A Review of Historical and Current Emissions of Radioactive Materials from Los Alamos National Laboratory into the Air

By Bernd Franke[1]

Introduction

This report addresses questions with respect to the emissions of airborne radioactive materials, historical releases, unresolved questions regarding the current releases from Los Alamos National Laboratory (LANL).

Given the limited resources available, the review focused on questions of public concern that were raised in discussions with Concerned Citizens for Nuclear Safety (CCNS) and other members of the community:

- How does currently available data on historical emissions from LANL compare with results from other Department of Energy (DOE) facilities?
- Is it reasonable to require a dose reconstruction of historical releases and an examination of subsequent health impacts?
- What conclusions can be drawn from the limited data on historical environmental monitoring?
- What issues with respect to current emissions of radioactive materials are still unresolved?

Historical Airborne Emissions of Radioactive Materials at LANL

Since its inception during World War II in the year 1943, radioactive materials have been released into the air, water and soil as a result of LANL operations. An in-depth analysis of the effects of these releases on human health and the environment requires an accurate account of the historical emissions. The Los Alamos Historical Document Retrieval and Assessment (LAHDRA) by the Centers for Disease Control and Prevention (CDC) is the first step in this direction. The objective of LAHDRA is a systematic review of all available documents related to LANL operations and the identification of records that contribute information about releases of chemicals and radionuclides from the site between 1943 and the present.

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Plutonium Releases into the Air

The LAHDRA project is carried out by ENSR International, a global provider of environmental services to industry and government, and Shonka Research Associates, Inc. A summary report was published in February 2002 (ENSR, 2002). The LAHDRA report focuses on plutonium releases, which form the basis for the evaluation in this report. Figure 1 displays the release estimates of plutonium isotopes, the values are listed in Table 1.

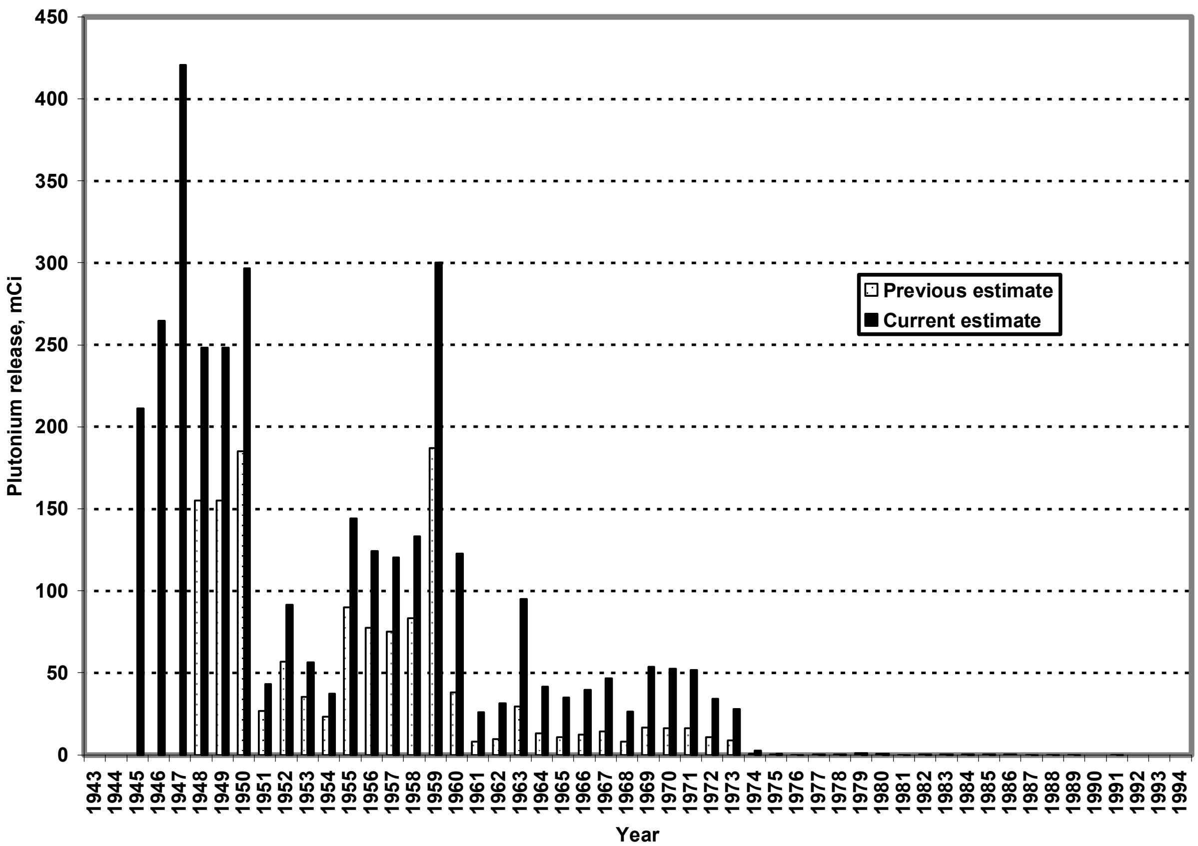

Figure 1 **Airborne releases of plutonium from LANL, 1945 – 1994 in millicuries per year (mCi/yr). Source: ESNR, 2002, p. 120**

Table 1 Airborne releases of plutonium from LANL, 1945 – 1994 in mCi/yr. Source: ESNR, 2002

Year	Previous estimate *mCi*	Current estimate *mCi*	Year	Previous estimate *mCi*	Current estimate *mCi*
1943			1970	16.42	52.65
1944			1971	16.11	51.66
1945		211.41	1972	10.69	34.28
1946		264.73	1973	8.7	27.88
1947		420.54	1974	0.79	2.55
1948	155	248.47	1975	0.25	0.79
1949	155	248.47	1976	0.07	0.07
1950	185	296.56	1977	0.13	0.13
1951	27	43.28	1978	0.11	0.11
1952	57.01	91.38	1979	1.09	1.09
1953	35.23	56.47	1980	0.75	0.75
1954	23.3	37.35	1981	0.06	0.06
1955	89.89	144.09	1982	0.11	0.11
1956	77.49	124.22	1983	0.11	0.11
1957	75.12	120.41	1984	0.14	0.14
1958	83.21	133.39	1985	0.21	0.21
1959	187.24	300.14	1986	0.21	0.21
1960	38.27	122.69	1987	0.07	0.07
1961	8.16	26.16	1988	0.07	0.07
1962	9.79	31.4	1989	0.05	0.05
1963	29.59	94.88	1990	0.03	0.03
1964	13.01	41.7	1991	0.04	0.04
1965	10.9	34.94	1992	0.01	0.01
1966	12.32	39.5	1993	0.01	0.01
1967	14.51	46.53	1994	0.01	0.01
1968	8.24	26.43			
1969	16.73	53.63	**Total**	**1,368**	**3,432**

The current LAHDRA estimate for the source term of 3.4 curies (Ci)[2] is more than twice the source term that ENSR estimated at an earlier date, while giving no further information as to when the estimate was made. Judging from the experience with source term estimates at other DOE facilities in which source term estimates were usually revised upwards, one can reasonably expect an upward revision for the LANL site as well.

[2] 1 Ci = 1 curie = 37,000,000,000 Bq (Becquerel) = 37,000,000,000 disintegrations per second.

The current estimate can be considered in several ways. One can compare the estimated releases with the fallout deposited on LANL property from atmospheric nuclear weapons tests. Based on data in the 1982 report of the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR,1982), the fallout of plutonium-239/240 from nuclear weapons tests in the northern hemisphere between 40°N and 50°N was about 1,600 picocuries[3] per square meter (pCi/m^2). Since LANL occupies 43 square miles, a total of 0.17 Ci of plutonium-239/240 is estimated to have been deposited on the LANL site from worldwide fallout using the UNSCEAR value. In other words, the current official estimate of the amount of plutonium emitted into the air by LANL is about 20 times the amount of plutonium that was deposited on the LANL site from nuclear weapons tests.

Another possibility to put the magnitude of plutonium emissions into perspective is to compare LANL's emission estimates with that of other DOE sites as shown in Table 2. The current estimate for LANL is about twice the official estimate for the Hanford site from the Hanford Environmental Dose Reconstruction Project (HEDR) for which the cumulative source term for plutonium-239/240 was estimated to be 1.78 Ci (Heeb, 1994). One should bear in mind that the Hanford site is much more remote than the LANL site; the closest offsite location is about 15 miles (24 km) east-southeast from the center of the Hanford 200 Area, whereas residential areas near LANL are less than a mile away from the source of historical emissions.

At the Rocky Flats site in Colorado, the official central estimate is 24.2 Ci, or about seven times the estimate for LANL (RAC, 1999). Whereas historical plutonium emissions were larger at Rocky Flats than at LANL, one should also bear in mind that the distance between the source of the emissions and the closest offsite area (about 5 miles) is far greater than at LANL. In addition, plutonium emissions were the only relevant source of radioactive materials at Rocky Flats, while at LANL other radioactive isotopes, such as tritium, uranium-235, uranium-238, iodine-131, cesium-137 and strontium-90, were emitted into the air as well. A careful accounting of the magnitude of historical releases of these materials into the air is still pending.

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[3] 1 pCi = 1 picocurie = 1×10^{-12} Ci = 0.037 Bq (Becquerel) = 0.037 disintegrations per second = 2.2 disintegrations per minute.

Table 2 Comparison of historical airborne releases of plutonium from three DOE facilities

DOE Site	Plutonium Source Term	Closest distance from source to offsite area, miles	Source
Hanford, WA	1.78 Ci	~ 15 miles	Heeb (1994)
Rocky Flats, CO	24.2 Ci	~ 5 miles	RAC (1999)
Los Alamos, NM	3.4 Ci	< 1 mile	ENSR (2002)

From the above, it is concluded that a careful reconstruction of historical releases and subsequent radiation exposures is necessary at LANL. Given the different circumstances and the lack of a final estimate for LANL, a precise evaluation of the potential impacts of LANL releases is not possible at this time.

It should be noted that dose reconstructions were performed at the Hanford and Rocky Flats sites, as well as at other DOE facilities, including the Nevada Test Site. The results for the estimates of the cancer incidence risk at various DOE sites were recently compared in an article by Reed, et al. (2003). The results are shown in Figure 2 for individuals defined as the "maximum receptor" (usually at the point of maximum impact and living entirely on locally produced food) and a "typical receptor" (usually a representative individual). The largest risk was calculated for iodine-131 releases from the Hanford site. The upper bound estimate of cancer incident risk for the maximum receptor near Rocky Flats is about 1 in 10,000, mainly due to airborne emissions of plutonium-239/240. Given the fact that additional radionuclides and toxic substances were also released from the LANL site along with plutonium-239/240, and because there are significant uncertainties in estimating releases and subsequent exposures due to site-specific circumstances, it is recommended to conduct a similar in-depth evaluation for the LANL site as well.

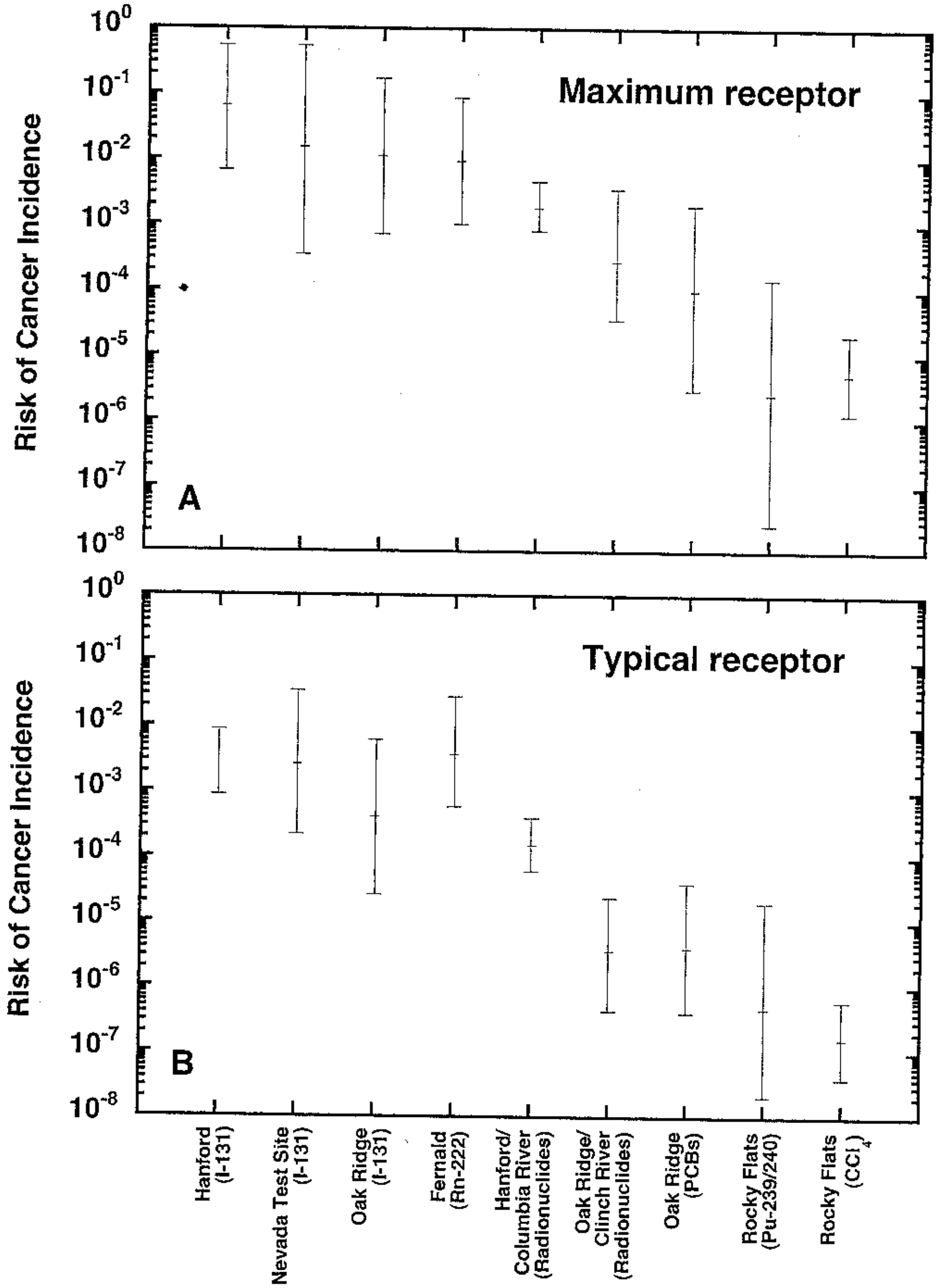

Figure 2 **Excess risk of cancer incidence for representative individuals due to contaminant releases from the indicated sites for (A) a maximally exposed receptor and (B) a typical receptor, vertical lines represent 90% or 95% confidence intervals (details in Reed, et al., 2003)**

Review of Selected Historical Environmental Monitoring Data for Airborne Radionuclides

The ENSR summary report contains data on concentrations of plutonium-238 and plutonium-239/240 in ambient air at various locations both onsite and offsite of LANL property in the year 1972. The reported data for annual averages is plotted in Figure 3. The highest concentrations were reported for onsite locations at Technical Area 3 (TA-3) and TA-6. At TA-6, the combined plutonium-238 and plutonium-239/240 activity was around 0.2 femtocuries[4] per cubic meter (fCi/m^3). The total plutonium emission from LANL in 1972 was estimated by ESNR to be 0.034 Ci compared to the maximum in 1947 of 0.42 Ci. Therefore, it can thus be expected that air concentrations were much larger in past years.

The expected concentration from weapons testing fallout for the year 1972 (0.04 fCi/m^3) is indicated in Figure 3, using the data that was reported for Denver in the Rocky Flats dose reconstruction (RAC, 1999). That data is shown in Figure 4 with the solid black line indicating the worldwide weapons testing fallout.

4 1 fCi = 1 femtocurie = 1 x 10^{-15} Curie.

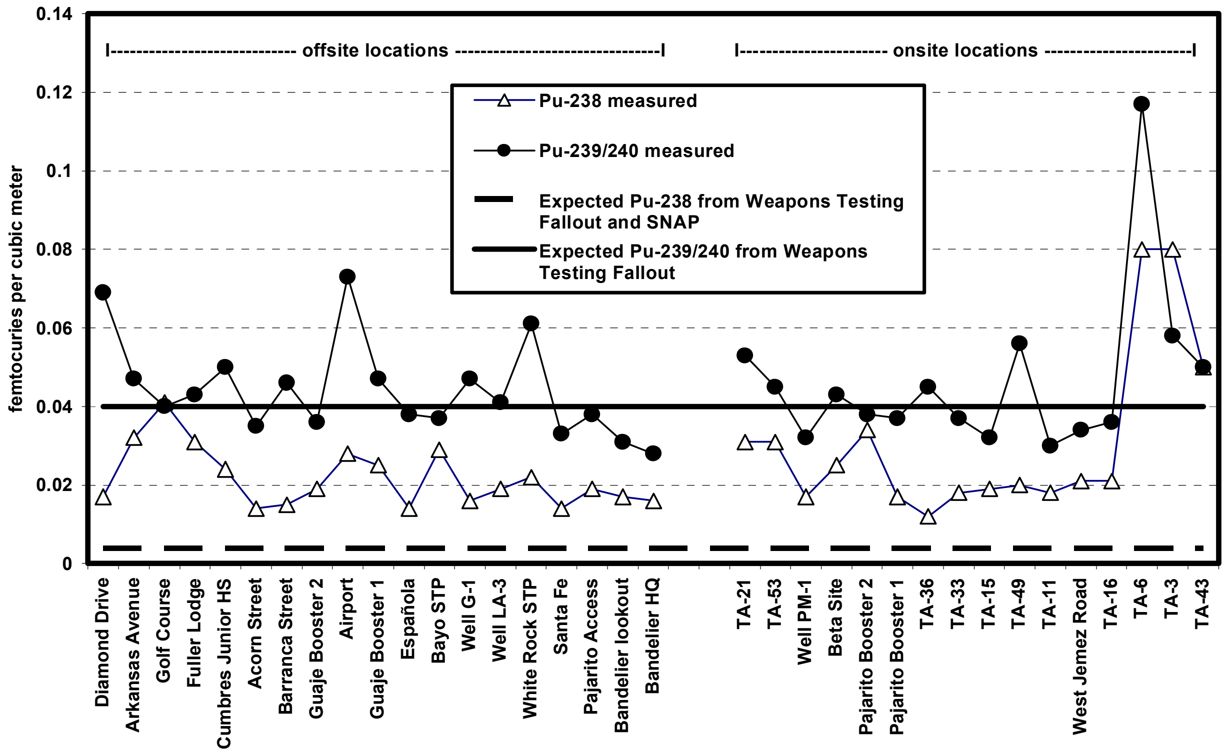

Figure 3 Activity of plutonium-238 and plutonium-239/240 in ambient air in fCi/m^3 reported by LANL for the year 1972. Source: ESNR, 2002

The expected amount of plutonium-238 can be derived from the ratio of the respective inventories in the atmosphere over the northern hemisphere. A total of 350,000 Ci of plutonium-239/240 was produced by worldwide atmospheric nuclear tests (UNSCEAR, 1982). Of this amount, about 4,000 Ci were still present in 1974 (Eisenbud, 1987). Of that, less than 80%, or about 3,000 Ci, was contained in the northern hemisphere. Hence, in 1972, the inventory in the stratosphere over the northern hemisphere was about 1% of the total production by atmospheric nuclear tests. Atmospheric nuclear tests produced much less plutonium-238, or about 9,000 Ci, of which 1% (or about 90 Ci) was present in the stratosphere over the northern hemisphere in 1972.

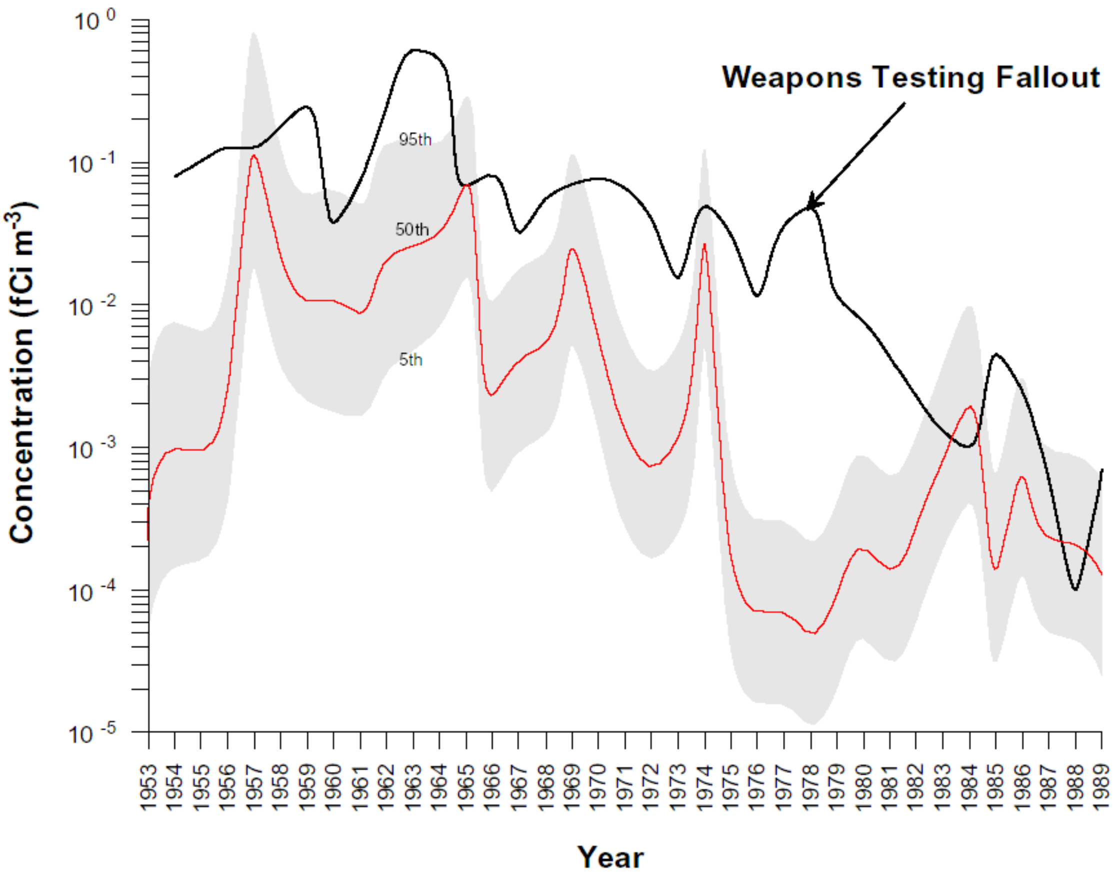

Figure 4. **Estimated concentrations of plutonium-239/240 in air at Rocky Flats, fCi/m^3. The values represent estimates for the location with the highest concentrations outside the current buffer zone (east of the plant boundary along Indiana Street). Source: RAC, 1999**

The major inventory of plutonium-238 in the atmosphere resulted from the re-entry of SNAP 9-A, a navigational satellite that was launched on April 21, 1964 and carried a radioisotope power generator. The satellite failed to reach orbital velocity and reentered the atmosphere over the Indian Ocean. The inventory of the source was about 17,000 Ci. In 1972, the inventory in the stratosphere over the northern hemisphere was about 300 Ci.

At first approximation, the ratio of concentrations in the atmosphere should be proportional to the stratospheric inventory. For the year 1972 in the northern hemisphere, the (plutonium-238) / (plutonium-239/240) ratio can thus be calculated (300 Ci) / (3,000 Ci) = 0.1. However, because the plutonium-239/240 air concentration from weapons testing fallout at LANL in 1972 was estimated to be 0.04 fCi/m^3, the plutonium-238 air concentration should have been 0.004 fCi/m^3.

While the reported concentrations of plutonium-239/240 at most offsite locations that were monitored by LANL are in the expected range, the same cannot be stated for the reported concentrations of plutonium-238. At all onsite and offsite locations, the reported concentrations exceed the expected concentrations by a factor between four and 20. This is also evident in the comparison of radionuclide ratios for (plutonium-238) / (plutonium-239/240). The ratios range from 0.2 to 1.4, rather than fluctuating around the value of 0.1 as one would expect, as shown in Figure 5.

This finding is quite unexpected and may be explained by major releases of plutonium-238 from LANL operations in the year 1972. The impact of LANL is indicated by the elevated concentrations at TA-3 and also in the elevated isotopic ratio. The ratio of ~1.0 for the offsite location at the golf course may indeed indicate a source upwind from that location. On the other hand, if the elevated plutonium-238 concentration was due solely to contributions from LANL, one would expect a sharper decline with distance from the site. The issue of plutonium-238 concentrations in ambient air is indeed puzzling, and a careful review of the quality of the data for 1972, as well as of data for other years, is clearly warranted.

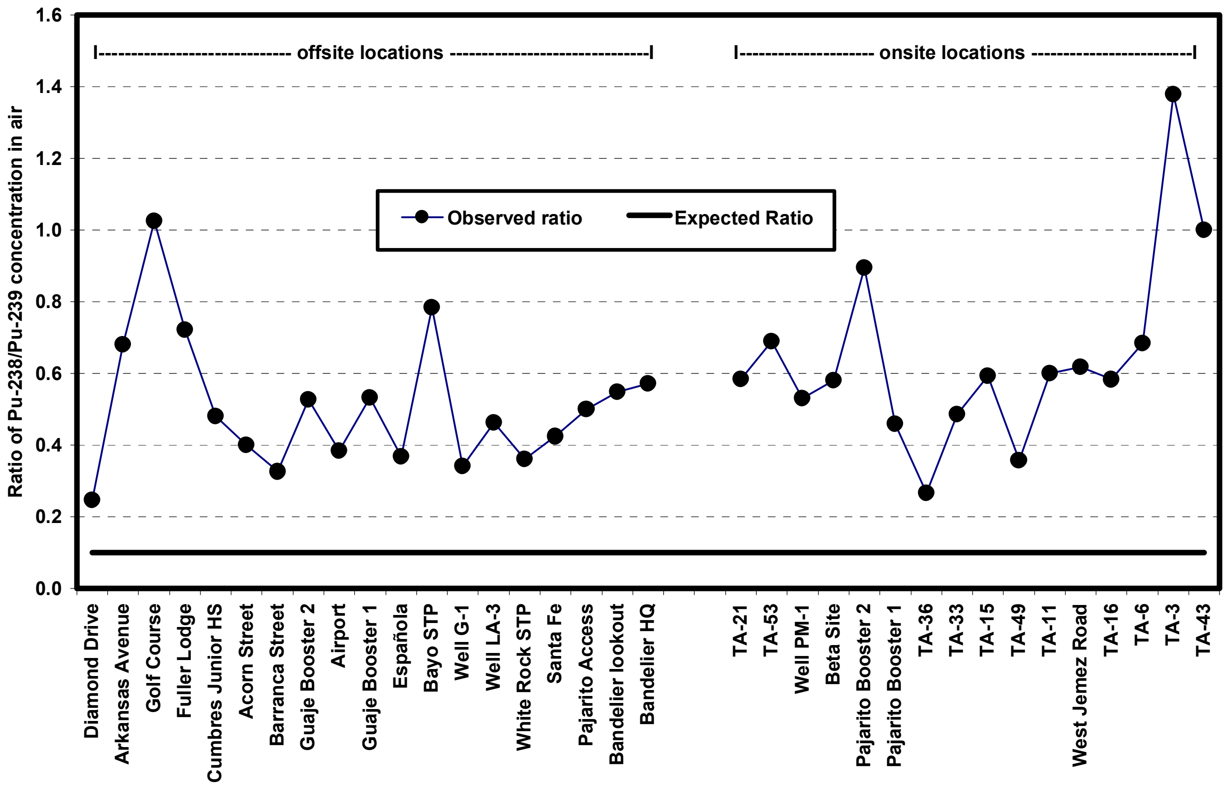

Figure 5 **Ratio of plutonium-238 and plutonium-239/240 activity in ambient air for the year 1972 at LANL**

Current Airborne Emissions of Radioactive Materials at LANL

This section of the report deals with two issues:

- Compliance requirements of the Clean Air Act, 40 CFR 61, Subpart H, and
- Radiation exposures to members of the public due to transient receptors.

Compliance with the Clean Air Act, 40 CFR 61, Subpart H

On January 21, 1997, DOE, former LANL director Siegfried S. Hecker, and CCNS reached an agreement to settle a citizens' suit concerning the status of LANL's compliance with the Clean Air Act (CAA), 40 CFR 61, Subpart H. Subpart H regulates releases of radioactivity to the atmosphere from DOE facilities. It was agreed in the settlement of the case and ordered by the U.S. District Court ordered that the Risk Assessment Corporation (RAC) would conduct a series of technical audits to evaluate LANL's compliance status as the Independent Technical Audit Team (ITAT). The first of these audits addressed whether LANL was in compliance in

1996 (Weber, et al., 1999); the second addressed LANL's compliance in 1999 (Aanenson, et al., 2000); and the third addressed LANL's compliance in 2001 (Aanenson, et al., 2002).

The Court also ordered that the audits be monitored by an independent monitoring team of the Institute for Energy and Environmental Research (IEER), Takoma Park, MD. The author was part of IEER's monitoring team; the monitoring report for the third audit was published in 2002 (Makhijani and Franke, 2002).

The ITAT concluded that LANL was in compliance with the 10 millirem[5] per year (mrem/yr) dose limit set forth in 40 CFR 61, Subpart H for the year 2001. The ITAT also found LANL to be in compliance with all other requirements of Subpart H and related Appendices. Further, the ITAT did not find any substantive technical deficiencies in LANL's compliance program. The ITAT did make some recommendations for "continued improvement" without finding that any of the areas in which these improvements were desirable constituted a substantive technical deficiency or a violation of Subpart H.

The report of the IEER monitoring team concluded:

> *IEER is in general agreement with only one of these overall conclusions of the ITAT. Despite the uncertainties and the technical deficiencies, as well as the essential lack of compliance in one area, IEER is in agreement with the ITAT regarding the 10 mrem/year dose limit compliance. This is because the maximum estimated dose is so much below 10 mrem per year (in part due to the fact that the main source of emissions, the Los Alamos Neutron Science Center (LANSCE), is not in full operation) that it is highly unlikely that the dose limit of 10 mrem per year was exceeded.*
>
> *In monitoring the audit and reviewing the final report, IEER has concluded that the ITAT should have called out four substantive technical deficiencies:*
> 1.) *a lack of quality assurance of the data on radionuclide usage supplied by the facilities to the Meteorology and Air Quality Group (MAQ),*
> 2.) *the problem of detecting radiologically elevated concentrations of plutonium-238 in samples in some cases,*
> 3.) *the need to provide continuous monitoring of airborne emissions from TA-54 waste characterization activities, and*
> 4.) *the significant uncertainties in the coverage of AIRNET stations with respect to Los Alamos North Mesa residences that justify an additional sampling station that has not been installed.*

[5] A millrem (mrem) is a measure of radiation dose. An exposure of 10 mrem/yr over 50 years would result in an increased cancer risk to an average individual of one in 2,600, using EPA's risk factors (EPA, 1994).

> *In relation to the first of these substantive technical deficiencies, IEER has also concluded that the ITAT should have found LANL to be in substantive breach of its compliance obligations under the Subpart H and related requirements under the Clean Air Act. As a result IEER finds that the main findings of the ITAT that LANL is in compliance with Subpart H and that the compliance program of LANL has no substantive technical deficiencies to be in error.*

A detailed description of the technical merits of IEER's conclusions can be found in Makhijani and Franke, 2002.

The Problem of Discontinuous Releases and Transient Receptors

One of the major problems with the compliance definition in Subpart H is that the compliance limit of 10 mrem/yr effective dose equivalent (EDE) only applies to a fixed location (a residence, school, business or office). In 2001, the closest "receptor," as these are called, was identified to be near the East Gate. This narrow definition poses a problem for the LANL site because of the following reasons:

- The LANL property is not totally fenced in as are other DOE sites (e.g., Savannah River Site).
- There are many point sources in proximity to access roads that can be frequented by members of the public.
- Many sources of radionuclide emissions are short-term and discontinuous in nature. For example, experiments conducted in laboratories can result in short-term airborne releases in batch form as compared to continuous releases from production facilities.

It is well established that past releases from several monitored stacks of the Chemistry and Metallurgy Research (CMR) building have indeed been discontinuous in nature, as a large portion of the total releases for one year occurred in less than a day. Given that the nature of LANL operations is more experimental than production-oriented, short-term releases can also be expected from at least some unmonitored point sources. In addition, there are a number of non-point sources, such as waste disposal sites, from which radioactive materials on the surface can be suspended into the air by gusty winds or fires.

This can be demonstrated by the TA-21 East area, which is suspected to contain waste materials from historical operations. Releases from a diffuse source, such as a waste site, are most likely to occur during high wind speeds (>10 miles per hour (mph)) when material can be suspended readily into air. Figure 6 shows the distributions of observations (averaged over 15 minutes) of the wind speed at the East Gate NEWNET station in August 2002 (Franke, 2002). High wind speed situations are more likely when the wind is blowing from the south-southeast. With regard to diffuse sources at TA-21 East, the wind speed/direction pattern suggests that the greatest impact to residents from such a source is likely at the North Mesa residences.

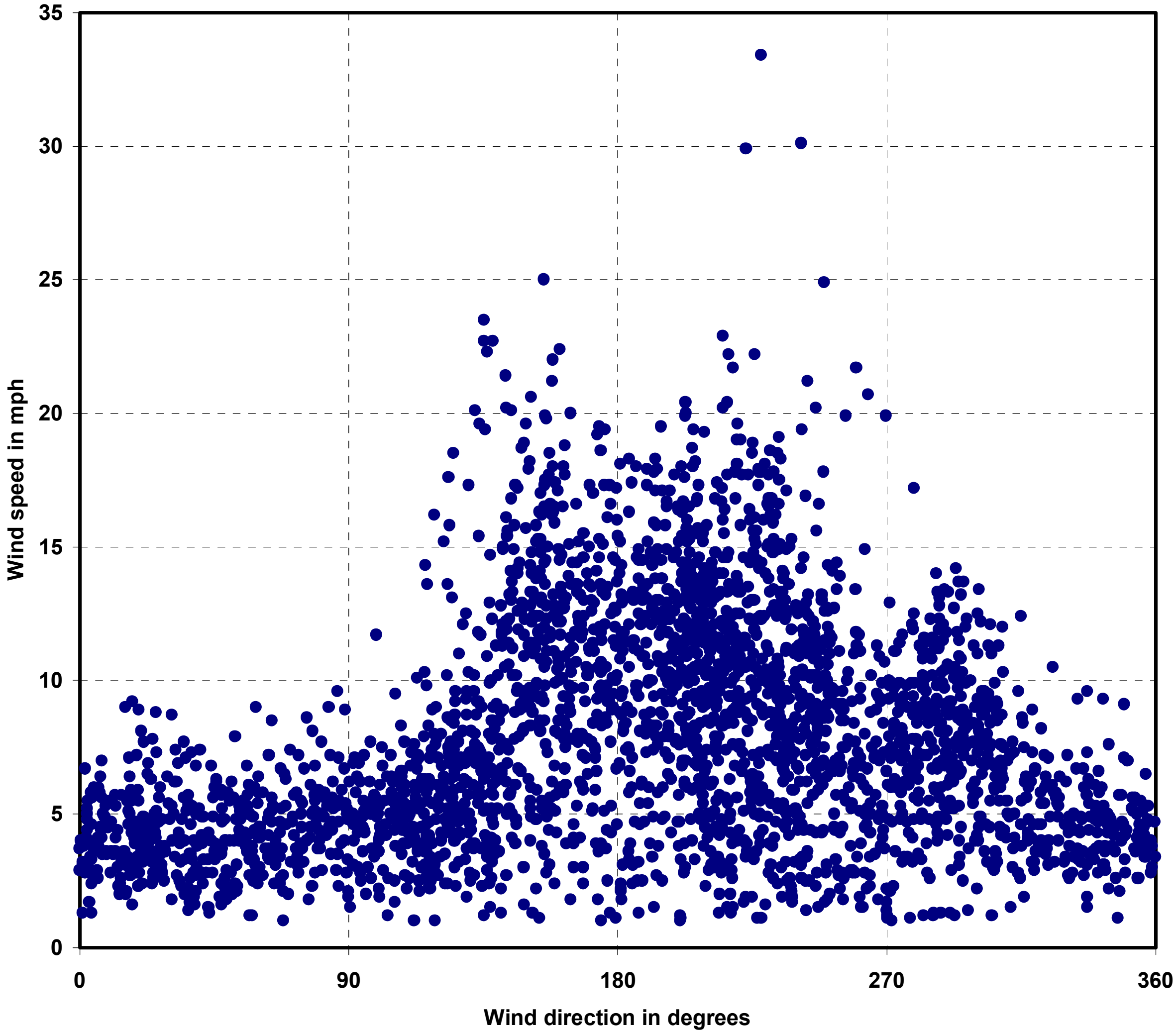

Figure 6 **Wind speed as a function of wind direction at East Gate NEWNET station, August 2002 (Franke, 2002)**

It is very difficult to determine the time-release function of the source term for diffuse sources with reasonable accuracy. This situation raises several questions:

- How large must a short-term release of radioactive material be in order to result in radiation exposures that exceed the 10 mrem/yr standard?
- Suppose that such a release goes undetected by stack monitoring, for example, an emission from an unmonitored source or a non-point source, such as a waste site. Would such a release be detected by the AIRNET monitoring network?

In answering the first question, dispersion calculations were performed for the following reference situation:

- A short-term release (assuming about one minute) from a source at an effective release height of 10 m (30 ft);
- An adult individual located downwind in the center of the plume where the highest concentration can be expected (plume centerline) at a distance of 50 m (150 ft.), at a normal breathing rate (about 1 cubic meter an hour (m^3/h)); and
- A release of plutonium-238 or plutonium-239/240 (medium solubility, class M) with a dose factor of 0.41 mrem/pCi (effective dose committed over a period of 50 years as a result of a single intake).

The results are shown for various atmospheric stability classes[6] for a low windspeed of 1 meter per second (m/s), which is equivalent to about 2.2 mph. The atmospheric dispersion coefficients that were used were based on dispersion experiments in Germany (Jülich and Karlsruhe). The data suggests that as little as 7.5 microcuries[7] (µCi) of plutonium would be sufficient to result in a dose that would be deemed noncompliant if the person were located at a residence or business. Furthermore, it is important to point out that the total airborne plutonium emissions from sampled stacks at LANL in 1995f were about 50 µCi (LANL, 1999). Total releases from LANL were larger because of the contribution from unmonitored point and area sources. In other words, a fraction of the reported plutonium releases from LANL, if released over a period of minutes, can be sufficient to result in an effective inhalation dose of 10 mrem if a person (e.g., a visitor passing by) happens to be present downwind near the location of the release.

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[6] Stability classes describe the dispersion of a plume. The range is from class 1 (unstable; wide plume) to class 6 (very stable, narrow plume).
[7] 1 µCi = 1 microcurie = 1 x 10^{-6} Curie.

Table 3. Airborne emissions of plutonium that would result in an effective inhalation dose of 10 mrem for an adult individual who is present at a distance of 50 m at plume centerline during a short-term release from a source height of 10 m

Stability class	Wind speed m/s	Number of experimental data points	Minimum amount of activity, µCi	Median amount of activity, µCi
1	1	5	30	150
2	1	12	48	330
3	1	11	14	210
4	1	14	23	86
5	1	8	7.5	140
6	1	1		140

Is it possible to properly monitor short-term releases from unmonitored sources, such as waste sites? In such a case, radioactive material would be transported to locations where exposure of the general public could occur. Thus, the AIRNET stations would be the only way to detect the releases. Depending on weather conditions, the AIRNET stations could be located in the plume centerline or could be bypassed by the plume. A careful analysis of the sensitivity of the AIRNET system would therefore assist in evaluating whether significant exposures of short-term releases could go undetected.

Given a unit release from a source in specific weather conditions, the sensitivity of the AIRNET system can be characterized by the ratio of (maximum potential dose) / (maximum detected dose) which can be expressed as

$$\frac{D_{max}}{D_{max,\ AIRNET}}$$

where

D_{max} = effective dose received by an individual present on generally accessible locations

$D_{max,\ AIRNET}$ = effective dose detected at the maximally exposed AIRNET station

Repeating the calculations for all weather conditions allows for calculation of the conditional cumulative frequency of the ratios for a given short-term emission. If the ratio (maximum potential dose) / (maximum detected dose) is close to one, this would mean that at least one

AIRNET station is close to the maximum exposure area accessible to the general public. However, if there is a significant probability that the ratio is large, care should be exercised in converting results of AIRNET measurements into dose. A detailed evaluation is recommended because of the chance that effective doses in the one to 10 mrem range could go underreported.

It may be beneficial to proceed by evaluating one set of facilities, such as the CMR building at TA-3, Building 29, and to proceed with other sources based on the outcome of such an evaluation. This issue should be further addressed in an in-depth review.

Conclusions and Recommendations

This report identified several issues pertaining to historical and current emissions of airborne radioactive materials at LANL. The primary conclusions and recommendations are:

- Based on current estimates, the historical plutonium emissions into the air from LANL of 3.4 Ci is about 20 times the amount of plutonium that was deposited onto the LANL site from worldwide atmospheric nuclear weapons tests.
- The reported concentration of plutonium-238 in air in the vicinity of LANL in the year 1972 was a factor of four and 20 higher than expected from other sources.
- The current information on historical emissions warrants a careful reconstruction of radiation doses to members of the public and the evaluation of the associated health risks.
- Visitors to the LANL site could receive an effective dose in excess of 10 mrem/yr from plutonium emissions under unfavorable circumstances (e.g., short-term emissions), even though the emissions would be reported to be in formal compliance with 40 CFR 61, Subpart H of the CAA.
- The suitability of AIRNET to detect releases from unmonitored point or diffuse sources, such as waste sites, needs to be carefully analyzed.

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