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Sampling and Analysis Protocols

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Introduction

Radiological sampling and analyses are performed to collect data for a variety of specific reasons covering a wide range of projects. These activities include:

- Effluent monitoring
- Environmental surveillance
- Emergency response
- Routine ambient monitoring
- Background assessments
- Nuclear license termination
- Remediation
- Deactivation and decommissioning (D&D)
- Waste management

In this chapter, effluent monitoring and environmental surveillance programs at nuclear operating facilities and radiological sampling and analysis plans for remediation and D&D activities will be discussed.

Most operating radiological facilities are required to have a radiological environmental monitoring program (REMP) in place that governs their effluent and environmental monitoring activities. The REMP serves two main purposes: to show compliance with applicable federal, state, and local regulations and to monitor any effects of plant operations on the environment-both on and off site.

Radiological effluent monitoring is conducted to

- Quantify source terms to show compliance with federal, state, and local regulations
- Verify commitments made in environmental impact statements
- Identify potential environmental problems
- Evaluate the need and/or effectiveness of effluent treatment and control practices
- Provide support for permitting activities and compliance
- Detect, characterize, and report unplanned releases

Radiological environmental surveillance is conducted to

- Verify compliance with environmental commitments
- Characterize and define trends in the physical and biological environs
- Establish baselines of environmental quality
- Assess pollution abatement and effluent control programs
- Assess the adequacy of plant operations and containment
- Identify and quantify new or existing environmental problems
- Verify or refine the predictions of environmental models
- Assess actual or potential contaminant exposures to critical groups and populations
- Conduct studies of the transfer of contaminants in the environment

For radiological facility license termination, remediation, D&D, and waste management the sampling and analysis of various media for residual radioactivity are required throughout the process. These activities include

- Scoping surveys
- Characterization surveys
- Remedial action support surveys
- Waste stream segregation
- Final status surveys

Numerous federal and state laws, regulations, and guidance documents are applicable to radiological sampling and analysis activities. Refer to Byrnes (2001) for a more complete discussion of these. However, two fairly recent, very detailed, multi-agency consensus documents have become the standard references for development of radiological sampling and analysis plans. They are the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) (EPA 2000a) and the Multi-Agency Radiological Laboratory Analytical Protocols Manual (MARLAP) (EPA 2004). MARSSIM was developed collaboratively by the U.S. Department of Defense (DOD), Department of Energy (DOE), Environmental Protection Agency (EPA), and Nuclear Regulatory Commission (NRC). MARLAP also was developed by these four agencies in collaboration with the U.S. Department of Homeland Security (DHS), Federal Drug Administration (FDA), United States Geological Survey (USGS), and the National Institute of Standards and Technology (NIST).

Data Life Cycle

Every year, government agencies and the regulated community spend billions of dollars collecting environmental radiological data for regulatory compliance, clean-up decision making, and research. Much of these data are required by laws and to ensure that human health and the environment are protected. Since most of these costs are paid for by taxpayers or nuclear power consumers, it should be the goal of all involved to minimize these costs by eliminating unnecessary and overly conservative data. However, the data collected needs to be of sufficient quantity and quality to allow defensible compliance and clean-up decision making. Therefore, all effluent and environmental radiological sampling and analytical protocols should be established as part of an overall structured process called the data life cycle approach. As shown in Fig. 1, there are four major phases to the data life cycle – planning, implementation, assessment, and decision (EPA 2000a).

In Fig. 1 the data life cycle is shown as a linear function but in actuality it is an iterative process with feedback cycles that allow for continuous improvement of the data quality. Without a complete understanding and integration of the four phases of the data life cycle, the analytical results obtained either may not meet the final decision making requirements or may be overly precise. Failure to properly perform and integrate all four phases will most likely increase project costs or lead to project failure.

The data life cycle process should be used for all projects where 1) the objective of the project is to collect radiological environmental data for regulatory compliance or 2) the results of the project will be used to make a specific clean-up decision. All parts of the data life cycle process may not be applicable to all projects, especially where specific final decisions can not be identified, such as for research or basic science. However, the data life cycle framework is still a

valuable tool to help plan these types of studies and to ensure the quality and defensibility of the data collected.

Data Life Cycle Process		
Process		Process Output
Planning	Planning Process	Development of Data Quality Objectives (DQO) and Measurement Quality Objectives (MQO)
	Plan Documents	Sampling and Analysis Plan (SAP) and Quality Assurance Project Plan (QAPP)
	Services	Statement of Work and Contracts
Implementation	Sampling	Laboratory Samples
	Analysis	Complete Data Package of Analytical Results including Quality Control Samples
Assessment	Verification	Data Verification Report
	Validation	Data Validation Report
	Data Quality Assessment	Assessment Report
Decision	Decide if the Final Data Results Demonstrate Compliance with Applicable Regulations	Annual Site Environmental Report or Project Close Out Report
Fig. 1 The data life cycle		

Planning Stage

The planning phase of the data life cycle is a performance based process that produces the final sampling and analysis plan, quality assurance project plan, and statements of work to be done for a given project. In this performance based approach, the project specific data requirements are determined and used to develop the appropriate sampling and analytical

protocols. This is done using the data quality objectives (DQO) process developed by the EPA (EPA 1994), which consists of the following seven steps:

1. State the Problem – Define the problem clearly so that the focus of the project will be unambiguous.
2. Identify the Decisions – Identify the principal study questions the project will resolve and what the alternative actions will result.
3. Identify Inputs to the Decision – Identify the information that needs to be obtained and the measurements (and the quality thereof) that need to be taken to resolve the decision statement.
4. Define the Study/Project Boundaries – Specify the time periods and spatial areas to which the decisions will apply. Determine when and where data should be obtained.
5. Develop a Decision Rule – Define the statistical parameter of interest, specify the action level, and integrate the previous DQO outputs into a single statement that describes the logical basis for choosing among alternative actions. These are usually "if…then" type statements.
6. Specify Limits on Decision Errors – Define the decision maker's tolerable decision error based on a consideration of the consequences of making and incorrect decision. All environmental data has associated sampling and analytical errors. Therefore, decisions based on these data also will have associated errors.
7. Optimize the Design – Iteratively evaluate the information from the previous steps and choose the most resource-effective design that meets all of the DQOs.

The final product of the DQO process is usually the projects' Sampling and Analysis Plan (SAP). The SAP includes a summary of the 1) DQOs, 2) Quality Assurance Project Plan, 3) Field Sampling Plan, and 4) Health and Safety requirements. The following suggested outline for a SAP is described in detail in Byrnes (2001).

1. Introduction
    a. Background
    b. Contaminants of Concern
    c. Data Quality Objectives
        i. Statement of the Problem
        ii. Decision Rule
        iii. Error Tolerance and Decision Consequences
        iv. Sample Design Summary
2. Quality Assurance Project Plan
    a. Project Management
        i. Project Organization
        ii. Quality Objectives and Data Measurement Criteria
        iii. Special Training Requirements
        iv. Documentation and Records
    b. Measurement/Data Acquisition
        i. Sampling Design
        ii. Sampling Methods
        iii. Sample Handling, Shipping, and Custody
        iv. Analytical Methods
        v. Quality Control Requirements
        vi. Instrument/Equipment Testing, Inspection, and Maintenance
        vii. Instrument Calibration
        viii. Inspection/Acceptance Requirements for Supplies
        ix. Data Acquisition Requirements
        x. Data Management
        xi. Sample Preservation and Holding Times
        xii. Field Documentation Requirements
    c. Assessment and Oversight
        i. Required Assessments and Response Actions
        ii. Reports to Management
    d. Data Validation and Usability
        i. Data Review, Validation, and Verification Requirements
        ii. Validation and Verification Methods
        iii. Reconciling Results with DQOs
3. Field Sampling Plan
    a. Sampling Objectives
    b. Sampling Locations and Frequency
    c. Sampling and Onsite Environmental Measurement Procedures

    d. Sample Management
    e. Management of Investigative Derived Waste
4. Health and Safety Issues
    a. Safety Procedures
    b. Hazard Assessments
5. References

Upon completion of the SAP, the Implementation Phase of the project begins.

Implementation Phase

During the implementation phase of the data life cycle, the collection of environmental radiological data entails two main processes, the sampling process and the analytical process (Fig. 1).

The sampling process involves collecting a small part of an environmental medium that is considered representative of that medium. The collected sample is then analyzed to quantify the radiological contamination present.

For some environmental surveillance projects and for most clean-up and decommissioning projects, the sampling process includes (and is usually preceded by) direct measurements and/or scanning. A direct measurement is performed by placing an appropriate detector near, on, or in the media being surveyed and reading the radioactivity level directly from a meter. Scanning is performed by moving an appropriate radiation detection instrument at a constant rate and distance above the surface to qualitatively and semi-quantitatively detect elevated areas of contamination.

The analytical process is a set of activities that begins at the time a sample is collected and ends with the final data package report. Sampling and analytical protocols will be discussed in more detail in later sections of this chapter.

Assessment Phase

The assessment phase of the data life cycle includes verification and validation, and an assessment of the overall data quality. Data verification and validation are the major components of the data quality assessment (DQA) report. In EPA's "Guidance on Environmental Data Verification and Data Validation" (EPA 2002), data verification and validation are defined as:

> **Data Verification** is the process of evaluating the completeness, correctness, and conformance/compliance of a specific data set against the method, procedural, or contractual requirements.
>
> **Data Validation** is an analyte- and sample-specific process that extends the evaluation of data beyond method, procedural, or contractual compliance (i.e. data verification) to determine the analytical quality of a specific data set.

The DQA process is the third and final step of the assessment phase and is described in EPA (2000b) as the "scientific and statistical evaluation of data to determine if data are of the right type, quality, and quantity to support their intended use." The DQA process is more global than verification and validation steps in that the focus is now on environmental decision making. The DQA report documents whether the datasets generated can credibly support the final required decision making process.

Decision Phase

During the decision phase of the data life cycle, final conclusions are drawn based upon the DQA report. The objective is to make technically defensible decisions concerning compliance

with applicable radiological effluent and environmental regulations or with dose/risk based clean-up regulations.

Many of the existing radiological regulations governing nuclear facility operations are deterministic. That is, compliance with a specific maximum annual dose or concentration level must be shown. Examples of these are EPA's National Emissions Standards for Hazardous Air Pollutants (NESHAP) dose standard of 0.1 mSv y^{-1} (10 mrem y^{-1}) for annual airborne emissions of radionuclides and drinking water maximum contaminant levels (MCLs) found in the National Primary Drinking Water Regulations (EPA 2000c). For operating facilities, compliance is usually documented in an annual site environmental report.

For clean-up activities, compliance with dose- or risk-based residual radioactivity requirements are usually done using parametric or non-parametric statistical tests to demonstrate that the mean concentration of radionuclides remaining in the survey unit complies with the release criteria. The typical null hypothesis for these tests is that the survey unit contaminant concentrations are above the criteria until proven clean. The final decisions for clean-up activities are usually documented in a site close out report.

Sampling Protocols

As discussed in the implementation phase of the data life cycle, the collection of radiological data may involve scanning, direct measurements, and sampling and analysis. Detailed discussions of the methods and instruments used for scanning and direct measurements can be found in Byrnes (2001) and in MARSSIM guidance (EPA 2000a). The focus of the remaining sections of this chapter will be on effluent and environmental media sampling and analysis.

Sampling Approaches

There are three basis approaches to environmental media sampling: judgmental, systematic, and random.

Judgmental sampling includes all aspects of effluent point monitoring and biased sampling performed in areas of known or suspected contamination. Judgmental samples usually require the least number of samples for obtaining the data required for the decision making process. However, biased sampling should not be used for projects requiring the mean concentration of a given area or for statistical hypothesis tests, such as would be required during the final status survey of a clean-up project.

Systematic sampling is used when the objective of the project is to 1) search for unknown or potential areas of contamination or 2) determine the spatial boundaries of a contaminated area. Because of the unknown location of the potential source of contamination, systematic sampling usually requires more samples to obtain sufficient statistically defensible data for the decision making process. Systematic samples are collected from an evenly spaced grid where the starting point is randomly or judgmentally selected. The grids may be square, rectangular, triangular, or radial. Triangular grids are usually the preferred pattern for clean-up projects because they reduce the interstitial area between sampling locations and have less potential pattern bias. The MARSSIM process (EPA 2000a) relies heavily on the triangular grid, systematic sampling protocol to help ensure that "hot spots" or areas of elevated contaminant concentration, do not go undetected during the final status survey of a clean-up project.

Simple random sampling is performed when little or no historical information about a site exists. All bias is removed because every point within the study area has an equal probability of being chosen as a sample location. The data obtained from simple random sampling can be used for most statistical hypothesis tests, such as comparing a mean radionuclide concentration to a

derived concentration guideline level. The disadvantage of random sampling is that the number of samples needed to satisfy the statistical tests will usually be more than the number required for other sampling methods.

There also are three main combinations of the three basic sampling approaches: 1) judgmental random (stratified random), 2) systematic random and 3) systematic judgmental. Judgmental random uses some historical knowledge to stratify the site into survey units of comparable concentration (to reduce heterogeneity) prior to the random samples being selected. Systematic random establishes a systematic grid after the first sample point is randomly selected. This approach is typical of the MARSSIM process (EPA 2000a). Systematic judgmental establishes a systematic grid after the first sample point is judgmentally chosen to conservatively ensure that previously known or potential areas of contamination are sampled.

Media Sampling

During the DQO process of the data life cycle, the number, location, size, and type of media sampling is determined and documented in the SAP. There are four basic types of media samples collected for environmental radiological investigations: 1) grab samples, 2) swipe samples, 3) composite samples, and 4) integrated samples.

A grab sample is simply the physical collection of a medium at a single location at a single point in time. The sample is typically transferred directly to a sample container and sent to a laboratory for radioanalysis. Mixing or compositing of the sample is usually avoided. Grab sampling is useful for the following applications:

- Periodic confirmatory measurements
- Batch sampling
- Scoping surveys
- Characterization surveys
- Remedial action design and support

- Waste stream segregation
- Baseline risk assessments

Swipe samples are areal surface samples that are taken to quickly estimate the amount of removable contamination on that surface. Swipe samples are taken from building, equipment, or material surfaces typically using filter papers that are swiped over an area of 100 cm^2. The filter paper samples are immediately bagged and sent to a laboratory for rapid gross alpha and/or gross beta analysis. Swipe sampling is an integral part of radiological control operations, but it also may be used in clean-up projects for characterization and remedial action support and confirmation.

Composite sampling combines portions of multiple samples taken from different locations or at different depths. It is often used to reduce a projects' cost by reducing the number of samples that are analyzed. Composite sampling should not be used when there is a potential for wide variation in residual contaminant concentrations. However, it has wide application in all aspects of environmental surveillance and research. Though it is typically not recommended, composite sampling may be used during clean-up projects, especially for scoping and characterization surveys, remediation support, and waste segregation. At the Savannah River Site (SRS), composite sampling has been used for facility D&D projects when the residual radioactivity has been shown to be fairly homogeneous (Lee et al. 2005).

Integrated sampling is distinct from composite sampling in that the media sample is collected from a single location over a specified period of time (days, weeks, months, years…). Continuous effluent monitoring of liquid and airborne emissions from a facility are examples of integrated sampling, where timed or flow-proportional samples are continuous taken and stored in bottles or on filter media until periodically collected and analyzed. Integrated sampling is not

normally used for clean-up projects, but has wide applications in effluent monitoring, environmental surveillance, long term stewardship, and research.

Liquid Effluent Monitoring

The generic term "effluent monitoring" is defined in DOE (1993) as ". . . the collection and analysis of samples or measurements of liquid and gaseous effluents for purposes of characterizing and quantifying contaminants, assessing radiation exposures to members of the public, and demonstrating compliance with applicable standards." Therefore, when used in this document, "effluent monitoring" can refer to:

- Continuous direct measurement of radionuclides in the effluent stream
- Sampling and analysis of the liquid waste prior to discharge as a batch release
- Continuous sampling, followed by laboratory analyses, to determine the quantity of radionuclides present in the effluent stream
- Periodic sampling, followed by laboratory analyses, to determine the quantity of radionuclides present in the effluent stream.

During the DQO process, a liquid effluent monitoring/sampling program must be designed to collect a representative sample so the results properly and accurately characterize the radiological emissions. The key components for any sampling scheme are selection of the sample location and selection of the type of sampling or monitoring to be performed. After evaluation of the potential for the emission point to release contaminants, the sampling equipment and sampling location can be determined based on applicable guidance (e.g. DOE 1991).

In general, a liquid effluent monitoring program is designed to directly monitor effluents and/or to collect and analyze samples from all site process outfalls that have the potential to release contaminants. Specifically, the program should ensure the following:

- Liquid effluent monitoring systems are based on the characterization of the source(s), pollutant(s), sample system(s), treatment system(s), and release points(s)
- Detection levels of the analyses and performance of the monitoring systems are sufficient to demonstrate compliance with regulatory requirements.
- The sampling systems are sufficient to collect representative samples.
- Continuous monitoring and sampling systems are calibrated and maintained appropriately.
- Environmental conditions and the nature of the effluents are not affecting the operation of the monitoring systems.
- Continuous-monitoring-system recorders and alarms are in locations continuously occupied by operations or security personnel and alarm set points are appropriate to prevent exceedances of applicable standards and recommendations.

Radiological Monitoring Locations. The monitoring of radioactive liquid effluents is normally required at the point of discharge and prior to dilution in the receiving surface waters. DOE (1991) defines a discharge point as "any discernable, confined, and discrete conveyance, including but not limited to any stack, duct, fissure, container, or vessel from which any radioactively contaminated gas or water is discharged to the atmosphere or to waters accessible by the general public."

At SRS, the interpretation of a liquid effluent discharge point is "the point at which a manmade conveyance (i.e., pipe, ditch, channel, conduit, well, or canal) discharges into (1) a naturally occurring body of water (i.e., site stream), (2) a manmade pond or lake that overflows into a naturally occurring body of water that ultimately is accessible by the general public, or (3) the Central Sanitary Sewer influent line from a properly licensed facility." By extension, the point of discharge can be located anywhere along the manmade conveyance-after the final process stream enters but prior to dilution by the receiving body of water. The small amount of rainwater that may fall directly into an open conveyance prior to a monitoring point is not considered to be dilution by a naturally occurring body of water. Additional monitoring,

although not required, may be conducted upstream of the point of discharge if it is determined by the applicable operating department to be technically justified during the DQO process.

Batch Release Sampling. Many radiological liquid effluent processes allow for batch releases of the effluent. The liquid effluent is collected in large tanks, usually after some type of filtration, mixed, sampled, and analyzed prior to discharge to ensure the effluent is within regulatory or recommended limits.

Continuous Discharge Sampling. For processes that require continuous aqueous discharges to the environment (e.g. cooling water blow down), periodic grab sampling or integrated sampling is performed. Depending on the DQO's, timed or flow-proportional integrated sampling may be employed.

Timed sampling is simply the automatic collection of a sample on a specific schedule (minute, hour…). The samples may be collected in individual vials or bottles, or they may be composited into a single sample bottle.

Flow proportional sampling requires the use of an effluent flow measurement or control device, such as a weir. The volume of sample taken, which can be timed or continuous, is then programmed to be proportional to the effluent flow volume. Because of the additional equipment requirements, flow proportional sampling is more expensive than other types of liquid effluent sampling. However, it is the preferred integrated sampling method for demonstrating regulatory compliance because the measured radionuclide concentrations in the sample will be more representative of the mean concentration of the total effluent volume released during the sample period.

Case Study - Radioactive Liquid Effluent Monitoring Requirements At SRS

Abstract. For Department of Energy (DOE) facilities, clear regulatory guidance exists for structuring radiological air emissions monitoring programs. However, there are no parallel regulations for radiological liquid effluent monitoring programs. In order to bridge this gap and to technically justify liquid effluent monitoring decisions at DOE's Savannah River Site, a graded, risk-based approach has been established to determine the monitoring and sampling criteria to be applied at each liquid discharge point (Jannik and Fledderman, 2001).

Airborne Effluent Monitoring

The design of an airborne effluent monitoring system begins with a characterization and documentation of the emission sources. The potential airborne emissions of radionuclide-emitting sources are required to be evaluated as part of the Clean Air Act's Radionuclide NESHAPs compliance demonstration process (EPA 1999). These evaluations determine the potential effective dose equivalent (PEDE) at each source and are based on normal emissions, including system upsets, assuming no control devices were operating.

The following important factors should be considered during these evaluations:

- Identification of the potential or actual radionuclides present
- Identification of fallout and naturally occurring (background) radionuclides
- Presence of materials (chemical, biological) that could adversely affect the sampling and monitoring system or the detection of radionuclides
- Internal and external conditions that could have a deleterious effect on the quantification of emissions (e.g. outside temperature, humidity, and ambient ionizing radiation; and gas-stream characteristics, such as temperature, pressure, humidity, and velocity)
- Process descriptions and variability
- Size distribution of particulate materials
- Cross-sectional homogeneity of radionuclide distribution at the sampling point

Results of the potential emissions evaluations are used to determine the monitoring requirements for specific emission points. The sources are grouped into categories based on the criteria found in EPA (1999). Fig. 2 shows the potential impact categories.

Potential Impact Category (PIC Level)	Monitoring and Sampling Criteria	PEDE (mrem/yr)	Actual EDE (mrem/yr)
1	Continuous sampling to include a real time monitor and alarm	>0.1	>1E-02
2	Continuous sampling with off-line periodic analysis	>0.1	≤1E-02
3	Periodic quarterly or annual sampling and off-line analysis	≤0.1	>1E-05
4	Annual administrative review of facility uses to confirm absence of radioactive materials in forms and quantities not conforming to prescribed specification and/or limits	≤0.1	≤1E-05

Fig. 2 Potential Impact Categories

Effluent Flow Measurement. The characteristics and conditions of gas flow can vary widely, and the frequency of the measurements needed to meet the required accuracy for flow-rate determination will be based on the stability of flow from that source. EPA Methods 1, 2, and 4 should be followed to measure and determine stack velocity, static pressure, temperature, and

moisture content. EPA Method 1 determines where and how many velocity measurements must be taken. EPA Method 2 is the actual procedure used to measure and determine stack gas velocity, static pressure, and volumetric flow rate. EPA Method 4 is used to determine moisture content in stack gases. Vane anemometers, Pitot tubes, and hot-wire anemometers are typically used for stack flow measurements.

Sampling Locations. Samples of gaseous effluents should be extracted from an accessible stack down-stream of any obstruction-and preferably in vertical sections of the stack and located near the outlet-so that concentrations of the material of concern are uniform. Where possible, effluents should be extracted at least eight stack or duct diameters downstream and two stack or duct diameters upstream from any major flow disturbances (e.g. bends, junctions, transitions, and probes).

Sampling Probes. If uniform flow and concentration can be demonstrated at a stack or duct location during all anticipated operating conditions, a single probe can be used, with the average velocity of the effluent flow integrated over the cross section of the probe opening (ANSI N13.1-1969). If uniform flow and concentration cannot be demonstrated, or if incomplete mixing is suspected in large-diameter stacks or ducts (diameters greater than 30 cm), the need for multiple-inlet probes under continuous sampling conditions should be considered. Extraction probes and nozzles for the sampling of particulate materials should be consistent with ANSI N13.1-1969 for particulate materials. Probes for aerosol sampling should be positioned isoaxially in the stack or duct and sized to extract at the same velocity as the effluent stream sampled (isokinetic sampling). The nozzles should be made of stainless steel, have no significant leakage or loss of material, and remain rigid to the point of collection, accumulation, or measurement.

Sampling Pumps and Flow Measurement. Because the intent of airborne effluent sampling is to extract a known fraction of the gaseous effluent being sampled, accurate and reliable measurement of the effluent flow is important. Air moving systems for airborne effluent sampling should be constant-displacement systems (rotary vane or gear pumps). The pumps and other mechanical components should be designed to operate continuously under anticipated operating conditions, with scheduled preventive maintenance and repair.

Sampler gas flows should be continuously measured, with the measurements recorded over the duration of the sampling period. The flow measurements should be accurate to ±10 percent by calibration with standards traceable to the National Institute of Standards and Technology (NIST), DOE/EP-0096. The most commonly used equipment for these measurements are rotameters. Flow measurements are typically taken downstream of the collection system since deposition and condensation can result in erroneous flow measurements.

Sample Collections Systems. The design and capabilities of an airborne effluent collection system will depend on the form of the radionuclides to be collected, the sampling conditions, and the analytical techniques to be used. Collector housing and equipment be designed to minimize sample loss. The radionuclides in airborne effluents can be gases, vapors, or particulate materials. Typical sample collection systems include filter papers for particulates, charcoal canisters for radioiodine, and silica gel columns for vapors. Gases are usually monitored with real time, in-line instruments, or periodic grab samples are taken to an laboratory.

Environmental Surveillance

An environmental surveillance program conducted at a radiological facility is designed to survey and quantify any effects that routine and nonroutine onsite operations may have on the

site, the surrounding area, or people living in the vicinity. In general, the program is conducted to meet the following criteria:

- Ensure compliance with all applicable environmental quality standards and public exposure limits
- Establish background levels, regional reference levels unimpacted by site operations, and site contributions of radioactive materials to the environment
- Verify the effectiveness of effluent treatment and controls in reducing effluents and emissions
- Verify the effectiveness and validity of dose calculation models
- Provide trending information on the build-up and migration of radioactive materials in the environment
- Detect and quantify unplanned releases

To accomplish these goals, the program typically monitors radioactive constituents in a wide range of environmental media, including atmosphere (including rainwater), surface water, soil, sediment, vegetation, food products, biota, drinking water, and groundwater.

Program Rationale. The technical justification and direction for the environmental surveillance program is accomplished during the DQO process. An important element of the justification is the performance of critical radionuclide/critical pathway analyses that consider factors such as site operating history, release pathways, source terms, transport, exposure pathways, dose, and the resulting risk. Based on these factors, an assessment is made to address the 1) proper media to be sampled; 2) sampling methods and frequency to ensure a representative sample is obtained; and 3) analytical methods to be used.

Atmosphere. The atmospheric surveillance program is typically divided into two main program areas: air and rainwater. The goal of this program is to quantify the amount of radioactivity in the atmosphere resulting from routine and nonroutine releases. Air sampling is

conducted on and off site. Sampling of rain also should be conducted at all locations. Typically, atmospheric surveillance stations are placed near the center of the site; in a ring around the site at the site perimeter; and at a regional reference locations assumed to be unimpacted by site operations. An analytical evaluation of placement, such as that recommended by Waite (1973a) is recommended to ensure adequate coverage.

Although the air surveillance program is not used to directly show compliance with applicable dose regulations, the program information is used to verify and modify the models that are used to show compliance with this limit.

At a typical environmental air sampling station, a regulated rotary-vane pump is used to move air through the sampling system at a specified flow volume. Flow is indicated by a rotameter downstream of the sampling media and a bellows-type volume totalizer also is placed in the sample stream. Control limits on sampling flow system performance (+/-10 percent) are verified and system calibration requirements are typically accomplished by a hot wire anemometer and performed as required under any of the following conditions: every 6-12 m (routine calibration frequency); equipment replacement; or system out of control limit.

The filter media used during environmental air sampling depends on the chemical and physical form of the radionuclide(s) of concern. A typical system would include a 1) filter paper for particulates, 2) charcoal canister for iodines, and 3) silica gel for tritium.

Rainwater is collected by stainless steel pans (0.6 m by 0.6 m) located on top of the environmental air monitoring stations. For particulates, the rainwater may be gravity fed through an ion exchange resin column, which is periodically collected and analyzed. 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. After passing through the column, (or directly from the

collection pan in the case of those locations not equipped to sample deposition), the rainwater empties into a polyethylene collection bottle for analysis of highly soluble radionuclides, such as tritium oxide.

Ambient Gamma Radiation. An ambient gamma radiation surveillance program is conducted to characterize the radiation levels at a site and to provide an indication of the effect, if any, of facility operations on the environment. It also is available for emergency response actions and special environmental surveys. Specific program objectives are to:

- Provide ongoing environmental radiation dosimetry during routine operations
- Measure photon radiation levels at a facility and in areas surrounding a site
- Provide an expedient and reliable means of establishing population exposure levels and doses in the event of a release of airborne radioactivity.

To quantify the ambient gamma radiation environment, thermoluminescent dosimeters (TLDs) are normally used. As an example, at SRS, Panasonic UD-801 badges are used. This badge contains two elements of copper-activated lithium borate (Li2B4O_7:Cu), and two elements of thulium-activated calcium sulfate ($CaSO_4$:Tm). Only the calcium sulfate elements are used for environmental dosimetry. These are covered by a 700 mg/cm^2 lead filter to attenuate low-energy photons to compensate for the over response of the phosphor to this energy spectrum.

TLDs are typically placed in sets of seven: five indicator badges and two badges used for fade correction. The badge sets are placed in free air on hangers 1 meter above the ground. They are exposed for one calendar quarter prior to processing.

ANSI 1975 specifies the requirements for monitoring environmental photon radiation with TLDs. This document specifies performance testing, procedure requirements, and data correction techniques.

In addition to monitoring ambient conditions, TLDs may be used as part of special surveys, for research, and for emergency response activities. The number, location, and exposure time of the TLDs will vary depending on the application.

Surface Water Monitoring. A surface water surveillance program consists of two divisions: 1) onsite bodies of water (streams, ponds, and lakes and 2) offsite bodies of water (streams, rivers, lakes, or ocean). The objective of the surface water surveillance program is to work with the effluent monitoring program to:

- Determine compliance with all applicable environmental quality standards and public exposure limits
- Establish background levels and quantify site contributions of radioactive materials in the environment
- Verify the effectiveness of effluent treatment and controls in reducing effluents and emissions
- Accumulate trending information on the buildup and migration of radioactive materials in the environment
- Detect and quantify unplanned releases

Sampling site locations for a surface water surveillance program are based on one or more of the following general criteria:

- Upstream or downstream of process effluents
- Upstream or downstream of the confluence of two bodies of water
- Control (background) locations, including upstream of the site and/or offsite bodies of water
- Obtaining representative samples considering stratification and complete mixing

The two major technical equipment areas of concern in a surface water surveillance program are sampling and flow measurement.

Sampling may be performed by grab sampling or by utilizing an integrated sampling system; the latter method is preferred and generally yields a more representative sample. Most integrated sampling systems use a time-programmable, peristaltic pump, composite-sampling system.

Stream flow is determined by the using one of two methods. The first method utilizes stage-to-flow discharge rating tables based on measured values in site streams. Stage is determined using a float system and stored by a data logger; the data are collected periodically or transmitted via a Geosynchronous Orbiting Earth Satellite (GOES) transmitter to a facility for final processing.

The second method utilizes a current velocity meter. The current velocity method relies upon the general equation:

$$\text{flow} = \text{velocity} \times \text{area} \qquad (1)$$

Both the stream velocity and cross-sectional area are measured, and flow rate is determined. This method provides an instantaneous measurement of the stream flow.

Groundwater Monitoring. Groundwater can be sampled using a direct push method (for grab samples) or, more commonly, with monitoring wells.

The direct push method utilizes a truck-mounted hydraulic press with a slide hammer. Depending on the composition of the unsaturated soil and the size of the truck and press, groundwater samples from near the surface down to 70 meters can be collected. The direct push method is not practical for integrated sampling or long term surveillance. However, for characterization surveys and research studies, the direct push method is economical and quick. It also produces very little investigative derived waste and causes only a small amount of environmental disturbance.

Monitoring wells are installed to provide a continuous and clean access point to the groundwater for integrated and long term surveillance. The basic components of a monitoring well are the well screen, sump, riser pipe, casing, and well cap. There are numerous tools and methods for collecting groundwater samples. Byrnes (2001) provides a comprehensive description of these. Overall guidance for implementation of groundwater surveillance monitoring can be found in DOE (2004).

Drinking Water Monitoring. A drinking water surveillance program performed at community water supplies is designed to monitor radiological contaminants for compliance with EPA (2000c) and applicable state or local regulations. Typically, both raw and finished water are sampled at these locations by water treatment plant personnel. Composited grab samples or integrated samples are collected using methods discussed for liquid effluent sampling. Depending on the applicable requirements, samples are analyzed for radionuclides monthly, quarterly, or annually. Compliance with EPA's drinking water dose standard and maximum contaminant limits (MCL) is demonstrated on an annual basis (EPA 2000c).

Food Product Monitoring. A food product surveillance program should be designed to determine any effects site releases may have on the food chain and quantify the exposure of the maximally exposed individual from the food pathway. Because of this, the program should concentrate on locally produced food products.

Terrestrial food products include milk, meat, fruit, grain, and vegetables. Collection of composited grab samples usually takes place annually and, where applicable, samples are collected from the farm or point of production during harvest. Samples of these items should be collected in each of the four quadrants surrounding the site and as close to the site boundary as

possible. In addition, control samples should be established at least 15 kilometers from the site center in the least prevalent wind direction.

Milk is a food product of special interest because, following atmospheric deposition, radioactive materials ingested by cattle and goats are quickly transferred into their milk, and the time between production of that milk and its consumption by humans usually is short. Additionally, milk is a major food product for children. Where it is available, grab samples of raw whole milk should be collected from individual dairies as close to the site as possible. All milk samples should be preserved by chilling.

An aquatic food product surveillance program may consist of both fish and shellfish. The program should be designed to quantify any effects of site operations on local and downstream edible fish. Grab samples of aquatic food may be obtained with line and pole, nets, traps, or electro-shocking systems. Normally, the mean concentration in fish and shellfish from a particular location is desired. Therefore, compositing of several fish/shellfish from the same location is usually performed to ensure adequate sample size and to reduce the number of radioanalyses performed. In review of related literature (Whicker et al. 1990) it is shown that the bioconcentration factor for radionuclides in fish and shellfish varies greatly depending upon the 1) chemical composition of the water, 2) amount of suspended solids in the water, 3) temperature of the water, 4) trophic level of the fish (piscivores bioconcentrate radionuclides much more that insectivores and benthivores), and 5) water level and flow rate. To take these variations into account, it is recommended that samples be taken several times per year and that they be composited by trophic level when practical.

Soil Surveillance Monitoring. A soil surveillance program performs two functions to:

- Observe and trend the deposition patterns of radioactive materials to the environment
- Provide an indication of concentrations of radioactive materials in the environment

Radioactive materials deposited in the environment come from two sources: site operations and worldwide fallout. Material is deposited by both dry and wet (rainfall) deposition processes.

A soil surveillance program is designed and best utilized to perform long-term trending of radioactive material levels in the environment, rather than quantifying local concentrations of activity. Because of the 1) limited number of samples usually collected, 2) variation in soil type (and the associated geochemistry), and 3) other sampling uncertainties, a direct comparison of soil data from year to year is not appropriate.

Soil grab samples are normally collected from onsite, site perimeter, and offsite locations. Samples should be collected from uncultivated and undisturbed areas. Hand augers or equivalent devices (i.e., pluggers) are used to obtain the grab samples and must be cleaned before reuse.

Sediment Surveillance Monitoring. Sampling and analysis of sediment provide a method to determine the movement, deposition, and accumulation of radioactive materials in stream and lake systems. Radionuclide levels in the sediment may show significant changes from year to year as stream and lake conditions change, resulting in increased deposition or remobilization.

Sediment grab samples are collected using either a dredge or scoop and must be cleaned before reuse. Sediment samples are often co-located with surface water samples.

Vegetation Surveillance Monitoring. Vegetation can accumulate radioactive contamination from either fallout or uptake from soil and water by the roots. Similar to soil surveillance, the purpose of vegetation surveillance is to monitor long term trends in the environment. Composited grab samples of local vegetation are usually taken on an annual basis and are often collocated with soil or environmental air samples.

Remediation and Decommissioning Monitoring

As previously stated, most clean-up and decommissioning projects rely heavily on scanning techniques and direct measurements of residual radioactivity. These protocols are outside the scope of this chapter and the reader is referred to the MARSSIM guidance document a (EPA 2000a) and Byrnes (2001) for a complete discussion of these methods.

However, volumetric sampling of applicable media is required for some types of preliminary and remedial support surveys and especially in support of the final status survey.

Soil Sampling. For remediation and decommissioning projects, soil sampling is performed to:

- Identify the type and extent of contamination (scoping surveys)
- Assessing the potential dose and risk to humans and the environment (characterization surveys)
- Determining the success of clean-up actions (remedial support)
- Verifying acceptability of clean-up (final status survey)

Near surface soil samples can be either grab or composites and are typically collected using a scoop, hand auger, slide-hammer, or tube sampler. Deep soil samples (depths greater than 2 m) are usually collected with tube samplers using a hydraulic press or auger drilling rig.

Concrete Sampling. For remediation and decommissioning projects, concrete sampling is usually performed to confirm scanning and direct measurements and to estimate radionuclide concentrations in floors, walls, and ceilings prior to demolition and disposal. However, in response to accelerated clean-up requirements at DOE sites, many facilities throughout the DOE complex will be decommissioned through removal of the facility structures leaving the concrete-slab foundations in place. These concrete-slab end states will be sampled and assessed in a similar manner to soil for final status surveys (Lee et al. 2005). At SRS, it has been shown that for many radionuclides, with the notable exception of tritium, most of the contamination is found

within the top 5 cm of the concrete slab (Roach et al. 2006) and this should be considered during the DQO process for concrete slab end-states.

Concrete samples are typically collected as composited grab samples to ensure sufficient sample mass. Sampling methods used include drilling, coring, scabbling, and chipping. It has been shown that rapid concrete sampling using a 2.5 cm rotary hammer drill is one of the best methods for obtaining samples that are amenable to acid dissolution without locally heating the sample, which may drive off volatile contaminants such as tritium (Hochel and Clark 2000).

Other Media Sampling. Other media sampling, such as paint and wall and floor coverings, usually required non-standard, site-specific sampling methods to ensure worker safety. Composited grab samples using scraping, scabbling, or chipping methods may be used.

Analytical Protocols

As part of the planning phase of the data life cycle (Fig.1), the development of measurement quality objectives (MQO) and analytical protocol specifications (APS) are a major part of determining analytical protocols. Volume I of the MARLAP guidance document (EPA 2004) provides detailed directions to project planners and managers for establishing MQOs and APSs and for preparing project plan documents such as sampling and analysis plans (SAPs) and quality assurance project plans (QAPPs). Volume II of MARLAP provides thorough and very detailed guidance to radioanalytical laboratory personnel concerning all aspects of the analytical process. Volume III of MARLAP deals with quality control and statistics.

Analytical Process

The analytical process is the general series of activities that are performed starting as soon as a sample is collected and ending with the final data report. Depending on the radionuclides of

concern, the sample matrix, and the MQOs, the following steps are followed in many radioanalytical processes:

1. **Field Sample Preparation and Preservation.** Immediately after a sample container has been filled in the field, it must be preserved, prior to shipment, in accordance with the SAP. Depending on the radionuclides of interest and the sample matrix, preservation techniques can range from none to cooling to the addition of acids or bases to maintain dissolution. During this step, Chain-of-Custody forms, labels, and seals are completed and will follow the sample all the way through the analytical process until the data are reported.
2. **Laboratory Sample Receipt.** The laboratory sample receipt process includes 1) radiological surveys of the sample containers and packaging, 2) physical inspection of the packaging, tracking labels and seals, and 3) laboratory sample tracking, which continues (inside the laboratory) the process started during sample collection and preparation.
3. **Laboratory Sample Preparation.** The laboratory sample preparation process typically introduces the most error into any radioanalytical process (EPA 2004). Preparation procedures may include 1) compositing, 2) mixing, 3) grinding, 4) filtering or screening, and 5) drying. Strict adherence to standard operating procedures must be followed to ensure that representative sample aliquots are prepared without sample loss, cross contamination, or loss of sample tracking.
4. **Sample Dissolution.** Sample dissolution is required for most inorganic solids and nonaqueous liquids to produce a uniform matrix for measurement or a homogeneous solution for chemical separation. There are three main techniques for sample dissolution: 1) Fusion, where the sample is mixed with a salt and heated to above the melting point of

the salt. The sample reacts and fuses with the salt flux, then is cooled and dissolved. 2) Wet ashing and acid dissolution, where an appropriate acid is added to the sample and then ashed, dissolved, or leached. 3) Microwave digestion, where the sample is decomposed by being heated to very high temperatures using microwaves.

5. **Chemical Separation.** Radiochemical separation methods are broad and range from the simple ion exchange column to complex multi-stage extractions. The separation of minute amounts (micro- to pico-grams) of a particular element from a large sample is an exacting science requiring strict adherence to procedures and quality control. The use of known carriers and tracers also is required to ensure adequate separation has occurred. Some common chemical separation techniques include oxidation/reduction, complexation, solvent extraction, volatilization and distillation, electrodeposition, chromatography, and precipitation and coprecipition. The reader is referred to chapter 14 of MARLAP (EPA 2004) for a thorough discussion of these methods and the use of carriers and tracers.
6. **Preparation of Samples for Measurement.** For gross alpha and beta counting and gamma spectroscopy of certain matrices such as filter papers and swipes, the sample is prepared by simply placing the filter or swipe on a standard-size planchet prior to counting. For other matrices and for radionuclide-specific analyses the sample preparation is typically the last step of the chemical separation process such as for 1) electrodeposition on a metallic surface, 2) precipitation/coprecipitation on a micropore filter and 3) evaporation on a planchet.
7. **Instrument Measurement.** In a typical, well-equipped radioanalytical laboratory, the following array of detectors will be available:

- Gas proportional detectors for gross alpha and beta emitters
- Sodium-iodide or high purity germanium (HPGe) detectors for gamma spectroscopy
- Solid-state detectors for alpha spectroscopy
- Liquid scintillation counters (LSC) for beta emitters
- Mass spectrometers for atom or ion counting of low-level samples

8. **Data Acquisition, Reduction and Reporting.** Data acquisition is process of collecting the raw data from the counting instrument. Data reduction is the mathematical process that takes into account background, detector efficiency, chemical separation yield, sample mass, and radioactive decay to determine the net count rate or radionuclide concentration. Data reporting is the presentation of the results in a format usable by the decision maker.

Alpha Detection Methods

Alpha particles, which are essentially positively charged helium atoms, are massive compared to beta particles and because of their size and charge they have a high rate of energy loss over short distances. They can travel only a few centimeters in air. Therefore, for solid matrices, the sample must be prepared in a very thin layer and be positioned extremely close to the detector window.

Typical alpha detection devices include ionization chambers, proportional counters, solid-state spectrometers, and scintillation counters. Absorption of the alpha particle by the sample material or by the air or detector window are major concerns in alpha detection.

Gas-Flow Proportional Counters. Gas-flow proportional (GP) counters are the most commonly used gross alpha detectors. They can not be used for determining alpha particle energy. When an alpha particle enters the gas-filled chamber of the detector primary ionizations occur. The electrons produced by this ionization are accelerated towards an anode using high

voltage bias. These free electrons gain sufficient energy to produce secondary ionizations as they move towards the anode where they are detected as a voltage pulse. This gas multiplication effect increases the total number of electrons produced in proportion to the original number of primary ion pairs produced. P-10 gas (90% argon and 10% methane) is typically used in GP counters.

Solid-State Detectors. Solid-state detectors are used for alpha spectroscopy applications. They are essentially solid ionization chambers, where the energy of the incident alpha particle produces electron-hole pairs in the semiconductor material. The freed electrons are collected in an electric field, amplified and counted. The detector provides a linear response to the energy of the alpha particle thus allowing radionuclide specific determinations based on the measured alpha energy.

Beta Detection Methods

Beta particles are electrons that have been emitted from the nucleus of an atom undergoing radioactive decay and are usually accompanied by gamma emissions, which are normally easier to detect and quantify. Unlike alpha and gamma emissions, beta particles are not emitted monoenergetically from the nucleus, but have a range of energies with specific mean and maximum values. This fact eliminates the ability to perform beta spectroscopy. Beta particle detectors are similar in style and type to alpha detectors and include: gas-flow proportional counters, end-window Gieger-Mueller tubes, liquid and plastic scintillators, and solid-state detectors.

Gas-Flow Proportional Counters. Because of the large differences in incident energies, most currently available GP counters can differentiate alpha and beta particle emissions.

Therefore, because of this dual use capability, GP counters are the most commonly used gross beta detectors in radioanalytical laboratories.

Liquid Scintillation. For low energy beta emitters such ^{3}H and ^{14}C, liquid scintillation counting (LSC) is commonly used. Sample preparation is fairly easy and simply requires the addition of a soluble or dispersable sample to a scintillation cocktail made up of appropriate organic phosphors and solvents. The beta particle energy is converted to photons in the scintillation cocktail. The photons are then detected in a photo-multiplier tube (PMT) and the photon events are counted. Two PMTs are normally used for coincidence detection in order to reduce background and electronic interferences. Because each molecule of the sample is surrounded by the liquid scintillator, LSCs have very high efficiencies.

Gamma Detection Methods

Gamma-rays are highly penetrating forms of radiation released monoenergetically from the nucleus of a radioisotope during decay. Relatively large solid detectors, instead of gas or liquids, are usually used to provide sufficient mass to stop the energetic gamma rays within the detector. The two most common gamma detectors in use are sodium-iodide (NaI) and high-purity germanium (HPGe).

Samples for gamma counting usually do not require destructive preparation. However, standard sample containers and configurations need to be maintained to ensure consistent geometry, volume, mass, and homogeneity between samples and with calibration sources.

Sodium Iodide Detectors. NaI crystals are very dense and when activated with a small amount of thallium (0.1%) are excellent scintillators for gamma radiation. The light photons caused by the incident gamma rays are detected and amplified into an electrical pulse by a PMT.

The measured pulse height size is directly proportional to the energy of the gamma ray captured in the NaI(Tl) crystal. Pulse-height analysis, which is simply counting the number of times a specific pulse height occurs, can then be performed on a multi-channel analyzer to identify radionuclide specific energy peaks. For laboratory use, NaI(Tl) crystals are cylindrical and typically 7.5 cm x 7.5 cm in size.

High Purity Germanium Detectors. HPGe detectors are cylindrical crystals that perform like a semiconductor. Incident gamma rays cause electron-hole pairs within the crystal and the freed electrons are collected in an electric field, amplified, and counted. The output electrical pulses from the system are directly proportional to the amount of energy deposited by the gamma ray in the crystal. HPGe crystals must be maintained in a metallic "can", under vacuum and kept cold using liquid nitrogen to reduce thermal and electronic effects. Because of these requirements, HPGe detectors are usually limited to laboratory use. HPGe detectors have lower efficiencies than the same size NaI(Tl) detectors, but they have much better energy resolution and, therefore, are the preferred detector for gamma spectroscopy applications.

Minimum Detectable Concentrations

One important part of the DQO process is to identify the minimum detectable concentrations (MDC) that a particular analytical process must meet to allow technically defensible decisions to be made at the end of the data life cycle. In some cases, definitive regulatory guidance exists, such as EPA's MDC requirement of 0.1 times the drinking water MCLs. By extension, this factor has been used for clean-up projects where the MDC must be at least 0.1 times the derived concentration guideline limit. However, there is no definitive MDC guidance applicable to

environmental surveillance. The following case study provides a risk-based approach to this determination.

Case Study of MDC Determination at SRS - Abstract. Other than drinking water, there is no definitive guidance in existing federal or state regulations concerning the appropriate minimum detectable concentrations (MDCs) that should be achieved for radiological analysis of environmental media. At the Savannah River Site (SRS), it has been proposed that MDCs for environmental samples be risk-based. Using

- Applicable and reasonable pathways to man
- Maximally exposed individual usage rates appropriate for the SRS area
- U.S. Department of Energy (DOE) approved dose factors
- Dose to risk factors from the International Commission on Radiological Protection (ICRP) Publication 60 (7.3 E-07 total risk per mrem)
- 30-year exposure time

Calculations were performed to determine radionuclide concentrations in environmental media that equated to a potential lifetime risk of 1E-06. This case study describes the process used to determine appropriate MDCs for selected environmental media. Also, a comparison of the risk-based MDCs with the SRS Environmental Monitoring Section's existing environmental media MDCs is provided and discussed (Jannik et al. 2000).

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