LA-UR- 11-01203

Approved for public release; distribution is unlimited.

Title:	Comments on the LAHDRA Report
Author(s):	Michael McNaughton, Jean Dewart, William Eisele, Philip Fresquez, David Fuehne, Andrew Green, Jeffrey Whicker
Intended for:	Health Physics

Los Alamos
NATIONAL LABORATORY
EST. 1943

Form 836 (7/06)

Comments on the LAHDRA Report

Michael McNaughton, Jean Dewart, William Eisele, Philip Fresquez, David Fuehne, Andrew Green, Jeffrey Whicker; February 17, 2011

We thank the team of the Los Alamos Historical Document and Retrieval Assessment, LAHDRA, for assembling an extensive and detailed set of documents describing historical operations at Los Alamos. A careful examination of these documents leads us to offer the following 19 clarifications of the LAHDRA report.

1. The LAHDRA report, page ES-11, states: "the LAHDRA team used the available release data as reported by LANL, and applied adjustments to account for effects such as loss of material in sample lines. The LAHDRA team performed no independent evaluation or reconstruction of LANL's reported releases." Similar words are on page 17-4. These "adjustments" are attributed to LANL by referring to specific email messages and documents by LANL staff members. However, these references are a small fraction of many LANL email messages and documents. There are extensive reports and data that contradict the "adjustments" selected by the LAHDRA team. Specific examples are discussed as follows.

2. The LAHDRA report, pages 17-33 to 17-36, discusses the use of the soil data obtained by Phil Fresquez to back-calculate plutonium releases and suggests that these should be corrected for weathering. On page 17-35 the LAHDRA report states that "LANL staff have asserted that plutonium soil concentrations are declining to levels indistinguishable from background." This is followed by a reference to Fresquez 1998. Phil Fresquez states that this is not a correct and complete statement. When the 1974 to 2009 record is studied at known plutonium-containing sites, there appears to be no significant decreasing trend. For plutonium at the locations sampled by Phil Fresquez, weathering is too small to be detected.

3. The LAHDRA report, in the discussion of mixed fission products (MFP) on page 17-16, says "MFP is a generic term, however; as of 1973, LANL was applying a definition for reporting purposes under which MPF releases were considered as four month decayed fission products at the time of release (Valentine, 1973). The 'primary biologically significant nuclides' were therefore ^{90}Sr and ^{137}Cs." The use by LAHDRA of the continuous tense, "was applying", suggests this was a continuing policy. On the contrary, these remarks are specific to this one memorandum. As stated in Attachment 2 of the memorandum, it is not applicable to short-lived nuclides such as ^{133}Xe, ^{41}Ar, ^{88}Kr, and its short-lived decay product, ^{88}Rb. Therefore, the LAHDRA adjustments to the 1969 data are incorrect, because these consisted of short-lived noble gases and their decay products (UHTREX 1969.) Furthermore, the line-loss corrections are not applicable to noble gases, and filter-burial corrections are not applicable to fission products measured by a sodium-iodide gamma detector (NIOSH 2004.) These errors need to be corrected.

4. The LAHDRA report, page 17-5, paragraph 3, discusses "a sample line loss correction factor equal to five" and states that "These factors were selected based on analyses performed by LANL staff." The reference is to Fuehne 2008, which is an email message from David Fuehne. David Fuehne states that this is not a correct interpretation of his email message. The factor of 5 applies only to a worst-case scenario for large diameter particles with poorly designed nozzles and sample line. In contrast, the LAHDRA report, page 17-5 line 9, says "No sample nozzle was likely used." This misinterpretation needs to be corrected. For the cases in the LAHDRA report, the correct factor is close to 1.

5. Sample line losses were calculated in LA-UR-09-4700 and were shown to be close to 1. The data reported in LAHDRA Tables 17-1, 17-2, and 17-5 need to be corrected accordingly.

6. The LAHDRA report, page 17-5, paragraph 4, states: "A filter dust loading and burial correction factor of 2.33 was also applied to plutonium release totals reported by LANL for 1948 through 1959 based on assessments performed by LANL staff (Fuehne 2008)." The reference is to an email message from David Fuehne. David Fuehne states that this is not a correct interpretation of his email message. The factor of 2.33 applies to the filters used at present, which are not optimal for front-face counting. This misinterpretation needs to be corrected.

7. In paragraphs 3 and 4 of page 17-5, the LAHDRA report states that factor-of-5 and factor-of-2.33 adjustments were applied to DP West Building 12 for the entire period of 1948 – 1975 "because Building 12 was not part of the HEPA filter exhaust system." This statement is made without explanation or references. It contrasts with the statements on page 4-27, page 17-5 paragraph 3, and page 21-A3 that "a single stage of high efficiency particulate air (HEPA) filters was added to the combined process exhaust system at DP West in 1959 (Maraman et al., 1975)." Maraman et al. repeatedly state that HEPA filters were installed on the primary containment during the 1950s (Maraman 1975 pages 472-3 and pages 475-6). On page 475 it says "In 1959, all gloveboxes had HEPA filters on exhausts." See also LA-9513-MS (1982.) These filter improvements are accompanied by an order of magnitude decrease in emissions between 1959 and 1961, as shown in Table 2 of Maraman 1975 and Table 17-1 of the LAHDRA report. Furthermore, the LAHDRA statement also appears contradictory to Hyatt's 1955 memorandum and several memoranda from Harry Jordan (Jordan 1955-6) which are focused on measuring and improving the filter systems. During 1955 and 1956, the conclusions are reinforced by the Health Division Leader (T.L. Shipman), the Industrial Hygiene Group Leader (Harry Schulte), and the CMR-7 Group Leader (James Lilienthal) (LAHDRA repository #199.) This misunderstanding regarding HEPA filters needs to be corrected.

8. Filter dust loading and burial were measured by several authors for the specific HV-70 filters used at the time. These measurements are reported on page 25 of LAHDRA repos. No. 189, in the H-Division Progress Report for Jan-March 1972 (LAHDRA Repos. No. 4992), and in two memoranda by the H-1 Group Leader, Dennis Vasilik: Vasilik 1976 and Vasilik 1978. The LAHDRA report ignores the previous measurements that specifically apply to the filters of the time. This omission needs to be corrected.

9. The LAHDRA report, pages 17-4 and 17-5, refers to the memoranda by Edwin C. Hyatt: Hyatt 1955a, Hyatt 1955b, and Hyatt 1956. Hyatt's work is relevant to several issues: the development and

installation of HEPA filters as discussed later, particle size as discussed later, and isokinetic sampling as follows. Regarding isokinetic sampling, the LAHDRA report (page 17-4) states: "After six months of sampling, results were compared, and correction factors were determined and applied to releases previously reported for 1948-1955." This is incorrect. The sampling was continued for only two months, and no correction factors were applied because the agreement between the old and new systems was excellent. These facts are clearly shown in Hyatt's memoranda as well as in the raw data from 1955 and 1956 that were provided to the LAHDRA team (LA-UR-09-4700.) The LAHDRA report, page 17-5, paragraph 1, acknowledges the agreement between the new and old systems with the statement: "when all the data from the test period is averaged, the two systems, on average, agree." The LAHDRA report adds disparaging comments, such as "individual measurements were found to differ by as much as a factor of 2900." Examination of the original data, which were provided to the LAHDRA team and are available from LANL, demonstrate that this was an individual anomaly, probably caused by a pump failure, and does not detract from the overall quality of the data, which are statistically robust with an overall uncertainty better than 10%. The LAHDRA report continues: "Since the new system was isokinetic, it is believed to be a better measurement, and the old system likely suffered from significant sampling errors and variability." On the contrary, the data demonstrate excellent agreement and overall errors less than 10%. Hyatt demonstrated that the correction factor is 1.0 ± 0.1. The LAHDRA correction factor of 5 needs to be replaced by a factor of 1, thus reducing the LAHDRA estimate by a factor of 5.

10. The LAHDRA report, page 17-5 paragraph 3, states that in 1959: "the particle size distribution likely shifts toward smaller particles." This sentence indirectly addresses the fact that the line loss correction factor of 5 obtained from Fuehne 2008 is for 10-micrometer particles. However, this sentence is directly contradicted by the particle-size measurements reported in Hyatt 1955a, which reported mass-median-aerodynamic-diameters (MMAD) of 0.38 to 1.3 micrometers, and also by Jordan and Black (1948) who stated that the MMAD was in the range 0.3 to 1 micrometer. These data are confirmed by Moss 1961.

11. LANL submitted comments on the draft-final LAHDRA report to CDC in the LANL report LA-UR-09-4700, dated July 17, 2009. Specifically, LANL presented evidence that the factors of 5 and 2.33 applied to the plutonium data are incorrect. The LAHDRA report needs to acknowledge the scientific evidence that contradicts their conclusions.

12. Table 17-1 on page 17-7 of the LAHDRA report is presented without adequate explanation. The LAHDRA report says the data were obtained from LANL data, and in previous drafts this table was obtained from Maraman 1975, Andrews 1973, or Valentine 1973. However, the final version does not have a clear relationship to any of these sources. If Table 17-1 is to be useful, scientists need to be able to reproduce the results. A detailed explanation is needed.

13. The draft-final LAHDRA report used the DOE handbook, US DOE 1994 to obtain an overall release fraction, ORF, of 0.001. The final report, page 17-9 and page 17-14, substitutes a value of 10%, obtained from Dahl and Johnson, 1977. Consequently, the release estimates for both uranium and radioactive lanthanum in the final report are 100 times larger than those in the draft-final report. It is important to evaluate the relative merits of the DOE handbook and the report by Dahl and Johnson, 1977. Other

evaluations of the radioactive lanthanum program (Jacobson 1995, Dummer 1996, Kraig 1997) should also be evaluated.

14. The LAHDRA report, page 17-9 and page 17-16, applies the sample line loss factor of 5 and the filter burial factor of 2.33 to both uranium and to mixed fission products. These factors do not apply to all particle diameters, tube sizes, configurations, and types of radiation. For example, according to page 17-16, correction factors for alpha particles have been used for the beta particles and gamma rays from mixed fission products. Explanations and corrections are needed.

15. The draft final report interpreted the term "mixed fission products" as a mixture of many fission products. The final LAHDRA report (pages 17-17, 21-A6, and 21-A9) substitutes two specific nuclides, cesium-137 and strontium-90, for "mixed fission products." As a result, the estimate has increased by a factor of 1000. It is a major error to equate the activity of short-lived nuclides with that of longer-lived nuclides. Although the fission yields for cesium-137 and strontium-90 are each about 6%, this fraction represents the number of atoms, not the activity; for a given number of atoms, the activity is inversely proportional to the half life. The most dramatic example is the reported emission of 416 Ci in 1969. This emission has been inflated by incorrectly applying line-loss factors where no sampling line was used and filter burial factors where no filters were used. As noted in NIOSH 2004 and UHTREX 1969, this emission consisted of noble gases, mostly Xe133 with smaller amount of Kr88 and its short-lived decay product, Rb88. The substitution of Cs137 and Sr90 for these noble gases is a major error. Mixed fission products are discussed more fully in Appendix A, below.

16. The LAHDRA report, page 21-A11, says "As far back as the 1940s, LANL staff were aware of the radon and thoron influence on plutonium air sampling results, and minimized their impact by allowing these daughters to decay before analyzing the filters." The word "minimized" should be replaced by "reduced" and "decay" should be qualified by the word "partially" because, at least during 1955 and 1956, day-shift filters were counted about 16 hours after they were collected and night-shift filters were counted a few hours after they were collected. In contrast, modern practice is to allow radon and thoron to decay for more than a week. To the best of our knowledge, radon corrections were not applied to the stack emissions. In this regard, the memorandum from J.F. Tribby (Tribby 1947) is interesting because it reveals an incomplete understanding of radon decay products at the time.

17. The LAHDRA report, page 17-26, says "This prioritization effort was intended to provide a first look," and page 17-35, presents "Suggestions for Improvement of the Current Analysis." We are implementing these suggestions (LANL 2011) and expect them to add significantly to our knowledge of historical releases. In particular, the results support the LANL estimates and contradict the LAHDRA estimates of Pu emissions.

18. The LAHDRA report, page 17-4, says: "In addition, there were other stacks beyond the four main stacks at Building 12 at DP West that were monitored; however, these data were not included in FEIS documentation, and were not compiled as part of the LAHDRA project." These data are listed in the handwritten notes supporting Andrews 1973 and Valentine 1973, and are much smaller than the releases from Building 12.

19. The LAHDRA report, pages ES-11 and 17-4, says that the LAHDRA team did not evaluate the LANL reports and data. However, they have selected some reports and ignored others. This selection constitutes a type of evaluation, and according to standard scientific procedures it should be accompanied by careful explanation. Furthermore, scientific ethics require that contradictory reports should be acknowledged, even if the team does not have the time or expertise to evaluate them. The most important issues that have been ignored are as follows.

- The factor-of-5 line loss correction is contradicted by Hyatt 1955a, Hyatt 1955b, Hyatt 1956, and LA-UR-09-4700.
- The 2.33 filter-burial and dust-loading factor is contradicted by Vasilik 1976, Vasilik 1978, and the reports listed in paragraph 6 of these comments.
- The 10% overall release fraction (ORL) is contradicted by US DOE 1994.
- The list of the isotopes in UHTREX stack emissions (UHTREX 1969) has been ignored.

Conclusion

We conclude by outlining the key issues.

The most significant radionuclide is plutonium-239, Pu-239. Most of the plutonium emissions were from D-Building in technical area 1 (TA-1) and also from DP West at technical area 21 (TA-21).

There are no definitive records of the releases from D Building at TA-1. The estimates on page 4-16 of the LAHDRA report seem reasonable: the median is 0.4 Ci and the 95th percentile is 1.05 Ci. They are also in good agreement with the data discussed on page 17-35 of the LAHDRA report (LANL 2011), and are consistent with the calculations reported in Miller 2008.

The best measurements of the releases from TA-21 are reported in Table 2 of the paper by Maraman, McNeese, and Stafford, 1975. These data are consistent with the estimates of Jordan 1958, Kennedy 1972, Andrews 1973, and Valentine 1973, though the Maraman data are the most complete (see the discussion on page 480.) These data are also confirmed by the data reported in LANL 2011.

The uncertainties associated with these data amount to a 60% uncertainty in either direction. The correction factor for filter burial is 1.4 to 1.6 (Vasilik 1976 and 1978.) On the other hand, the presence of the alpha-emitting decay products of radon-220 and radon-222 indicate a correction factor in the other direction. Furthermore, Maraman et al. assumed that the flow rate was equal to 200,000 cfm, which is the design capacity of the system. However, it is likely that the filters decreased the flow rate; if so, this would result in a smaller result for the amount of plutonium released.

The off-site impact of uranium is much smaller than that of plutonium because the distance to the receptors from the uranium sources was much larger than for plutonium; see Figure 20-3 on page 20-21 of the LAHDRA report.

Similarly, the off-site impact of radioactive lanthanum, RaLa, is much smaller because of the distance from source to receptor; see Kraig 1997.

The off-site impact of tritium is very small because almost all of the releases were of tritium gas, not tritium oxide, and the conversion of tritium gas to tritium oxide is slow (Steele 2002.)

The off-site impact of mixed fission products is very small because most of the activity consisted of short-lived radionuclides, especially noble gases. For details, see Appendix A, below.

The off-site impact of mixed activation products is very small because the releases consisted of short-lived radionuclides, mostly carbon-11, nitrogen-13, oxygen 15, and argon-41. These are well documented in the LANL Environmental Surveillance Reports and the Rad-NESHAP reports.

The off-site impact of radioiodine is small, as noted in the LAHDRA report; see Figure 17-9 on page 17-25.

The human dose impact of releases to water is small because surface water, storm water, alluvial groundwater, and intermediate perched groundwater are not used to supply drinking water.

References

Andrews, LL. Handwritten notes in LAHDRA repositories #1733 and #1813. Note: page 148 of #1733 and page 17 of #1813 are the same as page 18-3 of the LAHDRA report. 1973.

Dahl, David A and Johnson, Lamar J. Aerosolized Uranium and Beryllium from LASL Dynamic Experiments. Los Alamos report LA-UR-77-681; 1977.

Dummer JE, Taschner JC, Courtright CC. The Bayo Canyon Radioactive Lanthanum (RaLa) Program. Los Alamos Report LA-13044-H; 1996.

H-Division Progress Report for Jan-March 1972. LAHDRA Repos. 4992, page 1; 1972.

Hyatt EC. Total Activity Released from D.P. West Stacks. Memorandum to Dean D Meyer dated 26 October 1955. LAHDRA Repos. No. 0199 pp. 15-25 of 143; and No. 7099; also available as ERID #1710. 1955a.

Hyatt EC. Total Activity Released from DP West Stacks (1951 through September 1955). Memorandum to Dean D Meyer dated October 31, 1955. LAHDRA Repos. No. 0199 pp. 100-101; and No. 6513; and No. 6602; 1955b.

Hyatt EC. Total Activity Released from DP West Stacks, Bldg 12 from 1948 through 1955. Memorandum to Dean D Meyer dated January 3, 1956, LAHDRA Repos No. 6601; also available as ERID #5751. 1956.

Jacobson KW, Bossert JE, Letellier B, Stone GL. Modeling and Dose assessment of four high-explosive releases of radioactivity. Health Phys. Supplement to 68(6); 48-49; 1995.

Jordan 1955-6. Memoranda dated 1955 and 1956. LAHDRA repository 0199.pdf; 1955-6.

Jordan HS and Black RE. Evaluation of the air pollution problem resulting from discharge of a radioactive effluent. American Industrial Hygiene Association Journal 19, pages 20-25; 1958.

Kennedy WR and Purtymun WD. Plutonium and Strontium in Soil Near Technical Area 21, LANL report LA-4563, LAHDRA repos. No. 2141; 1971.

Kraig DH. Dose Reconstruction for Weapons Experiments Involving 140La at Los Alamos National Laboratory, 1944-1962. Health Physics vol. 73(4), pages 620-628; 1997.

LA-UR-09-4700. Comments on the draft final LAHDRA report; July 17, 2009. The cover letter from Craig Eberhart to Dr Charles Miller, reference EP2009-0367, was dated July 31, 2009. In addition to submission by US mail, these were also submitted by email to rsbinfo@cdc.gov on August 8, 2009. Further details are in LA-UR-09-5381, submitted with cover letter EP2009-0439 to rsbinfo@cdc.gov on September 1, 2009. A third document, LA-UR-09-07841, was prepared in November 2009 at the request of Tom Widner and submitted via the LANL LAHDRA office on December 2, 2009. A fourth document, LA-UR-10-00208, was submitted (together with the third document) to rsbinfo@cdc.gov on January 28, 2010.

LA-9513-MS. Garde, Raymond; Cox EJ; and Valentine, Allen M. Los Alamos DP West Plutonium Facility Decontamination Project 1978-1981. Los Alamos Report LA-9513-MS; 1982.

LAHDRA repository #199. Miscellaneous memoranda regarding the DP West exhaust systems from Harry Jordan, T.L. Shipman, and Harry F. Schulte, dated 1955 and 1956.

LANL 2011. Airborne Plutonium from Early Los Alamos Facilities, LA-UR-xxxx 2011.

Maraman WJ; McNeese WD; Stafford RG. Confinement facilities for handling plutonium. Health Phys 29: 469-80; 1975.

Miller G, et al. A Study of Early Los Alamos Internal Exposures to Plutonium, Radiat. Prot. Dosimetry vol. 130(4)503-509 (2008), doi:10.1093/rpd/ncn094 http://rpd.oxfordjournals.org/content/130/4/503.short ; 2008.

Moss, WD; Hyatt, EC; Schulte, HF. Particle Size Studies on Plutonium Aerosols. Health Physics Vol. 5, pp. 212-218; 1961.

NIOSH 2004. Technical Basis Document for the Los Alamos National Laboratory. ORAUT-TKBS-0010-2; 2004.

Steele CM and Mishima J. Oxidation of Tritium Gas Under Accident and Transport Conditions. LANL report LA-UR-02-3803; 18 June 2002.

Tribby 1947. Memorandum from J.F. Tribby to E.R. Jette, Airborne alpha radioactive contamination in U building. LAHDRA repository #197, pages 80-81. May 9, 1947.

UHTREX 1969. Isotopes Identified in UHTREX Stack Effluent and Log of Air Particle Activity Stack Releases, LAHDRA repository #5433. 1969.

US DOE 1994. Airborne Release Fractions/Rates and Respirable Fractions for Non-Reactor Nuclear Facilities, DOE-HDBK-3010-94. 1994.

Valentine AM. Memorandum from Allen Valentine dated November 9, 1973. Nuclide Inventory Data. LAHDRA repository #265 and #2106. These data are supported by handwritten notes in LAHDRA repositories #1733 and #1813, and also by the tables in repository #2750. 1973.

Vasilik DG. Depth of Burial Correction Factor for HV-70 Filter Media Used for Effluent Air Sampling of HEPA Filtered Plutonium Stacks. Memorandum to JE Drummer, H-1 Group Leader, May 27, 1976, Symbol H-1-DGV-18-76, LAHDRA Repos. 1019-1. 1976.

Vasilik DG. Depth of Burial Correction Factor for HV-70 Filter Media Used for Effluent Air Sampling of LASL Uranium Stacks. Memorandum to JE Drummer, H-1 Group Leader, August 24, 1978, Symbol H-1-DGV-15-78, LAHDRA Repos. 6107. 1978.

Appendix A: Mixed Fission Products

The LAHDRA report (page 17-17) states that mixed fission products were assumed to be primarily Sr90 and Cs137. The following paragraphs demonstrate that this assumption is incorrect.

1. The Chart of the Nuclides lists the "fission yield" for each isobar. The number listed is the percentage of fissions that produces each isobar. For example, for every U235 fission there are 0.058 nuclides with mass number $A = 90$ and 0.047 nuclides with mass number $A = 89$. Therefore, the number listed is proportional to the number of nuclei, N, not the activity, dN/dt.

2. The activity, dN/dt, is inversely proportional to the half life, as shown by the standard textbook decay equation: $dN/dt = -N \ln(2)/T$; (the variable t is time and the constant T is the half life.) For example, the half life of Sr89 is 50.5 days, and for Sr90 it is 29 years, so for the same number of nuclei, the Sr89 activity is 29*365/50.5 times the Sr90 activity.

3. In an operating reactor, the Sr89 activity will come to secular equilibrium with the fission rate. The time to reach secular equilibrium is usually assumed to be about 5 to 7 half lives. Therefore, after 250 to 350 days in an operating reactor, the Sr89 activity (in Bq) will be almost equal to 0.047 times the fission rate (in fissions per second.)

4. However, because of its much longer half life, the Sr90 activity will not reach secular equilibrium. After time t, the ratio of the Sr90 activity to the fission rate will be $(0.058)(\ln(2)*t/T)$ where T is the half life. If $t = 1.5$ years (a typical time) and $T = 29$ years, the activity is $(0.058)(\ln(2)*1.5/29) = 0.002$ of the fission rate. The ratio of Sr90/Sr89 = 0.002/0.047 = 0.04, which agrees with WASH1400 Table VI 3-1, page 19 of 481 of the following pdf file:
http://www.osti.gov/energycitations/servlets/purl/7134136-AEO6Lo/7134136.pdf

5. Similarly, the Cs137 activity will be $(0.062)(\ln(2)*1.5/30) = 0.002 = 0.2\%$ of the fission rate. This also agrees with WASH1400, which lists the Sr90 and Cs137 activities as 0.1% of the total fission-product activity. Therefore, Sr90 and Cs137 amount to a small fraction of mixed fission products.

6. The noble gases, krypton and xenon, are much more likely to be released than the others. For example, see the UNSCEAR-2000 report, http://www.unscear.org/docs/reports/annexc.pdf Annex C, Tables 31 through 39. Table 37 shows that the particulate activity amounts to less than 0.1% of the noble gas activity.

7. The Los Alamos UHTREX reactor was equipped with filters, charcoal traps, and molecular sieve to trap all types of fission products other than the noble gases, so almost all the releases were fission products, primarily Xe133, Xe135, Kr88, and its short-lived decay product Rb88. The nuclides released were measured with gamma spectrometry and are listed in LAHDRA repository 5433.pdf.