

ENVIRONMENTAL RADIOLOGICAL MONOTORING & SURVEILLANCE - VOL 2 OF 3

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NUC-119 EXAM PREVIEW

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Exam Preview:

- 1. The mobility of radionuclides and the likelihood of uptake into biota should also be considered. Tritium is the most mobile and is readily taken up by biota.
 - a. True
 - b. False
- 2. Using Table ABLE 6-1: Minimum Criteria for Determining Need for Environmental Surveillance, which of the following topics corresponds to the description: Use of data should be based on statistically significant differences between the point of measurement and background data?
 - a. Routine Surveillance of All Pathways
 - b. Periodic Confirmation
 - c. Pathway Measurements
 - d. Characterization of Background Unplanned Releases
- 3. When considering short-half-life radionuclides, ensure that the sampling and measurement intervals do not exceed three times the half-life of the radionuclide.
 - a. True
 - b. False
- 4. Which of the following radionuclides matches the following criteria: is mobile, readily taken up by mammals, and concentrates in the thyroid?
 - a. Cesium
 - b. Strontium
 - c. Iodine
 - d. Radium

- 5. When determining radionuclides of interest, staff should consider each category: transuranics, uranium, fission products, and activation products. Which of the following radionuclides corresponds to the description: Iodine isotopes are potentially important, though most have short half-lives; iodine-129 is naturally occurring and is very difficult to detect?
 - a. Uranium
 - b. Fission Products
 - c. Transuranics
 - d. Activation products
- 6. According to the reference material, the air sampling rate should not vary by more than ± ___ percent, and total air flow or total running time should be indicated or recorded; air sampling systems should be leak-tested, flow-calibrated, tested, and inspected on a routine basis.
 - a. 10
 - b. 15
 - c. 20
 - d. 25
- 7. In ICP-MS procedures, the sample, or a portion of the sample, digested in chemical reagents is transported by a noble gas (e.g., argon) through a high-electron density plasma. The sample is subsequently vaporized in the plasma region to separate its individual constituents.
 - a. True
 - b. False
- 8. Environmental samples selected for tritium analysis are typically counted for ______ or longer to enhance detection and to reduce measurement uncertainties due to the expected low environmental tritium concentrations.
 - a. 15 minutes
 - b. 30 minutes
 - c. 45 minutes
 - d. 1 hour
- 9. In terms of exposure potential, tritiated water vapor yields a dose equivalent approximately 10,000 times that of tritium gas for the same concentration.
 - a. True
 - b. False
- 10. Using TABLE 6-3: Global fallout activity relative to Pu-239, Pu-240; which of the following ratios corresponds to the activity of Cs-137?
 - a. 1.00
 - b. 0.42
 - c. 18
 - d. 28

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6 ENVIRONMENTAL SURVEILLANCE

The purpose of the environmental surveillance program is to characterize the radiological conditions of the DOE facility environs and, if appropriate, estimate public doses related to these conditions, and confirm predictions of public doses based on effluent monitoring data. Environmental surveillance data also may be useful in evaluating doses to the biota consistent with DOE O 458.1 and DOE-STD-1153-2002. The environmental surveillance program should be conducted in accordance with the requirements of DOE O 458.1 and other applicable regulations and DOE directives. Media routinely monitored in environmental surveillance include air, water, terrestrial foodstuffs, aquatic foodstuffs, soil and sediment.

The responsible DOE field organization needs to determine the scope of the environmental surveillance program by considering the following factors:

- Applicable regulations;
- Hazard potential of the effluents;
- Expected quantities and concentrations of effluents;
- Nature of potential or actual impacts on air, land, biota, and water;
- Extent to which facility operations are routine and unchanging;
- Need for supplementing and complementing effluent monitoring;
- Size and distribution of the exposed population;
- Cost effectiveness of modifications to environmental surveillance; and
- Availability of measurement techniques that provide sufficiently sensitive comparisons with the applicable standard and "ambient" measurements.

A lines of inquiry approach is provided to conduct self-assessments; to verify that the program is effective and in compliance with appropriate requirements; and to ensure the existence of continuous improvement of the program. Lines of inquiry are identified in Appendix B of this Handbook.

6.1 Key Requirements

DOE O 458.1, Radiation Protection of the Public and the Environment, requires that environmental monitoring conducted as part of demonstrating compliance with the Public Dose Limit include environmental surveillance.

6.2 Summary of General Criteria

The criteria listed in Table 6-1 can be used to establish environmental surveillance program elements for DOE sites. Any additional site-specific criteria should be documented in the site Environmental Monitoring Plan (EMP) or other supporting documentation.

An evaluation (e.g., critical pathway analysis) should be conducted and used as the basis for establishing environmental surveillance for DOE sites. The results of this evaluation should be documented in appropriate records to show the following:

- Environmental measurement and sampling locations used for determining ambient environmental levels resulting from facility operations;
- Procedures and equipment needed to perform the measurement and sampling;
- Frequency and analyses required for each measurement and sampling location;
- Minimum detection level and accuracy;
- QA components; and
- Investigation and alarm levels.

TABLE 6-1: Minimum Criteria for Determining Need for Environmental Surveillance

	<u>Criteria</u>				
Routine Surveillance of All	When feasible, all environmental media that, as determined by site-				
Pathways (Ingestion,	specific radiation exposure pathway analysis, might lead to a measurable				
Inhalation, and Immersion	annual dose of site origin at the location of the MEI (or a representative				
and Submersion Doses)	location) should be routinely sampled and analyzed for the radionuclides				
,	important to dose estimation, and routine measurements of penetrating				
	radiation should be performed at those sites that, as determined by site-				
	specific exposure pathway analysis, might result in an annual dose of site				
	origin at the site boundary, if the total exceeds:				
	a) 5 mrem effective dose, or				
	b) 100 person-rem collective effective dose to the affected				
	population (e.g., within a radius of 80 kilometers (km) of a central				
	point in the site).				
Periodic Confirmation	Environmental surveillance measurements may be performed				
	occasionally when potential dose is low, but should be performed at least				
	every five years even when the projected annual effective dose to th				
	public is less than 0.1 mrem. The frequency and magnitude of				
	environmental surveillance should be proportional to the potential annual				
	dose. Where potential annual dose represents a significant fraction of the				
	reference dose for routine surveillance, environmental sampling should				
	be more frequent. At 20 percent of the reference dose (e.g., 1 mrem				
	effective dose from emissions during a year), annual surveillance for				
	confirmation should be considered. Similarly, more frequent				
	measurements may be warranted if the biota screening levels are				
	challenged.				
Pathway Measurements	Actual measurements on two media for each critical radionuclide/pathway				
	combination, one of which might be the effluent stream, should be				
	performed as part of the site routine environmental monitoring and				
	surveillance program.				
Characterization of	Use of data should be based on statistically significant differences				
Background	between the point of measurement and background data.				
Unplanned Releases Provisions should be made, as appropriate, for the detection an					
	quantification of unplanned releases of radionuclides to the environment.				

6.2.1 Evaluation of Need for Sampling

The need for environmental sampling and analysis should be evaluated, by exposure pathway analysis, for each site radionuclide effluent or emission (liquid or airborne). This analysis with appropriate data, references, and site-specific assumptions, along with site-specific criteria for selection of samples, measurements, instrumentation, equipment, and sampling or measurement locations, should be adequately and appropriately documented as part of the ERPP. If actual releases are significantly greater than expected, or if unplanned or accidental releases occur, re-evaluate the environmental surveillance needs based on the actual releases. A critical pathway analysis (radionuclide/media) should be performed, documented, and referenced in appropriate documentation (e.g., ERPP-related documents, ASER). If the projected dose equivalent from inhalation of particulates exceeds the criteria of Table 6-1, a particle-size analysis of the sample should be conducted at least annually. In addition, the lung solubility class that is assumed for the particulates in question should be justified and resubstantiated on an annual basis if it is likely to vary with changing facility operations. If environmental surveillance data are to be used with (or in place of) effluent monitoring and modeling to support the assessment and demonstration of compliance with such regulations as 40 CFR Part 61, consider the special requirements of those regulations in the planning and implementation of the environmental surveillance system.

The radionuclides of interest are site specific and should be identified based on process knowledge, previous sampling results, and other pertinent information. Radionuclides of interest discussed in this document are provided as a general guide only. While it is tempting to include every suggestion, excessively long lists of radionuclides may lead to:

- Extra expense;
- Spectral interference; and
- False positives.

<u>Expense</u>: depending on the procedure, the analytical laboratory may charge more for extra analytes.

<u>Spectral interference:</u> occurs when the alpha-particle or photon energies for different radionuclides are too close. Peaks may overlap, or one peak can affect the estimated background of another, or unwanted data from one radionuclide may be within the region of interest of another. This is a frequent problem, and becomes more frequent with a long list of analytes.

<u>False positives:</u> typically, a few percent of the peaks reported by a computer program are false positives, and the more peaks the computer is asked to look for, the more false positives it will find. The problem of false positives is aggravated by the common practice of reporting the number of counts in a "region of interest" regardless of whether there is a well-defined peak in the right place and with the right shape. The human eye is good at pattern recognition; if a peak does not look real to a human eye, it probably is not real. If there are serious ramifications with a false positive, health-physics staff should examine the shapes and locations of the peaks, and consider whether all the expected peaks are credible, and have areas consistent with expectations.

Health physics staff should help to develop a list of radionuclides with a credible chance of being observed. Legacy materials are likely to predominate at most DOE sites. In this case, materials with short half-lives are unlikely unless there is a long-lived parent.

When determining radionuclides of interest, staff should consider each category: transuranics, uranium, fission products, and activation products.

<u>Transuranics</u>: the most common transuranics are: Pu-238, Pu-239, and Am-241. Transuranics should be measured by alpha spectrometry or elemental analysis techniques. Detection of transuranics by gamma spectrometry is unreliable because of the spectral interference between the low-energy gammas emitted by transuranics and the K-shell and L-shell x-rays emitted by many radioactive materials.

<u>Uranium:</u> may be categorized as natural uranium, uranium tailings, refined uranium, enriched uranium, and depleted uranium. Uranium should be measured by alpha spectrometry or elemental analysis techniques. The first two categories include Pb-214 and Bi-214, the other three do not. Refined uranium does not include measurable amounts of Pb-214 and Bi-214, because Th-230 and Ra-226 were removed during the refinement process and they take thousands of years to grow in. Therefore, at facilities that use only refined uranium, the presence of Pb-214 and Bi-214 indicate natural uranium. On the other hand, facilities that processed uranium ore will have Th-230, Ra-226, Pb-214, and Bi-214 in the tailings. The isotopic ratios of U-234:U-235:U-238 are useful, though staff should be aware that water is usually enriched in U-234 because the decay process causes the U-234 to become dislodged, and so makes it more soluble.

<u>Fission products:</u> normally occur together in a mixture known as "mixed fission products." At reactor facilities or re-processing facilities, the list of fission products will be long. In legacy

material, only two fission products are normally measurable: Sr-90 and Cs-137. However, Cs-134 is easy to detect by gamma spectrometry when sampling shortly after fission product accident events (e.g., Fukushima-Daiichi). Iodine isotopes are potentially important, though most have short half-lives; iodine-129 is naturally occurring and is very difficult to detect.

Activation products: Co-60 is easy to detect by gamma spectrometry. Na-22 is also easily detected by gamma spectrometry and may be measurable in more recently activated materials. Tritium is widespread and requires specialized detection techniques. At accelerators, the list of activation products will be long.

Radionuclides in sealed sources are unlikely to be found in the environment. Also, small quantities of radionuclides used in a well-managed modern facility are unlikely to be found in the environment unless there is a leak or spill. Discharges from a permitted outfall are monitored and the data should guide the environmental staff.

Measurement of background/ambient and near-site naturally-occurring radionuclides (e.g., Be-7, K-40, Tl-208, Pb-212, Pb-214, Bi-214, and Ac-228) may be useful as a reality check and to confirm that systems are behaving as expected.

The mobility of radionuclides and the likelihood of uptake into biota should also be considered. Tritium is the most mobile and is readily taken up by biota. Iodine is mobile, readily taken up by mammals, and concentrates in the thyroid, so it should be monitored if recent fission products are credible; several iodine isotopes are readily detected by gamma spectrometry when sampled shortly after fission product accident events (e.g., Fukushima-Daiichi). Strontium behaves like calcium and it is moderately mobile. Uptake of strontium into biota depends on the availability of calcium in the environment, though some plants do not easily discriminate between strontium and calcium. Cesium cations attach strongly to the soil matrix and are less mobile than strontium. Most biota can discriminate between cesium and potassium, so uptake of cesium is dependent on the availability of potassium. For example, cesium uptake is common near Savannah River where the soil is deficient in potassium, and less common where potassium is abundant. Transuranics are generally less mobile and not easily taken up by biota.

6.2.2 Emergency Monitoring Provisions

Emergency monitoring is beyond the scope of this document. However, provisions for monitoring during an emergency situation should be considered when planning for environmental monitoring and determining routine program needs. Further provisions should be

made, as appropriate, for the detection and quantification of unplanned releases to the environment of radioactive materials, including radionuclides that may be transported by storm water runoff, flooding, or re-suspension of ground-deposited material. It is important to establish an environmental surveillance program that will provide adequate data to compare to measurements taken to support a response should an emergency occur.

6.3 Performance Requirements for Environmental Surveillance Programs

For all new or modified DOE facilities, a pre-operational assessment should be made and documented to determine the types and quantities of effluents to be expected from the facility and to establish associated environmental surveillance programs. Calibration of dosimeters and exposure-rate instruments should be based on NIST traceable standards. Where significant variations in effluent releases are observed or expected, obtain environmental samples or perform measurements either continuously or at an interval less than one-half the expected peak-to-peak interval. Gross radioactivity analyses should be used only as trend indicators, unless documented supporting analyses provide a reliable relationship to specific radionuclide concentrations or doses. The overall precision (± percent uncertainty) of all measurements should be estimated, and the LLD at specified confidence levels for appropriate radionuclides should be determined and documented. Sample preservation methods used to assure integrity should be consistent with the analytical procedures used. All environmental surveillance programs and procedures should be designed to ensure representative samples or measurements of the radiation exposure pathway media are obtained.

6.3.1 Specific Performance Requirements

Sampling or measurement frequencies for each significant radionuclide - environmental medium combination (e.g., those contributing greater than 0.1 mrem effective dose or greater than or equal to 10 percent of the annual offsite dose from all emissions) should take into account the half-life of the radionuclides to be measured and should be documented in the site environmental surveillance description. When considering short-half-life radionuclides, ensure that the sampling and measurement intervals do not exceed twice the half-life of the radionuclide. "Background" or "control" location measurements should be made for every significant radionuclide and pathway combination for which environmental measurements are used in the dose calculations. An annual review of the radionuclide composition of effluents or emissions should be conducted and compared with those used to establish the site environmental surveillance monitoring program plan or other document that describes the

environmental surveillance. Deviations from established environmental surveillance requirements, including sampling or measurement station placement, should be documented.

6.3.2 Air Sampling System

Air sampling equipment calibrations should be performed by either a primary measurement device or a calibrated secondary measurement device at the field location. Recalibration should be performed on a pre-determined schedule. The air sampling rate should not vary by more than ± 20 percent, and total air flow or total running time should be indicated or recorded; air sampling systems should be leak-tested, flow-calibrated, tested, and inspected on a routine basis. At a minimum, the manufacturer's recommended calibration frequency should be followed.

6.3.3 Consultation with Game Officials

If protected species are selected through the critical pathway analysis to sample, analogous species should be selected and sampled in their place whenever possible. State and local game officials should be consulted when selecting appropriate protected species to sample.

6.3.4 Consultation with Local, State and Regional EPA Representatives

DOE field elements and contractor staff should ensure that ground water monitoring plans are consistent with applicable State and regional EPA ground water monitoring requirements. DOE Federal and contractor staff should consult as needed, with local, and State representatives and regional EPA offices, to ensure that applicable requirements are incorporated into environmental surveillance program documentation.

6.4 Design Criteria

It is important that overall objectives for environmental monitoring programs be established and documented. It is also important that the environmental surveillance program be reviewed periodically and modified as program needs change. The general design criteria for establishing an environmental surveillance program for radioactive materials released in the effluents or emissions from DOE-controlled facilities are discussed in the following sections.

6.4.1 Environmental Surveillance Program Objectives

Environmental surveillance programs conducted at all DOE sites should enable the following to be determined:

- Compliance with all applicable environmental quality standards and public exposure limits; and the requirements of DOE O 458.1;
- Background levels and site contributions of radioactive materials in the environment;
- Effectiveness of effluent treatment and controls in reducing effluents and emissions;
- Validity and effectiveness of models to predict the concentration of contaminants in the environment;
- Quantification of contaminant transport into the environment;
- Long-term buildup and prediction of environmental trends from site-released radioactive material; and
- Detection and quantification of unplanned releases.

In addition to determining the need for an environmental surveillance program based on the objectives noted above, certain subsidiary objectives should also be considered. For example, site history and current public interests might indicate the need for an environmental surveillance program that examines specific aspects of a site's environmental impact, even when no other need is indicated.

The following is a partial list of subsidiary objectives (as provided in ICRP 1985) that should be considered when establishing environmental surveillance program objectives:

- The environmental surveillance should provide information that is available to the public;
- The environmental surveillance program should provide data that enable distinguishing site radiation contributions from other local sources (natural or manufactured);
- The environmental surveillance program should be capable of obtaining data that may be needed to assess the consequences of an accident; and
- Elements of the environmental surveillance program should be capable of determining site-specific values for transfer parameters where appropriate. Information on transfer parameters is provided in IAEA (2010a), ICRP (2009), and Staven et al. (2003).

6.4.2 Program Planning and Design

Factors that affect the relative level of environmental surveillance and to some extent the points at which measurements are to be made, include:

- The potential hazard of the materials released, considering both expected quantities (including unplanned releases) and relative radiotoxicities;
- 2) The extent to which facility operations are routine and unchanging;
- 3) The need for supplementing and complementing effluent monitoring;
- 4) The size and distribution of the exposed population;
- 5) The cost effectiveness of modifications to the environmental surveillance program; and
- 6) The availability of measurement techniques that provide sufficiently sensitive comparisons with the applicable standard and "background" measurements.

The environmental surveillance media sampled or radiation measurements made should represent, as much as possible, the actual exposure vectors to people. Selection of locations, frequency, media and radionuclides to be measured, and measurement techniques are the basis of an environmental surveillance program. This program also should include any special monitoring required, such as trend indicators and additional samples/measurements required for quality assurance. The effort devoted to the environmental surveillance program should reflect the significance of the projected radiation doses.

Once the critical pathways and nuclides are identified (i.e., a critical pathway analysis is carried out), an annual review comparing reported effluent releases with those considered in the original analysis should be conducted and changes in the environmental surveillance program noted in a revised version of the appropriate program document and discussed in the ASER.

The effluents and the environment into which they are dispersed are dynamic, exhibiting both spatial and temporal variations of nearly all constituents. The importance of each individual radionuclide depends on its physical and chemical form, which determines its movement in the environment and eventual uptake, deposition, and retention by humans, and on the differential metabolism of the radionuclide by humans.

Providing site-specific tables of the environmental sampling/measurement locations per site as a function of calculated annual total effective dose (TED) to the representative person or to the MEI or collective dose is recommended. Any changes in site-specific factors (e.g., location of samples, type of samples, average temperatures, wind direction or velocities) and the basis for

the change(s) should be indicated in environmental surveillance program documentation. Information previously used should be preserved in historical records.

6.5 External Exposure Monitoring

A primary objective of external exposure monitoring is to assess and limit the actual or potential radiation dose to persons in the site environs. External exposure monitoring considerations include: (1) external exposure in air; (2) external exposure in water; (3) external radiation measurement locations and frequency; (4) factors in selection of indicator locations; (5) locations of background measurement stations; (6) onsite and offsite locations needed to characterize discharges or confirm effluent monitoring and modeling projections; (7) shoreline locations; and (8) height and frequency of measurements.

For most DOE facilities, the whole-body exposure is limited, and penetrating radiation measurements are satisfactory. Exceptions could include the atmospheric release of beta emitters such as uranium decay products or krypton from fuel manufacturing or reprocessing facilities, respectively. For DOE sites, the gamma (and, where applicable, neutron) exposure (or exposure rate) should be measured or calculated; any significant skin dose from airborne beta emitters should be calculated from effluent data. If external beta doses from deposition are considered to be significant, they should be estimated from effluent data, from beta-sensitive dosimeters, or by soil or vegetation sampling and laboratory analysis.

6.5.1 External Exposure in Air

One of the "critical pathways" of exposure for population groups living within the vicinity of DOE facilities is exposure to external radiation from those sites (Denham 1979). Exposure of population groups to external radiation from DOE operations includes: (1) cloud passage of airborne effluents; (2) previously released and deposited radionuclides on soil, vegetation, or sediments; (3) radiation-generating facilities, especially high-energy accelerators or industrial x-ray equipment, and large isotopic radiation sources; and (4) the storage, disposal, or movement of large sources of radioactive waste.

6.5.2 External Exposure in Water

External exposures from radionuclides in water generally are insignificant. However, unique situations could arise where recreational, commercial, or industrial use of a receiving body of water might cause exposure to certain individuals. Appropriate environmental measurements should be included in the routine program to better define an unusual "source" if the site-specific

pathway analysis shows this to be a significant (greater than 10 percent of the total offsite dose) source of exposure.

6.5.3 External Radiation Measurement Locations and Frequency

Considerable judgment needs to be used in locating environmental radiation measurement stations. Before final placement of any environmental radiation measurement station (background or control and indicator locations), an initial on-the-spot survey should be performed and documented to determine the absence of possible naturally occurring anomalies that could affect interpretation of later measurements. The recommended technique for making these pre-surveys is to use a low-level radiation survey instrument (e.g., micro-R meter) followed up with a pressurized ion chamber (PIC) measurement at those geographic locations selected on the basis of the preliminary screening by portable instrument survey. If desired, an in situ gamma- ray spectrometer (e.g., Nal, IGe (Intrinsic Germanium detector), or Ge(Li)) can be used to determine which terrestrial nuclides are contributing to the observed exposure rate.

Examples of dosimeter placement locations to be avoided, if at all possible, include the following:

- Locations of unique or different geology (i.e., reflecting changes in the terrestrial background);
- Locations where the altitude differs significantly (e.g., altitudinal differences between "background" or control locations and those indicator locations to be used around a given DOE site should not exceed 150 m (reflecting changes in the cosmic-ray background));
- Locations where the proximity of structures could alter the measurement results
 (reflecting changes from shielding or high background radiation levels due to naturally
 occurring radionuclides in building materials (e.g., thorium, uranium, radium)); and
- Valleys or hollows (where puddles of precipitation or runoff could accumulate, or where local topography could shield the dosimeters from the possible passage of airborne effluents).

6.5.4 Factors in Selection of Indicator Locations

Selection of the indicator locations⁴ for external exposure monitoring should be based on expected sources of external radiation – noble gas plumes, soil-deposited atmospheric particulates released from the site, onsite radiation-generating facilities or large radiation sources, or potential routes of waste transport from the site – and the local population distribution and prevailing wind directions. The technique described by Waite (1973a, 1973b) for placement of air samplers, based on average meteorological conditions and existing population distributions should be considered for determining external radiation measurement locations.

6.5.5 Location of Background Measurement Stations

Background or control measurement stations should be located a minimum distance of 15 to 20 kilometers (km) from the larger sites and 10 to 15 km from the smaller sites in the least prevalent wind direction. Control stations should also be placed in areas typical of local geology, away from buildings (which can shield the detectors), and at similar elevations to those for indicator stations. The emphasis here is on the placement of dosimeter stations such that the difference between background/control or pre-operational data and the data from those stations expected to be affected by site effluents/activities can be assessed accurately.

6.5.6 Offsite Locations

Offsite radiation measurement locations should be monitored for each DOE site where predicted external radiation doses exceed the 0.1 mrem effective dose criterion in Table 6-1. These offsite measurement locations include a background or control location, site perimeter or boundary locations, and locations in nearby communities (within a pre-determined distance from the site to include communities in the predominant transport regions). The site perimeter or boundary locations should include locations directly upwind from the maximum predicted

⁴ Indicator locations are monitoring locations intended for measuring radioactive material or radiation that has or may be present as a result of a DOE activity or operation. Background or ambient background locations are those monitored or sampled to establish to establish radiation or radioactive material levels that are not associated with DOE activities. Indicator locations also are used to verify or validate modeling projections and in such cases may indicate nothing above background.

ground-level concentration from atmospheric releases averaged over a period of 1 year. Offsite measurement locations should coincide with locations where maximum predicted levels occur and where any member of the public resides or abides.

For those sites larger than a few kilometers in radius, the maximum predicted concentrations may actually be onsite. In this case, radiation measurements may be made at the onsite location of predicted maximum air concentration(s), as well as other locations that may be helpful in the interpretation of offsite results.

6.5.7 Shoreline Locations

If exposure measurements are to be made at shoreline locations, dosimeters should also be placed to correspond to key water sampling locations (including the site boundary), as well as locations important for recreational, commercial, or industrial use. However, changes in water elevation caused by tides or fluctuating releases from dams may make this impractical, in which case intermittent exposure-rate measurements need to be used during the seasons in which recreational use of the shoreline (for hunting, fishing, sun-bathing) actually occurs.

6.5.8 Height and Frequency of Measurements

The recommended height for external radiation measurement is 1 m above the surface. If another height is used, the relationship to the 1 m height should be established and documented for the site. The frequency should be based on predicted exposure rates from site operations at the measurement locations. Integrating devices (e.g., dosimeters) should be exposed long enough (typically 1 calendar quarter) to produce a readily detectable dose (e.g., 10 × the minimum sensitivity of the dosimeter). If intermittent external radiation measurements are made, their frequency should be timed to coincide with batch atmospheric releases or the intermittent use of large sources or the operation of radiation-generating facilities.

6.6 Direct Radiation Measurement (Pressurized Ion Chamber-Type Instrumentation)

Factors for direct radiation measurement that need to be considered include: (1) continuous exposure monitoring; (2) neutron monitoring; and (3) instruments and methods to use.

6.6.1 Continuous Exposure Monitoring

Continuous environmental gamma-ray monitoring is available (Jackson et al. 1985; Urabe and Katsurayama 1984) and highly desirable, yet it cannot always be justified on the basis of initial

system cost or long-term maintenance. However, in situ gamma spectrometry should be used as a method of documenting environmental mixtures of radionuclides resulting from natural and manufactured sources (e.g., for dosimeter placement). Historical monitoring information also should be considered. The deployment of at least one continuously recording exposure-rate instrument is recommended, preferably near the site boundary in the expected direction of a potential plume. An array of continuously recording exposure-rate instruments should be considered if there is a potential for release of large inventories of gamma emitters.

6.6.2 **Neutron Monitoring**

For some sites, especially in the vicinity of high-energy facilities, neutron monitoring also may be necessary. Application of detection techniques to measure environmental levels of neutrons is limited. Commonly used materials for detection of slow or thermal neutrons include using a Bonner multisphere, etched track detectors, silicon diodes, and ionization detectors such as boron trifluoride proportional counters. Monitoring for fast neutrons will require use of Columbia Resin #39 (CR-39) detector material.

When neutron monitoring is necessary, the method of measurement should be based on the anticipated flux and energy spectrum. A fixed monitor (moderated boron trifluoride (BF₃) counter or rem counter) is recommended, yet site-specific conditions may warrant the use of intermittent portable instrument surveys only during the infrequent periods of machine operation. As with all external radiation measurements, neutron monitoring (or surveys) should be performed at the site boundary or location of nearest occupancy in the direction of maximum expected exposure rates, especially from beam dumps or accelerator targets.

6.6.3 Instruments and Methods

Instruments or dosimeters that have application to DOE environmental surveillance programs include Geiger-Muller (GM) and gamma scintillation systems, Pressurized Ion Chambers (PICs), Thermoluminescent Dosimeters (TLDs), Optically Stimulated Luminescence (OSL) Dosimeters and moderated BF₃ counters or rem counters. The method of measurement should depend on the anticipated type of radiation (beta, gamma, or neutron).

At high-energy particle accelerators, muon fields are monitored with scintillation counters located in the beam line. The muon fluence may fluctuate with the mode of operation of the accelerator. A typical application of in situ muon monitoring is to mount detectors on a vehicle

with the associated electronics and data acquisition system, and collect data while traversing the beam line.

Where integrating dosimeters are used, two or more dosimeters should be provided at each location (in the same package, if possible). Integrating dosimeters should be read without undue delay. It is critical that readings are made at a consistent time following collection.

DOE sites are encouraged to participate in international inter-comparison studies, such as the ones reported by dePlanque et al. (1976) and Gesell et al. (1982).

Only if adequate precautions are taken to avoid recording a significant exposure in transit can integrating dosimeters be sent to a distant location for processing.

6.7 Air Measurements and Sampling

The categories of airborne radionuclides that should be considered for measurement in air sampling systems include: (1) particulates; (2) gases (principally the noble gases); (3) halogens (principally radioiodines); and (4) tritium. These constituents are important because they account for virtually all of the radioactive materials released from DOE sites.

6.7.1 Basis for Air Sampling

Since air is a primary exposure pathway to humans from radionuclides released to the atmosphere, air sampling should be conducted to evaluate potential doses to populations from inhaled or ingested radionuclides or from exposure to radiation sources external to the body. The inhalation of airborne radionuclides, coming either directly from the source (facility) or from re-suspension following deposition, may result in their absorption from the lung or gastrointestinal (GI) tract. Absorption through the skin because of immersion in a "cloud" of gas and/or particulates may contribute to human exposure.

Radioactive materials in particulate form can result in radiation exposures to individuals both by direct inhalation and by deposition onto other environmental media. Therefore, wet/dry deposition monitoring should be included.

Although particle sizes range across a broad spectrum, with diameters ranging from about 0.01 to 10 micrometers (μ m), the optimum size for deposition in the upper respiratory tract (and subsequently the deep lung) tends to be in the range of 0.01 to 3 μ m, with 1 μ m often used for dose assessment. However, particulate filters used for sampling will function over the entire size spectrum, collecting particulates in the "respirable" range, as well as those that are not.

The collection efficiency of filters used to collect particulate materials should be considered when calculating the concentration of radionuclides in the air that was sampled. If releases of particulate materials could contribute significantly to environmental doses, measurements of particle size should be made. When inhalation of particulates may be significant, lung solubility class assumptions should be substantiated.

The filter type should be appropriate for the particles being emitted. EPA (2002a) identified some choices:

- Cellulose B a general-purpose filter, but not suited to alpha-emitting nuclides;
- Glass fiber high collection efficiency, without high airflow resistance, good for high temperature applications;
- Membrane good for alpha-emitting nuclides, but is fragile and has high airflow resistance; and
- Synthetic fiber special fibers tailored to specific needs and situations.

NCRP (2010) provides further discussion of the characteristics, advantages and limitations of selected filter media.

It is often more feasible to determine the impact of short-lived gamma-emitting gases (e.g., nitrogen-13 (N-13) and argon-41 (Ar-41)) by measuring the direct exposure (i.e., external radiation) resulting from them rather than by sampling and analysis. Gamma spectroscopy of grab samples (e.g., filling a previously evacuated Marinelli sampler) can be used to quantify the concentrations of short-lived gases, which can then be correlated with the observed increase in exposure rates. For longer-lived noble gases (e.g., krypton-85 (Kr-85) and xenon-133 (Xe-133)), one technique used is the collection of an air sample by compression or cryogenic techniques, separation and purification of krypton and xenon by adsorption on chromatographic columns, and analysis by liquid scintillation counting (Grossman and Holloway 1985; Trevathan and Price 1985). For noble gases, sampling using an activated charcoal is another method. For other halogens, noble gases, and water vapor, activated charcoal is an efficient absorber. Because the adsorption process is not radionuclide-specific, the analyses of other radioactive halogens and noble gases with charcoal will require analytical discrimination to measure the concentrations (NUREG-1400).

Rainwater surveillance could be included in the evaluation of the airborne pathway. Locations for rainwater sampling should be co-located with air, vegetation, and soil surveillance locations. Sampling for deposition should be collected over a predetermined area, such as a 2-foot by 2-

foot stainless steel pan located on top of the monitoring stations. Dry deposition on the pan prior to rainfall is washed through the system with the rainfall. Therefore, the sample represents both wet and dry deposition. The rainwater is collected beneath the station in a collection bottle and is then analyzed in the laboratory for the required analyses. Separation of tritium from non-tritium isotopes could occur during the field collection using an ion exchange resin column or at the laboratory.

6.7.2 Sampling Locations

Offsite air samplers should be employed at each DOE site having potential airborne releases that could result in an annual effective dose greater than 1 mrem to the MEI. The exact number of samplers will be determined by meteorology, demography, and the magnitude of projected doses to the surrounding population.

Sample locations should include the following: (1) a background or control location; (2) representative locations of maximum predicted ground-level concentration from stack (or vent) releases, averaged over a period of 1 year where members of the public reside or abide; (3) locations in the nearest community within a 15-km radius of the site's sources; and (4) locations necessary to confirm modeling or to characterize impacts of off-normal discharges.

For those sites larger than a few kilometers in radius, the maximum predicted concentrations may actually be onsite. In this case, onsite sampling may include the locations of predicted maximum concentration(s) and any other locations needed to help interpret the offsite sample results. However, if there are no receptors onsite, depending on the sampling network, it may be cost effective not to have an additional sample at the maximum predicted location.

Selection of background sampling and measurement locations for air should be made with special care. For measurements to be compared with the effects of airborne releases, a minimum distance of 15 to 20 km from the larger sites and 10 to 15 km from the smaller sites in the least prevalent wind direction (that is, upwind) is suggested for background sampling. If the MEI could receive a TED of more than 5 mrem, additional air samples should be collected in those communities within a 15-km radius of the site boundary for which the projected dose equivalents exceed the criteria in Table 6-1, and at a background location (10 to 20 km from the site in the upwind direction).

Unless documented site-specific evidence exists to justify otherwise, the sample(s) at each air sampling station should be collected at a height of 1-2 m above ground level (approximately the

height of inhalation for adults), in a location free from unusual localized effects or other conditions (e.g., in proximity to a large building, vehicular traffic, or trees) that could result in artificially high or low concentrations. If possible, locations should be selected to avoid areas where large-particle (non-respirable) fugitive dusts can dominate the sample (Ludwig 1976).

A method similar to that developed (Waite 1973b) and evaluated by Waite (1973a) should be used to determine the number of air sampling stations and their placement. Waite's method entails examining demographic and meteorological data for the site to determine the distance to local population centers, their population, and the wind frequency distribution and weighting factors that are scaled to equal the desired number of sampling locations. The application of this method to sites in coastal or agricultural areas requires only minor modification of the procedure illustrated (i.e., sites in coastal zones would adjust the number of radial divisions to the number required to cover the surrounding inhabited land mass).

6.7.3 Sampling Frequency

Many factors should be considered to determine sample frequency. In general, the frequency of collection for air samples is adjusted to take into account the limitations of the sample collectors (collection efficiency), the capabilities of the air movers (e.g., vacuum pumps), and the physical problem of retrieving samples from each location on a fixed frequency, typically 1 to 2 weeks. However, the operational status of relevant facilities should also be considered. Unless otherwise justified, the maximum air particulate filter exchange frequency should be biweekly.

A common practice, especially for the longer-lived radionuclides, has been to composite filters for subsequent analysis from several locations and/or successive time periods, taking advantage of the larger volume of air sampled to achieve the desired sensitivity. Use of compositing techniques assumes that the concentration of a given radionuclide at the locations or for the time composited is sufficiently constant for the end use of the data. NCRP (2010) identified the following disadvantages of compositing: (1) poor resolution in time or location for an elevated concentration; (2) masking a single spike of high concentration by large numbers of filters with normal low activity; and (3) difficult modifications to analytical procedures when large numbers of filters are combined. Since the applicable standards are annual standards, comparison of annual averages to the standards is appropriate. For dose calculation purposes, the annual average concentration for a location or for a group of locations can still be compared against an annual average for a background location as an indication of potential facility impact during the year in question. Also, averages for successive years can be compared for detection

of general trends. Requirements for sample collection and analysis, including the use of compositing, are shown in Table 6-2 as a function of effective dose to the MEI or representative person.

For air sampling of non-particulate material, the available tradeoff between sensitivity and frequency of sample removal is governed primarily by the fact that "breakthrough" can occur with the charcoal cartridges, molecular sieves and silica gel. These breakthrough phenomena can be based on flow rate, total volume, activity, or a combination of these. The sample exchange frequency for non-particulate sampling should be determined on a site-specific basis and should be documented.

For facilities with a significant release of iodine, measurements can be made at site-perimeter and control stations to characterize local site environs. It is also recommended that the relationship between iodine-129 (I-129) and natural iodine (I-127) be determined. However, it may be assumed that because of the extremely long half-life of I-129, its accumulation (if any) in the environment may be better observed in milk or soils than in air.

6.7.4 Sampling Methods and Criteria

Filtration is by far the most popular air-sampling method (Lee 1974) and the method generally required for air-particulate collection at DOE sites. Correct use of the International Commission on Radiological Protection (ICRP) lung model, as described in ICRP Publication 66 (ICRP 1994), requires knowledge of the chemical state and the particle size distribution. The need for particle size measurements is especially important at those sites where re-suspension of previously deposited material is or can be a significant factor in environmental air concentrations. Such particle size measurements will also be useful in distinguishing resuspended material from that of current emissions. Several methods, including impactors (e.g., multistage cascade impactor) and electrostatic precipitators, can be used to classify particle size (ISO 2010). Particulate filters can be made of any fibrous material, and a variety of filter media (e.g., cellulose, glass fiber, membrane, polystyrene) are commercially available. No single filter type is best for all purposes, but the specific filter to be used should be selected to meet sitespecific requirements, such as high collection efficiency, particle size selectivity, retention of alpha emitters on the filter surface, or ease of radiochemical analysis. According to ANSI/HPS N13.1-1999 filter media used to sample airborne radioactive particles should have a minimum efficiency of 95 percent. (See also Table 4-2).

Airborne radioiodines can be collected with charcoal or silver zeolite cartridges in series behind the particulate filter, and analyzed by gamma spectrometry, the method suggested by the American Public Health Association (1988). For greater sensitivity, I-129 extraction from the charcoal media for concentrated gamma spectrometry or liquid scintillation counting can be performed. (HASL 300, I-01 (modified), EPA (1980a), Method 901.1 [modified] (EPA 1980b)). Compound filter canisters of several designs (e.g., Keller et al. 1970) have been used to distinguish the several chemical forms of radioiodine that may be present in the atmosphere. Generally these canisters will contain a particulate filter and silver wire or mesh plus charcoal, each of which is analyzed separately. This type of collection device should be used if the levels of radioiodine or the cause of the release warrant.

Routine environmental surveillance for short-lived noble gases (e.g., Ar-41) should be performed by external radiation measurements. Laboratory analysis of periodic grab samples of ambient air (Denham et al. 1974) should be performed for the longer-lived radionuclides, principally Kr-85 when the critical pathway analysis indicates the potential dose exceeds the criteria given in Table 6-2. Suggested methods for radioactive gas (Kr-85) sampling, either grab or continuous, can be found in the *Proceedings of the Noble Gases Symposium* (Stanley and Moghissi 1974) and in reports by Grossman and Holloway (1985) and Trevathan and Price (1985). Atmospheric stability and wind speed and direction during the period in which the samples were collected should be recorded to aid in interpreting and using the data for dose calculations.

Several methods are available for collection of atmospheric tritium, such as bubblers, molecular sieves, and silica gel (Denham et al. 1974; Guthrie et al. 2001; Patton et al. 1997; Rosson et al. 1998; Rosson et al. 2000). The American Public Health Association (1988) method recommends the use of silica gel as a desiccant to remove moisture (water (H₂O), HTO) from air, followed by re-evolution, collection as a liquid, and liquid scintillation counting (Griffin et al. 1972; Ostlund 1970; and Osborne 1974). Measurement of the specific activity of tritium in atmospheric moisture, using a passive device such as a container of silica gel suspended in air to collect tritiated water vapor, is considered satisfactory as a qualitative device only. Measurements using an active air sampling system that includes flow through a silica gel column at measured flow rates and volumes are acceptable as a quantitative measure if: (1) the silica gel is pre-dried before field use, and (2) is evaluated for internal intrinsic moisture exchange with tritium oxide in the atmosphere.

TABLE 6-2: Minimum Air Sample Collections and Analyses to Be Performed as a Function of Estimated Total Effective Dose (TED) to the Maximally Exposed Individual (MEI) or Representative Person, as Determined from Gaseous Effluent Releases

	Sample Collection/Analysis Criteria			
Sample Analysis Type	TED < 1 mrem ^a	1 mrem < TED < 5 mrem ^a	TED > 5 mrem	
Air particulate:				
- Total beta	Yes, as indicators	Yes, ^b as indicators	Yes, ^b as indicators	
- Total alpha	Yes, as indicators	Yes, ^b as indicators	Yes, ^b as indicators	
- Gamma spectroscopy	Yes, annual composite	Yes, quarterly composite	Yes, monthly composite	
- Other ^c	No	Yes, quarterly or annual composite	Yes, quarterly composite	
- Alpha spectroscopy ^d	No	No	Yes	
- Particle size determinations	No	Yes	Yes, one sample per quarter	
Noble gases:				
- Direct radiation measurement	No ^e	No ^e	Yes	
- Sample collection	No	No	Yes, one sample per quarter	
Halogens (radioiodine):				
- Charcoal (KI impregnated) or silver zeolite	No	Yes	Yes	
- Species differentiation (I ₂ + CH ₃ I + HOI)	No	No	Yes, one sample per quarter	
Tritium	No	Yes	Yes	

a. Implemented when this TED is estimated to have been received during preceding 12 months

b. Assess relationships to specific radionuclide concentrations or use radiochemical analysis.

Some examples include Sr-90, Pu-239, U-natural or other radionuclides that need to be chemically separated prior to counting; the nuclides chosen need to be based on site-specific effluent data and contribution to dose.

d. Only if actinides other than Pu-239 contribute significantly to the dose as shown.

e. Routine environmental monitoring for incremental exposures of < 1 mrem/yr of direct radiation is not realistically achievable and levels < 5 mrem/yr are questionable.

6.7.5 Radioiodine

Thyroid and whole body exposure to atmospheric release of radioiodine can occur through several pathways including: (1) ingestion of foodstuffs such as milk; (2) inhalation; and (3) air submersion. The inhalation pathway is normally assessed by air sampling, while the external radiation component is assessed along with other external radiation sources by dosimeters. In certain instances, a special sampler and/or a multiple cartridge sampling train might be necessary to identify iodine species (elemental, organic, and hypoiodous acid (H0I)).

Species identification allows differentiation of those forms of iodine that are prone to deposition on vegetation and soil (elemental) from those that are not (organic forms and H0I). All chemical forms can be readily inhaled and contribute to thyroid exposure; however, it is primarily the elemental form that enters the food chain. The manner in which radioiodine concentrations are distributed among the various chemical forms is key input information for accurate environmental dose estimates.

6.7.6 Tritium

Environmental tritium is predominantly found in two forms: tritiated molecular hydrogen gas and tritiated water vapor (or tritiated oxide vapor). In terms of exposure potential, tritiated water vapor yields a dose equivalent approximately 25,000 times that of tritium gas for the same concentration (ISO 2010).

When tritiated water vapor is released to the environment, several exposure pathways including inhalation, ingestion, and absorption are possible. According to a model developed by Anspaugh et al. (1973), approximately 35 percent of the dose to individuals results from inhalation; the remaining 65 percent is due to ingestion (vegetable (36 percent), milk (13 percent), and meat (16 percent)). These percentages are modeled estimates. Actual values will vary from one site to another because of such factors as climate and land use.

For facilities that release tritium to the atmosphere, air sampling is an important medium, but clearly not the only one. Therefore, air-sampling techniques should employ methods that collect moisture from the air. Rosson et al. (2000) determined that a correction is needed to calculate the activity concentration of airborne tritium oxide when dried silica gel is used as the collector.

6.7.6.1 Precautions

A number of precautions should be taken when using the referenced methods and equipment for air sampling in the environment. Some of these relate to general air sampling and some relate specifically to the sampling of particulates, radioiodines, noble gases, or tritium:

- Sufficient material needs to be obtained for analysis of samples in a time frame set to meet reporting and data-retrieval requirements. The requirements of sufficient volume of air and number of samples should be evaluated and the need for compositing samples considered (DOE 1981).
- 2) Excessive material (sample or dust) collected on filters can invalidate the sample in several ways; the flow rate through the filter may be unknown, the pump may fail, the particulate material may penetrate the filter, the analysis for alpha emitters may be affected, or material on the surface may be lost when the flow is interrupted (DOE 1981).
- 3) Excessive sampling velocity can invalidate the sample if too much sample is collected during a specific time period.
- 4) Collection efficiency of an air filter is affected by flow rate; too low an air sampling velocity can produce reduced collection efficiency for specific filters (Keller et al. 1970).
- 5) Ambient levels of radon and the decay products can affect the analysis of a number of filter samples. These naturally occurring radon decay products are found on air particulate filters because they adhere to particulate matter and are thus efficiently trapped by the air sampling filter.
 - Therefore, it is necessary that any gas measurement system for other alpha and/or beta emitters (e.g., Sr-90 and plutonium-239 (Pu-239)) be able to discriminate against the typically much larger "background."
 - Rather than resorting to spectroscopic or chemical separation techniques, a common method of discrimination is to retain the filter from 1 to 7 days after collection and before counting, to allow for decay of the short-lived radon decay products.
 - A method for the determination of the minimum detectable activity of a sample for a two count method for stripping short-lived activity out of an air sample has also been developed for use (Allen 1997).
 - Application of patented techniques in portable instrumentation which apply autoadaptive, real-time spectrometric compensation to evaluate and reduce radon isotope interferences on fresh filters.

- 6) Too high a sampling rate reduces both the collection efficiency and retention time of charcoal filters, especially for the non-elemental forms of iodine (Bellamy 1974; Keller et al. 1970).
- 7) The monitoring of airborne radioiodines is complicated by the occurrence of several species, including particulate iodine (bound to inert particles), elemental iodine vapor, and gaseous (usually organic) compounds. Monitoring should take into account the probable occurrence of the different iodine forms, because their subsequent history in the environment will differ. While it may not be necessary to differentiate routinely between the various species, care should be taken so that no significant error results by neglecting one or more of them (DOE 1981).
- 8) Charcoal cartridges (canisters) for the collection of radioiodine in air are subject to channeling, as with any packing of loose materials.
 - Baffled-flow cartridge design, packing to a minimum required weight, and pre-testing of randomly selected cartridges for pressure drop before operation in the field should minimize the problem.
 - An alternative is to mount several cartridges in series to prevent loss of iodine; each cartridge needs to be counted in this case (DOE 1981).
- 9) For the short-lived radioiodines (mass numbers 132, 133, 135), environmental sampling is complicated by the need to obtain a sufficient volume for analysis while at the same time retrieving the sample soon enough to minimize decay (with half-lives ranging from 2 to 31 hours). Short-period grab sampling with charcoal cartridges is possible, with direct counting of the charcoal as soon as possible for gamma emissions, but radon isotopes including radon-220 will affect detection levels (DOE 1981).
- 10) Because of the extremely long half-life and normally low environmental concentrations I-129 determinations can be performed by neutron activation analysis after chemical isolation of the iodine or by mass spectrometry, liquid scintillation counter (LSC), or enhanced gamma spectrometry.

The following operational criteria relate to environmental sampling instrumentation and methods:

- The linear flow rate across particulate filters and charcoal cartridges should be maintained between 20 and 50 m/minute (DOE 1981).
- To the extent possible, the air sampling system should be protected from factors such as weather, tampering, and theft.

- Air sampling devices, such as "quick-disconnect" filter holders, should be designed so that the potential for loss of sample during the collection process is minimized.
- If impregnated, activated carbon is used as the adsorbent for radioiodine, the adsorber system should be designed for an average atmospheric residence time of 0.05 seconds per centimeter (0.25 second per 2 inch) of adsorbent bed (NRC 2012).
- NRC (1992a) contains guidance relative to determining errors associated with the total volume of air sampled.

6.8 Sampling of Terrestrial Foodstuffs

Sampling of terrestrial foodstuffs can provide information on the presence and movement of radionuclides released to the environment. Considerations for the sampling of terrestrial foodstuffs need to include the possibility of long-term buildup of radionuclides in the terrestrial environment, and the potential presence of radionuclides in agricultural products, milk, vegetation, meat, eggs, and game animals.

If the preliminary analysis of public dose indicates that the annual TED from ingestion of terrestrial foods is 5 mrem or greater, then sufficient sampling and analysis should be conducted so that the foods and radionuclides contributing at least 90 percent of this ingestion dose have been evaluated.

If the annual TED is between 1 and 5 mrem then sufficient sampling and analysis should be carried out to provide reasonable assurance that the doses are within this range.

If the annual TED is between 1 and 0.1 mrem then sufficient environmental surveillance should be conducted to show that the radionuclides are behaving in the environment as expected from historical measurements.

The principal pathways by which foods become contaminated are deposition from airborne materials and crop irrigation from surface or ground waters. The relative contribution of various pathways, foods, and radionuclides to the total dose depends on several factors, including:

- Agricultural uses of the land;
- Farming and gardening practices;
- Soil type;
- Climate (e.g., temperature, rainfall, growing season);
- Dietary habits; and

 Quantities of specific radionuclides released to air and water and their chemical and physical forms.

6.8.1 Possibility of Long-Term Buildup

Even in those instances where the annual TED from ingestion of terrestrial foods is less than 1 mrem, periodic sampling and analysis of indicator materials, such as soil or vegetation should be performed to determine if there is measurable long-term buildup of radionuclides in the terrestrial environment. Such long-term buildup could affect the relative contributions of certain radionuclides and foods to the total radiation dose of site origin. However, the availability of these radionuclides to plants grown in such soil may decrease with time as a result of several natural processes. These processes include changes in chemical or physical form of the radionuclides caused by weathering or the action of soil bacteria, fixation onto soil materials or the litter layer, migration below the root zone of the plant with irrigation water or rainfall, and removal of contaminated soil by wind or water erosion or by cultivation.

Unless terrestrial foods or indicator organisms are being analyzed routinely, the pathway evaluation should be repeated annually to reaffirm the original evaluation. Foods to be considered in the pathway analysis, listed in approximate descending order of importance, are: milk, vegetables, meat, eggs, grain, and fruit. If wild game, such as deer, game birds, or fish, is available locally then these should also be considered in the pathway analysis.

6.8.2 Agricultural Products

Representative samples of the pathway-significant agricultural products grown at locations surrounding the site should be collected and analyzed for radionuclides potentially present from site operations. These samples should be collected in at least two locations: the place of expected maximum radionuclide concentrations and a "background" location unlikely to be affected by radionuclide effluents and emissions released from the site.

Fresh produce, meat, poultry, and eggs can be purchased from local farmers or from commercial outlets if the local origin can be identified. Where warranted, and based on site-specific considerations, it may be necessary for individual DOE sites or facilities to conduct sampling at extended distances from the site or facility.

6.8.2.1 Milk

Cow milk, and in certain localities goat milk, is widely consumed by all age groups. Therefore, milk is frequently one of the most important foods contributing to the radiation dose to people if dairy animals are pastured near a DOE site. The source of where the dairy gets its feed for the cows should be documented. If dairy herds or "family" cows (or goats) are present in the vicinity of the site (within a distance normally downwind that would be impacted by releases of radioactive materials), representative milk samples should be taken and analyzed for radionuclides potentially present from site operations. The frequency of sampling will depend on the magnitude of the radiation doses potentially received via this source. Radionuclides of potential significance in milk include: strontium-89 (Sr-89), strontium-90 (Sr-90), iodine-131 (I-131), cesium-137 (Cs-137), and possibly H-3 and I-129.

The number of locations to be sampled depends on the number and distribution of the dairy herds or family cows in the potentially impacted vicinity of the site (i.e., one sample at highest annually averaged air concentration and in each area where estimated doses exceed the criteria in Table 6-1) but a minimum of one background and one potentially affected location should be sampled at least annually. For I-131 analyses, sampling should be at least biweekly during the local grazing season. The frequency should be increased if the I-131 release rate is highly variable. For longer-lived radionuclides such as Sr-90, I-129, and Cs-137, quarterly composite samples are usually adequate.

Milk samples should be as representative of the location of interest as possible. Commercially-available processed milk, while representative of consumption by the general public, may include milk produced in areas remote from the site. Information about the dates and distribution patterns of local milk production is essential if the analytical results are to be meaningful. Raw milk should be sampled for evaluation of potential radiation doses to individuals consuming milk produced by a family cow.

No particular sampling preservation techniques are necessary, other than to guard against cross-contamination and souring or curdling of the milk. However, specific requests should be made to the milk producer so that techniques are in accordance with the protocol accepted by the appropriate State agriculture department. A 4-liter sample of cow's milk is necessary to achieve the required detection level for the contamination expected at most DOE sites. However, for goat's milk, a 1-liter sample may be all that can be obtained, especially from a single goat. Milk samples should be refrigerated or frozen, or otherwise preserved (e.g., packed

in ice) prior to analysis; however, the analytical procedure to be used should be considered when choosing a sample preservation method. Radioanalysis of milk usually involves ion-exchange techniques (for concentration) followed by beta or gamma counting.

When fresh milk is not available, analytical results of leafy vegetable (or fresh forage) samples can be used to estimate concentrations in milk using transfer coefficients or concentration ratios for dose calculations.

6.8.2.2 Vegetation

Vegetation includes three categories: vegetables, grains, and fruit. If vegetation (i.e., vegetables, grains, and fruit) is not one of the contributing pathways involved in determining the dose to humans from the site, native (non-cultivated) vegetation can be used as indicator species. Collection and analysis of vegetation samples can serve three useful purposes: (1) evaluating the potential radiation doses received by people consuming such vegetation; (2) predicting the possible concentrations in meat, eggs, and milk from animals consuming contaminated forage (and resultant radiation doses to consumers of the animal products); and (3) monitoring trends in environmental contamination and possible long-term accumulation of radionuclides.

Radionuclides of interest in vegetation include those listed previously for milk (H-3, Sr-89, Sr-90, I-129, I-131, and Cs-137, and possibly ruthenium-106 (Ru-106)). Several kilograms of vegetation may be needed to provide a sufficient sample for analysis, depending on the analytical sensitivities for the radionuclides of interest. The particular samples collected will depend on species availability, seasonal growth patterns, farming practices, and the reasons for sample collection. Where actual measurement of radioactivity cannot be made (e.g., radioactivity levels are below minimum detectable concentrations), estimates of ingestion dose should be obtained using atmospheric dispersion and dose modeling computer programs such as EPA's CAP88 code.

The vegetable category includes common garden crops (e.g., corn, beans, potatoes, tomatoes, etc.). If the samples of garden vegetables are being collected for evaluation of radiation doses, then the edible portions of the vegetables should be analyzed for the radionuclides of interest. Analysis may include direct gamma measurement, or alpha or beta counting after drying, washing, and/or chemical separation of the desired radionuclide. The results should be expressed in terms of the radionuclide concentrations in the vegetables (consumed state) used in the dose calculation (e.g., fresh weight, peeled weight, etc.).

Samples of vegetables should be collected at local farms or from family gardens when the effective dose to individuals is being evaluated. When collective doses are being evaluated, fresh produce from commercial sources should be included in the samples. Care should be taken to collect vegetation from open, unshaded areas where radionuclide ground deposition would be expected. It is important that the origin of the materials sampled be within a 10- to 15-km radius of the site and be identified. Analyses of commercial food items of known origin can also provide data on concentrations of naturally occurring or fallout radionuclides.

The grain category includes sweet corn, field corn, wheat, and other cereal grains. It is not likely that field corn would need to be sampled, since it is used for animal feeds, and animal products would be more logical items to sample for evaluation of intake of radionuclides by humans. Strontium and cesium are usually the only radionuclides of interest in cereal grains. With the exception of fresh sweet corn from local farms, most grains, by the time they are consumed, would not be likely to contain any radionuclides with half-lives shorter than a few weeks. In addition, most pathway models use concentration ratios (picocuries per kilogram (pCi/kg) plant per pCi/kg soil) that reflect the average concentration of radionuclides in the whole plant.

Radionuclides of potential interest in fresh sweet corn include: zinc-65 (Zn-65), Sr-90, and I-131. Local sweet corn should be sampled annually at harvest time from a "background" farm and a farm where there is a potential for contamination with radionuclides released from the site. A 1-to 2-kg sample of corn should be sufficient for analysis unless the pathway analysis indicates an unusually high potential for contamination, other grains will probably not need to be sampled.

The category of "fruit" includes: tree fruits, berries, melons, and grapes. Unless the pathway analysis indicates that some unusual circumstances are present, it is normally not necessary to sample such fruit.

Samples collected for evaluation of intake of radionuclides by farm animals should be representative of the vegetation consumed by the animals. This includes silage and hay as well as fresh forage when available. Samples collected for monitoring of long-term trends in environmental contamination should be capable of accumulating the radionuclides of interest to permit detection at the desired level. Such samples should be collected from the locations of interest, including, but not necessarily limited to, a background location and a maximum location.

6.8.2.3 Meat

Due to the time delay for transfer of radionuclides from the point of release through vegetation to beef, pork, and poultry, samples of these meats are not good indicator materials. Therefore, frequent sampling of meat is normally required only when it is necessary to evaluate the radiation doses received via this foodstuff.

With a few exceptions, radiation doses from ingestion of radionuclides in meat are of secondary importance. (One such exception occurs when carbon-14 (C-14) from the facility's effluent is the predominant radionuclide present in the environment. In that instance, the doses from inhalation and external exposure would be small compared to those from ingestion of foods, and also the contribution from milk and vegetables would be less than that from meat.) The preliminary pathway analysis will determine whether frequent meat sampling is required.

Because of the time delay mentioned above, shorter-lived radionuclides (those with half-lives of less than 1 month) are not likely to be present in measurable concentrations in meat samples. The additional time lag (about 2 weeks for cattle and a few days for poultry) imposed between slaughter and delivery of the meat to retail outlets can be avoided by sampling directly at local farms or slaughterhouses. However, this time delay should be accounted for when the analytical results are used to calculate radiation doses from consumption of commercially available meat.

A 1- to 2-kg sample of meat is usually sufficient for analysis. Meat may be purchased from local farms, retail stores, or slaughterhouses with confirmation of local origin. All samples should be placed in plastic bags, sealed, and properly labeled before delivery to the analytical laboratory. Meat samples collected at farms or slaughterhouses should be reduced to edible portions in a manner similar to commercial and home preparation before analysis.

It should be noted that concentrations for several of the radionuclides of interest are generally lower in pork than in beef, despite the fact that many of the radionuclide concentration ratios (pCi/kg meat per pCi/kg feed) are somewhat higher for pork than for beef. The concentrations reflect the fact that the consumption rate of feed by swine is about 20 to 30 percent that of beef cattle. Similarly, the radionuclide concentrations in chickens are generally lower than those in pork because chickens have a much lower feed-consumption rate than swine.

6.8.2.4 Eggs

Under certain circumstances, eggs may make a contribution to radiation doses received from terrestrial foods. The preliminary pathway analysis will determine whether frequent sampling and analysis of eggs are required or whether annual sampling is sufficient. Eggs collected from small local farms where the chickens are free to range over open soil are more likely to contain detectable amounts of effluent radionuclides than eggs from large poultry farms where the hens are confined. As with other foods, it may be difficult to determine the origin of commercially purchased eggs. Do not purchase eggs for analysis unless their local origin is confirmed.

One dozen large eggs, which have a combined weight of about 600 to 700 grams (without the shells), is normally a large enough sample for analysis. Analysis should be done on the whole egg (without the shell). It is not necessary to analyze the yolk and white separately. Analytical results from local farm eggs, when available, should be used for estimating potential individual dose for the farming locations, while those from commercial eggs should be used for estimating collective dose to the affected population.

Several elements have relatively high concentration ratios in eggs (pCi/ kg egg per pCi/day intake) including isotopes of phosphorus, rubidium, iodine, calcium, cesium, barium, tellurium, copper, iron, cobalt, and nickel. Many radionuclides of these elements have such short radioactive half-lives that they would not be detectable in eggs. In addition, some of the radionuclides would not likely be present in the effluents from most DOE sites. Cesium, iodine, and barium could be present in both liquid and gaseous effluents from many different types of facilities. Phosphorus-32 and phosphorus-33, and iron, cobalt, and nickel could be released as activation products with liquid effluents from operating nuclear reactors.

6.8.3 Game Animals

At some sites, game animals are components of the diets of some individuals. Hunting of indigenous game (e.g., deer, small mammals, game birds) is permitted at or around several DOE sites. The practice of allowing hunting and harvesting of deer and other game is part of an ecosystem approach to local wildlife management and conservation efforts to restore and sustain the health, productivity, and biological diversity of a well-balanced ecosystem. Reptile species (e.g., turtles and alligators) that are categorized as game should also be considered if a pathway exists.

A review of the hunting habits in the local area should be included in the preliminary pathway analysis to determine if such game is an important part of the diet of the local population or of hunters from outside of the region. If the results of the preliminary survey indicate that local game could make an important dose contribution, then a more detailed survey of the amounts of each type of game harvested and the disposition of the meat should be made and documented.

It is also important to determine whether the meat is eaten, and if so, whether it is eaten fresh or frozen or given to others. If the results of the preliminary survey indicate that this pathway contributes a TED of less than 1 mrem/yr, then annual sampling and analysis of two or three representative species should be sufficient to determine whether or not this pathway is still insignificant. Radionuclides of interest in wild game are similar to those of interest in dairy cattle. Again, 1- or 2-kg samples should be sufficient for analysis.

Wild game samples can be obtained from wildlife that is trapped, acquired by hunters, or, for larger animals, such as deer, collected after accidental road-kills, or samples can be obtained from an appropriate State agency. Wildlife that is relatively rare locally should not be taken as environmental samples. When sampling deer and other game animals, it is important not to contaminate the meat sample with radionuclides that may be present on the animal's fur or in its gut. Wildlife samples should be kept on ice until transported to the laboratory for analyses.

Where this pathway exists it needs to be considered when demonstrating compliance with the DOE public dose limits and ALARA process requirements of DOE O 458.1. Information pertaining to radiological control and release of game for human consumption is contained in Appendix C of this Handbook.

The following should be considered for DOE sites where this pathway exists:

- The selected approach and level of effort should be commensurate with the importance of the pathway and needs to take into account the degree of uncertainties in dose estimates associated with each approach. Sites and facilities are responsible for documenting and implementing the approach used. The direct measurement approach provides a high level of confidence that the hunting pathway doses comply with DOE requirements.
- The selected approach should provide reasonable assurance that the dose attributable
 to the consumption of game will not exceed the DOE dose constraint of 25 mrem/yr from
 a single pathway and will be well below the public dose limit for all pathways of 100
 mrem/yr.

- Approaches may entail:
 - Process or site-wide knowledge approach;
 - Selective or statistical sampling approach; or,
 - o Direct measurement approach.
- Dose estimates and measurements compared to screening guidelines should be based on edible portions of the animal (e.g., muscle tissue).
- Although gamma measurements alone may be used to estimate alpha and beta
 contributions when radionuclide concentration ratios are generally uniform, if a
 consistent ratio (e.g., Cs:Sr) cannot be demonstrated from available data, then specific
 analyses may be necessary to measure concentrations of the alpha and beta emitting
 radionuclides.
- If potential doses associated with beta and alpha emitters are low (i.e., a small fraction of the gamma emitters) or a small fraction of the dose constraint (whether or not there is variability in the ratios), they need not be measured regularly.
- Tritium may be analyzed in a separated blood specimen and its content in meat estimated by assuming the same specific activity (pCi H-3 per gram of stable hydrogen).

Because strontium concentrates in the bone, strontium may be measured in non-edible tissue for screening purposes. If the concentrations are very low or non-detectable, no muscle tissue need be analyzed. Because it is difficult to estimate muscle concentrations and dose from consumption of the meat from analysis of non-edible tissue, analysis of strontium in edible tissue samples may be necessary only when analysis of the non-edible portions (e.g., bone) indicates the significant presence of the radionuclide.

6.9 Basis for Sampling Soil

Soil provides an integrating medium that can account for contaminants released to the atmosphere, either directly in gaseous effluents or indirectly from re-suspension of onsite contamination, or through liquid effluents released to a stream that is subsequently used for irrigation. Hence, soil sampling and analysis should be used to evaluate the long-term accumulation trends and to estimate environmental radionuclide inventories. In addition to radionuclides that are specific to a particular operation or facility, naturally occurring (e.g., the uranium and thorium decay chains and beryllium-7 (Be-7)) and fall-out radionuclides can be expected in soil samples. The relative importance of these contributors is dependent on site operations and site conditions including site geography, geology, and meteorology.

Radionuclides that are often detected include ³H, cobalt-60 (Co-60), Sr-90, zirconium-niobium-65 (⁶⁵Zr-Nb), Ru-106, Cs-137, cerium-praseodymium-144 (¹⁴⁴Ce-Pr), plutonium-238 (Pu-238), Pu-239, and americium-241 (Am-241). The relative abundance of these materials varies with the source and half-life. Analytical and sample preparation procedures should be tailored to the radionuclides of interest.

As pointed out in Denham et al. (1974), perhaps the greatest diversity among sites occurs in the techniques used for sampling and analyzing soil. Part of this diversity arises from different purposes for soil sampling and analysis (e.g., trend evaluation, projection of future plant uptake, contaminant inventory, and comparison with applicable standards).

Plutonium is one of the most commonly analyzed contaminants in soil (ASTM C998-05(2010)e1). However, there are many limitations of sampling and analysis of plutonium in soil, as indicated in NRC (2007a). Although concentrations of plutonium and other radionuclides in soil are generally readily detectable, the determination of their significance in terms of exposure to humans is less readily quantifiable, except perhaps for the gamma emitters, such as Co-60 and Cs-137. Therefore, it is desirable to assess, document, and periodically reassess the distribution and fate of radionuclides in the environment, especially plutonium in soil samples.

6.9.1 Soil Sampling Location and Frequency

Background determinations should be based on soil sampling and analysis at points corresponding to background (or control) air sampling locations. Where possible, soil sampling locations should be selected to coincide with air sampling stations, since the comparability of data may be important in achieving the objectives of the overall environmental sampling program. Except where the purpose of the soil sampling dictates otherwise, every effort should be made to avoid tilled or disturbed areas, locations near buildings, or areas of unusual wind or precipitation influences when selecting soil sampling locations.

An annual sampling frequency is recommended for long-term accumulation trends. In some situations a lower sampling frequency may be justified but generally should be at least once every three years. The sampling frequency of soil collected for purposes other than long-term environmental accumulation should be based on site-specific source terms and radionuclide half-life, with the purpose(s) and details documented.

6.9.2 Soil Sampling Methods

Several reports are available that provide useful information on sampling, preparing, and analyzing soil for plutonium (L'Annunziata 2003, Ohtsuka et al. 2006, Montero et al. 2000, Lee et al. 2007, ASTM C1001), for radium (GJ/TMC-13 1985; Meyer and Purvis 1985; Myrick et al. 1983; L'Annunziata 2003; IAEA 2010b), and for other radionuclides (ASTM C998-05(2010)e1 and C999-05(2010)e1; Mohrand and Franks 1982). Additionally, Healy (1984) has proposed a standard for comparing observed-to-allowable concentrations of plutonium.

It is recommended that trends in local environmental radionuclide levels be determined through routine soil sampling. Surface soil sampling should be conducted according to methods of NRC 2007a, ASTM C998-05(2010)e1, or HASL-300. Profile depths need to be established. For example, HASL-300 recommends profile depths up to 30 cm to measure the total amount of a radionuclide deposited on the soil, during pre-operational assessment, after a disturbance of the soil, and periodically as needed. Useful information about soil contamination levels can also be obtained using in situ gamma-ray spectrometry. Prior to counting and analysis, soil samples should be homogenized (by grinding and blending, as appropriate in the procedures), and the radioanalytical results reported on the basis of activity per dry weight.

Estimates of individual radionuclide contributions in soil can be made from field spectra, such as those developed by HASL-195, HASL-258, ICRU 1994, Tyler 2007 and reported in NVO-213. The soil concentration estimates depend on distribution of radionuclides with depth, soil density, soil moisture, and chemical composition.

When evaluating the airborne pathway and bioaccumulation in the environment, soil and grassy vegetation samples should be co-located with air surveillance sampling locations.

6.10 Water Sampling

When liquid effluents are released to streams (with continuous or intermittent flow), rivers, or lakes, samples of these surface waters should be collected according to the methods, locations, and frequencies specified in this section or in applicable permits if the releases are projected to result in radiation doses exceeding the criteria given in Table 6-1. Information related to sampling at the liquid effluent release point is provided in Chapter 3. Water sampling frequency and volume of water samples should be chosen to provide adequate sensitivity for the analysis using the general criteria in Table 6-1.

The principal exposure pathways to individuals and/or groups of individuals in the environment from waterborne radionuclides are: (1) consumption of fish, other aquatic species and ducks; (2) consumption of irrigated crops; and (3) ingestion of drinking water. Of lesser significance is external radiation from surface water (swimming, water-skiing, and boating).

Deposits of radionuclides are even more likely from facilities that discharge or have previously discharged liquid effluents to the ground via cribs, pits, or trenches. Routine laboratory analyses of water samples should include those radionuclides that are determined by pathway analyses to represent a significant fraction (e.g., more than 10 percent) of the potential dose from the water pathway. Where documented operating experience and/or system design show that no release (or significant potential for a release) will be made to surface waters that could cause the dose criteria presented in Table 6-1 to be exceeded, this portion of the environmental surveillance program may be reduced accordingly.

Potential for unplanned releases, including those caused by runoff, leaching, flooding, or resuspension should be considered in planning for monitoring.

6.10.1 Water Sampling Locations

The basic recommendations that follow should be applied at all DOE sites where radioactive liquid effluents are discharged to surface streams. Special studies, examining site-specific ground water and surface water flows, may be necessary to establish preferential sampling locations for ponds or lakes. Therefore, detailed hydrological and radiological studies should be conducted for each site on streams, ponds, and lakes to establish the best sampling locations and frequencies to determine radiological doses.

6.10.2 Surface Water

Surface waters can be divided into two basic types: (1) those that are constantly moving (e.g., rivers and streams), and (2) those that are stationary (or not constantly moving) (e.g., ponds and lakes). The type of surface water needs to be considered when specifying surface water sampling location requirements.

Samples should be collected at each location where water is withdrawn for public use.

Representative background samples from surface water sources including rivers, streams, ponds, and lakes should be collected routinely at locations unaffected by site operations.

An investigation should be conducted and documented to show that the surface water source is independent of local influence from site operations.

6.10.2.1 Moving Waters

Background samples provide control data used to compare data from potentially affected sampling locations. Care should be taken to avoid sampling from eddy currents. At a minimum, other offsite sampling locations for surface water should be at the edge of the effluent mixing zone and at the nearest down-current point of withdrawal for domestic or other uses. Continuous or flow-proportional samples may be needed at certain locations.

Multiple sampling points, based on diffusion and transport studies of the mixing zone, may be necessary to obtain a reliable and representative estimate for that location. Sampling at the first downstream point of withdrawal for public use provides an upper-bound estimate of the amount of radioactive material in the water supply (for drinking or irrigation) of the potentially affected population group(s).

For characterization studies, as compared to routine sampling:

- Samples should be taken on a traverse, at more than one depth, and at a number of
 points equidistant across the stream flow such that a representative set of samples are
 taken based on the size of the stream and the nature of the discharge.
- Each sample should represent no more than 10 percent of the total stream flow.
- This sampling strategy may not be applicable for very small streams.
- Traverse studies should be repeated whenever a significant change occurs either in the types or quantities of radionuclides (actual or expected) released or in the flow regime of

the stream (such as from the addition of hydroelectric or flood-control dams).

6.10.2.2 Stationary Waters

Another possible solution is to sample from another nearby pond or lake with the same water source (i.e., fed by the same stream or located within a similar runoff regime). If the **Ground water** may contain detectable or more highly radioactive materials (particularly Tritium) from liquid effluent storage systems (leakage) or discharges to surface water.

Drinking water supplied from any source (surface or ground water) that receives effluents from nuclear facilities is a potential source of radiation exposure to humans.

receiving pond or lake is onsite, an offsite counterpart pond or lake may be used to collect background samples.

Other offsite sampling locations for ponds or lakes should be at the edge of the effluent mixing zone (based on dye or other local transport studies) and at the nearest point of withdrawal for domestic or other uses. Sampling locations near the discharge outfall should be located beyond the turbulent area caused by the discharge. Multiple sampling points, based on diffusion and transport studies of the mixing zone, may be necessary to obtain a reliable representative estimate for that location.

Sampling a lake or pond at the nearest point of withdrawal (i.e., closest to discharge) for public use usually provides an upper bound estimate of the amount of radioactive material in the water supply (for drinking or irrigation) of the potentially affected population group(s). Samples on the traverse or axial sampling lines should be taken at more than one depth and at a minimum of three to five equally spaced points along each of four radials. Traverse or axial studies should be repeated whenever significant change occurs either in the types or quantities of discharges or in the water level of the pond or lake.

6.10.3 Storm Water Runoff

The potential impacts of storm-water runoff as a pathway of exposure to humans or biota should be evaluated. Where radioactive materials in storm water runoff could significantly increase the risk to humans or biota, the water and the receiving ecosystem should be monitored and the doses should be assessed.

Contaminated soil or sediment that could be moved by storm water should be considered for evaluation with RESRAD and RESRAD-Biota for potential impacts on the receiving ecosystem. Wildlife habitats, agricultural land, and intakes to drinking water systems are especially important. Table 6-1 should be used to assess the need for surveillance.

Storm water with significant, visually observable, settleable solids should be filtered, and concentrations in the sediment should be analyzed and reported separately from the filtrate. The concentrations of radionuclides in the sediment should be compared with those at the source of the storm water to determine the amounts from DOE activities. Where the storm water is a potential source for a public drinking-water system, the filtrate may be compared with the 15 pCi/L gross-alpha standard, and the concentrations should be kept ALARA.

Storm water can contain more than one hundred grams per liter of suspended sediment, in which case unfiltered samples are unsatisfactory for the following reasons:

- Many of the standard analytical methods are designed for drinking water with much less sediment than storm water.
- The amount of sediment that can be dissolved using standard methods may vary, leading to inconsistent results.
- Aliquots from a single sample will contain variable amounts of sediment, also leading to inconsistent results.

The 5-pCi/g and 50-pCi/g limits for settleable solids (DOE O 458.1) apply to "liquid discharge," which as defined in Attachment 2 of DOE O 458.1, does not apply to storm water.

When comparing concentrations of radionuclides in the sediment with those at the source of the storm water, the background concentrations provide a "fingerprint" that may be matched with that in the storm-water sediment. For example, naturally-occurring soil may contain ~1 pCi/g of each of the decay products in the uranium and thorium decay chains.

Natural uranium may be distinguished from refined uranium by methods such as process knowledge, isotopic analysis, and by the presence of Pb-214 and Bi-214 in the natural uranium decay chain. Pb-214 and Bi-214 are removed during the refinement process and remain with the mill tailings, together with their parents, Th-230 and Ra-226. The Pb-214 and Bi-214 decay products grow in with a half-life of 75,000 years so they are undetectable in the refined uranium used at many DOE sites.

Global-fallout radionuclides are likely to have higher concentrations than average for the following reasons.

- Fallout was deposited on the surface and usually remains near the surface, so it is preferentially swept into storm water.
- Fallout is mostly brought to earth by rain and snow fall, which are often higher than average where storms occur.
- Fallout often washes off rocky or impermeable slopes and accumulates in low-lying areas and in the path of storm-water runoff.
- Concentrations in ash are especially high because most fallout materials are refractory.

Global fallout may be distinguished from local contamination by methods such as process knowledge, isotopic analysis, and by the ratios of radionuclides, especially Pu-239, Pu-240, Am-241, and Cs-137. Sr-90 tends to be more mobile than Cs-137 but may provide useful indications in some cases. Pu-239 may be distinguished from Pu-240 by mass spectrometry.

Typical ratios of global fallout radionuclides, decay corrected to 2012 are shown in Table 6-3 (derived from UNSCEAR 2000, Annex C, Table 9). Generally, these radionuclides were deposited together and they are bound tightly to the soil matrix so they move together when the soil is physically disturbed. Under some conditions, they can go into solution, in which case the ratios will be different.

TABLE 6-3: Global fallout activity relative to Pu-239, Pu-240

Nuclide	Ratio
Pu- 239,	1.00
Pu-240	
Am-241	0.42
Cs-137	28
Sr-90	18

In solution, the activity ratios are likely to be considerably different from those in the sediment because of different solubility. For example, U-234 concentrations are usually higher than U-238 concentrations because the decay process dislodges the U-234 from the lattice and makes it more soluble (Arndt and West 2004, Eisenbud and Gesell 1997, Kraig and Gladney 2001).

6.10.3.1 Biota

For compliance with the biota dose limits, it may be helpful to consider the data for filtered water separately from the sediment residue because the bioaccumulation factors for soluble and insoluble material are different. For example, the default bioaccumulation factor (B_{iv}) for radium in aquatic animals is 3200 L/kg (DOE 2002a and RESRAD-Biota) but this high value may not be appropriate for insoluble radium in sediment.

Both man-made and naturally-occurring radioactive material should be assessed. However, "high background levels of naturally occurring radionuclides ... may be taken into account when determining compliance of DOE activities with biota dose limits" (DOE 2002a).

6.10.4 Drinking Water

Drinking water may be supplied from surface water sources or from ground water sources.

Thus, the drinking water sampling location requirements are presented according to the type of drinking water source that is available.

The sampling location for drinking water derived from surface water sources should be of the treated water at the point of maximum probable effluent concentration in the surface water.

Samples of untreated water prior to treatment should also be taken to determine any removal by water treatment and to improve the reliability of dose estimates. If surface water sampling and analytical results indicate that the dose criteria given in Table 6-1 are not exceeded, further drinking water sampling is not necessary. Such conditions should be documented and periodically (at least annually) reviewed to determine that the potential doses are still below the criteria in Table 6-1.

The sampling location for drinking water derived from ground water sources should be at the nearest domestically used well down-gradient from the surface (crib, pond, lake, or stream) discharge point. Another well up-gradient from the discharge point should be used for the control or background sample. When comparisons with control wells are conducted, the sampling stations should be located in the same hydrologic unit. If a significant number of wells are used domestically in the vicinity of the site, it may be necessary to sample several wells to determine if drinking water sources are affected by discharges from the site.

6.10.5 Ground Water

Ground water monitoring should be conducted onsite and in the vicinity of DOE facilities. Ground water monitoring should be designed and operated to:

- Obtain data for the purpose of determining baseline conditions of ground water quality and quantity;
- Demonstrate compliance with, and implementation of, applicable regulations and DOE Orders:
- Provide data for the early detection of ground water contamination;
- Identify existing and potential ground water contamination sources and maintain surveillance of these sources; and
- Provide data upon which decisions can be made concerning land disposal practices and the management of ground water resources.

The siting and number of ground water monitoring stations should be governed by the nature of ground water use and the location of known and potential sources of contamination. When possible, existing wells and historical data should be used. In the event that additional wells are necessary, their number and placement should be directly related to contaminant pathways. Quality control in well construction is essential. Predicting contaminant pathways requires a three-dimensional geologic, hydrologic, and geochemical analysis. General guidance for implementation of ground water surveillance monitoring can be found in DOE (2004). Additional

guidance may be found in documents and technical procedures prepared by regional EPA offices and State agencies.

Mechanisms for subsurface contaminant dispersal are not fully understood. The rate and extent of contamination are influenced by:

- Characteristics of the source of contamination;
- Nature of the geologic formations in the saturated and unsaturated zones;
- · Physical and chemical properties of the contaminants; and
- Phenomena that affect the fate of a contaminant which include capillary action, decay, adsorption, dispersion, and diffusion.

No comprehensive Federal statutes regulating ground water quality and monitoring currently exist. Ground water requirements are drawn from a number of distinct laws enacted to protect other resources or to regulate specific sources of contamination.

In addition to Federal statutes that authorize programs and activities for ground water protection, many States also are developing and implementing ground water policies, statutes, and strategies. Often States have the authority or "primacy" to administer several Federal environmental laws. Under this authority, States may, and often do, impose more stringent requirements than the Federal government. In many States, State agencies, regional authorities, and local governments share responsibilities for protecting ground water.

Contaminants covered by ground water quality standards vary from State to State. It is important that DOE Federal and contractor staff work closely with State and regional agencies when determining the specific monitoring requirements for each DOE facility.

6.10.6 Water Sampling Methods

Since most water measurements are made on samples taken in the environment and sent to laboratories for analysis, the two major concerns in water sampling are: (1) the collection of a representative sample, and (2) the maintenance of radionuclides in their original concentrations before analysis. Kahn (1972) discusses the general problem of the measurement of radioactive material in environmental water. Water sampling procedures are discussed in APHA (2012) and EPA (1983).

6.10.6.1 Water Sample Collection

Waste management practices often result in periodic or batch discharges of liquid wastes, rather than a continuous release.

The following factors should be considered when selecting water sampling equipment:

- Probability of significant fluctuations in concentration of the water sampled;
- Potential for significant human impact (dose);
- Potential for contaminating the environment; and
- Applicability to radionuclide(s) of interest.

The recommended practice for surface water samples is automated continuous sampling followed by analysis of the unfiltered sample. When the data are to be used for dose calculations, the method should use a fixed-time sampling frequency. If the data are to be used for radionuclide transport or inventory purposes, these samples should be taken with timing proportional to flow rate where practical.

When circumstances prohibit this type of automated continuous sampling (e.g., power restrictions, prohibitive pumping requirements, freezing temperatures, etc.), compositing should be performed by manual collection on a frequency based on effluent release and on information on the receiving body of water. An acceptable scheme is weekly grab samples of surface water composited for monthly analyses and daily grab samples of drinking water composited for weekly or monthly analyses.

Because the flow of most ground water systems is on the order of centimeters to meters per day (compared with tens or even hundreds of kilometers per day for surface stream flows), periodic grab sampling of ground water should be sufficient. Unless circumstances prohibit, ground water grab sampling should be done by pumping. Examples of pumps include pressure air lift, submersible pump and variable speed pump. In any case, the pump should be operated for a length of time sufficient to obtain a representative sample of water in the aquifer. In some cases, other innovative methods may be used, such as diffusion sampling and bailing. Diffusion sampling methodology is discussed in EPA (2009a).

To approximate conditions at the tap, finished drinking water conditions may require filtering of ground water samples to remove well-casing effects.

NOTE: For pre-analytical handling of water samples, it is common practice to well-mix all environmental water samples prior to splitting or transferring to holding containers (APHA 2012, Standard Methods for the Examination of Water and Wastewater, 22nd Edition, Section 1060, *Collection and Preservation of Samples*). Before some analytical procedures however, the sample is allowed to settle and then decanted to not include the settled fraction in the analysis. That fraction is not added to the water totals, but would be considered a "sediment" sample (APHA 2012, Section 2560, *Particle Counting and Size Distribution*). For some analyses (gamma spec and radiochemical isotopic) analytical steps incorporated into the methods eliminate interferences in nonhomogeneous samples. Therefore, the entire fraction of the mixed sample, including any sediment, can be considered as a "water sample," thus providing a conservative approach. (APHA 2012, Section 7010).

6.10.6.2 Sample Size

The size of water samples will be determined by the analytical procedures (see Chapter 7) to be used. A 3.5-liter (approximately 1 gallon) sample is usually adequate other than for tritium or gross activity measurements. The sample volume needs to be increased where splitting of samples for replicate analysis or individual radionuclide determinations is planned.

6.10.6.3 Representative Water Sampling

Natural waters are frequently two-phase systems (i.e., solid materials are suspended in, or floating on, the water). Therefore, all surface water samples should be carefully taken from beneath the water surface to avoid floating debris and any bottom sediments or growths. The soluble fraction provides an indication of possible stream transport, while the insoluble fraction can be used as an indication of potential sedimentary material. So that data are comparable, both fractions, if analyzed, should be added in reporting the total concentration.

Filtration of ground water samples is recommended because suspended material is usually an artifact of the sampling process (well-casing particles and dirt near water-soil interface) and is not representative of the ground water. Caution should be exercised to prevent water samples from different locations being cross-contaminated by reuse of sampling containers. When obtaining surface water grab samples, the sample container should be rinsed twice with the water being sampled before the actual sample is taken. When extracting aliquots from a larger

water sample, extra effort should be taken to provide that the aliquot is representative of the entire sample.

6.10.6.4 Water Sample Preservation

Continuing biological and chemical action in the sample during and after collection can cause changes in chemical form, deposition on container walls, and removal of radioactive material from solution by biological growths.

Known phenomena include the following:

- Cations, at very low concentrations, can be lost from solutions (e.g., cesium can exchange with potassium in the container (glass));
- Radionuclides can be absorbed by algae or slime growths in sample lines or on container walls, especially in sample containers that remain in the field for extended periods;
- Hydrolysis and sorption on container walls or on particles in the water can occur at low acidities (typical of many natural waters);
- Radio-colloidal phenomena may result in large flocculent particle formation or additional plate-out on container walls;
- Pretreatment may induce change in nuclide distribution (e.g., acidification can leach suspended particles in the original sample so that more radioactive material appears in solution);
- Acids used as biocides can oxidize iodide to iodine, resulting in its volatilization;
- Acids may guench standard liquid scintillation cocktails; and
- A change in counting geometry may occur for gamma-ray counting if finely divided particulate activity settles out or if soluble species become fixed on the container walls during counting.

EPA (1983), the Environmental Measurement Laboratory (EML) Procedures (HASL-300), the MARLAP Manual, and the Radiological and Environmental Sciences Laboratory procedures (DOE 1982) provide useful information on sample preservation, storage, and analysis methods. Radioiodine analyses should not be performed on an acidified sample because organic forms may be transformed to elemental forms that are more volatile.

6.10.7 Settleable Solids in Effluent Discharge

DOE O 458.1 requires that the radioactivity in the settleable solids in liquid discharge streams be limited to 5 pCi/g above background for alpha-emitting radionuclides, and to 50 pCi/g above background for beta-gamma emitting radionuclides.

The following method should be used to determine the radioactivity of settleable solids:

- Use the gravimetric test (APHA 2012, Section 2540 F, Settleable Solids⁵) to determine settleable solids in mg/L in the water sample. The gravimetric test method in Section 2540 F, 3.b, may be used to determine both the total suspended solids and non-settleable solids. The solid fractions of the total suspended solids and non-settleable solids samples should be retained for later radioactivity measurements.
- Determine the radioactivity of alpha-emitting radionuclides in pCi/g and the radioactivity
 of beta-emitting radionuclides in pCi/g in the recovered solid fraction of each of the total
 suspended solids and non-settleable solids samples.
- Determine the gross activity concentration of the settleable solids, using information obtained above and the equation:

$$A_{SS} = \frac{(M_{SS} \times A_{TSS}) - (M_{NSS} \times A_{NSS})}{M_{TSS} - M_{NSS}}$$

Where

 A_{SS} = activity concentration of settleable solids, pCi/g

 M_{TSS} = mass concentration of total suspended solids, mg/L

 A_{TSS} = activity concentration of total suspended solids, pCi/g

 M_{NSS} = mass concentration of non-settleable solids, mg/L

 A_{NSS} = activity concentration of non-settleable solids, pCi/g

- Since the sedimentation standard is presented as net settleable solid radioactivity, the
 activity of background settleable solids needs to be subtracted from the sample
 settleable solids activity.
- Determine the background radioactivity from an appropriately selected background water sample, using the same methods and equation.

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⁵ In some older editions of the APHA *Standard Methods*, discussion of *Settleable Solids* was identified as Section 209 E.

The settleable solids requirements in DOE O 458.1 are standards based. If using the method prescribed in the Order, settleable solids are not detected or the quantity of solids is so small that the radionuclides cannot be detected, the requirements are satisfied. Direct environmental monitoring of sediments, under the site environmental monitoring plan or other procedure in use, will further verify that radionuclides are not accumulating or increasing due to changing conditions.

6.11 Basis for Sampling for Aquatic Foodstuffs

Aquatic foods, including local fish, shellfish, and waterfowl, are eaten in relatively large quantities by residents of some regions of the country.

Aquatic plants are not normally a component of the human diet in the United States. However, there are exceptions; for example, along the California coast a particular species of seaweed is harvested and processed into a thickener for foods, such as milkshakes. Aquatic plants can be vectors in the water-plant-animal-human pathway.

If the preliminary analysis indicates that the potential annual TED from ingestion of aquatic foods is 5 mrem or greater, then sufficient sampling and analysis should be carried out to provide that the foods and radionuclides contributing at least 90 percent of this ingestion dose have been evaluated.

If the potential annual TED is between 1 and 5 mrem, then sufficient sampling and analysis should be carried out to provide reasonable assurance that the doses are in this range.

If the annual TED is potentially between 1 and 0.1 mrem, then sufficient surveillance should be done to show that the radionuclides are behaving in the environment as expected.

Only one generic concentration ratio for aquatic organisms (pCi/kg organism per pCi/L water) is less than 1; namely, 0.5 for uranium in marine plants. As a result, any radionuclide present in the water will be present in aquatic organisms, and most, but not all, radionuclides detectable in water will be present at detectable concentrations in the organism.

Aquatic animals, sediments, and other predictive environmental media should be sampled and analyzed at least annually to demonstrate compliance with the DOE O 458.1 requirements for

protection of biota. DOE (2002) provides practical screening and analysis methods for demonstrating compliance with the requirements for protection of biota. DOE (2002) and the RESRAD-BIOTA⁶ code are the preferred tools for estimating and evaluating doses to biota, unless there are site-specific requirements that necessitate the use of an alternative method or model, or it is determined that such alternate approaches will provide better results.

The sampling program should be determined on a case-by-case basis considering such factors as the estimated dose as determined from measured concentrations in organisms or predictive environmental media in comparison with the limit and any variation behavior of the contaminants involved.

Special permits from State fish and wildlife agencies are usually required for fish, shellfish, and waterfowl sampling for monitoring purposes.

6.11.1 Freshwater Foods

Concentrations of many elements in fresh water are highly site-dependent. This variation can affect the observed concentration ratios of radionuclides of these or biologically similar elements in freshwater organisms. (Except in estuaries, the elemental composition of seawater is relatively constant, and the concentration ratios of radionuclides in marine organisms are not nearly as site-dependent as they are for freshwater organisms.)

If the aqueous effluents are discharged into a surface body of fresh water (pond, lake, stream), then the background sampling point should be far enough from the discharge point for radionuclide concentrations in the water and sediment to be unaffected by the effluents.

The indicator sampling location should be downstream of the discharge point(s) at a location in which the water is determined to be well mixed (e.g., based on water-sample traverses). In choosing the locations to be sampled, consideration should be given to the possible migration of fish between upstream and downstream locations.

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⁶ The RESRAD-BIOTA code and its User's Guide serve as companion tools to DOE-STD-1153-2002 (DOE 2002a) for evaluating doses to biota.

6.11.1.1 Fish

The species of fish likely to contain the highest concentrations of radionuclides are those that feed at or near the bottom and do not migrate very far from the places having the highest water or sediment concentrations. These species are useful as indicator organisms for monitoring trends in aquatic contamination levels. However, they may not always be the ones that are consumed at the highest rate by the local population. Studies of fishing pressure and fish consumption, coupled with preliminary radiochemical analysis of the different types of available fish should be used to define the proper species to monitor for the purposes of dose calculation.

Fish can be collected by using nets, rod and reel, or other methods (e.g., slat traps, shocking), or they can be purchased from commercial sources if their origin can be determined. For use in dose calculations, the edible portions of the fish consumed by humans should be analyzed. In most instances that includes only the muscle. However, the whole fish should be analyzed if it is used for preparation of fishmeal or fish burgers. Fishbone should be analyzed when part of the local diet includes boiling the fish such as the preparation of catfish stew. It is also appropriate to analyze the whole fish when the data are used for trend indication. If fish are the critical pathway, then they should be analyzed by species. If the results are to be used as trend indicators, then the fish may be grouped by type for analysis (e.g., bottom feeders, insectivores, or predators).

The following factors should be considered when determining the frequency of sampling:

- Variability of the radionuclide release rates;
- Seasonal variations in the feeding habits of the fish and in the availability to consumers;
 and
- Where the freshwater habitat includes a flowing stream, the variability in the stream flow rate.

Radionuclides of potential interest in fish include H-3, phosphorus-32 (P-32), phosphorus-33 (P-33), Zn-65, cesium-134 (Cs-134), and Cs-137. Although the concentration ratio for ³H is only 1, it is often present in high concentrations in aqueous effluents. Strontium-90 (Sr-90) might be of importance in samples of whole fish since it concentrates mostly in bones. Phosphorous (P-32 and P-33) concentrates in fish flesh as well as in bones.

The sample size required for analysis will vary from 1 kg to several kilograms, depending on the specific radionuclides being measured and their concentrations.

6.11.1.2 Shellfish

Shellfish include mollusks, which live in or on the sediment, and crustacea, such as freshwater crayfish, which live on or near the bottom. Decisions on sampling locations and frequencies involve the same types of considerations as discussed above for fish (i.e., variability of radionuclide concentrations in water and sediment and inclusion of upstream and downstream locations).

Freshwater shellfish are usually not a significant diet item. However, they may be eaten by some individuals in certain specific regions of the United States. A preliminary pathway analysis will determine if shellfish are a potentially important contributor to the TED that might be received by residents of the region.

Radionuclide concentration ratios are generally higher in invertebrates than in fish, and in some cases significantly higher. Radionuclides of potential interest in freshwater mollusks and crustacea include: P-32, P-33, cobalt-58 (Co-58), Co-60, Zn-65, Sr-90, Ru-106, and the rare earth radioelements.

A 1- or 2-kg sample is normally sufficient for analysis. Samples of shellfish may have to be purchased commercially to avoid the difficulties associated with field collection.

6.11.1.3 Waterfowl

Waterfowl, such as ducks and geese, may acquire radionuclides from their food sources. Some species are bottom feeders and tend to accumulate those radionuclides associated with sediments, such as Co-60, Zn-65, and Cs-137. Others feed predominantly on surface plants, insects or fish. Depending on the specific diet, these species may accumulate P-32, P-33, Zn-65, Sr-90, and Cs-137.

The migratory habits of waterfowl species vary widely. Some may be year-round residents of the local waterways (and effluent ponds). These are usually species that are less desirable to hunters. Others may migrate long distances, and the limited amount of time spent in the local area may not be enough to cause significant contamination of their flesh. Because of these variables, it is often difficult to predict which species is most important in terms of potential exposure to local hunters.

The preliminary pathway analysis should include consideration of the amount of waterfowl hunting, if any, in the local area and the number of birds shot. It should be remembered that

even though some individuals may harvest a relatively large number of waterfowl, the collective dose to the local population from waterfowl consumption may still be small. If the potential TED is significant, a minimum of two or three birds of each type (bottom feeders, plant eaters, and fish eaters) should be sampled during hunting season.

The most common method of collecting waterfowl is by hunting. Sampling of non-migratory, non-game species can occasionally provide useful information on contamination trends.

During preparation of the samples for analysis, care should be exercised not to contaminate the edible portions with radionuclides present on the external surfaces of waterfowl. Analysis should include the radionuclides listed above plus any others that prove to be of special concern at a specific site.

6.11.2 Marine Foods

Sites that are located on the seacoast, an estuary, or a river upstream of an estuary should include consideration of the potential consumption of contaminated marine foods, such as sports and commercial fish and shellfish, in their preliminary pathway analysis. Considerations discussed for sampling of freshwater aquatic foods also apply to marine foods. These considerations include sample size and radionuclides of potential interest.

Sports fish and shellfish will be of interest primarily for calculation of radiation doses to the MEI, while commercial seafood is of interest for estimating the collective dose. It is important to document the origin of the commercial samples. It may be necessary to track the path of an effluent plume or contaminated river for many miles along the seacoast to identify the important locations for shellfish sampling. Arrangements can usually be made to buy seafood harvested at known areas from local packing houses. Certain marine fish, such as salmon and tuna, which migrate over large areas of the ocean, will not normally be measurably contaminated from aqueous effluents discharged along the shore or reaching the coast line. If fish are found to be contaminated, it might be difficult to determine the exact source of radionuclides detected in them.

6.12 Basis for Sampling Sediment

The basis for sampling, location and frequency, and sediment sampling methods are considerations that need to be included when establishing the process to sample sediment. The sampling of sedimentary material from streams or ponds can provide an indication of the accumulation of undissolved radionuclides in the aquatic environment. The accumulation of

radioactive materials in sediment can lead to exposure of humans through ingestion of aquatic species, through sediment resuspension into drinking water supplies, or as an external radiation source irradiating people fishing, wading, or sunbathing. Hence, the sampling and analysis of sediment, or the measurement of the external radiation emanating from the sediment, provide indications of the potential for human exposure from these indirect pathways.

Because of the accumulation of contaminants, sediment sampling is a more sensitive indicator of waterborne radionuclides than water sampling or, for some aquatic species, aquatic biota sampling. This sensitivity is especially true for radionuclides that are not significantly accumulated by fish or shellfish. Sediment sampling is particularly appropriate for most of the transuranics (especially Pu-239); such activation products as Mn-54, Co-58, Co-60, and Zn-65; and several fission products such as zirconium-niobium-95 (Zr-Nb-95), Cs-134, and Cs-137. Radionuclide concentrations may vary for a given location based on many factors including the chemistry of the radionuclides and the characteristics of the sediment and water (NCRP 2010) and the potential uncertainties recognized when these data are used in dose assessments.

6.12.1 Location and Frequency of Sediment Sampling

The need for sediment sampling and the choice of locations and frequency should be based on site-specific evaluations. These evaluations should consider the potential for offsite exposure of humans as well as the potential dose to onsite or offsite aquatic organisms.

Sediment samples are normally taken to detect the buildup of radionuclides by sedimentation. Sediment sampling locations should be based on the type of surface water receiving site liquid effluents. For moving bodies of water, such as streams or rivers, sediment sampling locations should include an upstream site beyond any possible facility influence and two downstream locations. The two downstream locations should be located such that one is near the discharge site and the other is in an area that favors sedimentation, such as the inner bank of a bend in the stream or river (EPA 1972), the region of a freshwater-saltwater interface, or at a dam impoundment. If liquid effluents from a DOE facility are discharged to a lake, pond, or arroyo, a sediment sample should be taken near the outfall but beyond the turbulent area created by the effluents. Because sediments are usually not in a critical exposure pathway, an annual frequency for sediment sampling should be sufficient.

For rapidly moving streams (e.g., rivers), sediment sampling should be considered in conjunction with the spring freshet (i.e., just before or just after) if one occurs locally. For arroyos, the sampling should take place after cessation of water flow (i.e., upon first drying in

the spring). For ponds or lakes, the timing of sediment sampling should be considered on a site-specific basis, but normally at about the same time each year.

6.12.2 Sediment Sampling Methods

Samples of deposited sediments in water can be collected manually (by hand in shallow water or by diving in deeper water) or mechanically (by dredge or with a core sampler). The manual methods are recommended where conditions permit, because the location and depth of the sample can be well-defined. The dredge and coring methods use a sampling device dropped from a boat that is activated when the device contacts the sediment (benthos). Three types of dredges commonly are used: Petersen dredge, Ponar dredge and Eckman dredge (NCRP 2010).

Except for cases where inventory estimation is desired, representative surface (top 5 to 10 cm) sediment samples should be collected along with water depth and stream flow (or pond/lake elevation) data at the time of sampling. Characteristics of the sample, such as particle-size distribution, sediment type, stream type (i.e., intermittent, creek, pond, river, reservoir, etc.), ion-exchange capacity, and organic content, may be useful for proper interpretation of the analytical results.

Every few years, samples (e.g., dredge or core) should be taken in areas in which sediments have been most heavily deposited to determine the profile of the historical depositions and to determine trends and changes in control of effluents and their impacts.

All sediment samples should be oven-dried, homogenized (by grinding and blending, as appropriate in accordance with procedures used) and the radio-analytical results reported on the basis of activity per unit dry weight (g or kg). To prevent cross-contamination, thorough cleaning of equipment between samples is necessary. Portions of the detailed EML procedures (ASTM C999-05(2010)e1) for preparing soil samples for analysis are equally applicable to sediment samples.

Disturbance of the sediment due to sampling activities generally can be reduced by moving slowly and always approaching the sample location from downstream (moving waters) or downwind (stationary water) (NCRP 2010).

6.13 Quality Assurance

As they apply to environmental surveillance activities, the general QA provisions of Chapter 11 should be followed.

7 SAMPLE HANDLING, PREPARATION, AND ANALYSIS PROCEDURES

The establishment of good sample handling, preparation, and analysis procedures is vital to obtaining quality results from samples collected under the effluent monitoring and environmental surveillance program. Laboratory procedures should be documented in the site environmental monitoring plan or other documentation describing the environmental monitoring. A lines of inquiry approach is provided in Appendix B to conduct self-assessments and verify that the program is effective and in compliance with the appropriate requirements and to ensure continuous improvement of the program.

7.1 Key Requirements and Supporting Documents

DOE O 458.1, *Radiation Protection of the Public and the Environment*, requires demonstration of compliance with the public dose limit using a combination of documented surveys, measurements and calculations to evaluate potential doses.

The Multi-Agency Radioanalytical Laboratory Analytical Protocols (MARLAP) Manual, is a multi-agency consensus document developed to provide guidance for project planners, managers, and laboratory personnel to ensure that radioanalytical laboratory data will meet a project's or program's data requirements. The MARLAP Manual offers a framework for national consistency in the form of a performance-based and graded approach for meeting project- or program-specific requirements. The MARLAP Manual includes guidance for handling, preparing, and analyzing laboratory samples, as well as additional guidance for a number of the topics addressed in this chapter of the Handbook.

The DoD/DOE Consolidated Quality Systems Manual (QSM) for Environmental Laboratories (2013) is a manual incorporating quality systems of both DoD and DOE. It is based on Volume 1 of The NELAC Institute (TNI) Standards (September 2009) which incorporates ISO/IEC 17025:2005, General Requirements for the Competence of Testing and Calibration Laboratories. QSM Checklist 4, Data Quality for Radiochemistry Analyses, is a checklist used by DOD and DoD during audits of the laboratories. The checklist is an excellent reference for laboratories to use to ensure their quality system meets the needs for both DOE and DoD.

7.2 Summary of Laboratory Procedure Requirements

A DOE site does not need to maintain a full laboratory, but it does need to have the necessary laboratory capabilities available to it. At a minimum, the following capabilities should be available.

7.2.1 Documentation

To ensure that the analyses performed are consistent and of the highest quality, site-specific policies, programs (e.g., Environmental Monitoring Plan, Environmental Radiological Protection Program) and standard operating procedures should be developed to promote and provide consistency in the handling, preparation and analysis of laboratory samples. Specific methods for handling, preparing, and analyzing laboratory samples should be identified based on the sample matrix, sample activity, and radionuclide of interest. These methods should be documented and used to identify and quantify all radionuclides in the facility inventory or effluent that contribute 10 percent or more to the total effective dose to individual members of the public from DOE operations.

When available, standard analytical methods (DOE, EPA, ASTM, etc.) should be used for radioanalytical analyses. Any modifications of standard methods or deviations from the procedures should be documented appropriately, along with demonstration of capability data. Additionally, sample handling, sample preparation, analytical methods, data requirements, and other necessary documentation should be specified in analytical contracts.

7.2.2 Sample Identification System

Each monitoring and surveillance organization should have a sample identification system that provides positive identification of samples and aliquots of samples throughout handling, preparation, analytical and data reporting processes. The system should incorporate a method for tracking all pertinent information obtained in the sampling and analysis processes.

7.2.3 Chain-of-Custody

The possession and handling of samples need to be traceable at all times. A sample chain-of-custody should be used to document sample possession and to demonstrate that the sample was maintained in a controlled and unaltered state. Sample custody should be assigned to one individual at a time. A sample is considered in custody if it is:

In the physical possession of the assigned individual;

- Remains in view of the assigned individual;
- Placed in a locked area or sealed to prevent tampering; or
- Is placed in a secure, controlled access area.

Chain-of-custody should be documented for all samples, data, and records used to demonstrate compliance. The chain-of-custody record may be a standardized form initiated by the individual collecting or overseeing the collection of samples and accompanies the samples throughout the handling, storage, transportation, analysis, and disposal processes. Any break in custody or evidence of tampering needs to be documented and investigated.

7.2.4 Screening of Samples

Environmental samples should be initially monitored (screened) to determine activity levels and to detect transferable contamination before transfer to an analytical laboratory to prevent contamination of materials and equipment with which they may come in contact. Samples with elevated screenings should be marked, along with the chain-of-custody.

7.2.5 Preventing Cross-Contamination

To prevent incorrect analysis results caused by the spread of contamination among samples, each laboratory should establish and adhere to written procedures to minimize the possibility of cross-contamination between samples. High-activity samples should be kept separate from low-activity samples to minimize the potential for cross contamination.

7.2.6 Sample Preservation

It is essential to maintain the sample integrity after collection to preserve the chemical and physical state of the sample, including the radiological constituents in the sample. The sample preservation practices should minimize degradation of the samples prior to analysis by using proper preservation and handling practices that are compatible with the sample matrix and analytical methods to be used. Examples of preservation practices include, but are not limited to:

- Biological samples should be kept frozen until they are processed.
- Water samples should have a small amount of acid (2-5% v/v HNO₃ is typical) added to inhibit biological growth and to prevent analyte hydrolysis, causing adherence to the sample container wall. However, acid should not be added if the radionuclide of interest is volatile in acidic solutions (e.g. tritium, C-14, and radioiodine).

- Water samples should be filtered if the analytical request is for dissolved solids. Water samples should not be filtered if the analytical request is for total analytes (dissolved plus not dissolved). Refrigeration should be used when necessary to prevent biological growth.
- Shielding from light may also be used to prevent chemical changes due to ultra-violet (UV) light exposures.
- Soil samples do not require special sample preservation. However, volatile radionuclides, such as H-3, C-14, and Tc-99, should not be dried prior to sample analysis.

7.2.7 Sample Packaging and Transportation

Samples that are sent offsite for analysis or for laboratory inter-comparison should be packaged, labeled, marked and transported in a manner that meets applicable transportation regulations and requirements. Samples that have been preserved with acids may be considered hazardous substances under RCRA and they should be packaged, labeled, marked and transported accordingly. Samples that show measurable surface contamination should be repackaged into uncontaminated containers before they are brought into the laboratory to prevent the spread of contamination or the loss of sample constituents.

7.2.8 Sample Handling

All samples for analysis should be handled in a manner that conforms to Section 7.2.5. of this chapter. All pertinent sample information and associated analysis should be recorded in a permanent laboratory record (e.g., logbook and/or computer system with backup). The sample identification number should enable tracking of the exact location of the record entry or computer file and indicate the chain-of-custody for the samples. Table 7-1 presents a summary of matrix-specific sample handling and preparation considerations as provided in the MARLAP Manual.

7.2.9 Sample Preparation

Sample preparation is a critical step for achieving quality analytical data and the type of sample preparation depends on the nature of the sample matrix and the radionuclide(s) of interest. Sample preparation is the physical manipulation of the sample to the point of chemical separation. Examples of sample preparation, depending on the sample matrix, include, but are not limited to: drying, screening, grinding, mixing, ashing, and acidifying. Whenever possible,

analytical methods that do not require complex sample preparations should be selected to avoid measurement uncertainties that can result from radionuclide losses during sample preparation. Carriers and/or tracers should be introduced as early as possible in the sample preparation procedures to facilitate adequate estimation of any loss of contaminants and the recovery efficacy (i.e., chemical yield) of the preparation procedure. Caution should be used when preparing soils, sediments, and biological materials (i.e., wet/dry washing, drying etc.) to prevent contaminant losses from volatilization, formation of dust or airborne particulates, or unexpected reactions such as combustion, foaming, and splattering.

Samples obtained for tritium and/or C-14 analysis should be sealed in airtight containers to prevent exchanges of H-3 and/or CO₂ vapor with atmospheric air. Biological samples selected for tritium and/or C-14 analysis may be preserved frozen (or refrigerated, at a minimum) to limit sample degradation if the sample is not analyzed immediately upon collection. Prior to analysis, tritium samples may require purification to eliminate contributions from environmental contaminants. For example, purification is necessary if: (1) color is visible, (2) organics are present, or (3) the sample or source history is unknown.

7.2.10 Instrumentation

All sites that release or could release radionuclides should have the capability—either internal to the organization or external—to analyze routine, special, and emergency samples to identify and quantify the radiological contaminants in a sample matrix of interest.

The instrumentation selected for assessing radiological contamination (screening and laboratory-based analyses) should be calibrated periodically in accordance with applicable procedures and national or international standards. Tables 16-19 of the QSM have the recommended calibration frequencies for laboratory instrumentation. At a minimum, the frequency recommended by the instrument manufacturer should be used as a starting point for periodic instrument calibrations. Additionally, instrument performance verifications (e.g., background counting, source checks) should be periodically performed on all instruments used to assess contamination or to identify and quantify radioactive materials to verify that the instrument(s) provide quality results meeting program- or project-specific requirements.

7.2.11 Laboratory Qualifiers

Laboratory personnel should recognize and respond to issues affecting the quality of sample results related to data validation (refer to Chapter 8). Examples include:

- Statistical uncertainty is too high to be accepted by the analyst;
- Radionuclide has no supporting photopeaks to make a judgment;
- Photopeak resolution is deemed unacceptable by the analyst;
- Result is below the decision critical level;
- Other radionuclides display gamma-ray interferences;
- A graphical display of analyzed photopeaks shows unacceptable fitting results;
- There is no parent activity, therefore the state of equilibrium is unknown and the radionuclide could not be quantified;
- Evidence of laboratory cross-contamination or quality control issues.

7.3 Uncertainty

The error of a measurement can be defined as the difference between the measured result and the actual value of the measurand (MARLAP). The measurement error is a result of systematic and random effects. The measurement error is a theoretical concept that implies knowledge of the actual value of the measurement (but in reality is unknown). Therefore, the International Organization for Standardization (ISO)/International Electrotechnical Commission (IEC) Guide 98-3:2008, *Guide to the expression of uncertainty in measurement (GUM)* (2008), and MARLAP suggest the term "uncertainty of measurement to denote a parameter associated with measurement results that characterizes the dispersion of the values that could be attributed to the measurand." In other words, the uncertainty of a measurement is a value used to estimate the size of the measurement error; the smaller the uncertainty, the smaller the error.

7.3.1 Estimation of the Measurement Uncertainty

Uncertainty associated with any measurement result is unavoidable due to introduction of systematic and random errors. The uncertainty of a measurement is an important parameter to be reported together with the measurement result. For example, radioactive decay and the measurement method (the combination of the selected instrumentation and measurement technique) are potential sources of uncertainty of a measurement. The larger the uncertainty of a measured value, the lower the probability that the measured value is close to the true value it represents.

TABLE 7-1: Common Matrix-Specific Analytical Planning Issues

Sample Matrix	Recommended Issues	Key Issues
Solids (soil, sediment, structural material, biota, metal, etc.)	Homogenization	Sample identification
	Subsampling	Container type
	Removal of unwanted material	Container material
		Sample preservation
		Surveying samples for health and safety
		Volatile compounds
		Sample identification
		Cross-contamination
		Sample size
		Compliance with radioactive materials license
		Compliance with shipping regulations
		Chemical and physical form of the substrate
Liquids (drinking water, ground	Is filtering required?	Sample identification
water, precipitation, solvents, oils, etc.)	Sample preservation	Volume of sample
	Should sample be filtered or preserved first?	Immiscible layers
		Precipitation
		Total dissolved solids
		Reagent background
		Compliance with radioactive materials license
		Compliance with shipping regulations
Filters and Wipes	Filter material	Sample identification
	Pore sizeSample volume or area wiped	Compliance with radioactive materials license
		Compliance with shipping regulations
		Subsampling
		Background from filter material

(Information from Table 3.1 of the MARLAP Manual)

The laboratory should report each measured value with either its combined standard uncertainty or its expanded uncertainty (MARLAP). Estimating the combined standard uncertainty of a measurement can be accomplished by propagating the standard uncertainty of the individual components of the measurement. Based on information provided in NRC Regulatory Guide 4.16, *Monitoring and Reporting Radioactive Materials in Liquid and Gaseous Effluents from Nuclear Fuel Cycle Facilities* (NRC 2010), the overarching goal should be to obtain an overall estimate of the uncertainty of the measurement by evaluating the important contributors to the uncertainty. The combined standard uncertainty may vary depending on the measurement method and instrument capabilities.

Measurement uncertainties may be classified as systematic (i.e., biased) or random. Random uncertainties are associated with the variation of the result when the measurement is repeated (e.g., random nature of radioactive decay) from one measurement to the next. Systematic uncertainties, on the other hand, are related to activities that cause variations in the result by a constant or relative amount (e.g., calibration, sample-to-detector positioning, voltage drifts, and measurements of weight, volume, time, and distance). Systematic uncertainties, and other uncertainties associated with human performance, should be identified and corrected to limit their effect on the combined standard uncertainty of the measurement.

The combined standard uncertainty can be estimated mathematically using empirical calculations or via computational tools that simplify the process without compromising the calculation. Uncertainty propagation can be performed fairly easily for simple measurements. However, when multiple measurements are performed or when complex algebraic operations are necessary, computational software may be utilized to propagate the uncertainties from multiple parameters or operations and estimate the combined standard uncertainty of the measurement. Procedural steps for evaluating uncertainty can be found in MARLAP.

7.3.2 Significant Figures

All results should be reported as obtained and accompanied by their corresponding uncertainty. The number of significant figures included in the reporting of the results depends on the uncertainty associated with the result. Measurement results and their corresponding uncertainty should be reported to no more than two or three significant figures (MARLAP). It is important to stress that rounding should only be performed when reporting the final result. Any rounding during intermediate calculations may introduce round-off errors. Figure 7-1 illustrates the application of this convention. Further, to preserve power of statistical hypothesis tests that may

be necessary during the data quality assessment phase of the data life cycle, the appropriate number of significant figures for the sample results should be maintained. Several of the tests introduced in Chapter 8 are based on ranking the pooled data and by maintaining significant figures; ties between rankings may be minimized.

Measured Value (y)	Expanded Uncertainty $U = ku_c(y)$	Reported Result
0.8961	0.0234	0.896 <u>+</u> 0.023
0.8961	0.2342	0.90 <u>+</u> 0.23
0.8961	2.3419	0.9 <u>+</u> 2.3
0.8961	23.4194	1 <u>+</u> 23
0.8961	234.1944	0 <u>+</u> 230

FIGURE 7-1: Example of reporting significant figures (MARLAP)

7.4 Analytical Procedures

The selection of appropriate analytical procedures and instrumentation depends on the sample matrix, the radionuclide(s) of interest, and associated radioactive emissions (i.e., alpha, beta, gamma). Some samples may require the addition of a "spike", that is, a known radionuclide(s) at a known concentration(s), to evaluate chemical/process recovery (yield) during sample preparation processes (e.g., chemical separation, filtration, evaporation, distillation) prior to analysis. Moreover, depending on the program- or project-specific goals, multiple analytical procedures may be needed to assess the radionuclide(s) of interest. Brief discussions of selected analytical procedures are included herein as examples. Additional analytical procedures, or a combination of multiple analytical procedures, may be needed to adequately assess the radiological components of the sample matrix of interest.

7.4.1 Alpha and Beta Measurements

7.4.1.1 Gross Alpha and Gross Beta Screening Measurements

Gross alpha and gross beta measurements are often used as a preliminary screening tool. Results above predetermined screening levels may lead to an increase in radiological controls. Gross alpha and gross beta measurements are typically performed after sample collection using hand-held instruments tailored to the emission of interest. Other instruments (e.g., gas proportional counters, liquid scintillation counters) may also be used for screening purposes in a laboratory setting.

7.4.1.2 Quantitative Alpha and Beta Measurements

Gross alpha and gross beta measurements are commonly reported in counts or as a count rate (i.e., counts per minute or cpm). When additional parameters are known (e.g., sampling area, mass, total efficiency), an activity or concentration can be calculated (e.g., disintegrations per minute per 100 cm², picocuries per liter, picocuries per gram). An approach such as that described in the International Organization for Standardization (ISO) Publication ISO/CD 7503-1, *Measurement of radioactivity – Measurement and evaluation of surface contamination – Part 1: General principles* (1988) can be used to quantify alpha and beta surface contamination.

A number of factors can impact the calibration of the instrumentation used for quantifying alpha and beta contamination; including, but not limited to, emission type, emission energies (particularly in the case of beta emitters), source-to-detector distances, detector type, and the calibration source geometry. For example, the instrument efficiency for Cs-137 (a high-energy beta emitter) and Co-60 (a low-energy beta emitter) will vary due to their distinct emission energies. If an instrument is calibrated for Cs-137 and Co-60 is measured, the resulting activity, and the calculated activity or concentration, would be erroneous without the application of appropriate energy response correction factors.

When a single radionuclide is present in the sample, the quantification of alpha and beta contamination is fairly straightforward. However, when multiple radionuclides are present (multiple alpha- or beta-emitters) a weighted instrument efficiency should be determined to account for the contributions to the measurement.

7.4.2 Gamma-Ray Spectroscopy

Gamma-ray spectroscopy is widely used in laboratory settings to identify and quantify gamma-emitting radionuclides. Gamma spectroscopy can be achieved by using scintillator detectors (i.e., sodium iodide thallium-activated crystals [NaI(TI)]), or semiconductor detectors such as high purity germanium (HPGe). The gamma spectroscopy instrument of choice depends on the radionuclide(s) of interest, the potential activity, gamma-ray energy, and required resolution.

Nal(TI) crystals have poorer energy resolutions and higher backgrounds than semiconductors. Because of these shortcomings, a limited number of radionuclides can be measured in complex radionuclide mixtures using Nal(TI) crystals. Nal(TI) detectors are best suited for counting known radionuclides with low activities and when the energy difference between the radionuclides in the sample matrix is large enough to allow identification of each energy

contribution (there is no overlapping between their distributions). HPGe detectors, on the other hand, are best suited for measuring unknown radionuclides independent of their activity.

Careful attention should be paid to the activity in the sample; the greater the radioactivity, the larger the dead time of the detector. Large dead times will affect the activity estimation in the sample matrix and could damage the detector. Proper sample screening should prevent large activity samples from being analyzed. When the level of radioactivity in the sample matrix is high, representative subsampling (lower sample mass or volume) may be used to reduce the sample activity to an acceptable counting range. When subsampling is not possible, the dead time can be reduced by increasing the detector-to-source distance. However, this will induce variations in the counting efficiency that may be corrected by performing an instrument calibration at the new distance of interest.

7.4.3 Alpha Spectroscopy

Alpha spectroscopy is used in a laboratory setting and is used when alpha emitting isotopes are the analyte(s) of interest. The separation chemistry leading to counting using alpha spectroscopy may be tedious and time consuming. However, detection limits for alpha spectroscopy are considerably lower than gamma spectroscopy.

After separation, the alpha emitting radionuclides are placed on a planchet. The sample should be made with as small thicknesses as possible. Increases in the sample thickness will increase self-absorption within the sample matrix. In turn, this will shield alpha particles emitted from the sample surface that are not facing the detector. Moreover, sample matrices for alpha spectroscopy should not be encapsulated. The encapsulation will shield the alpha particles and may prevent them from reaching the detector.

Examples of alpha spectroscopy detectors include Passivated Implanted Planar Silicon (PIPS), silicon surface barrier (SSB), or diffused junction type devices. Because of the nature of the sample matrix needed for counting, these detectors are operated under vacuum conditions to enhance detection. Prior to operation, the detector chamber should be vented from ambient air to reduce radon contributions or moisture that may introduce counting interferences.

7.4.4 Liquid Scintillation Counting

Liquid scintillation counters (LSC) are primarily used to measure beta-emitting radionuclides. Water and solid matrices may also be analyzed using LSC provided that adequate sample digestion and separation of the radionuclides is permissible. Alpha-emitting radionuclides may

also be measured using a combination of LSC and pulse shape discrimination to reduce the interferences produced by ambient gamma rays and beta emitters in the sample and scintillation cocktail.

7.4.4.1 Tritium Analysis

H-3 is a pure, low-energy beta emitter (E_{max} = 18.6 keV) with a half-life of 12.32 years. Tritium is typically analyzed via liquid scintillation counting methods due to its pure beta emissions and low-beta energies.

A representative sample, subsample, or purified subsample should be placed into a glass or transparent container (as required by facility-specific protocols and calibration requirements) and a pre-determined volume of scintillation liquid added to the sample container. For example, a liquid sample may be analyzed by obtaining a representative purified subsample of 1 milliliter (mL) of volume and mixing it with 10 mL of scintillation liquid. The LSC results are typically provided in counts or disintegrations, or counts or disintegrations per minute per mL, as programmed by the LSC user. Solid samples, biological materials, and semi-liquid samples can also be analyzed for tritium in a similar manner. However, the sample needs to be completely dissolved into the scintillation liquid prior to analysis.

The sample counting time will vary depending on the H-3 concentration in the sample. Environmental samples selected for tritium analysis are typically counted for 30 minutes or longer to enhance detection and to reduce measurement uncertainties due to the expected low environmental tritium concentrations. If the expected tritium concentrations are relatively high, the necessary count time can be reduced (e.g., 1 minute, 10 minute), based on minimum detectable concentration (MDC) considerations.

Blanks, reference, or background samples should also be prepared for each tritium sample matrix. These types of samples should be analyzed using the same count time of the sample, whenever possible, for direct comparison. When not possible, background counts should be subtracted manually during sample activity calculations. Check or calibration sources should also be periodically counted for quality control and quality assurance purposes and to document trends in the LSC response.

7.4.5 Elemental Analysis

Elemental analysis procedures can be used to identify and quantify radioactive, particularly hard-to-detect radionuclides, and non-radioactive contaminants present in a sample of interest.

Inductively coupled plasma mass spectrometry (ICP-MS) is an example of a commonly used elemental analysis technique. Another variation of ICP includes atomic emission spectroscopy (ICP-AES).

In ICP-MS procedures, the sample, or a portion of the sample, digested in chemical reagents is transported by a noble gas (e.g., argon) through a high-electron density plasma. The sample is subsequently vaporized in the plasma region to separate its individual constituents. The individual constituents are extracted by a differential vacuum pump and separated based on their respective mass-to-charge ratios. Because the sample, or a portion of the sample, is vaporized, interference corrections are necessary to compensate for potential ion contributions from the plasma gas, reagents, or sample matrix. Interference corrections can be achieved by using a series of "blanks" to establish a calibration curve, monitor for reagents in the sample or sample processing, and return the system to its initial state prior to evaluating the next sample.

Guidance for performing ICP-MS analyses is provided by EPA in SW-846 Methods 6020A, "Inductively Coupled Plasma-Mass Spectroscopy", and Method 200.8, "Determination of Trace Elements in Waters and Wastes by Inductively Coupled Plasma – Mass Spectroscopy". The previously mentioned methods are typically used in conjunction with other sample preparation methods specific for the sample matrix of interest. Examples of sample preparation methods for ICP-MS analysis include, but are not limited to: (1) EPA Method 3050B, "Acid Digestion of Sediment, Sludges, and Soils", (2) Method 3005A, "Acid Digestion of Waters for Total Recoverable or Dissolved Metals for Analysis by FLAA or ICP Spectroscopy", and (3) Method 3010A, "Acid Digestion of Aqueous Samples and Extracts for Total Metals for Analysis by FLAA or ICP Spectroscopy".

Although Methods 6020A and 200.8 are the primary guidance documents for performing ICP-MS analyses, site-specific Standard Operating Procedures (SOPs) should be developed for analysis of the contaminants of interest and safe handling of the chemical solutions used in the sample preparation and measurement processes.

7.5 Quality Assurance

As applicable, the general Quality Assurance provisions of Chapter 11 of this Handbook should be followed. Additionally, the *DoD/DOE Consolidated Quality Systems Manual (QSM) for Environmental Laboratories* (2013) may also be a useful reference.

12 REFERENCES

- 1. Title 42 U.S.C. 2011, et seq., *Atomic Energy Act of 1954*, as amended.
- 2. Title 42 U.S.C. 7401 et seq., Clean Air Act, as amended.
- 3. 40 CFR Part 60, Appendix A, Test Methods.
- 4. 40 CFR Part 61, National Emission Standards for Hazardous Air Pollutants.
- 5. 40 CFR Part 61, Subpart H. National Emission Standards for Emissions of Radionuclides Other Than Radon From Department of Energy Facilities.
- 6. 40 CFR Part 61, Subpart I, National Emission Standards for Radionuclide Emissions from Federal Facilities Other Than Nuclear Regulatory Commission Licensees and Not Covered by Subpart H.
- 7. 40 CFR Part 61, Appendix B, Method 114, Test Methods for Measuring Radionuclide Emissions from Stationary Sources.
- 8. 40 CFR Part 61, Appendix D, Methods for Estimating Radionuclide Emissions.
- 9. 40 CFR Part 61, Appendix E, Compliance Procedures Methods for Determining Compliance with Subpart I.
- 10. 40 CFR Part 192, Health and Environmental Protection Standards for Uranium and Thorium Mill Tailings.
- 11. Allen, D.E., "Determination of MDA for a Two Count Method for Stripping Short-Lived Activity Out of an Air Sample," *Health Physics*, 73:512-517, 1997.
- 12. American National Standards Institute (ANSI), *Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities*, ANSI N13.1-1969, American National Standards Institute, New York, NY, 1970 (Reaffirmed 1982).
- American National Standards Institute (ANSI), Performance Criteria for Instrumentation Used for Inplant Plutonium Monitoring, ANSI N317-1980, American National Standards Institute, New York, NY, 1980.
- 14. American National Standards Institute (ANSI), Sampling and Monitoring Releases of Airborne Radioactive Substances from the Stacks and Ducts of Nuclear Facilities,

- ANSI/HPS N13.1-1999, American National Standards Institute and Health Physics Society, New York, NY, 1999 (Reaffirmed in 2011).
- 15. American National Standards Institute (ANSI), Specification and Performance of Onsite Instrumentation for Continuously Monitoring Radioactivity in Effluents, ANSI N42.18-2004, American National Standards Institute, New York, NY, 2004.
- American National Standards Institute/ American Society for Quality (ANSI/ASQ),
 Quality Systems for Environmental Data and Technology Programs: Requirements with
 Guidance for Use, ANSI/ASQ E4-2004, American National Standards Institute, New
 York, NY, 2004.
- 17. American National Standards Institute (ANSI), *Determining Meteorological Information at Nuclear Facilities*, ANSI/ANS-3.11-2005; R2010, American National Standards Institute and American Nuclear Society, Washington, DC, 2005 (Reaffirmed in 2010).
- 18. American Public Health Association (APHA), *Methods of Air Sampling and Analysis*, 3rd ed., American Public Health Association, New York, NY, 1988.
- American Public Health Association (APHA), Standard Methods for the Examination of Water and Wastewater (22nd ed.), E.W. Rice, R.B. Baird, A.D. Eaton and L.S. Clesceri (Eds.), American Public Health Association; American Water Works Association; and Water Environment Federation, Washington, DC, 2012.
- American Society for Testing and Materials (ASTM), Standard Practice for Calibration of Type S Pitot Tubes, ASTM Standard D3796-90(2004), ASTM International, West Conshohocken, PA, 2004, www.astm.org.
- 21. American Society for Testing and Materials (ASTM), *Standard Test Method for Average Velocity in a Duct (Pitot Tube Method),* ASTM Standard D3154-00(2006), ASTM International, West Conshohocken, PA, 2006, www.astm.org.
- American Society for Testing and Materials (ASTM), Standard Test Method for Average Velocity in a Duct Using a Thermal Anemometer, ASTM Standard D3464-96(2007), ASTM International, West Conshohocken, PA, 2007, www.astm.org.
- 23. American Society for Testing and Materials (ASTM), *Standard Practice for Sampling Surface Soils for Radionuclides*, ASTM Standard C998-05(2010)e1, ASTM International, West Conshohocken, PA, 2010a, www.astm.org.

- 24. American Society for Testing and Materials (ASTM), *Standard Practice for Soil Sample Preparation for the Determination of Radionuclides*, ASTM Standard C999-05(2010)e1, ASTM International, West Conshohocken, PA, 2010b, www.astm.org.
- 25. American Society for Testing and Materials (ASTM), *Standard Practice for Rotameter Calibration*, ASTM Standard D3195/D3195M-10, ASTM International, West Conshohocken, PA, 2010, www.astm.org.
- American Society for Testing and Materials (ASTM). Standard Test Method for Radiochemical Determination of Plutonium in Soil by Alpha Spectroscopy. ASTM Standard C1001. ASTM International, West Conshohocken, PA, 2011, www.astm.org.
- 27. Anspaugh, L.R., J.J. Koranda, W.L. Robison, and J.R. Martin, *The Dose to Man Via Food-Chain Transfer Resulting from Exposure to Tritiated Water Vapor in Tritium*, Messenger Graphics, Las Vegas, NV, 1973.
- 28. Arndt, M.F. and L. West, "A Study of the Factors Affecting the Gross Alpha Measurement, and a Radiochemical Analysis of some Groundwater Samples from the State of Wisconsin Exhibiting an Elevated Gross Alpha Activity," Michael F. Arndt, Ph.D., Assistant Researcher, Radiochemistry Unit and Inorganic Chemistry Unit, Wisconsin State Laboratory of Hygiene, Lynn West, Radiochemistry Unit, Wisconsin State Laboratory of Hygiene, http://digital.library.wisc.edu/1711.dl/EcoNatRes.ArndtStudy http://www.slh.wisc.edu/ehd/radiochem/dnr reports/dnrfinal.pdf, 2004.
- 29. Barnett, J.M. and W.E. Davis, "Six Methods to Assess Potential Radioactive Air Emissions From a Stack," *Health Physics*, 71(5):773-778, 1996.
- 30. Barnett, J.M., V.I. Cullinan, D.S. Barnett, T.L.T. Trang-Le, M. Bliss, L.R. Greenwood, and M.Y. Ballinger, "Results of a Self-Absorption Study on the Versapor 3000 47-mm Filters for Radioactive Particulate Air Stack Sampling," *Health Physics*, 97(5): S161-S168, 2009.
- 31. Bellamy, R.R., "Elemental Iodine and Methyl Iodide Adsorption on Activated Charcoal at Low Concentrations," *Nuclear Safety*, Volume 1516, pages 711-723, U.S. Atomic Energy Commission Technical Information Center, Oak Ridge, TN, 1974.

- 32. Beres, D.A., *The Clean Air Act Assessment Package 1988 (CAP-88): A Dose and Risk Assessment Methodology for Radionuclide Emissions to Air*, U.S. Environmental Protection Agency, Office of Radiation Programs, Washington DC, 1990.
- 33. Briggs, G.A., "Plume Rise and Buoyancy Effects," in: D. Randerson (Ed.), *Atmospheric Science and Power Production*, DOE/TIC-27601, Technical Information Center, U.S. Department of Energy, Oak Ridge, TN, 1984.
- 34. Denham, D.H., "Environmental Radiological Surveillance in Perspective: The Relative Importance of Environmental Media as a Function of Effluent Pathway and Radionuclides," *Health Physics*, 36: 273-281, 1979.
- 35. Denham, D.H., D.A. Waite, and J.P. Corley, *Summary of Selected AEC Contractor Environmental Surveillance Techniques and Capabilities*, BNWLB-384, Pacific Northwest Laboratory, Richland, WA, 1974.
- 36. Department of Defense (DoD) and Department of Energy (DOE), Consolidated Quality Systems Manual (QSM) for Environmental Laboratories, 2013.
- 37. Department of Energy (DOE), A Guide for Environmental Radiological Surveillance at U.S. Department of Energy Installations, DOE/EP-0023, Washington, DC, 1981.
- 38. Department of Energy (DOE), Radiological and Environmental Sciences Laboratory, Analytical Chemistry Branch Procedures Manual, DOE/IDO-12096, 1982.
- Department of Energy (DOE), Enewetak Radiological Support Project- Final Report,
 NVO-213, Nevada Operations Office, Las Vegas, NV, 1982.
- 40. Department of Energy (DOE), A Guide for Effluent Radiological Measurements at DOE Installations. DOE/EP-0096, Washington, DC, 1983.
- 41. Department of Energy (DOE), *Procedures for Sampling Radium-Contaminated Soils*, Grand Junction Projects Office, Colorado, GJ/TMC-13 (UC-70A), 1985.
- 42. Department of Energy (DOE), *Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance*, DOE/EH-0173T, Washington, DC, 1991.
- 43. Department of Energy (DOE), Radiation Protection of the Public and the Environment, DOE 5400.5, Change 2, Washington, DC, 1993.

- 44. Department of Energy (DOE), U.S. Department of Energy and U.S. Environmental Protection Agency, Memorandum of Understanding Between the U.S. Environmental Protection Agency and the U.S. Department of Energy Concerning the Clean Air Act Emission Standards for Radionuclides 40 CFR Part 61 Including Subparts H, I, Q and T, joint DOE/EPA agreement, 1995.
- 45. Department of Energy (DOE), *Radioactive Waste Management*, DOE O 435.1 Chg 1, Washington, DC, 2001.
- 46. Department of Energy (DOE), A Graded Approach for Evaluating Radiation Doses to Aquatic and Terrestrial Biota, DOE STD-1153-2002, Washington, DC, 2002a.
- 47. Department of Energy (DOE), *Performance Analysis of the Environmental Monitoring,*Surveillance, and Control Programs within the U.S. Department of Energy, Office of Independent Oversight and Performance Assurance, Washington, DC, 2002b.
- 48. Department of Energy (DOE), *Comprehensive Emergency Management System*, DOE O 151.1C, Washington, DC, 2005.
- 49. Department of Energy (DOE), Radiological Control, DOE-STD-1098-2008, Washington DC, 2008.
- 50. Department of Energy (DOE), *Radiation Protection of the Public and the Environment*, DOE O 458.1, Washington, DC, February 11, 2011.
- 51. Department of Energy (DOE), *Derived Concentration Technical Standard*, DOE-STD-1196-2011, Washington, DC, April 2011.
- 52. Department of Energy (DOE), *Quality Assurance*, DOE O 414.1D, Washington, DC, April 25, 2011.
- 53. Department of Energy (DOE), *Environment, Safety and Health Reporting*, DOE O 231.1B, Washington, DC, June 27, 2011.
- 54. Department of Energy (DOE), *Quality Assurance Program Guide*, DOE G 414.1-2B, Washington, DC, August 16, 2011.
- 55. Department of Energy (DOE), Occurrence Reporting and Processing of Operations Information, DOE O 232.2, Washington, DC, August 30, 2011.

- 56. Department of Energy (DOE), *Records Management Program*, DOE O 243.1B, Washington, DC, 2013.
- 57. dePlanque, G., T.F. Gesell, and K. Becker, "Second International Inter-comparison of Environmental Dosimeters Under Field and Laboratory Conditions," In *Proceedings of* the Tenth Midyear Topical Symposium of the Health Physics Society, pages 555-574, Rensselaer Polytechnic Institute, Troy, NY, 1976.
- 58. Eastern Research Group, Methods for Estimating Fugitive Air Emissions of Radionuclides from Diffuse Sources at DOE Facilities, Report prepared for the U.S. Environmental Protection Agency, http://www.epa.gov/radiation/neshaps/pubs.html), 2004.
- 59. Eisenbud, M. and T. Gesell, *Environmental Radioactivity*, Academic Press, 1997.
- 60. Environmental Protection Agency (EPA), *Environmental Radioactivity Surveillance Guide*, EPA ORP/SID 72-2, Office of Radiation Programs, Washington, DC, 1972.
- 61. Environmental Protection Agency (EPA), *Prescribed Procedures for Measurement of Radioactivity in Drinking Water*, EPA 600/4-80-032, Environmental Monitoring and Support Laboratory, Cincinnati, OH, 1980a.
- 62. Environmental Protection Agency (EPA), Method 901.1 (modified), *Gamma Emitting Radionuclides in Drinking Water*, EPA 600/4/80.032, Environmental Monitoring and Support Laboratory, Cincinnati, OH, 1980b.
- 63. Environmental Protection Agency (EPA), *Methods for Chemical Analysis of Water and Wastes*, EPA-600/4-79/020, Environmental Monitoring and Support Laboratory, Cincinnati, OH, 1983.
- 64. Environmental Protection Agency (EPA), Turner, J., M. Branscome, R. Chessin, A. Damle, and R. Kameth, *Method for Estimating Fugitive Particulate Emissions from Hazardous Waste Sites*, EPA-600/2-87/066, Washington, DC, 1987.
- 65. Environmental Protection Agency (EPA), *User's Guide for CAP88-PC Version 1.0*, EPA 402-B-92-001, Las Vegas, NV, 1992.

- 66. Environmental Protection Agency (EPA), Federal Guidance Report No. 13, Cancer Risk Coefficients for Environmental Exposure to Radionuclides, EPA 402-R-99-001, Washington DC, 1999.
- 67. Environmental Protection Agency (EPA), *Meteorological Monitoring Guidance for Regulatory Modeling Applications*, EPA 454/R-99-005, Office of Air Quality Planning and Standards, Research Triangle Park, NC, 2000a.
- 68. Environmental Protection Agency (EPA), *Updated User's Guide for CAP88-PC, Version* 2.0, EPA 402-R-00-004, Environmental Protection Agency, Office of Radiation and Indoor Air, Washington DC, 2000b.
- 69. Environmental Protection Agency, *Guidance for the Data Quality Objectives Process*, QA/G-4. Washington, DC, 2000c.
- 70. Environmental Protection Agency, *Guidance for Data Quality Assessment*, QA/G-9. Washington, DC, 2000d.
- 71. Environmental Protection Agency (EPA), Radionuclide NESHAPs Subpart H Inspection Manual, available at:
 http://www.epa.gov/radiation/docs/neshaps/nshpinspman_092702.pdf, 2002a.
- 72. Environmental Protection Agency (EPA), *Guidance for Comparing Background and Chemical Concentrations in Soil for CERCLA Sites.* EPA 540-R-01-003/OSWER 9285.7-41, 2002b.
- 73. Environmental Protection Agency (EPA), *Data Quality Assessment: Statistical Methods for Practitioners, EPA QA/G-9S.* EPA/240/B-06/003, Office of Environmental Information, Washington, DC, 2006.
- 74. Environmental Protection Agency (EPA), Statistical Analysis of Groundwater Monitoring

 Data at RCRA Facilities: Unified Guidance, EPA 530/R-09-007, 2009.
- 75. Environmental Protection Agency (EPA), *ProUCL Version 5.0.00 User Guide Statistical Software for Environmental Applications for Data Sets with and without Nondetect Observations.* EPA/600/R-07/041, Office of Research and Development, Washington, DC, 2013a.

- 76. Environmental Protection Agency (EPA), *ProUCL Version 5.0.00 Technical Guide Statistical Software for Environmental Applications for Data Sets with and without Nondetect Observations*. EPA/600/R-07/041, Office of Research and Development, Washington, DC, 2013b.
- 77. Gifford, F.A., "Turbulent Diffusion-Typing Schemes: A Review," *Nuclear Safety* 17(1):68-86, 1976.
- 78. Gilbert, R.O., *Statistical Methods for Environmental Pollution Monitoring*, Van Nostrand Reinhold Co., New York, NY, 1987.
- 79. Gilbert, R.O. and R.R. Kinnison, "Statistical Methods for Estimating the Mean and Variance from Radionuclide Data Sets Containing Negative, Unreported or Less-Than Values," *Health Physics*, 40: 377-390, 1981.
- 80. Gilbert, T.L., C. Yu, Y.C. Yuan, A.J. Zielen, M.J. Jusko, and A. Wallo III., *A Manual for Implementing Residual Radioactive Material Guidelines*. ANL/ES-160, DOE/CH/8901, Argonne National Laboratory, Argonne, IL, 1989.
- 81. Griffin, W.R., J.A. Cochran, and A.A. Bertuccio, *A Sampler for Nonaqueous Tritium Gases*, U.S. Environmental Protection Agency, Office of Radiation Programs, Winchester, MA, 1972.
- 82. Grossman, R.F. and R.W. Holloway, "Concentrations of Krypton-85 near the Nevada Test Site," *Environmental Science and Technology*. 19:1128-1131, 1985.
- 83. Guthrie, E.B., N.C. Shen, and B.B. Bandong, *Isotope Exchange and Fractionation Corrections for Extraction of Tritiated Water in Silica Gel by Freeze-Drying Techniques,*Lawrence Livermore National Laboratory Report UCRL-ID-148610, 2001.
- 84. Hanna, S.R., G.A. Briggs, J. Deardorff, B.A. Egan, F.A. Gifford, and F. Pasquill, "AMS Workshop on Stability Classification Schemes and Sigma Curves Summary of Classification Recommendations," *Bulletin of the American Meteorological Society* 58:1305-1309, 1977.
- 85. HASL-195, *The Radiation Field in Air Due to Distributed Gamma-Ray Sources in the Ground,* New York Operations Office (Atomic Energy Commission) New York Health and Safety Laboratory, New York, NY, 1968.