FINAL REPORT

Independent Technical Audit of Los Alamos National Laboratory for Compliance with the Clean Air Act, 40 CFR 61, Subpart H in 2001

DOJ File Number: 90-5-1749A
Case Name: *CCNS v DOE*

October 2002

"Setting the standard in environmental health"
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EXECUTIVE SUMMARY

This report documents results of the third independent audit of Los Alamos National Laboratory regarding compliance with the Clean Air Act, 40 CFR 61, Subpart H for the year 2001. It was concluded that Los Alamos National Laboratory was in compliance with 40 CFR 61, Subpart H for the year 2001. The offsite dose limit under the Clean Air Act regulation is 10 mrem yr^{-1} (0.1 mSv yr^{-1}), and Los Alamos National Laboratory reported a dose for 2001 of 1.84 mrem (18.4 μSv). This dose was confirmed by independent audit team calculations.

This audit was conducted by a team led by Dr. John E. Till and other members of the *Risk Assessment Corporation* research team as part of a Settlement Agreement and Consent Decree that resolved a 1994 lawsuit filed by Concerned Citizens for Nuclear Safety against the U.S. Department of Energy regarding Clean Air Act compliance status at Los Alamos National Laboratory. A separate independent group, the Institute for Energy and Environmental Research, monitored the audit for completeness, quality, and thoroughness on behalf of Concerned Citizens for Nuclear Safety. The audit team divided its work into four distinct areas that addressed the major elements of the compliance program. These areas were:

- Radionuclide usage and associated emission estimates for unmonitored point sources
- Major release point effluent monitoring
- Environmental compliance sampling for non-point sources
- Dose calculation.

A number of general issues, in addition to specific regulatory requirements, were evaluated for each area, including traceability of data to their original source, documentation supporting compliance, technical competence, quality assurance, and overall confidence of the audit team in the compliance program. Thus the audit encompassed a scope much broader than a typical evaluation for regulatory compliance.

LANL has made significant changes to improve the compliance program over the period of the audits, many in response to recommendations made within our audit reports. As in the past, we have included a number of additional recommendations in this report.

As with past audits, the public's role in the process was critical. This success is due to all parties who cooperated fully with the audit team in pursuing its goals. We continue to believe the open and thorough process that was followed can be used as a model for other facilities where risk to the public is being evaluated.

The audit team commends LANL for addressing the findings of the first and second audits and also for the concerted effort they have put forth during this audit to make it an open, thorough, and responsive process. Credit for this achievement is also due to Concerned Citizens for Nuclear Safety, who, as a citizens' organization, helped initiate the audits and design their format.

It is noteworthy that this audit was conducted under unusually difficult circumstances created by the events of September 11, 2001 and critical issues with regard to security at the Los Alamos National Laboratory throughout this year. The audit's success is a direct reflection of the professionalism and dedication to this process by all involved parties.

This was the third audit of its kind. According to the Consent Decree, if "…the third audit identifies substantive deficiencies with compliance with Subpart H that the auditor believes

require corrective actions, a fourth technical audit will commence no later than the end of calendar year 2003. The scope of the fourth audit shall be limited to determining whether necessary corrective actions identified in the third technical audit have been satisfactorily accomplished." The audit team has concluded there were no substantive deficiencies requiring corrective actions that justify having a fourth audit under the Consent Decree. Therefore, we consider that audit requirements under the Consent Decree have been met and are concluded with this report.

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FINDING

This audit concludes that Los Alamos National Laboratory was in compliance with 40 CFR 61, Subpart H for the year 2001. The audit team commends Los Alamos National Laboratory for their implementation of the recommendations resulting from the first and second audits, and the cooperation they have shown during this audit to make it an open, thorough, and responsive process. We also commend Concerned Citizens for Nuclear Safety and the audit monitors for their cooperation, ideas, and review of the audit process. Several suggestions for continued improvement are detailed within this report.

INTRODUCTION

On January 21, 1997, the U.S. Department of Energy (DOE) and the Concerned Citizens for Nuclear Safety (CCNS) settled a suit filed by CCNS (CCNS v. DOE, D.N.M. Civ. No. 94-1039M) concerning the status of compliance of Los Alamos National Laboratory (LANL) with 40 CFR 61.90–61.97, Subpart H, *National Emission Standards for Emissions of Radionuclides Other Than Radon From Department of Energy Facilities* (http://www.epa.gov/docs/epacfr40/chapt-1.info/subch-C/40P0061/40P0061H.pdf). As part of that settlement, referred to as the Settlement Agreement and Consent Decree, a series of comprehensive technical audits were to be performed.

As stated in the Consent Decree, the purpose of the technical audits was to verify whether LANL is in compliance with the Clean Air Act, 40 CFR 61, Subpart H, set forth in the Code of Federal Regulations (CFR). It was agreed in the settlement that *Risk Assessment Corporation (RAC)* would conduct the audits as the independent auditor. Dr. John E. Till, president of *RAC*, assembled a multidisciplinary team of scientists, the Independent Technical Audit Team (ITAT), for this purpose.

The first audit, covering the year 1996, began in June 1997 and a draft report was issued in May of 1998. The final report on the first audit was issued in November 1999 (Aanenson et al. 1999). The findings of the first audit indicated that LANL was not in compliance with 40 CFR 61, Subpart H for 1996. The second audit began in June 2000 and covered compliance for the year 1999. The final report for the second audit was issued in December 2000 and concluded that LANL was in compliance for 1999 (Aanenson et al. 2000).

This third audit began in June 2002 and evaluated compliance for 2001. According to the Consent Decree, if "…the third audit identifies substantive deficiencies with compliance with Subpart H that the auditor believes require corrective actions, a fourth technical audit will commence no later than the end of calendar year 2003. The scope of the fourth audit shall be limited to determining whether necessary corrective actions identified in the third technical audit have been satisfactorily accomplished." The audit team has concluded there were no substantive deficiencies requiring corrective actions that justify having a fourth audit under the Consent Decree. Therefore, we consider that audit requirements under the Consent Decree have been met and are concluded with this report.

The audit was observed and monitored by an independent scientific group on behalf of CCNS, the Institute for Energy and Environmental Research (IEER), to ensure the audit was objective and comprehensive. IEER neither performed a separate audit nor were they responsible for the results of the audit. Their role was to monitor the audit for completeness, quality, and thoroughness.

LANL has made significant changes and improvements in its compliance program since the last audit. Many of these changes respond directly to recommendations that were outlined in the previous two audit reports and to issues raised by CCNS and IEER. As a result, the current program for compliance is significantly improved. We include some additional suggestions for improvements in this report and assume that LANL will consider them carefully. We outline the recommendations resulting from this audit in Appendix A. This is only a summary of the issues, and the main report should be referred to for details.

The audit team commends LANL for the cooperation they have shown during this audit to make it an open, thorough, and responsive process. We continue to believe the 40 CFR 61 Subpart H compliance program at LANL and this audit process could be considered as a model for other DOE facilities. We again give credit to CCNS, who, as a citizens' organization, initiated actions that helped make this achievement possible.

It is noteworthy that this audit was conducted under unusually difficult circumstances created by the events of September 11, 2001 and other important issues with regard to security at LANL throughout this year. The audit's success is a direct reflection of the professionalism and dedication to this process by all participating parties. The audit team expresses its appreciation for the spirit of cooperation that made this audit possible.

Background

The Los Alamos National Laboratory is a Department of Energy facility located in Los Alamos County in north-central New Mexico. LANL is located atop a mesa and is surrounded by canyons, making the topography of the site very complex. The primary mission of this facility has always been research and development of nuclear weapons, including weapons development, fission and fusion, and weapons safety. Associated with this is the responsibility of maintaining environmental controls to limit the release of radionuclides into the environment. These controls and the practice and procedures that accompany them are some of the questions around which this audit was focused.

Figure 1 is a map of the LANL site. This map shows some of the major release points to air for offsite dose calculations, the locations of major unmonitored point sources, and the locations of some of the environmental samplers used to demonstrate compliance with the 40 CFR 61, Subpart H, regulations.

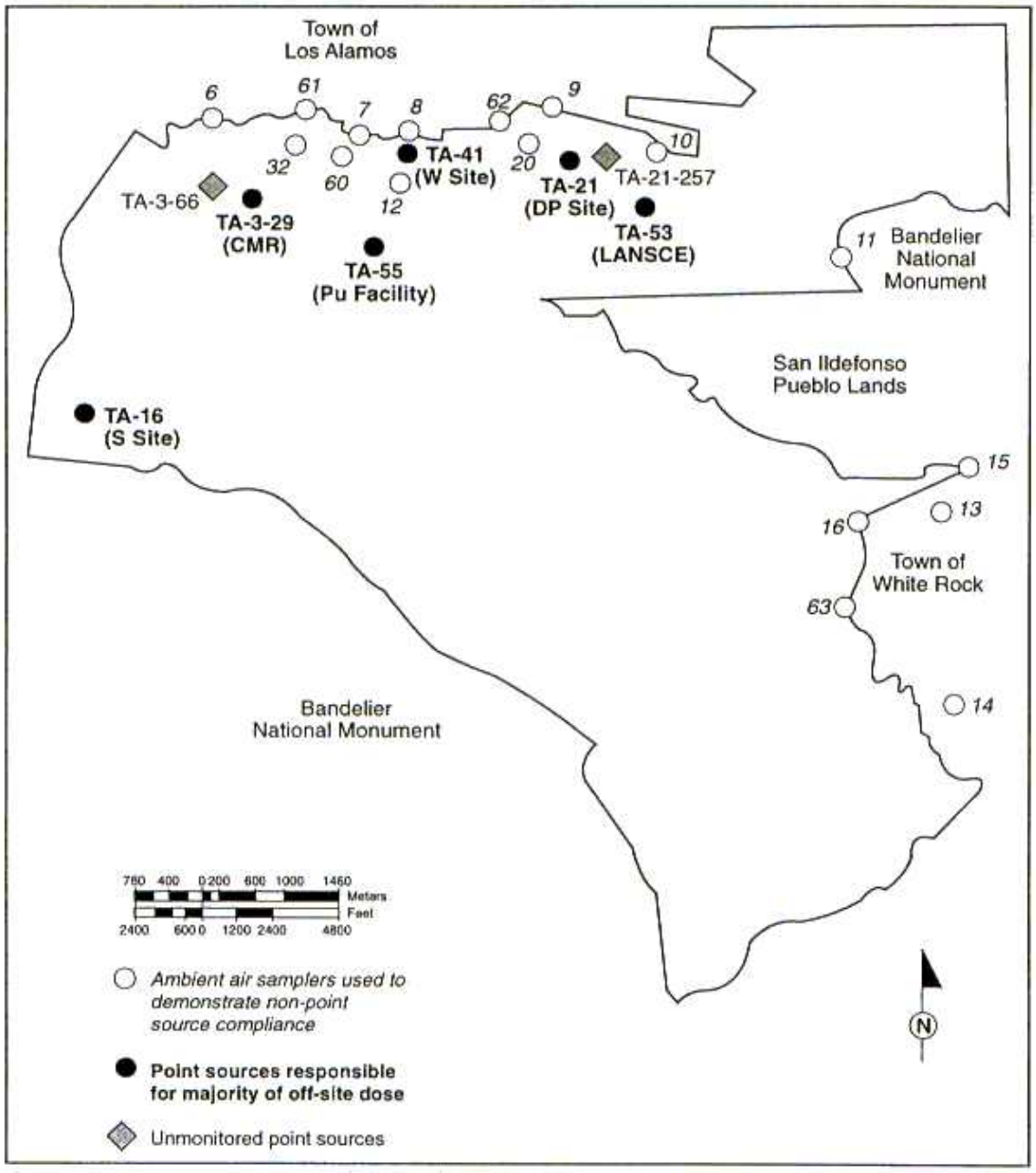

Figure 1. Los Alamos National Laboratory site.

The facilities identified in this figure are the major release points to air, which are discussed further in the chapter titled "Stack Sampling and Monitoring Evaluation." The Los Alamos Neutron Science Center, or LANSCE, is located at Technical Area (TA) 53 and has traditionally been the biggest contributor to offsite dose at LANL. This was again true for 2001.

For purposes of compliance, the offsite dose must be calculated at the location of maximum dose where a person could reside. The process of calculating dose is described in more detail in the chapter titled "Dose Assessment Evaluation." The location of maximum offsite dose for 2001

was near the sampler numbered 10 in Figure 1. This location, which can change from year to year, is commonly referred to as the maximally exposed individual, or MEI, for LANL. The calculated dose to the MEI in 2001 was 1.84 mrem (18.4 μSv).

The Independent Technical Audit Team

Because of the multidisciplinary nature of the audit, the audit team included scientists with a variety of backgrounds. These scientists have a broad range of skills including effluent monitoring, environmental surveillance, quality assurance, dose assessment and modeling, and source term development.

The audit team included scientists who are members of the *RAC* research team. *RAC* focuses on research related to risk associated with chemicals and radionuclides released to the environment. Key scientists on the audit team are listed below along with a description of the areas on which they focused during the audit.

- John E. Till, audit team leader
- H. Justin Mohler, radionuclide usage and associated emission calculations
- Paul G. Voillequé, stack monitoring and emissions
- Helen A. Grogan, environmental monitoring and diffuse source emissions
- Steven J. Maheras, dose calculation verification
- Arthur S. Rood, CAP-88 and CALPUFF comparison calculations
- Jill W. Aanenson, preparation of the final report.

For the most part, these scientists evaluated the same areas during each of the three audits, which streamlined the process considerably.

Purpose and Scope of Audit

The scope of work for the audits was established by the audit team. The scope of work stated that:

> This work will be performed independently by the audit team and the full cooperation of DOE, LANL, CCNS, and any other participants is expected. It should also be understood that this audit is the result of a legal settlement resulting from litigation brought about by CCNS, an environmental organization based in Santa Fe and representing the concerns of citizens residing near the Los Alamos National Laboratory. Therefore, to the greatest extent possible, the audit will be an open and fully documented process, providing both LANL and CCNS information that can be readily understood and traceable. Further, *RAC* recognizes the important role that will be played by the Institute for Energy and Environment Research (IEER), as a separate, independent group, responsible for monitoring the audit as it progresses. Therefore, throughout the course of the audit, we (will) provide whatever is necessary to allow IEER to fulfill its objectives.

Although this scope has remained essentially the same throughout the course of all three audits, the process has changed somewhat because of budget limitations. Principally, these

changes have been made to increase efficiency in the process and to concentrate on areas where we believed the most emphasis needed to be placed.

In each audit, the audit team assessed four technical areas of the compliance program:

- Radionuclide usage and associated emission estimates for unmonitored point sources
- Major release point effluent monitoring
- Environmental compliance sampling for non-point sources
- Dose calculation.

Quality assurance procedures were considered for each area, and the assessments are presented in their respective report chapters. The radionuclide usage, effluent monitoring, environmental sampling, and dose calculation chapters of this report present conclusions drawn by the audit team about LANL's compliance status.

One of the primary reasons to have independent scientists conduct the audit was to ensure that it addressed issues of scientific and technical merit as they applied to the compliance regulations. This audit was designed to not only verify compliance with regulations but also to assess whether the methodology chosen by LANL to demonstrate compliance was scientifically valid and defensible.

The scope of the audit was well defined and maintained, but the auditors also addressed several outstanding issues that were peripheral to the scope of the audit, that is, not related to LANL's compliance status, but of interest from a technical standpoint. These issues included the use of CAP-88 for calculation of doses for an environment with a complex terrain, use of thermoluminescent dosimeters (TLDs) for environmental measurements, uncertainty related to dose calculations, and future operations. These issues are discussed in the chapters titled "Complex Terrain Modeling Comparisons" and "Issues Peripheral to the Scope of the Audit."

Compliance as Defined by This Audit

Many aspects of compliance are discussed in the regulation. Most importantly, a facility must demonstrate a dose to the maximally exposed individual below 10 millirem per year (mrem yr^{-1}) (0.1 milliSievert per year or mSv yr^{-1}).[a] The regulation also contains many other requirements, such as measurement methods, procedures, quality assurance, and documentation that must be met for a facility to be in compliance.

As in the first two audit reports, we classified our findings and recommendations into three categories: (1) regulatory deficiencies, (2) technical or scientific deficiencies, and (3) specific observations. In previous reports, the third category of findings was titled additional observations, but we felt a revised title more accurately captured the intent of this finding category. Not all technical evaluation chapters identify findings for each category and some chapters may contain multiple findings.

A regulatory deficiency is a finding that tracks directly to a regulation or requirement that was not met by LANL for the year 2001. There were no regulatory deficiencies noted in this third audit.

[a] Throughout this report, we have redefined all units in their SI equivalent for universal readability, where applicable. In some cases, such as the discussion of a cited document, it was not appropriate to offer the conversion.

Technical or scientific deficiencies are items related to LANL's compliance program that are not specifically noted in the regulation but are critical to having a valid and defensible compliance program. Although not specifically outlined in 40 CFR 61, Subpart H, these technical issues directly affect LANL's ability to demonstrate compliance. There were no technical or scientific deficiencies noted in this third audit.

Specific observations point out practices for which the audit team recommends improvement or clarification. These issues are not noted or implied in the regulation, but they relate to good scientific practice, and we believe they need to be addressed by LANL. We highlight several specific observations and recommendations for improvement.

We have also included a section before the evaluation chapters to point out key areas where LANL has implemented significant changes in the program since the second audit.

Summary of 40 CFR 61, Subpart H and the Federal Facilities Compliance Agreement

A great deal of history accompanies the development of radionuclide standards for inclusion in the Clean Air Act, which was described in the first audit report (Aanenson et al. 1999).

The final radionuclide emission standards included in the Clean Air Act were published on December 15, 1989. The radionuclide emission standard for DOE facilities was established as 10 mrem yr^{-1} (0.1 mSv yr^{-1}); that is, emissions must be such that the resulting dose to any member of the public is less than this amount. The standard was codified in 40 CFR 61, Subpart H, *National Emission Standards for Emissions of Radionuclides Other Than Radon From Department of Energy Facilities*.

The regulatory guidance in 40 CFR 61, Subpart H, is designed to provide the rules that facilities must follow and guidance for the techniques that might be used to achieve compliance. These rules have some flexibility because the EPA can grant prior approval for alternative methodologies that the facility intends to use.

In 1991, the EPA conducted an audit of LANL's compliance status with 40 CFR 61, Subpart H. This EPA audit concluded that LANL was not in compliance with the 10 mrem yr^{-1} (0.1 mSv yr^{-1}) standard. In 1992, EPA issued a second notice of noncompliance to LANL and DOE. In 1994, DOE and EPA drafted a Memorandum of Understanding (MOU) that addressed the need for DOE facilities to reach an agreement with regional EPA offices regarding compliance. This MOU and the EPA audit findings brought about the development of the Federal Facilities Compliance Agreement (FFCA) between LANL and the EPA.

In 1996, LANL and DOE negotiated the FFCA, which was submitted to and approved by the EPA. This document provides more explicit guidance for the methodology to be used by LANL to implement their compliance programs as required by 40 CFR 61. In some cases, the FFCA constitutes prior approval for LANL to use alternate methodologies. In other cases, it simply identifies a more detailed discussion of the prescribed techniques identified in 40 CFR 61, Subpart H. The existence of the FFCA does not constitute compliance, but it provides LANL more detailed direction on methods to achieve compliance.

Part of the focus of the lawsuit that initiated the audit was a disagreement that CCNS had with the FFCA. Although a public comment period followed the release of a draft version of the FFCA, there was little opportunity for public input into the creation of the FFCA. In the eyes of

the public, the credibility of the FFCA was compromised by not including the public in the process of drafting the FFCA.

Nonetheless, the FFCA is an important part of the compliance process. Regulatory guidance is very limited for certain release scenarios that are important at LANL. LANL needed a framework for assessing the environmental impacts of releases for which compliance procedures either did not exist or were unclear. The FFCA provides this framework.

During the course of the first audit, the audit team reviewed the FFCA and found it, with one exception, to be technically sound. During all three audits, IEER challenged the technical validity of the FFCA with regard to its prescribed treatment of environmental transport and dose modeling of a complex terrain using a Gaussian plume model (CAP-88). Although the Clean Air Act regulation allows the use of CAP-88 for compliance modeling, the FFCA makes it a standard at LANL. We investigated the use of a complex terrain model and reported some initial results in the second audit. We continued to look at this issue during this audit and discuss our conclusions in the section titled "Complex Terrain Modeling Comparisons."

AUDIT PROCESS

For the LANL audit, we used a combination of methods to gather and analyze information. The audit team visited Los Alamos, New Mexico, on three occasions, touring facilities, talking with the LANL staff, and reviewing records. Members of the audit team performed independent calculations to verify dose estimates and to determine the validity of assumptions made. Although some work was carried out at the site, some analyses and reviews of documentation were performed at the individual offices of audit team members. The audit team believed this approach allowed a thorough evaluation of relevant aspects of LANL's compliance-related programs. Additionally, this evaluation was more detailed than a typical audit because the scientific and technical merit of each element of the program was closely examined.

In many cases, records to review were identified and sent to the audit team member requesting them. A list of all documents requested and received during the audit is included as Appendix B. Members of the public were encouraged to request documents related to compliance. To keep everyone informed of materials that were requested, LANL staff arranged for all interested individuals to receive copies of all requested material. Issues raised by IEER were distributed to *RAC*, and an attempt was always made to be certain that LANL personnel also received a copy. A list of all IEER, CCNS, and public issues raised during the third audit is included in Appendix C. These issues are discussed in specific chapters of this report or treated within the context of the appendix. The audit team worked to keep all parties informed of issues it was reviewing to the greatest extent possible. Since this audit was conducted under constraints of time and budget, all issues were not addressed to the same degree pursued during the first two audits. We believe, however, we have considered the most important and relevant issues and that the budget available and the schedule we established were sufficient to evaluate compliance.

During visits to the site, the audit team held meetings with LANL staff, CCNS, and IEER to discuss plans for the visit and to summarize findings at the end of each visit. These meetings were open to any individuals who wished to attend.

Layout and Purpose of Site Visits

The audit team planned and coordinated site visits through a joint effort with LANL, CCNS, and IEER. The audit team generally visited the site for three to four days at a time to gather information and tour facilities. Although different members of the team concentrated on different issues during the audit, site visits were planned to accommodate the interests of all concerned parties. Schedules and objectives for the visits were arranged and distributed to the appropriate people before the visits. Members of the public who participated in visits and tours were kept informed of progress through meetings at the beginning of each scheduled visit and summary meetings at the close of the day in a closeout briefing.

Site visits served as the primary information gathering mechanism during the audit. Relevant documents were identified, interviews were conducted, and facilities were toured. A number of processes and procedures were observed. The site visits allowed a thorough understanding of the methods by which LANL demonstrates compliance. More conventional audits are generally conducted with a single site visit and observational time. The audit team felt that scheduling multiple site visits with a chance to work, digest information, and read documents between the

visits enhanced the value of the time spent at LANL. This type of schedule streamlined the entire process and contributed to a more productive team effort and a more thorough audit.

LANL, IEER, and CCNS personnel accompanied the audit team members on facility visits. Additionally, invitations were extended to the public and to state agencies to attend these tours, and several individuals used this opportunity.

The audit team recognizes that considerable preparation was required on the part of LANL to meet security requirements and allow the audit to be conducted with unprecedented openness. The audit team appreciates this effort, and we believe that the ability to see facilities and explain our observations to members of the public was critical to credibility. We regret that for some tours, members of the audit team and the public did not participate as originally scheduled. This was an inevitable consequence of public involvement but also the result of an increasing confidence in the compliance program on the part of individuals as the audit process matured. Nevertheless, in every case a facility tour was scheduled, at least one member of the audit team was present and important information was gathered. At no point during the audit were interested parties denied access to facilities that we thought it necessary to visit. Table 1 lists the facilities visited.

Table 1. Facilities Visited During 2002 Audit Tours

Facility	Tour date	Purpose of tour
TA 43-1, Health Research Laboratory	June 4, 2002	Radionuclide survey
TA 3-1698, Materials Science Laboratory	June 4, 2002	Radionuclide survey
TA 53, LANSCE	June 6, 2002	Stack monitoring
AIRNET stations, White Rock loop	July 10, 2002	Non-point source monitoring
TA 54	July 10, 2002	Non-point source monitoring
TA 3-29, Chemical and Metallurgical Research Facility	August 21, 2002	Stack monitoring
TA 3-35, Press Building	August 22, 2002	Radionuclide survey
TA 35-213, Target Fabrication Facility	August 22, 2002	Radionuclide survey

Interviews

Throughout the audit, interviews were conducted with personnel from the Risk Reduction and Environmental Stewardship-Meteorology and Air Quality (RRES-MAQ) division[b] of LANL, facility managers, and other people responsible for compliance activities at LANL. Interviews were generally planned ahead of time and usually involved one or more members of the audit team, IEER, CCNS, LANL, and other interested parties. In keeping with the policy of openness, anyone could be a part of any interview. However, if either IEER or CCNS wanted to be involved

[b] Risk Reduction and Environmental Stewardship-Meteorology and Air Quality (RRES-MAQ) was formerly known as Environmental Safety and Health (ESH)-17.

in an interview and was unable to attend, an interview was rescheduled until all interested parties directly involved in the audit could attend. It is a credit to the groups involved that this procedure worked well.

Members of the audit team led interviews, and the interviews focused on procedures relevant to compliance issues. Interviews were quite specific because the first two audits had provided us with extensive background knowledge about LANL and its procedures, which enabled focused investigation of well-defined issues during this third audit.

Document Retrieval

It was necessary to obtain a large number of documents from LANL to support our research of compliance activities. Periodically, documents were requested in writing by the audit team, CCNS, or IEER. A copy of every document request was sent to all parties involved in the audit process so that all could be aware of ongoing research and information needs. Appendix B lists all documents requested and received during the course of the audit.

SIGNIFICANT CHANGES AND IMPROVEMENTS SINCE THE SECOND AUDIT

Because the RRES-MAQ Clean Air Act compliance program continues to improve as a result of these audits, we felt it was important to highlight a few key changes here. Details related to these and additional changes are discussed in the technical sections that follow. Many of the changes discussed here were in response to recommendations made or issues raised in the second audit.

LANL underwent a significant reorganization since the last audit. As a result of that reorganization, the name of the division that is responsible for maintaining the Rad-NESHAP compliance program changed from the Environment, Safety & Health division, Air Quality Group (ESH-17) to the Risk Reduction & Environmental Stewardship division, Meteorology & Air Quality group (RRES-MAQ). We believe this reorganization is beneficial to LANL. We would recommend, however, that RRES-MAQ update all procedure documents to reflect this change.

Substantial changes have been made since the second audit with regard to the unmonitored point source and usage survey aspects of the compliance program. Based on recommendations from the second audit, the database in which information about radionuclide usage is collected has been redesigned to further automate emissions and dose calculations. RRES-MAQ no longer collects information on inventory but has shifted the focus, more appropriately, to actual usage of radioactive materials in a facility. An important change in the dose calculation process involves identifying important progeny according to guidance provided by NCRP (1996).

Based on an issue raised by the IEER monitor during the second audit, improved stack sampling systems have been put in place at the Chemical and Metallurgical Research (CMR) facility. Additionally, a database has been developed to collect important stack information to automate the calculation of emissions.

Because of continued concerns about the adequacy of the AIRNET system to evaluate releases from all facilities, RRES-MAQ reviewed the locations of the AIRNET monitors and decided to place an additional monitor at the Los Alamos Airport to more effectively capture releases from TA 21. An issue raised by the IEER monitor with regard to silica gel collection efficiency led RRES-MAQ to further pursue this topic. LANL discovered that the recovery of sample from the silica gel was not as complete as originally estimated, and they applied a correction factor to all of the AIRNET tritium data to adjust for the limited recovery.

In all, we believe the changes made to the compliance program since the last audit have improved it. This is a credit to all involved in the audit process.

EVALUATION OF UNMONITORED POINT SOURCES AND EMISSION ESTIMATES BASED ON USAGE SURVEY DATA

As specified in 40 CFR 61, Subpart H, emissions must be estimated for all facilities with the potential to discharge radionuclides into the air. This includes all facilities conducting operations using radionuclides in an environment that discharge effluents through a forced ventilation system via a single exhaust stack or point source. This emission estimate is used to determine monitoring requirements for all point sources at each facility. Any point source with the potential to emit radionuclides in quantities that could cause a member of the public, or maximally exposed individual (MEI), to receive a potential effective dose equivalent (PEDE)[c] in excess of 1% of the standard (0.1 mrem yr^{-1} or 1 μSv yr^{-1}) is defined as a major point source and requires continuous monitoring. Point sources with a radionuclide emission potential and consequent dose of less than 1% of the standard are defined as minor point sources and may be evaluated for compliance with the standard by estimating the potential for emission during the year in question by one or a combination of the methods outlined in the FFCA. The requirements outlined in § 61.93 (b) for point sources that require monitoring state the following:

> Radionuclide emission rates from point sources (stacks or vents) shall be measured in accordance with [the following requirements] or other procedures for which EPA has granted prior approval.

The procedures outlined in 40 CFR 61, Subpart H, however, do not explicitly define the method to be used for estimating potential emissions from point sources that do not require continuous monitoring. In May 1996, the EPA and the DOE established the FFCA to provide further guidance to LANL with regard to making these point source potential emission determinations. The FFCA effectively serves as prior EPA approval regarding methods of estimating potential emissions and consequent doses from unmonitored point sources for comparison to the 10 mrem yr^{-1} (0.1 mSv yr^{-1}) standard specified in 40 CFR 61, Subpart H. It also establishes several options for determining monitoring requirements as well as verifying continued low emissions.

The methods for estimating emissions outlined in the FFCA include the use of historical stack sampling data, 40 CFR 61 Appendix D methodology, duct holdup studies, engineering estimates and judgments, and the need for operational flexibility. The text of Appendix D also refers to another procedure that may be used for demonstrating compliance and determining reporting and application to construct requirements (EPA 1989). The guidance provided by EPA (1989) that is applicable to unmonitored facilities involves an assessment based on annual possession quantities. Where facility operations are relatively stable, historical stack sampling data are considered by the FFCA to be the most accurate method for determining potential emissions. Appendix D methodology was originally designed to estimate emissions for new construction or modifications and changes to existing sources, but LANL also relies on this methodology for estimating potential emissions and determining monitoring requirements based

[c] For readability, the precise technical term potential effective dose equivalent is usually replaced with the general term dose in this document. However, the reader should be reminded that this is a possible dose and not an actual measured value that a member of the public received.

on respective facility radionuclide usage information. For determining monitoring requirements, estimated radionuclide release rates are based on the discharge of the effluent stream that would result if no pollution control equipment existed, but the facility operations were otherwise normal. For demonstrating compliance with the standard, the FFCA states that the filtration factors for pollution control equipment specified in Appendix D of 40 CFR 61 may be applied. Additionally, periodic confirmatory measurements are required to verify continued low emissions from unmonitored facilities. The FFCA (Section 2.1.3) specifies that these confirmatory measurements may be based on "…engineering estimates using current inventory measurements or determinations and the other methods described in Section 2.1.1."

It is important to make a clear distinction between radionuclide inventory and radionuclide usage. A number of concerns were raised during the first two audits regarding the use of the term "inventory" and whether the reported values for unmonitored point sources represented a snapshot of the inventory at a single point in time, the total inventory throughout the year, or usage during the year. The wording of 40 CFR 61, Subpart H and Appendix D, clearly states that radionuclide usage at facilities for the period under consideration (i.e., during the year for which compliance is determined) is necessary for reporting requirements as well as for use in estimating radionuclide emissions. The FFCA makes numerous references to 40 CFR 61, Subpart H and Appendix D, but its use of the term "inventory" is not clearly defined. It is imperative that radionuclide *use* throughout the year be the starting point for the purpose of estimating potential emissions, and we recommend that any revision or update to the FFCA incorporate a clear definition of the word "inventory" for the purpose of estimating emissions that is mutually agreed upon by EPA, LANL, and the public.

A defensible and credible record of radionuclide usage is clearly an integral part of demonstrating compliance as specified in 40 CFR 61, Subpart H, when Appendix D guidance is used to estimate emissions. It not only serves as the basis for determining monitoring requirements and verifying continued low emissions, but it is also used to directly estimate releases and consequent doses for unmonitored (minor) point sources. In addition, the usage survey data are used to determine those radionuclides contributing 10% or more of the total dose at monitored point sources and, therefore, evaluate the appropriateness of current monitoring equipment.

The following sections describe and evaluate the methodology LANL (RRES-MAQ) employed to compile 2001 usage information, estimate potential emissions, and calculate consequent doses from unmonitored point sources. These sections also discuss any changes LANL made to address concerns raised during the previous audit and as part of LANL's periodic procedure revision system. The audit findings and recommendations are presented at the end of each section.

Because of heightened security at the onset of the current audit, it was necessary to select facilities to visit before the radionuclide usage survey for 2001 was available. Therefore, a number of facilities were randomly selected, based on the 2000 unmonitored point source dose evaluations and a preliminary list of 2001 sources. We primarily selected facilities that had not been the focus of either of the first two audits.

Radioactive Material Usage Data Collection

Summary of LANL Methodology

Facility radionuclide usage data serve as the primary basis for evaluating annual potential emissions and doses from unmonitored point sources and for determining monitoring requirements for all point sources. The data collection, and hence our evaluation, focused on potential emission sources and associated data for 2001. Radionuclide usage data were gathered for all point sources with the potential to actively exhaust radionuclides through a forced ventilation system via a single stationary point.

To conduct work in a manner that helps focus appropriate emphasis on sources that have significant potential or actual emissions to the environment, RRES-MAQ (hereafter referred to in this section as MAQ) adopted (beginning with the 1999 usage survey) a graded approach to categorizing unmonitored point sources and updating radionuclide usage survey information based on the calculated potential doses for the previous year. The regularity with which usage information was collected and updated varied depending on the tier classification of the source, which is based on guidance provided by ESH-17-RN, R2. The following tier levels have been defined:

- Tier I – Any source with actual emissions that contribute greater than 1 mrem yr^{-1} (0.01 mSv yr^{-1}) to any member of the public (as defined by Subpart H) according to the previous rolling twelve month period.
- Tier II – Any source with the potential to contribute greater than 0.1 mrem yr^{-1} (1 μSv yr^{-1}) to any member of the public (as defined by Subpart H) according to the last usage survey.
- Tier III – Any source that does not have the potential to contribute greater than 0.1 mrem yr^{-1} (1 μSv yr^{-1}) but that does have the potential to contribute greater than 0.001 mrem yr^{-1} (0.01 μSv yr^{-1}) according to the last usage survey.
- Tier IV – Any source that does not have the potential to contribute greater than 0.001 mrem yr^{-1} (0.01 μSv yr^{-1}) to any member of the public according to the last usage survey.

Per 40 CFR 61 Subpart H requirements, all Tier I and II sources require continuous monitoring to determine emissions and consequent doses. Tier I and II sources are updated as part of the usage survey every 2 years, and need only meet the record keeping requirements for Tier IV sources. Tier III sources are updated and evaluated annually to confirm and verify that emissions and associated potential doses remain below Tier II classification requirements, and the information presented in the annual usage survey will be traceable to a secondary source of documentation (e.g., monitoring data, logbook, and database maintained at the facility level). Tier IV sources are evaluated at least every 2 years to confirm and verify that emissions and associated potential doses remain below Tier III classification requirements, and usage survey information does not need to be traceable to a secondary source of documentation. In summary, a partial usage survey (including only Tier III sources) will be conducted for calendar year 2002, a comprehensive usage survey (including Tier I, II, III, and IV sources) will be completed for calendar year 2003, a partial usage survey will be done for calendar year 2004, and so on.

The MAQ staff has taken additional steps to allocate resources to release points with the potential to impact the cumulative offsite dose and/or monitoring requirements and to more complex and dynamic operations throughout LANL. Certain point sources meeting established criteria will be removed from future usage survey reports (ESH-17:00-160). While full usage survey updating efforts will not be conducted for these point sources, they will remain as part of the usage survey update record and will be evaluated to ensure that these sources continue to meet the required criteria. The criteria required for removal from the full usage survey update are that the release point has a potential effective dose equivalent less than 0.0001 mrem yr^{-1} (0.001 μSv yr^{-1}) *and* the facility active operations have and are expected to remain constant, or are decreasing, or the facility is inactive.

Also in the interest of conserving time and resources, certain point sources meeting established criteria will not require onsite interviews or walkthroughs of facilities or laboratories for future usage survey updating purposes (ESH-17:00-071). A phone or e-mail interview will be considered adequate for these facilities, which include any facility that does *not* meet one or more of the following criteria:

- A new facility contact or operations that has not had an initial site visit interview by an MAQ representative *and* the calculated dose is a significant contributor to the stack's radionuclide emissions
- The operations and/or radioactive materials are continuously changing
- The operation and/or material have undergone a significant change
- A new release point of radionuclide emissions has been identified.

Point sources are identified through various mechanisms, including historical classification, the Air Quality Review process (discussed later), facility contacts, and field investigations. Collection of associated radioactive material usage data is intended to represent operations at each release point of interest during a specific year, as well as potential duct hold-up and residual contamination, that may impact or otherwise contribute to airborne emissions of radioactive materials. The usage information, in conjunction with information specific to each process related to radioactive material usage, forms the basis for estimating potential emissions and dose from both monitored and unmonitored release points at LANL (ESH-17-102, R3). Because of concerns remaining following the second audit regarding clear distinction between the definition and use of the term inventory, MAQ eliminated collection of "inventory" information and now attempts to collect information related only to radionuclide "usage" throughout the year (ESH-17:01-069).

To initiate the process of updating radionuclide usage information, MAQ personnel contacted the facility manager and/or the designated point of contact for the facility regarding the upcoming usage survey update. To facilitate data collection, the latest usage survey information was provided to the point of contact. If it was deemed necessary, a meeting was scheduled with the facility point of contact to explain the survey process. In addition to explaining the purpose of the survey during the meeting, a usage survey was completed and documented as described by ESH-17-126, R5. Following collection and compilation of updated usage information, discrepancies and/or incomplete data were resolved via phone and e-mail interviews or site visits. The compilation of all 2001 usage survey information was documented as Terp et al. (2002). Additional details related to each process contributing to potential emissions at each point source were collated in separate file folders and ESH-ID Air Quality Review documentation maintained by MAQ.

In addition to the primary radionuclide usage data collected as part of the survey process, MAQ staff reviewed ESH-ID Air Quality Reviews on a monthly basis to supplement survey data and incorporate relevant information into the current usage survey update. This additional review step was added to help ensure that new and modified operations are incorporated into the current survey. MAQ relies on identifying and capturing new or modified radionuclide air emission sources at LANL through the ESH-ID process or through notification directly to MAQ by facility project managers familiar with regulatory requirements.

Air Quality Reviews can be initiated through the ESH-ID process or by contacting MAQ directly. A facility must first determine that the proposed activity qualifies as a new source, new construction, or change in process and requires an Air Quality Review as documented in Laboratory Implementation Requirement (LIR) 404-10-01.1, which specifies that facility managers and supervisors identify and mitigate hazards associated with new activities and projects. Appendix 2 of this LIR requires contact of MAQ for an air quality requirements determination for any activity involving the use of radionuclides that includes a change in process or radionuclides, an increase in quantity, or relocation to a different exhaust system. New Source Review (NSR) project personnel assess new or modified radioactive air emission sources according to the procedures outlined in ESH-17-103.

In addition to the information described above, MAQ considered and incorporated into the usage survey report other elements that could influence emissions including actual and potential duct holdup data and room or area contamination data. The steps taken to estimate emissions from these sources are not repeated annually in all cases, and facility operational stability is considered in this regard. MAQ makes this determination based on information from facility representatives, best health physics practices, and best professional and engineering judgment.

Duct holdup was based on historic monitoring data, information derived from facility personnel, or actual duct holdup estimates, when available. When historic monitoring data were used to estimate duct holdup, data from the last 2 to 4 years of available monitored emissions are typically used, depending on the stability of operations. Monitored emissions were multiplied by the appropriate filtration factor (e.g., 2000 for single stage high-efficiency particulate air [HEPA] filtration, as specified in the FFCA) to estimate potential emissions. No physical state reduction factor was applied because this potential emissions estimate was based on actual monitoring data. For 2001, historic monitoring data, when available, were used to estimate potential duct holdup for a number of unmonitored point sources.

Residual contamination was also considered and was derived from sources including previous inventory/usage surveys, interviews with facility representatives, and radiological survey data. Best engineering judgment calculations attempt to estimate potential emissions resulting from residual contamination and take into account operational stability.

Although there are no explicit quality assurance (QA) requirements spelled out in either 40 CFR 61, Subpart H or the FFCA with regard to radioactive material usage information, assurance of data quality is important. Lacking specific regulatory guidance for assuring the quality of usage data, MAQ has implemented a project verification and peer review process to help assure the quality of reported data and to ensure that specific project requirements are met (ESH-17-RN, R2). This Quality Assurance Project Plan requires that MAQ personnel evaluate all point sources with the potential to emit radionuclides and ensure that all point sources are categorized and identified properly.

Point source identification requirements were evaluated by visits to selected LANL areas not included in the usage survey to verify that omission was appropriate, peer review of TA and building lists to ensure that the status was accurate, and peer review to verify that point source classification was valid. Completion of the requirements outlined for this process was documented in RRES-MAQ:02-159.

For the 2001 usage survey, MAQ staff evaluated 26 facilities not currently classified as point sources through field investigations to verify the absence of radioactive point sources (RRES-MAQ:02-155). In addition, to verify their classification as point sources, 7 exhaust systems at 7 facilities were inspected through field investigations (RRES-MAQ:02-153). Of the 7 exhaust systems evaluated, 5 met the definition of a point source, and 2 did not meet the definition of a point source. Point source radioactive materials usage survey requirements are also assessed by visiting selected facilities and spot-checking reported usage and process information. To verify 2001 usage survey information, 8 facilities were spot-checked to ensure that facility personnel agreed with the information compiled by MAQ (RRES-MAQ:02-153). The process description for one facility (TA 50-69) was found to be in error and was corrected.

Validation steps taken to verify data entry and electronic transfer are described in ESH-17-RN. The usage data reported by each facility and collected by MAQ were compiled using a relational Access® database developed for this purpose. Historical data from previous years are maintained as archived tables in the database. Data were verified and validated by project personnel by 100% verification of hand-entered data and 10% verification of electronically transferred data and professional evaluation (peer review) of all data for usability. Completion of this verification and validation process was documented in the file folders maintained by MAQ for each facility.

During the summer of 2001, the database application used to compile usage survey information was redesigned to further streamline data entry, potential emission and dose estimates, and quality assurance (QA) of data entry and associated calculations. A beta test and comparison calculations using the archived 2000 usage survey data and associated emission and dose estimates were instituted to demonstrate that the application was accurately calculating potential emissions and dose for each release point (ESH-17:01-451). Based on the results of the beta test, necessary modifications were made, and this new database application was used to compile all 2001 usage survey data and generate associated emission and dose calculations.

Evaluation of LANL Methodology

We assessed the methodology used by MAQ with regard to radioactive material usage data collection through visits and tours to four separate facilities (TA 43-1, 3-1698, 3-35, and 35-213). We also conducted detailed reviews of the supporting documentation maintained for the toured facilities and also those facilities with 2001 PEDEs greater than 0.01 mrem (0.1 μSv) (TA 3-66[ES 04], 3-102[ES 25], 21-150, 21-257, 48-1[ES 67], and 54-36).

LANL continues to make a concerted effort to evaluate all unmonitored exhaust systems with the potential to emit radionuclides. Specific steps were carried out to ensure that all facilities requiring an evaluation were properly assessed. MAQ has also implemented procedures that attempt to capture and review all new or modified sources that may impact monitoring requirements. Duct holdup and residual contamination were considered as potential emission contributors to a number of exhaust stacks. It is again noted that using historic monitoring data to

estimate duct holdup is a conservative approach because such data inherently include all operational releases in addition to any potential releases related to duct holdup.

Improvements have been made since the second audit with regard to the collection and presentation of radioactive material usage information. MAQ personnel no longer attempt to collect and distinguish between radionuclide inventory and usage information, and instead focus on compiling relevant usage information only. Because inventory information is not used to make emission estimates, this should alleviate much of the confusion related to differences between inventory and actual usage. A "Call Back Date" field was added to the database to document that facility personnel have provided retrospective radionuclide usage information for the preceding calendar year of interest, as opposed to estimates of future use. The usage survey database application has been equipped with a feature to enable automatic identification of 10% contributors for any point source to ensure appropriate monitoring capabilities are in place at each facility.

All processes with the potential to emit radioactive material are categorized separately for each facility, laboratory, or room vented by a given exhaust system. Source type (present or future), usage amounts and basis, and physical state information were collected for all radionuclides used in each separate process. Further, a description for each process was provided, as well as an assessment of any heating that was done and its potential impact on the chemical and physical state of each radionuclide involved in the process. This information was provided in the annual usage survey compilation. This represents a substantial improvement since the first audit in the amount of information provided to the public and, in most cases, helps provide a reasonably clear picture of each process potentially impacting radionuclide emissions. In combination with the file folders maintained by MAQ, which include documentation of e-mail communications, it was possible to understand how the data were collected, updated, revised (if necessary), and assembled into usage estimates.

A continued effort has been made to assure the quality of data reported by facilities and compiled by MAQ. The Access database developed to compile and document all radionuclide usage information has been revised and its utility expanded (this is discussed in more detail in the section "Potential Emissions and Dose Calculations"). We examined the structure of this database, and it is well designed and has resulted in a more efficient method for compiling and utilizing radionuclide usage data that will free up resources for use elsewhere. This move toward a more automated compilation process is wise, and it helps ensure data quality and reduce the potential for calculation errors. Our suggestion from the second audit to implement a LANL-wide database system for compiling radionuclide usage at the facility level was investigated by MAQ. The response from facility personnel indicated a desire for MAQ personnel to continue to maintain responsibility for data collection and data entry; therefore, implementing such a system was not pursued. MAQ is not solely responsible for demonstrating compliance with the Clean Air Act; rather it is the responsibility of the entire Laboratory. We continue to believe that implementing a Laboratory–wide system for compiling radionuclide usage at the facility level (by the users of radionuclides and at the time of or shortly following actual use) represents the most efficient, accurate, and defensible means by which to track this type of information.

Procedures have been implemented by MAQ to verify the accuracy of data provided by facilities, as well as to verify the accuracy of calculations made by MAQ and appropriateness of methodologies used to categorize point sources. Completion of these procedures is documented in

the file folders maintained by MAQ for each facility, and it is clear that these efforts continue to result in a more robust and defensible radionuclide material usage data compilation.

Findings and Recommendations

In many respects, the usage survey data and associated emission estimates represent one of the most complex and dynamic aspects of the Clean Air Act compliance program at LANL; therefore, these data are prone to a greater potential for error. The focus that MAQ is placing on increased automation of data compilation and associated emission and dose estimates is an important step in developing an efficient and credible method for determining radionuclide usage and associated emissions and potential doses to members of the public. The redesigned database application reduces the potential for error. There is little question that the new database will enable MAQ personnel to focus resources previously allocated to checking calculations, which are now largely automated, to other tasks that cannot be automated.

Additional requirements continue to be established and modified as the compliance program evolves in an attempt to focus resources most effectively and to address specific issues as they arise. This move toward an efficient and flexible program that continues to satisfy regulatory requirements is commendable, and we strongly encourage MAQ to maintain these efforts as part of future changes to compliance related processes. The MAQ program has actively followed through and addressed the recommendations and findings from the first and second audit reports, and the changes that have been implemented have resulted in a more defensible and understandable compliance program.

We identified a number of areas where modifications or refinements would result in continued improvement of the air quality program at LANL. The following discussion provides recommendations based on the findings of this audit that we believe will further strengthen the compliance program in several areas related to radioactive material usage data collection.

Specific Observations Related to Quality Assurance of Radionuclide Usage Data. Usage survey data form the basis for estimating potential emissions and dose for all unmonitored facilities, identifying 10% contributors to dose and evaluating monitoring status for all monitored facilities, and determining monitoring requirements for all facilities (including periodic confirmatory measurements to verify low emissions). Because of MAQ's reliance on usage data for a number of important purposes, establishing and maintaining effective methods to assure the accuracy of the data should be a fundamental goal of the compliance program. Lacking guidance from either 40 CFR 61 or the FFCA with regard to specific quality assurance requirements related to the collection of radionuclide usage information, MAQ has implemented a number of effective internal procedures to assure the quality of usage survey data and associated emission estimates.

In addition to the procedures designed to assure the quality of internally compiled data and calculations, MAQ personnel also conduct spot checks of the radionuclide usage information and process descriptions provided to them by facility contacts. During the course of facility visits, interviews, and document review during this audit, a number of instances were noted where usage information was changed as a result of MAQ inquiries to facilities, requests for documentation, and in one case because a facility contact had a hunch he may have made a mistake. However, none of these mistakes or changes to usage amounts resulted from the facility spot checks instituted by MAQ. To the credit of MAQ, all of these changes *were* captured in the 2001 usage

survey report, largely as a result of the internal procedures established and carried out by MAQ, and in one case simply because the facility contact realized a mistake was made. However, these issues, which are described in more detail by two IEER memos (see Appendix C), point to insufficient procedures designed to assure the quality of the data that are initially provided to MAQ by each facility.

Potential errors in data provided by facility contacts would, in nearly all cases, be identified only through additional follow-up by MAQ personnel. It is not reasonable to expect or assume that this type of follow-up by MAQ occur for all facilities, and in cases where such follow-up is not necessary or otherwise not conducted, it is difficult to know how prevalent errors in facility-level data may be. Because the data provided to MAQ by each facility form the basis for all subsequent calculations to estimate emissions and dose, and because LANL relies on emission and dose calculations based on usage data as a very integral part of their compliance program, establishing an effective mechanism to assure the quality of facility-level data when they are initially provided to MAQ is of high importance.

There are no specific requirements regarding performance (i.e., methods for achieving a specific degree of accuracy, precision, or completeness, such as are prescribed for effluent and environmental measurements) of quality assurance procedures related to radionuclide usage data in 40 CFR 61, Subpart H; the FFCA; or EPA (1989). Lacking specific regulatory guidance, MAQ has developed and implemented procedures designed to help assure the quality of usage data provided by facility contacts. As a result, this issue does not impact LANL's compliance with the Clean Air Act.

There are regulatory requirements specified by 40 CFR 61, part 61.95 for record keeping which state that it must be "…sufficient to allow an independent auditor to verify the accuracy of the determination made concerning the facility's compliance with the standard." Similar requirements are noted by EPA (1989) with regard to maintaining sufficient documentation "…for the EPA to judge the validity of the input used in the calculations." While we did not believe this record-keeping requirement was met during the year evaluated by the first audit (1996), we considered the program evaluated during this third audit (2001) satisfactory with regard to this regulatory requirement and believed the documentation maintained by MAQ was sufficient to allow us to assess the accuracy and validity of the emission calculations and determine compliance with the standard. The same conclusion of compliance with the record-keeping requirements was also made during the second audit (1999).

Our evaluation and assessment of the MAQ quality assurance program as it relates to usage data for this third audit has been consistent with the approach we have taken for the first two audits. In general, as with the first two audits, we believe that the procedures MAQ has adopted for assuring the quality of these data meet the underlying purpose of quality assurance in that they help minimize the occurrence of significant errors. As noted previously, the mistakes or changes in usage amounts identified during the course of this third audit did not result in MAQ using erroneous values and the correct values appear to have been used for estimating emissions and doses in 2001. However, the occurrence of these issues suggests that some additional procedures are needed to further assure the quality of usage data as used by MAQ.

The currently implemented procedures are effective for assuring the quality of data generated by MAQ, but we consider them insufficient with regard to assuring the quality of data generated at the facility level. We strongly recommend that MAQ, in conjunction with appropriate facility contacts, establish and institute a program to more effectively assure the

quality of usage data compiled at the facility level. We also encourage MAQ to seek additional EPA input in this regard because of the lack of guidance currently provided by 40 CFR 61 and the FFCA. In addition, we suggest that LANL work with CCNS and other members of the public to adopt specific procedures and requirements that will satisfy the public's wishes for establishing the quality of usage data.

It is simply not possible to establish a QA program that precludes the occurrence of all errors. In attempting to refine the current QA procedures, all stakeholders should recognize this fact and be prepared to balance the level of resources directed at and requirements specified for facilities with the magnitude of emissions and associated doses at each facility. It is also important to consider the sum of all compelling evidence with regard to potential public health impact from each facility, which includes AIRNET, both historic and current effluent data, and what appears to be a conservative method, in most cases, for estimating unmonitored releases (i.e., Appendix D).

Specific Recommendations for Improving the Quality Assurance of Radionuclide Usage Data. The spot checks currently carried out by MAQ staff do not conform to the general expectation that a quality assurance program involve review by multiple different individuals with knowledge of facility operations because the currently implemented spot checks simply revisit the same facility contact. Discussions with MAQ personnel during the course of this audit suggested a willingness to improve the effectiveness of their current compliance program in this regard. To this end, the currently established procedure for conducting facility spot checks should be modified and focused on facilities with the greatest potential for emissions to ensure that it is effective in assuring the quality of data provided by each facility. It may be necessary to adopt different approaches for different facilities, depending on the nature of operations at each facility. In some cases, a quality assurance plan may be easily implemented at the facility level, and in other instances it may be more effective to continue the policy of spot checks by MAQ personnel. Regardless, it is not reasonable to expect that MAQ personnel be responsible for assuring the quality of all data provided by radionuclide users, and individual facilities should be expected to shoulder a significant portion of this aspect of compliance.

Spot checks by MAQ could also be focused and directed in some cases by developing a flag that is triggered when a point source PEDE changes (increases or decreases) by some pre-determined "significant" fractional amount (e.g., a factor of five or more). It is important that this flag be triggered by either a decrease or an increase. Per current procedural documentation, MAQ personnel would typically follow up only on a facility whose PEDE increased such that its Tier level designation changed to a higher level (e.g., moved from Tier IV to Tier III). A potential error that resulted in an erroneously lower PEDE, however, could conceivably escape notice.

Because MAQ elects to initially calculate potential doses using assumptions presumed to be conservative, it is important that MAQ staff clearly emphasize to radionuclide users that usage estimates for Tier IV sources, which do not require documentation at the facility level, should be truly conservative. For example, a point of contact at TA 48-1, Room 430, ES-67 initially reported usage of 100 mg ^{242}Pu when asked for a conservative upper bound estimate. Then, when secondary documentation was requested by MAQ because the source changed from Tier IV to Tier III, the point of contact provided documentation indicating usage of 122.8 mg.

In addition to ensuring that initial user estimates, when not supported by facility records, be consistently upper bound estimates, it is important that MAQ staff not become overly dependent

on the documentation requirements established by ESH-17-RN, R2 for Tier IV sources such that facility records are not referenced simply because it is not required, particularly in cases when they are apparently readily available. For ^{239}Pu usage at TA 48-1, Room 430, ES-67, the point of contact initially estimated 450 mg, and then provided documentation of 380 mg actual usage. Similarly, the initial usage estimate provided by the facility contact at TA 35-213, Room F-11 was 400 mCi (1.5×10^{10} Bq) of tritium. Subsequently, this value was changed to 40 mCi (1.5×10^{9} Bq) after the facility contact referred to the original shipping manifest. Since the documentation in these cases apparently existed in the first place, it is not clear why the point of contact did not refer to it to provide the initial usage estimate. Establishing effective facility-level QA procedures would help ensure a consistent method for deriving and reporting usage estimates or documented values.

MAQ review of any facility-made calculations to derive usage estimates should also be part of all Tier III point source evaluations where secondary documentation is required. This was not done during 2001 in at least one case (TA 21-257). Discussions with MAQ personnel have indicated this was an oversight, and review of facility calculations is something that is usually done, and MAQ indicated that these reviews will be done in the future. We recommend that the process verification and peer review requirements established by the Quality Assurance Project Plan (ESH-17-RN, R2) be amended to make this review step mandatory.

In addition, it would be useful to establish a measure of the validity of the inventories for drums that are characterized or otherwise "used" at various facilities. Questions were raised during this audit regarding the accuracy of the drum inventories used by MAQ for potential emissions calculations (see Appendix C). Based on discussions with MAQ staff regarding drums characterized at TA 54-36 (which are noted to contain any type of waste that is generated by LANL), verbal reports from facility personnel indicate that post-characterization data for "newer" drums are very close to pre-characterization data, while post and pre-characterization data for "older" drums have a larger variability. MAQ staff noted that these "older" drums are, therefore, sampled (i.e., characterized) more frequently. The best available (i.e., most recent) information regarding drum inventories is then reported to MAQ for the purpose of making emission estimates. There is no official report generated to provide a quantitative measure of differences that exist between the pre- and post-characterization data. A quantitative measure of the accuracy of inventories assumed for "older" drums would help establish the validity of using pre-characterization data for the purpose of estimating potential emissions. If significant discrepancies are apparent, a more appropriate methodology may be to develop separate emission estimates (e.g., modified by some uncertainty factor) for "older" drums whose inventories are not based on post-characterization data and, therefore, not known to the same degree of accuracy.

Specific Observation Related to New or Modified Source Identification. Although MAQ staff review ESH-ID Air Quality Reviews on a monthly basis to supplement survey data and incorporate relevant information into the current usage survey update, it is still not clear that all new or modified operations are reviewed by MAQ personnel per LIR404-10-01.1 requirements. It was also noted during discussions with MAQ personnel that the ESH-ID process is not mandatory. The fact that the ESH-ID Air Quality Review process does not provide a fail-safe mechanism for identifying all new or modified operations is evidenced by the statement by Terp et al. (2002) that "As part of the usage survey process, MAQ personnel continue to identify several new/modified processes each year, which had not been identified through the Air Quality

Review program. Therefore, the interaction between MAQ personnel and facility points-of-contact continues to be a vital element of a thorough and complete usage survey, and is a valuable tool to supplement the Air Quality Reviews program."

It may not be efficient or practical to expect (or require, per LIR404-10-01.1 requirements) an Air Quality Review for every single new radionuclide, increase in amount, or new process in a given laboratory, particularly at a facility as dynamic and complex as TA 48-1. However, it is reasonable to expect that there be some mechanism in place to assess the potential impacts of operational changes before they occur. The annual (or biannual) usage survey does not serve this purpose; therefore, the ESH-ID Air Quality Review process is the primary tool that prospectively assesses new or modified operations. It may be possible to establish a facility-specific threshold for radionuclide use, below which an Air Quality Review is not required, rather than the currently instituted policy of an Air Quality Review requirement for any increase or change in use regardless of the magnitude. It may also be possible to relate this threshold to current facility operating limits, such as the handling limits for use of beta or gamma-emitting materials in a laboratory at any one time at TA 48-1 set forth by CST-SOP-037, R.7 (these limits require a health physicist consultation for any planned work involving quantities approaching 75% of 10 mCi [0.37 GBq]). Revising the Air Quality Review requirements in this manner would not preclude the inclusion of such use in the annual (or biannual) survey update, but it would help focus the Air Quality Review efforts, which are intended to assess operations before they happen, on those processes with the greatest potential for impacting monitoring requirements.

Specific Observation Related to Interaction with Radionuclide Users. There is little question that continued interaction by MAQ with facility contacts (i.e., radionuclide users) is a very important aspect of the compliance program. It keeps MAQ personnel abreast of potential future activities, and it helps establish a sense of importance at the facility level for the regulatory requirements to which they must adhere. During the course of this audit, however, it was clear in some cases that the facility level understanding gained through this interaction could be improved.

At TA 3-1698, radionuclide users reported information to MAQ personnel that suggested no particulate generation resulting from the operations. After some questioning, however, it became clear that the basis for this assumption was related to swipes taken to check for contamination after the equipment was cleaned. Clearly, cleaning a piece of equipment before taking a swipe would preclude the detection of any particulates generated during the process, so the information taken as meaningful by MAQ in fact had no bearing on demonstrating compliance. It is noted, however, that MAQ did assume a particulate release fraction, based on the nature of the process. At TA 21-257, the facility contact initially provided analytical data to MAQ that were based on a sample collected in 2002. The plutonium concentrations for this sample were believed to be incorrect (the higher than normal numbers instigated a follow-up), and the emission and dose calculations were revised to reflect the highest concentrations based on three representative samples from 2001. As noted previously, our suggestion from the second audit regarding implementation of a LANL-wide database that would allow radionuclide users to directly enter data was investigated by MAQ, and facility personnel indicated a preference for MAQ to maintain responsibility for data collection and entry.

These issues and responses suggest a lack of understanding in some cases at the facility level regarding the fundamental purpose of the usage data collected by MAQ. They also indicate that

facility personnel may not fully recognize their legal responsibility to keep track of and report all radionuclide usage. It is not possible for MAQ to be entirely responsible (or held accountable) for every aspect of the compliance program since it relies on information provided by many facility contacts. In fact, MAQ personnel have done an exemplary job of attempting to shoulder the burden of collecting and maintaining the information necessary to demonstrate compliance. However, considering LANL's reliance on radionuclide usage data reported by each facility for a number of different purposes, we recommend that further steps be taken to assist personnel at the facility level with developing a better recognition of the function and legal importance of usage data for which they are ultimately responsible. The preferences of facility personnel should be considered in this regard, but it is important that everyone associated with supplying and using data to demonstrate compliance with 40 CFR 61, Subpart H clearly understand their obligation to supply data that can be considered to provide a realistic (or conservative) representation of usage.

Potential Emissions and Dose Calculations

Summary of LANL Methodology

Based on the radionuclide usage survey information that is collected using the methodology described in the previous section, MAQ personnel estimate potential emissions from each unmonitored point source. These emission estimates are, in turn, used to estimate potential doses to the MEI for each unmonitored point source. MAQ staff used the procedures outlined in ESH-17-137, R1 to estimate potential emissions and doses from unmonitored point sources in 2001.

In addition to the changes discussed previously for compiling usage information, the database application, which was previously used only to compile radionuclide usage information, has been redesigned since the last audit to automate the process of calculating emission and dose estimates. This application has been designed to automatically calculate emissions for each radionuclide potentially contributing to emissions from a point source, based on usage and an assumed physical state reduction factor. In addition, the application is set up to estimate the PEDE for each radionuclide, based on the emission estimate and a derived millirem per curie factor[d] specific to each point source and radionuclide. Finally, a summed PEDE is calculated for each point source.

Appendix D methodology and FFCA guidance provide the basis for estimating emissions of materials that are used in various work processes at each facility, or are otherwise made available for release to the environment (e.g., waste drum inspection and characterization). This calculation considers the physical state of the material during the work processes that may involve radionuclide release, and appropriate reduction factors are used to estimate potential airborne emissions from solids (1×10^{-6}), particulates and liquids (1×10^{-3}), and gaseous materials (1). A material is considered to be a gas if it is heated to greater than 100°C, unless it is covered under the enhanced 100°C rule described in the FFCA. This methodology was reviewed during the course of the first audit, and it was deemed appropriately conservative (Aanenson et al. 1999).

Best engineering judgment, information from facility representatives, and other methods are used to estimate potential emissions based on smear and survey data. This may relate to surface contamination in a room or area that could be released via a point source to the environment or to

[d] Millirem per curie factors are LANL-derived factors that evaluate dose per unit activity.

contaminated equipment that could be a source of release to the environment. Historic monitoring data are used, when available and appropriate, to estimate potential duct holdup emissions. When historic monitoring data are used to estimate duct holdup, data from the last 2-4 years of available monitored emissions are typically used, provided the monitored operations are consistent with current operations. Monitored emissions are multiplied by the appropriate filtration factor (e.g., 2000 for single stage HEPA filtration, as specified in the FFCA) to estimate potential emissions.

Potential doses are calculated according to ESH-17-137, R1, and this value is the dose recorded in the usage survey for all unmonitored point sources with initially calculated doses less than or equal to 0.001 mrem (0.01 μSv). Because MAQ elects to initially calculate potential doses using assumptions presumed to be conservative, additional information may be obtained from facility representatives for point sources with calculated potential doses exceeding 0.001 mrem (0.01 μSv) to determine if a more realistic emissions estimate can be made. Additional information is obtained as necessary using best professional judgment until the calculated dose incorporates all relevant data, at which time it is recorded in the usage survey.

To estimate doses from all point sources, MAQ uses the potential emission estimates made for each facility (described above). Potential doses are determined by multiplying emission estimates by millirem per curie factors that have been calculated for the purpose of performing dose assessments for existing, new, or proposed facilities without having to run a dispersion and dose model each time. These dose factors have been calculated for up to 300 radionuclides at each LANL facility with the potential to emit radionuclides via a point source according to the procedures outlined in ESH-17-511 and ESH-17-512.

Calculating the millirem per curie factors is accomplished by first obtaining appropriate information for each release point, including the physical height, diameter, exit velocity, and X-Y location coordinates for each stack. Default CAP-88 values are used in cases where actual information is unavailable. A multi-year average of meteorological data is gathered for four meteorological towers at LANL for use in CAP-88 calculations. The maximally exposed individual or highest dose receptor location is determined and documented for each facility by performing preliminary CAP-88 runs, enabling identification of appropriate X-Y receptor location coordinates as well as the distance and direction from the source to the receptor. A generic list of radionuclides is appended to the CAP-88 input file for the source term input, and other radionuclides are added on a site-specific basis. A source term of 1 Ci (37 GBq) is assumed for each radionuclide, and progeny or decay products are included in the source term, based on guidance provided by NCRP (1996). The CAP-88 output is then electronically uploaded to tables in the database application.

These calculated potential doses are used for establishing point source tier level, determining monitoring requirements, and reporting annual dose from unmonitored point sources to the public. The decision to determine whether a sampling system should be installed or removed from a point source is made according to the guidance provided by ESH-17-138.

Potential emission calculations and dose estimates for each facility are peer reviewed to verify that calculations are accurate, assumptions are at least conservative, estimates are valid or at least conservative, and data entries into spreadsheets were performed correctly. These reviews are performed according to ESH-17-RN and are documented in the file folders maintained by MAQ for each facility.

Evaluation of LANL Methodology

As noted in the previous section, the MAQ program has actively followed through and addressed the recommendations and findings from the first and second audit reports. The methodology we suggested for estimating emissions at TA 21-213 was adopted. As discussed in the previous section, implementation of the redesigned database application has and will continue to significantly reduce the potential for making errors in emission and dose estimate calculations. MAQ has also taken steps to more thoroughly document the procedures used for calculating millirem per curie factors and has incorporated NCRP (1996) guidance to determine which progeny are important to consider for each millirem per curie factor.

We assessed the methodology used by MAQ with regard to potential emission and dose estimate calculations through detailed reviews of the file folders maintained for each facility. All calculations and assumptions related to estimating potential emissions are documented, as are data verification and peer reviews. It is apparent that MAQ expends a significant amount of effort in an attempt to calculate emissions in a thorough, accurate, and consistent manner, and this is supported by the documentation maintained by MAQ staff. In general, the calculations made by MAQ consistently adhere to the guidance and requirements set forth by 40 CFR 61, Subpart H and Appendix D, as well as the FFCA.

A database application has been developed to compile collected radionuclide usage information, assumed physical state reduction factors for each radionuclide, and millirem per curie factors for each radionuclide. Queries have been written enabling automated dose calculations for each facility. This trend toward increased automation by simplifying the dose calculations for the unmonitored point sources is encouraging and commendable.

For estimating duct holdup and potential emissions based on historic monitoring data, using the FFCA filtration factor (i.e., 2000 for single-stage HEPA filtration) is a more conservative approach than using the factor provided in Appendix D of 40 CFR 61, which corresponds to a single-stage HEPA filtration factor of 100 when used in this manner. As noted previously, using historic monitoring data to estimate potential emissions related to duct holdup is a conservative approach because such data inherently include all operational releases in addition to any potential releases related to duct holdup.

All unmonitored point source dose calculations are performed for the maximally exposed location or receptor for each point source, and they are based on potential emissions assuming no existing filtration mechanisms are functioning. The doses for each point source are then summed, and this collective potential dose is reported as the actual dose for all unmonitored point sources. This is a very conservative method of reporting doses for unmonitored facilities. The actual doses related to emissions from these facilities would in fact be considerably less than the 0.23 mrem (2.3 μSv) dose reported in the 2001 annual radionuclide air emissions report if existing filtration capabilities were taken into account and if the East Gate receptor (instead of release site receptor) was used to estimate the dose for each unmonitored point source.

Findings and Recommendations

As discussed previously, the MAQ program has demonstrated a willingness and desire to maintain a dynamic compliance program that responds effectively to the changing needs and goals of LANL. The MAQ program has actively followed through and addressed most of the

recommendations and findings from the first and second audit reports, and the changes that have been implemented have resulted in a more defensible and understandable compliance program.

We identified a number of areas where modifications or refinements would result in continued improvement of the LANL air quality program. The following discussion provides recommendations based on the findings of this audit that we believe will further strengthen the compliance program in several areas related to potential emission and dose calculations.

Specific Observation Related to Documentation for millirem per curie Factors. As was stated in the second audit report, it is again noted that the statement on page 10 of ESH-17-501 indicating that "…dose calculations for the rest of LANL…use the actual annual-average meteorology for the year in which emissions occurred" is not correct because of the use of calculated millirem per curie factors in dose calculations for unmonitored point sources, which are based on a multi-year average of meteorological data using the LANL met-tower nearest to the source (ESH-17-511). During interviews with MAQ personnel as part of the second audit, it was indicated that fluctuations of meteorological data from year to year are quite minimal and do not appreciably impact the calculated doses or the MEI locations. To reiterate our recommendation from the second audit, ESH-17-511 should include support for the assertion that the use of historical annual-average meteorological data is adequate for characterizing current annual conditions. One option for demonstrating this would be to use the archived millirem per curie factors to document that the changes resulting from assuming a different set of meteorological data are indeed insignificant. If they are not, sufficient runs of the model with representative meteorological data for a number of different years should be conducted and the most conservative (i.e., highest) millirem per curie factors should be used.

Specific Observation Related to Physical State Assumptions. In several instances (e.g., TA 54-36, TA 54-49, and the proposed characterization activities at TA 54 described by Fuehne 2002a), the physical state of solid that is assumed for drum contents is noted to be "consistent with ESH-17's (MAQ's) other analyses of operations at TA 54." During discussions with MAQ staff, it was realized that this assumption of a solid physical state for the drums handled at TA 54-36 was not consistent with information originally provided by the facility contact regarding the actual operations at TA 54-36, which indicated a particulate physical state. In this instance, the apparently incorrect assumption of a solid physical state did not impact the estimated PEDE because the dose was dominated by tritium, which was originally assumed to exist as a gas and, therefore, not affected by the change from solid to particulate. However, because of the six order of magnitude difference in estimated emissions that the physical state assumption can make, it is important that this assumption be sound in all cases. The rationale for assuming one physical state versus another should not be based on analyses of other operations, but should instead be based on facility-specific information related to the physical state of the actual materials used or processed at each facility. Additional procedures established for assuring the quality of data provided by each facility (discussed previously) should incorporate steps by which the physical state and process description information is also reviewed and checked by facility personnel.

Specific Observation Related to Appendix D Conservatism. Because MAQ operates under the assumption that emission estimates based on Appendix D guidance are extremely conservative, it would be useful to quantitatively demonstrate this conservatism. To this end, we

recommend that MAQ take steps to make relevant comparisons, wherever possible, of actual stack emission measurements with emission estimates made based on the methodology prescribed by Appendix D. Considering the questions regarding the accuracy of drum inventory estimates and the deviation from Appendix D methodology in certain cases [e.g., emission estimates for TA 54-33 and emission estimates made by Fuehne (2002a)], such comparisons could help confirm (or refute) the validity and assumed conservatism of the different methods by which emission estimate calculations are made.

The FFCA states that some minor point source emissions are measured for various reasons and that current (i.e., in 1996) sampling systems may be able to provide valid samples that could be used for periodic confirmatory measurements. The following is noted in ESH-17-138: "Historically, the Rad-NESHAP Project has de-energized and removed sample systems that were no longer needed. This, however, prevents the use of these systems for periodic confirmatory measurements. Therefore, this practice is no longer considered appropriate unless overriding factors (e.g., funding, access) necessitate it." In addition, MAQ calculates PEDEs based on usage data for all monitored point sources. As a result, it would appear that the opportunity exists to make a number of comparisons of emissions based on usage data and monitored emissions.

For illustration purposes, we make the following comparison using monitoring data for TA 50-69 (Size Reduction Facility). Releases of ^{241}Am and ^{238}Pu were reported in the 2001 LANL Radionuclide Air Emissions Report, corresponding to a release site receptor dose of 5.5×10^{-8} mrem (55×10^{-8} μSv). By comparison, the calculations for TA 50-69 in 2001 based on usage data and Appendix D guidance resulted in a PEDE of 43 mrem (0.43 mSv), which corresponds to an EDE of approximately 1.1×10^{-5} if the effect (i.e., reduction by a factor of 4,000,000) of existing two-stage HEPA filtration is considered. This comparison indicates that the emission estimate based on usage data and Appendix D guidance results in a dose more than 200 times greater than the dose based on measured emissions, and suggests that the emission estimate in this case is conservative.

Specific Observations Related to Continued Critical Review of Engineering Judgment Calculations. Clearly, the process of estimating potential emissions based on a usage value is not an exact science and necessarily involves making a number of assumptions. During the course of this audit, a number of questions were raised (see Appendix C) that instigated a closer examination of current assumptions or practices. One question related to the heat that may be generated as a result of machining operations involving depleted uranium at TA 3-102. Because no heat is applied externally, the emission estimate is based on the mass of the uranium pieces machined throughout the year and a solid physical state reduction factor. Based on an inquiry to the facility contact, the surface heating may reach 1100º C, which could lead to melting and/or oxidation and the generation of particulates. Therefore, a more conceptually accurate estimate of potential emissions could be to determine the fraction of each uranium piece that is subject to external heating, which would vary depending on the size and shape of the piece, the surface area to volume ratio, and the depth of cut. Because of the potential sensitivity related to this type of information, a demonstrated conservative estimate of the fraction of each piece that could be subjected to external heating would suffice. A liquid/particulate reduction factor could then be applied to this fractional amount. Alternatively, the weight of the uranium pieces before machining could be compared to the weight following machining plus the weight of any turnings or chips that are generated. The difference could be used to estimate emissions.

Another question that was raised related to the potential impact of trace contaminants of ^{99}Tc that could be present in uranium used in operations at TA 3-66 (Uranium Foundry). MAQ followed up on this question and examined historic monitoring data, which they noted to indicate very low actual emissions from the foundry operations into the early 1990s and no evidence of significant contamination of the depleted uranium fuel. MAQ personnel also followed up with the facility contact to better assess the potential impact of the presence of trace contamination in the uranium. The facility contact indicated that they could account for 99.999% of the material's composition as depleted uranium. For this calculation, MAQ assumed the remaining 0.001% was half ^{99}Tc and half ^{233}U, which would also be expected to be present as possible contamination in reactor-generated depleted uranium. The results of this calculation did not change the Tier III classification of this point source, but the PEDE did increase due to the significantly higher specific activity of ^{99}Tc.

It was also noted that the specific activity (e.g., Ci g^{-1}) used to convert ^{239}Pu usage by weight into activity was assumed to be that for pure ^{239}Pu in some, but not all, cases. By not considering the contribution of ^{240}Pu that is present with ^{239}Pu, the total activity is somewhat underestimated. Although MAQ personnel do distinguish weapons grade plutonium from other plutonium, it is not clear that the assumed specific activity for ^{239}Pu is applied consistently. For example, a value of 0.0622 Ci g^{-1} (2.30 GBq g^{-1}) was assumed to derive the usage estimate for TA 48-1 (ES 67) and a value of 0.0629 Ci g^{-1} (2.32 GBq g^{-1}) was assumed by Fuehne (2002a). Neither of these values is consistent with the value of 0.0613 Ci g^{-1} (2.27 GBq g^{-1}) noted by LANL (no date, DOE-STD-1027-92), which reportedly provides data tables to ensure consistency in LANL safety analysis work, and none of the values consider any contribution by ^{240}Pu. MAQ should adopt a consistent procedure for deriving activity estimates based on mass usage data for material containing plutonium isotopes.

Another issue raised during the audit related to the possibility of fires at unmonitored facilities using uranium, which is pyrophoric under certain conditions. Facilities where this may be a relevant issue are TA 3-102 (Chemistry and Metallurgy Research Laboratory) and TA 3-66 (Uranium Foundry). For unmonitored point sources, verbal notification of such an event (and subsequent reconstruction of its impact) would be the only mechanism by which the release could be quantified. For monitored point sources, the impact of such an event would be captured as part of the effluent monitoring data. MAQ staff investigated this question and indicated that the last fire at TA 3-102 was in the late 1980s. It was also noted that MAQ does have a staff member who responds to accidents or off normal occurrences, such as a fire, but because such an event would not be considered part of routine or normal operations it may not be reported to the MAQ personnel responsible for estimating annual emissions for compliance purposes.

This would appear to represent a misinterpretation of the regulation, which states that "Emissions of radionuclides to the ambient air from Department of Energy facilities shall not exceed those amounts that would cause any member of the public to receive in any year an effective dose equivalent of 10 mrem/yr." Therefore, all releases must be considered, including both routine and non-routine (e.g., episodic or accidental). Only in the sense that dose calculations for the majority of point sources use annual-average meteorology for the year in which emissions occurred (in some cases, multi-year average meteorological data are used) is the regulation focused on assessing releases as if they were routine or annual (i.e., occurring throughout the year). In other words, although the meteorological treatment of releases is not required by the regulation to be specific to the exact timing of the release, all releases occurring

during a given year must be considered regardless of their nature. It is recommended that MAQ incorporate additional steps into the radionuclide usage interview and survey processes described by ESH-17-102 and ESH-17-126 to ensure that any potential releases related to off normal or accidental occurrences are incorporated into the annual emissions estimates used to demonstrate compliance. In such circumstances, the treatment of atmospheric dispersion should be as specific to the timing of the release event as possible. In particular, it may not be appropriate to use annual average millirem per curie factors to evaluate the potential impact of a significant short-term release. This issue is discussed further in the section entitled "Complex Terrain Modeling Comparisons."

Critical review of engineering judgment assumptions is particularly important to maintain a technically defensible basis for demonstrating compliance. The evaluations we have completed as part of these audits have been beneficially augmented by the interaction of IEER and CCNS, as well as through participation by members of the public and local Pueblos. This interaction has resulted in a much more detailed and thorough evaluation than we could have completed without the participation of these stakeholders in the process of demonstrating compliance. We recommend that MAQ continue to actively support and seek critical review of engineering judgment calculations both internally and by other individuals with the experience or knowledge to provide meaningful input.

EVALUATION OF TA 54 WASTE CHARACTERIZATION DOSE CALCULATIONS

LANL Method for Dose Calculation

LANL completed two separate emission and dose estimates related to the waste characterization activities proposed for TA 54 (Fuehne 2002a). These characterization activities are intended to determine if waste (typically contained in 55-gallon [208 L] drums) meets transportation requirements and acceptance criteria for the Waste Isolation Pilot Plant (WIPP). The waste characterization activities will be performed in modular units (MUs) and may include temperature equilibration, headspace gas analysis, measurement of hydrogen generation, and repackaging. The first dose estimate for these activities was performed to determine if an application for pre-construction approval was required to be submitted to EPA Region IV. The second dose estimate was performed to determine if continuous monitoring is required for the operations. The procedures used to estimate doses for each of these purposes are briefly described below. Additional details regarding these calculations are provided in LA-UR-02-4138.

Application for Pre-Construction Approval

The dose calculations to determine the need for a pre-construction approval are based on 40 CFR 61, Subpart H, Appendix D methodology. These calculations employ an assumed usage amount for one year, an assumed emissions reduction fraction based on the physical state of the material, and an emission factor adjustment based on effluent controls (e.g., HEPA filtration).

The assumed usage amount is based on a reportedly conservative estimate of average drum inventory and operational estimates of the number of drums processed each year. The emission factor adjustment of 1×10^{-6} is based on the assumption that the drum contents are accurately characterized by a physical state of solid. The effluent control factor of 0.01 is based on the presence of HEPA filtration on both the inlet and exhaust of the ventilation system and Appendix D guidance this type of effluent control.

Analysis of Monitoring Requirements

The dose calculations to determine the need for a pre-construction approval are based on engineering judgment methodology (per the FFCA) and the emission determination methodology as prescribed in §61.107. These calculations employ an assumed usage amount for one year and an assumed emissions reduction fraction based on the physical state of the material. However, the calculations do not incorporate an emission factor adjustment based on effluent controls (e.g., HEPA filtration) and are to be "…based on the discharge of the uncontrolled effluent stream into the air" per §61.107 guidance.

The assumed usage amount for the monitoring requirement determination is different than for the pre-construction approval application and is noted to represent "…a more realistic scenario." For the monitoring requirement determination, usage is based on the same estimate of average drum inventory. However, the annual usage is not based on operational estimates of the number of drums processed each year. Instead, emissions are evaluated by assuming two drums in "steady state" operation for the entire year. The emission factor adjustment of 1×10^{-6} is based

on the assumption that the drum contents are accurately characterized by a physical state of solid. No effluent control factor was used.

Assessment of Methodology: Findings and Recommendations

The requirements of 40 CFR 61, Subpart H related to applications to construct or modify (§61.96) specify that Appendix D guidance must be used to determine the need for pre-construction approval from EPA. The requirements of 40 CFR 61, Subpart H related to determining monitoring requirements [§61.93(b)(4)(i)] specify that:

> "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 EPA approval, DOE may determine these emissions through alternative procedures."

Per Appendix D guidance, the methodology to determine emissions is to be based on "…the amount used at the facilities for the period under consideration…" The period under consideration is based on the standard prescribed in §61.92, which applies to a period of one year. Based on the requirements of 40 CFR 61, Subpart H, Appendix D methodology must be used to estimate emissions when determining the need for pre-construction approval, but "alternative procedures" may be used to determine monitoring requirements with prior EPA approval. Therefore, the emission estimate made to determine monitoring requirements for the proposed operations at TA 54 do not need to be based on Appendix D guidance, and can instead be based on engineering judgment calculations, as specified in the FFCA. However, if Appendix D methodology is going to be cited as the methodology used for estimating emissions, the amount "used" necessarily equates to the total drum throughput (i.e., usage) during the course of a year.

Fuehne (2002a) notes in the assessment to determine monitoring requirements that "The Appendix D release fractions assume that the entire inventory is available for release for an entire year;" however, it is not clear to us where this assumption is stated (or implied) in Appendix D. Further, it is not consistent with the MAQ treatment of usage to estimate emissions for other similar operations when Appendix D methodology is used. For example, the assessments at both TA 54-36 and TA 50-69 (where operations are similar to the proposed TA 54 operations) consider the total throughput for the year as the basis for estimating emissions.

The assumption of some number of drums in "steady state" conditions based on operational limits may be appropriate in some cases, such as the assessment done for TA 54-33 (Drum Vent System), where headspace gas sampling and hydrogen gas purging are the only activities done. In this case, engineering judgment is used to estimate dose per FFCA approval (Section 2.1.1.4). However, the operations at TA 54-33 are not consistent with the proposed operations at TA 54, which will involve both waste characterization and repackaging and are difficult to envisage as "steady state." As a practical note, assessment of emissions at TA 54-33 could perhaps be more realistically related to analyses of the headspace gas that is sampled from each drum.

In addition to estimating an effective annual throughput, it is also necessary to derive release fractions based on the physical state of the material that is used or otherwise made available for potential release. For the proposed activities at TA 54, a physical state of solid is assumed, and Fuehne (2002a) defends this assumption by stating that it "…is consistent with other waste handling operations at LANL TA 54 …" However, during the course of this audit, it was noted that this assumption led to an inappropriate physical state assignment at TA 54-36 (see recommendations in the Unmonitored Point Source and Usage Survey Evaluation section "Physical State Assumptions"). We previously recommended that the basis for assuming one physical state versus another should not be based on analyses of other operations, but should instead be based on facility-specific information related to the physical state of the actual materials used or processed at each facility. We reiterate that recommendation here.

Because 40 CFR 61, in combination with the FFCA, provide for the option of engineering judgment calculations in lieu of strict Appendix D methodology for determining monitoring requirements and annual dose estimates, it is not unreasonable to pursue this option in some instances. Particularly where engineering judgments are used to estimate emissions, though, it is important that the methodology be sound and defensible (see recommendations in the Unmonitored Point Source and Usage Survey Evaluation section "Specific Observations Related to Continued Critical Review of Engineering Judgment Calculations").

Currently, to our knowledge, engineering judgment calculations assuming some number of drums in "steady state" without any regard for the actual total number of drums processed have no demonstrated validity. The historic operations at TA 50-69 were noted during discussions with MAQ personnel to be similar to the waste characterization operations proposed at TA 54 (Fuehne 2002a). The fact that very low or no detectable emissions have occurred at TA 50-69 suggests that the proposed operations at TA 54 may result in similarly low emissions; however, this conclusion cannot be substantiated without additional investigation and comparison of throughput for the two operations.

The proposed operations evaluated by Fuehne (2002a) are noted to be part of a pilot program for consideration at other Department of Energy sites, and the dose estimated to determine monitoring requirements very closely approaches the dose that would require monitoring (0.08 mrem versus 0.1 mrem [0.8 μSv versus 1 μSv]). As a result, we recommend additional demonstration to show that the calculation done to determine monitoring requirements for the proposed TA 54 operations is valid so that it can be more thoroughly defended. Ideally, the operations would be monitored for a period of one year or more, during which the highest-wattage drums could be processed, to clearly demonstrate low emissions. We consider monitoring the operations to be the best possible mechanism to demonstrate the conservatism related to the potential emission estimates made using the different methodologies described by Fuehne (2002a) and strongly recommend that it be performed for at least the initial stages of this project, unless applicable data on potential release fractions can more clearly demonstrate that monitoring is not needed. As noted previously in the section "Specific Observation Related to Appendix D Calculations," we also recommend that MAQ take steps to make relevant comparisons, wherever possible, of actual stack emission measurements with emission estimates made based on usage data. These comparisons would be useful to MAQ in the future for demonstrating the level of conservatism associated with the various methodologies currently employed to estimate emissions.

STACK SAMPLING AND MONITORING EVALUATION

This chapter deals with point sources of radioactive releases to the atmosphere at the Los Alamos National Laboratory that are sampled or monitored. Unmonitored point sources were discussed in the previous chapter and non-point sources are discussed in the next chapter of this report.

Methods for monitoring, sampling, and analysis of effluents are specified in 40 CFR Part 61, § 61.93. Section 61.93 provides specific requirements that apply to monitoring or continuous representative sampling of discharges. Appendix B, Method 114, of 40 CFR Part 61 focuses on the requirements for sample collection, various types of analytical measurements made on collected samples, and real-time monitoring for radioactive gases discharged from stationary sources, such as stacks and building vents. The requirements of § 61.93 and Method 114 apply to the LANL effluent discharges that must be measured to comply with 40 CFR 61, Subpart H. Those discharges are measured, either continuously in real-time or by sequential collection and analysis of effluent samples throughout the year. This portion of the audit focused on evaluating the continuous effluent monitoring, sampling protocols, and analytical methods for locations that were estimated to be the main contributors to the offsite dose.

The sampled and monitored stacks were again the main contributors to the estimated offsite dose in 2001 (LANL 2002). At the East Gate business location, where the estimated dose was 1.84 mrem (18.4 μSv), the sampled and monitored stacks contributed about 84% of the total dose. Most (~97%) of that contribution was due to releases from the nearby Los Alamos Neutron Science Center (LANSCE) facility and the remainder resulted primarily from releases of tritium at other facilities in TA 16 and TA 21. Releases of long-lived alpha-emitters from the Chemical and Metallurgical Research (CMR) facility at TA 3 and the plutonium facility at TA 55 did not make significant contributions to the estimated dose.

The East Gate business location, north-northeast of LANSCE, has in most years been the place where the estimated offsite dose was highest. Radiation doses from LANSCE are estimated monthly to permit administrative control (ESH-17-610, R1) over the releases of short-lived activation gases that are only produced during accelerator operation.

Reliable estimates of the amounts of radionuclides released in effluents depend on knowledge of the effluent flow rate and the concentration of radionuclides in the effluent air. The quantity of radionuclides released is the product of its concentration in the air stream and the flow rate of the air out of the stack or vent. To measure some radionuclide concentrations properly, it is necessary to obtain a representative sample from the effluent stream, collect the radionuclides present in the sample, and measure the amounts of radioactivity collected. An alternative approach, used at the LANSCE facility, is to install instrumentation that can analyze the radionuclide concentrations and estimate releases as they occur. In either case, estimation of the release is a multi-step process, and there are EPA requirements related to each of the steps involved.

The following sections discuss the requirements that are most generic and apply to all sampling locations. These are the procedures for effluent flow measurements, selection of effluent sampling locations, extraction of effluent air samples, and transport of the sample to the collection device or measurement point. Subsequent sections address collection and measurement of radionuclide concentrations in effluent samples. Some measurement methods apply to more

than one facility. Each section includes a discussion of the applicable regulatory requirements, LANL methodology, an evaluation of the LANL approach, and any findings of this audit.

Effluent Flow Rate Measurements

Knowledge of the rate of discharge of effluent air is essential to accurately estimate radionuclide discharges. Because it is equally important for other pollutants regulated under the Clean Air Act, the EPA had established methods for measurements of the amount of air flowing in a stack or vent before radionuclide releases were regulated. The EPA had published appropriate methods in Appendix A of 40 CFR 60. Different methods are used to measure effluent flow rates (a) in large stacks and vents and (b) in pipes and small vents.

Summary of LANL Methodology

LANL uses the methods specified by the EPA in 40 CFR 60, Appendix A, to measure effluent air flow. The LANL methodology is described in detail in a recently updated procedure (RRES-MAQ-127, R3). A contractor organization, Johnson Controls of Northern New Mexico, conducts the air flow measurements for LANL. In 2001, stack flows were measured routinely once a quarter. Additional measurements are made when there are major changes in ventilation systems or at other times in response to requests by MAQ. The routine measurement frequency is not affected by the occurrence of special measurements.

Measured stack flow rates can differ from time to time because of changes in fan operation and effluent filtration units. For example, some stacks have both a primary exhaust fan and a back-up exhaust fan whose discharge flow rates may differ. To ensure that effluent releases are not underestimated, it has been LANL policy to use the highest flow rate measured during the previous three years in the release calculation.

Evaluation of LANL Methodology

The flow measurement techniques employed by LANL follow regulatory guidance established for the Clean Air Act. The approach is basically sound and the application of the flow measurement data is cautious. The policy of using the highest of the last twelve flow rate measurements leads to estimates of the amounts of radionuclides released that would nearly always exceed those that actually occurred.

Beginning in 2001, the data have been entered into a database called “stacks” from which the appropriate value can automatically be extracted for dose assessment calculations. Complete independent review of the data entry procedure is required. Beginning in 2002, the frequency of stack flow rate measurements was reduced to twice a year; however, s-type pitot tubes have been installed together with Magnahelic gages to provide visual indication of the stack flow rate. Some of the gages were observed during our tour of the CMR facility; they have been marked to indicate the expected range based upon historical measurements. The gages will be checked routinely during the weekly sample collection process. The new approach will provide more rapid identification of stack flow rate changes.

Selection of Effluent Sampling Locations

Because it is essential that collected samples of the air being discharged represent the properties of that effluent, it is important that the location for the sample be chosen carefully. The EPA identified this issue when regulating other pollutants before establishing 40 CFR 61, Subpart H. Subpart H employs guidance that was established earlier. It specifies in § 61.93 (b) (2) that Reference Method 1 of 40 CFR 60, Appendix A (or other method for which EPA has given prior approval) be used to select sampling sites. Reference Method 1 relies upon a rule-of-thumb to avoid disturbances of the effluent flow that could cause samples to be non-representative; namely, that the location should be at least eight duct diameters downstream and at least two duct diameters upstream of a major flow disturbance.

Summary of LANL Methodology

The preferred method used by LANL is an alternative method that has received prior approval from the EPA. Beginning in 1993, DOE requested EPA approval of a method of sampling point selection for all DOE facilities based upon quantitative measurement rather than the rule of thumb (Pelletier 1993). It is known from measurements of trace gas concentration profiles that a sampling location chosen using the rule of thumb does not guarantee that the effluent will be well mixed. The goal of the proposed approach was to identify a location where the effluent is well mixed, to sample at that location using a single highly efficient sample extraction method (the shrouded probe) to collect the sample, and to transport the sample to the collector using an optimized line. The sample extraction and transport line aspects are discussed in a later section.

Following the agreement on the Clean Air Act Memorandum of Understanding (Ogé 1994) (MOU) between the EPA and DOE in the fall of 1994, the EPA gave approval to use an alternative method for selecting sampling locations (Nichols 1994). That approval letter and subsequent correspondence (Kirkman 1995; Saginaw 1995) established the conditions under which single-point sampling using a shrouded probe could be used. Those conditions are all listed in our first audit report (Aanenson et al. 1999). When single point sampling using a shrouded probe is not feasible at a location because conditions listed above or others given below are not met, the approach of Reference Method 1 is employed. Because needs for effluent monitoring were not given adequate attention during facility design, construction, and renovation, there are some stacks at LANL where application of Reference Method 1 is impractical. Sampling locations for those stacks are selected under the provisions of § 61.93 (b) (3), which provides for prior EPA approval of documented procedures that will not significantly underestimate emissions.

Evaluation of LANL Methodology

The sampling location approach preferred at LANL that was approved by the EPA is part of the revised American National Standards Institute (ANSI) sampling guide (ANSI N13.1 1999) for releases of airborne radioactivity. The previous ANSI guide (ANSI N13.1 1969) is still incorporated by reference in the EPA regulations, but the agency has proposed incorporation of

the new guidance in the 40 CFR 61, Subpart H regulation. It is anticipated that a revised regulation will take effect in 2003, but it has not yet been published.

Selection of a sampling location where the effluent is well mixed is known to be satisfactory and is a clear improvement over the rule-of-thumb guidance that is part of Reference Method 1. For other stacks, the EPA-approved sampling locations are unlikely to lead to underestimates of the annual releases.

During 2001, most of the locations employed for sampling effluents from stacks and vents at LANL were the same as those used at the time of the previous two audits. The EPA has approved the sampling locations being used at LANL.

At the time of the last audit, LANL had identified new sampling locations, which met the requirements of EPA Reference Method 1 and ANSI standards (ANSI N13.1 1969), for ten stacks at the CMR facility. Since then, installation and testing of equipment were completed and measurements with the new systems were begun in 2000. The old and new sampling locations were both operated during 2001. Comparisons of the results of these measurements are discussed in a later section of the report.

Sample Extraction Techniques

The requirement that the effluent sample represent the properties of the air being discharged also affects the method of withdrawal of the sample from the airstream. In § 61.93 (b), the requirement states:

> Representative samples of the effluent stream shall be withdrawn continuously from the sampling site following the guidance presented in American National Standards Institute (ANSI) N13.1-1969 'Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities' (including the guidance presented in Appendix A of ANSI N13.1).

Appendix A of the ANSI guide deals specifically with sampling ducts and stacks. Briefly, the approach recommends sampling isokinetically at several points in the cross-section of the stack to ensure that the total sample collected is representative of a possibly non-uniform distribution of radionuclides in the air being discharged. Isokinetic sampling is achieved when the air velocity entering the probe is the same as the air velocity in the stack. As is the case for other methods presented in § 61.93 (b), alternative techniques may be used if prior approval is received from the EPA.

As noted above, the 1969 ANSI guide is part of current EPA regulations, which applied during 2001. Changes to the regulations are anticipated in the near future.

Summary of LANL Methodology

The preferred method used by LANL is an alternative method that has received prior approval from the EPA. As discussed above in the section on sampling location, the DOE requested and received prior approval for a technique that identifies a location where the effluent is well mixed and obtains samples using a single highly efficient sampling probe. This procedure employs a probe, called the shrouded probe, which intentionally avoids isokinetic sampling to reduce deposition of particles in the inlet of the sampling probe. The EPA approved the

alternative method of sample extraction using a single shrouded probe in November 1994 (Nichols 1994). The approval letter includes conditions for the use of shrouded probes. Those conditions were enumerated in the first audit report (Aanenson et al. 1999), which also included details related to the approval by the EPA Regional Office.

When single-point sampling using a shrouded probe is not feasible at a location because either a sample location condition or a shrouded probe condition is not met, LANL employs an approach that utilizes multiple sample extraction probes in accord with current EPA guidance (RRES-MAQ-127, R3). The sample extraction assembly with multiple probes installed is called a "sampling rake." LANL also uses ANSI sampling rakes with multiple probes to sample effluents from stacks for which Reference Method 1 cannot be used to select a sampling location.

In response to a recommendation from the first audit, LANL has selected a cautious correction factor for losses in rake sampling systems based on information for 5-µm aerodynamic diameter (AD)[e] aerosol particles.

Evaluation of LANL Methodology

During 2001, most of the sampling probes employed in stacks and vents at LANL were the same as those used at the time of the earlier audits. The shrouded probe, that is preferred by LANL and was approved by the EPA, is a superior approach and has been recommended in the revised ANSI guide (ANSI N13.1 1999). The EPA has also approved the use of multi-probe rakes in other sampling locations. The new sampling systems in the CMR facility employ new Kurz low loss sampling rakes. The LANL sample extraction techniques meet the regulatory requirements.

The corrections for losses in sampling systems most likely overestimate actual losses. The expected aerodynamic equivalent diameter of the particles being sampled is substantially less than the assumed 5-µm AD because aerosol filtration systems are installed upstream of the sampling points. The particles being sampled are more likely to have ADs in the sub-micron range, and sampling losses for those particles are much lower than for 5-µm AD particles. LANL's cautious approach is not likely to lead to dose estimates for particulate radionuclides that approach the 10-mrem (100-µSv) dose standard of 40 CFR 61, Subpart H. Nonetheless, the assumed correction factor must be remembered when making comparisons between concentrations of particulate radionuclides predicted using the estimated releases with the concentrations measured by the AIRNET stations.

During the second audit, the IEER monitors suggested that sampling rakes in both the CMR and TA 55 facilities should be improved (Aanenson et al. 2000). As noted, ten new systems have been installed in CMR facility stacks; however, the systems at the TA 55 plutonium facility have not been modified. The audit team had suggested that a decision about those systems should await the updated EPA regulations. When the anticipated changes in the regulations occur, then an appropriate decision can be made. The TA 55 sampling rakes were inspected during 2001 and found to be adequate for sampling in the interim.

[e] The aerodynamic diameter is the diameter of a unit density sphere that has the same terminal settling velocity in air as the particle of interest.

Sample Transport Lines

Transport lines carry sampled air from the outlet of the sampling probe to the point of sample collection or location of a continuous monitoring system. Deposition on the walls and in bends of the line can lead to losses of particulate radionuclides from the sample. Therefore, transport lines should be kept as short as is feasible given the conditions at the sampling location. Appendix A of the ANSI guide (ANSI N13.1 1999), which is included by reference in § 61.93 (b), deals specifically with sampling ducts and stacks. Appendix B of the same guide, which discusses particle deposition in sampling lines, is not included by reference in § 61.93 (b).

Summary of LANL Methodology

As part of the DOE request (Pelletier 1993) for EPA approval to use the alternative method, DOE proposed using the DEPOSITION computer code to optimize the transport line and to estimate transmission losses. This computer code was developed at Texas A & M University and had been accepted by the Nuclear Regulatory Commission for estimation of losses in transport lines. The EPA approved the proposed approach under certain conditions, which were all given in the first audit report (Aanenson et al. 1999).

For sampling locations that meet the requirements for use of the shrouded probe, LANL uses the DEPOSITION code to optimize the sampling line by selecting a line diameter that minimizes deposition for the flow rate needed for sampling. For these lines, LANL also uses the DEPOSITION code to estimate transmission losses in the probe and sample transport line.

Evaluation of LANL Methodology

The process of accounting for losses in transmission lines has been improved since the time of the first audit. Some of the sampling lines have been optimized; others have low transmission losses as the result of good practice based upon practical experience. Particle transmission fractions for sampling lines are listed in an attachment to the procedure for calculating emissions (ESH-17-114, R2).

Findings and Recommendations

Specific Observation. The procedure for calculating emissions (ESH-17-114, R2) gives a transmission factor of one (1.0) for vapors. While this is valid for the only location where such vapors were measured (LANSCE, which employs shrouded probes with very short sampling lines), it cannot be considered to be broadly applicable. Studies of vapor transmission losses have shown non-trivial deposition for elemental iodine, and it can be expected that the behavior of elemental bromine would not be greatly different. Mercury vapors may also deposit in sampling lines. If and when it is envisioned that operations in other facilities, such as CMR and TA 48, will lead to releases of reactive vapors, LANL will need to develop estimates of appropriate transmission factors for the sampling systems in question. The audit team made a related recommendation in the report of the second audit (Aanenson et al. 2000); namely, that (when feasible) the efficiency of the charcoal cartridges used for sampling these vapors should be

checked at other locations because the chemical forms of the airborne vapors may differ from those studied at LANSCE.

Collection and Analysis of Tritium Samples from LANL Stacks and Vents

The techniques used for collection and analysis of tritium in airborne effluents from stacks and vents are similar for all the LANL facilities with significant tritium emissions. Tritium sampling was discussed in some detail during the previous audit (Aanenson et al. 2000), which included onsite inspections of the sampling systems. The LANL systems and procedures for sampling tritium were found to follow EPA requirements and were not a focus of the current audit.

Analysis of Airborne Effluents from the Los Alamos Neutron Science Center (LANSCE)

The primary releases from the Los Alamos Neutron Science Center (LANSCE) are short-lived radioactive gases that are produced by operation of the linear accelerator. Lesser amounts of beta-gamma-emitters are released in particulate form or as vapors from the facility. Sections 2.2.3 and 2.2.4 of Method 114 in Appendix B of 40 CFR 61 apply to the short-lived gases.

> Radionuclides of argon, krypton, and xenon . . . are either measured directly by an in-line or off-line monitor, or are collected from the extracted sample by low temperature sorption techniques. Appropriate sorbers may include charcoal or metal zeolite.
> Radionuclides of oxygen, carbon, nitrogen, and radon . . . are measured directly using an in-line or off-line monitor. Radionuclides of carbon in the form of carbon dioxide may be collected by dissolution in caustic solutions.

An "in-line monitor" refers to a continuous measurement system in which the detector is placed directly in or adjacent to the effluent stream. An "off-line monitor" refers to a measurement system in which the detector is used to continuously measure an extracted sample of the effluent stream. Either may involve gross radioactivity measurements or specific radionuclide measurements.

The sampling protocol in Appendix B that is most appropriate for the reactive gases that are released from the facility is that for iodine, also a reactive gas. In both cases a collection medium such as impregnated charcoal or metal zeolite is used to trap the reactive forms (and less reactive forms that may exist in some exhausts). Particulate radionuclides are collected using filters as described earlier.

Summary of LANL Methodology

For the short-lived gas releases that are released through two stacks at LANSCE, LANL employs off-line monitors that employ Kanne flow through ion chambers and gamma spectrometers with high purity germanium (HPGe) detectors that are connected to multi-channel analyzers. Methods for testing and calibrating these systems are specified in LANL procedures (ESH-17-603, R4; ESH-17-604, R5). Each stack has a separate pair of off-line monitors. Outputs

of the Kanne ion chamber and gamma spectrometry systems are processed and recorded automatically using a computer system. The two measurements are routinely compared and agreement within ± 10% is required. The ion chambers provide a gross measurement of total activity released, while the gamma spectrometer is used to identify the radionuclides that are released. Several of the radionuclides of interest (^{11}C, ^{10}C, ^{15}O, ^{14}O, ^{13}N) are positron-emitters. When positron annihilation occurs, two 511-keV photons are emitted and those gamma rays are characteristic of all these radionuclides. Other nuclides (^{16}N, ^{41}Ar) that are released are beta-gamma emitters and are characterized by distinctive gamma ray energies.

Contributions of these radionuclides to the total release depend upon their production rates and half-lives. Typically, the longer-lived nuclides are most prominent in the discharge. For example, the half-life of ^{11}C, the predominant nuclide, is about 20 minutes, while the half-life of ^{10}C, a minor component of the mixture, is only about 9 seconds. The isotopic composition of the positron emitters is determined by observing the decay of the 511-keV peak using high-resolution gamma spectrometry.

Diffuse emissions of the radioactive activation gases leaking out of buildings, primarily near the main beam line, also occur and are estimated separately (ESH-17-611, R1). Kanne ion chamber data within the facility are used to estimate concentrations of radioactive gases, which are multiplied by estimated volumes of air released from the area as the result of leakage.

Although much less important to the annual dose assessment, releases of radioactive particles and vapors are sampled continuously, with weekly sample exchanges (ESH-17-601, R6; ESH-17-612, R0). Shrouded probes are used to extract those samples, which are collected by sampling trains, each of which consists of a particulate filter followed by an impregnated charcoal cartridge. These samples are analyzed by high-resolution gamma spectrometry in the Health Physics Analytical Laboratory.

Evaluation of LANL Methodology

The procedures cited above are now the responsibility of MAQ and most have been revised since the last audit. Based upon those updated procedures and information obtained during the audit tours of the LANSCE facility, we concluded that the LANSCE methods meet the EPA requirements for effluent monitoring and sampling for 40 CFR 61.

The short-lived gas monitoring systems are sophisticated and the dual measurement approach provides for high reliability for monitoring the important radioactive gas release points. The sampling system for particles and vapors employs shrouded probes at locations where the effluents are well mixed. As noted earlier, this is a superior approach. The vaporous radionuclide collection by the impregnated charcoal cartridge is assumed to be 65% efficient. Testing, using two identical cartridges in sequence to collect these nuclides from the LANSCE effluent, has shown the efficiency for these cartridges to be higher (87–100%) than the assumed value.

The procedure for estimating fugitive releases, which accounted for about 5% of the dose estimated for the East Gate site in 2001, is satisfactory and likely provides cautious estimates of releases. The Kanne ion chamber data that are used in those release calculations are considered quite reliable.

Because of the importance of LANSCE releases to estimated offsite doses, procedures were established in 1997 to track LANSCE emissions and estimated doses throughout the year and to carefully check those estimates as the projected dose increased (ESH-17-610, R1; ESH-17-609,

R1; ESH-17-608, R1). The graded administrative control was intended to assure that releases from LANSCE did not produce estimated doses that closely approached the EPA standard.

Collection and Analysis of Airborne Particles Released from Facilities at TA 3 and TA 55

Several radionuclides, primarily alpha-emitters, are released in the form of particles from many individual stacks at the CMR Facility, TA 3-29. As noted earlier, new sampling systems have been installed in ten of those stacks. Particulate alpha-emitting radionuclides are also released from other facilities, such as the TA 55 plutonium facility. Sample collection and analysis methods and the procedure for estimating releases of alpha-emitters from these facilities are the same and are discussed together in this section.

The requirements of 40 CFR 61, Appendix B, Method 114, describe sample collection and analysis methods for radionuclides present in particulate form:

> The extracted effluent stream is passed through a filter media to remove the particulates. The filter must have a high efficiency for removal of sub-micron particles. The guidance in ANSI N13.1–1969 shall be followed in using filter media to collect particulates.

Summary of LANL Methodology

Weekly samples are collected downstream of effluent filtration systems in the several exhaust stacks using LB-5211 filters. The collection efficiency for 0.3-µm unit density particles by the LB-5211 filters has been measured. In six tests, the collection efficiencies ranged from 99.2 to 99.8%, and the mean value was 99.6%. As a screening procedure, these sample collection filters are analyzed by direct counting to determine total alpha radioactivity. Half of each filter is included in a quarterly composite sent to an offsite radiochemical laboratory for determination, by chemical separation and alpha spectrometry, of the alpha-emitting radionuclides present in the composite sample. LANL retains the other half of the filter.

The results of the alpha spectrometric analysis are used to estimate isotopic releases from the stacks. In the calculations, corrections (discussed earlier) for losses of particles during sampling collection and transport to the filter are applied. As for other releases, the maximum value among the last twelve quarterly flow rate measurements is used when calculating releases.

Evaluation of LANL Methodology

The methods used to estimate releases of alpha-emitters have changed since the time of the first audit. Analysis of composite samples using alpha spectrometry following chemical separation is a more reliable method for determining the effluent radionuclide concentrations. The current method has been used since the beginning of 1997.

Corrections for particle collection efficiency are not made; the difference between the assumed collection efficiency of one and the measured average of 0.996 is not significant. The collection efficiency measurements were made using particles that were in the size range (0.1–0.5 µm equivalent aerodynamic diameter) most likely to penetrate the filter medium. Particles that are either smaller or larger than those in this range are collected more efficiently. Thus, the

collection efficiencies found for the 0.3-µm unit density particles can be applied broadly without fear of serious error. Performing some plant effluent measurements using two particulate filters in sequence could provide additional confidence in the collection efficiency of those filters under current field conditions.

LANL has continued the program of routine operational checks and preventative maintenance for the effluent air sampling pumps. New equipment has been obtained for field checks of sampling flow rates, which are documented. The sampling pumps are now equipped with run-time indicators to quantify the duration of any sampling system outages that may occur. Overall reliability of the sampling systems has been quite high.

During 2001, two sampling systems were operated in each of ten stacks of the CMR facility in TA 3. Seven older sampling locations are in short straight sections of duct between the filters and the exhaust fans; three older sampling locations are relatively close to the exhaust fan discharge into the stack. The new sampling locations are all near the tops of the stacks, further from the point at which the discharge from the fans enters the stacks and all satisfy the EPA requirements. Operation of both systems during the year produced a set of data that can be analyzed in several ways. After a series of increasingly refined comparisons, Fuehne (2002b) concluded that the estimates from the new sampling systems gave generally higher estimates of releases. He found 19 comparisons with higher estimates versus 11 that gave lower estimates; 24 comparisons were not statistically different. On the basis of this analysis it was decided to use the data from the new systems to prepare the release estimates for 2001.

Table 2 shows the results of another review. The net activities per composite sample and the associated uncertainties from the analytical reports of analyses of composite samples for the first two quarters and last two quarters of the year. The ratios of the results from the new to the old sampling systems were computed as were the uncertainties in those ratios for the individual isotopes reported to have detectable concentrations.

Some results were perturbed by maintenance activities. Fuehne (2002b) linked the difference for stack 3-29-32 to a filter change operation; the old sampling system is much closer to the filters and appears to have collected activity made airborne during the operation. At the same time the new sampling system was sampling air that was not affected by the maintenance. The set of high ratios for stack 3-29-28 was investigated and found to be associated with fan maintenance in October. The gross alpha data show that the new sampling system was measuring activity that was apparently resuspended from surfaces in the fan area and carried out the stack. Elevated concentrations measured by the new sampling system occurred during the week of the work and during the next three weeks as well. The total of these weekly gross alpha values is consistent with the value for the 6-month period. The fan is downstream of the old sampling system and fan operation would preclude movement of the activity toward that sampling point.

Table 2. Calculated Ratios of Results from New Sampling Systems to Those from Old Sampling Systems[a]

CMR	Ratio of sampling results (new system/old system ± one standard deviation) for radionuclides measured							
Stack	^{241}Am	^{210}Pb	^{210}Po	^{238}Pu	^{239}Pu	^{234}U	^{235}U	^{238}U
-14		0.78±0.25	0.78±0.15					
		0.99±0.22						
-15			0.80±0.17[b]					
-19	0.62±0.08	0.94±0.04	1.01±0.06	0.87±0.10	0.39±0.03			
	1.21±0.32	0.94±0.03	0.87±0.05	1.08±0.17	0.54±0.06			
-20		1.04±0.07	1.02±0.08	1.98±0.75	3.78±1.04			
		1.07±0.04	1.18±0.08					
-23	1.05±0.37	1.06±0.03	0.97±0.05	0.46±0.13		1.49±0.07	1.05±0.11	1.49±0.16
	1.58±0.68	1.03±0.02	0.90±0.05	1.43±0.46	0.95±0.73	1.95±0.11	2.03±0.33	1.52±0.23
-24	1.75±0.77	1.08±0.03	0.93±0.06	2.13±0.31		1.40±0.48		
	0.70±0.25	1.17±0.03	1.14±0.07	1.46±0.17	0.55±0.17	1.53±0.09		
-28	1.35±0.24	2.01±0.38	1.66±0.21	1.61±0.09	1.76±0.14			
	6.41±1.30	1.82±0.41		10.4±0.66	3.17±0.26			
-29	2.57±0.46	2.07±0.22						
		3.14±0.73						
-32		0.85±0.08	0.87±0.08	[c]				
		0.70±0.17						
-33		1.16±0.12	1.41±0.13	0.06±0.02				
		1.21±0.24						

[a] For each stack in the table, the first row contains data for the first two quarters of 2001, and the second row contains data for the final two quarters of 2001.

[b] Result for quarters 1 and 2.

[c] Ratio was 0.00017±0.0006.

It appears that the new systems are more efficient for sampling activity released from process areas: uranium in stacks 3-29-23 and 3-29-24, americium in stacks 3-29-28 and 3-29-29, and plutonium in stacks 3-29-20 and 3-29-28. Except for the values for stacks 3-29-28 and 3-29-29, most of the ratios for radon daughter products ^{210}Pb and ^{210}Po are reasonably close to one.

Table 3 summarizes the information about ratios for individual stacks and for particular radionuclides. Although most of the ratios can be said to differ significantly from unity, the means do show some tendencies. In particular, the grouped results for stacks whose old sampling point was upstream of the fan inlet have a mean ratio higher than the mean for stacks whose old

sampling point is downstream of the fan. The mean ratio for the grouped results for uranium, most of which are from a stack of the second type, is distinctly elevated (but not significantly).

Table 3. Summary of Ratios for Individual Stacks and Individual Radionuclides

Stack	Number of comparisons	Mean value	Standard deviation
	Summary for individual stacks		
-14	3	0.85	0.12
-15	1	0.80	Not applicable
-19	10	0.85	0.26
-20	6	1.68	1.09
-23	15	1.26	0.42
-24	11	1.26	0.46
-28[a]	5	1.68	0.24
-29	3	2.59	0.54
-32[a]	3	0.81	0.09
-33	4	0.96	0.61
Old before fan[b]	33	1.43	0.74
Both in stack[c]	28	1.07	0.40
	Summary for individual radionuclides		
^{241}Am	8	1.35	0.63
^{210}Pb	17	1.25	0.61
^{210}Po	13	1.04	0.25
^{238}Pu	9	1.23	0.68
^{239}Pu	6	1.33	1.30
^{234}U	4	1.59	0.24
^{235}U	2	1.54	0.69
^{238}U	2	1.51	0.02
All Uranium	8	1.56	0.31
All ^{210}Pb, ^{210}Po	30	1.16	0.49

[a] Ratios that were affected by maintenance operations were not included in the assessments of the mean and standard deviation.

[b] Summary for those stacks (–15. –20, –24, –28, –29, –32, and –33) in which the old sampling point was located upstream of the exhaust fan.

[c] Summary for those stacks in which the old sampling point was downstream of the fan but relatively close to the discharge point.

Findings and Recommendations

Specific Observation. When assembling the data to make these comparisons, the audit team discovered that the printed report from the contract laboratory that analyzed the 6-month composite samples did not include uncertainties for the analytical results. The uncertainties were included in the electronic files that were transmitted to LANL.

Concern About ^{238}Pu Particle Emissions

The IEER monitors expressed a concern (see Appendix C) that releases of large particles of ^{238}Pu, which has a high specific activity, could be a source of confusion that would affect response to increased emissions from a LANL facility. The comment cites a portion of the quality assurance plan (Section 5.5.6) that relates to increased emissions from LANSCE, which is not a source of ^{238}Pu. There is, however, a similar plan component, Section 5.4.6, that relates to increased releases from facilities that could release particles of ^{238}Pu. If an increased emission of ^{238}Pu were to occur, it would be detected when the gross alpha measurement of the air filter activity was performed at the LANL Health Physics Analytical Laboratory. The entire filter is counted directly so the gross alpha activity would detect a signal from all particles of ^{238}Pu collected on the filter.

We prepared a table of estimates of the radioactivity content of individual particles as part of our discussion of similar questions during the first audit (Aanenson et al. 1999). For a pure $^{238}PuO_2$ particle with an aerodynamic diameter of 10 μm, the estimated activity is 2800 pCi (104 Bq). In the Rad-NESHAP report to EPA for the year 2001 (LANL 2002), we found that releases from the CMR stack 28 were about 8×10^{-6} Ci (0.3 MBq) and the estimated dose for the associated offsite exposure point was 1.1×10^{-3} mrem (0.011 μSv). Thus a release of about 0.0073 Ci (of ^{238}Pu particles with a 1-μm aerodynamic diameter) [0.27 GBq] would produce a dose of 1 mrem (10 μSv) at that location. For simplicity, we ignored the fact that the dose per unit activity inhaled is lower for the larger particles and that the number required to produce a dose of 1 mrem (10 μSv) is larger, and we estimated that a release of at least 2.6×10^6 particles of pure $^{238}PuO_2$ would be required to produce such a dose. The nominal flow rate from that stack was about 40,000 $ft^3\ min^{-1}$ (1.9×10^5 L s^{-1}). An effluent air sample collected at 2 $ft^3\ min^{-1}$ (0.9 L s^{-1}) during the release would collect about 130 particles.

We note that the proposed scenario of releases of 10-μm particles is quite unlikely in practice. The discussion in the first audit report pointed out that releases from filtered exhaust are unlikely to consist of large particles. Filter penetration is most likely for particles with aerodynamic diameters between 0.1 and 0.5 μm. For the latter, the activity of ^{238}Pu in a pure oxide particle is 0.35 pCi (0.013 Bq), meaning that 8000 times more particles would be required to produce the same 1 mrem (10 μSv) dose. The number of particles sampled would be increased proportionally to approximately 1 million.

The fact that releases of 10^{-6} curies (3.7×10^4 Bq) of ^{238}Pu are routinely detectable demonstrates that the airborne particles are not of the size hypothesized in the IEER scenario. Larger particles could be present in the effluent together with much higher activity concentrations if both banks of high efficiency particulate air filters failed simultaneously.

NON-POINT SOURCE MONITORING EVALUATION

LANL has identified over 1500 potential sources for diffuse, or non-point, emissions within the boundaries of the LANL technical site. To meet the requirements of 40 CFR 61, Subpart H, releases from all sources that have the potential to release radionuclides must be kept below an annual dose limit of 10 mrem (0.1 mSv) to a maximally exposed individual. These releases include those from point sources as well as the 1500 diffuse sources onsite. Techniques for monitoring releases from point sources are specified in 40 CFR 61, Subpart H, but non-point sources are not explicitly addressed in the prescribed methods. Non-point sources include sources at LANL such as shallow land burials, surface impoundments, firing sites, unvented buildings, open burn sites, and storage tanks.

LANL evaluates dose from non-point sources using environmental monitoring. Assuming that the samplers are properly placed, LANL postulates that the environmental measurements should provide a comprehensive assessment of releases due to diffuse sources, as well as point source releases.

During this third audit, we reexamined LANL's use of environmental sampling to show compliance with the dose limit for diffuse sources. To evaluate the Laboratory's compliance with 40 CFR 61, Subpart H, and the viability of LANL's diffuse source monitoring program, the audit team carefully examined the AIRNET program once again, including a look at refinements and improvements that had been made since the first two audits.

The AIRNET System

AIRNET is a system of environmental air samplers located around the perimeter of LANL property and in other locations where monitoring the concentrations of radionuclides in air might be important. The AIRNET network has been in operation for over 20 years, long before LANL was legally bound to EPA and DOE requirements. The samplers are located between Laboratory facilities and potentially exposed members of the public or they encircle areas on the Laboratory property that have the potential to be major sources of diffuse emissions.

The compliance-related sampling network comprises only a small portion of the total AIRNET system. There are AIRNET stations located at other onsite locations as well as offsite locations that are not included in the compliance network. When the compliance sampler sites were being established, LANL tried to utilize existing AIRNET sites wherever possible to avoid duplication and added compliance sites as necessary to develop sufficient coverage.

Sample Collection and Handling

Summary of LANL Methodology

Each AIRNET sampler station collects filter samples of airborne particles and silica gel samples of water vapor, including tritium vapor, from ambient air. The filter housing and airflow equipment configuration were designed by the Laboratory from commercially available parts. The filter housing is a weather-tight, louvered design containing a particulate filter assembly, silica gel flow meter, vacuum pump, various connecting hoses, and a power supply circuit. The silica gel water vapor absorber is located outside of the sampler housing to better represent the

humidity in ambient air. The compliance sampler sites are equipped with Campbell data loggers to track the power supply to the station and alert the AIRNET laboratory within 10 minutes in the event of a power failure at a station. If a power failure is noted at a compliance sampler station, an AIRNET technician will go into the field and investigate the problem, fixing it immediately if possible, or collecting the appropriate equipment and returning to the field to resolve it. In the case of such a power failure, a note will be made in the AIRNET logbook (ESH-17-231, R0). Specifications for all parts of the sampling assembly are given in the sampling and analysis plan for AIRNET (ESH-17-AIRNET, R7).

Before placement in the field, filters and silica gel cartridges are prepared in the AIRNET laboratory (ESH-17-202; RRES-MAQ-204). The filter preparation involves cutting the filter paper using a tool to create filters with a uniform diameter. These filter papers are precut in a quantity to fill the filter heads for the upcoming 2-week sampling period. The filter paper is handled with tweezers on a clean surface to avoid contamination. The filter head assembly is cleaned of excess dust, the filter is inserted within the filter head assembly, and a plastic cap is placed over the filter to again protect it from contamination. Each filter head is designated to a specific sampler, and each assembly is placed on its designated sampler every other 2-week period.

The filter head design for the samplers was chosen to reduce the possibility of filter contamination when the filters were removed and transferred to the AIRNET laboratory. The filter head is removable in its entirety from the airflow system by a quick disconnect fitting. During sample change-out, the old filter head, already marked with the sampler number, is removed from the airflow system and covered with the cap from the new filter head. This new filter head, also appropriately labeled, is then placed on the airflow system. Capping the filter reduces filter contamination during the change-out and keeps the collected sample in place on the filter. The filter heads and silica gel cartridges are color coded to represent each 2-week period to reduce the potential for error by the field technician when identifying the old and new samples (ESH-17-202).

Silica gel containers are also prepared in the laboratory in the off-week during which samples are not collected. First, a can of silica gel material is baked in an oven at approximately 150°C for at least 2 days. This process removes any excess water from the silica gel, as such water would dilute the concentration of tritium collected in the sample. Under a fume hood, approximately 135 g of silica gel is placed into each sampler container. The container lid is replaced and the silica gel cartridge is then weighed on an electronic scale. This weight is automatically transferred electronically to the database.

Silica gels are safe from contamination if the gel beads are not exposed to moisture before being sealed in the casing. Once the silica gel is connected to the airflow system only moisture passing through the system during the 2-week sampling period will be collected by the gel. Gel weights have been selected based upon exposure duration and experience with these gels in the field. Silica gel cartridge holders include a mechanism that stops air flow and, thus, moisture flow, into the cartridge after it is removed from an airflow system. The change-out of silica gel cartridges proceeds similarly to the filter change-out. The importance of the color-coding system is even greater here, as there is no difference in appearance between the silica gels before and after placement in the field (RRES-MAQ-204).

During sampler change-out, important field information is entered into a palmtop computer. This information includes timer reading, portable calibrator filter flow rate, air flow rate

(downstream of the silica gel) before and after removal and replacement of the silica gel cartridge, and any other relevant information to describe conditions in the field that might affect sample readings, such as a power failure, a breaker that is off, or road construction activity. Since the last audit, a portable calibrator has been introduced to measure the particulate filter flow rate. The flow meters for the particulate filter heads are no longer used to record the flow rate because they were found to be inaccurate. Many of these have been removed from the system. The in-situ flow meter is still used to measure the silica gel cartridge flow rate. Additionally, a filter was inserted between the silica gel cartridge and the flow meter to remove silica dust since the last audit was completed. The MAQ Sampling and Analysis Plan for the Radiological Air Sampling Network (ESH-17-AIRNET, R7) needs to be updated to reflect the elimination of a Rotometer-type flow meter for assessing the particulate filter flow rate, and the installation of a filter to remove silica dust.

Once collected from the field, contaminated filters are removed from the filter head assembly using tweezers. All of this is done on a clean surface to minimize the potential for contamination. The filters are then placed in glassine envelopes that have been prelabeled to reflect the sampler number. During 2001, whole filters were face-front counted individually for gross alpha and gross beta, and counted in clumps for gross gamma by Wastren, Inc. in Grand Junction, Colorado. Wastren, Inc. retains these whole filters and at each quarter's end, and a representative from MAQ travels to the analytical laboratory and splits each filter in half, adding each filter half to a quarterly filter composite for that sampler. One half of the filter composite is digested and analyzed for isotopes of uranium and plutonium and ^{241}Am. The other half of the composite is maintained at Wastren for a period of time stipulated in the contract with Wastren Inc., in case it is needed for reanalysis.

The procedure for silica gel analysis had been revised since the second audit. A new software package was implemented in the AIRNET laboratory that connects the AIRNET database directly to the scale used to weigh the silica gels after removal from the field, eliminating the need for hand-entering the data. A second person rechecks all the silica gel weights. The silica gel is shipped directly to Paragon Analytics in Fort Collins, Colorado for processing and tritium analysis.

Silica gel cartridges are checked for leaks every 6 months. The leak checking process involves putting 10 pounds per square inch (0.7 kilograms per square centimeter) of pressure using helium gas on the silica gel cartridge. A sensitive helium detector is then used to check for any leaks. Leaks are repaired and documented in the AIRNET logbook.

In addition to the samples placed in the field, several quality assurance samples are prepared and used as a check for cross-contamination. For filters, these quality assurance checks include filter trip blanks that go out on the filter placement loops with the field technicians, and matrix blanks that stay in the AIRNET laboratory during the 2-week period. These help detect any contamination of filters in the field or in the laboratory. For the silica gels, 10-gram water samples representing tritium blanks are evaporated and absorbed onto silica gel and then sent to the analytical laboratory for distillation and analysis. This water is drawn from wells that sample the regional aquifer, where sensitive measurement techniques have been used to measure tritium concentrations that range from 1 to 2 pCi L^{-1} (0.037 to 0.074 Bq L^{-1}). These samples represent blanks for the AIRNET program, because the AIRNET detection limit for tritiated water is 500 pCi L^{-1} (18.5 Bq L^{-1}). Tritium-spiked water samples are prepared by another group at LANL. From these samples, 10-gram tritium spikes are evaporated and absorbed onto silica gel and then

sent to the analytical laboratory for distillation and analysis. In all, the filter program includes two different types of quality assurance samples, and the silica gel program includes three different types. The analytical laboratories also perform several quality assurance checks of their own to ensure that contamination is not introduced in their setting.

Evaluation of LANL Methodology

The audit team observed all aspects of the sample handling and preparation program described here. The techniques outlined in procedures were well followed in the laboratory. The audit team found the AIRNET sample collection process to be sound.

IEER raised an issue regarding the silica gel measurements during the second audit. Because of a change in silica gel methodologies, LANL revisited this issue during 2001. The results of that investigation are discussed in the "Data Validation and Verification" section.

The data loggers placed into the AIRNET compliance stations have considerably increased the completeness of the environmental data. The system has been further improved since the second audit because the data loggers transmit a message to the AIRNET laboratory within 10 minutes if the power supply is interrupted. Previously, the data loggers could continue to run for 2 days on an auxiliary power supply before transmitting a message to the AIRNET laboratory. The details about any occurring malfunction are recorded in the AIRNET logbook from the data loggers and transferred to the AIRNET database as part of the validation and verification process. The vacuum pumps are changed out and serviced every six months. The introduction of this active maintenance schedule has also improved the completeness of the environmental data.

Quality assurance of the AIRNET samples and field data is maintained through several avenues. The numerous quality control samples sent to the analytical laboratory are ample evidence to show that the laboratory measures values correctly and that the AIRNET facilities or personnel are not inadvertently introducing additional contamination onto the filters and silica gels. The data loggers have increased the completeness of the AIRNET data, reducing downtime of the samplers. Two AIRNET personnel go out on nearly every station change-out loop for safety reasons and to double check field procedures. The palmtop computers have built-in nominal values for the input parameters from the field, and alert personnel to any data points that are out of the typical range. The palmtops also print out a chain-of custody form, which the audit team observed to completely accompany each AIRNET sample from AIRNET laboratory to field and back, to shipping and to the analytical laboratory. It was obvious to the audit team that the AIRNET group works as a team and is dedicated to assuring the quality of data they are responsible for collecting. Constant improvements to the program are being made, mostly because of careful attention to detail and willingness to experiment and search for new and innovative ways to improve the program. Every quality assurance requirement is documented in AIRNET procedures. The audit team considers quality assurance as it relates to sample handling and preparation to be complete in both documentation and application.

An additional issue was raised by IEER regarding the potential for the presence of a single particle of ^{238}Pu on half a filter that would escape detection because only half of each filter is isotopically analyzed, and a large dose to a receptor could be missed. In spite of the fact that each filter is face-front counted for alpha particles and the probability of missing the signal during that analysis is small, we performed calculations to examine the dose impact of such a situation.

At a filter flow rate of 4 cubic feet per minute (1.9×10^{-3} m^3 s^{-1}), used in the AIRNET samplers, 163 cubic meters of air per day are sampled. This is seven times the average adult breathing rate of 23 cubic meters per day. The AIRNET samplers run for 2-week time periods. Conservatively, we assumed that the sampler collected one particle of ^{238}Pu per week. This would result in an average air concentration of 0.00245 pCi m^{-3} (9.1×10^{-5} Bq m^{-3}). At an average adult breathing rate, 0.39 pCi (0.014 Bq) of ^{238}Pu activity would be inhaled, which would result in a dose of only 0.0234 mrem (0.234 μSv). Additionally, the chance of an adult even inhaling this particle that the AIRNET system detected is only 14% (23 cubic meters per day / 163 cubic meters per day = 0.14). The IEER concern about a single particle of ^{238}Pu being missed and resulting in a significant dose seems to be unfounded.

Findings and Recommendations

Specific Observation. Leak-checking on the silica gel cartridges occurs with a frequency that concerned CCNS, and the audit team agrees with their suggested increase in frequency. Bi-annual leak checks seem inadequate for assuring that no sample is lost during the intervening six months. The audit team recommends that MAQ consider increasing the frequency of leak checks to once every quarter.

Data Validation and Verification

Data validation and verification is the process of reviewing the data to ensure that information has been transferred correctly to electronic form and to identify any issues that might make a data point unusable. There are four types of data that go through the validation and verification process: field data, analytical chemistry data, meteorological data, and concentration data.

Summary of LANL Methodology

The process of data validation and verification includes checking the data to make sure that transfer from one form to another is complete and accurate and initially separating the data into three categories: accepted, qualified, or rejected. The validation and verification process then involves evaluating the data included in each of these three categories to ensure that they do, in fact, belong in that category or to provide evidence to suggest that a qualified or rejected point may actually be useful.

First, field data are verified and validated. Data that go through field validation and verification include collection date and time, sampler number, timer reading, beginning and ending filter flow rates, beginning and ending silica gel flow rates, beginning and ending silica gel masses, and comments. Field data are transferred from the palmtop computers to the AIRNET database, and then 100% of the data are checked for accuracy. Any additional comments may be added at this time.

There are certain nominal ranges within which field data are expected to fall. If the data do not fall within these ranges, this could be evidence to suggest either qualifying or rejecting that data point. For instance, if a pump fails, the filter and silica gel data would be qualified, and

further review would be required to determine if those data points were valid and could be used in the annual calculations (ESH-17-216, R6).

The analytical chemistry data first arrive via email as an electronic data deliverable approximately one week before the hard copy is received. The hard copy is the legal copy. Once the final analytical chemistry data are received from the analytical laboratory, these data are electronically transferred to the AIRNET database. The biweekly data are 10% verified for accurate transfer, and the quarterly data are 100% verified. The quarterly data contain the isotopic information and are considered by the analytical chemistry coordinator to be too important not to verify at 100%, even though only 10% is required by LANL procedures. The analytical chemistry point person also checks to ensure that the chain of custody form is complete.

A number of calculations are made at this stage of the data analysis. Concentrations of alpha and beta are calculated for biweekly data packages, as well as run time and sample completeness. An analysis of the success of the analytical laboratory with the quality assurance samples sent with the data package is also made at this time. For quarterly data packages, isotopic concentrations are calculated.

The analytical data also go through the qualification, acceptance, or rejection process. Data are qualified or rejected based on analysis of expected ranges given historical data and professional knowledge and judgment. In addition, concentration values that are above the action or investigation levels are tagged. A memo documenting this entire analysis is included in the AIRNET data package and sent to the AIRNET managers and to the next step of the analysis. This analysis must be done within 3 weeks of receipt of the data package from the analytical laboratory (ESH-17-033).

One important function of data validation and verification from an analytical standpoint is ensuring that the analytical laboratory is meeting the data quality objectives set out in the statement of work. Audits of the analytical chemistry laboratories are done by RRES-MAQ annually. This is an important quality assurance step in data validation and verification.

The meteorological data are used for calculating 15-minute absolute humidity concentrations. These 15-minute concentrations are averaged over the 2 weeks that span the sampling period. These data are validated to ensure that they are within expected humidity levels for that time of year. An AIRNET staff member checks these data and would question the meteorologists if something appeared to be out of line.

Finally, air concentration data undergo a validation and verification along with a health physics review and check. During this process, any data points that exceed the action levels are assessed, and qualified and rejected data points are reviewed for possible acceptance. This process must be completed within 30 days of the completion of the field and analytical chemistry data verification.

Occasionally, an AIRNET station will suffer some partial loss of data during the 2-week sampling period. These cases may result from a power loss, a breaker being thrown, a pump failure, etc. In these cases, the AIRNET data are usually qualified and require review during this part of data validation and verification. Because some data from during the 2-week period is better than no data during the 2-week period, AIRNET staff research the data to see if they might be useable. For filters, beta concentration can be a good indicator. Beta concentrations vary seasonally, but at any given time of the year, they are quite consistent across stations. If the beta concentration is consistent with other stations, the data from that station can still be used (ESH-17-208, R2).

Concentrations that exceed action levels are also reviewed during this process to determine whether they are consistent with historical measurements, contemporaneous measurements, or known releases. There are two types of action levels: investigation and alert. Investigation levels are based on historical data, and alert levels are calculated as the concentration that would result in a dose of 0.1 mrem (1 μSv). In every case, the technical memo that is drafted as a result of this process contains information about values that exceed an action level and the cause. Derivation of investigation and action levels is documented in ESH-17-201.

Run-time and completeness are evaluated annually. The FFCA requires 95% run-time of stations and 80% completeness of samples throughout the year. The completeness requirement corresponds to no more than five individual samples being lost, rejected, or not analyzed during a calendar year.

Quality assurance for the verification and validation process, in addition to auditing the analytical laboratories, involves having another individual check and sign his/her approval of each step outlined here. This also applies for review of the database, in which case the database tracks when a review was completed and which individual completed it.

Evaluation of LANL Methodology

We found the data validation and verification process to be well managed and complete. Each individual is very familiar with the role they play in the validation and verification process, and we are confident that data are handled appropriately.

We reviewed approximately half of the data packages for 2001. We found chain of custody to be complete and promptness of data review to be satisfactory. When data were qualified or rejected, the reviewers did an excellent job of evaluating these data and including data points that may have otherwise been discarded. We also found the methodologies used to accept qualified or rejected data points to be sound.

The memos that summarize the data are very useful, but could be improved. In particular, the tables that summarize the data are not generated in a consistent manner. For example, this was done manually in the isotopic air concentration review for the second quarter of 2001 (AIRNET 01Q2 Isotopic Air Concentration Review, October 22, 2001) and resulted in an error in the summary table. Because all the data are contained in the AIRNET database, such tables should be generated automatically. It was also noted that the U-234/U-238 isotope ratio sheet which lists the site number, air concentration for each isotope, and the ratio, is generated automatically, however, it does not include details regarding the standard deviation difference. Inclusion of this information in the table would allow the significant sites to be identified directly.

In reviewing the analytical data packages for 2001, we noted an anomaly in the results for the laboratory matrix spikes for U-238. The percent recovery of U-238 was consistently above the defined upper limit of 110% for the analysis. LANL had noted this trend and reported it to the laboratory, but was not unduly alarmed because it would result in an overprediction of concentration and dose. However, this trend had continued for more than one year. When we inquired, the analytical laboratory rapidly identified the source of the problem. The spike solution the analytical laboratory used to prepare the laboratory matrix spike samples included inorganic beryllium, in addition to the uranium isotopes (U-234, U-235, U-238). The inorganic beryllium was found to contain a wide variety of elements including depleted uranium, which impacted the U-238 concentration in the spike. The analytical laboratory was directed to revise the composition

of the spike samples to eliminate this interference. The bias in the laboratory matrix spikes had no impact on the U-238 concentrations measured for 2001. We commend LANL for resolving this issue.

Comparison of concentrations to action levels is an important part of the data validation and verification process. Occasionally, exceeding an investigation level can be related to a known release. Often the values are consistent with historical data, and they reflect some seasonal change for which the action levels do not account.

The audit team also reviewed implementation of quality assurance and discovered it to be complete in all cases.

One issue raised by IEER during the second audit related to how total water vapor collected in the atmosphere on silica gels is measured and how tritium concentration is calculated using this information. Silica gel naturally retains some small amount of water that even baking the gel cannot remove; therefore, IEER postulated that this water may dilute the tritium concentration. It was difficult to show such a trend in 1999 because of inconsistencies and errors in the silica gel cartridge weight data. Several factors resulted in LANL conducting a number of experiments to investigate this issue more closely (Eberhart 2002). First, the procedure for analyzing the tritium spike samples was changed in 2001, so that instead of submitting 10-gram water samples with known tritium concentrations to the analytical laboratory for analysis, the 10-gram tritium spikes were evaporated and absorbed onto silica gel and then sent to the analytical laboratory for distillation and analysis. The average tritium concentration recovered from the silica gel dropped to 61% as compared to 96% when the earlier procedure was used. Also a paper was published in 2000 regarding this issue (Rosson et al. 2000). LANL determined that the tritium samples are "diluted" by two factors: 1) bound water – after distillation, hydrogen including tritium remains bound within the gel matrix; and 2) isotopic fractionation – tritiated water has a higher boiling point than water. The dilution by water is equal to 3.6% of the silica gel weight. For a 135 g cartridge of silica gel, this is 4.86 g of water. Therefore, the tritium concentration for a 10 ml sample of water vapor should be a factor of 1.486 higher [(10+4.86)/10]. The tritium concentration is multiplied by 1.03 (3% increase) to correct for the isotopic fractionation. These corrections were subsequently applied to the tritium ambient air concentrations in the AIRNET database. We commend LANL for continuing to research this issue, and successfully identifying the biases associated with the tritium sample analysis procedure.

Findings and Recommendations

Specific Observation. We recommend that LANL review the procedure used to generate the tables that summarize the air concentration data. These tables should be generated automatically from the AIRNET database. Also, the table formats should be reviewed to ensure that the pertinent useful information is captured in the summary tables.

Sampler Siting Analysis

The sampler siting analysis documented in the FFCA was thoroughly reviewed in the first audit. We found the siting analysis to be unsatisfactory and recommended that further justification of the sampler locations be completed. Despite enhancements by LANL to the sampler siting analysis, we concluded that this remained a technical deficiency in the second

audit. In particular, we stated "additional criteria for sampler siting to the north of the laboratory are necessary to make the siting technically sound."

Summary of LANL Methodology

A complete description of LANL's sampler siting analysis can be found in the FFCA and the first audit report (Aanenson et al. 1999). The location of AIRNET stations along the northern boundary of LANL relative to diffuse-emission locations had historically been evaluated against a grid of artificial diffuse-emission sites instead of actual diffuse-emission sites. In 2000, LANL reexamined sampler siting locations based on actual potential release points (ESH-17:01-032). As a result, one new station (#68) was added at the entrance to Los Alamos airport to supplement the AIRNET stations currently in place to address diffuse emissions at TA 21. The methodology used consisted of four parts: 1) Identifying diffuse-emission sites of interest, 2) Identifying the AIRNET sites of interest, 3) Plotting the information on maps and overlaying meteorological 22.5-degree sector grids centered on the diffuse-emission sites, and 4) Determining if any applicable sector along the northern boundary is unmonitored for any diffuse source.

In 2001, LANL established a procedure to determine the need for new AIRNET stations as a result of new or modified activities at LANL (ESH-17-238, R0). This was the procedure that was applied when we reviewed the adequacy of the existing AIRNET station coverage.

Evaluation of LANL Methodology

During the audit, the audit team, CCNS, and IEER reviewed the completeness of the existing AIRNET stations in relation to diffuse-emission locations across the LANL site with LANL staff. This was done using a map of the site, overlaying the 16-sector grid, identifying the distance to the MEI location, and determining if the AIRNET coverage was adequate.

In 2001, LANL established a procedure to determine the need for new AIRNET stations as a result of new or modified activities at LANL (ESH-17-238, R0). This was the procedure that was applied when we reviewed the adequacy of the existing AIRNET station coverage. The audit team commends LANL for reexamining and updating the AIRNET station sampling procedure, but we suggest one modification, as described below.

Findings and Recommendations

Specific Observation. Based on a concern raised by IEER and CCNS (Appendix C), the audit team reexamined the sampler siting evaluation done by LANL for placement of samplers at the North Mesa residences. The procedure developed by LANL used annual average wind speed and source term to evaluate the sampler coverage for diffuse sources. The audit team recommends that AIRNET station siting for diffuse sources should be evaluated considering the potential for resuspension.

The IEER monitors expressed concern regarding the diffuse source at TA 21. The only potential diffuse source at this location is MDA-U. We contacted LANL and learned that MDA-U was remediated in the 1980s, and that the majority of the source term was removed at that time. The Environmental Restoration Group plans to apply to the state, requesting that no further work

be done. If this application is successful, no further work will take place at MDA-U, and this location will no longer be identified as a potential diffuse source.

In spite of the limited potential for TA 21 and MDA-U to be a diffuse source, we recommend that LANL reevaluate the sampler siting with respect to the North Mesa residences and the MDA-U diffuse source using a year's worth of data where wind speed is above some threshold value at which resuspension of material from diffuse sources would become a factor. Using this situation as a test case for an evaluation of sampler necessity would provide both resolution of this issue raised by IEER and CCNS and a revised methodology for conducting New Source Reviews. Based on previous work done by the audit team on resuspension at LANL, we would suggest that an appropriate wind speed above which an evaluation would be instructive might be 10 mph (4.5 m s^{-1}). This wind speed should be sustained for some period of time (probably several hours) and no measurable precipitation should have occurred in the preceding 24 hours. These conditions would be most favorable for resuspension of material. This revision of the procedure for AIRNET sampler siting would then be appropriate for all New Source Reviews that might require AIRNET coverage.

DOSE ASSESSMENT EVALUATION

40 CFR 61, Subpart H, contains relatively few specifications for performing the dose assessments required for demonstrating compliance. For example, EPA-approved computer codes must be used. 40 CFR 61.93 specifically allows the use of CAP-88[f], AIRDOS-PC, and COMPLY. The EPA has also granted approval for the computer codes CAP88-PC and MICROAIRDOS. In addition, other computer codes or procedures could be used with prior approval by the EPA.

40 CFR 61, Subpart H, requires that doses are to be estimated at offsite points where there is a residence, school, business, or office. The highest dose to a member of the public at these locations is used to demonstrate compliance with the 10 mrem yr^{-1} (0.1 mSv yr^{-1}) standard contained in § 61.92. This person is referred to as the site-wide MEI. It should be noted that 40 CFR 61, Subpart H, is not an unrestricted area standard. This means that doses are to be estimated at fixed locations where members of the public are actually located and doses do not have to be estimated at locations such as roads to which members of the public merely have access for short periods of time. Short-term, episodic releases must be included as part of the annual emission estimates and modeled using an EPA-approved computer code such as CAP-88 and annual average meteorological conditions.

40 CFR 61.94 also contains reporting requirements. For example, the distances to the nearest residence, school, business, or office and the distances to the nearest farms producing vegetables, milk, and meat are to be included in the annual report submitted to the EPA. All user-supplied input data and the source of these data also are to be included in the annual report.

Due to limitations on resources for the third audit, our examination of the dose calculation methodology was not as exhaustive as it was for the first two audits. We conducted only a data validation and verification exercise on the dose calculations and a comparison of values between the CAP-88 input files and the Rad-NESHAP annual report for this audit.

Dose Calculation Validation and Verification

To verify LANL's use of CAP-88 to conduct the dose calculations, we obtained the input and output files for the CAP-88 runs for 2001 and confirmed the calculation of dose by running the files ourselves. The average absolute value of the percent difference was 0.13%, which shows extremely good agreement. See Table 4 for the details. The input files were very clean and the audit team found no deficiencies in the runs.

[f] Throughout this section, the term CAP-88 is used to refer to the mainframe version of the computer code CAP-88 used by LANL.

Table 4. Comparison of Audit Team CAP-88 Run Results to LANL Run Results

Release Point	LANL Dose (mrem yr^{-1})[a]	CAP88-PC Dose (mrem yr^{-1})	Percent Difference[b]
03002914	8.55×10^{-8}	8.56×10^{-8}	0.12%
03002915	1.11×10^{-7}	1.11×10^{-7}	0.00%
03002919	8.62×10^{-6}	8.62×10^{-6}	0.00%
03002920	1.27×10^{-6}	1.27×10^{-6}	0.00%
03002923	2.89×10^{-5}	2.90×10^{-5}	0.35%
03002924	1.92×10^{-5}	1.92×10^{-5}	0.00%
03002928	1.35×10^{-4}	1.36×10^{-4}	0.74%
03002929	3.49×10^{-7}	3.49×10^{-7}	0.00%
03002932	1.41×10^{-7}	1.41×10^{-7}	0.00%
03002933	6.18×10^{-8}	6.18×10^{-8}	0.00%
03002937	4.80×10^{-9}	4.81×10^{-9}	0.21%
03002945	4.69×10^{-7}	4.62×10^{-7}	-1.49%
03002946	1.83×10^{-7}	1.83×10^{-7}	0.00%
03010222	1.66×10^{-7}	1.66×10^{-7}	0.00%
16020504	4.13×10^{-2}	4.13×10^{-2}	0.00%
18000001	1.64×10^{-5}	1.64×10^{-5}	0.00%
21015505	1.43×10^{-3}	1.43×10^{-3}	0.00%
21020901	1.05×10^{-2}	1.05×10^{-2}	0.00%
33008606	1.29×10^{-3}	1.29×10^{-3}	0.00%
41000417	4.51×10^{-3}	4.51×10^{-3}	0.00%
50000102	1.01×10^{-6}	1.02×10^{-6}	0.99%
50006903	1.25×10^{-8}	1.25×10^{-8}	0.00%
55000415	3.95×10^{-6}	3.95×10^{-6}	0.00%
55000416	5.02×10^{-5}	5.03×10^{-5}	0.20%
53000303 May	1.61×10^{-5}	1.61×10^{-5}	0.00%
53000303 June	4.17×10^{-5}	4.18×10^{-5}	0.24%
53000303 July	6.02×10^{-5}	6.01×10^{-5}	-0.17%
53000303 August	3.40×10^{-5}	3.40×10^{-5}	0.00%
53000303 September	3.68×10^{-5}	3.68×10^{-5}	0.00%
53000303 October	2.68×10^{-5}	2.67×10^{-5}	-0.37%
53000303 November	7.60×10^{-5}	7.60×10^{-5}	0.00%
53000303 December	4.26×10^{-5}	4.26×10^{-5}	0.00%
53000303 PVAP	3.26×10^{-5}	3.26×10^{-5}	0.00%
53000702 May	2.76×10^{-3}	2.77×10^{-3}	0.36%
53000702 June	8.28×10^{-2}	8.28×10^{-2}	0.00%
53000702 July	1.39×10^{-1}	1.39×10^{-1}	0.00%
53000702 August	4.68×10^{-2}	4.68×10^{-2}	0.00%
53000702 September	1.06×10^{-1}	1.06×10^{-1}	0.00%
53000702 October	2.26×10^{-1}	2.25×10^{-1}	-0.44%
53000702 November	4.92×10^{-1}	4.92×10^{-1}	0.00%
53000702 December	3.10×10^{-1}	3.10×10^{-1}	0.00%
53000702 PVAP	7.35×10^{-4}	7.35×10^{-4}	0.00%

530003SY	4.95×10^{-3}	4.95×10^{-3}	0.00%
5300071L	6.73×10^{-2}	6.72×10^{-2}	-0.15%

[a]Divide each value in the table by 100 to get values in units of mSv yr^{-1}
[b]Average percent difference = 0.13% (absolute value).

Rad-NESHAP Annual Report

Summary of LANL Methodology

Each year, LANL is required to report the dose to a site-wide MEI via all pathways due to releases to air. The formal reporting of this dose is in the Rad-NESHAP report. This report must also contain

- The results of monitoring and dose calculations
- The name and location of the facility
- A list of radioactive materials used at the facility
- A description of the handling and processing that the radioactive materials undergo at the facility
- A list of the stacks or vents or other points where radioactive materials are released to the atmosphere
- A description of the effluent controls that are used on each stack, vent, or other release point and an estimate of the efficiency of each control device
- Distances from the points of release to the nearest residence, school, business, or office and the nearest farms producing vegetables, milk, and meat
- The values of all other user-supplied input parameters for the computer models and the source of these data
- A brief description of all construction and modifications that were completed in the calendar year.

According to the 2001 report, the effective dose equivalent for the site-wide MEI located at East Gate was 1.84 mrem (18.4 μSv) (LANL 2002).

Evaluation of LANL Methodology

We reviewed the annual report to confirm that the values matched those shown in the input files for the dose calculations. The following differences were discovered.

1. Table 3 in the Annual Report says that the exit velocity for 21015505 is 8.72 m/s, but the CAP-88 run used 9.41 m/s.
2. Table 5 in the Annual Report says that the distance to the receptor for 03002933 is 5,965 m, but the CAP-88 run used 5,995 m.
3. Table 3 in the Annual Report lists the diameter of 53000303 as 0.90 m. This agrees with the annual PVAP CAP-88 run for 53000303. However, the May through December CAP-88 monthly runs used a diameter of 0.91 m.
4. The PVAP and non-CAP-88 radionuclide runs for 53000303 and 53000702 used a full year's worth of atmospheric data (01/2001-01/2002), but the fugitive emissions from

530003SY and 5300071L used atmospheric data from 05/2001 through 12/2001. Shouldn't these use data over the same time period?

5. The monthly runs for 53000303 used an exit velocity of 11.2-11.4 m/s. However, the annual PVAP run used a velocity of 11.72 m/s. Why would the monthly velocities be lower than the annual velocity, when the emissions were presumably over the same time period?

These inconsistencies were shared with LANL, and explanations were given for the problems as follows, with the impact of the errors shown in Table 5:

1. The correct value was used in the CAP-88 run, the value in the report table is incorrect.
2. The correct value should be 5965, the value in the CAP-88 run is incorrect.
3. The correct value is 0.90, the value of 0.91 in the CAP-88 runs is incorrect.
4. No, we wrote into procedure 501 this is how we would model these emissions.
5. There was an error in calculating the annual average flow rate used in the comparison CAP-88 run. For LANSCE, the individual monthly runs are the official runs used for compliance purposes; the sum of these is the annual dose. These runs use the most recently measured flow measurement data, for the ventilation configuration that was in use during operations. That's where the 11.2 to 11.4 m/s values come from, which are correct for the CAP-88 runs. The annual LANSCE sum is merely a comparison to see if the annual number corresponds with the sum of the monthly emissions. The stack flow in this case is supposed to be an average value for the year, in the "operations" configuration. It appears that the average value used in this case included configurations which were not actual "beam operation" configurations. These configurations can have much higher flow rates, due to the operation of supplemental fan systems that may not operate during beam activities. The average value used in the CAP-88 annual comparison run did not reflect beam-operation ventilation, but the higher flow rates that could be used during extended maintenance outages.

Table 5. Impact of Errors on CAP-88 Dose

Release Point	LANL Dose (mrem/yr)	Corrected Dose (mrem/yr)	Percent Difference	Corrected Value
03002933	6.18×10^{-8}	6.22×10^{-8}	0.65	Receptor distance = 5,965 m
53000303 May	1.61×10^{-5}	1.61×10^{-5}	0.00	Stack diameter = 0.90 m
53000303 June	4.17×10^{-5}	4.18×10^{-5}	0.24	Stack diameter = 0.90 m
53000303 July	6.02×10^{-5}	6.02×10^{-5}	0.00	Stack diameter = 0.90 m
53000303 August	3.40×10^{-5}	3.41×10^{-5}	0.29	Stack diameter = 0.90 m
53000303 September	3.68×10^{-5}	3.69×10^{-5}	0.27	Stack diameter = 0.90 m
53000303 October	2.68×10^{-5}	2.68×10^{-5}	0.00	Stack diameter = 0.90 m
53000303 November	7.60×10^{-5}	7.62×10^{-5}	0.26	Stack diameter = 0.90 m
53000303 December	4.26×10^{-5}	4.27×10^{-5}	0.23	Stack diameter = 0.90 m

Findings and Recommendations

Specific Observation. The errors in transcribing data into the CAP-88 input files further demonstrates a general need for increased review with regard to the annual report and the dose

calculation input files. While these errors are not significant and transcriptional in nature, their existence stresses the importance of improved quality control checks.

COMPLEX TERRAIN MODELING COMPARISONS

The issue of complex terrain modeling was raised in the first two audits, and again during this third audit. The question from IEER involves the applicability of a Gaussian plume model for a region with complex terrain such as LANL.

As a means of exploring this issue during the second audit, we calculated the ratio of the air concentration (χ) divided by the source term (Q) for CAP-88 and the complex terrain model, CALPUFF. The ratio of concentration divided by source term is referred to as the Chi over Q (χ/Q). The CALPUFF model explicitly treats spatial variability in terrain and meteorological conditions. The comparison focused on annual emissions from Site D (facility stack 3002919) in a 12.42 km × 8.92 km model domain using meteorological data from 1999 for the CALPUFF simulation and a 5-year meteorological data set for the CAP-88 simulation (Figure 2). A limitation to this methodology was that CALPUFF χ/Q values were obtained from the computation node nearest the CAP-88 receptor location. The CALPUFF computational grid had a spacing of 175 m between grid nodes, and resulted in a potential error of 175 m/2 = 87.5 m between the location of the CAP-88 χ/Q and that of CALPUFF. As part of the third audit investigation, we corrected this inaccuracy and reevaluated the results along with an evaluation of terrain effects on estimated concentrations.

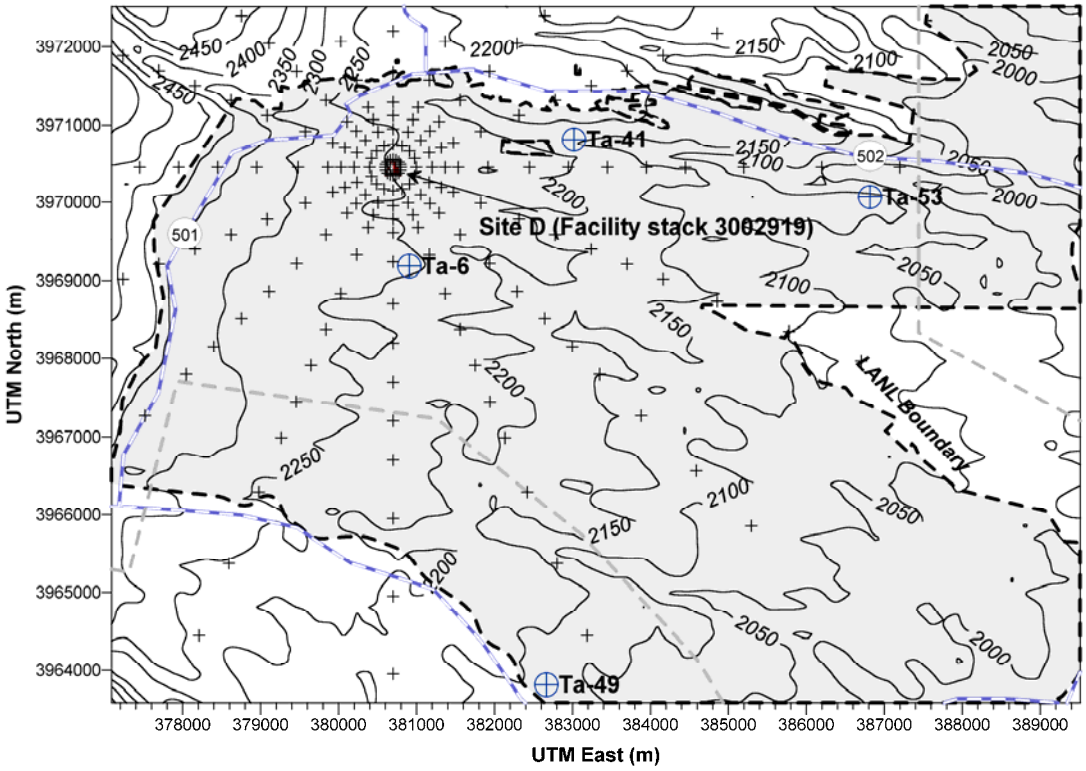

Figure 2. CALPUFF modeling domain showing location of Site D (Stack 3002919), meteorological towers (TA 6, TA 41, TA 49, and TA 53), terrain elevations (in meters), and CAP-88 receptors (shown by crosshairs).

Annual average CALPUFF χ/Q values were generally lower than those calculated with CAP-88 (Figure 3). Higher χ/Q values indicate less plume spreading and, consequently, higher pollutant concentrations. On average, CAP-88 χ/Q values were a factor of 1.85 higher than those calculated with CALPUFF. Approximately 9% (16 out of 179 values) of the CALPUFF/CAP-88 χ/Q ratios were greater than one, and of those 16 ratios, only seven were greater than two. A CALPUFF/CAP-88 χ/Q ratio >1 indicates the CALPUFF χ/Q is higher than the CAP-88 value. Twelve of CALPUFF/CAP-88 χ/Q ratios greater than one were 100 m from the source, two were 300 m from the source, and the remaining two were 3.75 km from the source. However, CALPUFF/CAP-88 χ/Q's ratios at the 3.75 km distance were ≤1.02, which means the χ/Q values from both codes were essentially the same.

Differences between CAP-88 and CALPUFF χ/Q values as a function of distance and direction from the source are shown in Figures 4 and 5. The terrain elevation has been superimposed on these graphs so that effects of terrain can be visualized. Figure 4 shows the south to north cross section of the annual-average χ/Q values. Note that the CAP-88 curve exhibits plume lofting over the 100-m receptor while the CALPUFF curve does not. Undulations in the CALPUFF curve are noted to occur at locations where the receptor was in the bottom of a canyon. Most notably, the receptors near Universal Transverse Mercator (UTM) coordinates 3970000 and 3972000 exhibit a drop in the χ/Q value followed by a rise in χ/Q value at the next downwind receptor. These undulations are suspected to be terrain induced. Both models have substantially higher χ/Q values north of the stack compared to south presumably due to the predominance of winds from the south.

The west to east cross section (Figure 5) depicts a slightly different pattern. There are few deep canyons that cut the cross section and the terrain slopes from west to east. Chi over Q values decrease sharply west of the stack. This sharp decline in χ/Q values appear to be a result of the meteorological conditions and not the influence of terrain, because both models exhibit similar behavior. East of the stack, concentrations drop-off slowly with distance for both models.

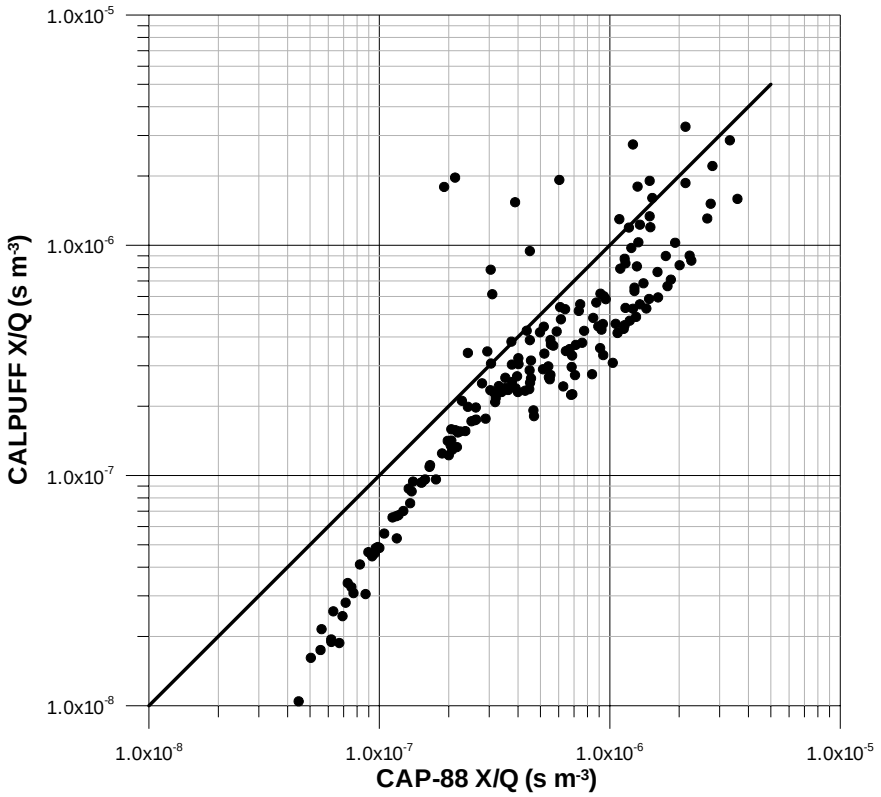

Figure 3. Annual average CAP-88 χ/Q values versus CALPUFF annual average χ/Q values for 1999 from a source located at Site D (Facility 3002919). Site D stack parameters are as follows: Stack height = 15.88 m, stack exit velocity = 25.9 m s^{-1}, stack diameter = 1.07 m, and stack exit temperature = 282 K.

One feature that is absent from the CALPUFF χ/Q values is the effect of plume lofting near the source. Originally, it was thought that the CALPUFF grid may have been too coarse to resolve plume lofting. Further investigation revealed that plume lofting occurs in the CALPUFF model for short-term averages (Figure 6), but is apparently absent when annual averages are considered. Therefore, grid resolution may not be as important as originally thought in terms of resolving plume lofting. One important distinction between the two models is that in CALPUFF, the plume may double back on itself. That is, if the wind direction shifts about 180 degrees, then the material emitted in the previous hour can advect toward the source, and thereby impact receptors near the source. It was not verified that the higher CALPUFF χ/Q values near the source were due to the plume doubling back on itself and it was beyond the scope of this exercise to investigate it any further. Nevertheless, these effects are incorporated into the CALPUFF χ/Q values. CALPUFF χ/Q values that significantly exceed those of CAP-88 are limited to receptors close to the source (<300 m). The nearest potential public receptor was estimated to be 600 m to the northwest of the Site D stack on State Highway 501.

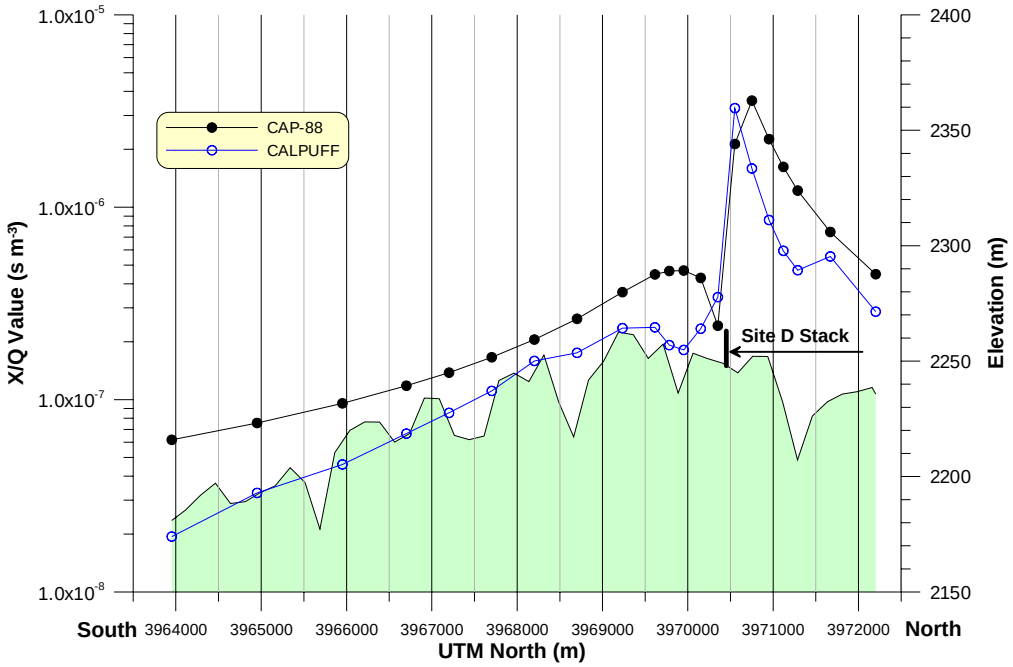

Figure 4. South-to-north cross section of terrain and annual average χ/Q values for CALPUFF and CAP-88. Terrain is represented by the shaded area.

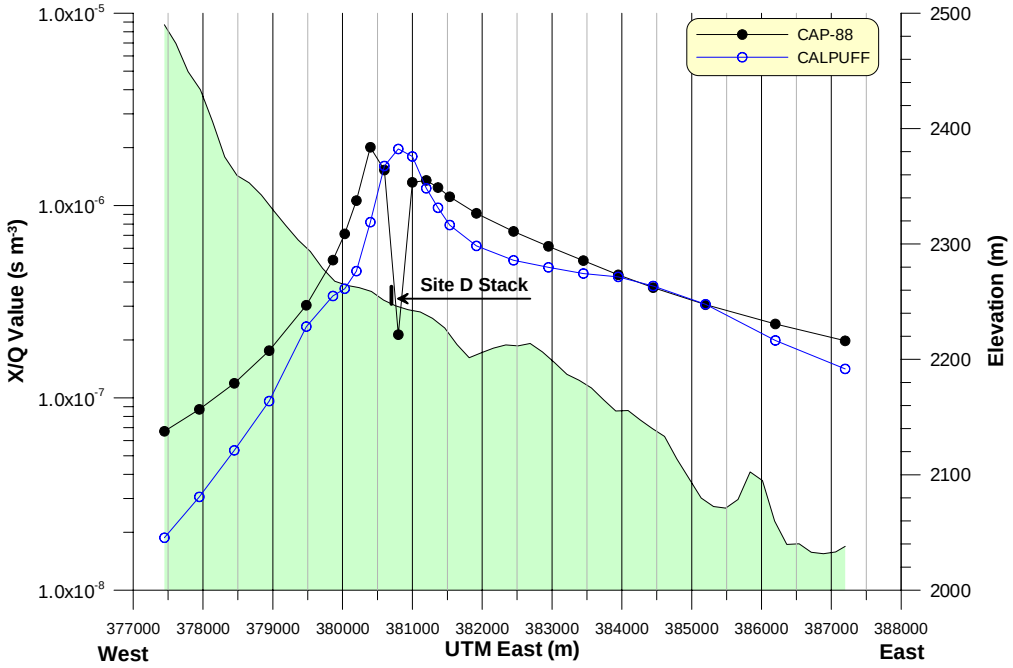

Figure 5. West-to-east cross section of terrain and annual average χ/Q values for CALPUFF and CAP-88. Terrain is represented by the shaded area.

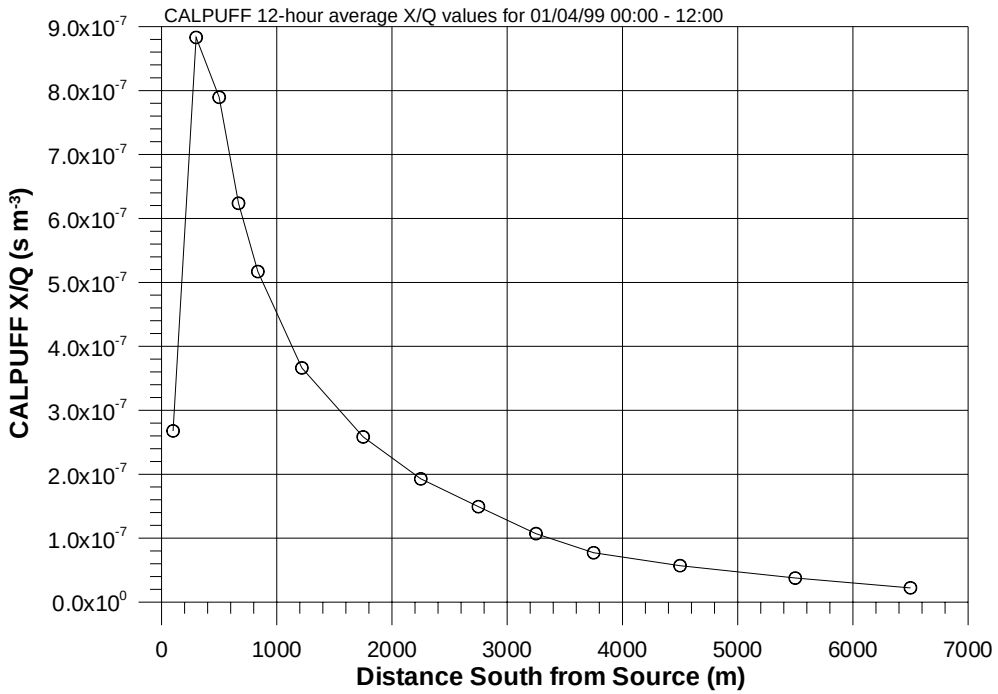

Figure 6. Twelve-hour average CALPUFF χ/Q values as a function of distance south from the Site D stack.

Overall, χ/Q values calculated by CAP-88 provide conservative estimates of dispersion at distances >300 m from the stack for annual average emissions. For some close-in receptors (<300 m), CALPUFF calculated higher dispersion factors. However, the nearest public access is estimated to be ~600 m from the stack, and therefore, CAP-88 would still provide a conservative estimate of annual-average dispersion at this distance.

An additional concern related to air dispersion modeling raised by IEER is that, in many cases, emission rates from LANL facilities are not uniform and vary with time. This may be particularly true for diffuse sources where resuspension is the major release mechanism. In such cases, doses may be underestimated because short-term dispersion estimates typically result in higher concentrations at some receptor locations compared to long-term averages. To evaluate these differences, we compiled output from CALPUFF for the maximum 1-hour and 24-hour average χ/Q values at each of the CAP-88 receptor points. The output CALPUFF χ/Q values represent the maximum χ/Q for the stated averaging period (1-hour or 24-hour) at CAP-88 receptor locations for the 1-year period (1999) simulated. The dose received by a hypothetical receptor is proportional to the amount of activity inhaled and can be calculated by

$$I = TIC \times BR, \qquad TIC = \chi/Q \times Q_T \tag{1}$$

where

I = the total radioactivity inhaled during the release period (Ci)
TIC = the time-integrated concentration (Ci-s m^{-3})
BR = the breathing rate (m^3 s^{-1})
χ/Q = the average concentration during the release period divided by the release rate (s m^{-3})

Q_T = the total activity released during the release period (Ci).

For demonstration purposes, we assumed the person remained stationary for the release duration and the total annual release quantity (Q_T) was 1 Ci (3.7×10^{10} Bq). We evaluated differences between radioactivity inhaled from annual-average and short-term releases by taking the ratio of I calculated for various release scenarios to I calculated for an annual-average release using CAP-88. Assuming the same breathing rate for annual-average and short-term releases, we have

$$R = \frac{I}{I_{AA}} = \frac{(\chi/Q_i)Q_{Ti}\,BR}{(\chi/Q_{AA})Q_{TAA}\,BR} = \frac{(\chi/Q)Q_T}{(\chi/Q_{AA})Q_{TAA}} \quad (2)$$

where

R = the ratio of I_i/I_{AA}
I_{AA} = radioactivity inhaled for annual average conditions calculated by CAP-88 (Ci)
I = radioactivity inhaled for a 1-hour or 24-hour release calculated with CALPUFF (Ci)
χ/Q = maximum 1-hour or 24-hour average concentration during the simulation year at a given CAP-88 receptor divided by the annual-average release rate (s m^{-3})
χ/Q_{AA} = annual average concentration divided by the annual average release rate calculated with CAP-88(s m^{-3})
Q_{Ti} = total release quantity for a 1-hour or 24-hour release period (Ci)
Q_{TAA} = total annual release quantity (Ci).

Three scenarios were considered:

1. The entire annual release quantity was released in a 1-hour and 24-hour period
2. Ten percent of the annual release quantity was released in 1-hour and 24-hour period
3. One percent of the annual release quantity was released in 1-hour and 24-hour period.

The first scenario was not considered credible under normal operating conditions, and should be considered only in an accident analysis. The ratio R was calculated for each of the 179 CALPUFF-CAP-88 receptor pairs were summarized in terms of the minimum, maximum, and average value of R (Table 6). Results of the calculation indicate that if 1% of the entire annual release quantity were emitted in a one-hour period, then, on average, doses would be little different from the annual-average dose estimate. However, at some receptor locations, doses could be a factor of 9.23 higher than the annual average dose estimate. Assuming 1% of the annual release quantity was released during a 24-hour period, then the annual average estimate would provide a bounding estimate of the dose.

Table 6. Results of Ratio of I to I_{AA} for the Three Release Scenarios and 1-hour and 24-hour Release Duration

	1-hour release duration			24-hour release duration		
Percent of annual release quantity	100%	10%	1%	100%	10%	1%
Average R	111.68	11.17	1.12	9.12	0.91	0.09
Maximum R	923.46	92.35	9.23	79.82	7.98	0.80
Minimum R	13.63	1.36	0.14	1.60	0.16	0.02

The data in Table 6 are by no means definitive, but can be used as a guide for future analysis. For example, if the release during any 24-hour period exceeded 1% of the annual release quantity, then a separate dose calculation may be needed for that particular release. As an additional example of how this methodology could be implemented, we also calculated R values for a 168-hour (1-week) release duration (Table 7). The maximum value of R multiplied by the annual dose would provide a crude (but bounding) estimate of the potential increase in doses due to short-term release rates that exceed the annual average release rate. For example, the stack examined in this application (Site D, facility stack 3002919) had an annual dose of 8.62×10^{-6} mrem yr^{-1} (8.62×10^{-8} mSv y^{-1}). If 100% of the annual release quantity were emitted in a 1-week period, the maximum possible dose would be $31.05 \times 8.62 \times 10^{-6}$ mrem $yr^{-1} = 2.68 \times 10^{-4}$ mrem yr^{-1} (2.68×10^{-6} mSv y^{-1}).

Table 7. Ratio of I_i to I_{AA} for a 168-hour Release

	Percent of annual release quantity					
	100%	75%	50%	25%	10%	5%
Average R	2.56	1.92	1.28	0.64	0.26	0.13
Maximum R	31.05	23.29	15.53	7.76	3.11	1.55
Minimum R	0.75	0.56	0.37	0.19	0.07	0.04

Values of R presented in Tables 6 and 7 were constructed for different facilities and different release scenarios and used to scale doses calculated by annual-average CAP-88 simulations. It was not necessary to use CALPUFF for these calculations since averaging time and not terrain complexity was at issue here. A straight-line Gaussian plume model was adequate for such calculations. Should doses exceed the specified limits, then additional investigation may be warranted. These investigations should consider the meteorology during the time of the release and the proximity of a potential receptor to the trajectory of the plume.

ISSUES PERIPHERAL TO THE SCOPE OF THE AUDIT

During the course of the audit, a number of issues were raised that were peripheral to the scope of the audit; that is, they are unrelated to LANL's demonstration of compliance. The audit team agreed to look at these issues and comment on them. These issues relate to the Neighborhood Environmental Watch Network (NEWNET), uncertainty in the dose calculations, the use of TLDs for environmental monitoring, and future operations.

The Neighborhood Environmental Watch Network

NEWNET is a system of environmental measurement stations located primarily in and around Los Alamos. The operation of NEWNET was funded by LANL in accordance with the Consent Decree, which expired September 30, 2002. NEWNET was originally funded as an environmental network at the Nevada Test Site as a means for the public to understand and appreciate environmental exposure from weapons testing. The goal of this network was principally educational in nature, but the network was also seen as a valuable source of raw and real-time environmental measurement data for members of the public. The NEWNET stations provide data on wind speed, wind direction, humidity, barometric pressure, and the external radiation exposure rate at the measurement location. These data are summarized as 15-minute averages and distributed via the Internet. Although NEWNET was developed at the Nevada Test Site for residents in that area, it was envisioned that the program could be implemented nationwide and worldwide as a tool to educate people about environmental radiation.

The NEWNET concept for the Nevada Test Site was made obsolete by the nuclear test ban treaty. The guiding philosophy and goal of NEWNET, however, were unchanged by this treaty, so the network was moved to the LANL area. Because LANL was already partially supporting the program, the move was both practical and relatively simple.

When NEWNET was originally developed, it was intended to provide real-time data on local meteorological conditions and environmental radiation fields. Ionization chambers were chosen as the measurement tool for environmental radiation. Ionization chambers measure charged particles created by gamma rays as they pass through the walls of the chamber and interact with the pressurized gas inside. The charged particles produce an electric signal proportional to the intensity of gamma-ray exposure, and that signal is recorded to enable an examination of temporal fluctuations in exposure rate.

At the time that NEWNET was moved to LANL, the primary source of offsite radiation exposure was the operation of the LANSCE facility. Short-lived radioactive gases released from LANSCE stacks are a source of external radiation exposure in nearby areas. The NEWNET radiation detectors are well suited to measure these radiation exposures. Another radionuclide of importance from a dose perspective at LANL is tritium, which emits only low-energy beta particles that are not detected by NEWNET's ionization chambers. Other radionuclides released from LANL, such as isotopes of plutonium and uranium, emit primarily alpha particles, to which the ionization chambers are also insensitive. Releases of these radionuclides are measured by the AIRNET samplers located around LANL.

As demonstrated by IEER during this audit, NEWNET is uniquely suited to provide a quality control check on the CAP-88 calculations done for releases from LANSCE. In IEER's July 3, 2002 memo to the audit team (Appendix C), they completed calculations which show

remarkable agreement between NEWNET measurements of dose at East Gate and CAP-88 calculations of the same dose. We thank IEER, and particularly Bernd Franke, for his detailed investigation and evaluation of NEWNET's ability to confirm dose calculations. We see this as an extremely useful application of NEWNET, and encourage LANL to continue to use it in this fashion.

During this audit, IEER again raised concerns (Appendix C) about the quality of the NEWNET data and the usefulness of the NEWNET web site. Based on our review of the program and interpretation of the IEER memos, we recommend that LANL continue to take steps to ensure the quality of web-posted data, state the capabilities and limitations of the NEWNET system for day-to-day operations and emergency release situations on the web site, resolve any apparent calibration problems, and investigate the most appropriate method for developing a representative characterization of background radioactivity levels. While not required for compliance purposes, we believe NEWNET to be a valuable tool for public use and participation in environmental monitoring. We suggest that LANL coordinate with CCNS and other citizens' groups to better develop the online description and discussion related to limitations and capabilities of the NEWNET system.

Uncertainties in Dose Calculations

Although the issue of uncertainty in CAP-88 dose calculations has been raised during this and past audits, we still consider this issue peripheral to the scope of the audit because estimates of uncertainty are not required by the regulation. As shown in the comparison of CAP-88 to CALPUFF earlier in this report, at typical receptor distances (>300 m), the CAP-88 model consistently provides conservative results (i.e. overestimates of dose). The calculations made by LANL using CAP-88 do not include estimates of uncertainty because the EPA has not deemed it necessary; however, in theory, the model does appear to compensate for not explicitly addressing uncertainty by producing results that are biased high.

What is lacking, however, should LANL pursue estimating uncertainties with doses calculated for compliance, is regulatory guidance on how uncertainties would be used. For example, what percentile value should be compared to the dose standard? A defensible argument could likely be made for the 50^{th}, the 95^{th}, or the 99^{th} percentile values. Without specific regulatory guidance on this issue, though, it is not possible to incorporate uncertainty into the estimation of dose for compliance purposes.

Despite the fact that we consider this issue peripheral to the question of compliance, we do believe it is an important issue that regulatory agencies should address. Therefore, as scientists, we intend to pursue this issue further with both national and international regulatory authorities.

Measurements of Direct Penetrating Radiation with Thermoluminescent Dosimeters

During the past year there has been continued interest about the monitoring of direct penetrating radiation around the LANL site. Direct penetrating radiation exposures are outside the scope of 40 CFR 61, Subpart H, however they remain of concern to stakeholders and the audit team tried to resolve as many questions as it could in this area.

One area of particular concern, related to TLD placement, was near the waste lagoons at TA 53. This area was toured by the audit team in June in an attempt to better understand the questions

being asked. Concerns were expressed by CCNS about worker protection and the use of the area TLD monitors and air sampling equipment for that purpose. Discussions revealed that the workers carried both individual TLD badges and personal air samplers.

Another area of discussion related to direct radiation monitoring along Pajarito Road, near TA 54. An aerial survey had reported a high gamma count rate at TA 54, presumably Material Disposal Area C, which was previously used for waste disposal (Rogers 1977). Color maps from the aerial survey report were distributed to the audit team by CCNS, and they showed a high gamma count rate for TA 54 East. Table 1 of the report indicated a large point source of ^{241}Am, but the figures did not confirm that. Unfortunately, without close examination of the gamma spectrum from the report itself, it was not possible to confirm ^{241}Am as the source of external radiation. Although this question about ^{241}Am is of interest to the audit team, it is outside the scope of our work and due to constraints of time and resources, we were not able to investigate this further.

The audit team offers the following comments and suggestions based upon its limited review of the relevant information:

1. Identification of a major gamma-ray source from the air does not necessarily imply a high exposure rate for persons on the ground in areas of public exposure; the cover over the top of the source may provide less shielding than the soil in the lateral path from the buried waste to a point of public exposure.
2. Occupancy time is an important component of dose assessment for sources that emit x- and gamma-rays at LANL. The majority of such sources are not close to areas that are occupied full time. This is considered in the LANL evaluation.
3. When documenting the criteria for location of TLDs it is very beneficial to include maps, for each area considered, that show the locations of (a) radiation sources, (b) the existing and proposed monitoring sites, and (c) nearby public access or transient exposure. Tabular summaries of data on which decisions are based are also helpful.
4. TLDs will not indicate the presence of airborne radioactivity at levels that are likely to be generated during decontamination and decommissioning activities. Similarly, TLD monitoring will not be an effective assessment tool in the event of a release of alpha-emitting radionuclides or tritium from LANL. If workers have individual badges, those provide a much better measure of the individual's dose than an area badge.
5. It seems that some cooperative field work to collect data on environmental exposure rates would be helpful. A tour of areas of public access and potential exposure with a sensitive hand-held instrument, such as a Ludlum μR Meter, could provide useful information about actual radiation levels at these locations. Comparison with background exposure rates and their variability at locations encountered en route to the points of interest will provide a frame of reference for such measurements.

Future Operations

Because operations at LANL are constantly evolving and changing, LANL is required by 40 CFR 61 to consider the potential impact of emissions related to proposed future operations. We stress, however, that future operations at the site have no bearing on our determination of compliance for 2001 as part of this audit. Each audit examines the statement of compliance for a year already past. It is instructive, as a part of the audit process, to review potential future

operations and evaluate LANL's methodology for estimating emissions related to those operations.

To examine the issue of future operations during this audit, we focused on two planned operations and evaluated LANL's emissions control and monitoring strategy. These operations were selected by the audit team following discussions with CCNS and IEER. The potential sources of future emissions discussed in detail during this audit were the TA 54 waste characterization project and the medical isotope facility.

The TA 54 waste characterization project is discussed in detail in an earlier section of this report. We evaluated and made recommendations regarding the methods used to estimate emissions for this project (Fuehne 2002a).

A presentation was given by MAQ on the medical isotope facility during our August visit. Discussions that took place during and after that presentation led the audit team to conclude that adequate steps have been taken to estimate potential emissions from this operation and determine monitoring requirements, including additional evaluations to address possible changes in scope of the intended operations, which are scheduled to begin in 2003.

VOLUNTARY COMPLIANCE EVALUATION INVOLVING STAKEHOLDERS: A MODEL FOR THE FUTURE

Introduction

The audit team believes that the audits conducted to verify compliance with 40 CFR 61 Subpart H at LANL have yielded many technical improvements and new ideas for calculating and documenting compliance. Most of these have already been implemented at LANL and some are ongoing. This report continues to identify areas where improvements can still be made.

It is evident that the audits under the Consent Decree have had a positive effect in helping LANL improve a number of technical areas related to compliance. Of equal importance, we believe the audits have helped stakeholders to be more aware of the requirements of 40 CFR 61 Subpart H and the role they can play in verifying compliance. There is no question that the audit process under the Consent Decree has become increasingly more effective and efficient as time has passed. It seems reasonable that many elements of the audit process should be documented for the future in the event similar audits are conducted at LANL or any facility where compliance with regulations involving exposures to the public are determined.

The audit team hopes and recommends that LANL and stakeholders will continue the compliance review process on a voluntary basis in the future. It is recognized that under voluntary participation, the content and extent of stakeholder participation may be somewhat different from the legal framework provided by the Consent Decree, and that there must be specific ground rules to guide all parties and to clarify expectations. The next section describes a framework under which a voluntary compliance evaluation program might exist and lists key elements that have contributed to the success of the audits under the Consent Decree. Together these provide a model for future stakeholder involvement in compliance evaluations in the future. This framework assumes that for a facility creating risk to the public, there are releases of contaminants to the environment and a pathway of exposure to the public.

Objective

The objective of a voluntary compliance evaluation program with stakeholder involvement is to increase the understanding of compliance with regulations and to seek stakeholder input to the process. It also makes compliance more transparent and helps authorities make better decisions with regard to compliance and reduction of risk.

There are many regulations governing compliance for radionuclides and chemicals in the environment. In this model, we are referring to compliance with regulations that focus on the release of radioactive materials to the environment and subsequent exposure to the public; however, in principle, the structure we propose would fit almost any situation of environmental exposure to the public. Furthermore, it is conceivable that variations of this model could be used for workers who are exposed occupationally as well.

The implementation of a voluntary compliance evaluation process involving stakeholders is especially important when public health is the objective of compliance. The series of audits under the Consent Decree have clearly demonstrated that public awareness of risk to exposure from radionuclides is greatly enhanced. In addition, these audits have significantly increased

stakeholder understanding of the compliance process. The audit team believes these achievements have been some of the most satisfying and noteworthy aspects of the audits.

Key Elements of a Voluntary Compliance Evaluation Program

Stakeholders, Key Participants, and Participants

Stakeholders are parties who have an interest in compliance of the facility. In this context, the term stakeholder has a broad interpretation and may include individuals from state and federal agencies, Tribes, citizens' groups, and private individuals who are concerned about compliance. Key participants are stakeholders who work with the facility to initiate a voluntary compliance evaluation. Key participants will be involved in selecting the mediating authority. Participants include other individuals who participate in the compliance evaluation once a commitment to the process has been made. However, to be considered as a participant in the compliance evaluation, individuals have a responsibility to become actively involved in the process.

At the initiation of a voluntary compliance evaluation, the facility and a group of key participants should meet to affirm their commitment to the process and take the steps necessary to begin the process. We believe that key participants should be representative of a wide spectrum of views and interests in the facility's existence. The key participants are individuals who will work with the facility to select a mediating authority and develop a draft agreement for the mediating authority to sign confirming initiation of the process.

During the LANL audit under the Consent Decree, a technical monitor (IEER) worked for CCNS to ensure the audit was objective and comprehensive. This arrangement worked well and improved the credibility of the process. For a voluntary compliance evaluation, participants could invite technical experts to assist them with the process.

Prescribed Framework for Implementation

The program must have a documented framework, which includes a clear scope, objectives within the scope, timeframe, and ground rules for implementation. This framework should be developed by the participants and the facility, with the assistance of a mediating authority. This framework must be in place before the compliance evaluation begins. Once the prescribed framework is developed, it can be adjusted prior to initiation of the compliance evaluation.

Source of Risk and Need for Compliance

In our model, there must be a source of risk, regulations that govern control of risk, and exposure to the public. Public in this sense implies individuals who live near the facility and who may, as a result, be subject to some incremental increase in risk because of the facility's existence. This definition may be expanded to include current risk (ongoing releases to the environment), historical risk (past releases to the environment), or future risk (potential releases to the environment). However, in order for this model to apply, there must be releases of contaminants to the environment and a pathway of exposure to the public from the facility.

For compliance under the Consent Decree, the primary regulation of concern was 40 CFR 61, Subpart H, which deals with releases of radionuclides to air. Under our model, the voluntary

compliance evaluation could focus on any area of compliance agreed to between the Key parties and the facility.

Primary Burden of Responsibility

It must be recognized that the burden of responsibility for implementation of a model for compliance evaluation rests primarily with the facility. This is important to acknowledge for several reasons. First, it must be recognized that the model we propose is voluntary; that is, there is no legal mandate for its implementation. A voluntary program could not be undertaken without the will of the facility. Resources committed by the facility demonstrate a willingness to be open about compliance and to help stakeholders understand how it is determined. Second, it is likely that most of the cost will be borne by the facility because it bears the burden of responsibility for demonstrating compliance, not the stakeholders. Because of this responsibility, stakeholders must recognize there will be limits to how far facilities can go in support of voluntary compliance evaluation. Finally, the burden of responsibility for compliance rests with the source of risk, the facility.

Scope

The scope of the compliance evaluation must be defined before the process begins. Although it is reasonable to expect that this model may work for a number of different organizations and facilities, we believe a succinct and well-defined scope is the key element of success. Therefore, in the model we propose, it is important to establish the scope for each compliance evaluation before it begins. The scope should be agreed upon between the participants and the facility with the assistance of a mediating authority. Issues will inevitably arise during the evaluation that may require the scope to be revised. This is normal in the course of openness and stakeholder involvement and should be expected. However, any changes to the scope must be agreed upon between participants and the facility.

During the Consent Decree, the audit team took the approach to listen to all ideas and consider all issues that arise that we believe are within the scope. It has been our job as auditor to make decisions about what issues we can address and those that, for whatever reason, we cannot consider. Our approach has been to try to respond to all questions we believe are within the scope and can reasonably be dealt with. We believe that for a voluntary model to be successful, adherence to a strictly defined scope is essential for success.

Mediating Authority

The audit team believes that a mediating authority will be required to carry out voluntary compliance evaluations. Over the course of the Consent Decree audits, we learned that there are many issues that arise that are viewed differently by participants and the facility. These differences are to be expected. Nevertheless, there must be a way to try to resolve differences when they arise, and this is the job of the mediating authority.

The selection of a mediating authority is critical to success and it must be completed well before the compliance evaluation takes place. The individual(s) must be accepted by the majority of participants and the facility. The mediating authority must be familiar with the technical area

being covered and understand the relevant regulations. Once the selection is made, the mediating authority has the responsibility to work on behalf of all parties and to see that the voluntary compliance evaluation meets its objectives.

The mediating authority should sign an agreement with the facility and key participants acknowledging the responsibility being undertaken. In order to improve the credibility of the voluntary compliance evaluation, financial support for the mediating authority should be from an independent organization and not directly from the facility if at all possible. This presents a problem for most agencies because there is no readily accessible avenue for providing independent support. We believe there are several options available to address this problem. One option is to provide funds to an independent party such as a university, a state department, or a foundation. Another avenue for funding that may be possible is that funds coming directly from the facility are provided as a grant and administered by the key participants and payment made to the mediating authority upon request. Once funding is arranged, it is the responsibility of the mediating authority to remain within the financial constraints allowed unless changes to the funds available are approved by the facility and the majority of participants upon mutual agreement.

It is the responsibility of the mediating authority to identify the participants who will be involved; to meet with the facility and participants; to establish the scope, objectives, timeframe, ground rules for implementation; to answer and try to resolve issues that arise during the evaluation; and to document the evaluation process.

Frequency and Duration of Voluntary Compliance Evaluations

The frequency of voluntary compliance evaluation depends on the type of facility and interest on the part of stakeholders. We believe the frequency of a voluntary compliance evaluation should be no more than every two years. The interval between evaluations could be longer should more frequent evaluations be deemed by the participants and the facility as unproductive. A frequency less than two years seems unreasonable in terms of costs and time needed by the facility to make adjustments in their program as a result of ideas contributed during an evaluation.

We also believe that the maximum length of time over which a voluntary compliance evaluation should take place is three months. We found that a sufficient period of time was needed to allow an exchange of documents to take place and time to review and question the information provided. Additionally, we believe the concept of participants viewing facilities that are potential sources, discussing the procedures and documentation with the facility personnel and having their questions answered about the issues at hand, and witnessing procedures used in the monitoring process are vital to any voluntary compliance program. Arranging for these activities to take place is time consuming, but we believe is worth the facility's investment. However, we also feel that a definite schedule has to be established that provides a clear termination of the evaluation.

Security

Security is a critical element of a voluntary compliance evaluation program and it requires the cooperation of all parties to abide by security requirements at the facility. This is especially important at the Department of Energy facilities, but security is also important at any facility that

agrees to participate in voluntary compliance evaluation. The audit team believes that security issues need to be clearly stated by the facility and must be complied with by all participants. It is important that all participants, but particularly those who can make a technical contribution to the process, not be denied the opportunity participate on the basis of security issues if at all reasonable and within the authority of the facility as was accomplished during the Consent Decree audits.

Documentation of the Compliance Evaluation

The audit team believes it is the responsibility of the mediating authority to document the voluntary compliance evaluation process. The facility and stakeholders should have an opportunity to provide input into a draft summary report resulting from the evaluation. Emphasis should be given in the documentation to ways in which the program could be improved and to specific issues that were raised that need to be considered in the future.

During the evaluation process, participants may submit written questions or issues they wish the mediating authority or the facility to answer. All questions within the scope should be documented and should be included in the final report with a written response. It is likely that some questions cannot be answered during the compliance evaluation or may not be within the scope of the evaluation. This is a decision the mediating authority must make. However, to the extent possible, questions asked should be documented and responses should be made.

Support for a Voluntary Compliance Evaluation

It is apparent that initiation of a voluntary compliance evaluation program requires resources to make it successful. We recognize this commitment and are confident that expenditure of these resources would be of great benefit to the facility. Resources are needed not only to support facility staff to undertake the responsibilities of a voluntary compliance evaluation program, but we believe some support should be set aside for participants as well. Financial support for participants is a difficult objective to achieve because there is not a good system established to provide independent support that ensures a fair and equitable distribution of resources. Nevertheless, providing support to participants should be considered by the facility. Some possible methods for independent financial support that might be considered include: (1) allowing the mediating authority to distribute funds to participants as grants; (2) having a group such as a facility's citizens' advisory board to administer funds; (3) providing funds to an independent foundation who would then distribute them to participants. There is no question that independent financial support to participants is a key element of success of a voluntary program but may also be the most difficult aspect to implement.

Accessibility to Documents

A key element to success of a voluntary compliance evaluation is access to documents by the mediating authority and the participants. Our experience during the three audits under the Consent Decree clearly shows that documents related to compliance are essential in verifying compliance. During the three Consent Decree audits, there was never a case where documents requested were not made available to all participants who wanted them. We believe that to the

greatest extent possible, participants should be able to request documents, with the approval of the mediating authority, as long as the documents are related to the scope of the compliance evaluation. Our approach to tracking documents requested was to develop a document request list that was continuously updated. The list indicated who requested the document and when it was delivered. We never had a situation where classified information was requested, nor was it required during the audits under the Consent Decree. It is recognized that the distribution of documents presents a burden to the facility, but we feel that this policy of openness and accessibility of information to the participants was critical to success.

Possible Tasks of a Voluntary Compliance Evaluation

When the audit team began its work at the beginning of the first audit in 1997, there were no specific guidelines to follow. Therefore, a workplan was developed that outlined the most important components to check to make a decision about compliance. Although the components of a voluntary compliance evaluation will vary from one facility to another and will depend upon the scope of work and regulatory issue being evaluated, we list below the most important tasks resulting from our audits under the Consent Decree. These may serve as a guide in the proposed model.

Task 1. Select the mediating authority. Identifying the mediating authority is the first step to take to begin the process of a voluntary compliance evaluation. The mediating authority should be selected with the agreement of the facility and the majority of the key participants. Once the mediating authority is selected, an agreement should be signed between the mediating authority, the facility, and the participants that states the scope, objectives, timeframe, and ground rules for implementation of the voluntary compliance evaluation. This agreement with the mediating authority is a public commitment that the facility and participants have agreed to the process.

Task 2. Identify additional participants and their responsibilities. Once the mediating authority is selected, it is necessary for the mediating authority and key participants to identify additional participants who wish to be involved in the voluntary compliance evaluation and to establish the ground rules and responsibilities for participation.

Task 3. Define the Scope. The first task of the mediating authority is to develop a draft scope of the compliance evaluation. The draft scope should be as detailed as possible. Once documented, the draft scope should be reviewed by the facility and participants, and their comments should be considered before the final scope is submitted. It is ultimately the responsibility of the mediating authority to establish a scope that meets the objectives of the evaluation and remains within the limits of the agreement.

Task 4. Request documents. One of the first tasks is to submit a document request to the facility. Documents requested might include

- Quality assurance plans and implementation procedures
- Procedures related to effluent monitoring, environmental surveillance, and dose assessment, including original data and calculations
- Annual site environmental reports and reports submitted to regulating agencies
- Environmental monitoring plans
- Effluent monitoring plans, including schematic drawings of effluent sampling systems
- Contaminant inventories, including current inventory and past inventories

- Results of self-assessments, internal audits, or external audits related to effluent monitoring, environmental surveillance, quality assurance, or dose assessments
- Results of any atmospheric dispersion validation, verification, benchmarking, calibration, or tracer studies

If the topic were something other than compliance with Clean Air regulations, this list would be appropriately modified.

Task 5. Kickoff workshop. A workshop should be conducted with the facility, participants, and technical advisors at the beginning of the evaluation. The purpose of the workshop would be to confirm the scope and schedule, discuss security requirements, establish channels of communication, and review the ground rules.

Task 6. Review sources of risk. One of the initial tasks in an evaluation of compliance where the regulation is concerned with limits on exposure to the public is to review the sources of contaminants released to the environment. Generally, this information forms the starting point for a calculation. This information should be documented and available for review by the mediating authority and participants. Additionally, it may be helpful for participants to visit sites where the sources originate. These visits, of course, will depend upon the security arrangements that can be made at the facility. However, we believe participants having an opportunity to view sources of contaminants is important and helps demystify the process of determining compliance.

Task 7. Evaluation of effluent monitoring. Effluent monitoring data and viewing effluent monitoring stations and procedures is another important task in compliance evaluation. As with viewing sources above, it is equally important for participants to have an opportunity to view monitoring equipment and original data.

Task 8. Evaluation of quality assurance. Quality assurance is a critical element of any compliance program. It is also an area that is of significant interest to participants for several reasons. Knowing that information is being checked independently gives participants confidence in the accuracy and completeness of the information. Most participants do not have the experience or background to verify compliance. However, when they review information in a compliance evaluation, they can often help identify where data do not seem correct. Therefore, it is natural that participants will expect to see how data are being checked. This verification is important in gaining credibility for the compliance process.

Task 9. Evaluation of compliance. Once information needed to evaluate compliance has been gathered and the mediating authority and participants have had an opportunity to review and understand the process, it is the responsibility of the mediating authority to develop conclusions about what has been learned. It should be recognized that the purpose of a voluntary compliance evaluation is to help participants understand how compliance is determined and to provide input to the facility about ways to improve the process, not to make a decision about compliance itself. In reality, only the facility and the authority responsible for the compliance regulation can determine if compliance is met.

In our implementation of the audits at Los Alamos National Laboratory, we divided our findings into three areas: (1) regulatory issues; (2) technical issues; and (3) specific observations and recommendations. Regulatory issues dealt with findings that could be directly linked to the non-compliance with the regulations. Such issues impacted the decision about compliance and required the facility's attention. Technical issues were findings of a technical nature that were not directly linked to the regulation but were important in establishing that compliance exists. Nearly

every technical issue we documented during the audits was implemented at LANL during the period between audits, significantly improving the collection of data and the documentation of compliance. Specific observations and recommendations are ideas that are based on good scientific principles and, although not required, would help make compliance more readily understood and defensible. Many of the recommendations we documented during the audits have been implemented at LANL.

Task 10. Final report. The mediating authority is responsible for issuing a final report on the voluntary compliance evaluation. The report should be issued as a draft and participants and the facility given a reasonable period of time to review it and provide written comments. Comments should be addressed and a final report issued by the mediating authority. Participants would be free to issue their own reports should they wish to do so.

Task 11. Final workshop. Once the final report is completed, the mediating authority should convene a workshop with the facility, participants to review the findings of the compliance evaluation.

CONCLUSIONS

This report documents the results of the third independent audit of LANL performed by *RAC*. The audit focused on LANL's compliance with 40 CFR 61, Subpart H, for 2001. The audit was conducted as part of a Settlement Agreement and Consent Decree that resolved a lawsuit filed against the DOE and LANL by CCNS. The audit team divided its work into four areas that addressed the major elements of the regulation. These elements comprise the major sections of this audit report. The audit team focused on the following four areas:

- Radionuclide usage and associated emission estimates for unmonitored point sources
- Major release point effluent monitoring
- Environmental compliance sampling for non-point sources
- Dose calculation.

The audit team also evaluated other areas as it assessed compliance with the regulations. These included traceability of data to their original source, documentation supporting compliance, technical competence, quality assurance, and overall strength of the compliance program.

This audit concluded that LANL was in compliance with 40 CFR 61, Subpart H for 2001. The audit team commends LANL for their implementation of recommendations provided in the first two audits and also the cooperation they have shown during this audit to make it an open, thorough, and responsive process. Furthermore, the Rad-NESHAP compliance program at LANL and this audit process could be considered as a model for other DOE facilities. Credit for this achievement is also due to CCNS, who, as a citizens' organization, helped to initiate the audits and design their format.

The audit team did still identify some areas for improvement. These findings are detailed throughout the technical evaluation sections and are summarized in Appendix A.

Additionally, the audit team evaluated some areas peripheral to the scope of the audit; that is, they do not pertain to LANL's compliance status. These issues included complex terrain modeling, NEWNET, use of TLDs for environmental monitoring, uncertainty related to dose calculations, and future operations. This report also provides important guidance for future voluntary evaluations of the compliance program at LANL.

It is noteworthy that this audit was conducted under unusually difficult circumstances created by the events of September 11, 2001 and important issues with regard to security at LANL throughout this year. The audit's success is a direct reflection of the professionalism and dedication to this process by all parties who participated. The audit team expresses its appreciation for the spirit of cooperation that made this audit possible.

The audit team believed that the public's role in the compliance process was critical. The positive interaction between the audit team, LANL, IEER, and the public confirmed that where regulations related to public exposures are being evaluated, the public can play an important role. The audit team also believed that IEER's role to monitor and verify the audit process was valuable in maintaining this atmosphere of openness. IEER challenged the audit team to conduct a thorough and fair evaluation of compliance and with the public raised a number of questions regarding important issues. These issues are detailed in Appendix C and are addressed within the text of this report.

This audit of 40 CFR 61, Subpart H was conducted by an independent audit team working under the auspices of the U.S. Department of Justice. This arrangement was critical to guarantee the independence of the audit team. The audit team learned much from the process used in the first two audits, and used these lessons to complete this audit much more efficiently.

It is emphasized that these audits were more rigorous and broader in scope than previous audits conducted for compliance with 40 CFR 61, Subpart H, at LANL and at other DOE sites. The degree of cooperation received from all parties involved was exemplary. The audit team especially commends the Meteorology and Air Quality Group of the Risk reduction and Environmental Stewardship Division at Los Alamos National Laboratory because supporting the audit process has required extraordinary effort on their part. The audit team also thanks and commends DOE, LANL, CCNS and IEER for their active involvement and support.

This is the third audit of its type. The audit team previously completed audits that covered compliance for 1996 and 1999. Many of the improvements made in the program and outlined in this report resulted from those first two audits. According to the Consent Decree, if "…the third audit identifies substantive deficiencies with compliance with Subpart H that the auditor believes require corrective actions, a fourth technical audit will commence no later than the end of calendar year 2003. The scope of the fourth audit shall be limited to determining whether necessary corrective actions identified in the third technical audit have been satisfactorily accomplished." The audit team has concluded there were no substantive deficiencies requiring corrective actions that justify having a fourth audit under the Consent Decree. Therefore we consider that audit requirements under the Consent Decree have been met and are concluded with this report.

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