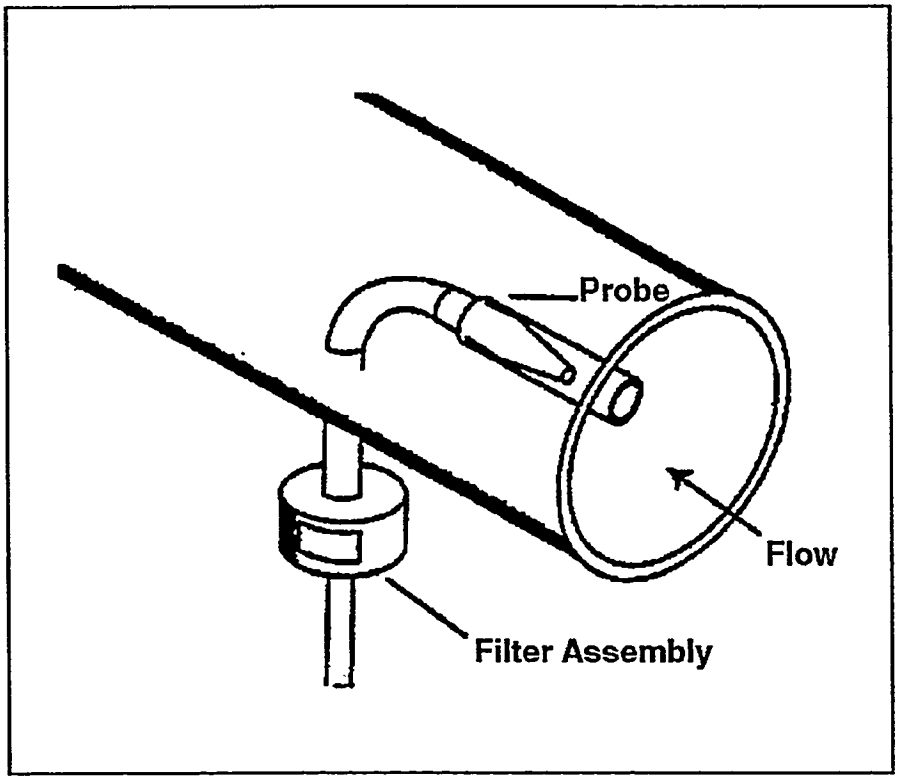
LA-13235-T
Thesis

Compliance Program for 40 CFR 61, Subpart H at Los Alamos National Laboratory



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Compliance Program for 40 CFR 61, Subpart H at Los Alamos National Laboratory

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ACKNOWLEDGMENTS

The physical upgrades of stack sampling systems to make them compliant with the requirements of 40 CFR 61, Subpart H have been conducted over the last four years. Although, as project manager, I have been responsible for the overall development and implementation of the project, I certainly did not accomplish this task on my own. This task was completed through the effective teamwork of the Los Alamos National Laboratory's entire Air Quality Group, ESH-17. This organization is a remarkable collection of dedicated professionals that all had a significant part in the implementation of a technically sound program within strict financial limits.

In particular, I would like to recognize the participation of Steve Story, Allen Meddles, Harold Martinez, Kevin Smale and Scott Miller. These gentlemen planned, coordinated, and supervised most of the day-to-day work. They were responsible for measuring exhaust system parameters, preparing the system designs, purchasing the materials, arranging for the system installations, developing and implementing many of the program's procedures, and ensuring quality control throughout the process.

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at Los Alamos National Laboratory

Eric A. McNamara

ABSTRACT

Effective on March 15, 1990, the Environmental Protection Agency established regulations controlling the emission of radionuclides to the air from Department of Energy facilities to limit the dose to the public to 10 mrem/yr. These regulations are detailed in 40 CFR 61, Subpart H, "National Emission Standards for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities" (USEPA 1993a). Part of these regulations require the operation of sampling systems on stacks meeting certain requirements. Although Los Alamos National Laboratory has a long history of stack sampling, the systems in place at the time the regulation became effective did not meet the specific design requirements of the new regulation. In addition, certain specific program elements did not exist or were not adequately documented.

The Los Alamos National Laboratory has undertaken a major effort to upgrade its compliance program to meet the requirements of USEPA 1993a. This effort involved: developing new and technically superior sampling methods and obtaining approval from the Environmental Protection Agency for their use; negotiating specific methodologies with the Environmental Protection Agency to implement certain requirements of the regulation; implementing a complete, quality assured, compliance program; and upgrading sampling systems. After several years of effort, Los Alamos National Laboratory now meets all requirements of the USEPA (1993a).

INTRODUCTION/BACKGROUND

Los Alamos National Laboratory (LANL or the Laboratory) is a 43 square mile Department of Energy (USDOE) site located approximately 60 miles north-northeast of Albuquerque, New Mexico. The site includes a large number of facilities supporting a wide variety of programs. Examples of these programs include support for the USDOE's nuclear weapons program, basic scientific research, the operation of a proton accelerator, and the production of radioisotopes for commercial and medical uses. These programs, and many others at LANL, involve the use of radioactive materials. As an unavoidable result of these programs, some of these radioactive materials are routinely released to the environment via the facilities' air exhausts.

Regulatory History

Prior to 1990, the release of radioactive materials from USDOE sites was regulated by USDOE orders. These orders limited the release of radioactive materials from a USDOE site such that no member of the public could receive a dose of greater than 100 mrem in any year from all pathways, including the air pathway. In 1990, the Environmental Protection Agency's 40 CFR 61, Subpart H, "National Emission Standards for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities" (Rad NESHAP) became effective. This regulation, an implementation of the federal Clean Air Act (CAA) National Emission Standards for Hazardous Air Pollutants (NESHAP), requires that no member of the public receive a dose

greater than 10 mrem in any year from the release of radionuclides from USDOE facilities via the air pathway (USEPA 1993a).

Sampling Program Prior to March 15, 1990

To demonstrate compliance with these requirements, as well as to support operational needs, LANL has historically sampled many of its stacks to determine the amount of radioactive material released from the associated operations. This program of sampling stacks has been in place for many years. In 1990, over ninety stacks were continuously sampled. These stacks represented the predominant sources of radioactive air emissions. The information obtained from these sampling systems was used in computer models to determine the dose a member of the public might receive.

In addition, the Laboratory has operated a series of environmental air sampling stations in and around the site to measure actual concentrations of radionuclides in the environment. This program has been in place since the 1970's and provides a redundant approach to determining dose to the public. The results of these measurements are reported in the Laboratory's annual Environmental Surveillance Reports.

New Technical Requirements

With the implementation of Rad NESHAP, the technical requirements for determining dose to the public became more rigidly defined. This regulation called out the requirements of the American National Standards Institute (ANSI) for the design of stack sampling systems (ANSI 1970). This guidance concentrates heavily on the ability of a sampling system to

representatively collect large particles (greater than about 3-5 μm Aerodynamic Equivalent Diameter (AED[1])) through isokinetic sampling. (The physics of large particles and isokinetic sampling are discussed in greater detail in Appendix A.) In addition, sampling sites are required to be selected following procedures in USEPA 1993b. It was clear in 1990 that the existing sampling systems were not necessarily designed to these rigid standards. This led to a substantial effort to fully characterize all radioactive air point sources at the Laboratory and to upgrade sampling systems to meet the new standards.

As a result of the new requirements, the USDOE and the Laboratory informed the Environmental Protection Agency (USEPA) that it could not meet the technical sampling requirements of the new regulation (USDOE 1991). This led to a Notice of Noncompliance (NON) issued by the USEPA on November 27, 1991 (USEPA 1991). This NON identified five specific shortcomings in the Lab's program:

1. Every release source from an operation which uses radionuclides has not been evaluated using the approved EPA computer model to determine a dose received by the public as required by 40 CFR 61.93(a).

2. DOE has failed to comply with 40 CFR 61.93(b)(4) because it has not determined each release point that has the potential to deliver more than 1% of the effective dose equivalent standard. The evaluation of emissions potential is to be performed by estimating the dose without taking any credit for any emissions controls on the effluent stream. The results of this modeling will then determine which release points

[1] AED is defined as the diameter of the unit density sphere that has the same settling velocity as the particle.

must be monitored in compliance with paragraph 61.93(b) and which release points must be monitored periodically to confirm continuing low emissions.

3. The facility currently has not installed stack monitoring equipment on all its regulated point sources in accordance with the above analysis and 40 CFR 61.93(b)(2)(ii) and (iii).

4. The facility has not conducted and is not in compliance with the appropriate quality assurance programs pursuant to 40 CFR 61.93(b)(2(iv).

5. The facility is in violation of 40 CFR 61.94 "Compliance and reporting" because it has not calculated the highest effective dose equivalent in accordance with the regulations cited above. EPA does acknowledge receipt of an annual report, as required by 40 CFR 61.94(a) but for the reasons specified above that report is incomplete.

It should be noted that these five items were identified based on self-reporting by the Laboratory and not by an EPA audit. The first formal EPA audit, which occurred in August of 1992, confirmed these findings. In addition, this audit identified the Laboratory's use of a shielding and occupancy factor in dose determination to be unauthorized. As a result, the USEPA issued a second NON regarding the use of shielding and occupancy factors. The Laboratory immediately terminated the use of these factors, and this issue will not be addressed further.

Federal Facilities Compliance Agreement

The first NON led the USDOE and USEPA to enter into active negotiations for the development of a Federal Facilities Compliance Agreement (FFCA). An FFCA is a means for a regulating agency (the USEPA) and a federal institution (the USDOE) to agree on a course of action that will lead to the federal institution's compliance with federal regulations (in this case Rad NESHAP). Negotiations were completed and the FFCA was signed on June 13, 1996 (USDOE and USEPA 1996).

The tasks described in the FFCA which would bring the Laboratory into compliance with Rad NESHAP have been actively pursued over the last several years. Much of the work actually began before the USEPA gave its concurrence for the Laboratory's approach. This entailed some risk that USEPA would disagree with the Laboratory's proposed methods and the work would have to be re-done. However, it was felt that the delay that would be caused by waiting for USEPA concurrence outweighed this risk. In retrospect, USEPA eventually concurred with all of the Laboratory's general approaches, requesting only very minor changes, and little or no work had to be done over again. All tasks related to compliance were completed by June 3, 1996, and the Laboratory is in compliance with Rad NESHAP as of that date. Although the FFCA includes future milestones regarding reporting of status and the completion of a management audit, these items are not strictly related to compliance with the regulation.

Sampling System Upgrades and Current Status

The information provided in this document summarizes the results of the last several years of work. Included are discussions on: source types; point source identification and characterization; major point source sampling; minor point source emission estimates and

periodic confirmatory measures; and accounting for nonpoint sources. The discussion include the rationale for selecting the approaches the Laboratory has used and summarizes the current status of stack sampling at the Laboratory.

SOURCES OF RADIOACTIVE AIR EMISSIONS AT LANL

There are two broad types of radioactive air emission sources at LANL: point sources and nonpoint sources. Typically, point sources are stacks and vents from buildings. Nonpoint sources are all other sources of radioactive air emissions and include, but are not limited to, unventilated buildings, outside waste storage sites, open burn sites, and firing sites. Both of these source types can provide a dose to a member of the public. Therefore, their emissions must be accounted for when determining compliance with the dose standard (10 mrem/yr) provided in §61.92 of USEPA 1993a. However, the treatment of point sources and nonpoint sources in the regulations is very different. Very specific requirements and procedures are given for dealing with point sources, but no methodologies are provided for nonpoint sources. LANL has chosen, with USEPA's concurrence, to account for nonpoint sources on a site-wide basis using environmental monitoring. Unlike point sources, individual nonpoint source identification, characterization, sampling, and modeling are not necessary using this approach. Therefore, nonpoint sources will be addressed separately from point sources.

POINT SOURCES

Point sources have been defined in the FFCA as "those sources of airborne radionuclides that discharge their effluent through a forced ventilation system via a single point" (USDOE and USEPA 1996). The following specific topics will be discussed for point sources: historical sampling systems, source identification, source characterization, major source sampling, and minor source emission estimates and periodic confirmation.

Historical Sampling Systems

Sampling has been conducted at many stacks at the Laboratory for over 20 years. All release points that are, or have been, required to have continuous sampling by Rad NESHAP have been sampled since at least 1989. All current release points are shown in Appendix B. However, these sampling systems were not designed in accordance with the requirements of ANSI 1970, nor were they located in the stack in accordance with the guidance of Appendix A, Method 1 of USEPA 1993b. In general, the probes had a single, small (¼-⅝ inch diameter) nozzle which sometimes led to superisokinetic sampling. These probes were essentially just a bent tube placed in the effluent stream. They were typically located in a convenient, accessible location without particular regard to flow and mixing characteristics at the sample site. Also, the sample transport lines were often lengthy and had numerous bends. These factors lead to the potential for undersampling of large particles.

Despite these shortcomings, these sampling systems can provide important, useful data for evaluating the release points in question. Some people have contended that since these

sampling systems did not meet the technical design requirements of Rad NESHAP, they did not provide "accurate" samples. This is not necessarily true. Since the deficiencies in the historic sampling systems only impact large particle[2] sampling, effluents that predominantly consist of small particles should still be providing representative samples. There are reasons to believe this is the case for the majority of the systems at the Laboratory. The most important of these reasons is that most exhaust systems handling radioactive air are filtered. These filters, whether High Efficiency Particulate Air (HEPA) filters or standard bag-type filters, generally have a high efficiency for removing large particles. Therefore, these filtered exhausts are expected to consist primarily of small particles that are not adversely impacted by the described sampling system deficiencies.

The assumption of small particle predominance is supported by comparing the results obtained from the old sampling systems with the results obtained from the new sampling systems. If these comparisons are made in facilities with fairly steady operations, the existence of significant numbers of large, radioactive particles should lead to an increase in measured emissions rates with the new systems. Selected exhaust systems are compared in this manner in Appendix C. This comparison has been conducted on those release points where a new probe (either ANSI-type rake or shrouded probe) has been installed and the exhaust system is otherwise unchanged. The comparison is a test of the null hypothesis: the hypothesis that the difference between the two means is zero. The test was conducted at a confidence coefficient of 95%. Note that only four of the release points fail the test (3-29-14, 3-29-19, 3-29-23, 3-102-18). In other words, in these four instances only has the average activity collected on the filter changed as a

[2] The terms "large particle(s)" and "small particle(s)" are used throughout this document. A small particle is

result of the new "improved" sampling systems. In fact, in these four instances the average activity collected on the filters has gone down, probably because of subisokinetic oversampling in the old sampling system. In addition, the measured sample filter activity both before and after installation of new sampling systems has typically been very low, often below statistical detection limits.

This comparison supports the contention that large, radioactive particles do not generally exist in sufficient quantity to have a significant impact on the validity of historical sampling results. Based on this conclusion, the Laboratory believes that the annual dose calculations prior to sampling system upgrades were reasonably accurate. In addition, decisions made based on these measurements are sound (see for example, "Source Characterization").

Source Identification

The identification of radioactive air point sources is required by §61.94(b)(4) of USEPA 1993a. In addition, this is a prerequisite to the source characterization specified in §61.93(b)(4)(ii) of USEPA 1993a and described below. The Laboratory uses two primary methods for this point source identification: periodic inventories and new project reviews. These processes have identified 117 current point sources at the Laboratory, which are listed in Appendix B.

Periodic inventories have been conducted at the Laboratory to determine the quantities of specific radionuclides at existing facilities. One purpose of these inventories is to establish

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considered one with an aerodynamic diameter smaller than about 3 μm AED.

which stacks and vents at the Laboratory have unsealed[3] radioactive materials. All of these stacks and vents are considered point sources and must be evaluated to determine if continuous sampling is required. The first comprehensive inventory was completed in 1992 and was updated in 1994/1995. Another inventory is underway and is expected to be complete before the end of calendar year 1996.

The 1992 inventory was completed over numerous months using several processes. At the time of this inventory, the facility manager concept had not yet been adopted at the Laboratory. Therefore, no single point of contact existed for each facility who was either knowledgeable of all operations at the facility or who could coordinate the inventory. Therefore, a variety of people were contacted for each facility. This included operational personnel, group leaders, radiological control technicians, building managers, etc., as appropriate. Information was obtained from these people through questionnaires, interviews, site visits, or other available information. Information collected included radionuclide type, physical form, quantity, containment, and process descriptions.

The inventory in 1994/1995 was conducted as an update to the 1992 inventory. Again, the facility manager concept had not yet been implemented throughout the Laboratory so numerous people were contacted. The information was collected using inventory update questionnaires and contacting a variety of facility personnel. Similar information as available in the 1992 inventory was obtained.

[3] The term "sealed" or "unsealed" sources is used as described in Appendix D of USEPA 1993a. Paragraph 2(a) of this appendix says: "Radioactive materials in sealed packages that remain unopened, and have not leaked during the assessment period should not be included in the calculation."

The inventory currently being conducted is again an update of the previous inventories. However, the Facility Manager concept has been instituted throughout the Laboratory. This has allowed the Laboratory's Air Quality Group (ESH-17) to utilize the facility managers as a single point of contact for the collection of information about their respective facilities. Copies of the 1994/1995 inventory results have been forwarded to the facility managers for their review and correction. It is anticipated that they will solicit input from many of the same people involved in earlier inventories, but the coordination should be improved with a single contact for each facility or group of facilities. The facility managers have been asked to update the inventory information to include identification of new radiological operations, modifications to the list of radionuclides (including quantity and form) at existing operations, and updated descriptions of operations.

In addition to these inventories, the Laboratory requires that all new or modified projects that could cause an increase in the release of radioactive materials be reported to ESH-17 for review (LANL 1991, 1995). ESH-17 reviews all of these projects to determine if a new point source will be created, or if a modification to an existing source will alter the sampling requirements for that source.

Source Characterization

A characterization of all point sources is required by §61.93(b)(4) of USEPA 1993a to determine the Potential Effective Dose Equivalent (PEDE). In particular, §61.93(b)(4)(i) of USEPA 1993a states: "Radionuclide emission measurements in conformance with the requirements of paragraph (b) of this section shall be made at all release points which have a potential to discharge radionuclides into the air in quantities which could cause an effective dose

equivalent in excess of 1% of the standard. All radionuclides which could contribute greater than 10% of the potential effective dose equivalent for a release point shall be measured. With prior USEPA approval, DOE may determine these emissions through alternative procedures. For other release points which have a potential to release radionuclides into the air, periodic confirmatory measurements shall be made to verify the low emissions". In addition, §61.93(b)(4)(ii) of USEPA 1993a states: "To determine whether a release point is subject to the emission measurement requirements of paragraph (b) of this section, it is necessary to evaluate the potential for radionuclide emissions for that release point. In evaluating the potential of a release point to discharge radionuclides into the air for the purposes of this section, the estimated radionuclide release rates shall be based on the discharge of the effluent stream that would result if all pollution control equipment did not exist, but the facilities operations were otherwise normal".

These requirements effectively split point sources into two categories: 1) Major point sources[4], which are those point sources with a PEDE greater than or equal to 0.1 mrem/yr (i.e. 1% of the standard), and 2) Minor point sources, which are those point sources with a PEDE less than 0.1 mrem/yr. Although the regulation clearly states this evaluation must assume "pollution control equipment did not exist" and "operations were otherwise normal", it does not otherwise provide any guidance or direction on methodologies that should be employed for determining estimated emissions or PEDE (USEPA 1993a). The Laboratory has therefore developed a multipronged approach for making these determinations. This approach is described in detail in

[4] The term "major point source" is a term defined in the FFCA for ease in distinguishing the two categories of point sources defined in 40 CFR §61.93(b)(4). It is not intended to be equivalent to the term "major source" as defined by

§2.1.1 of the FFCA and in the Air Quality Group procedure ESH-17-102, "Determination of Release Point Potential Effective Dose Equivalent". Essentially, this approach involves estimating the PEDE of a point source using one or both of two independent methodologies. The first method is based primarily on historical sampling results. The second method is an inventory based approach that conceptually follows the procedure described in Appendix D of USEPA 1993a.

Potential Emission Estimates Based on Historical Sampling Results

In developing the PEDE estimate based on historical sampling results, it is important to keep in mind the potential shortcomings of those sampling systems. These shortcomings were discussed above (see "Historical Sampling Systems"). Despite these potential shortcomings, this method can often provide the best estimate of PEDE. This method evaluated the measured releases of radioactivity from the point source over several years. The effect of filtration was then removed by dividing the measured release by the penetration (P) of the installed filtration system. Penetration is determined as follows:

$$\boxed{P = 1 - \eta}$$

where;

P = penetration

η = filter efficiency ($\eta = 0.9995$ for HEPA filters, $\eta = 0.80$ for ASHRAE-52 type filters)

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§112 of the CAA. The term "major source" as used in the CAA has no logical meaning with regards to 40 CFR 61, Subpart H.

This filtration effect was removed for each and every stage of filtration present in the exhaust system. The resulting emission estimates are shown in Supplement 1 of USDOE and USEPA 1996.

Estimates of PEDE based on historical sampling can only be developed for those point sources that have historically had sampling systems installed. There are several dozen point sources that have never had any sampling system installed. Therefore, estimates of PEDE for these sources have been based on inventories of radionuclides and estimated emission factors. In addition, this approach has been used as a redundant estimate for those point sources where historical sampling data exist.

Potential Emission Estimates Based on Inventories

The inventory based method estimates the release of radioactive materials by multiplying the inventory of material by emission factors presented in Appendix D of USEPA 1993a. These emission factors are 1.0/yr for gases, 1E-3/yr for particulate material, powders and liquids, and 1E-6/yr for solids.

Any material with a boiling point below 2000°C that was heated above 100°C was treated as a gas. Any material with a boiling point greater than 2000°C that was heated to within 1000°C of its boiling point was treated as a gas. Any material with a boiling point greater than 2000°C not heated to within 1000°C of its boiling point was treated as a liquid or solid, as appropriate. This treatment of heated materials is slightly different than the treatment in Appendix D of USEPA 1993a. This has been found acceptable by the

USEPA (USDOE and USEPA 1996). As the use of Appendix D for the determination of PEDE is not required by the USEPA, no alternative method approval was required.

Potential Effective Dose Equivalents

The resulting emission estimates from both methods were then modeled with CAP88 to determine the PEDE for each source. This determination was made to the sources' respective maximally exposed individual. This is more conservative than modeling the PEDE to LANL's MEI, which is generally much more distant.

If the facility's operations are relatively stable, the highest resulting PEDE based on historical emissions from the years evaluated were considered the PEDE based on historical emissions for that release point. If the facility's operations are changing, a judgment was made as to the most representative result from historical emissions. For instance, operations have been terminated or severely curtailed at some facilities. In this instance, a high result from several years ago may not be representative of current potential. Likewise, at facilities where operations are just starting, historically low emissions may not represent future potential.

The analysis resulted in two estimates of PEDE for historically sampled stacks: one based on historical sampling and one based on inventory. In general, the more conservative estimate of these two was used for determining if the point source required continuous sampling. In addition, sources with a PEDE between 0.01 mrem/yr and 0.1 mrem/yr were generally considered to require continuous sampling per Rad NESHAP. The resulting PEDE for each current point source is shown in Appendix B.

Major Sources

Again, major point sources are those point sources with a PEDE greater than 0.1 mrem/yr. The regulations require that these point sources be continuously sampled. In addition, the measurements at these point sources must account for all radionuclides that could contribute 10% or more of the PEDE (USEPA 1993a). This "rule" defines the radionuclides of interest at each stack.

At most major point sources at LANL this means that certain isotopes of uranium, plutonium, and other alpha emitters must be measured. At some major point sources this "10% rule" leads to the measurement of beta or gamma emitting radionuclides. The alpha and beta emitting radionuclides are exhausted in particulate form. The beta and gamma emitting radionuclides may be exhausted in particulate, vapor, or gaseous form. Both the decay scheme of the radionuclides of interest and the physical form were considered in the design of the associated sampling system. The systems are described below.

Sampling Systems

Under "emission monitoring and test procedures", Rad NESHAP calls out two references for radionuclide monitoring and extraction. The first citation states, "Reference Method 1 of appendix A part 60 shall be used to select monitoring or sampling sites". The second citation states, "The effluent stream shall be directly monitored continuously with an in-line detector or representative samples of the effluent stream shall be withdrawn continuously from the sampling site following the guidance

presented in ANSIN13.1-1969 'Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities' (including the guidance presented in appendix A of ANSI N13.1)" (USEPA 1993a).

For sample system design, ANSI (1970) states clearly that "some interpretation and judgment must be made in many situations which cannot be specifically described." In addition, it states the "appendices are not a part of . . . ANSI (1970), but are included for information purposes only." However, USEPA (1993a) requires a sampling system to be designed "following the guidance" of ANSI 1970 and specifically incorporates Appendix A of that standard. This does not make clear whether the ANSI recommendations then becomes strictly mandatory or remain guidance. LANL has taken the position that ANSI 1970 remains a guidance document outlining the issues to be considered in the design of a sampling system. When deviating from the guidance presented by ANSI (1970), every attempt has been made to provide a technical justification.

Sampling Location

Sample locations are to be selected using the procedures in Appendix A, Method 1 of USEPA 1993b. This reference method provides a framework for site selection based on the distance from the sampling site to the nearest upstream and downstream flow disturbances. These distances are defined in terms of "duct diameters" (linear distance divided by the diameter of the duct). Characterization of the proposed sampling site is performed by evaluating the flow profile of the sampling site cross-

section. The closer to a flow disturbance that the site is, the more points must be used for this characterization. This concept is shown in Figure 1.

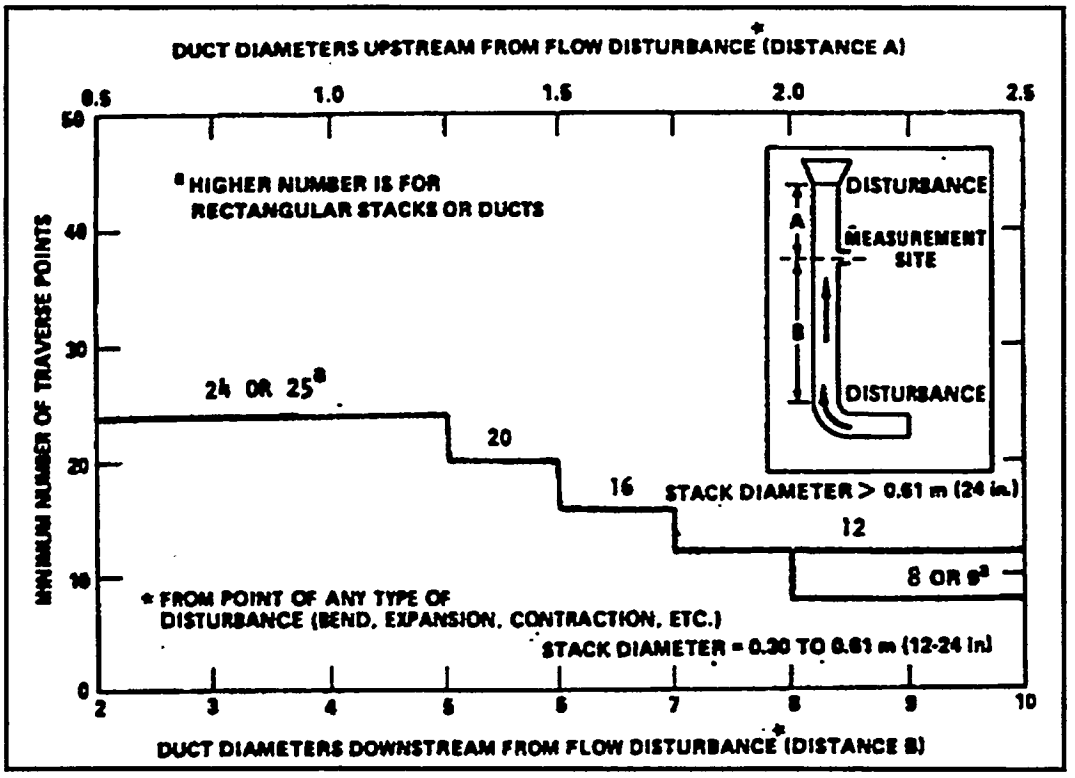

Figure 1: Particulate Traverse Point Requirements
(USEPA 1993b)

The techniques for characterization are clearly provided, but the criteria for evaluation of the characterization are not. That is, although the velocity profile at the sampling site is determined, there are no numerical criteria for deciding if the profile is adequate. There is an assumption inherent in the site selection process that the degree of flow development and mixing are directly related to the distance from disturbances. This, unfortunately, is not necessarily true. Hampl et al. (1986) have shown that flow development and mixing in a duct or stack are very dependent on the geometry of the

duct or stack. In some cases, very long runs of straight ducting (as many as fifty duct diameters) are required before mixing is achieved, whereas after two, out-of-plane, 90° bends in the duct mixing occurs fairly rapidly (less than two duct diameters). Therefore, the guidance presented does not always ensure an ideal sampling point. This is accounted for in the regulations by recommending the use of an ANSI-type probe (see "Sampling Probes").

Texas A&M University and LANL have developed an alternative approach to site selection that alleviates this weakness in site selection. In an USEPA approved alternative method, the sample site is characterized for both flow development and particulate mixing. The flow development is characterized as described in Method 1 with an established numerical performance criterion. Namely, the coefficient of variation (CofV)[5] must be less than 20%. The degree of particulate mixing is determined by injecting a surrogate aerosol upstream of the sampling point and measuring the surrogate concentration across a set of traverses at the sampling point. Numerical performance criteria are again established such that the CofV may not exceed 20% for the inside 2/3 area of the sampling site, and no point on the traverse may have a concentration outside the range of 70%-130% of the average concentration (LANL 1995b).

LANL uses the characterization information to determine the appropriate design for the sample probe. In the cases where these criteria are met, the sampling site is considered adequate for single point sampling and a shrouded probe is installed. If these criteria are not met, or if the measurements cannot be made due to physical constraints,

[5] The coefficient of variation is defined as the standard deviation of a sample divided by the mean of the sample.

single point sampling is not attempted and a standard ANSI-type rake is employed as described in "Sampling Probes".

Sampling Probes

Particulate Probes

The sampling probe is perhaps the most critical component in a sample system to ensure representative sampling when sampling for particulate matter. Improper design can easily lead to under-sampling of large particles. LANL has taken an aggressive approach in the design of sampling probes to maximize the representativeness of samples.

The discussion in Appendix A provides a strong basic framework for the concerns in sample probe design. The concept of isokinetic sampling is key in this framework. However, other characteristics of the bulk effluent (i.e., the degree of flow development and particulate mixing) are also critical to determine the design requirements of the probe. Using the site characterization methodology prescribed by USEPA (1993b), no assumptions can be made about these characteristics. These methods provide measurement procedures but no criteria. Therefore, USEPA (1993a) assumes nothing about flow development and mixing and calls out ANSI recommendations for probe designs. ANSI (1970) provides guidance for particulate sampling probes that utilize a multinozzle array to accommodate any deficiencies in the flow development or mixing. Figure 2 shows the typical layout of such a probe. The number of nozzles is determined based on

the size of the stack. This scheme has a significant drawback, however. As additional nozzles are added, the loss of particles increases due to impaction in the small nozzle inlet and tube bends. This is shown in Figure 3.

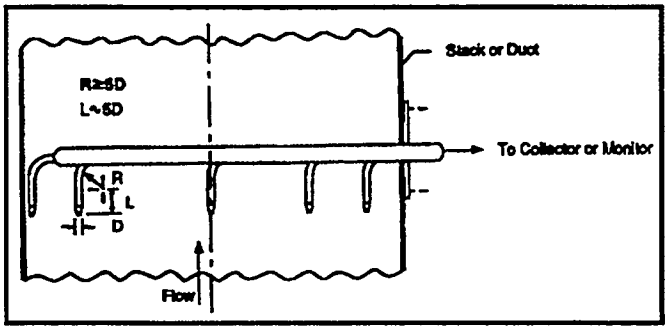

Figure 2: Typical ANSI-type Rake

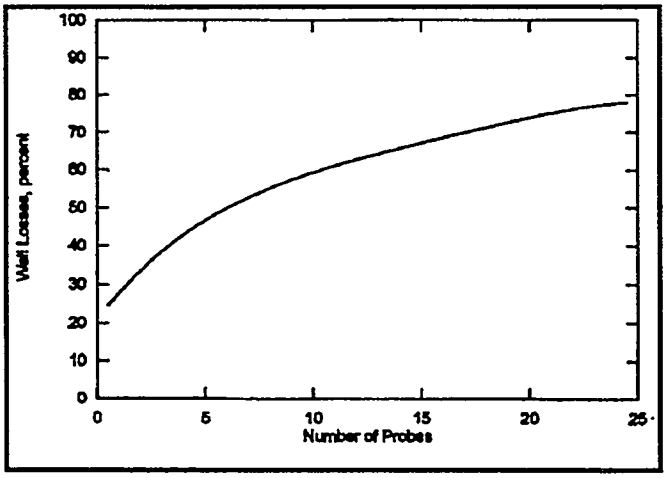

Figure 3: Effect of Number of Nozzles on Sample Loss

Texas A&M University and LANL have developed an alternative approach to particulate sampling probe design that quantifies flow development and mixing and improves on the concept of isokinetic sampling. As discussed in

"Sampling Location", this approach is based on establishing numerical criteria for sample site qualification. When properly qualified, a sample site can use single-nozzle sampling as opposed to multinozzle ANSI-type rakes. In addition, this alternative method employs the use of a shrouded nozzle as opposed to a simple isokinetic nozzle. This nozzle design reduces the loss of particulate matter at the nozzle inlet significantly. In addition, the shroud design ensures a high efficiency for particulate sample collection over a wide range of effluent flow, sample flow, and yaw angles (McFarland and Rodgers 1993). This approach has been approved by the USEPA (Nichols 1994).

LANL has used either ANSI-type rakes or shrouded probes, as appropriate. In some facilities it was not feasible to determine the characteristics necessary to meet the numerical performance criteria required for single point sampling with shrouded probes. In other facilities, the exhaust system is scheduled for reconfiguration in the near future and it was considered more timely and cost effective to install rakes. It is hoped that these systems can be back-fitted with shrouded probes in the future.

Tritium and Activated Gas Probes

The sampling of radionuclides in gaseous form is much simpler. As gaseous constituents have essentially no inertia, many of the concerns of particulate sampling do not apply. Therefore, any reasonable design of probe should be sufficient.

For activated gas sampling at LANL (i.e., at LANSCE), the Laboratory has chosen to install single-nozzle isokinetic sampling probes. Although isokinetic sampling is not technically necessary, it provides as good a sample as any other design. For tritium sampling at LANL, the Laboratory has chosen to use a simple extension of the sample lines. These stainless steel lines are bent 90° into the exhaust stream with a radius of bend equal to five times the tube diameter. This has provided a simple and economical installation that provides adequate sampling.

Transport Lines

Transport lines are the lengths of piping from the probe outlet to the sample collector inlet. Stainless steel has been used in most transport lines at the Laboratory. This helps ensure a smooth interior surface, minimizes corrosion concerns, and eliminates the build up of static charge on the transport line which can remove charged sample particles from the sample stream.

These transport lines, although typically required, can be a major source of sample loss. The design of the transport lines therefore requires some attention to minimize these losses. In non-particulate sampling systems, this normally only involves care to avoid unusual chemical reaction between the sample gas and the transport line. In particulate sampling systems, more attention is required to minimize the loss of sample due primarily to gravitational settling, Brownian diffusion, and inertial deposition.

Significant effort has been expended to minimize the length of transport lines when sampling for particulate material. In most cases, the probe outlet is connected to the sample collector through a short 90° bend only. The 90° bend is required to get the sample from the stack center to the stack exterior, but no other transport line is used, if possible.

In some cases, generally due to access restrictions, some additional transport line is necessary on particulate sampling systems. In these cases, the transport line has been carefully designed to minimize the loss of sample. Usually, the additional transport line is oriented vertically. This minimizes the impact of gravitational settling. The number of bends in the line are also minimized to avoid inertial deposition of large particles. Finally, the transport lines are designed to ensure smooth transitions from the probe to the transport line and the transport line to the sample collector.

Regardless, all particulate transport lines have been evaluated for large particle sample loss using the Deposition v2.0 computer code (Anand, et al. 1993). This computer code provides an estimate of particulate sample loss in the probe and transport line. In accordance with the alternative method approved by USEPA (LANL 1995b), all systems have been designed for minimum transport loss. A goal has been established for a minimum transport efficiency of 50%. All current systems have a transport efficiency of at least 50%.

An additional concern with the use of transport lines is the potential for condensation of the sample within the transport line. This can occur when relatively warm exhaust air is cooled as the transport line travels outside, especially at night. This

sample is not lost, as it will eventually evaporate and be reentrained when the outside air warms up during the day. However, this condensation can lead to a delay in accounting for the stack emissions. Therefore, where condensation of the sample in the transport line is a concern, insulation and heat tracing has been installed to maintain the sample gas above its dew point.

Sample Collectors

Samples must be collected in accordance with the principles described by ANSI (1970). Sections 5.2 and 5.3 of ANSI 1970 discuss the selection of both the collector itself and the sample collector holder. The selection or design of both of these items is important to ensure good sampling efficiencies. Three different sample collectors are used at LANL to account for the different form of the radionuclides to be collected. In all three cases, these sample collectors are exchanged weekly for off-line analysis.

The predominant collector is a glass-based filter paper used in particulate sampling systems. These systems typically sample at ~2 cfm. These filters have a greater than 99.95% efficiency for collecting 0.3 μm AED particles. Although standard filter holders are available commercially that will accommodate these filters, the Laboratory has designed special holders for better sampling efficiencies. The typical filter holder used in industry has a very small inlet (~⅜ inch diameter). The transition from the relative large diameter transport line (~1 inch diameter) to this filter holder can lead to large sample loss. The Laboratory's design incorporates a filter holder inlet the same diameter as the transport line. This ensures a smooth transition from the transport line to

the sample holder, thus minimizing the loss of large particles. Weekly changeout of these samples provides for timely analysis of the samples and minimizes self-shielding of the sample during alpha counting.

On some sampling systems, vapor materials must be sampled. These materials cannot be efficiently collected with a standard filter paper. In these cases a charcoal cartridge is used, generally in-line with and following the standard paper filter. These charcoal cartridges are commercially available. Although generally designed for the collection of radioiodine, they are effective for sampling the vapors present at the Laboratory. The holders for these charcoal cartridges are standard, commercially available holders. These samples are also exchanged weekly.

Tritium sampling is conducted at LANL using EG&G Labserco EL-700 bubblers. These instruments are designed to distinguish between the elemental form of tritium (HT or T_2) and the vapor form of tritium (HTO or T_2O). These instruments extract a small (~150 cc/min) sample from the stack or duct effluent. A pre-filter on the bubbler minimizes any interference from particulate matter. The sample gas is then bubbled through three consecutive vials of ethylene glycol. The ethylene glycol removes all water vapor from the sample gas, including the tritiated water vapor. Under normal circumstances, the third vial should collect little or no water vapor and should result in a near zero result from its tritium counting. This validates complete tritium collection. The vapor depleted sample gas is than passed over a heated palladium bed. This bed acts as a catalyst to oxidize the remaining elemental tritium to tritium oxide. This newly oxidized tritium is then passed through three more ethylene glycol vials to remove this vapor. The

first three vials contain all of the tritium oxide from the sample gas, and the second three vials contain all of the elemental tritium from the sample gas. As the ethylene glycol removes all water vapor (not just tritiated water vapor) from the gas stream, it is important that the vials be changed regularly to avoid expending the ethylene glycol. The vials are generally exchanged weekly.

Activated Gas Sampling

The Los Alamos Neutron Science Center (LANSCE) houses an 800 MeV, 1 mA proton accelerator. As a result of primary and secondary particle interactions with air, shielding materials, accelerator components, and experimental equipment, many potential activation products can be generated. These include gaseous, vapor, and particulate forms of radionuclides. The particulate, vapor, and tritium form radionuclides are collected as described above (see "Sample Collectors"). Activated gases from LANSCE are not "collected" for off-line analysis. The gaseous form of these radionuclides, as well as their short half-lives (less than 2 hours), preclude off-line analysis. Therefore, these radionuclides are monitored with a near real-time sampling system.

The activated gases generated by LANSCE account for over 95% of the dose to the Laboratory's maximally exposed individual (MEI). Therefore, special effort has been expended on the design and operation of the gas monitoring system at LANSCE. The primary constituents of the activated gas exhaust are shown in Table 1.

Nuclide	Decay Mode	Beta E_{max} (MeV)	Gamma Energy (MeV)	Half-life (min)	Fractional Composition
^{16}N	e^-	4.3	6.13	0.12	0.012
^{10}C	e^+	1.9	0.551, 0.72	0.32	0.039
^{14}O	e^+	1.8	0.551, 2.31	1.18	0.016
^{15}O	e^+	1.74	0.511	2.07	0.623
^{13}N	e^+	1.19	0.511	10.0	0.125
^{11}C	e^+	0.96	0.511	20.5	0.182
^{41}Ar	e^-	1.20	1.29	109.8	0.003

Table 1: Principle LANSCE Radionuclides

The two monitored stacks at LANSCE (53-3-3 and 53-7-2) each have a monitoring system that provides both continuous ion-chamber monitoring using the principles of Appendix B, Method 114, Analysis Method B-1 of USEPA 1993a for the total curies emitted from the stack and periodic determination of the radionuclide composition of these emissions by gamma spectroscopy using the principles of Appendix B, Method 114, Analysis Method G-1 of USEPA 1993a. Because of the unique nature of LANSCE emissions, measurements using Method B-1 alone are not sufficient to characterize emissions. Thus, Method G-1 is also needed to provide a nuclide-specific analysis and an ion-chamber calibration factor.

The essential components of the gas monitoring system consist of a high purity germanium (HPGe) gamma detector and sample can, a flow-through Kanne ionization chamber, electronics to collect data and monitor system status, and a pump system to maintain a constant air flow rate. The gamma system measures the concentration of radionuclides in the stack gas (μCi/cc) on a quasi-continuous basis, thus providing a means of calibrating the specific radionuclide quantities with the output of the ion chamber that continuously monitors the stack gas. When not in use for this purpose, the gamma system is

used to measure, by decay analysis, the composition fraction of the positron emitters in a grab sample of stack gas.

Vacuum Systems

Vacuum systems are employed on all sampling systems to withdraw the sample from the effluent stream in the exhaust system. The vacuum is supplied either by a dedicated vacuum pump or by the facility's vacuum system. These systems are connected to the clean side of the sample holder or chamber and generally have throttle valves to adjust the sample flow. The sample flow is monitored using in-line rotometers. The tritium bubblers have mass flow controllers to maintain stable sample flow. Sample flow is typically maintained at 2 cfm for particulate and vapor sampling and 150 cc/min for tritium sampling.

The vacuum lines from the sample holder or chamber to the vacuum source are typically stainless steel. Sizing of these lines is not critical other than to avoid overworking the vacuum source. A short length of flexible tubing is employed directly between the holder and the vacuum tubing to allow for manipulation of the holder during sample changeout. The exhaust from dedicated vacuum sources is directed back to the exhaust system.

Radionuclide Analysis

Sample counting techniques follow the requirements of Appendix B, Method 114 of USEPA 1993a. This method provides general requirements for counting techniques

depending on whether the radionuclide of interest is an alpha, beta, or gamma emitter. These requirements for some radionuclides are listed in Table 2. The approved methods are as follows:

A-1: Radiochemistry-Alpha Spectrometry

A-2: Radiochemistry-Alpha Counting

A-3: Direct Alpha Spectrometry

A-4: Direct Alpha Counting (Gross alpha determination)

A-5: Chemical Determination of Uranium

A-6: Radon-222-Continuous Gas Monitor

A-7: Radon-222-Alpha Track Detectors

B-1: Direct Counting in Flow-Through Ionization Chambers

B-2: Direct Counting With In-line or Off-line Beta Detectors

B-3: Radiochemistry-Beta Counting

B-4: Direct Beta Counting (Gross beta determination)

B-5: Liquid Scintillation Spectrometry

G-1: High Resolution Gamma Spectrometry

G-2: Low Resolution Gamma Spectrometry

G-3: Single Channel Gamma Spectrometry

G-4: Gross Gamma Counting

The regulation allows the use of the gross counting techniques "only when it is known that the sample contains a single radionuclide or the identity and isotopic ratio of the radionuclides in the effluent stream are well known" (USEPA 1993a). LANL uses a

combination of gross counting techniques and radionuclide discriminating techniques, as appropriate.

Filter papers at LANL, excepting those point sources at LANSCE which have no potential to exhaust alpha or pure beta emitting particulate matter, are counted on-site for gross alpha and gross beta activity using a Berthold LB770 gas flow proportional counter. This technique is consistent with methods A-4 and B-4. This provides a measure of total activity. To determine the isotopic ratios of ^{238}Pu, ^{239}Pu, ^{234}U, ^{235}U, ^{238}U, ^{241}Am, ^{210}Pb, and ^{90}Sr particulate matter, the weekly sample filters are composited at least annually for analysis by an off-site laboratory by dissolving the filter in an acid solution, chemically separating and concentrating the radionuclides onto sample planchets, and counting the concentrated samples with an alpha spectrometer. This follows methods A-1 and B-3. In addition, all filter papers are analyzed on-site by gamma spectroscopy with a high purity germanium detector following method G-1.

Charcoal cartridges are analyzed on-site by gamma spectroscopy with a high purity germanium detector following method G-1. This is the only required analysis for charcoal cartridges because the vapor form radionuclides of concern at LANL are all gamma emitters.

Tritium samples from the bubblers are analyzed on Packard/Canberra 2500 series liquid scintillation counters. This technique is consistent with method B-5.

The minimum detectable activity level (MDL) for these analyses is defined at two standard deviations above the average background count rate. Acceptable MDLs are set no higher than a corresponding release rate that could result in a dose of 0.01 mrem/yr to

any member of the public. This corresponds with the requirements of §61.93(b)(4)(i) of USEPA 1993a which requires that all radionuclides that could contribute greater than 10% of the stacks potential be accounted for. The actual tritium, alpha, and beta MDLs are actually set well below this point since they are readily attainable.

Radionuclide	Approved methods of analysis
Am-241	A-1, A-2, A-3, A-4
Ar-41	B-1, B-2, G-1, G-2, G-3, G-4
Ba-140	G-1, G-2, G-3, G-4
Br-82	G-1, G-2, G-3, G-4
C-11	B-1, B-2, G-1, G-2, G-3, G-4
C-14	B-5
Ca-45	B-3, B-4, B-5
Ce-144	G-1, G-2, G-3, G-4
Cm-244	A-1, A-2, A-3, A-4
Co-60	G-1, G-2, G-3, G-4
Cr-51	G-1, G-2, G-3, G-4
Cs-134	G-1, G-2, G-3, G-4
Cs-137	G-1, G-2, G-3, G-4
Fe-55	B-5, G-1
Fe-59	G-1, G-2, G-3, G-4
Ga-67	G-1, G-2, G-3, G-4
H-3 (H2O)	B-5
H-3 (gas)	B-1
I-123	G-1, G-2, G-3, G-4
I-125	G-1
I-131	G-1, G-2, G-3, G-4
In-113m	G-1, G-2, G-3, G-4
Ir-192	G-1, G-2, G-3, G-4
Kr-85	B-1, B-2, B-5, G-1, G-2, G-3, G-4
Kr-87	B-1, B-2, G-1, G-2, G-3, G-4
Kr-88	B-1, B-2, G-1, G-2, G-3, G-4
Mn-54	G-1, G-2, G-3, G-4
Mo-99	G-1, G-2, G-3, G-4
N-13	B-1, B-2, G-1, G-2, G-3, G-4
O-15	B-1, B-2, G-1, G-2, G-3, G-4
P-32	B-3, B-4, B-5
Pm-147	B-3, B-4, B-5

Radionuclide	Approved methods of analysis
Po-210	A-1, A-2, A-3, A-4
Pu-238	A-1, A-2, A-3, A-4
Pu-239	A-1, A-2, A-3, A-4
Pu-240	A-1, A-2, A-3, A-4
Ra-226	A-1, A-2, G-1, G-2
S-35	B-5
Se-75	G-1, G-2, G-3, G-4
Sr-90	B-3, B-4, B-5
Tc-99	B-3, B-4, B-5
Te-201	G-1, G-2, G-3, G-4
Uranium (total alpha)	A-1, A-2, A-3, A-4
Uranium (Isotopic)	A-1, A-3
Uranium (Natural)	A-5
Xe-133	G-1
Yb-169	G-1, G-2, G-3, G-4
Zn-65	G-1, G-2, G-3, G-4

Table 2: Approved Analysis Methods

(USEPA 1993a)

Monitors

In some cases, particularly in those facilities designated by the USDOE as "nuclear facilities", stack continuous air monitors (CAMs) have been installed. These instruments provide near real-time assessment of the radionuclide stack concentrations. This provides a particularly useful operational tool to identify potential problems with operations. However, the CAMs have several drawbacks in comparison to fixed filter sampling with off-line analysis. The CAMs do not have the sensitivity that off-line analysis can obtain, nor are they as accurate. In addition, commercially available CAMs generally cannot provide radionuclide specific analysis, rather they provide gross measurements of alpha activity, beta activity, or gamma activity. Therefore, these CAMs

are generally not utilized as record samplers. On the other hand, they do provide a valid backup and confirmation to fixed air sampling results (filter papers, charcoal cartridges, and bubblers). Therefore, the installation of these CAMs has followed the same requirements as the record samplers.

A critical exception to the exclusion of CAMs as record samplers is the sampling installation at LANSCE. As discussed above (see "Activated Gas Sampling"), the activated gases generated at LANSCE cannot reasonably be collected for off-line analysis. Therefore, the custom built gas monitoring system described in "Activated Gas Sampling", a form of CAM, is used as the record monitor.

Minor Sources

Minor point sources are those point sources with a PEDE less than 0.1 mrem/yr. The regulations do not require that these point sources be continuously sampled. However, other requirements do exist.

Emission Estimates

The emissions from minor point sources must be accounted for in the comparison with the 10 mrem/yr standard. Since they are not sampled, other methods must be used. No methods are provided in the regulations, so the Laboratory is free to develop technically sound methods, subject to the review of the USEPA. In addition, the concept of "grab samples" is discouraged (USEPA 1993a).

The Laboratory has defined a set of methods in the FFCA that the USEPA has reviewed and agreed are acceptable. These methods follow the same concepts as the determination of point source PEDE given above. However, in this case, the effects of filtration may be included. In general, these estimates are solely based on inventory determinations and emission factors.

Periodic Confirmation

For minor point sources, the regulations also state: "periodic confirmatory measurements shall be made to verify the low emissions" (USEPA 1993a). The methodology for periodic confirmatory measures is not given. Again, the concept of "grab samples" is discouraged. Therefore, the Laboratory will use the emission estimates, with the effect of filtration excluded, to periodically verify that the emissions from minor point sources remain low and are not subject to the continuous sampling requirements. In addition, the Laboratory's environmental monitoring system (see "NONPOINT SOURCES" below) provides a very effective confirmation of total emissions from the Laboratory. Any particulate or tritium emissions from minor point sources are accounted for in environmental monitoring. Therefore, any significant release from minor point sources will show up as a major discrepancy between the modeled results from point sources and the environmental measurements.

Modeling and Dose Determinations

For each source of radioactive air emissions, the measured or estimated emissions must be modeled using CAP88. CAP88 conducts both air dispersion modeling and radionuclide concentration to dose conversions. This modeling is conducted without modification to the computer code. Input files are modified to account for local meteorology using locally collected data. This ensures more accurate dispersion results and is in accordance with the recommendations of the user's manual for CAP88. In addition, a variety of population files can be used, each representing population distances and bearings relative to specific Laboratory sites.

No modifications are made to the results of this modeling. This results in several very conservative effects. Perhaps the most significant of these is that the modeling is evaluated at "a residence, school, business or office" (USEPA 1993a). At LANL, this results in the evaluation of dose at an infrequently occupied building as if it were occupied twenty-four hours per day, 365 days per year.

NONPOINT SOURCES

Requirements

Nonpoint sources have been defined in the FFCA as "those diffuse and fugitive sources of airborne radionuclides that are not discharged through a forced ventilation system via a stack or vent" (USDOE and USEPA 1996). As with minor point sources, the regulations do not call for continuous sampling of nonpoint sources. Again, however, the emissions from nonpoint sources must be accounted for in comparisons with the 10 mrem/yr standard. No methodologies are provided for these determinations. This sets the Laboratory free to develop technically sound methods, subject to the review of the USEPA. This has been done in the FFCA.

In the FFCA, the Laboratory has detailed the advantages and disadvantages of attempting to identify, characterize, and estimate emissions from individual nonpoint sources as compared with conducting environmental monitoring. It has been determined that environmental measurements provide more accurate, comprehensive, and timely information than any program of source specific measurements or estimates. Therefore, a portion of the Laboratory's existing environmental monitoring system (AIRNET) will be used for this evaluation. However, the environmental monitoring system cannot sample the short-lived activated gases that are generated at LANSCE (see Table 1). As these radionuclides are a significant nonpoint source, as much as 0.5 mrem/yr to the LANL MEI, they must be accounted for separately. This is done using local measurements and modeling with CAP88.

Environmental Monitoring

The regulations have provided some criteria for the use of environmental monitoring in lieu of stack sampling for demonstration that the 10 mrem/yr standard is not exceeded. Although this is not strictly the purpose of the environmental monitoring presented here, these criteria provide a valid framework for developing our environmental sampling requirements. These requirements, presented in §61.93(b)(5) of USEPA 1993a are as follows:

i) 'The air at the point of measurement shall be continuously sampled for collection of radionuclides.'

ii) 'Those radionuclides released from the facility, which are the major contributors to the effective dose equivalent must be collected and measured as part of the environmental measurement program.'

iii) 'Radionuclide concentrations which would cause an effective dose equivalent of 10% of the standard shall be readily detectable and distinguishable from background.'

iv) 'Net measured radionuclide concentrations shall be compared to the concentration levels in Table 2 of appendix E to determine compliance with the standard. In the case of multiple radionuclides being released from a facility, compliance shall be demonstrated if the value for all radionuclides is less than the concentration level in Table 2, and the sum of the fractions that result when each measured concentration value is divided by the value in Table 2 for each radionuclide is less than 1.'

v) 'A quality assurance program shall be conducted that meets the performance requirements described in appendix B, Method 114.'

vi) 'Use of environmental measurements to demonstrate compliance with the standard is subject to prior approval of EPA. Applications for approval shall include a detailed description of the sampling and analytical methodology and show how the above criteria will be met.'

System Description

This monitoring program consists of seventeen of the AIRNET stations around the Laboratory's perimeter. The AIRNET system has been in operation since the 1970's, and twelve of the existing stations were used as part of the NESHAP compliance program. In addition, four new stations were added and a previously discontinued station was restarted. All of these stations continuously sample the ambient air for particulate matter and tritium oxide. Particulate matter is collected on a filter paper with low natural uranium content (i.e., not fiberglass). A sample flow of 4 cfm is maintained through this filter paper which is collected approximately every two weeks. Tritium is collected by drawing 200 cc/min of air through a silica gel column to remove all water vapor. This column is also exchanged approximately every two weeks.

These sample collectors are maintained in a weather house for protection from the environment. Included in these weather houses are the sample collectors, sample pumps and motors, flow meters, and vacuum actuated timers to record total sample time. The systems are maintained to ensure at least 90% availability.

Sample Station Siting

Sample stations are sited using two general sets of criteria. These criteria will be referred to as "macro-siting" and "micro-siting" criteria. These criteria are intended to ensure the requirements of §61.93(b)(5)(i) and (ii) of USEPA 1993a are met.

Macro-siting

Macro-siting criteria are used to determine what regions adjacent to the Laboratory require sampling. To identify these regions, a network analysis was performed. The primary consideration of this network analysis was the placement of samplers in all sectors that contain a potential MEI. Assumptions and criteria for this analysis included the following:

1. Maximum off-site concentrations for nonpoint source emissions will occur at the site boundary since all such emissions are considered ground-level releases.
2. All populated areas around LANL will be sampled.
3. All residence/business "islands" within the LANL boundary will be sampled.
4. A standard 16-sector radial array (22.5-degree sector angle) from potential release sites will be used to evaluate potential MEI sectors.

5. As a conservative measure, samplers will also be located in the sectors adjacent to the sectors of maximum off-site impact if any population exists in those sectors.

From this network analysis, seventeen potential MEI sectors were identified. These seventeen locations have a sample station on or near the LANL boundary between the release point and the potential MEI sector for any given nonpoint source within LANL. These stations are shown in Table 3. This arrangement effectively provides a "wall" of samplers along the LANL boundary and all adjacent populated areas around or enclosed by LANL. Such a large number of sampling sites greatly exceeds the USEPA's (1993a) requirement to determine the dose at the single LANL MEI each year. As long as LANSCE operates, the LANL MEI location will always be in the sector where the East Gate sampler is located. However, if LANSCE were to cease operation, the MEI sector could shift each year between several locations. Consequently, LANL has chosen to sample all MEI sector possibilities, even though most sites are unlikely ever to be selected as the LANL MEI.

Identification (#)	Location New Mexico State Plane Coordinates	
	Northing	Easting
48th Street (6)	1776556	476714
LA Hospital (61)	1776438	480506
Shell Station (7)	1775843	483461

Identification (#)	Location New Mexico State Plane Coordinates	
	Northing	Easting
McDonald's (8)	1774932	485436
Trinity Bible Church (62)	1775964	488473
LA Airport (9)	1776244	492348
East Gate (10)	1773918	498438
Royal Crest (12	1772810	485106
County Landfill (32)	1774936	480120
DP Road (20)	1774793	490964
LA Canyon (60)	1774810	482878
Well PM-1 (11)	1768257	507327
WR Pinon School (13)	1754745	511266
WR Fire Station (15)	1756934	513180
WR Nazarene Church (16)	1754506	508401
WR Pajarito Acres (14)	1743891	512275
WR Monte Rey South (63)	1750376	507380

Table 3: Identification and Location of Ambient Air Samplers

Additionally, the three regional background AIRNET samplers located in Espanola, Pojoaque, and Santa Fe will be included in the program for background determinations other than uranium. Natural background uranium levels at the regional background stations are actually higher than the uranium levels near LANL. If the regional values were used for background determinations, the reported MEI sector dose from LANL uranium operations would usually be negative. Therefore, for the special case of uranium background determinations,

the AIRNET perimeter station with the lowest annual uranium concentration will be used as a more conservative indicator of natural background uranium levels.

Micro-siting

Micro-siting criteria are established to ensure a representative sample of the ambient air is obtained at each sample station. These criteria establish an acceptable sampling location within the region to be sampled. These criteria include the following (LANL 1995d):

1. **Favorable surface characteristics:** To reduce the loading of filters by particulate matter, ideal sites will have minimal extraneous material prone to air suspension in the immediate area.
2. **Trees acceptable:** According to guidance from 40 CFR Part 58, samplers "must be 10 m from the dripline when the tree(s) act as an obstruction."
3. **Distance to obstructions (primarily buildings) greater than two times the height of the sampler:** The distance between the sampler and the obstruction must be at least twice the height difference between the sampler and the obstruction (equivalent to a rise angle from the sampler to the top of the potential obstruction of approximately 27°).
4. **Unrestricted airflow in 270° arc containing source direction:** The object (excluding trees, which are addressed under criterion #2) must not fall within the 270° arc, relative to the sampler, that contains the specific source that is being monitored (40 CFR Part 58).
5. **Good topographic location:** The area surrounding a site should be as level and flat as possible.

Radionuclides of Interest

LANL air emission reports for prior years provide a detailed list of the LANL site radionuclides of potential interest (USDOE 1991, 1992, 1993B, 1994, 1995, 1996). These are shown in Table 4.

H-3	O-15
C-10	Ar-41
C-11	P-32
N-13	Mixed fission/activation products
N-16	U-234/235/238
O-14	Pu-238/239/240/241

Table 4: Environmental Radionuclides of Interest

By far the most dosimetrically significant of these radionuclides are the activated gases of carbon, nitrogen, oxygen, and argon, all coming from LANSCE. These radionuclides typically contribute several mrem combined annual dose to the LANL MEI (from all sources, point and nonpoint). The next most dosimetrically significant radionuclides are U (mixed), ^{3}H, and Pu (mixed). Typically, none of these latter radionuclides, nor any of the remaining radionuclides, contribute as much as 1 mrem/year to either the LANL MEI or to any other person. Tritium, uranium, and plutonium were the most "significant" of the other emissions. These very small doses are contributed almost entirely by isotopes of uranium (approximately 0.1 mrem), with tritium contributing a dose about ten times lower, and isotopes of plutonium contributing a dose ten times less than tritium.

Thus, for NESHAP compliance determinations using environmental measurements for nonpoint sources and in compliance with §61.93(b)(5)(iii) of USEPA 1993a, LANL will consider tritium and isotopes of uranium and plutonium to be "significant," even though they are not expected to contribute a combined dose exceeding

1.0 mrem/year to any MEI. This is a conservative approach that has been reviewed and accepted by the USEPA.

Radionuclide Analysis

Individual particulate filters from each sampling location are counted for gross radioactivity (alpha and beta) by gas flow proportional counting and for gamma-emitting radionuclides by gamma spectroscopy. These alpha and beta prompt counts are done only to provide an early indication of unexpected types or quantities of radioactive materials that may be present from an unplanned release or unusual occurrence and are not used in dose determinations. These methods comply with Appendix B, Method 114, Analysis Methods A-4, B-4, and G-1 of USEPA 1993a.

On a quarterly basis, a composite sample of biweekly particulate filters is submitted for radiochemical analysis. The filters are analyzed for ^{238}Pu, $^{239/240}Pu$, ^{234}U, ^{235}U, ^{238}U, and ^{241}Am by dissolving the filter in an acid solution, chemically separating and concentrating the radionuclides onto sample planchets, and counting the concentrated samples with an alpha spectrometer. The method complies with Appendix B, Method 114, Analysis Method A-1 of USEPA 1993a. Only selected stations receive ^{241}Am analyses (americium is a daughter of plutonium); these stations include background stations and selected other stations near sites that contain plutonium or plutonium-contaminated waste.

After collection, the biweekly silica gel sample are heated to distill and collect the water, which is analyzed for tritium by liquid scintillation counting to comply with Appendix B, Method 114, Analysis Method B-5 of USEPA 1993a.

The result of these analyses is that all radionuclides that the Laboratory has defined as "significant" in accordance with §61.93(b)(5)(iii) of USEPA 1993a are measured using USEPA approved methods. Again, it is noteworthy that no radionuclides from nonpoint sources truly meet this definition.

Dose Calculations

The measured net annual average ambient airborne concentrations of the "significant" radionuclides will be determined for the MEI and compared with Appendix E, Table 2 of USEPA 1993a. Using the table values, the concentrations will be converted to equivalent dose using the valid assumption that each table value is equivalent to 10 mrem/year from all appropriate pathways. This complies with the requirements of §61.93(b)(5)(iv) of USEPA 1993a.

It is important to note that these dose calculations effectively cause point source emissions (other than LANSCE activated gases) to be double counted. This is because point source emissions are accounted for by continuous stack sampling for major point sources or emission estimates for minor point sources. However, the environmental monitoring stations will also measure the impact from these sources. The Laboratory does not consider this conservative effect to be significant because the total annual dose

to the MEI from particulate and tritium emissions is small (approximately 0.05 mrem in 1995).

The operation of these environmental sampling stations has been incorporated into LANL's complete air quality program for quality assurance. This ensures compliance with §61.93(b)(5)(v) of USEPA 1993a. Finally, although prior approval in accordance with §61.93(b)(5)(vi) of USEPA 1993a is not technically required because environmental monitoring is not being used in lieu of stack sampling, the Laboratory has received USEPA's concurrence in the FFCA (USDOE and USEPA 1996).

Activated Gas Nonpoint Sources

CAP88 is used to determine any activated gas dose from nonpoint sources to the MEI. The activated gas emission used in CAP88 is determined by measuring the activated gas concentrations with ion chambers representatively placed in buildings where the emissions originate and conservatively estimating the diffusion of the activated gas into the environment. Currently, this is only necessary at LANSCE. No other significant source of activated gases exist at LANL.

QUALITY ASSURANCE

USEPA (1993a) requires a comprehensive quality assurance (QA) program to document the adequacy of this program. The Laboratory has implemented a tiered QA program to accomplish this. The Air Quality Group has a Quality Management Plan (QMP) that provides a broad description of how the group performs air quality activities for LANL. Under this document are a group of quality assurance project plans (QAPPs). These QAPPs are project specific and provide detailed requirements for each project. Their are currently five project plans that relate to compliance with Rad NESHAP. These include the following:

1. Quality Assurance Project Plan for LANSCE Radioactive Air Emissions Monitoring, 53 FMM 104-01.3.

2. Quality Assurance Project Plan for the Radiological Air Sampling Network (AIRNET), ESH-17-AIRNET.

3. Quality Assurance Project Plan for Tritium Stack Emissions Monitoring, ESH-17-TRIT.

4. Quality Assurance Project Plan for Radioactive Particulate and Vapor Stack Emissions Monitoring, ESH-17-PARTIC.

5. Quality Assurance Project Plan for Unmonitored Point Source Radioactive Air Emissions, ESH-17-UMS.

These five project plans provide more detailed information on all issues presented in this document. Included in these documents are descriptions of the project organization,

responsibilities, general training and certification requirements, general technical requirements, and data quality objectives. The project plans provide all information required by §4 of Appendix B, Method 114 of USEPA 1993a.

These five project plans are each supported by a myriad of implementing procedures. These procedures are intended to describe specific responsibilities and step-by-step descriptions of specific tasks. Examples of these include procedures covering sample collection and handling, chemistry data package evaluation, stack sampling design and installation, modeling and dose calculation, deficiency reporting and correction, program assessments, and many others. These procedures cover all aspects of the project.

In addition, all external organization (e.g., analysis laboratories) are required to demonstrate an adequate quality assurance program. All procedures performed by these organizations are reviewed by the Air Quality Group for adequacy. Each of these organizations is also subject to audit by the Air Quality Group, or EPA, to ensure adequate implementation of their QA program and procedures.

Appendix A

The Physics of Aerosols in Sampling Systems

Radioactive materials in the air exhaust of nuclear facilities can exist in several forms. These include particulate matter, gaseous radionuclides, water vapor, and other vapor materials. Particulate matter is typically the most common form, and it provides the greatest challenge in sampling. Therefore, a discussion of the behavior of particulate matter suspended in a gas (i.e., an aerosol) is appropriate. This discussion is only a cursory review of the physics of aerosols, but should provide a basis for understanding the discussions in the body of the document. For further information, Hinds (1982) provides a thorough presentation on aerosol physics.

The principle concern in extracting a sample of the radioactive effluent from a stack or duct is to ensure that a representative sample is obtained. This is the primary concern addressed by ANSI (1970) in its recommendations. Representative sampling is defined by ANSI as "faithfully showing the quality and characteristics of the entire volume from which a sample is drawn." In particular, the concern in sampling stacks for radioactive constituents is ensuring that the sample has a similar distribution of particle sizes as the entire volume. This is critical because a 10 μm AED particle has 10^6 times the activity of a 0.1 μm AED particle. Therefore, if the sample is depleted of large particles, the analysis will grossly underrepresent the activity in the entire effluent volume. The majority of the technical issues addressed in the ANSI guidance are intended to minimize the loss of these large particles.

Large particles are particularly difficult to sample in a representative manner because they have significant inertia. As a result, they can be deposited on the probe interior surfaces or lost through other mechanisms when the gas flow makes rapid changes in direction. The first of

these mechanisms addressed by ANSI (1970) are the effects of anisokinetic sampling. These effects are illustrated in Figure 4, Figure 5, and Figure 6.

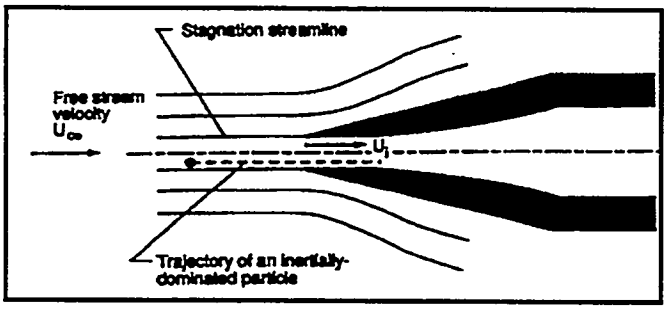

Figure 4: Isokinetic Sampling

Isokinetic sampling is defined as withdrawing a sample from an effluent such that the velocity of the withdrawn sample at the nozzle inlet plane is identical to the velocity of the bulk effluent in the stack or duct at the sample location. This is shown in Figure 4. The flow lines of the bulk effluent are not disturbed and any particles associated with a flow line entering the probe nozzle also enter the probe. Therefore, the concentration of any particle size in the nozzle is the same as in the bulk effluent.

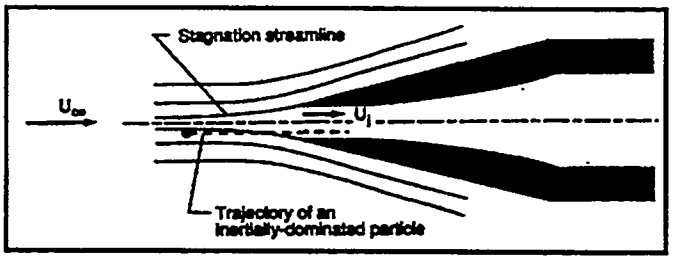

Figure 5: Subisokinetic Sampling

Figure 5 shows the effect of subisokinetic sampling; that is sampling such that the velocity at the nozzle inlet is lower than the velocity of the bulk effluent. In this case, some of the bulk effluent air streams bend around the probe nozzle to account for the lower velocity at the nozzle inlet. However, the associated larger particles (with correspondingly greater inertia) cannot change direction quickly enough and diverge from the path of the gas. These particles may enter the probe. As a result, the concentration of large particles in the probe is higher than the concentration of large particles in the bulk effluent. Because of the disproportionate radioactivity associated with large particles, this can lead to an activity concentration in the withdrawn sample that is much higher than in the bulk effluent.

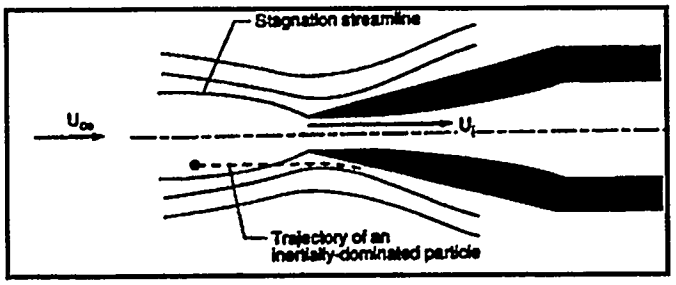

Figure 6: Superisokinetic Sampling

Finally, Figure 6 shows the effect of superisokinetic sampling; that is sampling such that the velocity at the nozzle inlet is higher than the velocity of the bulk effluent. Large particles are unable to change direction into the nozzle inlet along with the converging flow lines, thus depleting the sample of large particles. In this case, the withdrawn sample can have a significantly lower activity concentration than the bulk effluent. This case is particularly troublesome because it provides a non-conservative measure of the effluent radioactivity.

The effects of anisokinetic sampling drop off rapidly as particle size decreases. ANSI (1970) says, "except in very unusual situations, particles smaller than an aerodynamic diameter of about five microns are able to follow the streamlines of the air and the fractionation error is not great. In particular, when the ratio of probe inlet velocity to free stream velocity is 2.0 (very superisokinetic), 4 μm particles are still collected with an 86% efficiency.

Large particles are also affected in other components of a sampling system. In particular, large particles can be removed from the sample through gravitational settling in horizontal runs of sample transport lines, through inertial impaction in bends of transport lines, and through turbulent deposition throughout the sample system. All of these mechanisms are important due to the large fraction of activity that may be represented by large particles, if present. Therefore, these issues must be addressed in the design of any system intended to representatively sample particulate matter.

Small particles can also be removed from the sample stream. This removal will primarily be due to Brownian diffusion to the sample system walls. However, small particles represent a

much smaller fraction of the total activity and this mechanism is of correspondingly less importance. Regardless, minimizing the total length of the sample system will reduce this impact.

Appendix B

Radioactive Air Point Sources as of 06/01/96

ID[6]	Primary Radionuclides[7,8]	PEDE (mrem/yr)[9]	Sampling	Comments
3-16-CD1	^{3}H	< 0.001	None	Operations at this facility have been terminated.
3-16-8	^{3}H	< 0.001	None	Operations at this facility have been terminated.
3-16-9	^{3}H	< 0.001	None	Operations at this facility have been terminated.
3-16-14	^{3}H	0.003	None	Operations at this facility have been terminated.
3-16-16	^{3}H	0.006	None	Operations at this facility have been terminated.
3-16-B1	^{3}H	< 0.001	None	Operations at this facility have been terminated.
3-16-none	^{3}H	< 0.001	None	Operations at this facility have been terminated.
3-29-13	^{99}Tc	< 0.001	None	
3-29-14	$^{238/239}Pu$	15.7	Particulate	ANSI-type rake.
3-29-15	$^{238/239}Pu$	15.7	Particulate	ANSI-type rake.
3-29-17	$^{238/239}Pu$	< 0.001	None	
3-29-18	$^{238/239}Pu$	< 0.001	None	
3-29-19	$^{238/239}Pu$	15.7	Particulate	ANSI-type rake.
3-29-20	$^{238/239}Pu$	15.7	Particulate	ANSI-type rake.
3-29-21	$^{238/239}Pu$	< 0.001	None	
3-29-22	$^{238/239}Pu$	< 0.001	None	
3-29-23	$^{238/239}Pu$	15.7	Particulate	ANSI-type rake.
3-29-24	$^{238/239}Pu$	15.7	Particulate	ANSI-type rake.
3-29-26	$^{238/239}Pu$	< 0.001	None	
3-29-27	$^{238/239}Pu$	< 0.001	None	
3-29-28	$^{238/239}Pu$	15.7	Particulate	ANSI-type rake.

[6] The release point ID is in the form Technical Area-Building-Exhaust Stack. For example, 3-29-14 refers to TA-3, Building-29, Exhaust Stack-14.

[7] This represents the radionuclide(s) expected to account for greater than 10% of the PEDE. For many buildings with a low PEDE, this is not calculated for the whole building rather than stack by stack.

[8] PVAP: Particulate and Vapor Activation Products - Generally Gamma Emitters
Alpha: Various Alpha Emitters
D-38: Depleted Uranium

[9] Any PEDE less than 0.001 mrem/yr is shown as "<0.001" for brevity only. An actual PEDE has been developed.

ID[6]	Primary Radionuclides[7,8]	PEDE (mrem/yr)[9]	Sampling	Comments
3-29-29	$^{238/239}$Pu	15.7	Particulate	ANSI-type rake.
3-29-30	$^{238/239}$Pu	< 0.001	None	
3-29-31	$^{238/239}$Pu	< 0.001	None	
3-29-32	$^{238/239}$Pu	15.7	Particulate	ANSI-type rake.
3-29-33	$^{238/239}$Pu	15.7	Particulate	ANSI-type rake.
3-29-34	$^{238/239}$Pu	< 0.001	None	
3-29-35	$^{238/239}$Pu	< 0.001	None	
3-29-37	None	< 0.001	Particulate	ANSI-type rake. Sampled per DOE requirements only.
3-29-44	^{235}U/^{137}Cs	> 0.1	Particulate	ANSI-type rake.
3-29-45	^{235}U/^{137}Cs	> 0.1	Particulate	ANSI-type rake.
3-29-46	^{235}U/^{137}Cs	> 0.1	Particulate	ANSI-type rake.
3-34-1	^{3}H	< 0.001	None	
3-34-7	Alpha	< 0.001	None	
3-34-23	D-38	< 0.001	None	
3-34-33	D-38	< 0.001	None	
3-34-35	D-38	< 0.001	None	
3-35-1	^{235}U	0.05	Particulate	Operations at this facility have been terminated. ANSI-type rake.
3-40-25	^{3}H	< 0.001	None	
3-66-1	D-38	0.002	None	
3-66-6	D-38	< 0.001	None	
3-66-8	D-38	0.03	None	
3-66-9	^{238}U	< 0.001	None	
3-66-13	D-38	< 0.001	None	
3-66-18	D-38	< 0.001	None	
3-66-25	D-38	0.007	None	
3-66-26	D-38	< 0.001	None	
3-66-28	U-238	< 0.001	None	
3-66-43	Alpha	< 0.001	None	
3-102-18	D-38	0.4	Particulate	ANSI-type rake.
3-102-25	Alpha	0.005	None	
3-141-6	D-38	< 0.001	None	
3-141-9	D-38	< 0.001	None	
3-141-10	D-38	< 0.001	None	
9-21-3	^{3}H	< 0.001	None	
15-183-18	D-38	< 0.001		
15-233-1	D-38	< 0.001	None	
16-205-3	^{3}H	4030	Tritium	
16-248-none	^{3}H	< 0.001	None	
16-410-1	D-38	< 0.001	None	

ID[6]	Primary Radionuclides[7,8]	PEDE (mrem/yr)[9]	Sampling	Comments
16-410-2	D-38	< 0.001	None	
16-410-B1	D-38	< 0.001	None	
18-127-2	^{3}H	< 0.001	None	
18-168-1	$^{137m/140}Ba/^{140}La$	< 0.001	None	
21-5-7	$^{90}Sr/^{90}Y$	0.06	None	
21-150-1	$^{234/238}U$	0.07	None	
21-155-5	^{3}H	0.05	Tritium	
21-209-1	^{3}H	0.06	Tritium	
21-213-B1	^{3}H	< 0.001	None	
21-257-4	$^{241}Am/^{234}U$	< 0.001	None	
21-313-1	^{234}U	< 0.001	None	
21-315-1	$^{238/239}Pu$	0.04	None	
33-86-2	^{3}H	< 0.001	None	
33-86-6	^{3}H	< 0.001	Tritium	Sampled per DOE requirements only.
35-34-1	^{3}H	0.009	None	
41-1-4	^{3}H	0.004	Particulate, Tritium	Sampled per DOE requirements only.
41-4-17	^{3}H	< 0.001	Tritium	Sampled per DOE requirements only.
43-1-13	$^{32/33}P$	0.002	None	
46-24-none	^{235}U	< 0.001	None	
46-31-25	^{235}U	< 0.001	None	
46-31-41	^{235}U	< 0.001	None	
46-41-6	^{238}U	< 0.001	None	
46-154-5	^{234}U	< 0.001	None	
48-1-7	P/VAP	0.4	Particulate, Vapor	Shrouded Probe.
48-1-11	^{239}Pu	< 0.001	None	
48-1-15	$^{241}Am/^{239}Pu$	0.001	None	
48-1-35	^{75}Se	< 0.001	None	
48-1-45	$^{234}U/^{239}Pu$	< 0.001	None	
48-1-51	$^{90}Sr/^{90}Y$	0.01	None	
48-1-54	P/VAP	45.1	Particulate	Shrouded Probe.
48-1-60	P/VAP	0.3	Particulate, Vapor	Shrouded Probe.
48-1-61	^{239}Pu	< 0.001	None	
48-45-1	^{241}Am	0.004	None	
50-1-1	$^{241}Am/^{238/239}Pu$	< 0.001	None	
50-1-2	$^{241}Am/^{239}Pu$	160	Particulate	ANSI-type rake.
50-37-1	$^{90}Sr/^{90}Y$	17	Particulate	Shrouded Probe.
50-69-1	^{239}Pu	0.01	None	
50-69-2	^{239}Pu	0.008	None	

ID[6]	Primary Radionuclides[7,8]	PEDE (mrem/yr)[9]	Sampling	Comments
50-69-3	^{238}Pu	5.7	Particulate	Shrouded Probe.
53-1-16	P/VAP	0.003	None	
53-3-3	P/VAP	1740	Particulate, Vapor, Gas	Shrouded Probe.
53-7-2	P/VAP	3	Particulate, Vapor, Gas	Shrouded Probe.
53-30-none	P/VAP	< 0.001	None	
54-1009-1	^{241}Am/ ^{239}Pu	0.001	None	
54-1009-2	^{241}Am/ ^{239}Pu	0.001	None	
55-4-15	$^{238/239}$Pu	1.5E5	Particulate	ANSI-type rake.
55-4-16	$^{238/239}$Pu	1.1E5	Particulate, Tritium	ANSI-type rake.
59-1-4	$^{228/232}$Th	0.07	None	
59-1-14	$^{228/232}$Th	0.07	None	
59-1-30	$^{228/232}$Th	0.07	None	
59-1-B1	$^{228/232}$Th	0.07	None	
59-1-B2	$^{228/232}$Th	0.07	None	
59-1-B3	$^{228/232}$Th	0.07	None	
59-1-B4	$^{228/232}$Th	0.07	None	
59-1-B5	$^{228/232}$Th	0.07	None	
59-1-B6	$^{228/232}$Th	0.07	None	
59-1-B7	$^{228/232}$Th	0.07	None	

Appendix C

Comparison of Alpha Particle Collection between Old Sampling Systems and New Sampling Systems

I.D.	1995 Count	1995 Ave. Filter Activity (µCi)	1995 Variance	1996 Count	1996 Ave. Filter Activity (µCi)	1996 Variance	Pooled Variance	df[10]	t[11]
03002914	47	1.55E-06	1.62E-12	38	-1.10E-07	1.64E-13	9.71E-13	83	**7.73E+00**
03002915	47	8.26E-08	5.00E-13	38	7.93E-08	4.83E-13	4.92E-13	83	2.16E-02
03002919	46	1.41E-05	2.14E-10	38	8.09E-06	1.45E-10	1.83E-10	82	**2.03E+00**
03002920	47	6.91E-07	6.72E-12	38	2.57E-07	2.63E-13	3.84E-12	83	1.02E+00
03002923	47	1.30E-05	9.67E-11	38	5.67E-06	4.56E-11	7.39E-11	83	**3.89E+00**
03002924	47	2.45E-05	4.95E-09	38	1.44E-05	1.05E-09	3.21E-09	83	8.17E-01
03002928	47	1.70E-07	7.46E-13	38	1.98E-08	1.30E-13	4.71E-13	83	1.00E+00
03002929	47	5.59E-08	2.95E-13	38	7.97E-08	2.13E-13	2.58E-13	83	-2.15E-01
03002932	47	1.37E-07	6.84E-13	38	5.56E-08	1.08E-13	4.27E-13	83	5.71E-01
03002933	47	3.86E-07	9.58E-13	38	2.61E-07	4.21E-13	7.19E-13	83	6.71E-01
03002937	47	-1.83E-08	9.37E-13	38	-9.62E-08	8.91E-14	5.59E-13	83	4.77E-01
03002944	49	6.31E-07	1.04E-11	18	1.57E-07	1.09E-13	7.68E-12	65	6.21E-01
03002945	49	3.97E-07	8.93E-13	18	1.52E-07	2.62E-13	7.28E-13	65	1.04E+00
03002946	49	7.00E-08	4.66E-13	18	7.05E-08	3.93E-13	4.47E-13	65	-2.88E-03
03003501	50	1.42E-06	8.41E-12	17	1.78E-06	2.04E-12	6.84E-12	65	-4.91E-01
03010218	50	2.51E-06	7.18E-12	19	1.07E-06	1.39E-12	5.62E-12	67	**2.26E+00**
48000154	42	1.37E-07	8.20E-13	37	2.09E-08	1.30E-13	4.98E-13	77	7.29E-01
48000160	50	-1.17E-07	5.34E-13	20	-3.63E-08	3.72E-13	4.89E-13	68	-4.35E-01
50003701	42	-1.05E-07	3.76E-13	38	3.05E-07	2.75E-12	1.50E-12	78	-1.50E+00
50006903	41	5.38E-08	3.14E-13	38	2.09E-07	4.63E-13	3.85E-13	77	-1.11E+00

[10] df: degrees of freedom

[11] t_{α}=1.960, α=0.05

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