
September 2012
www.epa.gov/narel

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Radiological Laboratory Sample Analysis Guide for Incident Response – Radionuclides in Soil

EPA 402-R-12-006
www.epa.gov/narel
September 2012
Revision 0

Radiological Laboratory Sample Analysis Guide for Incident Response – Radionuclides in Soil

National Air and Radiation Environmental Laboratory
Office of Radiation and Indoor Air
U.S. Environmental Protection Agency
Montgomery, AL 36115

This report was prepared for the National Air and Radiation Environmental Laboratory of the Office of Radiation and Indoor Air, United States Environmental Protection Agency. It was prepared by Environmental Management Support, Inc., of Silver Spring, Maryland, under contract EP-W-07-037, Work Assignment I-33, managed by David Garman and Daniel Askren. Mention of trade names or specific applications does not imply endorsement or acceptance by EPA.

PREFACE

The need to ensure an adequate laboratory infrastructure to support response and recovery actions following a major radiological or nuclear incident has been recognized by a number of federal agencies. The Integrated Consortium of Laboratory Networks (ICLN), created in 2005 by 10 federal agencies,[1] consists of existing and emerging laboratory networks across the Federal Government. ICLN is designed to provide a national infrastructure with a coordinated and operational system of laboratory networks that will provide timely, high quality, and interpretable results for early detection and effective consequence management of acts of terrorism and other events requiring an integrated laboratory response. It also designates responsible federal agencies (RFAs) to provide laboratory support across response phases for chemical, biological, and radiological agents. To meet its RFA responsibilities, the U.S. Environmental Protection Agency (EPA) established the Environmental Response Laboratory Network (ERLN) to address chemical, biological, and radiological threats during nationally significant incidents (www.epa.gov/erln/). EPA is the RFA for monitoring, surveillance, and remediation of radiological agents. EPA will share responsibility for overall incident response with the U.S. Department of Energy (DOE).

This document is one of several initiatives by EPA's Office of Radiation and Indoor Air designed to provide guidance to radioanalytical laboratories that will support EPA's response and recovery actions following a radiological or nuclear incident. This guide examines the analysis of soil samples following a radiological incident. The guidance provided in this document for the screening, pretreatment, and analysis of soil samples should assist those federal, state, and commercial radioanalytical laboratories that will be challenged with a large number of such samples when responding to a radiological incident. This document discusses three different types of events; a radiological transportation incident, a radiological dispersal device (RDD) ("dirty bomb"), or the detonation of an improvised nuclear device (IND). These samples will be contaminated with varying levels of radionuclides, and will represent soil matrices of varied composition. Advance planning by national and regional response teams, as well as by radiological laboratories, will be critical to ensure uninterrupted throughput of large numbers of radioactive samples and the rapid turnaround and reporting of results that meet required data quality objectives associated with the protection of human health and the environment. EPA's responsibilities, as outlined in the *National Response Framework*, *Nuclear/Radiological Incident Annex*, include response and recovery actions to detect and identify radioactive substances and to coordinate federal radiological monitoring and assessment activities.

While the recommendations in this Guide may be implemented by radiochemistry laboratories using their standard analytical procedures, EPA has developed a suite of validated rapid methods for selected radionuclides in soil, water, air filters, and swipes. These methods can achieve a required relative method uncertainty of 13% at, or above, an analytical action level of 10^{-4} risk. The methods also have been tested to determine the time within which a batch of samples can be analyzed. For these radionuclides, results for a batch of samples can be provided within a turnaround time of hours instead of the days to weeks required by some previous methods. References to these methods are found throughout this Guide and citations are listed at the end of this Preface and in Section I.E (References). In particular, laboratories needing to analyze soils

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[1] Departments of Agriculture, Commerce, Defense, Energy, Health and Human Services, Homeland Security, Interior, Justice, and State, and the U.S. Environmental Protection Agency.

are encouraged to consider *Rapid Method for Fusion of Soil and Soil-Related Matrices Prior to Americium, Plutonium, and Uranium Analyses; Rapid Method for Radium-226 Analyses in Soil Incorporating the Fusion of Soil and Soil-Related Matrices;* and *Rapid Method for Sodium Carbonate Fusion of Soil and Soil Related Matrices Prior to Strontium-90 Analysis* (EPA 2012a) and validate them as recommended in the *Method Validation Guide for Radiological Laboratories Participating in Incident Response Activities* (EPA 2009a).

Detailed guidance on recommended radioanalytical practices can be found in the *Multi-Agency Radiological Laboratory Analytical Protocols Manual* (MARLAP), which provides detailed radioanalytical guidance for project planners, managers, and radioanalytical personnel based on project-specific requirements (www.epa.gov/radiation/marlap/links.html). Familiarity with Chapters 2 and 3 of MARLAP will be of significant benefit to users of this guide. This document is one in a planned series designed to present radioanalytical laboratory personnel, Incident Commanders (and their designees), and other field response personnel with key laboratory operational considerations and likely radioanalytical requirements, decision paths, and default data quality and measurement quality objectives for analysis of samples taken after a radiological or nuclear incident. Documents[2] currently completed or in preparation include:

- *Radiological Laboratory Sample Analysis Guide for Incidents of National Significance – Radionuclides in Water* (EPA 402-R-07-007, January 2008)
- *Radiological Laboratory Sample Analysis Guide for Incidents of National Significance – Radionuclides in Air* (EPA 402-R-09-007, June 2009)
- *Radiological Laboratory Sample Screening Analysis Guide for Incidents of National Significance* (EPA 402-R-09-008, June 2009)
- *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities* (EPA 402-R-09-006, June 2009)
- *Guide for Laboratories – Identification, Preparation, and Implementation of Core Operations for Radiological or Nuclear Incident Response* (EPA 402-R-10-002, June 2010)
- *A Performance-Based Approach to the Use of Swipe Samples in Response to a Radiological or Nuclear Incident* (EPA 600-R-11-122, October 2011)
- *Guide for Radiological Laboratories for the Control of Radioactive Contamination and Radiation Exposure* (EPA 402-R-12-005, August 2012)
- *Uses of Field and Laboratory Measurements During a Radiological or Nuclear Incident* (EPA 402-R-12-007, August 2012)

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[2] All the documents in this series are available at www.epa.gov/erln/radiation.html and at www.epa.gov/narel/incident_guides.html.

ACKNOWLEDGEMENTS

This guide was developed by the National Air and Radiation Environmental Laboratory (NAREL) of EPA's Office of Radiation and Indoor Air (ORIA). Dr. John Griggs was the project lead for this document. Special acknowledgment and appreciation are extended to Dr. Keith McCroan, ORIA/NAREL, and Dr. Lowell Ralston and Mr. Edward Tupin, CHP, both of ORIA/Radiation Protection Division, for their assistance in developing the protective action guides (PAGs) used in this study. Several individuals provided valuable support and input to this document throughout its development. We wish to acknowledge the external peer reviews conducted by Cecelia DiPrete and Scott C. Moreland.

Numerous other individuals within EPA provided peer review of this document, and their suggestions contributed greatly to the quality and consistency of the final document. Technical support was provided by Environmental Management Support, Inc.

Dedication

This report is dedicated to the memory of our friend and colleague, David Garman. Dave administered nearly three dozen separate contracted radiochemistry projects for EPA dating back nearly 17 years, beginning with the *Multi-Agency Radiological Laboratory Analytical Protocols* (MARLAP) in 1994. Dave put up with countless changes of prime contractors, priorities, subcontractors, and budgets, all with good cheer, diligence, and all while keeping up with his "day job" as counting room lead for alpha-spectrometry analysis at NAREL.

Dave started with EPA's National Air and Radiation Laboratory in 1992. He left many friends throughout EPA and the radioanalytical community, and he will be greatly missed.

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ACRONYMS AND ABBREVIATIONS

(Excluding chemical symbols and formulas)

α	alpha particle
α	probability of Type I decision error
AAL	analytical action level
ADL	analytical decision level
AL	action level
β	beta particle
β	probability of Type II decision error
Bq	becquerel (1 dps)
CFR	*Code of Federal Regulations*
Ci	curie
cm	centimeter
cpm	counts per minute
d	day
DL	discrimination limit
DOE	United States Department of Energy
DP	decay product(s)
dpm	disintegration per minute
dps	disintegration per second
DQO	data quality objective
DRL	derived response levels
DRP	discrete radioactive particle
$E_{\beta max}$	maximum energy of the beta-particle emission
EDX	energy dispersive X-ray analysis
ERLN	Environmental Response Laboratory Network
EPA	United States Environmental Protection Agency
FRMAC	Federal Radiological Monitoring and Assessment Center
γ	gamma radiation
g	gram
Ge	germanium semiconductor
GM	Geiger-Mueller (detector)
GPC	gas proportional counting
GS	gamma spectrometry
Gy	gray
h	hour
H_0	null hypothesis
H_1	alternate hypothesis
HF	hydrofluoric acid
HIC	high integrity container
HPGe	high-purity germanium [detector]
IC	incident commander
ICLN	Integrated Consortium of Laboratory Networks
IND	improvised nuclear device (i.e., a nuclear bomb)
in	inch
kg	kilogram (10^3 gram)

keV	thousand electron volts (10^3 eV)
L	liter
LEPD	low-energy photon detector
LSC	liquid scintillation counter/counting
MARLAP	*Multi-Agency Radiological Laboratory Analytical Protocols Manual*
MARSSIM	*Multi-Agency Radiation Survey and Site Investigation Manual*
mCi	millicurie (10^{-3} Ci)
MeV	million electron volts (10^6 eV)
mg	milligram (10^{-3} g)
mL	milliliter (10^{-3} L)
mrem	millirem (10^{-3} rem)
μCi	microcurie (10^{-6} curie)
μg	microgram (10^{-6} g)
MDC	minimum detectable concentration
Micro-XANES	micrometer-scale X-ray absorption near edge structure spectroscopy
Micro-XRF	micrometer-scale X-ray fluorescence
min	minute
MQO	measurement quality objective
NaI(Tl)	thallium-activated sodium iodide detector
NAREL	EPA National Air and Radiation Environmental Laboratory
NORM	naturally occurring radioactive materials
ORIA	EPA Office of Radiation and Indoor Air
φ_{MR}	relative required method uncertainty
PAG	protective action guide
pCi	picocurie (10^{-12} Ci)
PM	project manager
QA	quality assurance
QC	quality control
rad	radiation absorbed dose
RDD	radiological dispersal device (i.e., “dirty bomb”)
RFA	responsible federal agency
rem	roentgen equivalent: man
RTG	radioisotope thermoelectric generator
s	second
SI	International System of Units
SOF	sum of fractions
SOP	standard operating procedure
SR	synchrotron radiation
Sv	sievert
$t_{½}$	half-life
TEDE	total effective radiation dose equivalent
TRU	transuranic elements
tSIE	transformed spectral index of the external standard
u_{MR}	required method uncertainty
y	year
$z_{1-\alpha}$; $z_{1-\beta}$	$1-\alpha$ and $1-\beta$ quantiles of the standard normal distribution function

RADIOMETRIC UNIT CONVERSIONS

To Convert	To	Multiply by	To Convert	To	Multiply by
years (y)	seconds (s)	3.16×10^{7}	s	y	3.17×10^{-8}
	minutes (min)	5.26×10^{5}	min		1.90×10^{-6}
	hours (h)	8.77×10^{3}	h		1.14×10^{-4}
	days (d)	3.65×10^{2}	d		2.74×10^{-3}
disintegrations per second (dps)	becquerels (Bq)	1	Bq	dps	1
Bq	picocuries (pCi)	27.0	pCi	Bq	3.70×10^{-2}
Bq/kg	pCi/g	2.70×10^{-2}	pCi/g	Bq/kg	37.0
microcuries per milliliter (µCi/mL)	pCi/L	10^{9}	pCi/L	µCi/mL	10^{-9}
disintegrations per minute (dpm)	µCi	4.50×10^{-7}	pCi	dpm	2.22
	pCi	4.50×10^{-1}	µCi		2.22×10^{6}
cubic feet (ft^3)	cubic meters (m^3)	2.83×10^{-2}	m^3	ft^3	35.3
gallons (gal)	liters (L)	3.78	L	gal	0.264
gray (Gy)	rad	10^{2}	rad	Gy	10^{-2}
roentgen equivalent man (rem)	sievert (Sv)	10^{-2}	Sv	rem	10^{2}

NOTE: Traditional units are used throughout this document instead of International System of Units (SI) units. PAGs and their derived concentrations appear in official documents in the traditional units and are in common usage. Conversion to SI units will be aided by the unit conversions in this table.

I. BACKGROUND

This guide deals with the analysis of soil[3] samples that may have been contaminated as the result of the deployment of a radiological dispersion device (RDD) or improvised nuclear device (IND), or a radiological materials transportation event, accident at a nuclear facility, or an intentional release of radioactive materials onto surface soil. In the event of a major incident that releases radioactive materials to the environment, EPA may need to turn to qualified commercial radioanalytical laboratories to support national response teams in determining the radionuclide source term(s), extent and magnitude of contamination, and the possible actions to be taken based on the potential human radiation doses compared to national guidelines. In order to expedite sample analyses and data delivery to the client, the laboratories will need guidance on EPA's expectations.

An incident response to a release of radioactivity to the environment likely will occur in three phases: "early", "intermediate", and "recovery." Each phase of an incident response will require different and distinct radioanalytical resources to address the different consequences, management, priorities, and requirements of each phase. Some of the more important radioanalytical laboratory resources germane to an incident response consist of radionuclide identification and quantification capability, sample load capacity, sample processing turnaround time, quality of analytical data, and data transfer capability.

The exposure to individuals from soils is assessed in a different manner than either air or water exposure pathways. Exposure from soil is based on both direct radiation (i.e., "ground shine") and inhalation due to resuspension of the contaminated soil. Both of these mechanisms are affected by weathering of the soil. This combination yields slightly different protective action guides (PAGs) concepts than for air and water. Part of this difference is that decay and weathering are taken into account when calculating doses based on the soil concentration. For soils, the early phase begins at the initial event and lasts for approximately 96 hours. The PAG is 1,000 mrem during this phase. Because this aspect of the event is so short-lived laboratories that do become involved with soil sample analysis will need to respond promptly with analytical values for samples that are conservative[4] in their assessment of the concentration.

Once the early phase has concluded and soil analyses are in progress, the specific action levels are still dose or risk based but the time period of exposure based on weathering is used to establish specific dose- or risk-based concentrations.

During the intermediate phase the source term radionuclides will have been qualitatively identified, however radionuclide concentrations and the extent of the contaminated zone still may not be well defined. The radioanalytical resources that are needed will depend on the PAGs that are implemented for the incident by the project team. These PAGs may depend upon

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[3] For purposes of this document, "soil" refers to loose, unconsolidated material consisting of sand, silt, and clay mineral particles together with organic matter of various types, which has been subjected to weathering and biological processes, without regard to morphology, moisture content, particle size, or mode of origin.

[4] In this discussion the term "conservative" refers to measurement results where the acceptable error rate for *risk of deciding that analyte is not present at the action level when it is indeed present at or above the action level,* is very low (e.g., < 0.05). See Appendix VI for details.

location and habitation. For the intermediate phase, PAGs have been established to limit the projected exposures for different periods not to exceed:

- 1,000-mrem total effective radiation dose equivalent (TEDE) over the first few days,
- 2,000-mrem TEDE over the first year,
- 500-mrem TEDE during the second year, or
- 5,000-mrem over the next 50 years (including the first and second years of the incident).

In addition, radionuclide concentration limits for food, water, and air as regulated by the Food and Drug Administration and EPA would be applicable. For the analysis of soils the PAG values are in units of pCi/g of soil to correspond to these doses.

The final, or "recovery," phase occurs as part of a radiological incident site remediation effort. The PAG that may be applicable for this recovery phase has as a limit of 5,000 mrem over the next 50 years (including the first and second years of the incident response). Risk-based exposure limits for each of these time periods also have been established, and in turn converted into equivalent soil concentrations. This document also addresses concentrations of radionuclides in soils at the 10^{-4} and 10^{-6} risk factors. During this final phase, when site characterization and remediation cleanup effectiveness is determined, there is a potential need for more extensive radiochemical analyses at these lowest levels of detection.

During all phases of an incident response, radioanalytical resources are needed for identifying the radionuclide source term(s), quantifying the radionuclides in soils, and screening the gross radiation of samples for prioritization of sample processing or for information related to the general level of contamination. This guide has been developed to provide the incident responder (Project Manager, On Scene Coordinator, Incident Commander) and the laboratories used during an incident with a logical processing scheme to prioritize sample processing in relation to the radionuclide concentration action levels corresponding to established PAGs.

A. Structure of the Document

Background (this section). This section describes the radiological event phases, goals of this document, the scenarios that may be encountered during a radiological event, and the sampling issues that may ensue long after the event has terminated.

Discussion. This section provides a perspective on the types of concerns that will arise when dealing with soil samples resulting from a radiological event. Effects of particle non-homogeneity, weathering, oxidation state, and the buildup of radioactive progeny are discussed as background material for potential radionuclide scenarios. The scenarios are based on historical events that are similar to potential future events. Additionally, specific information regarding the type of screening instrumentation and potential background interferences that will be present is discussed. This material should be considered for inclusion when writing standard operating procedures for laboratory analysis.

Scenarios. There are three different situations described in this document. These are:

- The first scenario assumes unknown radionuclides and elevated unknown concentrations in soil samples. Sample priority flow path is determined using methods based on identifying gross radionuclide activity concentrations that exceed a PAG during the early to intermediate phase of an event.
- The second scenario deals with samples that are being used to determine the extent of contamination of identified radionuclides dispersed on the surface of specific soils over a wide area. The immediate need is to identify those samples taken from areas with concentrations below a designated action level (e.g., less than 10^{-4} risk).
- The third scenario assumes that an event has occurred where fresh fission products have been released. Such an event could be the result of a containment breach at a nuclear power facility or from a terrorist cell detonating an improvised nuclear device. The analytical support from the laboratory will be focused on identifying and quantifying gamma-ray emitting radionuclides and on principal alpha and beta emitters.

These scenarios may be applicable in different phases of the event, although usually the associations are: Scenario 1 – early phase, Scenario 2 – intermediate phase, and Scenario 3 – the result of a mixed fission product event that may span a long time period well after the recovery phase. The first two scenarios deal with events that would involve single or multiple radionuclides that result from individual or unrelated sources. In the third scenario, the radionuclides to be determined are intermediate- and long-lived fission and activation products that will be present with short-lived fission products. This represents a specific closed set of potential radionuclides, most of which will be determined via gamma ray spectrometry. Some of the radionuclides *not* determined by gamma-ray spectrometry initially will be ^{90}Sr, ^{234,235,238}U, and $^{239+240}$Pu, while radionuclides such as ^{99}Tc, ^{129}I, and ^{135}Cs will be examined under long-term surveillance. Other radionuclides are also possible and likely will be investigated, but those cited here probably have the greatest significance to dose.

Examples. Each of the scenarios will be addressed with a specific example that includes analytical values for the samples so that the reader will be able to see how the flow charts may be implemented. Each example also will provide a timeline for the laboratory processes so that the response times can be put into the perspective of the incident.

B. Goals

The ultimate purpose of the overall process described in this guide is to ensure that public health is protected. The recommendations in this guide are based upon PAGs for the early phase and first year of exposure, or 10^{-4} to 10^{-6} cancer risks for longer term exposure to each of the listed radionuclides. Sampling of soil to a specified depth and over a certain area needs to be integrated into the project data quality objectives (DQOs) as these concentrations of soil are used to calculate the dose/risk-based corresponding soil concentrations. The depth and areal distribution form the basis of the models for resuspension/inhalation and direct radiation exposure found in Tables 6 to 9. The specific methods and assumptions used in these models are cited in the Federal Radiological Monitoring and Assessment Center (FRMAC) Assessment Manual, Volume 1 (FRMAC 2010). The incident commander (or his designee) will need to specify what the depth of sample is for the incident.

This document does not address long-term distribution and migration of radionuclides from the surface of the soil due to weathering over a long period of time. The sampling regime required for soil penetration beyond the surface will likely be an event-specific measurement quality objective that is based on the type of soil, the specific climate of the area, and the type of radionuclide under investigation.

C. Radiological Scenarios

The response to radiological events can be subdivided into three phases: early (onset of the event to about day 4), intermediate (about day 4 to about day 30), and recovery (beyond about day 30). This guide concentrates on the time from the end of the early phase, through the intermediate and recovery phases. During the early phase, analytical priorities need to address the protection of the public and field personnel due to potentially high levels of radioactivity. During the intermediate phase, the radionuclides and matrices of concern have been *identified,* and the *quantitative* levels suitable for making decisions based on action levels need to be rapidly determined. Laboratories performing analyses must focus on rapid turnaround of sample results and optimized sample analysis so that rapid determination can be made of whether or not PAGs have been exceeded. During the recovery phase, the screening techniques used for sample prioritization may be less effective because activity concentration will begin to approach background. The focus here will be on the latter portions of the flow charts, which entail the specific radionuclide analyses and a prioritization of samples for processing.

Three distinct radioanalytical scenarios and associated flow charts are presented for soil potentially contaminated with radionuclides. The first two assume that the radioactive material is unknown.

- In the first scenario, samples are being taken close-in to where the event occurred. The soil will be highly contaminated with an unknown quantity of yet unidentified radionuclides. The first priority is to determine how the health and safety of the public and emergency workers may be affected by the level of contamination.

- The second scenario deals with samples that are taken either later in the event or further from the initial event site. The laboratory analyses of these samples help to determine the extent of areal contamination of the identified radionuclides.

- The third scenario examines the sampling issues that may arise following an event where fresh fission products have been released from a power plant containment breach or an improvised nuclear device (IND). The major screening technique in this event will be gamma spectrometry because so many of the fission products are gamma emitters. This scenario poses a much different challenge from the previous two not only because of the number of radionuclides that result from such an event, both short- and long-lived, but also because of the radionuclides' wide range of chemical reactivity in the environment.

The priority in the first radioanalytical scenario is to identify all the radionuclides present near the incident center and their estimated concentrations in the soils sampled as compared to the

PAGs. This is most likely to occur in the early phase. The need to identify the extent of the spread of contamination (Radioanalytical Scenario 2) most likely will occur in the intermediate phase and into the recovery phase. Once the radionuclides are identified, the modified flow chart (Radioanalytical Scenario 2) for either the intermediate or recovery phase may be used depending upon the direction from the incident commander or on-scene coordinator.

Radioanalytical Scenario 3 deals with the processing of samples following an IND or nuclear power plant incident. Here, the most effective preliminary screening will be performed using gamma spectrometry analysis. The flow chart in Scenario 3 may be used from the early through the recovery phase. Although there are specific fission products that are not gamma ray emitters (^{90}Sr, ^{99}Tc, etc.), this flow chart provides a good general approach to sample prioritization as both classes of radionuclides will appear in samples.

The attached charts and accompanying numbered notes and data tables depict the anticipated analytical flow required to respond rapidly and consistently. In keeping with concepts of the *Multi-Agency Radiation Laboratory Analytical Protocols Manual* (MARLAP 2004), this guide does not prescribe the use of specific analytical methods. A performance-based approach for the selection of appropriate analytical methods by the laboratory will be used to achieve measurement quality objectives (MQOs) specified by this document and incident responders.

MQOs are statements of performance objectives or requirements for selected method performance characteristics. Method performance characteristics include the method's:

- uncertainty;
- detection capability;
- quantification capability;
- applicable concentration range;
- specificity; and
- ruggedness.

An example MQO for the method uncertainty at a specified concentration, such as the action level, might be:

> "For radionuclide specific analysis of ^{241}Am, a required method uncertainty (at 1σ) of 8.2 pCi/g or less is required at the 10^{-4} risk level (first year exposure) action level of 65 pCi/g."[5]

The MQOs and any other analytical requirements serve as the basis for the laboratory's selection of a method under a performance-based approach. The laboratory should have performance data to demonstrate the method's ability to achieve the project-specific MQOs.

The scenarios presented in this document are examples of how to establish MQOs based on the tables of soil equivalent concentration values for different PAGs. MQOs specific to an event will be developed by the incident command and project personnel to address a particular event. However, in order to have an analytical approach in place to address a variety of incident

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[5] This assumes a tolerable error rate for Type I and II errors of 1% and 5%, respectively and a discrimination level of 32.5 pCi/g.

scenarios, the identified decision points in the accompanying flow diagrams provide some specified MQOs — primarily in the form of required method uncertainties — for analyzing the radionuclides of concern. For example, at most of the decision points in the diagrams where a quantitative value is needed for a radionuclide specific analysis, a required method uncertainty of 13 percent of the action level is used. In a few cases, an MQO in the form of a required detection limit is used. Once the appropriate method has been selected, then based on the required method uncertainty or detection limit, the laboratory can select the proper aliquant size, counting time and other parameters to meet the MQOs in the most efficient manner.

D. Analytical Response Time

Decisions regarding the extent of contamination in surface soils will need to be made in a timely manner. Approximate times required for laboratory processing of these samples and finalizing the sample results are shown in Appendix IV. This identifies the workflow for making qualitative and quantitative measurements of high-activity concentration contaminated soil samples (Radioanalytical Scenario 1). In addition, results of the sample radioanalytical measurements need to be communicated promptly by the laboratory to the project manager (PM) so that decisions regarding movement of population, sheltering, and additional sampling can be made.

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Consensus Methods

Some recognized methods (i.e., published through a standards organization either nationally or internationally) and some published in the refereed literature for the analysis of radionuclides in soil, are included below. The American Society for Testing and Materials (ASTM) methods may be purchased online from www.astm.org. However in every case the individual laboratory must perform method validation in their own laboratory for a soils matrix to ensure that the results will conform to the needs of the incident.

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II. RADIONUCLIDES

Table 1 identifies potential radionuclides[6] that could be used in a radiological dispersal device (a “dirty bomb” or RDD), and Table 2 identifies those that would result from a release of fresh fission products (such as a nuclear power plant breach or an IND). These radionuclides could subsequently contaminate soils in the vicinity, and downwind of, the event.

Several of the radionuclides in these tables have progeny that also are radioactive. Thus, if ^{241}Pu is found, ^{241}Am will be present.[7] However the extent to which progeny may be present will depend on the age of the material used in the RDD.

Similarly, several pairs of radionuclides are shown in Table 2. In a fission event, if the parent (listed first) is found in a sample then the progeny also will be present. However, they may not be in radiochemical equilibrium due to differences in the chemical reactivity in the environment and the time elapsed since the event.

Table 1 – Possible Radionuclides Resulting from an RDD

Alpha Emitters [5]		Beta/Gamma Emitters	
Am-241[1]	Ra-226 [1]	Ac-227	Ir-192
Cm-242[1]	Th-228	Bi-210 [4]	P-32[2]
Cm-243[1]	Th-230	Bi-212 [4]	Pd-103
Cm-244[1]	Th-232	Bi-214 [4]	Pb-210 [4]
Np-237	U-234	Co-57	Pb-212 [4]
Po-210 [4]	U-235 [1]	Co-60	Pb-214 [4]
Pu-238 [1]	U-238 [1]	Cs-137	Pu-241 [2]
Pu-239 [1]	U-Nat	I-125	Ra-228 [2]
Pu-240 [1]		I-129	Se-75

Notes:

The following notes are for both Tables 1 and 2:

[1] Principally an alpha emitter with low abundance gamma rays; see Table 2.

[2] Beta only emitter (a small fraction of ^{241}Pu also decays by alpha emission).

[3] Parent is a low abundance or non-gamma emitter; progeny used for quantification by gamma spectrometry.

[4] These radionuclides are found in many environmental samples as a result of being decay progeny of ^{226}Ra or ^{224}Ra. Care should be taken in the assignment of their half-lives in gamma spectrometry libraries.

[5] Some alpha-emitting radioisotopes (e.g., Pu-239 and Pu-240) cannot be resolved even through alpha spectrometric measurements. The activities for both radionuclides under these circumstances are usually reported as one value for $^{239+240}$Pu.

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[6] Radionuclides with half-lives less than about 12 hours have not been included in this list (unless they are short-lived progeny of a long-lived progenitor) as it is unlikely that they will be deposited on soil and still detectable shortly after the event.

[7] The production of the $^{239+240}$Pu isotopes results in the production of ^{241}Pu as a result of multiple neutron capture. Pu-241 has a 14-year half-life. If the nuclear material is “old” measurable activity of ^{241}Am (decay product of ^{241}Pu) may be present, even though it was not the radionuclide originally part of the event.

Table 2 – Radionuclides Resulting from a Fission Event

Alpha Emitters	Beta/Gamma Emitters		
Am-241[1]	Ba-140/ La-140	Nd-147/Pm-147	Ru-106/Rh-106 [3]
U-234	Ce-141	Eu-155	Sb-125
U-235 [1]	Ce-143/Pr-143	H-3	Sr-89 [2]
U-238 [1]	Ce-144/Pr-144	I-131/Xe-131	Sr-90/Y-90 [2]
Pu-238	Cs-134	I-133	Tc-99 [2]
Pu-239 [1]	Cs-137 [3]	Np-239	Te-132/I-132 [3]
Pu-240 [1]	Eu-154	Pm-151/Sm-151	Zr-95/Nb-95
Pu-241	Mo-99/Tc-99m	Ru-103/Rh-103	Zr-97/Nb-97
Activation Products			
Co-58	Ag-110m	Cr-51	Mn-54
Np-239	Co-60	Fe-59	Na-24

a. See numbered notes following Table 1.
b. This table only represents the most likely radionuclides, with half lives greater than about one day, to be detected following a fission event.

An RDD event may not necessarily have one radionuclide; several different radionuclides and their progeny may be present. In a fresh fission product event, activation products (such as those listed in Table 2) also may be present dependent upon the type of material involved in the incident.

The flow charts that accompany each scenario enable the laboratory to evaluate the activity concentration from the specific radionuclides identified against the initial screening results to ensure large contributors to the total activity concentration have not been missed.

III. DISCUSSION

Scenario Types

Appendix I contains 14 tables that identify the analytical action levels (AAL), analytical decision levels (ADL) and required method uncertainty for the radionuclides listed in Tables 1 and 2. Analytical measurements are compared to the ADL when making decisions regarding exceeding an AAL. When the results are compared to the ADL, it is expected that the required (absolute) method uncertainty requirement has been met for the measurement. In this document the required method uncertainties are typically different for gross and specific radionuclide measurements because a different assumption is made regarding tolerable error rates (see Appendix VI).

The selection, validation, and execution of a particular analytical method rely on the ability of that method to produce a result with the required specified uncertainty, u_{MR}, at the AAL. These conditions assure that the quality of the final sample analysis data will be adequate for making critical decisions. Whenever the reported sample activity or concentration exceeds a pre-defined decision level (the ADL), appropriate action is warranted. The derivation and use of AAL, u_{MR}, and ADL are discussed in detail throughout this guide. While closely interrelated, it is important to note that the use of AAL (and associated u_{MR}) and ADL represent distinct concepts; they may not be used interchangeably but rather should be interpreted and applied according the guidelines of this document.

Action Levels: AALs and ADLs

This guide relies heavily on the use of the terms "analytical action level" (AAL), "required method uncertainty" (u_{MR}), and "analytical decision level" (ADL) in characterizing the desired levels of performance of analytical methods and the radioanalytical results for use in decisions.

The term "analytical action level" is used as a general term denoting the radionuclide concentration at which action must be taken by incident responders. The AAL will correspond to a PAG value (short-term dose-based) or a risk-based value (related to long-term health effects). Ideally, the Incident Commander (IC) will provide the laboratory with the dose- or concentration-based action level and the acceptable decision error rates. If not, this guide provides "default" values. For example, the air concentration of ^{226}Ra corresponding to the 500-mrem PAG is 1.8 pCi/m^3. Tables 7A, 7B, 7C, and 7D show the AALs associated with the 2-rem, 500-mrem, 10^{-4} or 10^{-6} risk values for selected alpha, beta, and gamma-emitting radionuclides in soil.

Incident-specific action levels different from the ones used in the tables may be promulgated. In these cases, the corresponding AALs can be calculated as a linear function of either the 500-mrem AALs or the 10^{-4} or 10^{-6} risk values (see Scenario 3 for an example of an event-specific AAL calculated in this manner).

The required method uncertainty and ADL will change depending upon the acceptable decision error rate. Tables provided in Appendix I list the AAL, ADL, and u_{MR} values for the radionuclides of concern. The tables present gross screening and radionuclide-specific measurements for alpha and beta/gamma-emitting radionuclides. Derivation of the ADL values for each of these tables can be found in Appendix VI. The listed AALs are applicable as default values based on generic conversions of the dose level to concentration in soil for a specific radionuclide. The Incident Commander (IC) may provide incident-specific action levels or decision error rates that would supersede these values. In this case, the laboratory will need to develop new tables for all values, using the process described in Appendix VI.

Three generic scenarios for soil contaminated with radionuclides are presented together with a flow chart and description of the processes associated with the laboratory handling of these samples based on direction from the incident command. The direction to be followed may be one of the following —

- Analyze the highest activity concentration samples first, or
- Analyze the lowest activity concentration samples first, or
- Analyze those samples first that have alpha activity concentration (by screening) of more than 10^3 pCi/g —

or some other characteristic that the laboratory can measure using their screening instrumentation.

In order to illustrate the typical decisions and actions to be taken by a laboratory for each scenario, examples of the scenarios using theoretical samples and measurement results are provided in Appendices II, III, and IV. These examples represent only three of many different possible permutations and should not be construed as limiting. Each example is keyed back to the steps in its respective generic flow diagram and notes.

These scenarios assume that the time period from taking the sample to the actual beginning of the analysis by the laboratory will be short (on the order of one day). Samples received by the laboratory will not have had any pretreatment performed. Sample drying, homogenization, and representative sub-sampling will need to be accomplished by the laboratory staff based on their established standard operating procedures and the guidance from the incident commander based on the project MQOs. Any sample-specific preservation or storage requirements for samples that are not to be analyzed immediately should be communicated to the laboratory by the incident commander no later than the time of the sample shipment.

For the three scenarios discussed in this guide, it is assumed that field personnel have performed some type of radiation detection survey of the samples prior to sending them to the laboratory. Laboratory staff should request field-screening measurements and descriptions of the instruments used if not received with the sample shipping papers. If appropriate, field personnel may determine which samples are to be submitted first to the laboratory based on these survey results. The laboratory's surveys and analyses of the samples are not intended to confirm the field survey results. Significant differences between field and laboratory screening measurements may exist as a result of:

- Short half-life of a particular radionuclide (either anthropogenic or naturally occurring radioactive materials [NORM]).
  - Samples saturated with ground water may have thorium progeny ^{212}Pb ($t_{½}$ = 10.6 h) and ^{212}Bi ($t_{½}$ = 1 h) that are unsupported.
- High background radiation levels in the area where the samples were initially obtained.
  - If the samples were screened *in situ*, the background from contamination over the large area where the samples are taken may be promoting higher readings of the samples.
- Ingrowth of progeny radionuclides.

  - For an IND or incident at a nuclear power plant, several radionuclides form a transient equilibrium (See Table 2). For many of these it will take days to weeks for the equilibrium to develop after an incident. The activity will increase by a factor of about two if transient equilibrium is in its early stages at the time the sample was taken.
- Different responses by the field and laboratory instrumentation to the possible radiation emission types. The response of an alpha/beta survey instrument may vary considerably depending upon:
  - The energy of the alpha or beta particle emitted by the radionuclides in the sample,
  - Self shielding within the sample matrix, which will have an indeterminate effect, and
  - The effect on the measured response.

  Therefore, screening instrumentation should be calibrated for target radionuclides when they have been identified. See Section 6 of the *Uses of Field and Laboratory Measurements During a Radiological or Nuclear Incident* (EPA 2012c).
- Uncertainty of the measurements based on the sample to detector geometry, background radiation, type of instrument used, etc.
- Dealing with soil samples, specific issues like sample self-shielding, hot particles, and non-uniform distribution can create a large measurement uncertainty dependent upon the type of screening instrument used and the skill of the analyst.

Laboratory staff may want to compare the two screening results to assess how the above issues can affect their sample prioritization.

Specific Soil Sampling Protocols

Soil sampling will likely have two general techniques. The first will take place in all phases of the incident. *The Federal Manual for Assessing Environmental Data during a Radiological Emergency* (see references) expresses PAGs for soil in terms of activity per cm^2. The manual describes collecting a sample from an area 10 cm by 10 cm to a depth of 2 cm and assumes that the soil density will be approximately 1.6 g/cm^3. Based on these assumptions the total sample size will be approximately 320 grams.[8]

Radioanalytical results and depth of deposition will be used to determine if the PAG or risk value Analytical Action Levels identified in the tables in Appendix I have been exceeded. These values are based on direct exposure and inhalation due to resuspension. The values given in these tables are based on the assumption that the exposure period is going forward from the time of sampling.

A second technique may be used late in the intermediate to recovery phase. This would involve core sampling to a specified depth of the soil. The specifics for this type of sample such as depth of core, thickness of soil slices from the core, core diameter, and fraction of material to be sampled/combined, as well as associated Action Levels would be identified in the MQOs/DQOs by the incident command staff. No specific guidance on subsurface AALs for core sampling of soil currently exists.

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[8] The exact mass needs to be measured as it will vary based on factors such as organic content, soil composition and packing, and moisture content of the sampled material.

Importance of Sample Radiological Screening and Particle Size Distribution

Neither an IND nor an RDD has been deployed, however, there have been several incidents involving radioactive materials and explosives that yielded soil contamination that would be similar to such an event. Two of the most serious events were the crashes at Palomares, Spain, in January 1966 and at Thule Air Force Base, Greenland, in January 1968.

In both incidents, the conventional explosives that accompanied the bombs detonated during the crash without setting off a nuclear explosion. The explosions in both cases ignited the pyrophoric plutonium, producing a cloud of radioactive materials that was dispersed over wide areas. These areas were contaminated with the radioactive material from the bomb components, principally uranium, plutonium, and americium. The size and distribution of particles that resulted from these events, and settled on the soils after the aerosols were dispersed, were non-uniform.

Figure 1 shows an autoradiograph of a soil sample from Palomares after it had been homogenized.

The sample (used for the autoradiograph shown in Figure 1) was homogenized and brought to grain size of between 125 and 250 µm. The white spots indicate the particles of plutonium while the entire dark area is due to soil particles without radionuclides. Visually the soil was homogeneous. However when the autoradiograph was developed the difference in size of the plutonium particles was evident even though the soil was ground to a fine mesh. It is clear from the autoradiograph that the size and distribution of these particles was non-uniform.

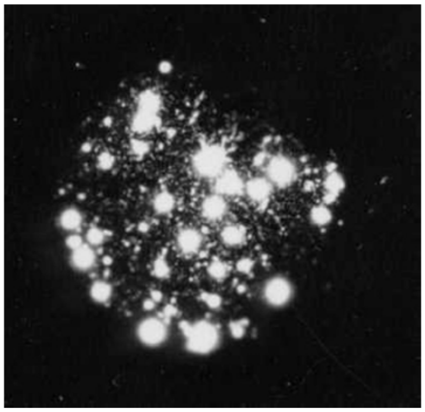

Figure 1 – Autoradiograph of Particles from Palomares Incident

In another recent research study[9] using soils from both incidents, other techniques were employed to assess not only the particle size but oxidation states of the plutonium and other actinides.

> Furthermore, results from electron microscopy with Energy Dispersive X-ray analysis (EDX) and synchrotron radiation (SR) based micrometer-scale X-ray fluorescence (micro-XRF) 2D mapping demonstrated that U and Pu coexist throughout the 1–50 micron sized particles, while surface heterogeneities were observed in EDX line scans. SR-based micrometer-scale X-ray Absorption Near Edge Structure Spectroscopy (micro-XANES) showed that the particles consisted of an oxide mixture of U (predominately UO_2 with the presence of U_3O_8) and Pu ((III)/(IV), (IV)/(V) or (III), (IV) and (V)).

Also from that study,

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[9] O.C. Lind, et al., "Characterization of U/Pu particles originating from the nuclear weapon accidents at Palomares, Spain, 1966 and Thule, Greenland, 1968," *Science of the Total Environment*, 376 (2007) pages 294–305.

> Using SEM in SEI and BEI-mode (Figs. 2 and 3), the size of the particles is estimated to be between 1 μm and 50 μm for Palomares and 20–40 μm for Thule (Table 1). However, the number of particles is limited and the ^{241}Am activity concentration of submicron particles are below the detection limits (~0.1 Bq) for γ-spectrometry. In addition, the particles tend to be imbedded in larger soil and sediment aggregates and imbedded small sized particles are difficult to identify in SEM–EDX. Thus, the particle sizes reported herein should reflect the high end of the size distribution pattern.
>
> Taking all XANES results into account, it appears that the Palomares and Thule particles contain a mixture of U (predominately IV with the presence of VI) and Pu ((III)/(IV), (IV)/(V) or (III), (IV) and (V)), most probably in the form of mixed-oxides/oxyhydroxides and not as ordered actinide-O_2 structures.

In both studies, the soils were initially dried and then brought to a specific particle size range, followed by analysis for gross radioactivity. The gross measurements were made either by survey instrument, gamma spectrometer, or using autoradiography to assess sample homogeneity. It also is important to note that the radioactive material in some cases had become incorporated with larger soil aggregates. This likely occurred as a result of aging and weathering of the soils *before* the sampling occurred.

The time dependence of the change in particle size, change in oxidation state of the radionuclides present, or depth of penetration into the sampled medium was not a part of either of these studies. Depending upon the type of incident, radioactive material involved, chemical reaction leading to the dispersal of materials and weather conditions, different soil penetration and particle aggregation will occur. However, no other easily-obtainable references have been reported in the open literature since these incidents occurred that identify how the concentration in the surface soil would change as a function of time.

The importance of these studies as they relate to either an IND or RDD is that they:

- Identify the *non-homogenous* distribution of the particulate matter (soil) that may exist in the soil samples,
- Demonstrate the variety of particle sizes (associated with the radionuclide) that may be formed or exist, and
- Show the variation in oxidation state for actinides if present.

Analysis of these samples by gamma spectrometry and radiography (as in the above references) requires adequate homogenization but does not require rigorous dissolution of the samples to identify the gamma emitting radionuclides or “hot” spots. The radiographic evidence has shown that discrete radioactive particles (DRPs) do exist in an RDD-type incident. In any radiological event where temperatures are elevated to the thousands of degrees Fahrenheit, DRPs would also form and likely be refractory. Such DRPs would be of an entirely different composition than the soil they contaminate. This makes the analysis of non-gamma-emitting radionuclides more difficult and emphasizes the need for soil dissolution techniques that completely dissolve the sample, including DRPs. Because chemical separation techniques are required for the non-gamma-emitting radionuclides, additional oxidative-reductive processes may be necessary in order to bring the radioisotopic species present to one, common oxidation state.

Laboratory Preparation of Samples

Only laboratories using validated radioanalytical methods (see MARLAP, Chapter 6) should be used in order to process samples in a timely and effective manner. These laboratories will have the necessary radioanalytical capability and sample-processing capacity to conduct the gross screening and radionuclide-specific analyses defined for the radioanalytical scenarios in a timely and effective manner. This guide recommends the following analytical process flow by the laboratories when soil samples are received.

1. General screening based on radiation emitted from the sample (i.e., alpha, beta, and gamma).
2. Initial homogenization of the bulk sample so that aliquants removed will have a high probability of being representative of the whole sample if analysis of volatiles is needed.
3. Sample pretreatment involving drying (when applicable), further homogenization (e.g., ball mill treatment), and sieving and blending (to permit taking of smaller representative subsamples if required by the incident data quality objectives).
4. Sample treatment by leaching or complete dissolution by hydrofluoric acid digestion or salt fusion technique.
5. General radiological screen of individual fractions using liquid scintillation or gas proportional counting following dissolution.
6. Comparison of the pre- and post-sample treatment/dissolution screening values.
7. Specific radioanalytical techniques applied after dissolution for the samples designated as priority by the incident command (may be highest or lowest activity concentration samples that are the priority).

These steps are shown in Figure 2.

This is the sample preparation sequence used for radioactivity screening in the flow diagrams for each radioanalytical scenario described in this document. A sample aliquant may be taken following any of the steps in this process as no sample preparation except for homogenization is necessary. Thus an analysis by gamma spectrometry could be done after Step 2, 4, or 6. Each of the yellow boxes in Figure 2 represents an action that should be stipulated by the incident DQO and MQO statements. For example yellow box B states the detritus removed from the sample is separately assessed for radioactivity. This action would be a project-specific requirement and separate MQOs would need to be established for this situation.

The three scenarios described in Sections IV, V, and VI each contain a flow diagram where a decision point in the flow diagram relates to an action level (PAG, regulation, or risk-based dose limit). It also is important to note that the AALs and MQOs will likely be different for the screening process versus the radiochemical determination steps. Many of the flow diagram boxes have numbers indicating the sequence of the analytical process. The boxes are color-coded indicating the most important flow path (red) to the least important (yellow) based on the time requirements for returning the analytical results.

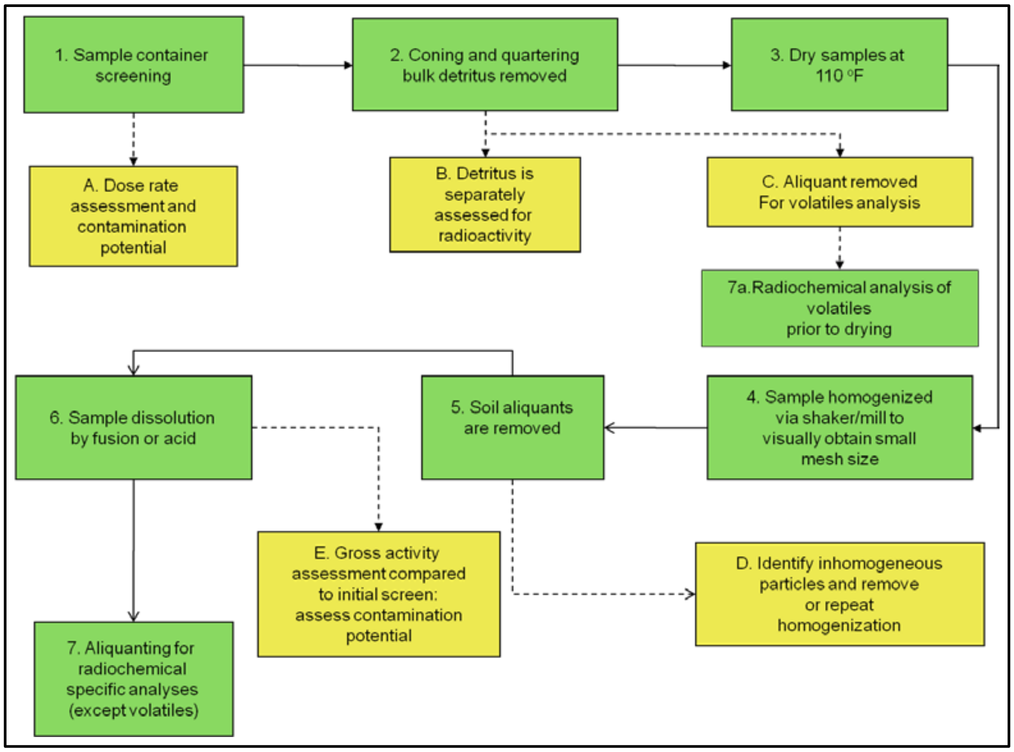

Figure 2 – Initial Screening and Pretreatment Process

The screening techniques[10] outlined in the first steps of the flow charts assume that the laboratory is equipped with instrumentation that can perform the screening functions identified below:

- Micro-R meters that can be used to evaluate radiation exposures or doses on incoming samples.
- Hand-held gross alpha detector that can be used to assess the alpha contamination of container surfaces by swipe analysis.
- Determine the alpha count rate directly on the sample surface (although this may not be very effective except for those cases of very high alpha contamination levels).
- Thallium-activated sodium iodide (NaI(Tl)) or high-purity germanium (HPGe) detectors that can be used in energy or total counting modes to perform gross gamma activity analysis of samples. (Laboratory staff must ensure that they are using library data from the National Nuclear Data Center [www.nndc.bnl.gov/] or another recognized source of nuclear data that will be comparable.)

The laboratory also should have the instrumentation to perform radionuclide-specific analyses (e.g., liquid scintillation , gamma spectrometry, and alpha spectrometry).

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[10] Specific recommendations for sample screening processes are identified in *Radiological Laboratory Sample Screening Analysis Guide for Incidents of National Significance* (EPA 402-R-09-008, June 2009).

Each numbered box has associated with it a note that provides additional detail for that particular part of the process. Clarification also is provided in these notes as to when parallel paths of analysis should be followed to help expedite the processing of samples.

Appendix V contains an example of a spreadsheet using generic assumptions that can help laboratory personnel in assessing count times for samples. The spreadsheet demonstrates how the user can determine the time it will take to achieve the required method uncertainty for the decision points in the flow diagram for two different screening methodologies. Laboratories should prepare their own spreadsheets, in advance of an event, using their preferred methodology. Laboratories also should determine (in advance) whether their individual analytical protocols will need to be revised to accommodate this process. The flow charts used in this document that describe the screening process use gas proportional counting for the aliquant sizes and count times. However, liquid scintillation also may be used for this process. In both cases the laboratory should have a validated method that provides appropriate guidance to laboratory staff on how the screening results relate to overall sample activity concentration. Appendix V has approximate times and sample sizes for this method of analysis as well.

The number of samples that will be analyzed, and their level of contamination, may be significantly higher than normal samples. Laboratories should use the guidance provided in *Guide for Radiological Laboratories — Identification, Preparation, and Implementation of Core Operations for Radiological or Nuclear Incident Response* (EPA 2010b)*; Guide for Radiological Laboratories for the Control of Radioactive Contamination and Radiation Exposure* (EPA 2012b)*;* and *A Performance-Based Approach to the Use of Swipe Samples in Response to a Radiological or Nuclear Incident* (EPA 2011) when:

- Assessing the need for separate sets of procedures for sample handling and storage.
- Increasing the frequency of detector background analyses.
- Increasing the frequency of quality control (QC) checks.
- Adjusting the QC check activity concentration level to more closely align with the activity concentration of the anticipated samples.
- Increasing the frequency of contamination assessments (i.e., smears/swipes) on working surfaces in the laboratory.
- Writing separate protocols for personnel protective equipment.
- Writing separate protocols for personnel and sample radiation monitoring.
- Creating separate storage location for high activity concentration samples or a large group of samples that would increase laboratory background for detectors or increase exposure to personnel.

It should be noted that the procedures that have been in place for the last 30 years may have been modified to account for the low concentrations of anthropogenic radionuclides normally encountered. Should an RDD or IND be deployed, and it contains a radionuclide that has radioactive progeny, it is possible that the radioactive equilibria involved with these progeny will have been established. This means that not only will there be considerably higher concentration of the parent but of each of the progeny. Furthermore, if multiple radionuclides are involved, the cross-contamination factor during separations must be minimized, a phenomenon that current day analysts may not have previously experienced.

A specific example of such a phenomenon would be the elimination of ^{140}Ba ($t_{½}$ = 12 d) during the ^{90}Sr separation process. Currently, several methods in use do not account for ^{140}Ba removal. Strontium-specific resin separations (for example using an Eichrom™ resin) use 8 M HNO_3 to minimize retention of barium. However, this method is not designed to remove large amounts of interfering beta emitters (~1,000 times greater than ^{90}Sr levels). In these instances of samples with very high beta activity, it will be beneficial to perform a double column separation to ensure adequate removal of such beta interferences. Generally, the most likely source of this radionuclide would be a release from a nuclear power plant.

Analysis of soil samples for radionuclides represents a more difficult process than for water or air samples. This is not only due to the different types of soil presenting challenging matrices to digest during sample preparation, but also due to the presence of measurable concentrations of several different naturally occurring radionuclides. The decay products of uranium and thorium can provide a significant gross activity measurement to some soils when no anthropogenic sources are present.

Each natural series has lead, bismuth, and polonium radionuclides that are related via direct equilibrium. The radium isotopes decay to these radionuclides via different radon gas radioisotopes. This can cause significant differences between *in situ* field measurements and laboratory screening measurements. Such differences may have an effect on the prioritization assigned to a specific sample.

Furthermore, soils contain ^{137}Cs and ^{90}Sr from atmospheric bomb testing that occurred during the time period of 1950–1980. Although the mean concentrations are typically very low (~0.2 and ~0.05 pCi/g, respectively) and will not affect screening results, the soil concentrations of these radionuclides should be established before using new data to assess spread of contamination in the event these radionuclides were used in the RDD event. EPA's *Response Protocol Toolbox* (EPA 2004) provides additional recommendations concerning planning and threat management, site characterization and sampling, and sample analysis to assist utilities and state and local agencies. If laboratory protocols for non-emergency situations cannot ensure that the DQOs and MQOs are achievable with the laboratory's standard operating procedures (SOPs) under emergency conditions, then a separate set of SOPs for incident conditions will need to be developed.

IV. RADIOANALYTICAL SCENARIO 1 (IDENTIFYING SAMPLES WITH HIGHEST ACTIVITIES)

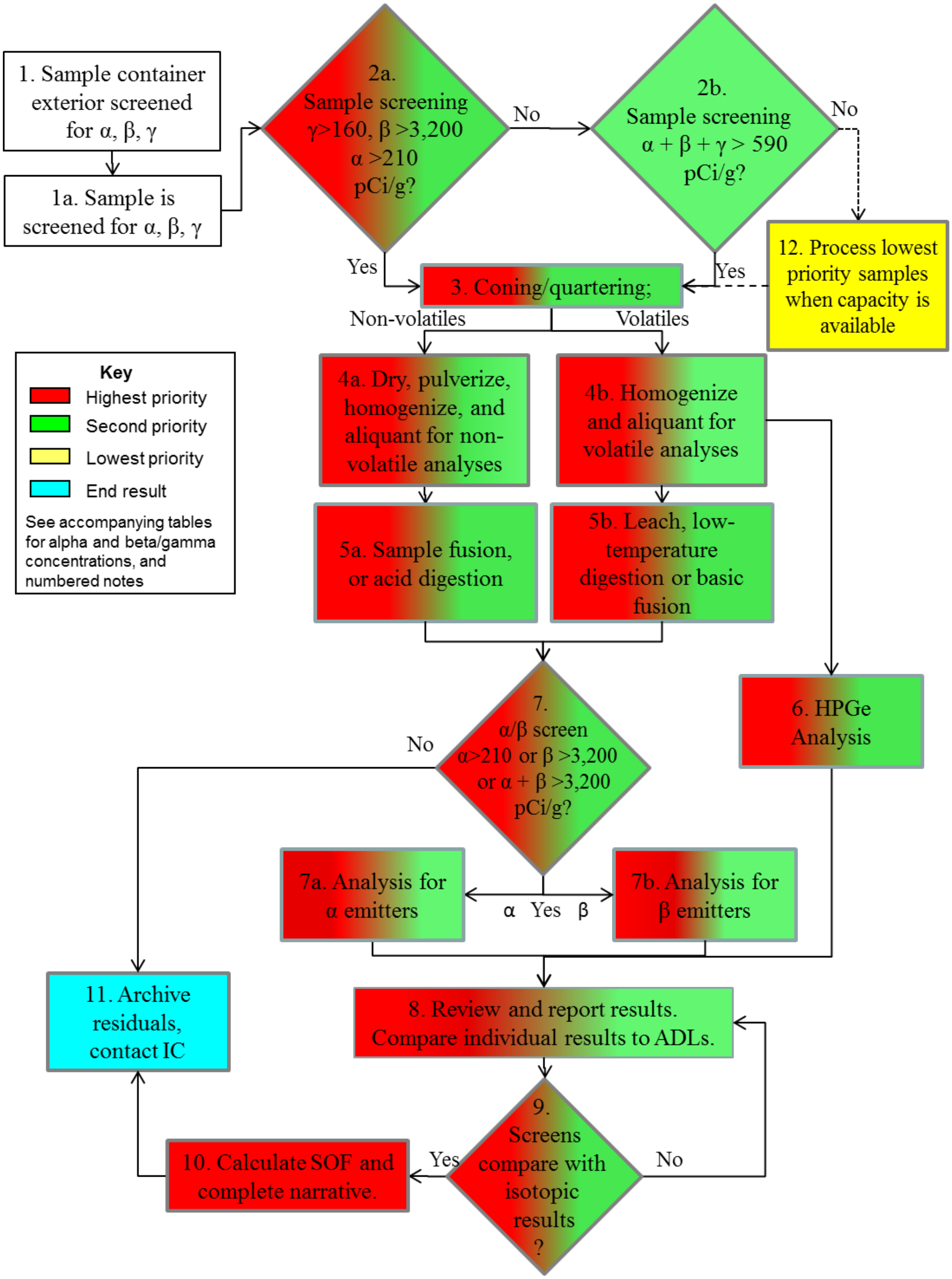

Figure 3 – Radioanalytical Scenario 1 (Identifying Samples with Highest Activities)

Notes to Scenario 1:	**Source of Contamination: Unknown**
Purpose:	**Priority to Those Samples with Highest Activities (early into intermediate phase) — Greater than the 2,000-mrem PAG**

Highest priority samples should be analyzed first. Only after the highest priority samples have at least been started (excess capacity available) or completed should lower priority samples be addressed. The samples may arrive over several days. Lower priority samples (those following the green and yellow flow paths on this chart) may need to be stored for several days until the highest priority samples have been analyzed.

The radionuclide activities corresponding to PAG values for this scenario can be found in Tables 6A and 6B for screening and 6C and 6D for radionuclide-specific activities.

1 The determination of gross γ, α, and β sample concentration relative to the PAG values is left until the next step. Sample container exteriors are surveyed to assess whether there is indication that contamination is present.

These might include a Geiger-Mueller counter with appropriately calibrated beta and gamma detector probes or a micro-roentgen meter (gamma only).[11] This step should be performed with the sample container unopened. Surface swipes of the container are taken to assess for external gross α/β and γ contamination. These surveys are for contamination control purposes and not for the assessment of sample processing flow path.

Any container with measurable external contamination should be decontaminated before proceeding with subsequent analysis. This will help to minimize sample cross-contamination as well as laboratory contamination.

1a *Gamma Screening*

Unless the identity of the radionuclide contaminant(s) is known, the screening instrument should be calibrated for ^{60}Co.[12] MQOs will correspond to those defined for ^{60}Co first year 2,000 mrem PAG for screening measurements (see Table 6B). A ^{60}Co source distributed through a 300-gram soil matrix counted in the same geometry as the sample should be used for calibration of the screening instrument.

If a limited list of radionuclides of concern is known, it may be practicable to use a calibrated NaI(Tl) detector to assess gross gamma activity concentration and relate this measurement directly to a PAG concentration. The gross gamma activity concentration can be determined using either a NaI(Tl) or HPGe detector using the total counts between 40 and 2,000 keV accumulated over a short time period (about 10 minutes).[13]

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[11] Some manufacturers have developed kits that include the survey meter plus an alpha-beta-gamma pancake GM detector and a NaI gamma detector.

[12] The gross gamma activity as determined by integrating the entire spectrum of a NaI(Tl) or HPGe detector above about 40 keV. Appropriate efficiency corrections for each detector would need to be applied.

[13] *Radiological Laboratory Sample Screening Guide for Incidents of National Significance*, EPA 402-R-09-008 (June 2009).

Some gamma-emitting radionuclides may not be detected at their PAG concentration levels if the sensitivity of the instrument used is inadequate, or for very low-abundance or low-energy gamma emitters. Radionuclides that do not emit γ- or X-rays greater than approximately 40 keV (including pure alpha and beta emitters such as ^{210}Po or ^{90}Sr) may not be detected. If multiple gamma ray emitting radionuclides are known to be present, it may be difficult to assess if an AAL has been exceeded with a gross gamma measurement. It may be possible to identify samples where an AAL for a gamma emitter will not be exceeded. By calibrating the survey instrument using the beta/gamma radionuclide having the "limiting AAL" for the 1000 mrem PAG listed in Table 6B and the geometry used for the screen the gross gamma measurement can be compared to the respective "limiting" ADL.[14]

1a *Gross Alpha and Beta*

The methods described here present a rapid, positive approach to screening of soils, involving digestion or aliquanting methods commonly used in the radiochemistry laboratory; it requires open handling and processing of samples that may contain elevated levels of radioactivity. Laboratory staff will need to take specific precautions while performing these techniques to avoid personal contamination (including inhalation), laboratory surface contamination, and sample cross-contamination since the radioactivity level of pure alpha or beta emitters in the sample is unknown. The lab should develop guidelines for their staff outlining the personal protective equipment required for different levels of gross alpha or gross beta activity based on screening results. Additional precautions should be taken to have a separate area for the screening process.

The samples received from a radiological incident may come in a variety of containers: plastic bags, plastic bottles, glass bottles, Petri-style containers, aluminum cans, etc. The masses of the samples received also will vary significantly. If possible, using several different style containers with different masses of standards for use in calibration of screening equipment will be helpful in minimizing the time for the initial sample screening process.

Screening of the sample for α and β activity must be done carefully as the gamma screen may not yield any indications of elevated activity concentration when only pure alpha- or beta-emitting radionuclides, or radionuclides that only emit γ- or x-rays below the calibrated range of the instrument. A very rudimentary screening process, performed in a hood, should be used prior to removing the entire sample from its container for homogenization and sub-sampling. Some options for making such a measurement may be:

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[14] The limiting AAL and ADL for gross gamma measurements cannot be used to demonstrate that an individual AAL has been exceeded, rather it can only show that an AAL will not be exceeded obviating the need to perform further gamma measurements. The limiting AAL is the one that produces the lowest gamma response at its respective AAL (i.e., the lowest product of AAL and gross gamma efficiency). It is most reliably determined by calibrating the detector for gross gamma in the geometry to be used with each gamma-emitting radionuclide that may be present. This approach may not be practicable if a large number of gamma emitters may be present.

- Take a swipe of the inside of the container cap. Then, using an alpha-beta probe, determine the count rate of the swipe.
- Insert a tongue depressor (with double-sided tape on one surface) into the container and remove for direct measurement.
- Take a small aliquant of the top surface (after the sample container is shaken) and spread it onto a planchet.
- Take a measured aliquant and place it into a scintillation vial with cocktail for liquid scintillation analysis.
- Perform a rapid nitric-hydrofluoric acid digest (total dissolution is not necessary) on a small aliquant of the sample and evaporate onto a planchet.

For the first three bullets above, an initial estimate of the mass transferred using the specific technique should be made with a soil of similar composition. The probe used for such techniques should have been calibrated with soil samples spiked to a level simulating the AAL values.

The last two options above, LSC and gas proportional counting (GPC) may provide more quantitative measures for gross alpha/beta screening of samples. Note that a screening method that evaluates gross alpha/beta activity concentration directly on soil is not included.

LSC can provide a faster screening process and avoid some of the shortcomings of a simple sample leaching process needed for GPC because the effects of self-absorption in the sample test source are minimized using LSC. The procedure used must correct for quench from various sources in the soil, and the method must be validated prior to use on samples from an incident. It may be impractical to separate alpha from beta signals using pulse-shape analysis in the case of high levels of quench. Instead, a total activity measurement (combined alpha and beta) across the entire energy spectrum may provide the most reliable estimate of activity in highly quenched samples.

For soil samples, the question arises of how to introduce a representative sub-sample into the liquid scintillation matrix. Although there are no standard methods that specifically address this issue, several general approaches are commonly used. The first, perhaps more conventional approach, involves direct measurement of the solid; the second involves leaching or digesting a representative aliquant of the solid sample and processing the leachate / digestate as if it were an aqueous sample. The third approach involves directly suspending a small, representative aliquant of the soil in a gelling liquid scintillation cocktail.[15]

The limiting factor in using gross alpha beta measurements for reliable decisionmaking is obtaining a representative aliquant. In contrast to the gamma measurement where the entire sample is usually measured, the relatively small portion of unhomogenized sample may not be representative of the entire sample. The small aliquant used may fail to

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[15] Cocktails such as Insta-Gel, or Quicksafe A may be used since they will suspend the solid and allow it to be counted in a 4-π geometry.

identify non-homogenously distributed activity present in a sample. The alpha / beta screen may still provide important information with regard to handling and the need for personal protective measures since the next steps involve working with the entire sample outside of the sample container.

> The gross radioactivity measurements used for comparison in Step 2 are taken from the ADL values shown in Tables 6A and 6B (^{60}Co for gamma; ^{226}Ra for α, and ^{90}Sr for β) for the 2,000-mrem PAG. These represent the ADL values for the listed radionuclides that are likely to be present.
>
> No conclusions about the presence or absence of these or other radionuclides should be made at this point in the analytical process.
>
> The laboratory should weigh the time spent performing screening at these low concentrations versus the time taken to perform radiochemical-specific sample analysis.

2a

While gamma measurements may meet MQOs for required method uncertainty prior to complete homogenization of the sample, high levels of uncertainty associated with the alpha/beta screening techniques discussed above may preclude use of the gamma measurements for making reliable screening decisions until a representative aliquant can be obtained following homogenization of the sample. It is important to ensure that estimates of the measurement uncertainty are robust, and that MQOs for uncertainty have been met before using data to decide that a sample concentration exceeds an AAL. If estimates of uncertainty are deemed to be unreliable, the results should not be used for critical decisionmaking. Thus it may be necessary to delay decisions about whether AALs for non-gamma-emitting radionuclides have been exceeded until the sample is milled and homogenized and a representative aliquant can be obtained.

Measurements resulting from the screening process that satisfy the MQOs for required method uncertainty are compared to the limiting ADL corresponding to the first year 2,000 mrem PAG[16] to determine if an AAL may have been exceeded. Samples that exceed the default gross screening ADL values for gross alpha of 210 pCi/g, or gross beta of 3,200 pCi/g, or gross gamma of 160 pCi/g will take the red path (highest priority). If the values for gross alpha and beta are taken as an aggregate measure (i.e., the sum of the gross alpha and beta activities), then the more restrictive ADL value of 210 pCi/g for alpha response is used to assess if the screening ADL has been exceeded. Samples that exceed the values identified in this step stay on the red path and go to Step 3.

Sample results that exceed either of these values should be communicated immediately to the IC so that decisions regarding the elevated activity concentration from these sample locations can be made in a timely fashion. This feedback also will reinforce the priorities assigned to each sample and further enhance decisionmaking.

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[16] Depending on the time of the response, a 1,000-mrem PAG for the first four days of exposure may be requested by the incident command. If so, use the radionuclide concentrations corresponding to the 1,000-mrem PAG in Tables 6A and 6B.

2b Samples with a combined alpha + beta + gamma activity of greater that 590[17]pCi/g are placed on the green flow path and analyzed when time permits after completing the red path analyses. Lower activity samples are stored for analysis after all higher priority samples have been processed (see Step 12).

> NOTE: The dashed lines connecting Boxes 2a, 12, and 3 in the flow chart indicate that analyses for short-lived or volatile radionuclides may need to proceed more quickly due to concerns about short half-lives or chemical stability of the target radionuclides.

3 A process described as “coning and quartering” is used to break the entire bulk sample down into representative subsamples. Coning and quartering of the sample provides reasonable assurance that subsamples will be representative of the whole sample. Unless the project has a specific MQO to the contrary, remove extraneous detritus (e.g., sticks, twigs, rocks larger than 1/4", etc.) prior to “coning and quartering.” The material removed from the sample should be retained for potential future analysis and an appropriate comment added to the case narrative.

Fractions are isolated for volatile radionuclides and gamma spectral analysis (Step 4b), and for analysis of non-volatile radionuclides (Step 4a), where required. The volatile fraction should be processed quickly and immediately sealed in containers to minimize potential losses of volatile radionuclides.

The details of subdividing the sample can be found in *Rapid Method for Fusion of Soil and Soil-Related Matrices Prior to Americium, Plutonium, and Uranium Analyses; Rapid Method for Radium-226 Analyses in Soil Incorporating the Fusion of Soil and Soil-Related Matrices; and Rapid Method for Sodium Carbonate Fusion of Soil and Soil Related Matrices Prior to Strontium-90 Analysis* (EPA 2012a). The volatile fraction should be processed quickly and immediately sealed in containers to minimize potential losses of volatile radionuclides.

Precautions should still be taken (e.g., opening the sample container and handling in a hood) since the alpha/beta activity concentration has only been assessed by bulk screening techniques.

4a The homogenization and size reduction can be accomplished using an approach such as that described in the above-referenced method (EPA 2012a). This approach has been used on smaller scales than that described in ASTM C-999[18] and would facilitate preparation of representative samples for a batch in approximately 2 hours time.

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[17] The value of 590 pCi/L is derived from, 160(gross gamma) +210 (gross alpha) +220 (lowest ADL for a beta only emitter ^{227}Ac). The ^{227}Ac is used since there is a potential for this being an IND type scenario.

[18] ASTM C-999, *Standard Practice for Soil Sample Preparation for the Determination of Radionuclides*.

Place the non-volatile fraction isolated in Step 3 in a tared can. Remove the lid and dry to constant weight in an oven at 110±10 °C. Drying samples provides for consistent comparison of results regardless of moisture content.

This is the dry-weight basis for the analysis. This mass is also used to calculate the fractional solids content for each sample (i.e., percent solids) so that results from the volatile fraction, analyzed "as received" can be converted to and reported on a dry-weight basis.

Constant weight may be determined by removing the container from the oven and weighing repeatedly until the mass remains constant to within about 1%. This also may be achieved operationally by observing the time needed to ensure that 99% of all samples will obtain constant weight.

Once the sample is dried, milling and homogenization continues using the same vessel (the paint can). Stainless-steel or ceramic balls or rods are added to the can and the can is shaken for about 5 minutes, or as needed to produce a finely-milled, well-homogenized, sample. The precise particle size of the milled sample is not critical as long as the milled sample is fine enough to facilitate rapid and complete dissolution of the soil matrix during the dissolution process.

NOTE: A qualitative, visual standard can be prepared by passing quartz sand or other milled material through a 50-mesh and then a 100-mesh screen. The portion of the sample retained in the 100 mesh screen can be used as a qualitative visual standard to determine if samples have been adequately pulverized. The process is complete once 95% of the sample (or greater) is as fine, or finer, than the qualitative standard. If, by visual estimation, more than ~5% of total volume of the particles in the sample appear to be larger than the particle size in the standard, return the sample to the shaker and continue milling until the process is complete.

If, by visual inspection, the sample appears to contain larger particles that may not be effectively dissolved during the fusion or digestion process, those particles may be preferentially removed prior to aliquanting. In most cases, removal of a small fraction of larger particles will still provide representative results because the surface area of larger particles is relatively low and the surface will be abraded during milling. As a result, the activity associated with sample fines should be representative of that found in the original sample. A comment should be added to the sample narrative addressing removal of the solids.

The details of the milling and homogenization process are presented in *Rapid Method for Fusion of Soil and Soil-Related Matrices Prior to Americium, Plutonium, and Uranium Analyses*; *Rapid Method for Radium-226 Analyses in Soil Incorporating the Fusion of Soil and Soil-Related Matrices*; and *Rapid Method for Sodium Carbonate Fusion of Soil and Soil Related Matrices Prior to Strontium-90 Analysis* (EPA 2012a).

At this point, representative aliquants of the dried, homogenized sample can be taken for radionuclide-specific analysis for non-volatile components. A separate aliquant is also removed for gross alpha and beta screening and subjected to the same dissolution process as are samples for radionuclide-specific analyses. Note that no tracers/carriers are added to the gross alpha beta aliquant as these will interfere with the determination of gross alpha and beta activity.

Prior to or concurrent to Step 5a, soil samples with significant organic content should be ashed to completely combust organic matter. This can be accomplished using a programmable furnace where a temperature of approximately 600 °C can be achieved. Ramping up the temperature in time intervals is necessary to avoid uncontrolled ignition and excessive smoke generation during the process. If the sample is ashed prior to aliquanting, record the wet, dry, and combusted weights so that the wet/dry ratio (i.e., percent solids) and the percent organic content (if required) can be calculated and used to convert final results to a dry-weight basis

4b Representative portions of the undried sample fraction are aliquanted as needed for analysis of volatile radionuclides and for HPGe gamma analysis. The sample should be mixed, homogenized, and subsampled rapidly and in a manner that ensures that minimizing loss of volatiles and that aliquants will be representative of the original sample.

Aliquant size should be planned to optimize count times and throughput while ensuring that method uncertainty requirements are met.

Analyses for volatile radionuclides should be started as soon as possible to minimize loss of analyte. If sample analysis cannot be started immediately, a project-specific means of sample storage for volatiles should be in place so that even short-term storage does not significantly reduce their concentration.

Steps 5 and 6 can be performed concurrently.

5a Non-volatile radionuclides are prepared for chemical separations using validated techniques for total dissolution of the sample. The dried or ashed aliquant of the sample is dissolved to create one or several stock solutions, as appropriate for the analyses to be performed.

For samples resulting from an RDD, it is important to achieve complete dissolution. Certain materials such as those used in a radioisotope thermoelectric generator (RTG), brachytherapy sources, or matrices that have been exposed to a high temperature detonation, may be difficult to dissolve using conventional acid digestion techniques. Sodium carbonate or sodium hydroxide fusions are two good methods for obtaining complete dissolution of soil samples.[19] Fusions help ensure isotopic exchange of analyte

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[19] Fusion processes are presented in detail in *Rapid Method for Fusion of Soil and Soil-Related Matrices Prior to*

with the tracer or carrier in the homogeneous melt formed. Soil dissolutions using hydrofluoric acid (HF) may not effectively solubilize certain refractory materials, but they can be very effective in completely dissolving a variety of matrices including poorly soluble silica-based materials.

5b Volatile radionuclides are prepared for chemical separations using validated techniques for dissolution of the sample. The sample aliquant is dissolved to create one or several stock solutions as appropriate for the analyses to be performed.

To prevent volatilization of analyte during dissolution, samples should be dissolved prior to chemical separations using techniques validated for the volatile radionuclides of concern. Low temperature digestion, basic leaching, or basic fusion techniques, combined with appropriate oxidation/reduction control may be used to maintain radionuclides in a non-volatile state throughout the process.

6 The sample fraction isolated for gamma spectrometric analysis in Step 4b is counted on a HPGe spectrometer. Aliquant size should be planned to optimize count times and throughput while ensuring that method uncertainty requirements are met.

The HPGe analysis is routinely performed on the “volatile” or “as-received” fraction of the sample. Analysis of this fraction will produce results that are valid for all gamma emitters, volatile or non-volatile. Results performed on the “as-received” sample are converted to a “dry-weight” basis by applying percent moisture results determined in Step 4a.

The volumetric configuration and density of the sample should be as close as possible to the calibration standard. Every soil sample will have some naturally-occurring radionuclides present. A sample with similar soil composition from a representative non-impacted area may help in determining the radionuclide concentration levels of background radionuclides in uncontaminated soil.

If sample size is limited, the prepared sample from this step may need to be shared with the fusion, digestion, leaching processes in Steps 5a or 5b.

7 Once the sample is homogeneous, it is possible to withdraw a representative aliquant of the fusion, digestion, or leaching processes in Steps 5a and 5b for analysis for gross alpha and gross beta activity. After the source radionuclide(s) has been identified, it may be possible to eliminate one of the screens depending on the volatility of radionuclides that may be present in the sample. If it is known that only gamma-emitting radionuclides are present in the sample, and if all MQOs can be met, screening measurements in Step 7 and radionuclide specific measurements in Steps 7a and 7b may be skipped in lieu of the gamma measurement in Step 6.

Americium, Plutonium, and Uranium Analyses; Rapid Method for Radium-226 Analyses in Soil Incorporating the Fusion of Soil and Soil-Related Matrices; and Rapid Method for Sodium Carbonate Fusion of Soil and Soil Related Matrices Prior to Strontium-90 Analysis (EPA 2012a).

Assuming that the MQO for required method uncertainty for alpha screening of 130 pCi/g at the AAL of 410 pCi/g is met (corresponding to the first year 2,000 mrem PAG for ^{226}Ra in Table 6A)[20], and the alpha concentration is greater than the ADL of 210 pCi/g, the sample will be analyzed for α-emitting radionuclides using radionuclide-specific methods. Assuming that the MQO for required method uncertainty for gross beta screening of 3,200 pCi/g at the AAL of 330 pCi/g (first year 2,000 mrem PAG for ^{60}Co in Table 6B) is met, and the beta concentration is greater than 3,200 pCi/g, the sample will be analyzed for radionuclide-specific β-emitting radionuclides.

If a screening technique is used that provides an aggregate value for the radioactivity (i.e., it does not differentiate between alpha and beta activity but reflects their summed total) greater than 210 pCi/g, the sample should be analyzed for radionuclide-specific alpha-only and beta-only emitters. If MQOs are met, but the gross activity is less that the above values, archive the remainder of the fused sample for analysis at a later time and report results to the IC per step 11.

The laboratory should validate screening techniques for concentrations at the AAL for the 1,000-mrem and 2,000-mrem PAG values.

7a
7b

If the gross alpha or beta concentration exceeds the ADL for either of these groups of emitters, chemical separations should be started immediately using validated procedures for each analysis to be performed. If the project manager does not specify the sequence of analyses, laboratory personnel should use their best professional judgment, based on the characteristics of the samples, to determine the order of processing the samples so that the results are obtained in the timeliest manner.

8

All analytical results should be collected for each sample (both the screening values and the final radiochemical-specific analyses). The results should be reviewed and reported by knowledgeable personnel per the laboratory's QA program.

As reviews are completed, and finalized radionuclide-specific results become available, each individual result can be compared to project-specific MQOs and ADL values (see Tables 6C or 6D for default values). For example, from Table 6C, the required method uncertainty for ^{241}Am is 140 pCi/g at the AAL of 1,100 pCi/g. The ADL for the 2,000-mrem PAG is 780 pCi/g. If the MQO for required method uncertainty is met, and the activity concentration exceeds its ADL, the 2,000-mrem AAL has been exceeded. The IC should be promptly notified (broken line to Step 11) while the remaining analytical work is completed.

9.

Compare the sum of all final analytical results that are above their respective critical level concentrations with the sum of the respective gross radioactivity measurements. This is done to verify that no major radionuclide contributor to dose has been missed. Isotopic and gross screening results should agree with each other within a factor of about two.

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[20] This is the limiting default value for all the listed radionuclides. If a shorter list of possible radionuclides is known, use the limiting value from that list.

Discrepancies in agreement may indicate that a major radionuclide may have been omitted from the analysis sequence in Steps 6, 7a, or 7b, or they may be attributable to problems with the analytical process. They may also indicate that the screening process needs to be adjusted based on the specific radionuclides identified in the samples.

Comparability may not be realized for volatile radionuclides, when radionuclides with low-energy emissions (beta-gamma, conversion electrons, x-rays, etc.) are present. Difference may also result when radionuclides with short-lived decay progeny are in the mix of radionuclides in the sample. For example, if ^{103}Ru is detected by gamma-ray spectrometry, its progeny ^{103}Rh may not be specifically identified by the software, even though it is present in secular equilibrium within hours. Significant unreconciled differences between the screening and the summed radionuclide-specific results should be noted in the report to the IC.

10 The sum of fractions (SOF) for all radionuclides present is calculated at this point. This is done by dividing each individual radionuclide concentration that exceeds its critical level by its respective AAL value (see Table 6C or 6D for default values). The calculation of the sum of fractions uses the following equation:

$$Sum\ of\ Fractions = \sum_{i=1}^{n} \left[\frac{R_i}{AAL_i} \right]$$

Where R_i is the activity concentration of the individual radionuclide and AAL_i is the analytical action level of that radionuclide for the 2,000-mrem PAG concentration at one year of exposure. If the sum of fractions is greater than 1.0, the 2,000-mrem PAG AAL concentration for the first year exposure may have been exceeded. The case narrative is updated accordingly.

11 Contact the IC and report all final results, and whether or not individual analysis, or the sum-of-fractions results, identifies that a PAG AAL has been exceeded. All sample residuals or residual solutions from dissolution should be archived in the event that additional analyses are required.

12 Samples that do not exceed the gross screening values in Step 2b are given lower priority. Analysis of these samples may be delayed at least until analyses for red and green path samples have been started unless there is concern about decay of short half-lived radionuclides in the sample. Samples that fall into this category based on initial screening results may be communicated to the IC so that decisions regarding the first-year exposure pathway PAGs level of activity concentration from these sample locations can be made in a timely fashion. Although there may be some question regarding the representativeness of these results, this feedback may help shape priorities assigned to each sample and thus enhance decisionmaking.

Additional Points for Scenario 1:

Volatiles

Volatile radionuclides that are most likely to be encountered are ^{131}I, ^{129}I, ^{125}I, ^{3}H (tritium), and ^{99}Tc. The normal preservation techniques of a sealed container and low temperature will minimize the loss of these radionuclides in a soil sample during transport and sample handling (both in the field and in the laboratory). If it is determined that radioisotopes of these elements are present, sample acquisition techniques, handling, storage, and shipping may need to be modified to further minimize potential losses. Such techniques may include:

- Sample techniques that minimize sample aeration when transferring from the ground to the sample container.
- Use of small containers so that the sample completely fills the container.
- Use of an electronic or dry ice cooler to immediately reduce sample temperature.

For measurement of tritium in soil samples (where ^{3}H is incorporated solely as part of a water molecule), relatively simple techniques such as freeze drying or distillation following equilibration with a minimal amount of water carrier provide good separation from the remainder of the matrix. More involved chemical methods are necessary to ensure adequate separation and capture of the radioiodines or technetium.

Screening of Soil Samples

Screening samples for gross γ activity concentration is much easier than screening for gross α or gross beta activity concentration. Analysts should recognize that the degree of self-absorption will limit the size and representativeness of sample that can be effectively analyzed for gross alpha or gross beta, and may introduce substantial bias and uncertainty into direct measurements of soil samples. Gamma rays, however, are not as significantly affected by the soil matrix, which means that larger more representative samples can be taken. This allows γ emitters to be more reliably detected resulting in lower bias and uncertainty, and a lower rate of false negative measurements due to non-homogeneity for γ measurements. It may be advisable to consider making standards for the gamma-screening equipment that are incident-specific in sample/ container size and in radionuclide(s) content.

Low-energy gamma emitters (with energy less than ~40 keV) and those radionuclides decaying via electron capture (followed by X-ray emission) will not be effectively identified by most screening tests due to sample self-absorption. This issue may only come to light when radionuclide-specific analyses have identified these low-energy emitters even when gross activity screens do not indicate any activity. The final comparison between the screening result and the sum of all radionuclide-specific activity concentration will show that the alpha/beta/gamma screen has underestimated the activity concentration present in the sample.

In counterpoint to low-energy gamma-emitting radionuclides not contributing to the gross activity concentration is the presence of varying degrees of NORM in soil samples. Uranium and thorium decay products emit gamma rays and will contribute to the overall gross gamma activity. Some short-lived radionuclides such as ^{212}Pb and ^{212}Bi may be present as unsupported progeny of ^{220}Rn (the thorium decay chain) and their activity will decrease rapidly between the time of sampling and the time of counting. Other NORM progeny have long enough half-lives to be present at the time of receipt of the laboratory and may be decaying during the screening process.

This means that gross radioactivity screening results for gamma and alpha or beta may change as a function of time. For example, unsupported ^{224}Ra progeny would decay with a half-life of approximately 10 hours (based on the half-life of ^{212}Pb).

Table 3 provides information about gamma rays that come from radionuclides not usually determined by gamma spectrometry because their gamma ray abundances are relatively low. The event that resulted in these radionuclides being deposited may have caused micron sized particles to form with high specific activity concentration.[21] These are referred to as discrete radioactive particles (DRP) or "hot" particles. Thus an aliquant that contains a DRP could allow these less-abundant radionuclide gamma rays identified in Table 3 to become a measureable feature in the gamma ray spectrum. As such it is advisable to review spectra for the presence of these gamma rays, or to add these radionuclides to the gamma spectrometry library. A longer count time for this step than for the initial scan is appropriate if there are alpha or beta emitters with low abundance gamma rays present. The staff can prepare sample aliquants for these analyses after the soil dissolution steps are completed.

Table 3 – Radionuclides with Low-Abundance Gamma Rays Not Usually Used for Analysis

Radionuclide	**^{89}Sr**	**^{90}Y**	**^{129}I**	**^{210}Po**	**^{226}Ra**	**^{228}Th**
Principal Decay	β^-	β^-	β^-	α	α	α
Gamma, keV	909.0	2186.2	39.6 (29.8 and 29.5 x-ray)[1]	803.1	186.2 [3] (262.3) [1]	84.4 (131.6)
Abundance, %	9.56×10^{-3}	$1.4\text{x}10^{-6}$	7.51 (36.7 and 19.9)[1]	1.03×10^{-3}	3.59 ($5.0\text{x}10^{-3}$) [1]	1.22 (0.130)
Radionuclide	**^{232}Th**	**^{235}U**	**^{237}Np**	**^{238}U**	**^{238}Pu**	**^{239}Pu**
Principal Decay	α	A	α	α	α	α
Gamma, keV	911.2[2] (338.3) [2], from ^{228}Ra/^{228}Ac	143.8 (185.7 [3], 163.3)	86.5 (311.9[2] from ^{233}Pa)	1001.0[2] from ^{234m}Pa (63.3[2] from ^{234}Th)	152.7 (43.5)	51.6 (129.3, 375.1, 413.7)
Abundance, %	25.8 (11.3)	11.0 (57.2, 5.08)	12.4 (38.5)	0.842 (3.7)	9.29×10^{-4} (3.92×10^{-2})	2.72×10^{-2} (6.31×10^{-3}, 1.55×10^{-3}, 1.47×10^{-3})
Radionuclide	**^{240}Pu**	**^{241}Am**	**^{241}Pu**	**^{242}Pu**	**^{243}Cm**	
Principal Decay	α	A	β^-	A	α	
Gamma, keV	45.2 (104.2, 160.3)	59.5	148.6 (103.7)	44.9 (103.5, 158.8)	277.6 (228.2, 209.8)	
Abundance, %	4.47×10^{-2} (7.14×10^{-3}, 4.02×10^{-4})	35.9	1.86×10^{-4} (1.01×10^{-4})	3.73×10^{-2} (2.55×10^{-3}, 3.00×10^{-4})	14.0 (10.6, 3.29)	

Notes:

[1] Values in parentheses represent the next most abundant gamma ray.

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[21] Any radionuclide can form a DRP. However the significance of an alpha or beta emitter with a low abundance gamma ray emission forming a DRP is that the radionuclide will be concentrated rather than spread out creating the possibility of identifying these low abundance gamma rays in the spectrum.

[2] Based on activity of the progeny. Can only be used for quantification when the parent and progeny are undisturbed for 5-10 progeny half-lives and they are known to be in secular equilibrium. For ^{237}Np (by ^{233}U) and ^{238}U (by ^{234}Th/^{234m}Pa) this requires about 4–9 months. For ^{232}Th (by ^{228}Ra) it requires 30–60 years and generally applies only to undisturbed geological materials. Although these photopeaks cannot be used for quantification, their presence in the spectrum provides valuable evidence and may confirm the presence of the radionuclide in a sample.

[3] Note that the 186.2 keV photopeak from ^{226}Ra cannot be resolved from the 185.7 keV photopeak of ^{235}U. Unless the interference can be resolved, or is determined to be trivial, these photopeaks should not be used for quantification. Their presence in the spectrum, however, provides valuable evidence and may confirm the presence of the radionuclide in a sample. Also, in samples with high ^{137}Cs activity, the 661 keV backscatter peak may be detected at 186 keV and could be mistaken for either ^{226}Ra or ^{235}U.

Additionally, certain α- and β-emitting radionuclides have very low-abundance γ rays. These γ rays are not normally used for analysis of those radionuclides, and may not necessarily be identified in gamma spectrometry software. Thus, if sufficient amounts of the radionuclides noted in Table 3 are present in a soil sample, the alpha or beta measurement may not be commensurate with the gross screen for gamma radiation.

However, as the activity concentration of these radionuclides decreases, it is a combination of γ ray abundance and half-life that makes the gamma ray of little utility at these lower-activity concentrations. It is recommended that a separate library for incident response samples be created that has these low-abundance γ rays for radionuclide specific analyses. Table 3 provides some examples.

These γ rays can be used for qualitative identification of these radionuclides. Their presence in the γ-ray spectrum should direct the analyst to perform chemical separations followed by alpha- or beta-specific detection.

Aluminum absorbers can be used to qualitatively identify the presence of radionuclides based on penetrating ability. Thus, if an aluminum absorber of 6.5 mg/cm^2 is used, and the measured activity concentration is reduced to background, one could qualitatively state that the beta particle energy of the radionuclide is < 0.067 MeV. Conversely, if the absorber has little effect on the count rate, it can be stated that the beta particle energy is >0.067 MeV. Table 4 identifies some beta-only emitters with their energies and range in aluminum absorbers.

Table 4 – Beta-Only Emitters

Radionuclide	**^{241}Pu**	**^{63}Ni**	**^{129}I**	**^{35}S**	**^{99}Tc**	**^{32}P**	**^{90}Sr/^{90}Y**
Maximum Beta Energy, MeV	0.021	0.067	0.150	0.167	0.294	1.711	(0.546)/2.28 [1]
Range [2], mg/cm^2 for $E_{\beta max}$	0.8	6.5	27	32	75	800	1,100

Notes:

[1] It may be assumed that ^{90}Sr/^{90}Y will be in secular equilibrium by the time any analysis is started. Thus, the 2.28 MeV beta particle of ^{90}Y will be present.

[2] U.S. Department of Health, Education and Welfare (HEW). 1970. *Radiological Health Handbook*, p.123.

V. RADIOANALYTICAL SCENARIO 2 (IDENTIFYING UNCONTAMINATED SOIL DISTANT FROM THE RDD SITE OR FOLLOWING INITIAL REMEDIATION)

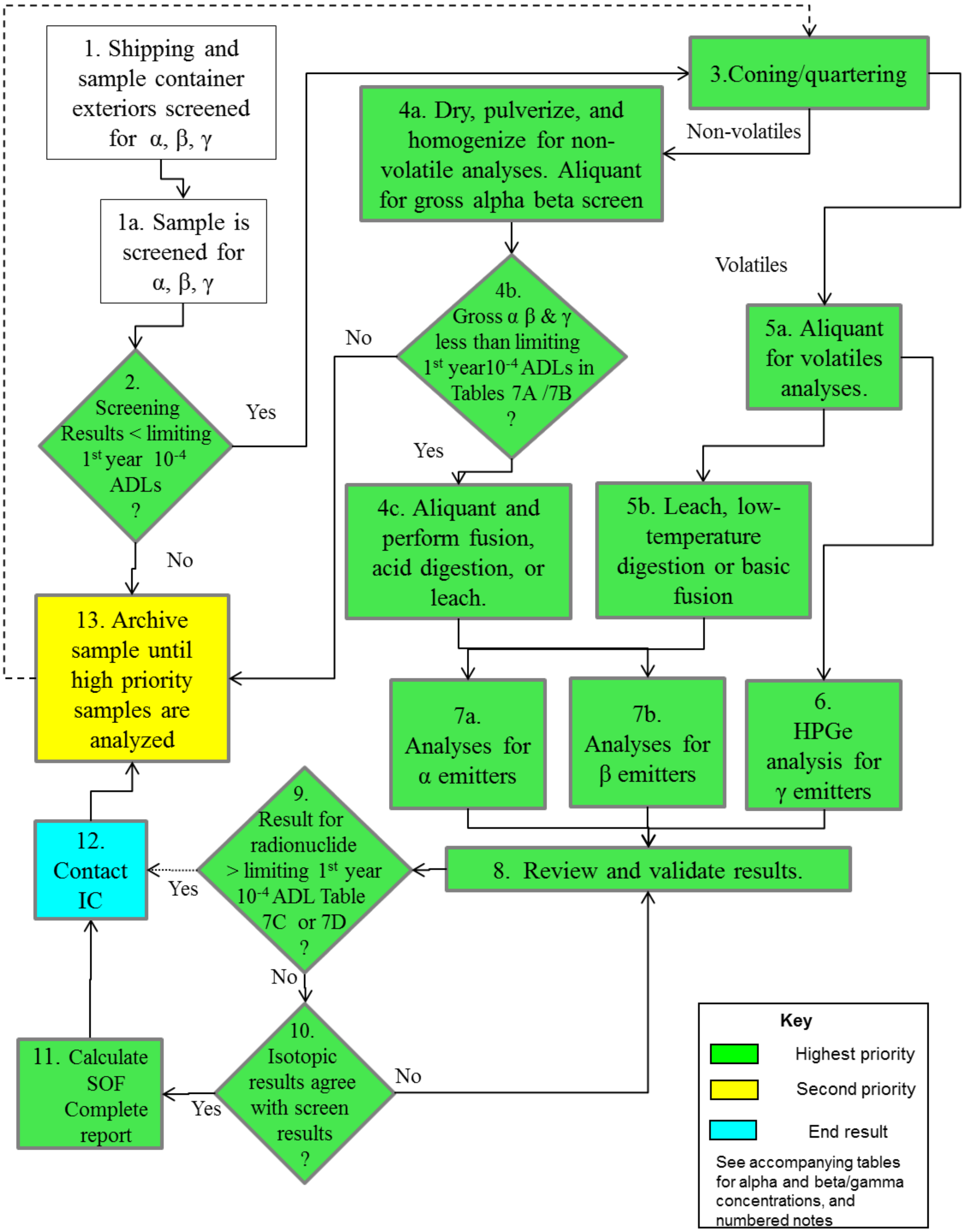

Figure 4 – Radioanalytical Scenario 2 (Identifying Samples with Activities less than 10^{-4} Risk – First Year)

Notes for Scenario 2: Several weeks to months following the initial event. Radionuclide contaminants are known.

Purpose: Identify areas where contamination has *not* occurred, or areas that have been successfully remediated.

The samples may arrive over several days. It is likely that detailed screening of samples at this point will not provide any significant insight into the radionuclide content of the samples because the AALs identified in Tables 7A and 7B for first year exposure at 10^{-4} risk will yield screening values close to background for normal soils (these may well be above the ADL in these tables). If volatile radionuclides such as ^{125}I, ^{129}I, ^{131}I, ^{99}Tc, or ^{3}H are expected to be part of the sample matrix, an alternate, parallel sample processing path specifically for these volatile radionuclides is performed. Only after an analytical step or procedure has been completed for the highest priority samples should lower-priority samples be addressed. Lower-priority samples (yellow on this chart) may need to be stored for several days until the highest priority samples have been analyzed. The samples with the highest priority in this instance will be the ones with the lowest activity concentration.

The radionuclide activities corresponding to PAG values for this scenario assumes a first year exposure and can be found in Tables 7A and 7B for screening and 7C and 7D for radionuclide-specific activities.

1. The exterior surfaces of the containers are swiped to determine if any contamination is present. The actions taken and the limits for those actions should be established by the individual laboratory according to their radiation safety manual.

These might include a Geiger-Mueller counter with appropriately calibrated beta and gamma detector probes or a micro-roentgen meter (gamma only).[22] This step should be performed with the sample container unopened. Surface swipes of the container are taken to assess the presence of external gross α/β and γ contamination. These surveys are for contamination control purposes and not for the assessment of sample processing flow path.

Any container with measurable external contamination should be decontaminated before proceeding with subsequent analysis. This will help to minimize sample cross-contamination as well as laboratory contamination.

1a. *Gamma Screening*

An initial screen is performed by directly counting the unopened sample container for a short count time on a HPGe detector.[23] MQOs required method uncertainty and ADLs for the 10^{-4} risk level are used (see Table 7B for screening measurements of beta emitters).

Some laboratories may use a NaI(Tl) detector to determine whether the gross γ activity indicates that the activity of gamma emitters in a sample is less than an AAL

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[22] Some manufacturers have developed kits that include the survey meter plus an alpha-beta-gamma pancake GM detector and a NaI gamma detector.

[23] The potential configurations for these measurements are outlined in *Radiological Laboratory Sample Screening Analysis Guide for Incidents of National Significance* (EPA 2009b).

concentration. The survey instrument should be calibrated for the geometry used and for the gamma-emitting radionuclide of concern having the limiting AAL listed in Table 7B.

In order for gamma measurements to be used to quickly assess whether sample activity will be less than first year 10^{-4} risk AALs, it must be known that only gamma-emitting radionuclides are present in the sample.

It may not be possible to determine some gamma-emitting radionuclides with low enough uncertainty at the AAL concentration to meet MQOs when the sensitivity of the instrument used is inadequate, or for very low-abundance or low-energy gamma emitters. Radionuclides that do not emit γ- or X-rays greater than 40 keV, including pure alpha and beta emitters such as ^{210}Po or ^{90}Sr, will not be detected.

Gross Alpha and Beta

NOTE: The techniques described here present a rapid, positive approach to screening of soils. They involve digestion and aliquanting methods commonly used in the radiochemistry laboratory and require open handling and processing of samples that may contain elevated levels of radioactivity. Laboratory staff need to take specific precautions while performing these techniques to avoid personal contamination (including inhalation), laboratory surface contamination, and sample cross-contamination especially since the radioactivity level of pure alpha or beta emitters in the sample is initially unknown. Additional precautions should be taken to dedicate a segregated area for screening samples.

The samples received from a radiological incident may come in a variety of containers: plastic bags, plastic bottles, glass bottles, Petri-style containers, aluminum cans, etc. The masses of the samples received also will vary significantly. If possible, using several different style containers with different masses of standards for use in calibration of screening equipment will be helpful in minimizing the time for the initial sample screening process.

Screening of the sample for α and β activity must be done carefully as the gamma screen may not yield any indications of elevated activity concentration when only pure alpha- or beta-emitting radionuclides, or radionuclides that only emit γ- or x-rays below the calibrated range of the instrument. A very rudimentary screening process, performed in a hood, should be used prior to removing the entire sample from its container for homogenization and sub-sampling. Some options for making such a measurement may be:

- Take a swipe of the inside of the container cap. Then, using an alpha-beta probe, determine the count rate of the swipe.
- Insert a tongue depressor (with double-sided tape on one surface) into the container and remove for direct measurement.
- Take a small aliquant of the top surface (after the sample container is shaken) and spread it onto a planchet.

- Take a measured aliquant and place it into a scintillation vial with cocktail for liquid scintillation analysis.
- Perform a rapid nitric-hydrofluoric acid digest (total dissolution is not necessary) on a small aliquant of the sample and evaporate onto a planchet.

For the first three bullets above, an initial estimate of the mass transferred using the specific technique should be made with a soil of similar composition. The probe used for such techniques should have been calibrated with soil samples spiked to a level simulating the AAL values.

The last two options above, LSC and GPC provide more quantitative measures of gross alpha/beta activity. LSC can provide a faster screening process and avoid some of the shortcomings of a sample leaching process needed for GPC because the effects of self-absorption in the sample test source are minimized using LSC. The procedure used must correct quench from various sources in the soil, and the method must be validated prior to use on samples from an incident. It may be impractical to separate alpha from beta signals using pulse-shape analysis when high levels of quench are present. Quench will shorten the duration of alpha pulses making them appear more like a beta pulses. Instead, a total activity measurement (combined alpha and beta) across the entire energy spectrum may provide the most reliable estimate of activity in highly quenched sample.

For soil samples, the question arises of how to introduce the solid sample into the liquid scintillation matrix. Although there are no standard methods that specifically address this issue, several general approaches are commonly used. The first, perhaps more conventional approach, involves direct measurement of the solid. The second involves leaching or digesting a representative aliquant of the solid sample and processing the leachate / digestate as if it were an aqueous sample. The third approach involves directly suspending a small, representative aliquant of the soil in a gelling liquid scintillation cocktail.[24]

The limiting factor in using gross alpha beta measurements for reliable decisionmaking is obtaining a representative aliquant. In contrast to the gamma measurement where the entire sample is measured, the relatively small portion of unhomogenized sample may not be representative of the entire sample. The small aliquant may indeed completely fail to identify non-homogenously distributed activity present in a sample. The alpha / beta screen may still provide important information with regard to handling and indicate a need for personal protective measures since the next steps involve working with the entire sample in unencapsulated form.

While gamma measurements may meet MQOs prior to complete homogenization of the sample, high levels of uncertainty associated with the alpha beta screening techniques discussed above, however, may preclude use of those results for making reliable decisions until a representative aliquant can be obtained following homogenization of the

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[24] Cocktails such as Insta-Gel, or Quicksafe A may be used since they will suspend the solid and allow it to be counted in a 4-π geometry.

sample. It is important to ensure that estimates of the measurement uncertainty are robust, and that MQOs for uncertainty have been met before using data to decide that a sample concentration exceeds an AAL. If estimates of uncertainty are deemed to be unreliable, the results should not be used for critical decisionmaking. Thus it may be necessary to delay decisions about whether AALs for non-gamma-emitting radionuclides have been exceeded until the sample is milled and homogenized and a representative aliquant can be obtained.

2. Any of the above techniques will yield a good estimate of the potential α and β activity concentration of the sample. If pure alpha or beta emitters may be present, or if the gross gamma result is less than the limiting ADL at the 10^{-4} risk level , or if the MQOs for required method uncertainty cannot be met, proceed with coning, quartering and homogenization to obtain a representative aliquant of the sample.

If only gamma emitters are present, and the gamma screening performed in 1b satisfies the MQOs for required method uncertainty, and the gross γ activity is less than the limiting ADL listed in Table 7B, the IC may be notified that no AALs are exceeded and the sample may be archived.

3. A process described as "coning and quartering" is used to break the entire bulk sample down into representative subsamples. Unless the project has a specific MQO stating the contrary, remove extraneous detritus (e.g., sticks, twigs, rocks larger than 1/4", etc.). The removed material should be retained for potential future analysis and an appropriate comment added to the case narrative.

Fractions are isolated for volatile radionuclides and gamma spectral analysis, and for analysis of non-volatile radionuclides, where required. The volatile fraction should be processed quickly and immediately sealed in containers to minimize potential losses of volatile radionuclides.

This process is described in the Appendix to *Rapid Method for Fusion of Soil and Soil-Related Matrices Prior to Americium, Plutonium, and Uranium Analyses*; *Rapid Method for Radium-226 Analyses in Soil Incorporating the Fusion of Soil and Soil-Related Matrices*; and *Rapid Method for Sodium Carbonate Fusion of Soil and Soil Related Matrices Prior to Strontium-90 Analysis* (EPA 2012a).

Precautions should still be taken (e.g., opening the sample container and handling in a hood) since the alpha/beta activity concentration has only been assessed by bulk screening techniques which may have very high uncertainties.

Steps 4 through 7 may be performed concurrently.

4a. After the coning and quartering, place the sample in a tared can (without lid) and dry the soil to constant weight in an oven at 110 ±10 °C. Drying samples provide for consistent comparison of results regardless of moisture content.

Note: Constant weight may be determined by removing the container from the oven and weighing repeatedly until the mass remains constant to with within 1%. This also may be achieved operationally by observing the time needed to ensure that 99% of all samples will obtain constant weight.

This is the dry-weight basis for the analysis. This mass is also used to calculate the fractional solids content for each sample (i.e., percent solids) so that results from the volatile fraction, analyzed "as received" can be converted to and reported on a dry-weight basis.

Once the sample is dried, homogenization continues using the same vessel (the paint can). Stainless-steel or ceramic balls or rods are added to the can and the can is shaken for about 5 minutes, or as needed to produce a finely-milled, well-homogenized, sample. The precise particle size of the milled sample is not critical as long as the milled sample is fine enough to facilitate a rapid and complete dissolution of the soil matrix.

NOTE: A qualitative, visual standard can be prepared by passing quartz sand or other milled material through a 50-mesh and then a 100-mesh screen. The portion of the sample retained in the 100 mesh screen can be used as a qualitative visual standard to determine if samples have been adequately pulverized. The process is complete once 95% of the sample (or greater) is as fine, or finer, than the qualitative standard. If, by visual estimation, more than ~5% of total volume of the particles in the sample appear to be larger than the particle size in the standard, return the sample to the shaker and continue milling until the process is complete.

If visual inspection of the milled sample shows elevated presence of larger particles that may not be effectively dissolved during the fusion or digestion process, the sample should either be milled for a longer period of time, or those particles preferentially removed prior to aliquanting. In most cases, preferential removal of a small fraction of larger particles will still provide representative results since the surface area of larger particles is relatively low, and that surface will be abraded during milling, and the activity associated with sample fines should be representative of that found in the original sample. A comment should be added to the sample narrative addressing removal of the solids.

NOTE: In this scenario, the radionuclides that are potential contaminants are known. This may mean that if a single type of emitter is present that the ADL for that specific radionuclide should be used. However, when mixed decay modes are present, it may be necessary to compare alpha, beta, or gamma emitting radionuclide screening results based on gross gamma or combined gross ($\alpha + \beta$).

Prior to aliquanting samples in Step 4c, the sample is screened for gross alpha and beta. A stainless steel planchet to which an adhesive backed cloth swipe has been applied is tared, and a small aliquant of dried, pulverized solid (e.g., <0.1 gram) is transferred to the upward-facing textured surface of the swipe. Using a hand-held survey meter, the planchet is screened to detect the presence of elevated alpha and beta activity that would be a concern to handling outside the hood and could result in contamination of the low-background proportional counter. The planchet, swipe, and solid are reweighed to determine the net amount of sample on the planchet. The residues are fixed to the planchet by applying a very light layer of hair spray. The planchet is dried for a minute under a heat lamp and reweighed to determine the final mass to use for self-absorption corrections for the gross alpha and beta measurement. The sample test sources are counted on a gas flow proportional counter to determine the gross alpha and beta activity.

Prior to, or concurrent with, Step 5a, soil samples with significant organic content should be ashed to completely combust organic matter. This can be accomplished using a programmable furnace where a temperature of approximately 600 °C can be achieved. Ramping up the temperature in time intervals is necessary to avoid uncontrolled ignition and excessive smoke generation during the process. If the sample is ashed prior to aliquanting, it is important to record the wet, dry, and combusted masses so that the wet/dry ratio (i.e., percent solids) (and the percent organic content if required) can be calculated and used to convert final results to a dry-weight basis

4b. The results of the alpha/beta screen are reviewed to determine whether any limiting ADLs have been exceeded. If the MQO for required method uncertainty for alpha screening is met, and the gross alpha concentration is less than the lowest applicable 10^{-4} ADL in Table 7A, the sample will be analyzed for α-emitting radionuclides using radionuclide-specific methods.

If the MQO for the required method uncertainty for gross beta screening is met, and the beta concentration is less than the lowest applicable 10^{-4} ADL in Table 7B, the sample will be analyzed for radionuclide-specific β-emitting radionuclides. If more than one radionuclide is of concern, or if a screening technique is used that provides an aggregate value for the radioactivity (i.e., it does not differentiate between alpha and beta but reflects their summed total), and the activity is less than the lowest applicable 10^{-4} ADL in Tables 7A or 7B, the sample will be analyzed for all applicable radionuclide-specific alpha-only and beta-only emitters.

The screen also will provide the most reliable information yet on levels of activity that are present. This can be used to determined appropriate levels of contamination control and personal protective gear needed. The results may also be used to determine whether tracer levels should be adjusted when aliquant size cannot be reduced due to concerns about the representativeness of the sample (e.g., 0.5-1 gram).

4c. Individual non-volatile radionuclides (with the exception of gamma emitters) are prepared for analysis using validated techniques for dissolution and chemical separation. Separate aliquants may be removed for radionuclide-specific analysis for non-volatile components. Aliquant size should be coordinated with planned count times to ensure that the required method uncertainty will be met, and to optimize throughput. The dried samples are dissolved so they may be aliquanted for analysis of remaining potential radionuclides appropriate to the radionuclide analyses to be performed.

For samples resulting from an RDD, it is important to ensure complete dissolution of potentially refractory materials. Certain materials such as those used in a radioisotope thermoelectric generator (RTG), brachytherapy sources, or radionuclides that have been exposed to a high temperature detonation may be difficult to dissolve using conventional acid digestion techniques.

Sodium carbonate or sodium hydroxide fusions are two good methods for complete dissolution of soil samples. It is important to achieve complete dissolution as refractory materials could result from an RDD, which would not be dissolved by simple acid dissolution. Fusions help ensure isotopic exchange of analyte with the tracer or carrier in the homogeneous melt formed. Soil dissolutions using hydrofluoric acid (HF) may not effectively solubilize certain refractory materials, but they can be very effective in completely dissolving a variety of matrices including poorly soluble silica-based materials.

5a. Representative portions of the “as received” (i.e., undried) sample are aliquanted as needed for HPGe gamma analysis, and where required, for analysis of volatile radionuclides. The sample should be mixed, homogenized, and subsampled rapidly and in a manner that minimizes loss of volatiles while ensuring that aliquants will be representative of the original sample.

Analyses for volatile radionuclides should be started as soon as possible to minimize loss of analyte. If sample analysis cannot be started immediately, a project-specific means of sample storage for volatiles should be in place so that even short-term storage does not significantly reduce their concentration. Volatile radionuclides are prepared for chemical separations using validated dissolution techniques that will prevent loss of analyte due to volatilization. These processes should be performed in a manner that also minimizes loss of volatiles. Low temperature digestion, basic leach, or basic fusion techniques, combined with appropriate oxidation/reduction control are used to create one or several stock solutions while ensuring that radionuclides are in non-volatile form.

Note that all analysis results from the volatile fraction should be calculated and reported on a dry-weight basis. For example, the volatile fraction can be analyzed “as received” and the results converted to dry-weight basis by applying the percent solids value determined in Step 4a.

6. Gamma spectrometric analysis is performed on the undried fraction isolated in Step 4b using a HPGe gamma detector. Aliquant size and count times should be optimized to ensure that the required method uncertainty is met, and to optimize throughput. Since an “as received” sample is counted, results are calculated and reported by applying percent solids values determined in Step 4a.

The volumetric configuration and density of the sample should be as close as possible to the calibration standard. Every soil sample will have some naturally-occurring radionuclides present. A sample with similar soil composition from a representative non-impacted area may help in determining the radionuclide concentration levels of background radionuclides in uncontaminated soil.

The sample needs to be counted long enough to achieve the respective MQO for the required method uncertainty indicated in Table 7D. For example, if ^{60}Co were the radionuclide of concern, a required method uncertainty of 2.5 pCi/g would be required at the AAL of 20 pCi/g. The final result would be compared to the predicted ADL for ^{60}Co of 14 pCi/g.

7a.
7b.
If gross alpha or beta concentrations are less than their respective ADLs, chemical separations should be started immediately using validated procedures for each alpha or beta emitter to be determined. Sample test sources should be counted such that the measurement quality objectives for the event will be met. If the project manager does not specify the sequence of analyses, laboratory personnel should use their best professional judgment, based on the characteristics of the samples, to determine the order of processing the samples so that the results are obtained in the timeliest manner.

8. Analytical results should be collected for each sample (both the screening values on the raw sample and the final radiochemical-specific analyses). The results should be reviewed and reported by knowledgeable personnel (per the laboratory's QA program).

9. As reviews are completed, and finalized radionuclide-specific results become available, each individual result is compared to project-specific MQOs and ADL values (see Tables 7C or 7D for default values). For example, for the 10^{-4} risk PAG in Table 7C, the required method uncertainty for ^{241}Am is 8.2 pCi/g at the AAL of 65 pCi/g and the ADL of 46 pCi/g. If the MQO for required method uncertainty is met, and the activity concentration exceeds the ADL, the 10^{-4} risk AAL has been exceeded. The IC should be promptly notified (broken line to Step 11). If any AAL is shown to be exceeded, proceed with archiving sample residuals, digests and sample test sources in Step 13 and transfer available resources to the continued analysis of potentially uncontaminated samples.

10. Compare the sum of all final analytical results that are above their respective critical level concentrations with the sum of the respective gross radioactivity measurements. This is done to verify that no major radionuclide contributor to dose has been missed. Isotopic and gross screening results should agree with each other within a factor of about two.

Discrepancies in agreement may indicate that a major radionuclide may have been omitted from the analysis sequence in Steps 6, 7a, or 7b, or they may be attributable to problems with the analytical process. They may also indicate that the screening process needs to be adjusted based on the specific radionuclides identified in the samples.

Comparability may not be realized for volatile radionuclides, when radionuclides with low-energy emissions (beta-gamma, conversion electrons, x-rays, etc.) are present. Difference may also result when radionuclides with short-lived decay progeny are in the mix of radionuclides in the sample. For example, if ^{103}Ru is detected by gamma-ray spectrometry, its progeny ^{103}Rh may not be specifically identified by the software, even though it is present in secular equilibrium within hours. Significant unreconciled differences between the screening and the summed radionuclide-specific results should be noted in the report to the IC.

If the screening process indicated $<10^{-4}$ risk whereas radioanalytical results show $>10^{-4}$ risk, consider whether the screening process may not be sufficiently robust at the 10^{-4} and 10^{-6} risk levels to identify low activity concentrations for the radionuclides present.

11. Calculate the sum of fractions at this point. The calculation of the sum of fractions follows the following equation:

$$Sum\ of\ Fractions = \sum_{i=1}^{n} \left[\frac{R_i}{AAL_i}\right]$$

Where R_i is the activity of the individual radionuclide and AAL_i is the analytical action level of that radionuclide for the 10^{-4} risk level at one year of exposure.

If the sum of fractions is > 1.0 then the AAL for 10^{-4} risk level for the first year exposure may have been exceeded.

> The dashed line connecting Box 9 with Box 12 indicates that that checking SOF may not always be needed.

12. Contact the IC with the results of the analysis, in this case indicating whether the results confirm that the radionuclides present pose $<10^{-4}$ risk based on the radionuclide activity concentrations identified.

13. Sample residuals, unused dissolved sample, and sample test sources should be archived if potential analysis is needed at a future date.

VI. RADIOANALYTICAL SCENARIO 3 (IDENTIFYING SOIL CONTAMINATED WITH FISSION PRODUCTS FOLLOWING AN IND INCIDENT)

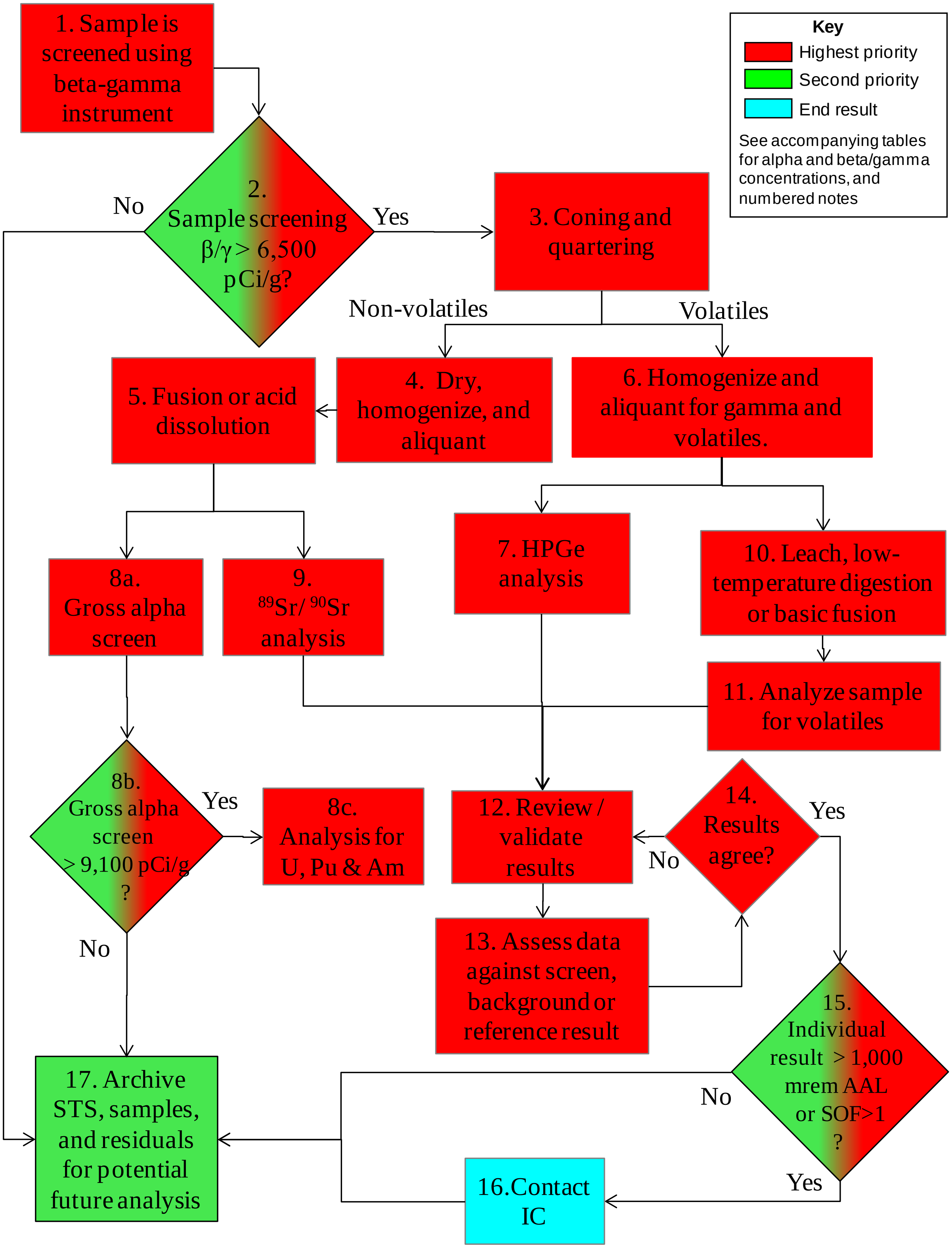

Figure 5 – Radioanalytical Scenario 3 (Samples Resulting from an IND)

Notes for Scenario 3: **An IND has been detonated. It is within the first week of the incident.**

Purpose: **Identify principal fission and activation products and their concentrations in samples with the highest activity.**

Samples will arrive and will require rapid screening to identify the most highly contaminated first. The majority of the fission products will be gamma emitters. The principal volatile species present will be tritium, iodine, and xenon isotopes. The iodines and xenons are all gamma emitters. The principal U and transuranic isotopes of concern will be ^{234}U, ^{235}U, ^{238}U and $^{239/240}$Pu from the original nuclear material and some neutron-capture progeny of each of these isotopes.

The radionuclide activities corresponding to early phase 1,000 mrem PAG values for this scenario can be found in Tables 6A and 6B for screening and 6C and 6D for radionuclide-specific activities.

1. An IND probably will have a lower explosive yield than a military nuclear weapon. This will mean that not only will there be fission products dispersed, but also dispersal of material that has either not undergone fission or fissionable material that has undergone activation instead of fission. Table 2 lists radionuclides resulting from a fission event (half-lives greater than ~ 1day). Table 9 lists several short-lived radionuclides that may be present in the first days following an IND. There is some overlap between the tables.

   These radionuclides, as well as other principal fission products, are beta/gamma emitters and will be able to provide a good response to screening on a beta/gamma survey instrument at the 1,000-mrem PAG concentration (Early Phase) and the 2,000-mrem PAG (first year) identified in Table 10a. This could be a Geiger-Mueller type device or a handheld Micro-R meter. The radionuclide that has the lowest PAG value is ^{60}Co. Although ^{60}Co is not a fission product, it may be formed as a result of neutron activation of stable ^{59}Co, a trace element in steel and other building materials. Initial calibration of screening instruments with this radionuclide would be acceptable until more specific information is known about the sample composition.

2. It is very likely that IND material that did not fission will have fission products associated with it. Since all fission products are beta or gamma emitters, the sample may be screened by measurement of its gamma emissions. The measurement should meet the Early Phase 1,000-mrem Early Phase PAG MQO for required method uncertainty for ^{60}Co (3,900 pCi/g at the AAL of 13,000 pCi/g) shown in Table 10A.

   If the gross gamma activity concentration exceeds the ADL for ^{60}Co of 6,500 pCi/g, proceed with additional radionuclide-specific analyses. If radionuclide-specific gamma analysis is performed using a HPGe spectrometer, MQOs are satisfied and any radionuclide exceeds the respective 1,000-mrem PAG radionuclide-specific ADLs identified in Table 10B, proceed with additional radionuclide-specific analyses. Otherwise archive the sample for potential future analysis per Step 17. Report analysis results to the IC and indicate that measurements show sample activity below the 1,000 mrem Early Phase PAG AAL values.

3. The entire sample should be coned and quartered to break the bulk sample down into representative subsamples. Unless the data quality objectives for the project state the contrary, remove detritus (e.g., sticks, twigs, rocks larger than 1/4", etc.) prior to coning and quartering. Save the removed material for potential future analysis and make an appropriate note of this in the case narrative.

Fractions are isolated for volatile radionuclides and gamma spectral analysis, and for analysis of non-volatile radionuclides. The volatile and gamma fraction should be processed quickly and immediately sealed in containers to minimize potential losses of volatile radionuclides.[25]

Steps 4 through 8 may be performed concurrently.

4. Transfer the non-volatile fraction isolated in Step 3 to either a 1-pint or 1-quart paint can (depending upon the sample size). Place the can (without lid) in an oven at 110±10 °C and dry the soil to constant weight. Drying samples provide for consistent comparison of results regardless of moisture content.

Note: Constant weight may be determined by removing the container from the oven and weighing repeatedly until the mass remains constant within 1%. This also may be achieved operationally by observing the time needed to ensure that 99% of all samples will obtain constant weight.

This is the dry-weight basis for the analysis. This mass is also used to calculate the fractional solids content for each sample (i.e., percent solids) so that results from the volatile and gamma fractions, which are analyzed “as received,” can be converted to and reported on a dry-weight basis.

Once the sample is dried, homogenization continues using the same vessel (the paint can) by adding stainless-steel or ceramic balls or rods to the can and shaking for about 5 minutes, or as needed to produce a finely-milled, well-homogenized, sample. The precise particle size of the milled sample is not critical.

NOTE: A qualitative, visual standard can be prepared by passing quartz sand or other milled material through a 50-mesh and then a 100-mesh screen. The portion of the sample retained in the 100 mesh screen can be used as a qualitative visual standard to determine if samples have been adequately pulverized. The process is complete once 95% of the sample (or greater) is as fine, or finer, than the qualitative standard. If, by visual estimation, more than ~5% of total volume of the particles in the sample appear to be larger than the particle size in the standard, return the sample to the shaker and continue milling until the process is complete.

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[25] This process is presented in detail in *Rapid Method for Fusion of Soil and Soil-Related Matrices Prior to Americium, Plutonium, and Uranium Analyses; Rapid Method for Radium-226 Analyses in Soil Incorporating the Fusion of Soil and Soil-Related Matrices; and Rapid Method for Sodium Carbonate Fusion of Soil and Soil Related Matrices Prior to Strontium-90 Analysis* (EPA 2012a).

If, by visual inspection, the sample appears to contain larger particles that may not be effectively dissolved during the fusion or digestion process, those particles may be preferentially removed prior to aliquanting. In most cases, removal of a small fraction of larger particles will still provide representative results since the surface area of larger particles is relatively low, and that surface will be abraded during milling. As a result, the activity associated with sample fines should be representative of that found in the original sample. A comment should be added to the sample narrative addressing removal of the solids.

A representative aliquant of the dried, homogenized sample is taken for gross alpha screening and subjected to the same dissolution process used for radionuclide-specific analyses. Separate aliquants are also removed for radionuclide-specific analysis of ^{89}Sr and ^{90}Sr, and for potential alpha spectrometric analysis of Pu and U isotopes. Note that no tracers/carriers should be added to the aliquant for gross alpha as these will interfere with the determination of gross alpha activity.

Prior to or during Step 5, soil samples with significant organic content should be ashed to completely combust organic matter. This can be accomplished using a programmable furnace where a temperature of approximately 600 °C can be achieved. Ramping up the temperature in time intervals is necessary to avoid uncontrolled ignition and excessive smoke generation during the process. If the sample is ashed prior to aliquanting, it is important to record the wet, dry, and combusted weights so that percent solids and the percent organic content (if required) can be calculated and used to convert final results to a dry-weight basis.

5. Aliquants of the dry sample from Step 4 are taken for total dissolution. For samples from an IND, it is important to achieve complete dissolution of refractory materials prior to chemical processing. Sodium carbonate or sodium hydroxide fusions offer two good methods for complete dissolution of soil samples. Fusions help ensure isotopic exchange of analyte with the tracer or carrier in the homogeneous melt formed. Soil dissolutions using hydrofluoric acid (HF) may not effectively solubilize certain refractory materials, but they can be very effective in completely dissolving a variety of matrices including poorly soluble silica-based materials.

A clear melt of the sample, or a clear solution from acid digestion should be achieved. If the dissolution process does not completely dissolve the sample, undissolved material potentially should be set aside for additional analysis and an appropriate note made in the case narrative.

6. The "as-received" sample fraction isolated for gamma and volatile radionuclides in Step 3 should be mixed, homogenized, and subsampled rapidly and in a manner that minimizes loss of volatiles while ensuring that aliquants will be representative of the original sample. In addition, if radionuclides are present in organic compounds, aliquants may be removed for separate analysis.

Representative portions of the "as received" fraction are aliquanted as needed for HPGe gamma analysis, and for radionuclide-specific analysis of volatile radionuclides such as ^{3}H, ^{129}I or ^{99}Tc. Aliquant size should be planned to optimize count times and throughput while ensuring that MQOs for required method uncertainty are met.

Analysis for short-lived species or volatile radionuclides, such as ^{131}I and noble gases, should begin as soon as possible to minimize decay and optimize count times, and to minimize losses due to volatility of the analytes. If analysis for volatiles cannot be started immediately, a project-specific means of sample storage for volatiles should be in place so that even short-term storage does not significantly reduce their concentration.

7. An aliquant of the sample from Step 6 is taken and counted using an HPGe detector. HPGe analysis is performed on the "as-received" fraction of sample isolated in Step 3. Analysis of the volatile fraction will produce results that are valid for all gamma emitters, volatile or non-volatile.

The volumetric configuration and density of the sample should be as close as possible to the calibration standard. Every soil sample will have some naturally-occurring radionuclides present. A sample with similar soil composition from a representative non-impacted area may help in determining the radionuclide concentration levels of background radionuclides in uncontaminated soil.

The sample needs to be counted long enough to measure the AAL concentration of 9,200 pCi/g of ^{60}Co with a required method uncertainty of 1,600 pCi/g. Note that ^{60}Co is the gamma emitting radionuclide with the limiting activity concentration for 1,000-mrem PAG in Table 10B.

8a. The product of the dissolution from Step 5 is dissolved in acid and the sample screened for gross alpha to assess if alpha-specific analyses should be done. The sample should be counted long enough to meet an MQO for required method uncertainty for ^{239}Pu of 410 pCi/g at the AAL of 1,300 pCi/g. (Note that ^{239}Pu is the alpha emitting radionuclide with the limiting activity concentration for 1,000-mrem PAG in Table 6A.)

If liquid scintillation screening is used, alpha / beta discrimination can be used to significantly reduce interference from beta emitters. The pulse-shape discriminator is pushed past the classic cross-over point used for simultaneous counting of alpha and beta to a point where beta-to-alpha crosstalk is nearly eliminated. Additional information may be available from the gamma spectrometry analysis performed in Step 7 as there are several gamma lines from either U or Pu fissile materials that will give an indication of the presence of these materials.

8b.
8c. Results of the gross alpha screen are compared to the ADL of 9,100 pCi/g (1,000-mrem PAG in Table 6A). If the alpha screening ADL is exceeded, analysis for uranium and plutonium isotopes is performed using a rapid method such as the *Rapid Radiochemical Method for Selected Radionuclides in Water for Environmental Restoration Following*

Homeland Security Events (EPA 2010). MQOs and ADLs for the 1,000-mrem PAG are stated in Table 6A.)

9. If gamma-specific analysis in Step 7 indicates the presence of fission products, then analysis for ^{89}Sr and ^{90}Sr should be started immediately as they are principal fission products and neither is a gamma ray emitter. The product of the dissolution of the dried sample from Step 5 is dissolved in acid and analyzed for radiostrontium. The sample may be initially analyzed for total radiostrontium. The analysis should be configured to meet the MQO for required method uncertainty at the 1,000 mrem level in Table 10B (2.7×10^4 pCi/g at an AAL of 2.1×10^5 pCi/g). If radiostrontium activity is detected, above the ADL of 150,000 pCi/g, ^{89}Sr and ^{90}Sr should be speciated and compared to their respective ADLs.

10. Volatile radionuclides are prepared for chemical separations from the aliquants in Step 6 using validated dissolution techniques that will prevent loss of analyte due to volatilization. These processes should be performed in a manner that also minimizes loss of volatiles. Low temperature digestion, basic leach, or basic fusion techniques, combined with appropriate oxidation/reduction control are used to create one or several stock solutions while ensuring that radionuclides are in non-volatile form.

    Note that all analysis results from the volatile fraction should be calculated and reported on a dry-weight basis. For example, the volatile fraction can be analyzed "as received" and the results converted to dry-weight basis by applying the percent solids value determined in Step 4a.

11. Chemical separations should be started immediately using validated procedures for each volatile alpha or beta emitter to be performed. Sample test sources should be counted such that the measurement quality objectives for the 1,000 mrem PAG in Tables 6C and 10B will be met. If the IC does not specify the sequence of analyses, laboratory personnel should use their best professional judgment, based on the characteristics of the samples, to determine the order of processing the samples so that the results are obtained in the timeliest manner.

12. When all analyses are completed, validate the individual results and determine if all information requested has been finalized.

13. Review the screening and analytical process to assess where there may be a discrepancy between the screening process that indicated < 1,000 mrem (short-term exposure PAG) and the radioanalytical results that indicate > 1,000 mrem.

14. If the results of screening and background measurements compared with the sample activity are in reasonable correspondence, proceed with Step 1. If not re-review the analytical results prior to proceeding.

15. The final results for each sample are compared to project-specific criteria. Each individual radionuclide analyzed will be compared to its ADL value cited in Table 6C or 10B. Also calculate the sum of fractions using the following equation:

$$Sum\ of\ Fractions = \sum_{i=1}^{n} \left[\frac{R_i}{AAL_i} \right]$$

Where R_i is the activity of the individual radionuclide and AAL_i is the analytical action level of that radionuclide for the 1,000-mrem PAG at one year of exposure.

If the sum of fractions is > 1.0 then the 1,000-mrem PAG for short-term exposure has been exceeded. This means that there may be discrepancy between the screening and the final results, or that the screening process may need to be adjusted based on the specific radionuclides identified in these samples.

16. Contact the IC with the results of the analysis, in this case indicating that these results confirm that the radionuclides present are greater than the 1,000-mrem PAG based on the radionuclide activity concentration identified.

17. Sample aliquants and unused dissolved fusion melts should be archived for analysis at a future date if needed.

Additional Points for Scenario 3

Gamma Ray Analysis

The detonation of an IND will create many fission products that decay in mass chains by β^{-}/gamma emission. The heads of each mass chain are generally very short-lived (seconds to hours) and sampling of soil following such an event would not identify any of these “heads-of-chain.” However, several fission products form parent-progeny pairs that establish radiochemical equilibria after the event over the course of days to months. Some of the pairs that form these equilibria are identified in Table 5. Some pairs that are specifically *not* noted in this table are those that form secular equilibria with their progeny in time frames that are very short. Examples of these are ^{137}Cs–^{137}Ba (equilibrium achieved in ~10 min), and ^{106}Ru–^{106}Rh (equilibrium achieved in ~4 min).

Table 5 – Time for Certain Radionuclide Pairs to Achieve Maximum Progeny Activity

Radionuclide Pair	λ Parent (Days)	λ Progeny (Days)	Time to Peak Progeny Activity (Days) [1]	Type of Equilibrium	Decay Correction (post equilibrium)
Ba-140/La-140	5.435×10^{-2}	4.130×10^{-1}	5.7	Transient	λ Parent
Zr-95/Nb-95	1.082×10^{-2}	1.981×10^{-2}	67.3	Transient	λ Parent + Equation
Te-132/I-132	2.166×10^{-1}	7.295	0.50	Transient	λ Parent
I-131/Xe-131m	8.641×10^{-2}	5.824×10^{-2}	14.0	No	λ Progeny + Equation
Nd-147/Pm-147	6.311×10^{-2}	7.232×10^{-4}	71.6	No	λ Progeny + Equation
Ce-143/Pr-143	5.033×10^{-1}	5.107×10^{-2}	5.1	No	λ Progeny

[1] These times are calculated using the equation for the maximum number of progeny atoms, N_2, to achieve equilibrium:

$$N_2 = (\ln \lambda_1 - \ln \lambda_2)/(\lambda_1 - \lambda_2)$$

Practical times to reach equilibrium (e.g., 99% of maximum) are shorter than those identified above.

When analyzing samples, if either member of the pair is present the other must have been present (no equilibrium cases) or is still present (transient and secular equilibrium cases). For example if ^{140}La is identified by gamma ray spectrometry, ^{140}Ba must also be present as it is the longer-lived progenitor of the ^{140}La. Once the activity of each member of the radiochemical equilibrium is identified, the decay correction to a point in time may need to be determined. For the radionuclides identified in Table 5 (and those others with very short equilibration times), decay correction either to the time of sampling or forward to a future date has several different cases that need to be considered. These cases depend on the:

- Time that has passed from the time of the incident to the time of deposition,
- Chemical solubility differences of the pairs during the transport to the soil,
- Chemical solubility/transport in the soil matrix from deposition to sampling, and
- Length of time in the soil until sampling occurs.

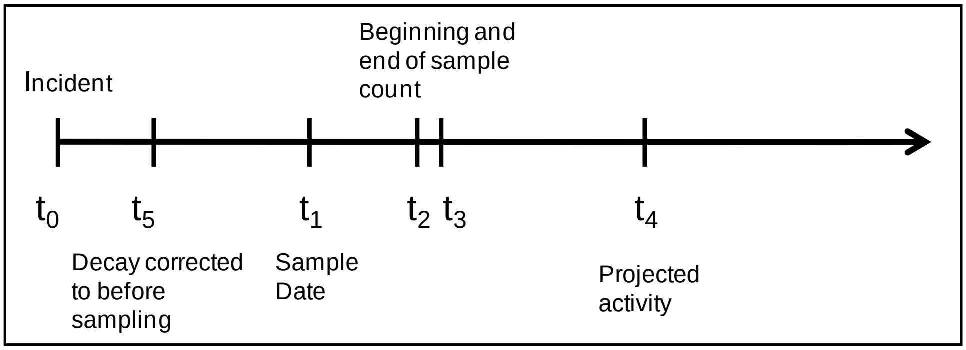

Figure 6 – Timeline for IND Parent-Progeny Activity Determinations

Each combination of the times and element solubilities noted here will represent a different case and will require a different solution (some much more complicated than others). The last column of Table 5 identifies a general method to be used *once it has been established* that equilibrium has been achieved. Figure 6 shows the sequence of events on a general timeline. These will be used in examples further on in this discussion.

Radionuclide results are generally decay corrected to the date and time of the sample collection. However, since the radionuclides in the decay chain will form the parent-progeny relationships starting with the event, it will take several hours to weeks until transient equilibrium[26] for the fission product pairs noted above will take effect. A simple default decay correction factor based on delta days between collection and analysis will require additional calculation outside the normal software options used in most gamma spectrometry systems.

For the transient equilibrium cases the half-life of the parent is used for decay corrections (backwards and forwards in time). Note for the ^{95}Zr/^{95}Nb pair that the time to achieve equilibrium is about 67 days. This means that some additional assumptions may need to be made and additional calculations involving an ingrowth formula would be needed, if equilibrium has not yet been established (see Example 1, below).

For the no equilibrium cases the half-life of the progeny is used for decay corrections (backwards and forwards in time). Note that for the ^{147}Nd/^{147}Pm pair, the time for ^{147}Pm to decay with its characteristic half-life is rather long: about 72 days. This means that some additional assumptions may need to be made and additional calculations involving an ingrowth formula would be needed, if equilibrium has not yet been established. Also note that for the ^{131}I/^{131m}Xe pair that xenon is a noble gas and this may cause issues with sample storage and handling, as well as with decay correction.

For all of these cases, the project MQOs will need to specify the assumptions to be made regarding the level of equilibrium in the sample that is to be assumed at the time of sampling. Additionally, specific equations that support the MQOs should be established in the project plan so that the assumptions and equations are consistently applied.

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[26] Some secular equilibrium pairs are not noted in the table as their time to equilibrium is measured in minutes. Examples of these are ^{137}Cs/^{137m}Ba, ^{103}Ru/^{103}Rh, and ^{106}Ru/^{106}Rh.

Two examples will provide some guidance on the details of correct decay calculations for these parent-progeny relationships.

Example 1

An incident has occurred on Day 0 at 1200 hours (t_0). A soil sample is taken on Day 5 at 1200 hours (t_1) and sent to a laboratory for analysis of ^{95}Zr–^{95}Nb. The laboratory decay corrects the sample activity concentration to the start of the count time, Day 7 at 1200 hours (t_2). The IC wants to know what the activity concentration will be on Day 15 at 1200 hours (t_4). The measured activity concentration of ^{95}Zr is 1×10^4 pCi/g and for ^{95}Nb is 600 pCi/g, decay corrected to the beginning of the sample count time (t_2).

Step 1. Assumptions. The chemical reactivity of the zirconium and niobium may not be exactly the same. Thus, from the time of the event to the time of soil deposition some chemical separation of the two may occur resulting in a non-predictable activity concentration of the progeny based on the parent.

Question: Is all of the ^{95}Nb in the soil sample supported, or is there unsupported ^{95}Nb?

Answer: Decay-correct the activity concentration of the sample for the parent (^{95}Zr in this case) to the time the sample was taken. Use that value to determine what the activity concentration of supported ^{95}Nb will be at the time of analysis. If the activity concentration at the time of analysis *is different* from that calculated based on the parent, two conditions can exist:

- Sample progeny activity concentration is greater than calculated (unsupported progeny is present)
- Sample progeny activity concentration is less than calculated (chemical separation of parent and progeny has occurred before after deposition)

Both of these can be solved for the activity concentration of the progeny using the generalized formulas

$$\lambda_d N_d = \frac{\lambda_d}{\lambda_d - \lambda_p} A_p^0 \left[e^{-\lambda_p \Delta t} - e^{-\lambda_d \Delta t}\right] + \lambda_d N_d^0 e^{-\lambda_d \Delta t} \tag{1}$$

$$A_n = A_{n_2}^0 e^{-\lambda_n (\Delta t)} \tag{2}$$

Where: λ_d is the decay constant of the progeny
λ_p is the decay constant of the parent
λ_n is the decay constant of radionuclide n
Δt is the time between t_0 and the time of analysis
N_d is the number of progeny atoms
N_p is the number of parent atoms
$_0$ refers to the parameter at time $t = 0$

Step 2. Decay-correct parent activity concentration. The activity concentration of the parent is unaffected by that of the progeny. Decay correct the activity concentration of the parent from time $t_{2\ (\text{analysis date})}$ to time $t_{1\ (\text{collection date})}$.

$$A_1 = A_2 e^{\lambda_1(t_2-t_1)} = (1x10^4\ pCi/g)\left(e^{(.693x2days)/(64\ days)}\right)$$

$$= 1.02\times10^4 \text{ pCi/g}$$

Step 3. Calculate the theoretical progeny activity concentration. Using Equation 1, where A_1 is A_p^0 and the term, $\lambda_d N_d^0 e^{-\lambda_d t}$, is set to zero (assumes progeny is only present from parent), the theoretical value of the progeny activity concentration at the time of analysis (t_2) is 392 pCi/g.

Step 4. Calculate the unsupported activity concentration for day 2. This is the difference between theoretical and measured progeny activity concentration or (600 – 392) = 208 pCi/g.

Step 5. Calculate the activity concentration for both radionuclides at Day 15.

$$\lambda_{Nb} N_{Nb} = \frac{\lambda_d}{\lambda_d - \lambda_p} A_p^0 \left[e^{-\lambda_p t} - e^{\lambda_d t}\right] + \lambda_d N_d^0 e^{-\lambda_d t}$$

$$= 2.2056 \times 1 \times 10^4 pCi/g\left[e^{-0.01081\ d^{-1}\times 8\ d} - e^{-0.01982\, d^{-1}*8\ d}\right] + 208 \times e^{-0.01982\ d^{-1}*8\ days}$$

$$= 1.58\times10^3 \text{ pCi/g}$$

$$A_{Zr} = A_2 e^{-\lambda_1(t_2-t_1)} = (1x10^4\ pCi/g)\left(e^{-0.01081\, d^{-1}*8\ d}\right) = 9.17x10^3\ pCi/g$$

The activities on Day 15 for the radionuclides are: ^{95}Zr = 9.17×10^3 pCi/g and ^{95}Nb = 1.58×10^3 pCi/g.

Example 2

An IND is detonated on Day 1. A soil sample is taken and sent to the lab on Day 30. Gamma spectrometry measurements at 1200 hours on Day 30 for ^{140}Ba and ^{140}La are 1.03×10^6 pCi/g and 1.18×10^6 pCi/g, respectively. Several members of the public were exposed in the area where this soil was taken on Day 5 of the event. This was not discovered until the samples were taken. The IC wants to know what the activities of these two radionuclides were on Day 5 1200 hours for the purposes of dose reconstruction.

Step 1: Assumption: the deposition of the material occurred within the first 5 days of the detonation. Is any ^{140}La in the soil sample analyzed on Day 30, unsupported? Although the two radionuclides are chemically different, after 25 days any unsupported lanthanum would be reduced to ~ 3×10^{-3}% (14.9 half-lives) of what it was on Day 5.

Step 2: Decay correct the parent ^{140}Ba back to Day 1 using Equation (2).

$$A_{Ba} = A_{Ba}^0 e^{\lambda_1(t_2-t_0)} = 1.03x10^6 \times e^{0.05435\times30} = 5.26\times10^6 \text{ pCi/g}$$

Caution: although the progeny is decaying *forward* with the half-life of the parent, it is not advisable to decay correct *backwards* in time. This is because the curve goes through a maximum that cannot be accounted for by simple exponential decay using Equation (2).

Step 3: Calculate the activity of the progeny on Day 5 using Equation (1)

$$\lambda_d N_d = A_{La}$$

$$= \left(\frac{0.413}{0.413 - 0.05345}\right) 5.26 \times 10^6 pCi/g \left[e^{-0.05345\,d^{-1} \times 5\,d} - e^{-0.413\,d^{-1} \times 5\,d}\right] = 3.85 \times 10^6 \text{ pCi/g}$$

Step 4: Calculate the activity concentration of the parent on Day 5.

$$A_{Ba} = A_{Ba}^0 e^{-0.05345\,d^{-1} \times 5\,d} = 4.03 \times 10^6\ pCi/g$$

Alpha and Beta Analysis

The most significant alpha emitter concentrations will be due to fissile material that has not undergone fission and some transuranic elements (TRU) formed as a result of multiple neutron capture on fissile material during the detonation. The principal alpha emitting radionuclides will likely be ^{234}U, ^{235}U, ^{238}U, and ^{239}Pu (if ^{235}U is used as the fissile material), and ^{239}Pu, ^{240}Pu, and ^{241}Am (if ^{239}Pu is used as the fissile material). A gross alpha screen directly on samples will suffer from significant self-shielding compromising any assessment of the alpha contribution. In Scenario 3, this assessment is postponed until after the samples have been dried, oxidized, fused, and dissolved as this will eliminate a good deal of material that would attenuate the alpha signal. It also is also possible that the leftover fissile material will be present as discrete radioactive particles (DRPs) and may be missed on an initial screening based on the particle size.

The most significant beta-only emitters from such an incident will be ^{89}Sr and ^{90}Sr. Both radionuclides will distribute in the same manner (chemically and physically) in the environment since they are chemically identical. Both radionuclides have high fission yields, thus their activity concentration will be significant. However, the ^{89}Sr contribution will decrease measurably due to its half-life (50 days). At the time of the incident the ratio of activity of ^{89}Sr/^{90}Sr will be approximately 167, and after 180 days will be 14.3.

There are several other beta-only emitting radionuclides: ^{99}Tc, ^{129}I, ^{93}Nb, ^{135}Cs, ^{241}Pu, and others. However, their long half-lives translate into low-activity concentrations relative to the gamma emitters and the radiostrontiums. Thus, from the IND perspective they are a much less significant contributor to the total activity concentration and dose. Therefore, for the short-term dose assessment (1,000-mrem PAG) in terms of activity concentration they will be insignificant contributors. When the 50-year exposure risk is assessed their activity concentration may need to be determined as they may contribute significantly to the long-term dose.

APPENDIX I. TABLES OF RADIOANALYTICAL PARAMETERS FOR RADIONUCLIDES OF CONCERN

The following tables list the AAL, ADL, and u_{MR} values for the radionuclides of concern. The tables contain calculated activity concentration values for gross screening or radionuclide-specific measurements for alpha, and beta/gamma-emitting radionuclides, respectively. Derivation of the ADL values for each of these tables can be found in Appendix VI. Tables 6, 7, 8, and 10 contain calculated activities for specific radionuclides. These values were calculated based on the Type I and Type II error rates presented in Appendix VI.[27] Table 9 identifies some radionuclides and their constants associated with an IND event.

The AALs in the tables in this appendix may be used as default values. They are based on generic conversions of the PAGs and risk-based dose levels to concentration in soil for a specific radionuclide based on decay and weathering.

The u_{MR} and ADL will change depending upon the acceptable decision error rate. The IC may provide incident-specific AALs or decision error rates that would supersede the values presented here. In this case, the laboratory will need to develop new tables for all values using the process described in Appendix VI.

The values represent the concentration found at the time of sampling. The PAG or risk-based numbers are derived from that concentration, for total dose at a time in the future, taking into account radioactive decay, resuspension, and weathering. All values in the tables have been rounded to two significant figures. The methodology, assumptions, and calculations that were used to derive these numbers can be found in FRMAC (2010).

FRMAC provides examples of using "marker radionuclides," i.e., gamma ray emitters, when screening samples to assess the potential of an individual sample exceeding PAG or risk-based limits without processing the sample for each radionuclide that may be present. Such an assessment relies on the fact that the distribution of radionuclides in the soil samples will be similar to those that have already been analyzed using radionuclide-specific methods. The screening process can then be used to estimate the sum of the fractions for all radionuclides assumed to be present and provide insight to incident command staff as to the degree of contamination. Decisionmakers should keep in mind that a full radiochemical analysis of the samples needs to be performed before these assumptions can be verified. Other factors to be considered are based on very broad assumptions, such as:

- Is the radionuclide mixture the same?
- Are there hot particles?
- How will weathering and resuspension affect dose?

Thus, it may be possible to draw preliminary conclusions from screening, but only if the relation of the marker radionuclide to other radionuclides is shown to be constant.[28]

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[27] A Type I error refers to rejecting the null hypothesis when it is true. A Type I decision error is sometimes called a "false rejection" or a "false positive."

[28] An example of this is that ^{137}Cs/^{89}Sr ratios may have one value close to the event site in time and location while mobility and atmospheric transport/chemistry will change the ratios at points distant in time and location. Thus, location and time factors should be used in assumption/estimates.

FRMAC also has doses for a two-year exposure period that are not included in this document. One reason is that several of the radionuclides that are short lived (half-lives of a few hours to several days) yield absurdly large values for the PAGs and action levels at two years. The proposed analytical action levels in this document can be easily adapted to most situations. If a specific event requires the two-year value PAGs of 500 mrem, FRMAC (2010) should be consulted.

Some of the assumptions made in the document to arrive at the values in these tables are:

- EPA Lifetime Excess Total Cancer Risk = 8.46×10^{-7} risk/mrem (FGR-13,[29] Table 7.6, p. 182)
- Risk Level: 1×10^{-4} or 1×10^{-6}
- PAGs: Early Phase = 1,000 mrem, 4 days
- First Year = 2,000 mrem
- Second Year = 500 mrem
- Fifty Year = 5,000 mrem
- Deposition Derived Response Level (DRL) (Dep_DRL) taken from Appendix C of the FRMAC Manual (FRMAC 2010) or calculated using TurboFRMAC 2010 (µCi/m^2)
- Sample size: 100 cm^2 × 2 cm deep (200 cm^3)
- Soil density: 1.6 g/cm^3

Table 6A – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to Early Phase and First Year PAGs in Soil Using Gross Alpha Screening Methods

	Early Phase 1,000-mrem PAG			**First Year 2,000 mrem PAG**		
Radionuclide	**AAL pCi/g**	**ADL pCi/g**	**u_{MR} pCi/g**	**AAL pCi/g**	**ADL pCi/g**	**u_{MR} pCi/g**
Gross Alpha[1]	8.6×10^{3}	4.3×10^{3}	2.6×10^{3}	410	210	130
Am-241	1.7×10^{3}	830	500	1.1×10^{3}	550	340
Cm-242	2.7×10^{4}	1.4×10^{4}	8.2×10^{3}	2.2×10^{4}	1.1×10^{4}	6.7×10^{3}
Cm-243	2.3×10^{3}	1.1×10^{3}	690	1.3×10^{3}	630	380
Cm-244	2.8×10^{3}	1.4×10^{3}	850	1.9×10^{3}	970	590
Np-237	3.1×10^{3}	1.6×10^{3}	950	1.3×10^{3}	650	400
Po-210	3.8×10^{4}	1.9×10^{4}	1.1×10^{4}	3.2×10^{4}	1.6×10^{4}	9.6×10^{3}
Pu-238	1.5×10^{3}	740	450	1.0×10^{3}	510	310
Pu-239	1.3×10^{3}	670	410	930	460	280
Pu-240	1.3×10^{3}	670	410	930	460	280
Ra-226	8.6×10^{3}	4.3×10^{3}	2.6×10^{3}	410	210	130
Th-228	3.2×10^{3}	1.6×10^{3}	960	500	250	150
Th-230	1.6×10^{3}	780	480	1.1×10^{3}	540	330
Th-232	Activity concentration for this radionuclide is not available.					
U-234	1.7×10^{4}	8.5×10^{3}	5.2×10^{3}	1.2×10^{4}	5.8×10^{3}	3.5×10^{3}
U-235	1.7×10^{4}	8.5×10^{3}	5.2×10^{3}	3.3×10^{3}	1.6×10^{3}	990
U-238	1.8×10^{4}	9.1×10^{3}	5.6×10^{3}	4.0×10^{3}	2.0×10^{3}	1.2×10^{3}
U-Nat	1.8×10^{4}	9.1×10^{3}	5.6×10^{3}	4.0×10^{3}	2.0×10^{3}	1.2×10^{3}

Note:

[1] The AAL, ADL and u_{MR} values for gross alpha shown in this table are for ^{226}Ra. Once specific radionuclides have been identified for an incident, the radionuclide with the lowest AAL, ADL and u_{MR} values should be selected for the respective gross activity screening measurements.

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[29] Federal Guidance Technical Report 13, *Cancer Risk Coefficients for Environmental Exposure to Radionuclide: Updates and Supplements*, www.epa.gov/radiation/federal/techdocs.html#report13.

Table 6B – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to Early Phase and First Year PAGs for Soil Using Gross Beta/Gamma Screening Methods

	Early Phase 1,000-mrem PAG			First Year 2,000 mrem PAG		
Radionuclide	**AAL pCi/g**	**ADL pCi/g**	**u_{MR} pCi/g**	**AAL pCi/g**	**ADL pCi/g**	**u_{MR} pCi/g**
Gross Beta[1] [2]	2.1×10^{5}	1.1×10^{5}	6.4×10^{4}	6.4×10^{3}	3.2×10^{3}	1.9×10^{3}
Gross Gamma[2]	1.3×10^{4}	6.5×10^{3}	3.9×10^{3}	330	160	100
Ac-227+DP	900	450	270	440	220	130
Ce-141	4.4×10^{5}	2.2×10^{5}	1.3×10^{5}	7.5×10^{4}	3.7×10^{4}	2.3×10^{4}
Ce-144	1.6×10^{5}	8.0×10^{4}	4.8×10^{4}	5.9×10^{3}	3.0×10^{3}	1.8×10^{3}
Co-57	2.7×10^{5}	1.4×10^{5}	8.3×10^{4}	9.9×10^{3}	5.0×10^{3}	3.0×10^{3}
Co-60	1.3×10^{4}	6.5×10^{3}	3.9×10^{3}	330	160	100
Cs-134	2.0×10^{4}	1.0×10^{4}	6.1×10^{3}	560	280	170
Cs-137	5.4×10^{4}	2.7×10^{4}	1.6×10^{4}	1.3×10^{3}	660	400
I-125	9.3×10^{5}	4.6×10^{5}	2.8×10^{5}	9.1×10^{4}	4.5×10^{4}	2.8×10^{4}
I-129	1.1×10^{6}	5.6×10^{5}	3.4×10^{5}	3.5×10^{4}	1.8×10^{4}	1.1×10^{4}
I-131	9.6×10^{4}	4.8×10^{4}	2.9×10^{4}	5.7×10^{4}	2.8×10^{4}	1.7×10^{4}
Ir-192	3.9×10^{4}	2.0×10^{4}	1.2×10^{4}	3.1×10^{3}	1.6×10^{3}	950
Mo-99	1.7×10^{5}	8.5×10^{4}	5.2×10^{4}	2.2×10^{5}	1.1×10^{5}	6.6×10^{4}
P-32	3.8×10^{5}	1.9×10^{5}	1.2×10^{5}	1.4×10^{5}	6.8×10^{4}	4.2×10^{4}
Pd-103	3.8×10^{6}	1.9×10^{6}	1.1×10^{6}	1.2×10^{6}	5.8×10^{5}	3.5×10^{5}
Pu-241	7.0×10^{4}	3.5×10^{4}	2.1×10^{4}	4.9×10^{4}	2.4×10^{4}	1.5×10^{4}
Ra-228	7.8×10^{3}	3.9×10^{3}	2.4×10^{3}	800	400	240
Ru-103	6.5×10^{4}	3.2×10^{4}	2.0×10^{4}	9.1×10^{3}	4.6×10^{3}	2.8×10^{3}
Ru-106	8.4×10^{4}	4.2×10^{4}	2.5×10^{4}	2.8×10^{3}	1.4×10^{3}	860
Se-75	8.5×10^{4}	4.2×10^{4}	2.6×10^{4}	4.7×10^{3}	2.3×10^{3}	1.4×10^{3}
Sr-89	4.4×10^{5}	2.2×10^{5}	1.3×10^{5}	4.9×10^{4}	2.5×10^{4}	1.5×10^{4}
Sr-90	2.1×10^{5}	1.1×10^{5}	6.4×10^{4}	6.4×10^{3}	3.2×10^{3}	1.9×10^{3}
Tc-99	1.2×10^{7}	5.8×10^{6}	3.6×10^{6}	4.7×10^{6}	2.4×10^{6}	1.4×10^{6}

Note:

[1] Several nuclides in this table decay by electron capture. These radionuclides cannot be detected using gross β analysis. The electron capture decay leads to characteristic X-rays of the progeny radionuclide. The most effective way to detect the specific X-rays from these electron-capture-decay radionuclides is either with a low-energy photon detector (LEPD) or a reverse electrode germanium detector (N-type semiconductor detector). The lower range of energy with these detectors is about 10 keV. Alternatively liquid scintillation counting could be used as a gross indicator of activity concentration.

[2] The AAL, ADL and u_{MR} values for gross beta shown in this table are for ^{90}Sr. The AAL, ADL and u_{MR} values for gross gamma shown in this table are for ^{60}Co. Once specific radionuclides have been identified for an incident, the radionuclide with the lowest AAL, ADL and u_{MR} values should be selected for the respective gross activity screening measurements.

Table 6C – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to Early Phase and First Year PAGs for Soil Using Radionuclide-Specific Methods for Alpha Emitters

	Early Phase 1,000-mrem PAG			**First Year 2,000 mrem PAG**		
Radionuclide	**AAL pCi/g**	**ADL pCi/g**	**u_{MR} pCi/g**	**AAL pCi/g**	**ADL pCi/g**	**u_{MR} pCi/g**
Am-241	1.7×10^3	1.2×10^3	210	1.1×10^3	780	140
Cm-242	2.7×10^4	1.9×10^4	3.4×10^3	2.2×10^4	1.6×10^4	2.8×10^3
Cm-243	2.3×10^3	1.6×10^3	290	1.3×10^3	890	160
Cm-244	2.8×10^3	2.0×10^3	350	1.9×10^3	1.4×10^3	240
Np-237	3.1×10^3	2.2×10^3	390	1.3×10^3	920	160
Po-210	3.8×10^4	2.7×10^4	4.7×10^3	3.2×10^4	2.2×10^4	4.0×10^3
Pu-238	1.5×10^3	1.0×10^3	190	1.0×10^3	720	130
Pu-239	1.3×10^3	950	170	930	660	120
Pu-240	1.3×10^3	950	170	930	660	120
Ra-226	8.6×10^3	6.1×10^3	1.1×10^3	410	290	52
Th-228	3.2×10^3	2.2×10^3	400	500	360	63
Th-230	1.6×10^3	1.1×10^3	200	1.1×10^3	760	140
Th-232	Activity concentration for this radionuclide is not available.					
U-234	1.7×10^4	1.2×10^4	2.1×10^3	1.2×10^4	8.2×10^3	1.5×10^3
U-235	1.7×10^4	1.2×10^4	2.1×10^3	3.3×10^3	2.3×10^3	410
U-238	1.8×10^4	1.3×10^4	2.3×10^3	4.0×10^3	2.8×10^3	510
U-Nat	1.8×10^4	1.3×10^4	2.3×10^3	4.0×10^3	2.8×10^3	510

Table 6D – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to Early Phase and First Year PAGs for Soil Using Radionuclide-Specific Methods for Beta/Gamma Emitters

	Early Phase 1,000-mrem PAG			**First Year 2,000 mrem PAG**		
Radionuclide	**AAL pCi/g**	**ADL pCi/g**	**u_{MR} pCi/g**	**AAL pCi/g**	**ADL pCi/g**	**u_{MR} pCi/g**
Ac-227+DP	900	640	110	440	310	55
Ce-141	4.4×10^5	3.1×10^5	5.6×10^4	7.5×10^4	5.3×10^4	9.4×10^3
Ce-144	1.6×10^5	1.1×10^5	2.0×10^4	5.9×10^3	4.2×10^3	750
Co-57	2.7×10^5	1.9×10^5	3.5×10^4	9.9×10^3	7.0×10^3	1.3×10^3
Co-60	1.3×10^4	9.2×10^3	1.6×10^3	330	230	42
Cs-134	2.0×10^4	1.4×10^4	2.5×10^3	560	400	71
Cs-137	5.4×10^4	3.8×10^4	6.8×10^3	1.3×10^3	930	170
I-125	9.3×10^5	6.6×10^5	1.2×10^5	9.1×10^4	6.4×10^4	1.1×10^4
I-129	1.1×10^6	7.9×10^5	1.4×10^5	3.5×10^4	2.5×10^4	4.5×10^3
I-131	9.6×10^4	6.8×10^4	1.2×10^4	5.7×10^4	4.0×10^4	7.1×10^3
Ir-192	3.9×10^4	2.8×10^4	4.9×10^3	3.1×10^3	2.2×10^3	390
Mo-99	1.7×10^5	1.2×10^5	2.2×10^4	2.2×10^5	1.5×10^5	2.7×10^4
P-32	3.8×10^5	2.7×10^5	4.8×10^4	1.4×10^5	9.7×10^4	1.7×10^4
Pd-103	3.8×10^6	2.7×10^6	4.7×10^5	1.2×10^6	8.2×10^5	1.5×10^5
Pu-241	7.0×10^4	4.9×10^4	8.8×10^3	4.9×10^4	3.4×10^4	6.1×10^3
Ra-228	7.8×10^3	5.5×10^3	980	800	560	100
Ru-103	6.5×10^4	4.6×10^4	8.2×10^3	9.1×10^3	6.5×10^3	1.1×10^3
Ru-106	8.4×10^4	5.9×10^4	1.1×10^4	2.8×10^3	2.0×10^3	350
Se-75	8.5×10^4	6.0×10^4	1.1×10^4	4.7×10^3	3.3×10^3	590
Sr-89	4.4×10^5	3.1×10^5	5.5×10^4	4.9×10^4	3.5×10^4	6.2×10^3
Sr-90	2.1×10^5	1.5×10^5	2.7×10^4	6.4×10^3	4.5×10^3	810
Tc-99	1.2×10^7	8.3×10^6	1.5×10^6	4.7×10^6	3.3×10^6	5.9×10^5

Table 7A – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to First Year Exposure at 10^{-4} and 10^{-6} Risk for Soil Using Gross Alpha Screening Methods

Radionuclide	**AAL 10^{-4} risk pCi/g**	**ADL 10^{-4} risk pCi/g**	**u_{MR} 10^{-4} risk pCi/g**	**AAL 10^{-6} risk pCi/g**	**ADL 10^{-6} risk pCi/g**	**u_{MR} 10^{-6} risk pCi/g**
Am-241	65	33	20	0.65	0.33	0.20
Cm-242	1.3×10^3	650	390	13	6.5	3.9
Cm-243	75	37	23	0.75	0.37	0.23
Cm-244	110	57	35	1.1	0.57	0.35
Np-237	77	38	23	0.77	0.38	0.23
Po-210	1.9×10^3	930	570	19	9.3	5.7
Pu-238	60	30	18	0.60	0.30	0.18
Pu-239	55	27	17	0.55	0.27	0.17
Pu-240	55	27	17	0.55	0.27	0.17
Ra-226	24	12	7.4	0.24	0.12	0.074
Th-228	30	15	9.0	0.30	0.15	0.090
Th-230	64	32	19	0.64	0.32	0.19
Th-232	Activity concentration for this radionuclide is not available.					
U-234	690	340	210	6.9	3.4	2.1
U-235	190	96	58	1.9	0.96	0.58
U-238	240	120	72	2.4	1.2	0.72
U-Nat	240	120	72	2.4	1.2	0.72

Table 7B – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to First Year Exposure at 10^{-4} and 10^{-6} Risk for Soil Using Gross Beta/Gamma Screening Methods

Radionuclide	AAL 10^{-4} risk pCi/g	ADL 10^{-4} risk pCi/g	u_{MR} 10^{-4} risk pCi/g	AAL 10^{-6} risk pCi/g	ADL 10^{-6} risk pCi/g	u_{MR} 10^{-6} risk pCi/g
Ac-227+DP	26	13	7.9	0.26	0.13	0.079
Ce-141	4.4×10^3	2.2×10^3	1.3×10^3	44	22	13
Ce-144	350	180	110	3.5	1.8	1.1
Co-57	590	290	180	5.9	2.9	1.8
Co-60	20	9.8	5.9	0.20	0.098	0.059
Cs-134	33	17	10	0.33	0.17	0.10
Cs-137	78	39	24	0.78	0.39	0.24
I-125	5.4×10^3	2.7×10^3	1.6×10^3	54	27	16
I-129	2.1×10^3	1.0×10^3	640	21	10	6.4
I-131	3.3×10^3	1.7×10^3	1.0×10^3	33	17	10
Ir-192	180	92	56	1.8	0.92	0.56
Mo-99	1.3×10^4	6.4×10^3	3.9×10^3	130	64	39
P-32	8.1×10^3	4.0×10^3	2.5×10^3	81	40	25
Pd-103	6.9×10^4	3.4×10^4	2.1×10^4	690	340	210
Pu-241	2.9×10^3	1.4×10^3	870	29	14	8.7
Ra-228	47	24	14	0.47	0.24	0.14
Ru-103	540	270	160	5.4	2.7	1.6
Ru-106	170	83	51	1.7	0.83	0.51
Se-75	280	140	84	2.8	1.4	0.84
Sr-89	2900	1.5×10^3	890	29	15	8.9
Sr-90	380	190	110	3.8	1.9	1.1
Tc-99	2.8×10^5	1.4×10^5	8.5×10^4	2.8×10^3	1.4×10^3	850

Table 7C – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to First Year Exposure at 10^{-4} and 10^{-6} Risk for Soil Using Radionuclide-Specific Methods for Alpha Emitters

Radionuclide	AAL 10^{-4} risk pCi/g	ADL 10^{-4} risk pCi/g	u_{MR} 10^{-4} risk pCi/g	AAL 10^{-6} risk pCi/g	ADL 10^{-6} risk pCi/g	u_{MR} 10^{-6} risk pCi/g
Am-241	65	46	8.2	0.65	0.46	0.82
Cm-242	1.3×10^3	920	160	13	9.2	1.6
Cm-243	75	53	9.4	0.75	0.53	0.094
Cm-244	110	81	14	1.1	0.81	0.14
Np-237	77	54	9.7	0.77	0.54	0.097
Po-210	1.9×10^3	1.3×10^3	230	19	13	2.3
Pu-238	60	43	7.6	0.60	0.43	0.076
Pu-239	55	39	6.9	0.55	0.39	0.069
Pu-240	55	39	6.9	0.55	0.39	0.069
Ra-226[1]	24	17	3.1	0.24	0.17	0.031
Th-228	30	21	3.7	0.30	0.21	0.037
Th-230	64	45	8.0	0.64	0.45	0.080
Th-232	Activity concentration for this radionuclide is not available.					
U-234	690	490	87	6.9	4.9	0.87
U-235	190	140	24	1.9	1.4	0.24
U-238	240	170	30	2.4	1.7	0.30
U-Nat	240	170	30	2.4	1.7	0.30

Note:

[1] The AAL for ^{226}Ra at the 10^{-6} risk is less than that concentration normally found in most soils. While these calculations are technically correct, it is unlikely that such an action level concentration would ever be applied to incident-response samples.

Table 7D – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to First Year Exposure at 10^{-4} and 10^{-6} Risk for Soil Using Radionuclide-Specific Methods for Beta/Gamma Emitters

Radionuclide	**AAL 10^{-4} risk pCi/g**	**ADL 10^{-4} risk pCi/g**	**u_{MR} 10^{-4} risk pCi/g**	**AAL 10^{-6} risk pCi/g**	**ADL 10^{-6} risk pCi/g**	**u_{MR} 10^{-6} risk pCi/g**
Ac-227+DP	26	18	3.3	0.26	0.18	0.033
Ce-141	4.4×10^{3}	3.1×10^{3}	560	44	31	5.6
Ce-144	350	250	44	3.5	2.5	0.44
Co-57	590	410	74	5.9	4.1	0.74
Co-60	20	14	2.5	0.20	0.14	0.025
Cs-134	33	24	4.2	0.33	0.24	0.042
Cs-137	78	55	9.8	0.78	0.55	0.098
I-125	5.4×10^{3}	3.8×10^{3}	680	54	38	6.8
I-129	2.1×10^{3}	1.5×10^{3}	260	21	15	2.6
I-131	3.3×10^{3}	2.4×10^{3}	420	33	24	4.2
Ir-192	180	130	23	1.8	1.3	0.23
Mo-99	1.3×10^{4}	9.1×10^{3}	1.6×10^{3}	130	91	16
P-32	8.1×10^{3}	5.7×10^{3}	1.0×10^{3}	81	57	10
Pd-103	6.9×10^{4}	4.8×10^{4}	8.6×10^{3}	690	480	86
Pu-241	2.9×10^{3}	2.0×10^{3}	360	29	20	3.6
Ra-228[1]	47	33	5.9	0.47	0.33	0.059
Ru-103	540	380	68	5.4	3.8	0.68
Ru-106	170	120	21	1.7	1.2	0.21
Se-75	280	200	35	2.8	2.0	0.35
Sr-89	2.9×10^{3}	2.1×10^{3}	370	29	21	3.7
Sr-90	380	270	48	3.8	2.7	0.48
Tc-99	2.8×10^{5}	2.0×10^{5}	3.5×10^{4}	2.8×10^{3}	2.0×10^{3}	350

Note:

[1] The AAL for ^{228}Ra at the 10^{-6} risk is less than that concentration normally found in most soils. While these calculations are technically correct, it is unlikely that such an action level concentration would ever be applied to incident-response samples.

Table 8A – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to Risk Limits for 50-Year Exposure for Soil Using Gross Alpha Screening Methods

Radionuclide	AAL 10^{-4} risk pCi/g	ADL 10^{-4} risk pCi/g	u_{MR} 10^{-4} risk pCi/g	AAL 10^{-6} risk [2] pCi/g	ADL 10^{-6} risk pCi/g	u_{MR} 10^{-6} risk pCi/g
Am-241	15	7.7	4.7	0.15	0.077	0.047
Cm-242	1.3×10^3	640	390	13	6.4	3.9
Cm-243	14	7.1	4.3	0.14	0.071	0.043
Cm-244	52	26	16	0.52	0.26	0.16
Np-237	6.3	3.1	1.9	0.063	0.031	0.019
Po-210	1.8×10^3	920	560	18	9.2	5.6
Pu-238	19	9.4	5.7	0.19	0.094	0.057
Pu-239	15	7.5	4.6	0.15	0.075	0.046
Pu-240	15	7.5	4.6	0.15	0.075	0.046
Ra-226[3]	1.0	0.52	0.31	0.010	5.2×10^{-3}	3.1×10^{-3}
Th-228[3]	12	6.2	3.8	0.12	0.062	0.038
Th-230	17	8.7	5.3	0.17	0.087	0.053
Th-232	Activity concentration for this radionuclide is not available.					
U-234	180	89	54	1.8	0.89	0.54
U-235	10	5.0	3.0	0.10	0.050	0.030
U-238	13	6.5	3.9	0.13	0.065	0.039
U-Nat	13	6.5	3.9	0.13	0.065	0.039

Notes:

[1] Values are based on the dose conversion factors taken from Appendix C of the FRMAC Manual (FRMAC 2010) or calculated using TurboFRMAC 2010 available from Sandia National Laboratory.

[2] The stated Ra, U, and Th AALs for the 10^{-6} risk levels may be below the inherent concentrations in typical soils. While these calculations are technically correct, it is unlikely that such an action level concentration would ever be applied to incident-response samples.

[3] Includes decay products originating from the ^{226}Ra or ^{228}Th in the body.

Table 8B – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to Risk Limits for 50-Year Exposure for Soil Using Gross Beta/Gamma Screening Methods

Radionuclide	AAL 10^{-4} risk pCi/g	ADL 10^{-4} risk pCi/g	u_{MR} 10^{-4} risk pCi/g	AAL 10^{-6} risk pCi/g	ADL 10^{-6} risk pCi/g	u_{MR} 10^{-6} risk pCi/g
Ac-227+DP	4.4	2.2	1.3	0.044	0.022	0.013
Ce-141	4.4×10^3	2.2×10^3	1.3×10^3	44	22	13
Ce-144	220	110	68	2.2	1.1	0.68
Co-57	380	190	120	3.8	1.9	1.2
Co-60	3.5	1.7	1.1	0.035	0.017	0.011
Cs-134	12	5.9	3.6	0.12	0.059	0.036
Cs-137	4.9	2.4	1.5	0.049	0.024	0.015
I-125[1]	5.3×10^3	2.6×10^3	1.6×10^3	53	26	16
I-129	86	43	26	0.86	0.43	0.26
I-131	3.3×10^3	1.7×10^3	1.0×10^3	33	17	10
Ir-192	180	90	54	1.8	0.90	0.54
Mo-99	1.3×10^4	6.4×10^3	3.9×10^3	130	64	39
P-32	8.1×10^3	4.0×10^3	2.5×10^3	81	40	25
Pd-103[1]	6.9×10^4	3.4×10^4	2.1×10^4	690	340	210
Pu-241	1.4×10^3	710	430	14	7.1	4.3
Ra-228[2]	8.7	4.3	2.6	0.087	0.043	0.026
Ru-103	540	270	160	5.4	2.7	1.6
Ru-106	93	46	28	0.93	0.46	0.28
Se-75[1]	250	120	75	2.5	1.2	0.75
Sr-89	2.9×10^3	1.5×10^3	880	29	15	8.8
Sr-90	24	12	7.3	0.24	0.12	0.073
Tc-99	2.2×10^4	1.1×10^4	6.7×10^3	220	110	67

Notes:

[1] I-125, Se-75, and Pd-103 decay by electron capture. These radionuclides cannot be detected using gross β screening analysis. They emit low-energy gamma rays and also X-rays from their respective progeny radionuclides. Liquid scintillation counting could be used as a gross screening technique for the presence of these radionuclides and other electron capture decay radionuclides.

[2] The AAL for ^{228}Ra at the 10^{-6} risk is less than that concentration normally found in most soils. While these calculations are technically correct, it is unlikely that such an action level concentration would ever be applied to incident-response samples.

Table 8C – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to Risk Limits for 50-Year Exposure for Soil Using Radionuclide-Specific Methods for Alpha Emitters

Radionuclide	AAL 10^{-4} risk pCi/g	ADL 10^{-4} risk pCi/g	u_{MR} 10^{-4} risk pCi/g	AAL 10^{-6} risk pCi/g	ADL 10^{-6} risk pCi/g	u_{MR} 10^{-6} risk pCi/g
Am-241	15	11	1.9	0.15	0.11	0.019
Cm-242	1.3×10^{3}	900	160	13	9.0	1.6
Cm-243	14	10	1.8	0.14	0.10	0.018
Cm-244	52	37	6.6	0.52	0.37	0.066
Np-237	6.3	4.4	0.79	0.063	0.044	7.9×10^{-3}
Po-210	1.8×10^{3}	1.3×10^{3}	230	18	13	2.3
Pu-238	19	13	2.4	0.19	0.13	0.024
Pu-239	15	11	1.9	0.15	0.11	0.019
Pu-240	15	11	1.9	0.15	0.11	0.019
Ra-226[1]	1.0	0.73	0.13	0.010	7.3×10^{-3}	1.3×10^{-3}
Th-228[1]	12	8.8	1.6	0.12	0.088	0.016
Th-230[1]	17	12	2.2	0.17	0.12	0.022
Th-232[1]	Activity concentration for this radionuclide is not available.					
U-234[1]	180	130	23	1.8	1.3	0.23
U-235[1]	10	7.1	1.3	0.10	0.071	0.013
U-238[1]	13	9.2	1.6	0.13	0.092	0.016
U-Nat[1]	13	9.2	1.6	0.13	0.092	0.016

Note:

[1] The stated Ra, U, and Th AALs for the 10^{-6} risk levels may be below the inherent concentrations in typical soils.

Table 8D – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to Risk Limits for 50-Year Exposure for Soil Using Radionuclide-Specific Methods for Beta/Gamma Emitters

Radionuclide	AAL 10^{-4} risk pCi/g	ADL 10^{-4} risk pCi/g	u_{MR} 10^{-4} risk pCi/g	AAL 10^{-6} risk pCi/g	ADL 10^{-6} risk pCi/g	u_{MR} 10^{-6} risk pCi/g
Ac-227+DP	4.4	3.1	0.55	0.044	0.031	5.5×10^{-3}
Ce-141	4.4×10^{3}	3.1×10^{3}	550	44	31	5.5
Ce-144	220	160	28	2.2	1.6	0.28
Co-57	380	270	48	3.8	2.7	0.48
Co-60	3.5	2.5	0.44	0.035	0.025	4.4×10^{-3}
Cs-134	12	8.3	1.5	0.12	0.083	150
Cs-137	4.9	3.4	0.61	0.049	0.034	6.1×10^{-3}
I-125	5.3×10^{3}	3.7×10^{3}	670	53	37	6.7
I-129	86	61	11	0.86	0.61	0.11
I-131	3.3×10^{3}	2.4×10^{3}	420	33	24	4.2
Ir-192	180	130	23	1.8	1.3	0.23
Mo-99	1.3×10^{4}	9.1×10^{3}	1.6×10^{3}	130	91	16
P-32	8.1×10^{3}	5.7×10^{3}	1.0×10^{3}	81	57	10
Pd-103	6.9×10^{4}	4.8×10^{4}	8.6×10^{3}	690	480	86
Pu-241	1.4×10^{3}	1.0×10^{3}	180	14	10	1.8
Ra-228	8.7	6.1	1.1	0.087	0.061	0.011
Ru-103	540	380	68	5.4	3.8	0.68
Ru-106	93	65	12	0.93	0.65	0.12
Se-75	250	180	31	2.5	1.8	0.31
Sr-89	2.9×10^{3}	2.1×10^{3}	370	29	21	3.7
Sr-90	24	17	3.0	0.24	0.17	0.030
Tc-99	2.2×10^{4}	1.6×10^{4}	2.8×10^{3}	220	160	28

Table 9 – Additional Radionuclides Potentially Present in Soil Immediately Following an IND

Radionuclide	Half-Life, Days	Principal Gamma Ray Energy, keV	Source
^{239}Np	2.4	106	Activation of ^{238}U
^{24}Na	0.63	1369	Activation of ^{23}Na
^{133}Xe	5.2	81	Decay of ^{133}I($t_{1/2}$ 0.87 d)
^{131m}Xe	11.9	164	Decay of ^{131}I($t_{1/2}$ 8.04 d)
^{132}Te – ^{132}I [1]	3.2	668	A = 132 decay chain
^{143}Ce	1.4	293	A = 143 decay chain
^{105}Rh	1.475	319	A = 105 decay chain
^{147}Nd – ^{147}Pm [2]	11	91	A = 147 decay chain

Note:

These radionuclides are in addition to other fission products identified in the previous and subsequent tables.

[1] The half-life is expressed as that of the parent, ^{132}Te, because they will be in transient equilibrium within about 12 hours of the incident.

[2] These radionuclides represent a no equilibrium case. The half-life is that of ^{147}Nd. The progeny, ^{147}Pm, has a 2.6-year half-life but has very low gamma ray abundance and would likely be detected only after several months of sample decay using a long count time.

Table 10A – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to Early Phase 1,000-mrem and First Year 2,000-mrem PAG Values for Fission Products in Soil Using Gross Beta/Gamma Screening Methods

	1,000-mrem PAG[1]			**2,000 mrem PAG[1]**		
Radionuclide	**AAL pCi/g**	**ADL pCi/g**	**u_{MR} pCi/g**	**AAL pCi/g**	**ADL pCi/g**	**u_{MR} pCi/g**
Ce-141	4.4×10^{5}	2.2×10^{5}	1.3×10^{5}	7.5×10^{4}	3.7×10^{4}	2.3×10^{4}
Ce-143	Activity concentration for this radionuclide is not available.					
Ce-144/Pr-144m/Pr-144	1.6×10^{5}	8.0×10^{4}	4.8×10^{4}	5900	3.0×10^{3}	1.8×10^{3}
Co-60	1.3×10^{4}	6.5×10^{3}	3.9×10^{3}	330	160	100
Cs-134	2.0×10^{4}	1.0×10^{4}	6.1×10^{3}	560	280	170
Cs-137/Ba-137m	5.4×10^{4}	2.7×10^{4}	1.6×10^{4}	1.3×10^{3}	660	400
I-129	1.1×10^{6}	5.6×10^{5}	3.4×10^{5}	3.5×10^{4}	1.8×10^{4}	1.1×10^{4}
I-131	9.6×10^{4}	4.8×10^{4}	2.9×10^{4}	5.7×10^{4}	2.8×10^{4}	1.7×10^{4}
I-132	4.0×10^{5}	2.0×10^{5}	1.2×10^{5}	7.9×10^{5}	4.0×10^{5}	2.4×10^{5}
I-133	1.6×10^{5}	8.0×10^{4}	4.9×10^{4}	3.1×10^{5}	1.5×10^{5}	9.3×10^{4}
La-140/Ba-140	1.4×10^{4}	7.1×10^{3}	4.3×10^{3}	5.6×10^{3}	2.8×10^{3}	1.7×10^{3}
Mo-99/Tc-99m	1.7×10^{5}	8.5×10^{4}	5.2×10^{4}	2.2×10^{5}	1.1×10^{5}	6.6×10^{4}
Np-239	3.1×10^{5}	1.6×10^{5}	9.5×10^{4}	4.3×10^{5}	2.2×10^{5}	1.3×10^{5}
Nd-147/Pm-147 [2]	2.2×10^{7}	1.1×10^{7}	6.6×10^{6}	1.0×10^{7}	5.2×10^{6}	3.2×10^{6}
Rh-105	Activity concentration for this radionuclide is not available.					
Ru-103/Rh-103m	6.5×10^{4}	3.2×10^{4}	2.0×10^{4}	9.1×10^{3}	4.6×10^{3}	2.8×10^{3}
Ru-106/Rh-106	8.4×10^{4}	4.2×10^{4}	2.5×10^{4}	2.8×10^{3}	1.4×10^{3}	860
Sr-89	4.4×10^{5}	2.2×10^{5}	1.3×10^{5}	4.9×10^{4}	2.5×10^{4}	1.5×10^{4}
Sr-90/Y-90	2.1×10^{5}	1.1×10^{5}	6.4×10^{4}	6.4×10^{3}	3.2×10^{3}	1.9×10^{3}
Tc-99	1.2×10^{7}	5.8×10^{6}	3.6×10^{6}	4.7×10^{6}	2.4×10^{6}	1.4×10^{6}
Zr-95/Nb-95m/ Nb-95	2.1×10^{4}	1.1×10^{4}	6.5×10^{3}	1.9×10^{3}	970	590

Notes:

[1] The values in this table for some radionuclides may be different from values shown in previous tables as these values assume equilibrium between the progenitors and the progeny. The values in the previous tables assume single radionuclides except where noted.

[2] This is the dose equivalent activity concentration from the ^{147}Pm.

Table 10B – Analytical Action and Decision Levels (AAL and ADL) and Required Method Uncertainty Corresponding to Early Phase 1,000-mrem and First Year 2,000-mrem PAG Values for Fission Products in Soil Using Radionuclide-Specific Methods for Beta/Gamma Emitters

	1,000-mrem PAG[1]			2,000 mrem PAG[1]		
	AAL pCi/g	ADL pCi/g	u_{MR} pCi/g	AAL pCi/g	ADL pCi/g	u_{MR} pCi/g
Ce-141	4.4×10^{5}	3.1×10^{5}	5.6×10^{4}	7.5×10^{4}	5.3×10^{4}	9.4×10^{3}
Ce-143	Activity concentration for this radionuclide is not available.					
Ce-144	1.6×10^{5}	1.1×10^{5}	2.0×10^{4}	5.9×10^{3}	4.2×10^{3}	750
Co-60	1.3×10^{4}	9.2×10^{3}	1.6×10^{3}	330	230	42
Cs-134	2.0×10^{4}	1.4×10^{4}	2.5×10^{3}	560	400	71
Cs-137	5.4×10^{4}	3.8×10^{4}	6.8×10^{3}	1.3×10^{3}	930	170
I-129	1.1×10^{6}	7.9×10^{5}	1.4×10^{5}	3.5×10^{4}	2.5×10^{4}	4.5×10^{3}
I-131	9.6×10^{4}	6.8×10^{4}	1.2×10^{4}	5.7×10^{4}	4.0×10^{4}	7.1×10^{3}
I-132	4.0×10^{5}	2.8×10^{5}	5.0×10^{4}	7.9×10^{5}	5.6×10^{5}	1.0×10^{5}
I-133	1.6×10^{5}	1.1×10^{5}	2.0×10^{4}	3.1×10^{5}	2.2×10^{5}	3.9×10^{4}
La-140/Ba-140	1.4×10^{4}	1.0×10^{4}	1.8×10^{3}	5.6×10^{3}	4.0×10^{3}	700
Mo-99/Tc-99m	1.7×10^{5}	1.2×10^{5}	2.2×10^{4}	2.2×10^{5}	1.5×10^{5}	2.7×10^{4}
Np-239	3.1×10^{5}	2.2×10^{5}	3.9×10^{4}	4.3×10^{5}	3.1×10^{5}	5.5×10^{4}
Nd-147/Pm-147[2]	2.2×10^{7}	1.7×10^{7}	2.8×10^{6}	1.0×10^{7}	7.4×10^{6}	1.3×10^{6}
Rh-105	Activity concentration for this radionuclide is not available.					
Ru-103/Rh-103m	6.5×10^{4}	4.6×10^{4}	8.2×10^{3}	9.1×10^{3}	6.5×10^{3}	1.1×10^{3}
Ru-106/Rh-106	8.4×10^{4}	5.9×10^{4}	1.1×10^{4}	2.8×10^{3}	2.0×10^{3}	350
Sr-89	4.4×10^{5}	3.1×10^{5}	5.5×10^{4}	4.9×10^{4}	3.5×10^{4}	6.2×10^{3}
Sr-90/Y-90	2.1×10^{5}	1.5×10^{5}	2.7×10^{4}	6.4×10^{3}	4.5×10^{3}	810
Tc-99	1.2×10^{7}	8.3×10^{6}	1.5×10^{6}	4.7×10^{6}	3.3×10^{6}	5.9×10^{5}
Zr-95/Nb-95m/Nb-95	2.1×10^{4}	1.5×10^{4}	2.7×10^{3}	1.9×10^{3}	1.4×10^{3}	240

[1] The values in this table for some radionuclides may be different from values shown in previous tables as these values assume equilibrium between the progenitors and the progeny. The values in the previous tables assume single radionuclides except where noted.

[2] This is the dose equivalent activity concentration from the ^{147}Pm.

APPENDIX II. EXAMPLE OF HIGH RADIONUCLIDE CONCENTRATION IN SOIL (RADIOANALYTICAL SCENARIO 1: HIJACKED RESIN SHIPMENT DISPERSED BY IGNITION)

Description

A shipment of resins used at a nuclear power plant to decontaminate the reactor coolant system was being transferred from the power plant to a long-term storage facility on May 5. The shipment contained 750 ft^3 of resin estimated to contain over 1,500 curies of mixed radioactive fission products at the time of the incident. It had been stored at the facility for over 6 years to allow the level of activity to decay down to a more manageable transport level. The table identifies the breakdown of the radioactive contaminants contributing most significantly to the total curies.

Table 11 – Principal Radionuclides Identified in Resins

Radionuclide	**^{90}Sr**	**^{241}Am**	**^{137}Cs**	**^{99}Tc**	**Other Radionuclides**[1]	**Total**
Half-Life	28.8 [1]	433 [1]	30.1 [1]	2.1×10^5 [1]	—	—
Activity, Ci	400	100	250	600	150	1500

Note:

[1] A mixture of ^{60}Co, ^{65}Zn, ^{106}Ru, ^{14}C, ^{3}H, ^{129}I, ^{241}Pu, ^{238}Pu and $^{239+240}$Pu

The IC has indicated that the ratios of ^{241}Pu, ^{238}Pu and $^{239+240}$Pu isotopes in the source term are known.

The shipment was hijacked *en route*, by terrorists. The high integrity containers (HIC) used to hold the radioactive materials were opened by drilling holes in the outer shell and charges of C4 explosive were implanted into the HIC. The charges on the HIC were detonated in an open field. This caused a spread of resins in an area of about 1 acre. The field was set ablaze with gasoline. The meteorological conditions during the 134 hours of active burning and smoldering of the field were steady wind from the SSW at 4–11 mph (6–18 kph), no precipitation, and average daily temperature of 81 °F (27 °C). Soil samples in several downwind and upwind directions are being collected to assess the spread and extent of contamination. The incident command has decided that the soil samples are to be analyzed for first year exposure PAG values (e.g., the first year 2,000 mrem AAL is 330 pCi/g for ^{60}Co, and 6,400 pCi/g of ^{90}Sr and 930 pCi/g ^{239}Pu). All values should be reported on a dry weight soil basis in pCi/g. Samples that are screened and found to be above that level based on screening results are to be analyzed first and all other samples held in suspense until further notice.

Event Sequence

It is Day 7 of the event. The incident responders have established a field office for coordinating response efforts including a laboratory project manager. At 1000 hours of Day 7, the incident-response team sends the laboratory five, 320 gram soil samples taken from the affected area downwind of the burn area that have been field screened at significantly above background radiation levels. The samples arrive at the laboratory at *Day 8, 1300 hours.* A 24-h turnaround time has been requested.

Analysis Paths

Step 1. Sample receipt screening uses a micro-R meter. Laboratory personnel perform an initial screen of the sample shipping container prior to opening it. The contact reading at the surface of the transport container is 4 mR/h. The sample transport container is moved to a mini-hot-cell.

The sample containers are removed from the transport container, and assessed for external contamination using standard swipe techniques. None are externally contaminated.

Day 8, 1330 hours

Step 1a. Each container has approximately 320 g of soil. Containers are counted for gross gamma for 1 minute each on a shielded 3×3" NaI(Tl) detector. The lower level discriminator is set at 50 keV and all counts up to 2000 keV will be captured as gross gamma activity.

A 2-g aliquant of each sample is transferred to a liquid scintillation vial with cocktail and counted with a wide open window without discrimination between alpha or beta. It is recognized that the risk of false non-detection of analyte, and the measurement uncertainty for a 2-gram subsample of unprocessed, unhomogenized soil will be high. Still, this test can provide important indications about the overall activity of samples under the plume and will be used to help make decisions about safe handling and contamination control.

Day 8, 1400 hours

Steps 2a and 2b. Example results of screening analysis for gamma and gross alpha/beta are shown in Table 12.

Table 12 – Results of Screening Analysis for Soil Samples

Container ID	Total counts, NaI(Tl) detector	Gross cpm Liquid Scintillation Counter
1	28,830	142
2	1,167,550	1,352
3	110	32
4	543,550	1,782
5	2,047,550	3,892
Background, (± 1σ)	30 ± 6	12 ± 2

The NaI(Tl) detector has been calibrated using a ^{60}Co standard in 320 grams of soil in the same type of container in which samples are received and has an efficiency of 31%. The background is 30 counts for a 10 minute count time. The sample activities (± CSU, *k*=1) estimated are shown here:

Sample 1:

$$\text{Gross gamma}, \frac{\text{pCi}}{\text{g}} = \frac{(28{,}830 - 30)\ \text{counts}}{(0.31 \times 1\ \text{minute} \times 320\ \text{g}\ \times 2.22)} = 131\ (\pm\ 33)\ \text{pCi/g}$$

Sample 2: 5,300 (± 1,300) pCi/g
Sample 3: 0.36 (± 0.10) pCi/g
Sample 4: 2,470 (± 620) pCi/g
Sample 5: 9,300 (± 2,300) pCi/g

The liquid scintillation counter was calibrated for single channel analysis with a ^{241}Am standard and yielded an efficiency of 4.5%, and with a ^{90}Sr/^{90}Y soil standard and yielded a beta efficiency of 9.5%. Due to high quench, samples were not counted in alpha/beta discriminating mode. The lower of the two efficiencies was used to conservatively minimize the risk of underestimating measured activities.

Following the removal of detritus and sub-sampling, a 2-g aliquant is suspended in 15 mL of a gelling liquid scintillation cocktail and counted for 10 minutes on the LSC. This yields a gross activity concentration (± CSU, k=1) for total alpha plus beta of:

Sample 1: $Gross\ Alpha + Beta\ Activity, \frac{pCi}{g} = \frac{[142\ cpm\ - 12\ cpm\,]}{0.045 \times 2\ g \times 2.22} = 650\ pCi/g$
Sample 2: 6,700 pCi/g
Sample 3: 100 pCi/g
Sample 4: 8,800 pCi/g
Sample 5: 19,000 pCi/g

In Steps 2a and 2b, samples are to be assessed based on the alpha/beta, and gamma results.

All samples meet the MQO for required method uncertainty for gross gamma (i.e., 100 pCi/g at or below the AAL of 330 pCi/g, or less than 31% above the AAL). Due to the small aliquant of a potentially non-homogenous sample, there is significant risk that uncertainty estimates will not adequately reflect the non-representative aliquant, and that alpha- and beta-emitter activity present in the sample might go unidentified. Although gross alpha and beta results will not be used to make any final decision about whether an AAL has been exceeded, this data will be used to make decisions about handling and processing samples (i.e., priority and minimizing the risk of contamination and cross-contamination). Comparing gross alpha and beta results based on a small aliquant against the gross gamma results based on the entire sample may show indications of potential non-homogeneity.

The measured combined standard uncertainty is less than the required method uncertainty, and the measured sample activity for samples 2, 4, and 5 are above the gross gamma ADL of 160 pCi/g (based on the limiting case of ^{60}Co). These samples are put on the red path to Step 3. The combined alpha/beta for sample 1 is above the limiting ADL for combined gross alpha+beta activity of 210 pCi/g so it is also placed on the red path to Step 3. Sample 3 falls below the ADL for both gross gamma and gross alpha +beta and is placed on the green path to Step 2b. In Step

2b both the sum of the gross gamma and gross alpha+beta activity is less than the limiting ADL of 210 pCi/g and sample 3 is relegated to the lowest priority yellow path.

Results of both screens indicate that samples 1, 2, 4 and 5 are of higher yet similar enough activity levels that they may be processed together using elevated levels of contamination control. The gross alpha+beta activity parallels the gross gamma activity and does not provide any preliminary indications of issues with potential gross non-homogeneity. The gross gamma activity is somewhat greater than the gross alpha+beta indicating the possible presence of pure alpha, beta, or low-energy emitters or in the samples.

The time is Day 8, 1500 hrs.

Step 3. The soil contains twigs and stones greater than about ½" in diameter. These are removed with the aid of a stainless steel forceps and probe and set aside as there has yet to be direction on how to address an activity concentration in these materials. There is considerable moisture and organic content to the soil. The samples are individually coned and quartered into two 160 gram fractions.

Working quickly, the volatile fraction is isolated and transferred to an air-tight sample container to minimize the loss of volatile radionuclides. The non-volatile fraction is transferred to a tared half-liter paint can. The non-volatile samples are placed in a drying oven at 110±10 °C.

The time is Day 8, 1525 hours

Step 4a, Samples 2, 4, and 5.

After the samples have dried to constant weight, percent solids are calculated for each sample for correction of “as-received” aliquants to “dry-weight” aliquants in Steps 6 and 7b. Stainless steel balls are added to each paint can and the cans are shaken for about 5-10 minutes to mill and homogenize the samples to solids that conform to the visual size standard. Aliquants for each radionuclide-specific test are transferred to crucibles. Additional aliquants are set up without tracers for gross alpha and beta screening in Step 7. Quality controls are also set up for each batch, and appropriate tracers and carriers added to all sample aliquants. The crucibles are transferred to a furnace where a ramped program is used to incrementally raise the temperature to ~600 °C to remove all organic materials.

The time is Day 8, 1550 hours.

Step 4b, Samples 2, 4, and 5.

Samples are aliquanted for gamma spectral analysis from the volatile fraction isolated in Step 3. Working quickly to minimize losses, the sample is mixed in the container and 75 mL of “as received” soil is transferred to a tared container matching calibrated gamma geometry and sealed. The sample is weighed to determine the net mass of the “as received” aliquant.

Representative 1-gram aliquants are taken from the mixed sample for analysis of volatile radionuclides. Based on general knowledge about the source term, analyses performed will be for ^{129}I and ^{99}Tc. Quality controls are also set up for each batch and all samples spiked with appropriate tracers and carriers. Samples are digested using the validated methods that minimize losses to volatilization for each of these radionuclides.

The dry-weight equivalent for each of the volatile radionuclide aliquants is calculated by applying the percent solids determined in Step 4a. This "dry-weight" aliquant will be used to calculate gamma results on a dry-weight basis in Steps 6 and 7b.

The time is Day 8, 1625 hours.

Step 5a, Samples 1, 2, 4, and 5.

The samples dried and oxidized in Step 4a are dissolved using a sodium carbonate fusion technique. The melt is redissolved in hydrochloric acid and visually inspected to verify that complete dissolution of the solids was obtained.

The time is Day 8, 1945 hours

Step 5b, Samples 1, 2, 4, and 5.

The samples aliquanted in Step 4b are digested using a basic wet digestion technique. The product of the dissolution is visually inspected to verify that complete dissolution of the solids was obtained.

The time is Day 8, 1800 hours

Step 6, Samples 1, 2, 4, and 5. The samples prepared for gamma analysis in Step 4b are counted 5 cm from the surface of the HPGe detector for approximately 30 minutes to meet the limiting first year 2,000 mrem MQO for required method uncertainty for ^{60}Co of 42 pCi/g at the AAL of 330 pCi/g. The detector used is calibrated for the same 75-mL geometry in which the samples are counted. As the samples count, the 59.5 keV gamma ray for ^{241}Am is identified, and the most abundant characteristic gamma ray from ^{241}Pu is present but with low very peak area. Although there is high uncertainty, these both tentatively indicate the presence of Pu isotopes in the sample. Since the IC has indicated that the ratios of Pu isotopes in the source term are known, the more complex and time intensive analysis for ^{241}Pu will not be required.

The "dry-weight" equivalent is entered into analysis software as the sample aliquant since results will be reported on a "dry-weight" basis. Following the count, all unidentified peaks in the spectrum with a standard uncertainty less than 50% are identified and added to the library if they will significantly impact final quantitation of results.

The time is Day 8, 1725 hours

Step 7, Samples 1, 2, 4, and 5.

Aliquants of the sample set up for gross alpha and beta screening in Steps 4a and 4b are analyzed for gross alpha and beta on a liquid scintillation counter. Given the smaller size of the aliquants and the cleaner matrix after dissolution, alpha beta discrimination is practicable. The liquid scintillation counter has been optimized to reliably detect low levels of alpha activity in the presence of elevated beta activity by using a longer discriminator time setting for the pulse shape discriminator (longer than the cross-over point) to minimize beta-to-alpha crosstalk. The LSC is calibrated with pure ^{238}Pu (since other isotopes of Pu present in ^{239}Pu mixtures interfere with cross-talk measurements) and with ^{90}Sr/^{90}Y standards. The dissolved melt from the fusion is taken to dryness and redissolved in 1 mL of 0.1 M nitric acid. 19 mL of liquid scintillation cocktail are added, the vial is capped, shaken, and the samples counted for 5 minutes each. The results of the screen are listed in Table 13.

Table 13 – Results of Screening Analysis for Soil Samples

Sample ID	Gross Alpha Activity (pCi/g dry-weight)	Gross Beta Activity (pCi/g dry-weight)
1	44 ± 30	191 ± 53
2	1,000 ± 260	5,400 ± 1,300
4	1,330 ± 340	7,100 ± 1,800
5	2,910 ± 730	15,500 ± 3,900

All four gross alpha and beta results satisfy the MQO for required (relative) method uncertainty (based on limiting ADLs ^{239}Pu and ^{60}Co in Tables 6A and 6B). All four samples have combined alpha+beta activities that exceed the limiting ADL for screening, so they continue on the red path to Step 7a.

The time is Day 8, 2045 hours

Step 7a, Samples 1, 2, 4, and 5.

Although ^{241}Am was identified by the gamma spectral analysis, it still will be determined by alpha spectrometry due to the possibility of bias for the low-energy 59.5 keV gamma ray.

Samples are aliquanted for the Pu and Am analysis. The results of gross alpha and gross beta screening indicate, however, that a reduced aliquant for sequential analysis of Am and Pu isotopes will be necessary to prevent the tracer from being overwhelmed by analyte, and to minimize the risk of contamination of the detector. The size of aliquant containing ~50 pCi of alpha is calculated from the gross alpha results. Samples 2, 3 and 5 are diluted to a known volume of 100 mL in dilute hydrochloric acid and 50, 1, 20 and 0.5 mL aliquants are taken, respectively. Since the amount of tracer originally added to the samples prior to dissolution is no longer be sufficient to produce the minimum number of tracer counts needed for the yield determination, an additional 60 dpm ^{236}Pu and ^{243}Am tracers is added to each aliquant. The tracer activity used for calculations is adjusted to reflect the total of original and added tracer present in the samples. The lower activity aliquants are then processed using a validated rapid sequential method for Pu and Am analysis.

Although the late addition of tracer is not optimal, chemical separations have yet to be performed and the dissolution process should be near quantitative as run, so there should be little impact on the data quality. Still, the need to perform the dilution and replenish tracer in the samples will be clearly noted in the case narrative.

The time is Day 9, 0145 hours

Step 7b, Samples 1, 2, 4, and 5. The product of the sample dissolutions from Step 5b is taken for analysis using validated rapid methods of ^{99}Tc and ^{129}I.

The time is Day 9, 0200 hours

Step 8, Samples 1, 2, 4, and 5. Each set of results is reviewed to ensure that the correct process was followed, that all manual data entry is accurate, that all QC criteria are met, results are appropriately qualified, and that all anomalous situations or deviations from standard operating procedures are narrated. These results are approved by the laboratory supervisor.

The analytical results from the samples are shown in Table 14 in units of pCi/g, dry weight.

Table 14 – Results of Radionuclide Specific Analysis in Soil Samples

Sample	^{241}Am	^{238}Pu	$^{239+240}$Pu	**Total α Activity**	**α fraction**	^{90}Sr	^{90}Y[30]	^{99}Tc	^{60}Co	^{137}Cs	^{129}I	Total β + γ activity	β + γ fraction	Sum of Fractions >1.0
1	39	—	24	63	0.1	45	45	0	41	52	99	327	0.2	N
2	310	20	205	535	0.5	1,000	1,000	580	900	1,160	2,200	7,840	3.8	Y
3	—	—	—	—	—	—		—	—	—	—	—	—	—
4	4,680	75	800	5555	2.1	1,100	1,100	400	150	330	100	4,280	0.9	Y
5	7,000	80	1,000	8,080	2.9	3,800	3,800	2,600	1,250	4,700	1,500	21,450	8.0	Y
2,000 mrem ADL	780	720	660	—	—	4,500	n/a	3.3×10^6	230	930	2.5×10^4	—	—	—
2,000 mrem AAL	1,100	1,000	930	—	—	6,400	n/a	4.7×10^6	330	1,300	3.5×10^4	—	—	—

Results are evaluated to determine whether AAL values have been exceeded based on individual radionuclide results (as batches of data are completed). The MQO for required method uncertainty has been met for all analyses. Based on individual results, one or more values for either an alpha or gamma emitter is greater than the ADL in samples 2, 4, and 5 and it is concluded that the respective AALs have been exceeded. The IC is notified of the preliminary results while the remaining analyses proceed to completion (broken line to Step 11).

The time is Day 9, 0230 hours

Step 9, Samples 2, 4, and 5. The total α and β+γ activity columns represent the sum of all the activities from those radionuclide groupings. The sum of the radionuclide-specific results is

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[30] Assumes secular equilibrium of ^{90}Y with ^{90}Sr. Note that the derivation of AAL values for ^{90}Sr assume secular equilibrium with decay progeny (see Table 6B).

compared with the original gross gamma and final alpha beta screen by LSC screening results to ensure that major contributors to dose have not been overlooked.

Note that ^{129}I does not contribute to the gamma sum since it decays by electron capture and its 30 keV X-rays are below the threshold of the gross gamma ray analysis. Low-energy radiation does provide some response in the LSC analysis. Assuming that ^{129}I was not lost during oxidation of the sample, its activity may be added to the beta-gamma total. It is also assumed that ^{90}Y is present in secular equilibrium with ^{90}Sr although this may or may not be the case shortly after the incident.

When the values in Table 14 are compared to those in Table 13 for the screening results, the sums of radionuclide specific activities of (alpha + beta) or gamma are within a factor of two of their respective screening results. It is concluded that no major emitters have been overlooked.

The time is Day 9, 0245 hours

Step 10, Samples 1, 2, 4, and 5. Once all data is complete and reviewed, the sum of the fractions (i.e., the "unity rule") is calculated for all radionuclides in each sample. The sum of the fractions for all radionuclides in samples 2, 4, and 5 exceed unity it is concluded that the first year at 2,000 mrem AAL has been exceeded. In sample 1, the sum of the fractions calculation indicates that the AAL has not been exceeded.

No radionuclide-specific analysis has been done for sample 3. it will be analyzed for radionuclide specific activity at a later date once all higher priority samples have been analyzed.

The time is Day 9, 0300 hours

Step 11, Samples 2, 4, and 5. Any residual materials from the original sample, the dried or dissolved residues from the dissolution, and the sample test sources, are archived in case other analyses are necessary in the future. A concise report and case narrative with the analytical values and their associated uncertainties is sent to the IC.

The time is Day 9, 0315 hours

Step 12, Samples 3. At this time, sample screening results identifies sample 3 as below the 2,000-mrem AAL. It is stored and will be sent through the complete analytical scheme when analytical capacity permits.

APPENDIX III. EXAMPLE OF SOILS ANALYZED TO ASSESS THE EXTENT OF CONTAMINATION (RADIOANALYTICAL SCENARIO EXAMPLE 2: SOIL ANALYSIS IN THE RECOVERY PHASE FOLLOWING AN RDD)

Description[31]

During the recovery phase following the detonation of an RDD, the environs around the detonation site and beyond will be evaluated for radioactive contamination. For this scenario, the priority switches from high priority for high-activity samples (clearly contaminated) to high priority for low-activity samples (indicating zones that may be uncontaminated). Thus, samples are screened for gross gamma, alpha, and beta radioactivity based on first-year 10^{-4} risk-based action levels. Those samples having gross radioactivity concentrations below the 10^{-4} risk-based action levels have priority for specific radionuclide analyses. The radionuclide contaminants that initiated the incident have largely been characterized by now under Radioanalytical Scenario 1, and their results lead into the specific radioanalytical processes. However, it is possible that these samples may have other radionuclide contaminants, either related to the initial incident or from naturally-occurring sources, which will need to be characterized. Again, it is important to emphasize that the priority flow path for this scenario is set up in the reverse of Radioanalytical Scenario 1: the high-priority flow path is for those samples that have low activity concentration.

The micro-R meter used in the receipt screening of the samples has been calibrated with an ^{192}Ir source as this has been identified as the principal gamma-emitting radionuclide.

Event Sequence

On May 3 an RDD was detonated in a metropolitan area. Attribution sample analyses have confirmed that the device contained ^{90}Sr, ^{238}Pu, and ^{192}Ir as the major contaminants. It is Day 30 following the event and the recovery phase is ongoing. The Incident Command Center has dispatched three soil samples that need to be assessed for contamination levels acceptable for re-habitation.

The time frame for results is not as critical as in Radioanalytical Scenario 1 but prompt identification of “clean” areas (areas deemed by the IC to be re-habitable based on first year 10^{-4} risk) is important in rebuilding public confidence in the cleanup effort. The only radionuclides that have been identified above background in any of the contaminated samples in or outside the blast zone are ^{238}Pu, ^{192}Ir, and ^{90}Sr. *The three samples arrive at the laboratory at 0800 on Day 31.*

Analysis Paths

Step 1. Sample receipt involves screening using a micro-R meter. Laboratory personnel perform an initial screen of the transport container prior to opening it. The contact reading at the surface of the transport container is 140 µR/h. The sample transport container is moved to a laboratory hood so that the sample containers may be individually assessed for dose.

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[31] Radioanalytical Scenario Example 2 is unrelated to Radioanalytical Scenario Example 1.

The three samples are individually surveyed as they are removed from the transport container, using a micro-R meter, yielding the following results for total dose (Table 15).

Table 15 – Screening Results for Soil Samples Following an IND

Sample Container	Container 1	Container 2	Container 3	Instrument Background
μR/h	65 ± 5	25 ± 4	100 ± 6	15 ± 3

The sample containers are also assessed for external contamination using standard swipe techniques. None of the containers are found to be externally contaminated.

Step 2. Each container has approximately 500 g of soil. The soil contains twigs and stones greater than about ½" in diameter. These are removed with the aid of stainless steel forceps and a probe and are reserved as there has yet to be direction on how to address activity contained in the twigs or stones. There is considerable moisture and organic content to the soil.

Each sample is counted for 1 minute on a shielded 3×3" NaI(Tl) detector. The detector's lower level discriminator is set at 50 keV and all counts up to 2000 keV will be captured as gross gamma activity and referenced to ^{192}Ir. The results of the screening analysis for gross gamma are shown in Table 16.

Table 16 – Results of Gross Gamma Analysis of Soil Samples

Container ID	Total counts, NaI(Tl) detector
1	10,254
2	8,298
3	48,139
Background, **(**± 1σ**)**	40 ± 6

The NaI(Tl) detector has been calibrated in a geometry conforming to the full sample container and has an efficiency of 0.375 counts per disintegration when using a 500-g soil matrix containing ^{192}Ir.[32] The sample activities are estimated as shown here:

Sample 1: $$Gross\ Gamma, \frac{pCi}{g} = \frac{(10{,}254-40)}{(2.22 \times 0.375 \times 500\ g)} = 24.5\ \pm 6.1\ pCi/g$$

Sample 2: 19.8 ± 5.0 pCi/g
Sample 3: 116 ± 29 pCi/g

Although the lowest ADL in Table 7B is for ^{60}Co at 9.8 pCi/g, ^{60}Co has not been identified as a contaminant so it is not considered limiting. In this case the screening results are compared to the MQOs and ADL value for ^{192}Ir (the sole gamma emitter). The required method uncertainty is 56 pCi/g at the AAL of 180 pCi/g. The ADL is 92 pCi/g.

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[32] The only gamma-ray emitter expected in samples from this event is ^{192}Ir.

MQOs for required method uncertainty are met for all samples. The conclusion is made that ^{192}Ir is not present above the AAL in samples 1 and 2 since their activity is less than the ADL. Sample 3, however, shows activity above the screening ADL for ^{192}Ir.
Since those areas that are "clean" are the priority, this sample is archived to be analyzed if needed at a later date. Samples 1 and 2 will need to be analyzed for ^{192}Ir by HPGe gamma spectrometry in Step 6.

Step 3. Samples 1and 2. The samples are individually coned and quartered. Two 125-g fractions of bulk sample are isolated during coning and quartering. The fraction isolated for non-volatile analysis is transferred to a tared 1-pint paint can. A second portion is transferred to a container for "as-received" analyses. The unused half of the sample is archived.

Step 4a. Samples 1and 2. The steel can containing the 125-gram "non-volatile" fraction is placed in an oven at 110 ± 10 ˚C and the sample dried to `constant weight. Percent solids are calculated as the ratio of the dry-weight mass to the pre-drying "as-received" mass of sample. This will be applied to convert "as-received" aliquants to "dry-weight" aliquants in Steps 6 and 7b.

Stainless steel balls are added to each paint can containing the dried sample and the cans are shaken for about 5-10 minutes to mill and homogenize the samples to solids that conform to the visual size standard.

A representative aliquant of dried, milled, and homogenized sample is taken for gross alpha and beta screening prior to aliquanting samples for dissolution. About 70-120 mg of homogenized solid will be transferred to the textured surface of a "sticky" swipe applied to a stainless steel planchet using the adhesive backing of the swipe (i.e., cloth-side facing up). The swipe plus planchet are tared. The same paint can used for drying, pulverizing and storing the sample has an opening marginally smaller than 2" in diameter. The 2" diameter planchet (containing the swipe) fits neatly into the groove for the lid sealing the opening of the can. By shaking the can containing the pulverized solids and tapping the planchet to remove loose solids from the surface of the swipe, a small amount of solid is quickly transferred to the swipe with minimal use of equipment and relatively low risk danger of cross-contamination.

Before they are removed from the hood, the sample test sources are checked for elevated levels of radioactivity using a handheld survey meter. The planchet, swipe, and solid are weighed to determine the net amount of sample on the planchet for activity calculations. The solids are then fixed to the planchet by applying a very light layer of hair spray. The planchet is dried for a minute under a heat lamp and reweighed to determine the mass to use for self-absorption corrections.

Step 4b. Samples 1and 2. The planchet is counted on a gas flow proportional counter dedicated to screening samples that have low to slightly elevated levels of radioactivity. The results of the alpha/beta screen are calculated applying corrections for crosstalk, efficiency and the dry-weight aliquant. The screen results with associated combined standard uncertainty are reviewed and are presented in Table 17.

Table 17 – Results of Gross Alpha and Beta Analysis of Soil Samples

Container ID	Gross Alpha pCi/g, dry weight	Gross Beta pCi/g, dry weight
Sample 1	20.7 ± 6.1	14.1 ± 7.4
Sample 2	16.4 ± 5.8	16.1 ± 7.9

Since there is only one pure alpha and one pure beta emitter, and neither of them is volatile, the results of the alpha and beta screen can be used to determine whether the activity of ^{238}Pu and ^{90}Sr is below the respective AALs and can potentially be used to eliminate the need for radionuclide-specific testing. Table 7a shows that the u_{MR} for screening for ^{238}Pu is 18 pCi/g at the AAL of 60 pCi/g, with an ADL of 30 pCi/g. Similarly for gross beta screening for ^{90}Sr, u_{MR} is 110 pCi/g at the AAL of 380 pCi/g, and the ADL is 190 pCi /g. None of these results exceed those values and samples 1and 2 are all kept on the hot path for the alpha- and beta-emitting radionuclide-specific analyses.

Step 4c. Samples 1and 2. One-gram aliquants of milled homogenate are transferred to nickel crucibles for plutonium and radiostrontium analysis. Quality controls are set up for each batch, and tracers and carriers added to sample aliquants as required by each of the procedures being run. The crucibles are transferred to a furnace where a ramped program is used to incrementally raise the temperature to ~600 °C to remove all organic materials. The plutonium and strontium samples are dissolved using a sodium carbonate fusion technique. The melt is redissolved in hydrochloric acid and visually inspected to verify that complete dissolution of the solids was obtained.

Step 5a. Sample 1and 2. Although no volatile constituents were identified in the attribution analysis, the volatile fraction will be used to prepare the sample for gamma spectral analysis. It is thoroughly mixed and a representative 75-mL volume of the sample is transferred to a tared counting container for which there is a calibrated geometry on the gamma spectrometer. The filled container is reweighed and the “as received” aliquant mass calculated. Since all results must be reported on a dry-weight basis in Steps 6 and 7b, the “as-received” mass will be converted to “dry-weight” equivalent by applying the percent solids determined in Step 4a.

Step 6, Sample 1and 2 The sample prepared for gamma analysis in Step 5a is counted above the endcap of the HPGe detector for approximately 30 minutes to meet the first year 10^{-4} risk-based MQO for required method uncertainty for ^{192}Ir of 56 pCi/g at the AAL of 180 pCi/g. The detector used is calibrated for the same 75-mL geometry in which the samples are counted. As the samples count, it is noted that no other gamma rays except for those from ^{192}Ir, and low-levels of natural radionuclides in the background soil, are present.

Step 7a, Samples 1and 2. After completion of the dissolution techniques, the dissolved samples are processed for ^{238}Pu using the laboratory's validated procedures.

Step 7b, Samples 1and 2. After completion of the dissolution techniques, the dissolved samples are processed for total radiostrontium using the laboratory's validated procedures.

Step 8, Samples 1and 2. Each set of results is reviewed to ensure that the correct process was followed, that all manual data entry is accurate, that all QC criteria are met, required method uncertainties have been met, results are appropriately qualified, and that all anomalous situations or deviations from standard operating procedures are narrated. The analytical results along with the associated combined standard uncertainty for the samples are shown in Table 18 in units of pCi/g, dry weight. These results are approved by the laboratory supervisor.

Table 18 – Results of Soil Analysis Following an RDD

Sample	^{192}Ir	^{238}Pu	^{90}Sr	Σγ	Σα	Σβ†	Sum of Fractions
1 (Radiochemical)	39 ± 4.29	0.6 ± 1.0[&]	6.2 ± 2.8	39	0.6	51.3	0.25
1 (Screen)	24	20	14	—	—	—	—
2 (Radiochemical)	32 ± 3.52	3.7± 1.2	8.1 ± 3.3	32	3.7	48.2	0.26
2 (Screen)	20	16	16	—	—	—	—
3 (Radiochemical)	Not Analyzed						
3 (Screen)	[116 ± 29][%]	—	—	—	—	—	—
First year 10^{-4} ADL	130	43	270				
First year 10^{-4} AAL	180	60	380				

[%] Gross gamma results

[&] Result less than critical level - not considered when comparing screen and radionuclide-specific results

†The gross beta results include activity from ^{90}Y assumed to be in secular equilibrium with the ^{90}Sr

Step 9, Samples 1and 2. Individual results are compared to the respective ADLs as they are completed. All MQOs are met and no results exceed 10^{-4} risk ADLs.

Step 10, Samples 1and 2. The Σ columns are the simple sums of the individual α, β, or γ activities. These are compared with the original screening values by gross gamma and final LSC screen. Gross alpha is greater than the ^{238}Pu results for samples 1 and 2. The alpha and beta data from Samples 1 and 2 are reviewed again (Step 8) to check for errors and no issues are identified. The gamma ray results for sampes 1 and 2 compared well with the screening results. The discrepancy between the screen and radionuclide-specific results is consistent with typical levels of naturally-occurring radioactive materials in soils, however, so a notation is made in the case narrative and the data are reported without further qualification.

Step 11, Samples 1 and 2. The sum of the fractions is calculated for samples 1 and 2. The gross gamma results that demonstrated in Step 2 that ^{192}Ir was not present in samples above the AAL are substituted for the radionuclide-specific results when calculating the SOF since ^{192}Ir could be present at those levels in samples. If the SOF had exceeded unity for either of these two samples, however, a determination would have to be made if samples should be sent back for radionuclide specific analysis (in the case of gamma, Step 6) to determine the actual concentration of ^{192}Ir present.

Because the SOF does not indicate that the AAL have been exceeded, the results for these two samples are reported to incident command.

Step 13, Samples 1, 2, 3. All sample residuals, digests and sample test sources are archived for potential future analysis.

APPENDIX IV. EXAMPLE OF SOILS ANALYZED FOLLOWING AN IND INCIDENT (RADIOANALYTICAL SCENARIO EXAMPLE 3)

Description[33]

An IND detonation spreads contamination in the form of fission products with a minor contribution of activation products and TRU elements. Some initial fissile material that did not undergo fission is likely present as particulate matter and it is anticipated that this material will be in the form of DRPs. The initial samples from the blast zone are sent for attribution analysis.

Fission products are negatron and gamma-ray emitters. Screening using a micro-R meter will provide a good measure of the degree of contamination from the fission products but probably not for any of the TRU elements. Thus, gamma screening will be used as the primary tool for determining contamination controls. The determination of gross alpha is left until later in the analysis sequence.

Additionally there are many fission, activation, and TRU radionuclides that result from such an event that have half-lives measured in hours to a few days. This means that the screening results will change rapidly as a function of time. Several notable parent-progeny relationships exist for transient or secular equilibrium, which will create some difficult calculation issues. The analytical results for these radionuclides will require review by someone knowledgeable in these relationships and the capabilities of the detection systems used. These pairs are found in Tables 10A and 10B along with their AAL values.

Event Sequence

An IND was detonated four days ago in an urban setting on the Atlantic Seacoast. Immediately following the event a nor'easter hit the area of the blast zone spreading the contamination over a wider area up and down the Eastern Seaboard as well as inland for about 150 miles.

The Incident Command Center has dispatched five 200-g soil samples that need to be analyzed so that isopleths of contamination can be established to help facilitate recovery efforts. The time frame for results is critical as rehabitation of large suburban areas need to be made. The focus is to identify the most contaminated samples. First year 1,000 mrem PAG AAL, ADL and u_{MR} values apply.

The samples arrive at the laboratory at 0800 on Day 4.

Analysis Paths

Step 1. Five samples (all placed in 200-mL plastic containers filled to the top, compacted with hand pressure and the lids sealed on top with duct tape) have arrived, all of which have been determined by the field sampling teams to be significantly above background radiation levels.

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[33] Radioanalytical Scenario Example 3 is unrelated to Radioanalytical Scenario Examples 1 and 2.

The laboratory staff has a specific-sample geometry for their micro-R meter so that each individual sample can be scanned for gross gamma contamination as it arrives using a 10-second pass through a shielded chamber. This is sufficient to meet the MQO for required method uncertainty for ^{60}Co. Although ^{60}Co is not anticipated to be a major contributor, its ADL value of 6.5×10^3 pCi/g is conservatively selected to identify samples that will need additional, prompt radiochemical analysis. The conversion factor that is being used for the dose reading is based on a 200 g soil sample with a ^{60}Co standard homogeneously distributed within it. The factor is 175 pCi/ (µR/h).

Table 19 identifies the samples received, their measured dose rates and estimates of the equivalent gamma activity referenced to ^{60}Co.

Table 19 – Initial Gamma Dose Rate Survey in Soil Following an IND

Sample ID	A680	A685	A702	A719	A736	Background
µR/h	60 ± 5	460 ± 35	395 ± 35	746 ± 45	250 ±15	34 ± 3
pCi/g	4,550	74,550	63,300	124,600	37,800	—

Step 2. All samples have screening results that are greater than 6.3×10^3 pCi/g except for A680. That sample is archived for analysis at a future time. The remaining samples are put on the fast path for immediate analysis.

The IC is notified that samples A685, A702, A719, and A736 will be analyzed first as their initial gamma screening results place them in the high activity concentration category, potentially challenging the AALs. Sample A680 is to be archived until analysis of the first samples is underway.

Step 3. Each container has approximately 200 g of soil. The soil contains twigs and stones greater than about ½" in diameter. The samples are processed individually with the aid of a stainless steel forceps and probe to remove any detritus. The detritus is set aside to be analyzed separately if required by the analytical protocol specified in the project plan.

Care should be used (both from the personal exposure and analytical cross-contamination standpoint) when isolating non-soil like materials as it is possible that these materials, like the soil, may contain hot particles (DRPs) of TRU oxide from the original IND device.

The soil samples have considerable moisture and organic content. They are individually coned and quartered.

Step 4. The samples from each of the containers are dried at about 110 °C. After constant weight has been achieved (and the dry-weight basis determined), the dried samples are put into a pint paint can with some stainless steel balls and shaken for about 5 minutes to mill and homogenize the samples.

Aliquants are removed from this homogenate and spiked with Sr carrier for radionuclide-specific analysis. Three times the normal Sr carrier is added since the sample will be split in three following dissolution. The samples are transferred to a furnace where the temperature is raised using a program to ~600 °C to remove all organic materials. Aliquants for each sample are taken.

Step 5. The aliquants from each of the samples that was dried and oxidized in Step 4 is placed into nickel crucibles, and each is dissolved using a validated sodium carbonate fusion technique. The samples are dissolved in an HCl solution following the cool down of the melt from the fusion. Separate aliquants are taken for analysis in Steps 8a and 9.

Step 6. The "as-received" sample is mixed well and a 75-mL aliquant of each sample is transferred to a counting container for which there is a calibrated gamma geometry. Gamma analysis proceeds at Step 7.

Step 7. Each sample is counted for 15 minutes 7.5 cm from the endcap of an HPGe gamma spectrometer to meet MQOs for required method uncertainty for ^{60}Co. The analysis will determine all gamma emitters, volatile and non-volatile. The gamma spectrometric results are tabulated in Table 20.

Table 20 – Results for Gamma Spectrometry in Soil Samples Following an IND

	A685	**A702**	**A719**	**A736**
Radionuclide [1]	**pCi/g**	**pCi/g**	**pCi/g**	**pCi/g**
^{95}Nb	62 ± 7	72 ± 7	110 ± 12	50 ± 4
^{95}Zr	1.2×10^{3} ± 65	1.43×10^{3} ± 70	2.2×10^{3} ± 100	937 ± 43
^{99}Mo	1.2×10^{4} ± 400	1.42×10^{4} ± 420	2.2×10^{4} ± 750	8.20×10^{3} ± 260
^{99m}Tc	1.0×10^{4} ± 500	1.20×10^{4} ± 580	1.8×10^{4} ± 900	7.78×10^{3} ± 390
^{131}I	2.3×10^{3} ± 240	3.4×10^{3} ± 300	5.3×10^{3} ±390	2.25×10^{3} ± 200
^{132}I	730 ± 40	840 ± 50	1.3×10^{3} ± 80	479 ± 35
^{133}I	630 ± 29	730 ± 37	1.2×10^{3} ± 65	488 ± 25
^{137}Cs	2.3×10^{4} ± 1100	1.4×10^{4} ± 800	2.2×10^{4} ± 1000	8.00 ± 490
^{140}Ba	1.5×10^{4} ± 800	1.9×10^{4} ± 900	2.8×10^{4} ± 1200	1.17×10^{4}±570
^{140}La	1.3×10^{4} ± 710	1.5×10^{4} ± 790	2.3×10^{4} ± 1100	1.06×10^{4} ± 500
^{239}Np	1.7×10^{3} ± 120	550 ± 48	2.9×10^{4} ± 1200	293 ± 32

Step 8a. Gross alpha will be measured to determine if plutonium, americium or uranium analysis are necessary. Since gross alpha measurements on gas proportional counters operating in *simultaneous* counting mode, or on liquid scintillation counters operating in pulse-shape or pulse decay counting mode, will be overwhelmed by high levels of beta into alpha crosstalk. Therefore, the gross alpha samples will be counted on a gas proportional counter operating at the alpha voltage.

An aliquant of the dissolved sample from Step 5 is treated with concentrated nitric acid to remove chlorides, evaporated to dryness, and quantitatively transferred to a planchet. The samples are counted for 30 minutes each to meet MQOs for required method uncertainty for ^{239}Pu of 410 pCi/g at the AAL of 1,300 pCi/g. The gross alpha activities for each of the samples are shown in Table 21.

Table 21 – Results for Gross Alpha in Soil Following an IND

Sample ID	Gross Alpha Activity ± CSU pCi/g
A685	720 ± 130
A701	418 ± 78
A719	265 ± 42
A736	251 ± 41

Step 8b. Gross alpha screening results for Step 8a are assessed to see if they exceed the ^{239}Pu AAL of 1,300 pCi/g. All results satisfy the MQO for required method uncertainty at the AAL. Only sample A685 exceeds the gross alpha screening ADL of 670 pCi/g. An aliquant of this sample is taken for sequential alpha spectrometric analysis in Step 8c.

Step 8c. The initial attribution analyses for the event indicate that this was a ^{239}Pu device. The elevated gross alpha activity in sample A685 indicates that Pu, and ^{241}Am analyses need to be performed. Plutonium and americium analyses are begun shortly after strontium analysis is commenced using an aliquant of the fused sample.

The size of the aliquant is reduced to minimize the likelihood that the tracer will be overwhelmed by analyte, and to keep the levels of activity introduced into alpha spectrometers as low as possible. The plutonium analysis is run with ^{236}Pu tracer which is not easily overwhelmed by high levels of $^{239+240}$Pu. These radionuclide-specific flow paths use rapid methods that produce analytical results within 24 hours. The results of these analyses are shown in Table 22. Note that the U results showed a negligibly small contribution to both the total activity and the SOF/dose and are not included here.

Table 22 – Results for Isotopic Am and Pu in Soil Following an IND

	A685	**A702**	**A719**	**A736**
Radionuclide[1]	**pCi/g**	**pCi/g**	**pCi/g**	**pCi/g**
$^{239+240}$Pu [34]	480 ± 41	430 ±12	280 ±10	332 ±11
^{238}Pu	32 ± 4	29 ± 4	14 ± 2	17 ± 2
^{241}Am	75 ± 4	190 ±9	85 ±4	110 ±9

Step 9. The only significant beta-only emitters that need analysis are ^{89}Sr and ^{90}Sr. An aliquant of the dissolved melt from Step 5 is used to start analysis for both these radionuclides. This step is done in parallel with Steps 7 and 8a. The ^{89}Sr and ^{90}Sr activity concentration are shown in Table 23.

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[34] The results of plutonium analysis using alpha spectrometry can only provide a result which is the sum of the two activities for ^{239}Pu and ^{240}Pu. This is because their alpha peaks are too close together to be resolved. By convention the sum of the two activity contributors will be compared to the ADL and AAL for ^{239}Pu.

Table 23 – Results for ^{90}Sr and ^{89}Sr in Soil Following an IND

	A685	A702	A719	A736
Radionuclide [1]	**pCi/g**	**pCi/g**	**pCi/g**	**pCi/g**
^{89}Sr	410 ± 35	740 ± 56	1070 ± 92	499 ± 38
^{90}Sr	2.6 ± 0.4	5.3 ± 0.7	6.7 ± 0.8	3.3 ± 0.5

Step 12. Each set of results is reviewed to ensure that the correct process was followed, that all manual data entry is accurate, that all QC criteria are met, required method uncertainties have been met, results are appropriately qualified, and that all anomalous situations or deviations from standard operating procedures are narrated. The analytical results along with the associated combined standard uncertainty for the samples are shown in Table 24 in units of pCi/g, dry weight. These results are approved by the laboratory supervisor.

Table 24 – Summary and Evaluation of Results for Soil Samples Following an IND

	A685		A702		A719			
Radionuclide	pCi/g	Fraction of AAL	pCi/g	Fraction of AAL	pCi/g	Fraction of AAL	pCi/g	Fraction of AAL
^{89}Sr	410 ±35	9.3×10^{-4}	740 ± 56	1.7×10^{-3}	1070 ± 92	2.4×10^{-3}	499 ± 38	1.1×10^{-3}
^{90}Sr	2.6 ±0.4	1.2×10^{-5}	5.3 ± 0.7	2.5×10^{-5}	6.7 ± 0.8	3.2×10^{-5}	3.3 ± 0.5	1.6×10^{-5}
^{95}Nb	62 ±7	—	72 ± 7	—	110 ± 12	—	50 ± 5	—
^{95}Zr	1.2×10^{3} ±65	0.057	1.43×10^{3} ±70	0.068	2.2×10^{3} ±100	0.10	937 ±43	0.045
^{99}Mo	1.2×10^{4} ±400	0.071	1.42×10^{4}±420	0.084	2.2×10^{4} ±750	0.13	8.20×10^{3}±260	0.048
^{99m}Tc	1.0×10^{4} ±500	—	1.20×10^{4}±580	—	1.8×10^{4} ± 900	—	7.78×10^{3}±390	—
^{131}I	2.3×10^{3} ±240	0.024	3.4×10^{3} ±300	0.035	5.3×10^{3} ±390	0.055	2.3×10^{3} ±200	0.023
^{132}I	730 ±40	1.8×10^{-3}	840 ±50	2.1×10^{-3}	1.3×10^{3} ±80	3.2×10^{-3}	479 ±35	1.2×10^{-3}
^{133}I	630 ±29	3.9×10^{-3}	730 ±37	4.6×10^{-3}	1.2×10^{3} ±65	7.0×10^{-3}	488 ±25	3.0×10^{-3}
^{137}Cs	2.3×10^{4}±1100	0.43	1.4×10^{4} ±800	0.26	2.2×10^{4}±1000	0.41	8.0×10^{3} ±490	0.15
^{140}Ba	1.5×10^{4} ±800	1.07	1.9×10^{4} ±900	1.4	2.8×10^{4}±1200	2.0	1.17×10^{4}±570	0.84
^{140}La	1.3×10^{4} ±710	—	1.5×10^{4} ±790	—	2.3×10^{4}±1100	—	1.05×10^{4}±500	—
^{239}Np	1.7×10^{3} ±120	5.5×10^{-3}	550 ±48	1.8×10^{-3}	2.9×10^{4}±1200	0.094	293 ±32	9.4×10^{-4}
^{238}Pu	32 ± 4	0.021	29 ± 4	0.019	14 ± 2	0.093	17 ± 2	0.011
$^{239+240}$Pu	480 ±41	0.37	430 ±12	0.33	280 ±10	0.22	332 ±11	0.25
^{241}Am	75 ±4	0.044	190 ±9	0.11	85 ±4	0.050	110 ±9	0.060
SOF Total		2.1		2.3		3.2		1.4
Gamma total (pCi/g)	8.1×10^{4}		8.3×10^{4}		1.5×10^{5}		5.2×10^{4}	
Dose rate survey(pCi/g)	7.5×10^{4}		6.3×10^{4}		1.2×10^{5}		3.8×10^{4}	
Sum of isotopic alpha (pCi/g)	587		649		379		459	
Gross Alpha (pCi/g)	720 ± 130		418 ± 78		265 ± 42		251 ± 41	

Steps 13 and 14. The sum of the individual results compares favorably with the screening results and no additional action is required for these samples.

Step 15. The results from the analyses completed in Steps 7, 8b, 8c, and 9 are compared to their respective ADL values in Tables 6C, 6D, and 10B.

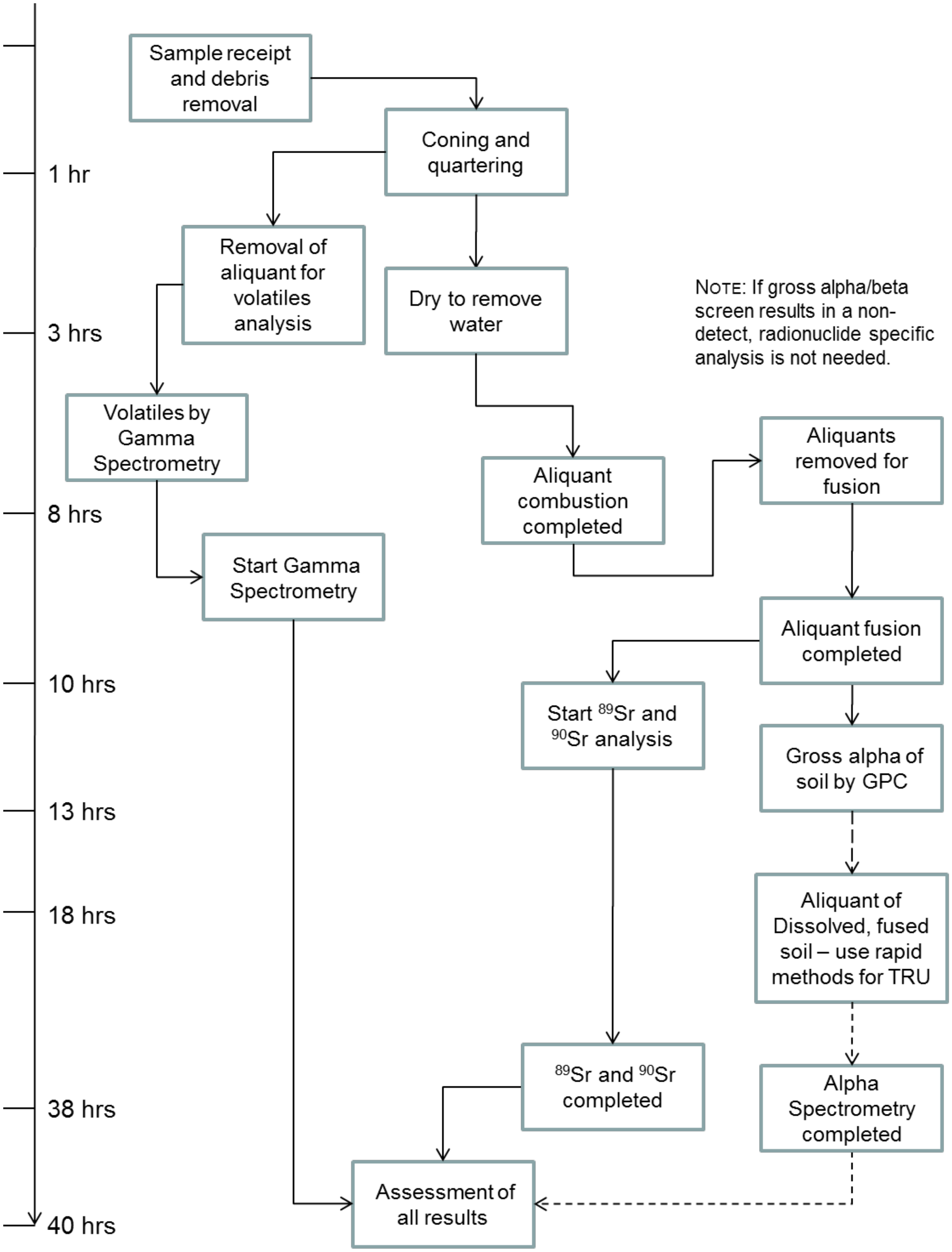
The comparison shows that that the measured result for ^{140}Ba exceeded the ADL for three of the four fast track samples. This means that the 1,000-mrem AAL concentration for that radionuclide has been exceeded at the tolerable error selected for the MQO. However, the AAL concentration assumes that the ^{140}Ba is in equilibrium with its progeny ^{140}La. Based on the results of the laboratory analysis, it is apparent that equilibrium has not yet been achieved. Equilibrium, when achieved, will yield an activity ratio of ^{140}La/^{140}Ba of approximately 1.2. This means that the activity of the combination will increase over the next several days until maximum is achieved. Thus the conclusion is conservative for the time of analysis. This fact is noted in the case narrative to the IC.

The calculation of the fractional AAL for each radionuclide, as well as the sum of fractions (SOF) for all radionuclides, are shown in Table 24. The SOF for all four of the samples exceeds unity. Note even if the ^{140}La/^{140}Ba pair were not part of the analytical results (i.e., not detected above the critical level) that the sum of fractions for the remainder of the radionuclides would yield values greater than one for two of the four samples even though individually each radionuclide would be less than its AAL value.

Step 16. The results in Table 24 are transmitted to the IC indicting that these four samples exceed the AAL values

Step 17. The samples are archived, and the sample that was found to have screening values less than the ADL is now analyzed. The information from the high activity concentration samples are used to assess both sample size and count times for these lower activity concentration samples.

APPENDIX V. REPRESENTATIVE ANALYTICAL PROCESSING TIMES



NOTE: If gross alpha/beta screen results in a non-detect, radionuclide specific analysis is not needed.

Figure 7 – Analytical Processing Timeline for Soil Sample Following an IND

APPENDIX VI. ESTABLISHING DQOS AND MQOS FOR INCIDENT RESPONSE ANALYSIS

Three distinct radioanalytical scenarios are presented for soil potentially contaminated with radionuclides. The first two assume that the mixture of radionuclides in the sample is unknown. In the third situation, the result of an IND, the radioactive contaminants are known (fission products). In each scenario there is special emphasis on the implementation of the decision trees presented within that scenario for prioritizing sample processing by the laboratory. This emphasis on the decision trees is to support timely decisionmaking by the IC regarding actions to protect human health and to expedite analysis so that decisions made to support the data quality objectives can be made with confidence. Specific MQOs associated with the flow diagrams in Figures 2, 3, and 4 are given in Tables 23, 24, and 25.

This appendix covers single-sample screening measurement decisions by the laboratory. The IC may need to make decisions based on the final radionuclide-specific concentrations based on the mean of the set of samples taken from an area. MQOs would need to be developed separately for this case. The required method uncertainty (u_{MR}) should be smaller in this case compared to the laboratory's screening decisions, perhaps by a factor of three (see MARLAP Appendix C).

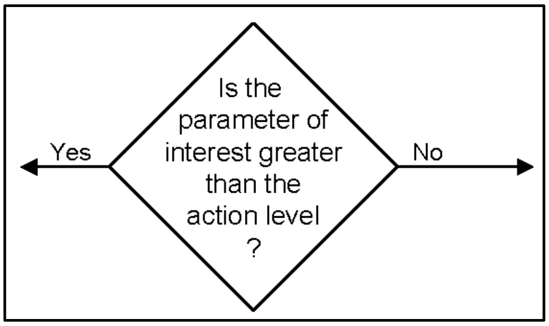


Figure 8 – Decision Symbol Used in Flow Diagrams

The flowcharts depicted in this document contain decision points. There are three basic symbols on these flowcharts: rectangles, which represent activities or tasks; decision point diamonds, which represent decision points; and arrows, which represent flow of control. In these flow diagrams, there are many diamond-shaped decision points. Most often they are of the form shown in Figure 8. This is the general form of a theoretical decision rule as discussed in Step 5 of the DQO process. The parameter of interest usually is the "measurand" of the radiochemical analysis being performed (e.g., concentration of a radionuclide, total activity concentration, etc.). The AALs will have been set according to criteria involving the appropriate PAGs. The arrows specify the alternative actions to be taken.

The DQO process may be applied to all programs involving the collection of environmental data with objectives that cover decisionmaking activities. When the goal of the study is to support decisionmaking, the DQO process applies systematic planning and statistical hypothesis testing methodology to decide between alternatives. Data quality objectives can be developed using EPA (2006) *Guidance on Systematic Planning Using the Data Quality Objectives Process (EPA QA/G-4).*

The DQO process is summarized in Figure 9 and Table 19. From this, MQOs can be established using the guidance in MARLAP. The information in Table 19 should be sufficient to enable the decisionmaker and laboratory to determine the appropriate MQOs. The output should include an AAL, discrimination limit, gray region, null hypothesis, analytical decision level (ADL, referred to in MARLAP as "critical level"), and required method uncertainty at the AAL. A table summarizing DQO process for each decision point diamond can be prepared in advance and summarized as shown in Tables 20 and 21.

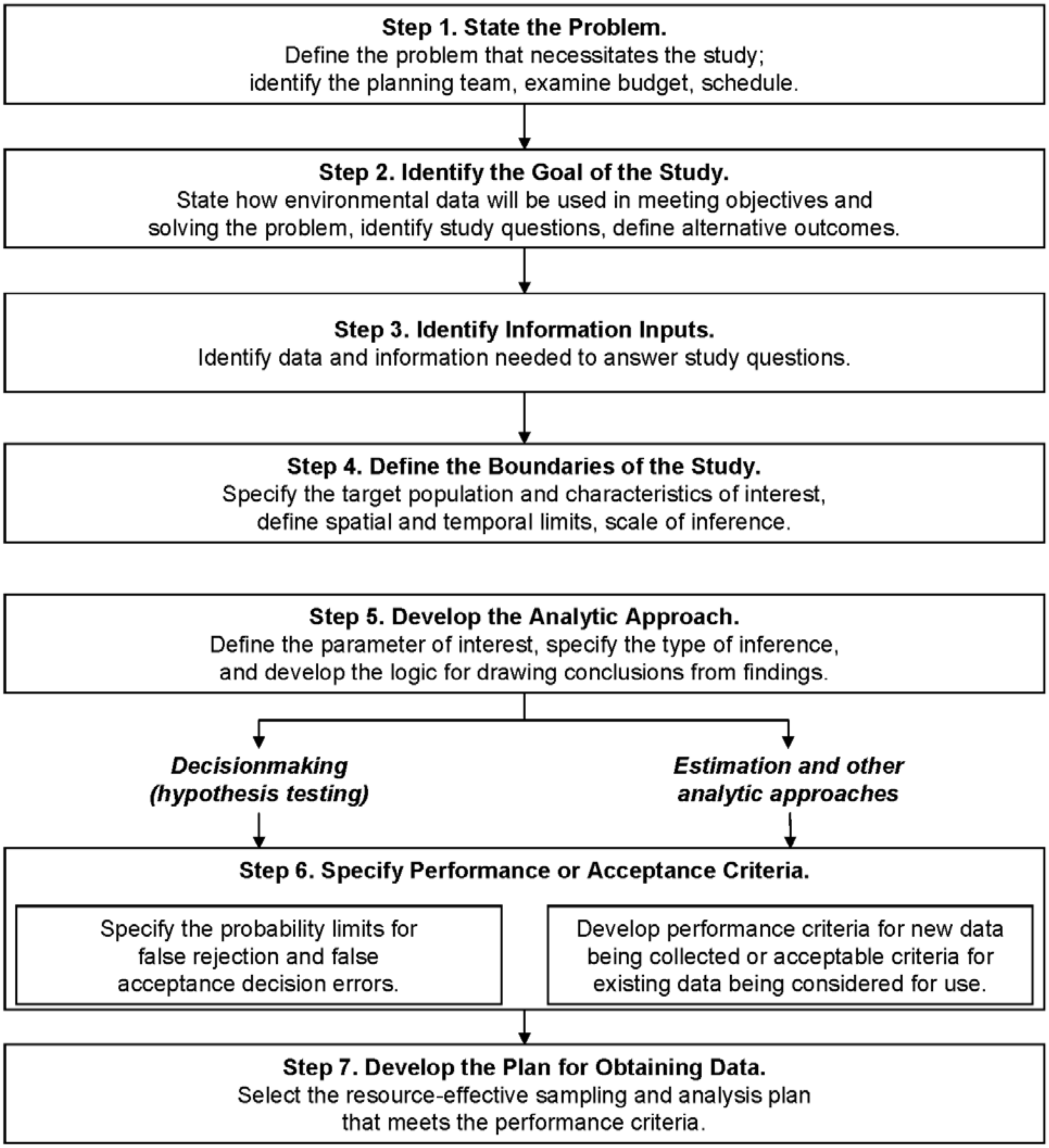


Figure redrawn from EPA G-4 (2006).

Figure 9 – The Data Quality Objective Process

Table 25 – The DQO process Applied to a Decision Point

STEP	OUTPUT
Step 1. Define the problem	… with a preliminary determination of the type of data needed and how it will be used; identify decisionmaker.
Step 2. Identify the decision	…among alternative outcomes or actions, and a list of decision statements that address the problem.
Step 3. Identify information needed for the decision	Analytical action levels that will resolve the decision and potential sources for these; information on the number of variables that will need to be collected; the type of information needed to meet performance or acceptance criteria; information on the performance of appropriate sampling and analysis methods.
Step 4. Define the boundaries of the study	Definition of the target population with detailed descriptions of geographic limits (spatial boundaries); detailed descriptions of what constitutes a sampling unit timeframe appropriate for collecting data and making the decision or estimate, together with any practical constraints that may interfere with data collection; and the appropriate scale for decisionmaking or estimation.
Step 5. Develop a decision rule. ***This defines the decision point diamond.***	Identification of the population parameters most relevant for making inferences and conclusions on the target population; for decision problems, the "if..., then...else..." theoretical decision rule based upon a chosen AAL.

The theoretical decision rule specified in Step 5 can be transformed into statistical hypothesis tests that are applied to the data. Due to the inherent uncertainty with measurement data, there is some likelihood that the outcome of statistical hypothesis tests will lead to an erroneous conclusion, i.e., a decision error. This is illustrated in Table 20.

Table 26 – Possible Decision Errors

	True Value of the parameter of interest	
Decision Made	**Greater than the action level**	**Less than the action level**
Decide that the parameter of interest is greater than the action level.	Correct decision	*Type I Decision Error*
Decide that the parameter of interest is less than the action level.	*Type II Decision Error*	Correct decision

In order to choose an appropriate null hypothesis (or baseline condition), consider which decision error should be more protected against. Choose the null hypothesis that, if falsely rejected, would cause the greatest harm. Then the data will need to be convincingly inconsistent with the null hypothesis before it will be rejected, and the probability of this happening (a Type I error) is more easily controlled during the statistical design. Failing to detect a sample that exceeds the AAL could have consequences to public health. But screening additional samples will slow the overall process and therefore also may impact the public health. The probability that such decision errors occur is defined as the parameters α and β in Steps 6.1 and 6.2 in Table 21. Values of alpha and beta should be set based on the consequences of making an incorrect decision. How these are balanced will depend on the AAL, sample loads, and other factors as specified by the IC. The most commonly used values of alpha and beta are 5%, although this is by tradition and has no sound technical basis. These values may be used as a default, but should be optimized in Step 7 of the DQO process according to the actual risk of the decision error being considered.

Table 27 –The DQO Process Applied to a Decision Point

STEP	OUTPUT
Step 6. Specify limits on decision errors	

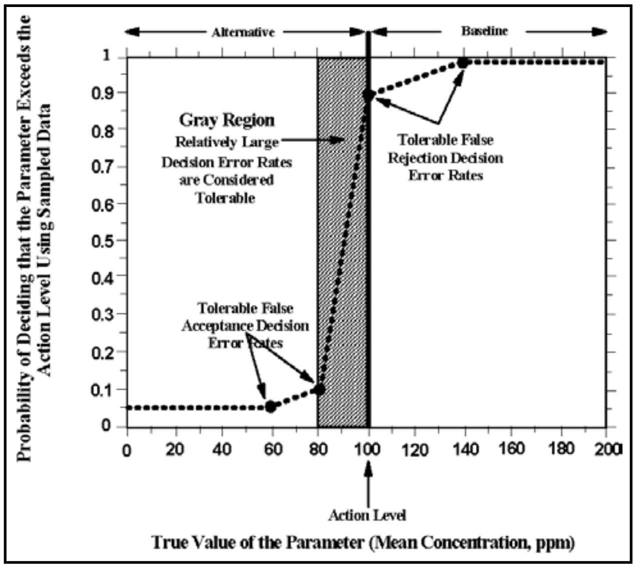
Step 6.1 Determine analytical action level (AAL) on the gray region boundary and set baseline condition (null hypothesis, H_0)	Which is considered the worse: decision error (a) deciding that the parameter of interest is less than the AAL when it actually is greater, or (b) deciding that the parameter of interest is greater than the AAL when it actually is less? Case (a) is usually considered to be a conservative choice by regulatory authorities, but this may not be appropriate in every case.
	If (a), the AAL defines the upper boundary of the gray region. The null hypothesis is that the sample concentration is above the AAL. (All samples will be assumed to be above the AAL unless the data are convincingly lower.) A desired limit will be set on the probability (α) of incorrectly deciding the sample is below the AAL when the sample concentration is actually equal to the AAL.
	If (b), the AAL defines the lower boundary of the gray region. The null hypothesis is that the sample concentration is below the AAL. (All samples will be assumed to be below the AAL unless the data are convincingly higher.) A desired limit will be set on the probability (β) of incorrectly deciding the sample is above the AAL when the sample concentration is actually equal to the AAL.

6.2 Define the discrimination limit (DL)	If (a), the discrimination limit defines the lower boundary of the gray region.[1] It will be a concentration below the AAL where the desired limit will be set on the probability (α) of incorrectly deciding the sample is above the AAL. If (b), the discrimination limit defines the upper boundary of the gray region.[2] It will be a concentration above the AAL where the desired limit will be set on the probability (β) of incorrectly deciding the sample is below the AAL.
6.3 Define the required method uncertainty at the AAL	According to MARLAP Appendix C, under either case (a) or case (b) above, the recommended required method uncertainty is: where $z_{1-\alpha}$ and $z_{1-\beta}$ are the $1-\alpha$ and $1-\beta$ quantiles of the standard normal distribution function. Values of $z_{1-\alpha}$ (or $z_{1-\beta}$) for some commonly used values of α (or β), taken from tables of the cumulative normal distribution (EPA 2009), are shown in Table 22.
Step 7. Optimize the design for obtaining data	Iterate Steps 1–6 to define optimal values for each of the parameters and the measurement method required.

Notes:

[1] The discrimination limit (DL) is the point where it is important to be able to distinguish expected signal from the AAL. When one expects background activity, then it might be zero. If one expects activity near the AAL, however, it might be at 90% of the AAL.

[2] The DL is the point where it is important to be able to distinguish expected signal from the AAL. If the AAL is near zero, the DL would define a concentration deemed to be too high to be undetected. Thus, the DL may be set equal to the minimum detectable concentration (MDC). If one expects activity near the AAL, however, it might be at 110% of the AAL.



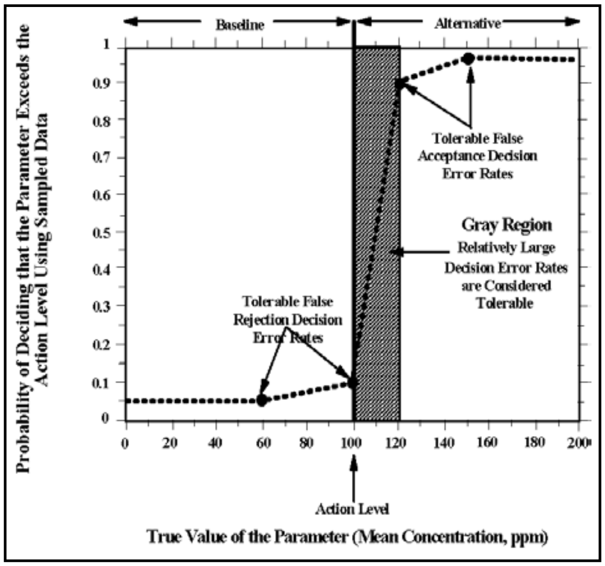


Figure 10 – Example Illustrating Case (a) Baseline Condition (null hypothesis): Parameter Exceeds the AAL

See Table 27, Step 6.1. Figures 10 and 11 taken from EPA QA/G-4 (2006).

Figures 10 and 11 illustrate the concepts above for case (a) and case (b) respectively. In Figure 10, the AAL = 100, the DL = 80, Δ = 100 – 80 = 20 $\alpha = \beta = 0.1$ and

$$u_{MR} \leq \frac{\Delta}{(z_{1-\alpha} + z_{1-\beta})} = \frac{20}{(1.282 + 1.282)} = 7.8$$

In Figure 11, the AAL = 100, the DL = 120, Δ = 120–100 = 20 $\alpha = \beta = 0.1$ and

$$u_{MR} \leq \frac{\Delta}{(z_{1-\alpha} + z_{1-\beta})} = \frac{20}{(1.282 + 1.282)} = 7.8$$

Table 28 – Values of $z_{1-\alpha}$ (or $z_{1-\beta}$) for Some Commonly Used Values of α or β

α or β	$z_{1-\alpha}$ (or $z_{1-\beta}$)
0.001	3.090
0.01	2.326
0.025	1.960
0.05	1.645
0.10	1.282
0.20	0.842
0.30	0.524
0.50	0.000

The concentration that indicates the division between values leading to rejecting the null hypothesis and those that do not is termed the “critical level.” Possible values of the concentration can be divided into two regions, the acceptance region and the rejection region. If the value of the concentration comes out to be in the acceptance region, the null hypothesis being tested is not rejected. If the concentration falls in the rejection region, the null hypothesis is rejected. The set of values of a statistic that will lead to the rejection of the null hypothesis tested is called the critical region. Critical region is a synonym for rejection region. In the context of analyte detection, the *critical value* (MARLAP 2004, Attachment 3B.2) is the minimum measured value (e.g., of the instrument signal or the *analyte* concentration) required to give confidence that a positive (nonzero) amount of *analyte* is present in the material being analyzed. The critical value is sometimes called the *critical level*.

In case (a), the critical value (or "Analytical Decision Level" - ADL) will be UBGR – $z_{1-\alpha}\ u_M$, where u_M is its combined standard uncertainty of the measurement result, x. Only measurement results less than the critical value will result in rejecting the null hypothesis that the true concentration is greater than the AAL. This process can be completed for each diamond in each flowchart to fill in Tables 29, 30, and 31. In these tables, values have been rounded to 2 significant figures.

In case (b), the critical value (ADL) will be LBGR + $z_{1-\alpha}\ u_M$, where u_M is its combined standard uncertainty of the measurement result, x. Only measurement results greater than the critical value will result in rejecting the null hypothesis that the true concentration is less than the AAL.

In the following tables, MQOs were determined for screening using a discrimination level of zero and Type I and Type II error rates of $\alpha = \beta = 0.05$. These are the MQOs usually associated with developing MDCs and result in a relative method uncertainty of 30% at the AAL, and an ADL value of 0.5 times the AAL. For radionuclide-specific measurements the requirements are more stringent, using a discrimination level of one-half the AAL and Type I and Type II error rates of $\alpha = 0.01$ with $\beta = 0.05$. These result in a relative required method uncertainty of 13% at the AAL and an ADL value of 0.71 times the AAL. Note that gamma spectrometric measurements using an HPGe are always radionuclide-specific, and therefore have the more stringent MQOs.

Table 29 – DQOs and MQOs for Radioanalytical Scenario 1. Laboratory Prioritization Decisions Based on Screening (Gross α, β, or γ) and Radionuclide-Specific Measurements

Measurement Rectangl0065x	Decision Point Diamond	Type of Analysis, α, β, or γ	Analytical AL (pCi/g)	Null Hypothesis H_0 Choose > AAL or <AAL i.e., case (a) or case (b)	DL < AAL in case (a) and DL > AAL in case (b)	Δ= UBGR-LBGR	Type I error rate *α*	Type II error rate *β*	u_{MR}	φ_{MR} (relative required method uncertainty)	Analytical Decision Level (Critical Level) (pCi/g)[1], [2]	Source of AAL
1a	2a	α	410	a	a	410	0.05	0.05	130	0.30	210	AAL for 2,000 mrem First Year Exposure)
1a	2a	γ	330	a	a	330	0.05	0.05	100	0.30	160	
1a	2a	β	3200	a	a	3200	0.05	0.05	970	0.30	1600	
1a	2b	α + β + γ	590	a	a	590	0.05	0.05	180	0.30	290	
-	7	α	See Table 6A	a	a	AAL	0.05	0.05	0.30×AAL	0.30	0.50×AAL	
-	7	β	See Table 6B	a	a	AAL	0.05	0.05	0.30×AAL	0.30	0.50×AAL	
-	9	γ	See Table 6C and 6D	a	a	0.5×AAL	0.01	0.05	0.13×AAL	0.13	0.71×AAL	

Notes:

[1] All calculated values are rounded to two significant figures.

[2] The value of 590 pCi/L is derived from, 160(gross gamma) +210 (gross alpha) +220 (lowest ADL for a beta only emitter ^{227}Ac). The ^{227}Ac is used since there is a potential for this being an IND type scenario.

Table 30 – DQOs and MQOs for Scenario 2. Laboratory Prioritization Decisions Based on Screening (Gross α, β, or γ and Radionuclide-Specific Measurements)

Measurement Rectangle	Decision Point Diamond	Type of Analysis, α, β, or γ	Analytical AL (pCi/g)	Null Hypothesis H_0 Choose > AAL or < AAL i.e. case (a) or case (b)	DL < AAL in case (a) and DL > AAL in case (b)	Δ= UBGR-LBGR	Type I error rate *α*	Type II error rate *β*	u_{MR}	φ_{MR}	Analytical Decision Level (Critical Level) (pCi/g) [1]	Source of AAL [1]
2	3	γ	20	a	a	20	0.05	0.05	5.9	0.30	10	
4a	4b	α and β	44	a	a	44	0.05	0.05	13.4	0.30	22	
7a	9	α	See Table 7C	a	a	0.5×AAL	0.01	0.05	0.13×AAL	0.13	0.71×AAL	
7b	9	β	See Table 7D	a	a	0.5×AAL	0.01	0.05	0.13×AAL	0.13	0.71×AAL	
7c	9	γ	See Table 7D	a	a	0.5×AAL	0.01	0.05	0.13×AAL	0.13	0.71×AAL	

Note:

[1] All calculated values are rounded to two significant figures.

Table 31 – DQOs and MQOs for Radioanalytical Scenario 3. Laboratory Prioritization Decisions Based on Screening (Gross α, β, or γ) and Radionuclide Specific Measurements

Measurement Rectangle	Decision Point Diamond	Type of Analysis, α, β, or γ	Analytical AL (pCi/g)	Null Hypothesis H_0 Choose > AAL or <AAL i.e., case (a) or case (b)	DL < AAL in case (a) and DL > AAL in case (b)	Δ= UBGR-LBGR	Type I error rate *α*	Type II error rate *β*	u_{MR}	φ_{MR}	Analytical Decision Level (Critical Level) (pCi/g) [1], [2]	Source of AAL
1	2	γ	13,000	a	a	13,000	0.05	0.05	3,900	0.30	6,500	AAL for 1,000 mrem (First Year Exposure)
6	8b	α	1,300	a	a	1,300	0.05	0.05	390	0.30	650	
8c	15	α	See Table 6C	a	a	0.5×AAL	0.01	0.05	0.13×AAL	0.13	0.71×AAL	
9	15	β	See Table 6D	a	a	0.5×AAL	0.01	0.05	0.13×AAL	0.13	0.71×AAL	
7	15	γ	See Table 10B	a	a	0.5×AAL	0.01	0.05	0.13×AAL	0.13	0.71×AAL	

Notes:

[1] All calculated values are rounded to two significant figures.

[2] The gamma screening ADL is derived from the AAL ^{60}Co.

[3] The alpha screening ADL value is derived from the AAL of ^{239}Pu. Pu-239 represents a radionuclide that would either be part of the IND or would be formed as a result of neutron capture on ^{238}U.

APPENDIX VII. NET COUNT RATE AND COUNTING TIMES FOR TYPICAL NUCLEAR INSTRUMENTATION USED FOR RAPID ANALYSES (EXAMPLE) – 10^{-4} AND 10^{-6} RISK

Table 32 – Net Count Rate and Counting Times for Typical Nuclear Instrumentation Used for Rapid Analyses (Example) – 10^{-4} and 10^{-6} Risk

Radionuclide	Sample Size, g	AAL 10^{-4} Risk, pCi/g	u_{MR}, pCi/g	Count Time, (min)	AAL 10^{-6} Risk, pCi/g	u_{MR}, pCi/g	Count Time, (min)
^{241}Am [1]	1	65	3.5	4	0.65	0.035	350
^{241}Am	2	65	3.5	2	0.65	0.035	180
^{243}Cm [1]	1	75	9.7	4	0.75	0.097	310
^{243}Cm	2	75	9.7	2	0.75	0.097	160
^{239}Pu [1]	1	55	7.1	4	0.55	0.071	430
^{239}Pu	2	55	7.1	2	0.55	0.071	220
^{226}Ra [1]	1	24	3.2	9	0.24	0.032	1000
^{226}Ra	2	24	3.2	5	0.24	0.032	500
^{238}U [1]	1	240	31	1	24	0.31	100
^{238}U	2	240	31	<1	24	0.31	50
^{144}Ce [3]	250	350	46	<1	3.5	0.46	4
^{60}Co [3]	250	20	2.5	<1	0.20	0.025	40
^{228}Ra [2]	1	47	6.1	3	0.47	0.061	very long
^{228}Ra	2	47	6.1	2	0.47	0.061	520
^{89}Sr [2]	1	290	38	<1	2.9	0.38	80
^{89}Sr	2	290	38	<1	2.9	0.38	30

Notes: A 7% relative standard uncertainty is assumed for the other standard uncertainties such as for a tracer or a gamma efficiency curve.

[1] Alpha spectrometry radionuclides assumptions: detector efficiency of 0.24; chemical yield of 0.70; abundance/branching ratio of 1.0; background count rate of 0.005 cpm; background counting time of 1,000 min.

[2] Beta only radionuclide assumptions: detector efficiency of 0.40; chemical yield of 0.70; abundance/branching ratio of 1; background count rate of 1.0; background counting time of 1,000 min.

[3] Gamma radionuclides assumptions: detector efficiency, ^{60}Co – 0.035 and ^{144}Ce – 0.12; branching ratio, ^{60}Co – 1.0 and ^{144}Ce – 0.108; background count rate, ^{60}Co – 1 cpm and ^{144}Ce – 5 cpm; background counting time of 100 min.