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Airborne Plutonium from Early Los Alamos Facilities

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Abstract

We have calculated the airborne plutonium emissions from the two earliest Los Alamos facilities: the original plutonium facility used during the World-War-II Manhattan Project known as D-Building, and its replacement known as DP or D-primed. For the DP facility our calculations are in substantial agreement with previous estimates by Los Alamos scientists, and for D-Building they are in substantial agreement with the estimates in the report by the Los Alamos Historical Document Retrieval and Assessment (LAHDRA) project.

Introduction

There is widespread interest in the historical releases from the earliest nuclear facilities and the resulting doses to workers and the public. For example, the Health Physics Journal recently devoted an entire issue to the Mayak Worker Dosimetry Study (Mayak 2007). Plutonium doses to early Los Alamos workers have been calculated by Miller (2008), Voelz (1997), Campbell 1973, and McInroy (1979 and 1995).

The authors of the Los Alamos Cancer Rate Study (Athas and Key 1993) referring to the first few decades of the Los Alamos laboratory, lamented: "The extent to which county residents were exposed to ionizing radiation beyond background levels during these time periods is not known." Such information impacts both dosimetry and epidemiology, which are especially important for the first nuclear facilities such as Los Alamos.

In the field of radiation protection, "conservative" or upper-bound estimates of releases and doses are sometimes acceptable. However, in sciences such as epidemiology, accuracy is essential. The following work is intended to provide the most accurate estimates of historical releases that are possible, given the limited information available.

1. The first Plutonium Facilities

The first plutonium facility was established in technical area #1 (TA-1) of Los Alamos in 1943, during the Second World War. The buildings were constructed hurriedly to avoid delaying the wartime scientific and engineering efforts, and they were not built to modern standards. Consequently, emissions were larger than those from modern facilities.

Compared with modern facilities, there are several unusual aspects of the first plutonium-processing building, known as D-Building. (See Figures 1 and 2.)

- The building was constructed of wood.
- The ventilation stacks were relatively short, ending approximately 10 m above ground level, and they were equipped with rain caps.
- Initially, there were no filters; when filters were installed, they were of relatively low efficiency.

- The stack emissions were not monitored.
- The nearest residence was 200 meters from the facility.

Neither the stacks nor the ambient air were monitored. Consequently, very little is known about the airborne plutonium emissions at Los Alamos during the earliest years from 1943 to 1945.

In late 1945, after the war, the main plutonium-processing operations were moved to a new facility known as DP. DP is sometimes referred to as D-primed because it was a replacement for D-Building. DP is also sometimes referred to as Technical Area 21 or TA-21.

Though still not up to modern standards, the DP facilities at TA-21 included some improvements compared with the original D-Building at TA-1.

- The main buildings were of concrete.
- The main ventilation stacks (TA-21-12, see Figure 3) were 17.86 m high with an exit velocity of 16 m/s.
- The filter efficiencies gradually improved, until high-efficiency filters were eventually developed in the 1950s.
- The stack emissions were monitored, though some of the earlier records are incomplete or missing.
- The nearest residence, built in 1957, was 480 m from the stacks.

Over the years, the facilities improved. The original D-Building continued to be used for plutonium research until it was replaced by the Chemistry and Metallurgy Research (CMR) building in 1953. D-Building was demolished in 1954. During the 1970s, the DP facility was replaced by the present Los Alamos plutonium facility, known as TA-55. The DP buildings at TA-21 are presently being demolished.

The airborne plutonium emissions from TA-21-12 were summarized by Maraman et al., (1975.) Data have not been found for 1945 through 1947, and many of the earlier data are incomplete and uncertain. The efficiency of the filters and the quality of the data improved during the 1960s and early 1970s, until the completion of the modern plutonium facility at TA-55.

The following data provide independent verification of the airborne plutonium emissions from the Los Alamos DP facility, TA-21-12. Furthermore, we present the first measurement of the airborne plutonium emissions from the original D Building in the original technical area, TA-1.

2. Method

The airborne emissions were not monitored at the time, so we calculate the emissions using the plutonium concentrations deposited on the ground near the sources. Plutonium concentrations are significantly above the global-fallout background in the soil surrounding the major sources, TA-1 and TA-21, and the following paragraphs describe how we used these data to calculate the emissions.

In principle, we use a standard air-dispersion model, CAP88 (EPA 1988) to calculate the expected deposition that would result from a nominal release of plutonium from each of the sources, and then use a statistical "maximum-likelihood" method (Bevington 1969) to find the release that corresponds most closely to the data.

The soil deposition data vary because of hot particles, soil disturbance, and weathering. These variations are discussed in later sections. Our general approach is to use data from many locations with various soil conditions and a large range of distances and directions. The variance of the final result is an indication of the magnitude of the uncertainties.

The objective is to find the best estimate of the emissions from two sources, as follows. Assume S_1 is the amount of plutonium emitted from TA-1 and S_2 is the amount emitted from TA-21. There are many soil locations, and at each location there is a measured deposition, d_i, and a calculated deposition, D_i.

At each location, the calculated deposition D_i is related to the emissions S_1 and S_2 by the equation

$$D_i = c_{i1} S_1 + c_{i2} S_2$$

where the coefficients, c_{ij}, are calculated by the air-dispersion program, CAP88.

According to standard maximum-likelihood methods (Bevington 1969) the best estimates for S_1 and S_2 are calculated by minimizing the function

$$f(S_1,S_2) = \chi^2 = \Sigma\, (c_{i1}S_1 + c_{i2}S_2 - d_i)^2 / \sigma_i^2$$

where σ_i is the combined uncertainty of the calculations and the measurements.

To find the minimum, find the partial derivatives with respect to S_1 and S_2 and set them equal to zero.

$$\partial f(S_1,S_2)/\partial S_1 = 2\Sigma\, c_{i1}(c_{i1}S_1 + c_{i2}S_2 - d_i)/\sigma_i^2 = 2[\Sigma\, c_{i1}^2 S_1/\sigma_i^2 + \Sigma\, c_{i1}c_{i2}S_2/\sigma_i^2 - \Sigma\, c_{i1}\, d_i/\sigma_i^2] = 0$$

$$\partial f(S_1,S_2)/\partial S_2 = 2\Sigma\, c_{i2}(c_{i1}S_1 + c_{i2}S_2 - d_i)/\sigma_i^2 = 2[\Sigma\, c_{i2}c_{i1}S_1/\sigma_i^2 + \Sigma\, c_{i2}^2 S_2/\sigma_i^2 - \Sigma\, c_{i2}\, d_i/\sigma_i^2] = 0$$

Then solve for S_1 and S_2:

$$S_1 = [(\Sigma\, c_{i2}^2/\sigma_i^2)(\Sigma\, c_{i1}\, d_i/\sigma_i^2) - (\Sigma\, c_{i1}c_{i2}/\sigma_i^2)(\Sigma\, c_{i2}\, d_i/\sigma_i^2)] / [(\Sigma\, c_{i1}^2/\sigma_i^2)(\Sigma\, c_{i2}^2/\sigma_i^2) - (\Sigma\, c_{i1}c_{i2}/\sigma_i^2)^2]$$

$$S_2 = [(\Sigma\, c_{i1}^2/\sigma_i^2)(\Sigma\, c_{i2}\, d_i/\sigma_i^2) - (\Sigma\, c_{i1}c_{i2}/\sigma_i^2)(\Sigma\, c_{i1}\, d_i/\sigma_i^2)] / [(\Sigma\, c_{i1}^2/\sigma_i^2)(\Sigma\, c_{i2}^2/\sigma_i^2) - (\Sigma\, c_{i1}c_{i2}/\sigma_i^2)^2]$$

Thus, in order to calculate the sources, S_1 and S_2, we need the set of soil data, d_i, the coefficients for source #1, c_{1i}, and the coefficients for source #2, c_{2i}.

3. Calculating the coefficients

We calculate the coefficients using the standard EPA program, CAP88 (EPA 1988.) The input parameters for CAP88 are as follows.

For each location, we require the distance and direction from the source to the location of the soil measurement. These are listed in Table 1.

The meteorological data were obtained by averaging 16 years of current data. We assume that this average is similar to the historical meteorological data.

The TA-1 stacks were 10 m high. The stacks were equipped with rain caps so we assume no plume rise. The TA-21 stacks were 17.86 m high. There were four stacks, each with a diameter of 1.07 m and an exit velocity of 16 m/s. The agricultural data required for CAP88 do not affect the deposition and are not relevant. We used the CAP88 default deposition velocity: 1.8 mm/s.

CAP88 calculates all results on an annual basis but lists the deposition in units of $pCi \cdot cm^{-2} \cdot s^{-1}$, so we multiply by 3.15×10^7 s (which is the number of seconds in a year) to get the total deposition in units of $pCi \cdot cm^{-2}$. To compare with results measured in pCi/g, we divided by the sampling depth of 5.0 cm and by the soil density of 1.5 g/cm^3. The coefficients then have the units of pCi/g of soil per Ci released.

4. The soil data

Ideally, the soil data should satisfy the following conditions.

- The soil should be minimally affected by other sources of contamination, such as spills, runoff, or discharges of contaminated water.
- The soil should have been subjected to minimal disturbance between deposition and measurement.
- The samples should be obtained using the ASTM protocol from a known surface area and to a depth of 50 mm (ASTM C998-90).

Suitable soil samples obtained with the ASTM protocol are described in Fresquez 1996 (LA-13149-MS), in Chapter 7 of the Los Alamos Environmental Surveillance reports, and in Ahlquist 1977 (LA-6887).

Many of the locations have been sampled repeatedly and the results vary considerably. However, over the course of 35 years of sampling, at most sites there has been no general downward trend; this indicates minimal movement of plutonium either laterally or vertically. The variability arises from the presence of discrete "hot particles" as follows.

As an example, consider a 1-µm Pu-239 particle, which has an activity of about 0.1 pCi. If a single 0.1-pCi particle is contained in a 2-gram aliquot the concentration will be reported by the analytical laboratory as 0.05 pCi/g, in addition to the background from global fallout, 0.015 pCi/g. Most samples contain zero or one hot particle. The resulting concentrations are 0.015 pCi/g for a sample with zero hot particles, or (0.05+0.015) pCi/g for a sample with one hot particle.

According to Hyatt (1955 and 1956), Jordan (1958), and Moss (1961) the mass median aerodynamic diameters (MMAD) of the particles emitted from TA-21 were from 0.3 to 1 µm. However, at locations close to TA-21, the observed variability of the soil data indicates 2 µm MMAD, probably resulting from emissions during the 1940s when the filters were less efficient.

When all the data are analyzed, the standard deviation indicates the variability of the data, which is 50%. Furthermore, when the data are separated into categories based on a variable, the differences between the results from different categories also vary by a factor of 2. The important variables are: distance, direction, soil type, amount of disturbance, and proximity to other sources of plutonium.

Within the boundaries of TA-1, There are many possible sources of contamination such as pipes containing contaminated wastes, leaks, and spills. There are also many types of soil disturbance such as vehicle and pedestrian traffic, or storm-water runoff that moved contamination. This raises the possibility that the standard sample depth of 5 cm may not have included all the plutonium that was

originally deposited on the surface. The concentration was measured at different depths (Ahlquist 1977) and the results indicate that at these locations the total amount deposited on the surface was 1.55 times the amount measured in the top 5 cm of soil.

On the other hand, at some locations leaks, spills, and other waterborne sources of contamination added to the plutonium that was deposited from the air. The variability of the data indicate that our estimate of the total airborne deposition has an uncertainty of about a factor of two in either direction.

For example, at TA-1 several samples are exceptionally high. Sample #4 was taken near Tank #138, which fed Hillside #138, a known area of water-borne contamination. Sample #5 was taken at a distance of 15 m from the edge of D building on the southwest side, down-gradient from D Building. The rain caps on the D-building stacks deflected contamination onto the roof, and the contamination ran off the roof to the surrounding areas. It is likely that sample #5 was contaminated by this runoff. And sample #23 was taken next to the radioactive waste line that connected to the radiochemistry building. It is likely that leaks from this line contaminated this sample. Nevertheless, we have included these samples in the analysis.

The data other than those at TA-1 are averages of several samples, often with considerable variability. For example, at TA-21, TA-54, and at some of the TA-73 locations, the variation is a factor of 10, which is far more than the uncertainty calculated by the analytical laboratory. This variation indicates patchy contamination caused by hot particles, spills, runoff, or other sources.

The samples at TA-73 are generally high, probably because of other sources of contamination such as material disposal areas (MDA) A, B, T, U, and V, that are closer to the sample locations than the major sources represented by S_1 and S_2. Releases from these minor sources will be included in the estimates of S_1 and S_2, but the estimates will be biased high. Ideally, the coefficients, c_{ij}, should be adjusted upwards to include these closer sources, and this adjustment would reduce the estimates of S_1 and S_2. However, this would greatly complicate the analysis.

Similarly, the TA-21 samples are high because of the nearby waste-research laboratory, TA-21-45, as well as MDA B and MDA V. The TA-50 samples are high because of the radioactive liquid waste facility, TA-50-1, as well as MDA C. And the TA-54 samples are high because of the nearby radioactive waste area, MDA G. All of these samples have been included in the analysis so releases from these sources will add to S_1 and S_2, but the estimates will be biased high. When these locations are excluded, the results decrease by 35%.

In summary, by including all locations, including data averaged from 700 samples, we have a robust analysis that is insensitive to individual anomalies. The overall uncertainty is estimated to be about a factor of two.

5. Results

The maximum-likelihood analysis using the method described in Section 2 leads to the results: $S_1 = 0.5 \pm 0.1$ Ci and $S_2 = 0.6 \pm 0.2$ Ci. Figures 4 and 5 show χ^2 versus S_1 and S_2. The value of χ^2 at the minimum is 78, which is 1.2 per degree of freedom. This shows that the estimates of the uncertainty, σ, are reasonable.

The uncertainties, ± 0.1 Ci for S_1 and ± 0.2 Ci for S_2, correspond to the statistical variation of the data from 69 sample locations. The small uncertainties indicate good agreement for a wide variety of conditions such as soil type, direction, and distance.

Systematic errors that might bias the data are considered in the following sections.

5.1 Global Fallout

Almost all of the soil concentrations used for these calculations are substantially above those that result from global fallout, so the uncertainty in global-fallout background does not change the results greatly. For example, if we halve the value of global-fallout background from 0.015 pCi/g to 0.0075 pCi/g, S_1 remains at 0.5 Ci and S_2 increases to 0.7 Ci.

5.2 Deposition velocity

We used the CAP88 default deposition velocity of 1.8 mm/s. For comparison, Napier (PNNL-14599) recommends a larger value, 3 mm/s, while Till and Grogan (Till 2008) recommend a smaller value, 1 mm/s, which is the same as the value used by RESRAD-Offsite (RESRAD 2010.) Hotspot (Hotspot 2010) uses 3 mm/s for respirable particles and 8 cm/s to calculate groundshine.

The effective deposition velocity depends on the type of terrain, the effective surface roughness, and the particle size (Sehmel 1980). The terrain includes ponderosa pine forest toward the west, pinon-juniper forest toward the east, and occasional buildings that are intermediate in height between these types of forest.

The particle sizes are affected by the types of filters used in the exhaust ducts. It is possible that the particle sizes were larger during the 1940s, before the development of HEPA filters. As filter efficiency improved during the 1950s it is likely the particle sizes were close to those reported by Hyatt 1955, Hyatt 1956, Jordan 1958, and Moss 1961.

In summary, the deposition velocity could be lower by a factor of two, or possibly higher by more than a factor of two. Note that if use a higher deposition velocity it would increase the coefficients, c_{ij}, and consequently decrease our estimates of the sources, S_1 and S_2.

5.3 Daytime winds compared with 24-hour winds

We have performed the analysis using two sets of wind data. The first represents wind data collected during both day and night; the second represents wind data collected during the daytime. Using both day and night wind data, the results were: S_1 = 0.5 Ci and S_2 = 0.5 Ci. Using daytime wind the results were: S_1 = 0.5 Ci and S_2 = 0.6 Ci. Thus, the choice of wind data does not make a significant difference.

5.4 Weathering, disturbance, or other contamination

It is possible that the soil samples are affected to some degree by weathering, soil disturbance, or other sources of contamination. Weathering and soil disturbance would decrease the estimates of S_1 and S_2. Sources of contamination such as spills, leaking pipes, and outfalls would increase the estimates. In general, the consistency of the results indicates these conditions affect the results by no more than 50%

in either direction. The correction factor for weathering is estimated to be 1.5 and the correction for other sources of contamination is 0.65. The product of these two factors is 1.0 ± 0.5.

6. Conclusions

We have used measurements of the plutonium deposition on the ground to calculate the airborne emissions from Los Alamos technical areas TA-1 and TA-21. The total emissions amount to: S_1 = 0.5 Ci from TA-1 and S_2 = 0.6 Ci from TA-21. The overall uncertainties are about a factor of two.

The value for the total releases from TA-1, S_1 = 0.5 Ci, agrees well with the range of value shown in Figure 4-13 and described on page 4-16 of the LAHDRA report. According to the LAHDRA report, the most probable value is 0.2 Ci, the median value is 0.4 Ci, and the 95th percentile upper limit is 1.05 Ci. These values are also within the range of 0.08 to 6.12 Ci estimated on page 4.22 of the LAHDRA report.

The value for the total releases from TA-21, S_2 = 0.6 Ci, is somewhat lower than the total of 1.2 Ci calculated by Andrews 1973 and Maraman 1975. It is possible that their estimates were high because their data were based on gross-alpha measurements obtained a few hours after the filters were removed, so they included contributions from radon and thoron. Also, Maraman et al. assumed the stack flow rate was equal to the design capacity: 3×10^9 m^3/year.

At TA-1, the largest amounts of plutonium were processed during the summer of 1945 (Hammel 1998). After World War II, D Building at TA-1 was used for chemistry and metallurgy research with relatively small amounts of plutonium. The release of 0.5 Ci of plutonium is consistent with the 19-mSv plutonium dose received by a clerical worker during this time (Miller 2008.) At TA-21, most of the releases occurred prior to the development and installation of HEPA filters during the 1950s (Maraman 1975.) The TA-21 releases resulted in smaller doses than the TA-1 releases because the TA-21 stacks were more effective and the distances to the residential areas were larger (Section 1.)

In summary, the total airborne releases of plutonium amounted to approximately 1 Ci, which is consistent with previous estimates by Los Alamos scientists.

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Table 1. Measured and predicted soil concentrations for 69 locations. Column 1 is the location of the sample; columns 2 through 5 are the distances (meters) and directions (sector) from TA1 and TA21 to the sample location; column 6 is the measured concentration (pCi/g); Columns 7 and 8 are the predicted concentrations (pCi/g) for 24-h wind and for daytime wind.

Location	from TA1		from TA21		Measured	24-h wind	Day wind
	meters	dir	meters	dir	pCi/g	pCi/g	pCi/g
TA1-1	310	ESE	2200	W	0.196	0.114	0.060
TA1-2	240	ESE	2300	W	0.076	0.135	0.076
TA1-3	220	E	2300	W	0.186	0.166	0.107
TA1-4	114	SE	2400	W	1.15	0.135	0.106
TA1-5	50	SW	2500	W	48.3	0.316	0.654
TA1-6	50	NW	2500	W	1.00	0.266	0.584
TA1-7	111	NNW	2500	W	0.392	0.233	0.449
TA1-8	120	WSW	2600	W	1.40	0.289	0.556
TA1-9	117	WNW	2600	W	0.50	0.204	0.371
TA1-10	160	NW	2600	W	0.373	0.142	0.245
TA1-11	160	WSW	2700	W	2.16	0.227	0.400
TA1-12	176	WNW	2700	W	0.232	0.140	0.230
TA1-13	220	WNW	2700	W	0.135	0.113	0.172
TA1-14	280	NW	2700	W	0.164	0.086	0.121
TA1-15	270	W	2800	W	0.148	0.118	0.177
TA1-16	293	WNW	2800	W	0.029	0.086	0.117
TA1-17	330	NW	2800	W	0.069	0.074	0.097
TA1-18	350	W	2850	W	0.276	0.091	0.123
TA1-19	354	WNW	2850	W	0.012	0.073	0.092
TA1-20	380	WNW	2850	W	0.726	0.068	0.084
TA1-21	425	W	2900	W	0.074	0.075	0.095
TA1-22	410	WNW	2900	W	0.234	0.064	0.076
TA1-23	465	WNW	3000	W	4.63	0.057	0.066
TA1-24	525	WNW	3000	W	0.137	0.052	0.057
TA1-25	540	WNW	3000	W	0.166	0.050	0.055
TA1-26	565	WNW	3000	W	0.148	0.049	0.053
South Mesa 1	2219	ESE	632	SW	0.034	0.046	0.062
South Mesa 2	1887	ESE	897	SW	0.158	0.041	0.046
South Mesa 3	1518	ESE	1171	WSW	0.031	0.038	0.040
South Mesa 4	1243	ESE	1402	WSW	0.043	0.040	0.036
South Mesa 5	987	SE	1721	WSW	0.030	0.042	0.033
South Mesa 6	685	SE	1996	W	0.083	0.056	0.033
South Mesa 7	503	SSE	2300	W	0.048	0.082	0.039
South Mesa 8	477	S	2510	W	0.110	0.100	0.050
South Mesa 9	423	SSW	2663	W	0.135	0.111	0.081
South Mesa 10	459	SW	2833	W	0.068	0.089	0.088
South Mesa 11	677	WSW	3132	W	0.102	0.053	0.058
South Mesa 12	785	W	3250	W	0.045	0.041	0.045
South Mesa 13	1134	W	3610	W	0.098	0.031	0.033
TA73-1	2000	E	633	NW	0.330	0.038	0.049

TA73-2	2300	E	395	NNW	0.171	0.054	0.096
TA73-3	2400	E	295	NNW	0.351	0.058	0.111
TA73-4	2670	E	300	NE	0.242	0.059	0.057
TA73-5	3000	E	523	ENE	0.172	0.049	0.042
TA73-6	3860	E	1251	E	0.221	0.033	0.024
TA73-7	4000	E	1431	E	0.029	0.031	0.023
East of TA-53	3086	ESE	827	SE	0.053	0.055	0.024
West of TA-53	1646	ESE	1060	WSW	0.030	0.047	0.043
Two-Mile Mesa	3231	WSW	5383	WSW	0.019	0.020	0.021
R-Site Rd East	4080	S	4272	SSW	0.019	0.022	0.018
North Mesa	2356	NE	1854	NNW	0.018	0.031	0.035
Sportsman Club	5411	NE	4388	NNE	0.024	0.022	0.019
Frijoles	11550	SSE	10790	S	0.025	0.017	0.016
PM1	7530	ESE	5100	ESE	0.012	0.018	0.016
TA16	5240	SSW	6280	SW	0.021	0.019	0.018
TA21	2090	E	420	WNW	0.131	0.045	0.071
TA33	11440	SSE	10480	S	0.013	0.017	0.016
TA36	5900	SSE	4700	S	0.012	0.019	0.017
TA49	7260	SSE	6870	S	0.018	0.018	0.017
TA49-S	6260	SSW	6960	SSW	0.027	0.018	0.016
TA50	1930	SSE	2450	SW	0.055	0.029	0.024
TA51	5190	SE	3720	SSE	0.017	0.020	0.016
TA52	3150	SE	2080	S	0.034	0.026	0.020
TA54	8310	SE	6390	SE	0.190	0.017	0.016
TA60-1	1230	SSE	2420	WSW	0.034	0.037	0.026
TA60-2	1740	SE	1850	SW	0.032	0.031	0.027
TA8	5010	WSW	7280	WSW	0.035	0.018	0.018
Kwage	5361	E	2945	ENE	0.028		
Barracas	5380	NE	3762	NNE	0.021		
Guaje	9926	ENE	8056	NE	0.024		
Chupaderos	12475	NE	11113	NNE	0.021		
Marias	13293	ENE	11159	ENE	0.017		
WR	10730	ESE	8480	SE	0.012	0.017	0.016

Figure 1. The first plutonium facility, D Building at Los Alamos technical area 1, viewed from the north-east.

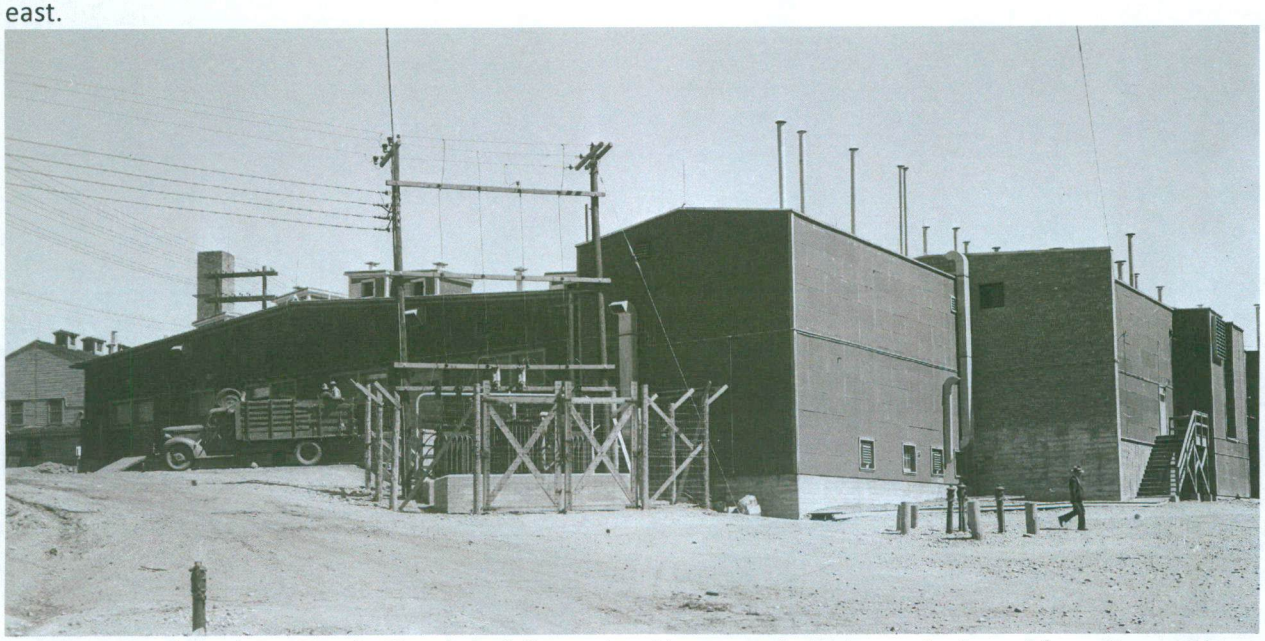

Figure 2. The first plutonium facility, D Building at Los Alamos technical area 1, viewed from the south.

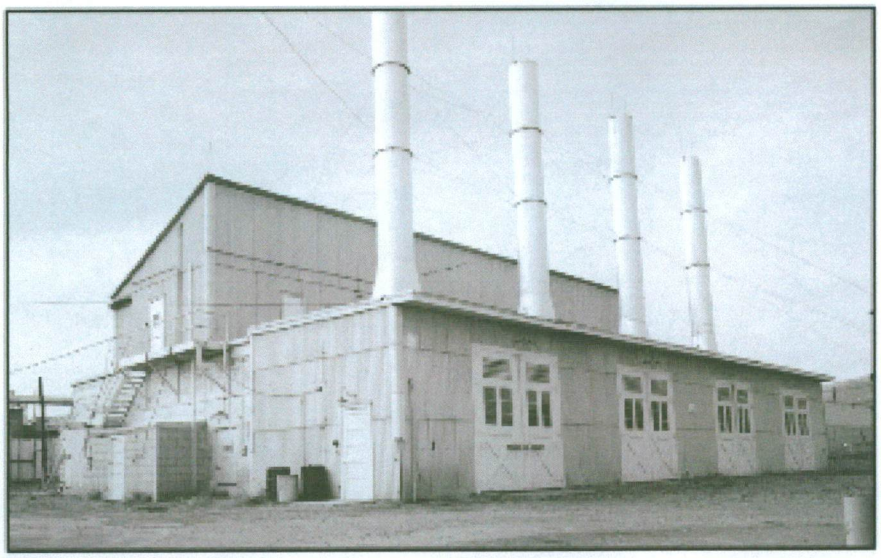

Figure 3. The TA-21 stacks, TA-21-12, viewed from the north-east.

Figures 4 and 5: variation of χ^2 as a function of the source terms, S_1 and S_2. The theoretical curve is a parabola of the form $y = y_0 + ((x-x_0)/\sigma)^2$, so $\pm 1\sigma$ corresponds to a χ^2 increase of 1. For example, the empirical curve shown in Fig. 1 is: $\chi^2 = 78.3 + ((S-0.457)/0.064)^2$; therefore $S_1 = 0.457 \pm 0.064$.