Los Alamos NATIONAL LABORATORY EST.1943

National Nuclear Security Administration

Associate Director for Operations
P.O. Box 1663, A104
Los Alamos, New Mexico 87545
505-667-0079/Fax 505-665-1812

Date: May 20, 2004
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Charles Miller
Senior Scientist, NCEH/EHH
Centers for Disease Control and Prevention
1600 Clifton Road, NE
Atlanta, GA 30333

Reference: Comments on the DRAFT Interim Report of the Los Alamos Historical Document Retrieval and Assessment (LAHDRA) Project

Dear Dr. Miller:

We appreciate the opportunity to review and comment on the subject report before it is finalized and published as part of the Centers for Disease Control and Prevention (CDC), Los Alamos Historical Document Retrieval and Assessment (LAHDRA) Project. We understand this is intended as an interim report, but believe it must be current, accurate and technically sound, as it will play an important role shaping public confidence in this project.

Our comments are limited to two areas, as follows:

1. The section "Measurements of Plutonium in Soil as Indicators of Historical Releases" (pages 78 through 83) is based on several assumptions and methods that we do not find to be scientifically defensible. We believe the assumptions and methods selected by CDC's contractors create serious deficiencies in the analysis, and we request that the report not be published without formal, CDC-managed resolution of our comments. The heart of our concern is the magnitude of the airborne plutonium release estimate derived from soil contamination results. According to the first paragraph on page 79, the CDC's contractors concluded that selected soil contamination data "indicated that airborne plutonium releases from LANL [Los Alamos National Laboratory] operations could have been hundreds of times higher than the 1.2 Ci officially reported." In the attached analysis, we show that with a defensible set of assumptions the calculated plutonium release using the same data sources is approximately 1 Ci. The major points of our concern are (1) the process of selecting 37 soil contamination measurements out of 697 introduces significant bias into the data set; (2) most of the locations used for the analysis in the Interim Report are confounded by close-by sources of plutonium contamination that render the data inappropriate for estimating airborne releases from TA-1 or TA-21; (3) the Interim Report fails to account for the impact of variable rainfall levels (thus variable background levels) on and near the Pajarito Plateau; and (4) dispersion modeling in the Interim Report with the RSAC code results in release estimates that are substantially higher than those predicted by the EPA-standard CAP-88 code. In the attachment, we examine each of these concerns in greater detail. As part of comment

resolution we recommend that CDC consider obtaining an independent technical review prior to final publication of this report. If you concur with the importance of independent review, we will make available staff in LANL's Risk Reduction and Environmental Stewardship Division to provide copies of all the references cited in the attachment and to discuss these matters with CDC, Shonka Research Associates, or other reviewers. Please contact the LANL CDC-LAHDRA Project Office at (505) 665-0750 to request such assistance.

2. The section of the Interim Report titled "Challenges to Information Gathering at Los Alamos" does not indicate the significant progress that LANL and CDC have jointly made since August 2003 streamlining document release for this project. As you well know security incidents and emerging concerns about homeland security made the task of accessing classified information substantially more difficult in the 2000–2003 timeframe, not only for CDC and its contractors but for LANL staff as well. We believe that beginning August 2003 these difficulties have been substantially resolved and we are confident that your staff concurs. We would appreciate your considering an update to this section of the report to indicate the very real and necessary security restrictions that slowed document release, and most importantly to indicate the recent progress.

Finally let me assure you that despite significant concerns about this Interim Report, LANL recognizes the importance of this study and CDC's public service commitment. We have committed significant resources to this project and will continue to work closely with the CDC and its contractors to bring the LAHDRA project to a successful conclusion.

Respectfully,

Barbara A Stine for

James L. Holt,
Associate Director for Operations

Joe Vozella

Joe Vozella,
Assistant Manager for Facility Operations
Los Alamos Site Office

CMH

Distribution w/att:
Phil Green, CDC
C. M. Wood, CDC
Joe Shonka, Shonka Research Associates
Attn: Debby Shonka
4939 Lower Roswell Road, Suite 106
Marietta, GA. 30068
E. Wilmot, NNSA/DOE LASO, A316
F. Ward, NNSA/DOE LASO, A316
M. Johansen, NNSA/DOE LASO, A316
B. A. Stine, ADO, A104
R. Brake, ADO, A104
C. Holmes, ADO, K560
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Title: Comments on the DRAFT Interim Report of the Los Alamos Historical Document Retrieval and Assessment (LAHDRA) Project

Author(s): Michael McNaughton, RRES-MAQ
Andrew Green, RRES-MAQ

Submitted to: Charles Miller
Centers for Disease Control and Prevention
May 20, 2004

Comments on the "Draft Interim Report of the Los Alamos Historical Document Retrieval and Assessment (LAHDRA) Project" dated March 21, 2004, obtained from http://www.shonka.com/ReConstructionZone/

Michael McNaughton and Andrew Green
Meteorology and Air Quality Group
Risk Reduction and Environmental Stewardship Division
Los Alamos National Laboratory
May 15, 2004

Abstract

We have estimated the airborne plutonium released from Los Alamos using the same basic method as described in the draft interim report of the LAHDRA Project, except we have corrected some misunderstandings and changed some assumptions. Our estimate is consistent with previous estimates and reports from LANL, whereas the LAHDRA estimate is "hundreds of times higher than the 1.2 Ci officially reported." Because of the significant public health implications of the Los Alamos Historical Document Retrieval and Assessment (LAHDRA) Project, the authors also strongly recommend that the reports generated by the Project go through an independent peer review process, such as by the National Academy of Sciences. This will greatly enhance the quality and public acceptance of LAHDRA reports.

Introduction

The following comments are limited to the methodology and assumptions used to develop the airborne release estimates for plutonium, as these are critical to any eventual health risk conclusions.

The section "Measurements of Plutonium in Soil as Indicators of Historical Releases", (pages 78 through 83) is based on several assumptions and methods that we do not find to be scientifically defensible. According to the first paragraph on page 79, the LAHDRA results "indicated that airborne plutonium releases from LANL operations could have been hundreds of times higher than the 1.2 Ci officially reported." In contrast, in the following paragraphs we show that with a defensible set of assumptions the calculated plutonium release is approximately 1 Ci.

The Draft Interim Report of the LAHDRA Project is not sufficient for detailed analysis, so to complete this review the Laboratory's Risk Reduction and Environmental Stewardship air quality (RRES-MAQ) personnel contacted Joseph Shonka and obtained the document "Back-calculation of Airborne Plutonium Releases from LANL soil data", (Shonka Research Associates number SRA-03-005, dated 02/04/04).

The major points of concern are as follows:

- The process of selecting 37 measurements out of 697 introduces a bias into the data set used for the LAHDRA calculations.
- For most of the locations impacted by LANL, there are sources of plutonium much closer than TA-1 or TA-21, and pathways much more likely than airborne release.
- Global fallout is a function of rainfall, so background concentrations measured in the arid Rio Grande valley cannot be directly applied to soil samples on the wetter Pajarito Plateau.
- The soil concentrations predicted by the RSAC code disagree with those predicted by the EPA-standard CAP-88 code.

In the following sections, we examine each of these concerns in detail.

Selection of data

The LAHDRA Project used a set of criteria to select 37 measurements out of the original 697. Thus, 660 measurements, or 95% of the data have been excluded. The 697 measurements consist of about 25 measurements at each of about 24 locations; some locations were omitted, and at each individual location, between 1 and 7 of about 25 measurements were selected.

One of the selection criteria was the ratio of the plutonium concentration to the cesium-137 concentration. To be selected, the ratio must be greater than 0.065. The selected data have either an unusually large plutonium concentration or an unusually low cesium-137 concentration, which tends to bias the data toward large plutonium concentrations. By comparing the selected data with average data at each location, we calculate the bias to be a factor of 2.

It is instructive to consider the likely cause of the variation of measured concentrations at one location. The soil-sampling procedure begins with the collection of 2.5 liters of soil. After thorough mixing, an aliquot is dissolved in acid for further analysis; at various times in the past, the aliquot has been either one or ten grams. Apparently, the aliquot sometimes contains a "hot particle" with activity on the order of 0.01 to 1 pCi and diameter on the order of 0.3 to 1 micrometer (which is the range of particle sizes suggested by Jordan and Black in their 1958 paper). This would explain the variability in the data at each location impacted by LANL emissions. Suppose 1 sample out of 10 contains a hot particle; then a representative sample contains on average one-tenth of such a particle, and the data are better represented by an average than by the single sample containing the hot particle.

There are many possible ways to handle the data in an unbiased way. For simplicity, we have used the LAHDRA criteria to select the locations impacted by LANL and used the average concentration from all sampling data at each location.

Other sources and other pathways

The LAHDRA Project assumes that all plutonium contamination at all the soil-sample locations came from TA-1 or TA-21 by the air pathway. This assumption is incorrect for the soil sample locations at TA-21, TA-49, TA-50, TA-52, TA-54, Test Well DT-9, and Otowi. These locations have all been contaminated by a water pathway or by releases from nearby TAs.

For example, on July 9, 1974, the soil at TA-50 was contaminated by a spill of about 200 mCi of plutonium from the radioactive liquid waste treatment facility; during the subsequent cleanup, 600 cubic yards of this contaminated soil with plutonium concentrations up to 300,000 pCi/g were transported to TA-54. Although most of the contamination was contained, remnants of this and other spills still measure a few pCi/g at TA-50 and TA-54. These and other sources of contamination are discussed in Appendix A.

At individual locations far from TA-1 and TA-21, this incorrect assumption that the measured contamination came by the air pathway from TA-1 or TA-21 results in some very large individual estimates of the plutonium released. For example, the assumption that 0.19 pCi/g at TA-50 came by air from TA-21 results in an estimated release of 650 Ci. Generally, the locations farthest from the purported source lead to the largest estimates. This fact is acknowledged in the discussion in the second paragraph on page 82 of the draft interim LAHDRA report, and as a result the LAHDRA analysis selected only locations within a 5.5 km boundary. If the boundary were to be extended or the criteria changed, even larger estimates would be obtained, so we consider it important to review all the soil sample locations and describe the reasons for rejection, which we have done in Appendix A.

As it stands, many locations have already been omitted from the most recent LAHDRA study. Only 3 others need to be omitted because of alternate pathways: TA-21, TA-50, and Sigma Mesa; omitting these decreases the final result by less than a factor of 2.

Global fallout

As stated in the fourth paragraph on page 78 of the LAHDRA report, there have been several attempts to estimate airborne plutonium releases based on soil measurements, and these have used different estimates of the background from global fallout. The background value 0.006 pCi/g is mentioned on page 78 of the LAHDRA report, and two values, 0.006 pCi/g and 0.0144 pCi/g, are mentioned on page 10 of the SRA report, accompanied by the statement that "this change [from 0.006 to 0.0144] is a small contributor to the difference because data for the current study involved samples with higher levels of plutonium". However, after we correct the data set as described in the two previous sections, the results are sensitive to the choice of the background concentration; we find the change from 0.006 to 0.0144 pCi/g changes the estimate of plutonium released by a factor of 1.7.

Global fallout from atmospheric nuclear weapons testing is a function of rainfall. Radionuclides from nuclear tests were injected into the stratosphere or the high troposphere and were deposited on the earth's surface over the course of months, mostly by the action of rain or snowfall. Therefore, regional soil concentrations from locations with low annual precipitation cannot directly be used to estimate the background near Los Alamos. Even within ten miles spanned by LANL, the rainfall near the Jemez Mountains is about a factor of two greater than it is near White Rock.

The most recent estimate of soil background data is in the report "Inorganic and Radionuclide Background Data for Soils, Canyon Sediments, and Bandelier Tuff at LANL", LA-UR-98-4847, by R.T. Ryti, P.A. Longmire, D.E. Broxton, and E.V. McDonald (1998); according to Table 3.3-2, the mean plutonium concentration in regional and perimeter soils is 0.015 pCi/g, with a standard deviation of 0.013 pCi/g. However, this estimate is not adjusted for rainfall.

We have also estimated the background at the individual locations in the present study by averaging the data that have a plutonium-to-cesium-137 ratio consistent with global fallout. For the key locations, this estimate is consistent with the previous estimate of 0.015 pCi/g. We also considered global estimates such as UNSCEAR-2000, see Appendix B. These results are also consistent with 0.015 pCi/g.

For the present, we have assumed a background of 0.015 pCi/g at all locations used in this study. This is almost the same as the value 0.0144 pCi/g used by the latest iteration of the LAHDRA analysis, so this does not change the results. However, it would be better to use individual background values for each location, which would likely range from 0.01 to 0.03 pCi/g. This uncertainty in background corresponds to about a factor of 2 uncertainty in the estimates of plutonium released.

Predictions

The method described in the last paragraph on page 78 of the draft LAHDRA report compares the prediction of the RSAC computer program (see http://www.inel.gov/rsac/) with the measured soil concentrations. The LAHDRA predictions are shown in Figure 9 of the Shonka Research Associates report, SRA-03-005, together with a power-series fit, $y=11469501x^{-2.7}$, where y is the plutonium concentration in pCi/g in the downwind sector from 1 Ci released, and x is the distance from the source in meters.

The exponent of –2.7 is surprising, especially considering the last paragraph of page 78 of the LAHDRA report that says it is based on Pasquill-Gifford stability-class C, because the standard Gaussian-plume model for class C results in an exponent of –1.8. An important question is: which exponent is most consistent with the data? At present, we don't have a definitive answer.

We begin with a specific example taken from the SRA report. Using Figure 9 together with a value of 0.04 for the fraction of the time the wind is in the right direction (called the "DP Fraction" in Table 2 of the SRA report), the prediction for a 1 Ci release from

TA-21 is 0.00003 pCi/g at Two-Mile Mesa. In Table 2, one of the 25 measurements at Two-mile Mesa is selected by the LAHDRA criteria: 0.016 pCi/g. Background is subtracted (presumably 0.0144 pCi/g) and the net concentration is listed as 0.00 pCi/g (presumably 0.0016 pCi/g). The ratio 0.0016/0.00003 is approximately 50, so this measurement is taken to indicate a release 50 times the original prediction, i.e., 50 Ci.

Several points can be drawn from this example.

- First, the estimated release is sensitive to the background. Changing the background from 0.0144 pCi/g to 0.006 pCi/g increases the estimated release from 50 Ci to 300 Ci.
- Second, substituting CAP88 for the RSAC program changes the prediction by a factor of 50 and so decreases the estimated release from 50 Ci to 1 Ci.
- The RSAC program predicts that most of the plutonium was deposited close to the source; consequently it predicts very small concentrations far from the source, which leads directly to the large estimates of the plutonium released.

To examine this third bullet quantitatively, we first note that all the soil concentration measurements used by LAHDRA are outside a radius of 0.41 km, but the RSAC program predicts only 10% of the plutonium is outside this radius. Therefore, 90% must be within this radius, and we should be able to find it.

The equation $y=11469501x^{-2.7}$ may not be strictly valid at small x, but nevertheless if 100 Ci of plutonium is released and 90% of this is deposited inside a radius of 410 m, the average concentration inside this radius must be 2,300 pCi/g, an amount that would certainly have been noticed. At TA-21, this possibility is contradicted by data from environmental restoration measurements at TA-21 (outside the SWMUs) that show the maximum soil concentration is less than 3 pCi/g. Environmental restoration teams have also measured the alpha activity near TA-1, for example when they removed the radioactive-liquid-waste pipe from TA-1 to Acid Canyon, and when they remediated the hillside south of TA-1. Isolated patches of waterborne contamination were reported, but generally the data show that the concentrations are less than 20 pCi/g.

The inescapable conclusion is that the concentration cannot fall as sharply as predicted by the LAHDRA model and that the releases cannot exceed several curies.

To make a numerical comparison, we performed a calculation using the EPA-standard computer program CAP88. Compared with the LAHDRA model, the soil concentrations predicted by CAP88 were larger by a factor of 2 at the smaller distances (410 m) and by a factor of 50 at the larger distances (5000 m).

Estimate of plutonium released

We have estimated the plutonium released from TA-1 and TA-21 using the basic method described at the bottom of page 78 of the LAHDRA draft interim report except with the changes discussed above. Specifically:

- we used averages in place of a subset of data;

- we discarded data from locations affected by other sources and other pathways (see Appendix A);
- we used a single background value of 0.015 pCi/g; and
- we used CAP88 in place of the RSAC program.

Our estimate is summarized in Appendix C. If we assume TA-1 was the source, we estimate 1.4 Ci was released; and if we assume TA-21 was the source we estimate 1.0 Ci was released, which is consistent with the value of 0.8 Ci obtained by Harry Jordan and Ralph Black and published in the American Industrial Hygiene Association Journal, Volume 19, pages 20-25 (February 1958).

Summary

In summary, we have serious concerns about the methods and assumptions used to develop the airborne plutonium release estimates in the LAHDRA draft interim report, and we ask that CDC management consider obtaining independent technical peer review prior to final publication.

To support such a technical review, staff in the Laboratory's Risk Reduction and Environmental Stewardship Division can provide copies of all the above references and are available to discuss these matters with CDC, Shonka Research Associates, or other reviewers.

Appendix A: Discussion of Soil-Sample Locations

On-site locations

TA-16 (S-site), TA-11 (K-site)

Although sometimes listed as TA-16, this location is actually at the western edge of TA-11, near the boundary with TA-16. TA-16 has been in use since the 1940s, and TA-11 since the 1950s. There are 12 SWMUs at TA-11 and 37 SWMUs at TA-16, though there is no reason to expect plutonium contamination at this location. This location was not used by the LAHDRA Project. However, in a future analysis, it may be useful in determining the background from global fallout.

Two of the 30 soil measurements could be interpreted as either low-level plutonium contamination or fallout: the 1978 measurement of 0.086 +/- 0.091 pCi/g, and the 1981 measurement of 0.076 +/- 0.009 pCi/g. We discount the 1978 measurement because it is not statistically significant and the uncertainty is more than ten times the uncertainty of the other 29 measurements. The 1981 measurement is accompanied by 2.9 pCi/g of cesium-137 and 1.4 pCi/g of strontium-90, which suggests an unusually high concentration of global fallout at this location. The mean concentrations are: 0.021 +/- 0.021 pCi/g of plutonium-239-240, 0.82 +/- 0.81 pCi/g of cesium-137, and 0.54 +/- 0.37 pCi/g of strontium-90; these mean values are larger than average but consistent with global fallout.

TA-21

The TA-21 soil-sampling location is on the site of the former waste research lab, TA-21-45, and an old surface disposal area, SWMU TA-21-13(d); it is also close to SWMU 21-15 (also known as material disposal area MDA-B) and to SWMU 21-18 (also known as material disposal area MDA-V). A laundry for contaminated anti-C clothing was located on the other side of the road from the soil-sample location; the laundry drained to a nearby surface waste disposal area. The soil-sampling location was subject to a voluntary corrective action in 2003 and the surface soil was transported to TA-54. In summary, the TA-21 data cannot be used to deduce airborne emissions from TA-1 and TA-21.

The LAHDRA analysis used 7 of the 27 measurements at this location with an average net concentration of 0.25 pCi/g, whereas the corresponding average of all the data at this location is 0.12 pCi/g.

Near TA-33

The location designated as "near TA-33" is a part of TA-39 enclosed by a loop of State Road 4 as the road climbs toward TA-33. There is no reason to suspect plutonium contamination at this location, none is evident, and the LAHDRA Project did not use this location. The mean plutonium concentration, 0.013 +/- 0.013 pCi/g, is consistent with global fallout.

Potrillo Drive, TA-36

The "Potrillo Drive" location is close to TA-18. Small amounts of airborne contamination from TA-18 are known to have been released so this location cannot be used to estimate emissions from TA-1 and TA-21. The mean plutonium concentration, 0.012 +/- 0.009 pCi/g, is consistent with global fallout. LAHDRA did not use this location.

TA-50

The TA-50 location is close to the radioactive liquid waste treatment facility. The report LA-UR-96-1283, appendix I, describes a spill on July 9, 1974 that spread over 1000 m^2; gross alpha data ranged from background to 300,000 pCi/g. It is estimated that 200 mCi of plutonium was spilled. During the cleanup, 600 cubic yards of soil were removed and transported to Area G, TA-54. Although most of the contamination was removed, the area is not pristine. Therefore, the TA-50 soil data cannot be used to deduce airborne emissions from TA-1 and TA-21.

The LAHDRA analysis used 4 of the 27 measurements at this location with an average net concentration of 0.07 pCi/g, whereas the corresponding overall average at this location is 0.09 pCi/g.

TA-51

The TA-51 location is near Pajarito Road and close to TA-18, TA-53, and TA-54, which are known to have released small quantities of plutonium. The 1978 data have very large uncertainties, are not statistically significant, and should be omitted. The remaining data have a mean value of 0.017 +/- 0.011 pCi/g, which is consistent with global fallout. Nevertheless, they cannot be used to estimate the emissions from TA-1 and TA-21 because of the proximity to TA-18, TA-52, and TA-54

The LAHDRA analysis used 2 of the 11 measurements at this location with an average net concentration of 0.017 pCi/g, whereas the corresponding overall average at this location is 0.003 pCi/g.

East of TA-52

The location designated "East of TA-52" is near the boundary of TA-52 and TA-5. TA-52 was the LANL reactor development site. The remains of some early reactor components have been buried at material disposal areas nearby. There are many SWMUs and potential sources of plutonium contamination in the vicinity, including an old firing site. Therefore, the TA-52 data cannot be used to deduce airborne emissions from TA-1 and TA-21. LAHDRA did not use this location.

West of TA-53

There are no known sources of plutonium near TA-53. The 1978 data, 0.150 +/- 0.193, has an uncertainty more than ten times the other data and should be discarded. The remaining data range from background to 0.109 pCi/g, which indicates airborne emissions from TA-1 or TA-21. After discarding the 1978 measurement, the remaining 23 measurements have a mean plutonium concentration of 0.030 +/- 0.025 pCi/g.

The LAHDRA analysis used 4 of the 24 measurements at this location with an average net concentration of 0.027 pCi/g, whereas the corresponding overall average at this location is 0.015 pCi/g.

East of TA-53

There are no known sources of plutonium near TA-53. The data from several years have plutonium concentrations greater than is expected from global fallout, which could indicate airborne emissions from TA-1 or TA-21. The mean plutonium concentration is 0.053 +/- 0.055 pCi/g.

The LAHDRA analysis used 3 of the 11 measurements at this location with an average net concentration of 0.033 pCi/g, whereas the corresponding overall average at this location is 0.038 pCi/g.

East of TA-54

The TA-54 location is adjacent to LANL's largest waste site, Material Disposal Area G, which is known to contain plutonium contamination. Therefore, the TA-54 soil data cannot be used to deduce airborne emissions from TA-1 and TA-21. LAHDRA did not use this location.

East of Sigma I and II (TA-60)

The locations designated "East of Sigma I and II" are on Sigma Mesa (TA-60), a few hundred meters north of Effluent Canyon, so called because it receives effluent from the Radioactive Liquid Waste Treatment Facility at TA-50. There are 7 SWMUs on Sigma Mesa. SWMU 60-005(a) is an inactive evaporation pond that received effluent from TA-50 during the 1970s. Therefore, the "East of Sigma I and II" soil data cannot be used to deduce airborne emissions from TA-1 and TA-21.

The LAHDRA analysis used 1 of the 3 measurements at this location with a net concentration of 0.04 pCi/g, whereas the corresponding overall average at this location is 0.02 pCi/g.

Near Test-well DT-9

The DT-9 location is at TA-49, a few hundred meters east of Area AB, which contains plutonium, some of which has reached the surface; therefore it cannot be used to deduce

the emissions from TA-1 and TA-21. The mean plutonium concentration is 0.018 +/- 0.015 pCi/g. LAHDRA did not use this location.

Near R-site Road

The R-site Road location is at TA-15, several hundred meters west of TA-16-40. There are no known sources of plutonium nearby so it is reasonable to use these data to estimate airborne emissions from TA-1 or TA-21. The mean plutonium concentration is 0.019 +/- 0.014 pCi/g.

The LAHDRA analysis used 1 of the 28 measurements at this location with a net concentration of 0.020 pCi/g, whereas the corresponding overall average at this location is 0.004 pCi/g.

Two-mile Mesa

The Two-mile Mesa location is at TA-6 near the meteorological tower. There are 8 SWMUs at TA-6, including some burial pits dating from the 1940s. Although it is not known what is in these pits, there is no reason to suspect plutonium, so it is reasonable to use these data to estimate airborne emissions from TA-1 or TA-21. The mean plutonium concentration is 0.019 +/- 0.010 pCi/g, and the mean ratio of plutonium to cesium-137 is 0.003, which indicates the plutonium is from global fallout. One measurement out of 25 at this location is listed in Table 2 of the Shonka Research Associates report, SRA-03-005. The plutonium concentration for this measurement is 0.016 pCi/g, which is less than the average at this location; (the plutonium to cesium-137 ratio is high because the cesium-137 concentration is low). Plutonium background is in the range 0.015 to 0.02 pCi/g, so the net plutonium concentration is small and consistent with zero. If this measurement is used to estimate plutonium emissions from TA-1 and TA-21, the uncertainty will be very large.

The LAHDRA analysis used 1 of the 25 measurements at this location with a net concentration of 0.002 pCi/g, whereas the corresponding overall average at this location is 0.004 pCi/g.

TA-8

The TA-8 location is about a hundred meters east of TA-8. There are no known sources of plutonium nearby so it is reasonable to use these data to estimate airborne emissions from TA-1 or TA-21. After discarding the 1977 measurement because of its large uncertainty (0.122 +/- 0.108 pCi/g), the mean plutonium concentration is 0.035 +/- 0.024 pCi/g. The average cesium-137 and strontium-90 concentrations at this location are also relatively large: 1.0 pCi/g and 0.85 pCi/g, respectively, and the ratios suggest that all three radionuclides are from global fallout. This raises the interesting question of why this location has larger amounts of fallout than the average in Los Alamos County. Possible answers include: higher rainfall, and fallout washed down from the nearby hills.

LAHDRA did not use this location.

TA-49

The TA-49 location is on Bandelier National Monument land southwest of State Road 4, across the road from TA-49. TA-49 has a large source of plutonium in Material Disposal Area AB, a few hundred meters from the sample location. Therefore, these data cannot be used to estimate air emissions from TA-1 and TA-21. LAHDRA did not use this location.

Otowi

The Otowi location is on low-lying land where Los Alamos Canyon joins the Rio Grande. Los Alamos Canyon receives water contaminated with plutonium from TA-1, TA-21, and TA-45, via Acid Canyon, Pueblo Canyon, and DP Canyon. Therefore, these data cannot be used to estimate air emissions from TA-1 and TA-21. LAHDRA did not use this location.

East and West of Airport

There are no known sources of plutonium near the airport other than TA-21. It is reasonable to use these data to estimate airborne emissions from TA-21.

The LAHDRA analysis used 3 of the 11 measurements at "East of Airport" with a net concentration of 0.032 pCi/g, whereas the corresponding overall average at this location is 0.014 pCi/g.

The LAHDRA analysis used 7 of the 12 measurements at "West of Airport" with a net concentration of 0.107 pCi/g, whereas the corresponding overall average at this location is 0.156 pCi/g.

Other locations

None of the other locations used by the LAHDRA project are known to have plutonium from sources or pathways other than the air pathways from TA-1 or TA-21.

The LAHDRA analysis used 1 of the 22 measurements at North Mesa with a net concentration of 0.035 pCi/g, whereas the corresponding overall average at this location is 0.003 pCi/g.

The LAHDRA analysis used 3 of the 27 measurements at Sportsman's Club with a net concentration of 0.015 pCi/g, whereas the corresponding overall average at this location is 0.009 pCi/g.

The environmental restoration group of RRES division has data at other locations that we are in the process of collecting and understanding.

The best data are close to the source. Considering the data used by the LAHDRA project, the most accurate estimates are derived from the East- and West-Airport and East- and West-TA-53 locations, which are relatively close to TA-21 and do not have any other likely source. According to our analysis, the data at all of these four locations are consistent with a release of 1 Ci from TA-21. We are seeking data close to TA-1.

Appendix B

Calculational estimate of background activity due to fallout plutonium.

The plutonium background from weapons testing fallout is estimated as follows. Latitude specific deposition factors are taken from UNSCEAR 2000 Annex C, Table 8. Los Alamos lies in the band 30 - 40 N which is noted as having a deposition density of 5.09 Bq/m^2 for each PBq deposited in the northern hemisphere. The global depositions of Pu-239 and Pu-240 are 6.52 PBq and 4.35 PBq respectively (taken from UNSCEAR 2000 Annex C, Table 9), totaling 10.9 PBq. Fallout deposits in the northern hemisphere (as evidence by the Sr-90 data in UNSCEAR 2000 Annex C, Table 8) form 76% of the global depositions. This amounts to 8.3 PBq. Using the deposition density for this latitude we should expect an areal depositional density of

$$5.09 \times 8.3 = 42 \text{ Bq/m}^2 \text{ which is about } 1200 \text{ pCi/m}^2.$$

Deposition is a surface phenomenon, and the varying depth to which a sample is measured can obscure matters. For example, the CAP88 results are in pCi/m^2, which must be converted to pCi/g for comparison with the data used by LAHDRA.

Converting the areal density to a volume or mass density requires one to know the depth to which the sample was measured. The Soils and Biota Group in the RRES Division at LANL samples to 5cm. Further, to get to a mass density (measured in pCi/g) one must assume a soil density. The typical range is 1.3 to 1.8 g/cm^3; in the following calculation we used 1.6 g/cm^3. Putting these data together gives

$$1200 / (1.6 \times 100 \times 100 \times 5) = 0.015 \text{ pCi/g}$$

as the background concentration level for soil contamination in the Los Alamos region.

Note that the soil density and sample depth are crucial in deriving this estimate, yet both are somewhat debatable. Soil density depends on packing, granule porosity, wetness, rock type and variety of granule size. Sample depth is a matter of choice for the experimenter. Depth of penetration depends on the chemical form of plutonium including solubility, granule chemistry, rainfall, porosity, presence of other chemicals in the soil, plutonium uptake by vegetation, and plutonium particle size. In rainy regions with porous soil one would need to do deeper sampling than in drier regions with non-porous soil.

Most of the factors mentioned in the preceding paragraph do not affect the areal contamination density. Factors that move the plutonium laterally (heavy rainfall, wind) or remove it from the soil (vegetation uptake) have an effect. As a result, the background varies according to the location and the choice of a single background value of 0.015 pCi/g is an approximation. A complete scientific study would investigate the causes of the variation, assign an uncertainty to each measurement, calculate weighted averages, and propagate the uncertainty to the final result. In the present study, we estimate that the

uncertainty in background results in an uncertainty of a factor of 2 in the estimate of plutonium released from Los Alamos.

Appendix C: Calculation of plutonium released from TA-1 and TA-21

The table columns are as follows:

1. Location
2. Distance to location in meters
3. Direction to location (sector)
4. Air concentration (pCi/m^3) predicted by CAP-88, assuming 1 Ci is released at a uniform rate during 1 year
5. Areal deposition rate [pCi/(cm2·s)] predicted by CAP-88, assuming 1 Ci is released at a uniform rate during 1 year
6. The soil concentration (pCi/g) is the areal deposition rate (column #5) multiplied by 365*24*60*60 seconds per year, divided by 5 cm sample depth, and also divided by 1.5 g/cm^3 soil density. The time of 1 year is arbitrary; for example, if 2 years is chosen, the air concentration would be halved and the number of seconds in two years would be twice as much so the soil concentration would be the same.
7. Measured soil concentration (pCi/g): average value for the location minus 0.015 pCi/g background
8. Ratio of column 7 to column 6.

Following the method used by the LAHDRA Project, the ratio in column 8 is the estimated release of plutonium that would result in the measured concentration. Generally, the ratios are near 1 so the results are consistent with a release of 1 Ci.

The Two-Mile-Mesa data are unusual. The LAHDRA criteria selected only one measurement as indicating plutonium from LANL; the others appear to indicate slightly higher background, probably because of higher rainfall. If the background is assumed to be 0.017 pCi/g instead of 0.015 pCi/g, the ratio is halved. This illustrates that the distant locations are more sensitive to background.

Table 1. Calculation assuming unit release from TA-1

From TA-1			air conc	deposition	soil conc	measured	ratio
	dist (m)	dir	pCi/m3	pCi/cm2/s	pCi/g	pCi/g	
West of TA-53	1646	ESE	6.50E-02	1.30E-08	5.47E-02	1.50E-02	0.3
East of TA-53	3086	ESE	2.40E-02	4.90E-09	2.06E-02	3.80E-02	1.8
East of Airport	3883	E	1.70E-02	3.60E-09	1.51E-02	1.40E-02	0.9
West of Airport	2312	E	3.90E-02	7.70E-09	3.24E-02	1.56E-01	4.8
North Mesa	2356	NE	2.10E-02	4.40E-09	1.85E-02	3.00E-03	0.2
Sportsman's Club	5411	NE	5.60E-03	1.30E-09	5.47E-03	9.00E-03	1.6
R-Site Road East	4080	S	5.90E-03	1.30E-09	5.47E-03	4.00E-03	0.7
Two-Mile Mesa	3231	WSW	3.00E-03	8.10E-10	3.41E-03	4.00E-03	1.2
average							1.4

Table 2. Calculation assuming unit release from TA-21

From TA-21	dist (m)	dir	air conc pCi/m3	deposition pCi/cm2/s	soil conc pCi/g	measured pCi/g	ratio
West of TA-53	1060	WSW	2.10E-02	4.60E-09	1.93E-02	1.50E-02	0.8
East of TA-53	827	SE	1.50E-01	2.90E-08	1.22E-01	3.80E-02	0.3
East of Airport	1431	E	8.10E-02	1.60E-08	6.73E-02	1.40E-02	0.2
West of Airport	467	NNW	1.30E-01	2.60E-08	1.09E-01	1.56E-01	1.4
North Mesa	1854	NNW	1.20E-02	2.90E-09	1.22E-02	3.00E-03	0.2
Sportsman's Club	4388	NNE	6.30E-03	1.50E-09	6.31E-03	9.00E-03	1.4
R-Site Road East	4272	SSW	3.90E-03	9.10E-10	3.83E-03	4.00E-03	1.0
Two-Mile Mesa	5383	WSW	1.30E-03	3.90E-10	1.64E-03	4.00E-03	2.4
average							1.0

In conclusion, the estimated airborne release from Los Alamos is consistent with 1 Ci.