# **FINAL REPORT**

# Review of Routine Releases of Plutonium in Airborne Effluents at Rocky Flats

Task 2: Verification and Analysis of Source Terms

August 1999

Submitted to the Colorado Department of Public Health and Environment Disease Control and Environmental Epidemiology Division Rocky Flats Health Studies in Partial Fulfillment of Contract No. 100APPRCODE 391

"Setting the standard in environmental health"



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#### SUMMARY

This review is part of Phase II of the Historical Public Exposure Studies on Rocky Flats. It concentrated on routine releases of plutonium in years when the largest releases occurred. The term "routine" is used here to distinguish these releases from those during the 1957 and 1969 fires, which are presented in separate reports. Releases from all other unplanned events and conditions are included as part of the routine releases. During the period of most interest (before 1975), releases were primarily from the Building 771 stack and from the roof vents of Buildings 776-777. The main issue examined was the effect of nonuniform concentration distributions in large exhaust ducts on the estimates of routine plutonium releases. Possible bias and uncertainty in the self-absorption correction factor used in the analysis of effluent filter counting data were also studied.

A review of effluent monitoring data indicated that concentrations of plutonium were not constant over the cross-section of the large ducts and that the single point sampling system that was used before mid-1963 did not obtain a representative sample of the effluents. Detailed analysis of two sets of data, collected after multiple point sampling was introduced at Rocky Flats, led us to develop correction factors for nonrepresentative sampling. We used these factors and the associated uncertainties to make new estimates of plutonium releases.

The search for measurements of self-absorption of alpha particles in filter samples uncovered several old documents that contained relevant information. We used the data to estimate a central value and range of possible values for the self-absorption correction factor. Analysis showed that there was a small bias in the point estimate of the self-absorption correction factor previously used at Rocky Flats. In the present analysis, we considered the full range of self-absorption correction factors.

Revised estimates of plutonium releases from the Building 771 stack and Buildings 776-777 and other exhaust vents are shown as distributions in Figures <u>S-1</u> and <u>S-2</u>. In the figures, the median release estimate is shown as a short horizontal bar. For each year, the vertical line indicates the separation between the 5<sup>th</sup> and 95<sup>th</sup> percentiles of the distribution.

The revised Phase II release estimates are higher and reflect a broader range of uncertainty than those developed in Phase I. The greatest uncertainties are for years before 1964 and reflect the correction for nonrepresentative sampling. Revised estimates of releases during four decades, which reflect the distributions of the annual estimates, are shown in Table S-1. The estimated median release for the entire period (1953–1989), also shown in the table, is about 3 times greater than previous estimates.

	ised Estimates of Roenj 1	
Period	Median estimate (Ci) <sup>a</sup>	5 <sup>th</sup> –95 <sup>th</sup> percentile values (Ci) <sup>a</sup>
1953–1959	0.059	0.026-0.17
1960–1969	0.058	0.045-0.086
1970–1979	0.0043	0.0034-0.0056
1980–1989	0.00076	0.00066–0.0090
1953–1989	0.12	0.087-0.24
<sup>a</sup> To convert to g	rams, divide by 0.072 Ci g <sup>-1</sup>	(for example, $0.12 \text{ Ci} = 1.7 \text{ g}$ ).

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#### **REVIEW OF ROUTINE RELEASES OF PLUTONIUM IN AIRBORNE EFFLUENTS AT ROCKY FLATS**

#### **1. INTRODUCTION**

This investigation is a part of Phase II of the Historical Public Exposures Studies on Rocky Flats, performed by *Radiological Assessments Corporation* for the Colorado Department of Public Health and Environment. The Health Advisory Panel for the Studies recommended that an independent review of the estimates of releases of plutonium in airborne effluents be conducted as part of Phase II. Also recommended were separate reviews of the release estimates for the 1957 fire in Building 771 and the 1969 fire in Buildings 776-777. Effluent releases other than those during the two fires were considered in this review.

The releases considered in this report are frequently called "routine" to distinguish them from larger, short-term accidental releases associated with the two major fires at Rocky Flats. That semantic convention is followed in this review and in Phase II in general. However, we are aware that routine releases included discharges that were due to a variety of unplanned events and conditions that arose during facility operations. For example, there have been many small fires involving plutonium metal, which is pyrophoric, at Rocky Flats. There were also more significant events, such as the peroxide tank explosion in Building 771 in 1957 and the glovebox drain fire in Building 776 in 1965. Releases from these events, which were much smaller than those from the major fires, are not being analyzed separately and have been included in the category of routine releases. Similarly, plutonium releases during the extended cleanup work that followed both the 1957 and 1969 fires are placed in the same category. These releases were also much smaller than those that occurred during the fires.

The largest routine releases of plutonium from Rocky Flats facilities occurred before 1975 and were primarily from Building 771 stack and roof vents on Buildings 776-777. This review focuses on that period and those buildings. It builds upon investigations of plutonium releases that were performed during Phase I. The Phase I investigations (<u>ChemRisk</u> 1994; <u>Ripple et al.</u> 1996) can be grouped into three categories:

- 1. Flow rate measurements: included evaluation of measurements of sampler flow rate and of building exhaust flow rate information
- 2. Sample collection issues: included possible losses due to anisokinetic sampling of the discharge, deposition of particles in the sampling line, and incomplete collection of particles by the filters used for sampling
- 3. Factors affecting sample analysis: included detection efficiency of counters used to measure plutonium activity collected on the filters, adjustments for other alpha-emitters that could be on the filters, correction for the absorption of alpha particles emitted by plutonium that had penetrated into the sampling filter or had

been covered by dust, and partitioning of the gross alpha activity into plutonium and americium fractions.<sup>a</sup>

For some years, comparisons were also made of the releases reported in official documents with calculations based upon detailed effluent sampling data found in plant archives. The plutonium releases estimated in Phase I using archival data were generally in reasonably good agreement  $(\pm 20\%)$  with the officially reported releases (<u>ChemRisk</u> 1994).

ChemRisk estimated biases and uncertainties in the individual factors affecting the release estimates and assigned an uncertainty distribution to each factor. Calculations of overall bias associated with release estimates were made using Monte Carlo techniques. The median value of overall bias in plutonium release estimates for the years 1953–1973 was estimated to be 1.3. The geometric standard deviation (GSD) of the distribution of estimates was 1.6. For later years, when releases were generally much lower, the median estimate of bias was the same but the GSD of the distribution was slightly smaller, 1.4 (ChemRisk 1994). The reduction in uncertainty during that period was due primarily to the practice of routine determinations of the plutonium and americium fractions by alpha spectrometry. The revised plutonium release estimates developed by ChemRisk are shown in Table 1. Examination of the data shows very clearly that nearly all of the plutonium was released before 1975.

	Release		Release		Release
Year	estimate (µCi) <sup>a</sup>	Year	estimate (µCi) <sup>a</sup>	Year	estimate (µCi) <sup>a</sup>
1953	2.1 (0.81–5.3)	1965	6,900 (2,700–18,000)	1977	5.2 (2.0–13)
1954	69 (27–180)	1966	340 (130-870)	1978	3.6 (1.4–9.3)
1955	77 (30–200)	1967	430 (170–1,100)	1979	7.2 (2.8–18)
1956	250 (97-630)	1968	520 (200-1,300)	1980	16 (6.1-40)
1957	16,000 (6,100-40,000)	1969	1,400 (560–3,700)	1981	11 (4.2–27)
1958	3,300 (1,300-8,300)	1970	400 (160–1,000)	1982	26 (10-67)
1959	1,400 (560–3,700)	1971	79 (31–200)	1983	100 (40–260)
1960	1,400 (560–3,700)	1972	65 (25–170)	1984	100 (40–260)
1961	1,600 (610-4,000)	1973	66 (26–170)	1985	12 (4.7–31)
1962	3,300 (1,300-8,300)	1974	1,200 (490–3,200)	1986	38 (15–97)
1963	3,900 (1,500–10,000)	1975	13 (5.1–33)	1987	20 (7.6–50)
1964	3,000 (1,200–7,700)	1976	5.2 (2.0–13)	1988	20 (7.6–50)
				1989	5.9 (2.3–15)

Table 1. Phase I Estimates of Routine Plutonium Releases (ChemRisk 1994)

<sup>a</sup> Median value with 95% confidence interval in parentheses. For weapons grade plutonium the activity to mass ratio is 0.072  $\mu$ Ci  $\mu$ g<sup>-1</sup>. Example mass releases corresponding to the activity values are 222 mg in 1957, 42 mg in 1964, 17 mg in 1974, and 1.4 mg in 1984.

<sup>&</sup>lt;sup>a</sup> In this context, the word plutonium means weapons grade plutonium, which consists primarily of <sup>239</sup>Pu (~93.8%), <sup>240</sup>Pu (~5.8%), and <sup>241</sup>Pu (~0.36%). Both <sup>239</sup>Pu and <sup>240</sup>Pu emit 5.15-MeV alpha particles and cannot be separated by alpha spectrometry. Releases of these isotopes were the most important sources of radiation exposure of the nearby population. The beta decay of <sup>241</sup>Pu leads to formation of <sup>241</sup>Am, also an alpha-emitter, which was estimated to account for about 18% of the total alpha activity in routine releases.

One aspect of effluent sampling for plutonium that was not thoroughly investigated in the Phase I work was the question of representative sampling. That is, were the concentrations of plutonium in the samples of effluent that were collected representative of the average concentrations in the discharges? That question, which is the main focus of the present investigation, arises because the concentrations in very large exhaust ducts were based upon samples initially taken from single points in the centers of the ducts. Later, three sample withdrawal points were used to measure concentrations in the principal discharges.

The answer to the question about representative sampling is important to another Phase II investigation, namely the assessment of the release from the 1957 fire (Voillequé 1999). The 1957 fire analysis depends, in part, upon measurements of plutonium concentrations in large ducts leading to filter systems in that building. Those measurements were also based upon samples collected in a manner similar to that used for effluent sampling.

<u>Section 2</u> describes the exhaust duct and effluent sampling arrangement in Building 771 during the early years of operation and shows why there is a question about the representativeness of the effluent samples. <u>Section 3</u> contains an analysis of sets of effluent sampling data obtained after mid-1963, when 3-point sampling began, and presents estimates of the bias due to nonrepresentative sampling.

Uncertainty in the correction for self-absorption of alpha particles in air sampling filters is another aspect of the Rocky Flats effluent measurement results that was not investigated previously. Information on evaluation of self-absorption factors given in historical documents is discussed in <u>Section 4</u>. The historical data are used to estimate the uncertainty in the self-absorption correction factor.

Revised estimates of routine plutonium releases for the period of interest, which reflect the findings regarding representative sampling and the self-absorption factor, are given in <u>Section 5</u>. <u>Section 6</u> contains references to the technical literature. Detailed numerical data are included in <u>appendices</u>.

#### 2. BASIS FOR PLANT ESTIMATES OF ROUTINE PLUTONIUM RELEASES

Routine releases of plutonium to the atmosphere from Building 771 and from Buildings 776-777 were initially based upon samples of the effluent drawn from one point in the exhaust air stream. Figure 1 shows the discharge sampling locations for Building 771, both at startup (1953) and after modification of the sampling system in May 1963. During the first 10 years of operation, release estimates were based on the single sampling point, labeled Main 2, in the center of the large (90 by 96 in.) rectangular exhaust duct. For the next 20 years, release estimates were based upon three samples of the exhaust air withdrawn at the three locations indicated in the figure. Nearly all the routine releases from the Building 771 stack were measured using the single- or 3-point sampling procedure. Measurements in the main plenum of Buildings 776-777 were similar. At first, a single sampling point formed the basis for plant estimates of releases. Three-point sampling was also introduced there in 1963.

If the concentration of plutonium in the exhaust air had been uniform across the duct, then the concentration in a sample withdrawn at any location would be representative of that in the exhaust air. The number and location of sampling points would not be important in that case; however, the physical arrangement of the exhaust ductwork suggests that concentrations may well not have been uniform.



**Figure 1.** Locations of sampling points in a cross-section of the Building 771 exhaust duct after modification on 16 May 1963. Before that time samples had been collected only at the point labeled Main 2.

A schematic diagram (top view) of the exhaust system is shown in Figure 2. There were many ducts that carried air from all parts of the building to the large plenum where the filters were located. The direction of air flow is indicated by the arrows. Four exhaust fans are shown as circles containing the numbers 1–4 on the near side of the filter bank. These fans pulled air



**Figure 2.** Schematic diagram of Building 771 exhaust system and location of effluent sampling points. This is a top view; it is not drawn to scale.

through the filter bank and discharged air into the exhaust duct. The increase in size of the exhaust duct is indicated (not to scale) in the figure. After the connection to the outlet of the last fan the duct remained the same size (90 in. high by 96 in. wide) until it reached the larger (120 in. high by 96 in. wide) tunnel that led to the stack. Sampling points were located near the entrance to the tunnel. The sampling points are shown in <u>Figure 1</u>, which illustrates the cross-section of the duct at that point.

The size changes of the exhaust duct are shown in Figure 3, which has four parts. These show duct cross-sections at points just downstream of the connection of each of the fans. The shaded area in each section of the figure indicates the location and approximate size of the fan outlet connection that has entered the main exhaust duct.



**Figure 3.** Changes in exhaust duct cross-section as fan exhaust outlets enter the discharge. Shaded areas show the approximate sizes and locations of the exhaust fan outlet ducts entering the main exhaust duct.

In general, exhaust fans act to mix the contaminants in the air that passes through them. Downstream of Fan 1 it could reasonably be expected that the plutonium air concentration would be reasonably constant across the cross-section of the duct. However, the concentrations discharged by the four fans may differ, reflecting differences in concentrations in the inlet ducts or different decontamination factors for portions of the filter bank. If that were the case, it is quite possible that the concentration of plutonium in the duct was not uniform when the air reached the location of the samplers.

If the ducts were still in place, testing to determine whether mixing was complete could be performed using tracer gases under a variety of fan operating conditions. Unfortunately, the ductwork has been replaced and such measurements are not possible. Modeling of mixing of the fan exhaust flows within the duct was also considered. This approach would require records of fan operation during the early years of plant operation to determine the numbers and pairings of fans operating at any particular time. Such records are not available for the period of interest, so that technique is also not feasible.

The routine effluent measurement program carried out after the time when three sampling probes were used provides information about the degree of mixing in the exhaust ducts. Those data, discussed in the next section, provide substantial insight into the question of representative sampling of exhaust ducts for Buildings 771 and 776-777.

#### 3. ANALYSIS OF EFFLUENT CONCENTRATION MEASUREMENTS

As indicated in Figure 1, the change from one to three effluent samples for Building 771 occurred in 1963. A similar change occurred in Buildings 776-777, also in 1963. Effluent concentration measurements made when three samplers were operating provide information about the degree of mixing within the ducts and about the representativeness of the single sample taken in earlier years from the center of the duct. We evaluated two sets of effluent sampling results, as described below.

Before examining details of the measurement data, we define effluent concentrations  $C_1$ ,  $C_2$ , and  $C_3$  to be the estimates based upon measurements at locations Main 1, Main 2, and Main 3 that are shown in Figure 1. Then we examined the ratios of concentrations at the side locations to those on the centerline. These concentration ratios (CR) were defined to be

$$CR_4 = \frac{G}{G_2} \text{ and } CR = \frac{G}{G_2}$$
 (1)

These two ratios were part of a larger array of nine ratios, each of which represented a sector of the cross-section of the exhaust duct. The entire array is shown in <u>Figure 4</u>. For any given sector, the concentration ratio,  $CR_i$ , for that sector was the ratio of the average concentration in the sector to the average concentration in the center sector. The concentration ratio for the center sector,  $CR_5$ , was defined to be 1.

#### 3.1 Concentration Ratios Derived Using Measurements Made during 1963

<u>Table 2</u> contains effluent concentration data measured in the exhaust duct in Building 771 during the first 5 weeks following the modification of the sampling system in May 1963 (<u>Dow</u> 1963). <u>Table 2</u> is shown here to illustrate the data that were collected; <u>Table A-1</u> in <u>Appendix A</u> contains data for the first 7 months following the sampling system modification. The measured concentrations were originally reported in units of disintegrations per minute per cubic meter of air (dpm m<sup>-3</sup>), and those units are used here.

CR1	CR2	CR3
CR4	CR5	CR <sub>6</sub>
CR7	CR <sub>8</sub>	CR9

**Figure 4.** Illustration of the division of the exhaust duct into sectors. For the i<sup>th</sup> sector, the concentration ratio  $(CR_i)$  is the ratio of the concentration of plutonium in that sector to the measured concentration of the central sector.

During the period covered by <u>Table 2</u>, daily sample changes were the normal practice, although there are a few longer sampling periods (see <u>Table A-1</u>). Tables <u>2</u> and <u>A-1</u> also contain the concentration ratios,  $CR_4$  and  $CR_6$ , computed from the daily measurements and from the 7- or 8-day average concentrations. For those longer periods, the dates, mean concentrations, and ratios are intentionally offset to distinguish them from the daily results.

Figure 5 illustrates the dependence of the mean daily concentration ratios, designated  $CR_{D4}$  and  $CR_{D6}$ , on the average measured centerline concentration. The data in <u>Table A-1</u> were used to compute these ratios. Figure 5 shows that when the centerline concentration was small, there was more variability of concentration of plutonium in the duct and the centerline concentration was consistently smaller than that measured at the other two points. The magnitudes of the concentration ratios  $CR_{D4}$  and  $CR_{D6}$  decreased as the average concentration increased.

Mean daily concentration ratios for six ranges of centerline concentrations in the Building 771 exhaust during 1963 are shown in <u>Table 3</u>. Concentrations are listed in the units used for the measurements; for conversion to plutonium mass concentrations, 0.01 dpm corresponds to 0.063 pg. The grouping of data in the table differs slightly from that for <u>Figure 5</u>, but the tabulation shows the same decrease in ratios as the concentration increases. The ranges given in <u>Table 3</u> show the variability of the concentration ratios. At the highest concentrations, the spread in computed concentration ratios is lowest.

Mean concentration ratios for longer averaging times were also computed. Averages for 7or 8-day periods are termed "weekly" averages and are designated by  $CR_{W4}$  and  $CR_{W6}$ . In Tables 2 and A-1, the averaging period, mean concentration, and concentration ratios are displaced to the right in the row beneath the daily values used to compute the averages. Thirty weekly concentration ratios were computed for the Building 771 measurements in 1963. The mean values of  $CR_{W4}$  and  $CR_{W6}$  and the ranges of values of these ratios are presented in Table 4. Concentration ratios for seven monthly periods were also computed from the data in Table A-1. The means and ranges for these ratios, designated  $CR_{M4}$  and  $CR_{M6}$ , are shown in the lower portion of Table 4.

	Concentration Ratios for Bunding 771				
1963	Measured P	u concentrations	$(dpm m^{-3})$	Concentrat	tion ratios
Date	Main 1 $(C_1)$	Main 2 $(C_2)$	Main 3 $(C_3)$	$CR_4 \ (C_1/C_2)$	$CR_{6} (C_{3}/C_{2})$
16 May	1.33	1.90	3.09	0.70	1.6
17 May	0.86	1.62	4.48	0.53	2.8
18 May	2.33	1.29	2.95	1.8	2.3
19 May	1.00	0.43	1.81	2.3	4.2
20 May	0.86	0.95	1.71	0.91	1.8
21 May	0.86	0.86	1.24	1.0	1.4
22 May	0.95	1.29	4.10	0.74	3.2
23 May	4.50	1.20	7.40	3.8	6.2
16–23 May	1.59	1.19	3.35	1.3	2.8
24 May	2.20	0.90	2.90	2.4	3.2
25 May	1.33	2.10	1.71	0.63	0.81
26 May	0.86	2.33	2.19	0.37	0.94
27 May	1.95	2.33	3.48	0.84	1.5
28 May	0.71	1.00	2.00	0.71	2.0
29 May	2.00	1.31	6.48	1.5	4.9
30 May	1.90	1.57	2.29	1.2	1.5
31 May	1.14	0.95	5.43	1.2	5.7
24–31 May	1.51	1.56	3.31	1.0	2.1
1 Jun	0.19	0.005	2.48	38	500
2 Jun	0.86	0.71	2.33	1.2	3.3
3 Jun	0.90	1.57	3.10	0.57	2.0
4 Jun	1.71	3.76	2.33	0.45	0.62
5 Jun	0.86	1.29	1.76	0.67	1.4
6 Jun	1.29	2.38	2.67	0.54	1.1
7 Jun	2.57	0.38	1.48	6.8	3.9
1–7 June	1.20	1.44	2.31	0.83	1.6
8 Jun	2.24	1.24	3.52	1.8	2.8
9 Jun	0.33	0.10	1.00	3.3	10
10 Jun	0.38	0.81	1.10	0.47	1.4
11 Jun	1.00	0.90	2.00	1.1	2.2
12 Jun	2.33	1.10	0.76	2.1	0.69
13 Jun	0.38	0.48	0.33	0.79	0.69
14 Jun	1.19	0.05	1.38	24	28
15 Jun	0.29	0.43	3.10	0.67	7.2
8–15 June	1.02	0.64	1.65	1.6	2.6
16 Jun	0.57	0.38	3.62	1.5	9.5
17 Jun	0.95	0.81	2.70	1.2	3.3
18 Jun	0.71	0.52	2.33	1.4	4.5
19 Jun	1.48	1.05	2.19	1.4	2.1
20 Jun	0.10	0.52	0.005	0.19	0.010
21 Jun	0.24	0.14	1.52	1.7	11
22 Jun	0.005	0.005	0.005	1.0	1.0
16–22 June	0.58	0.49	1.77	1.2	3.6

## Table 2. Measured Effluent Concentrations and<br/>Concentration Ratios for Building 771



**Figure 5.** Dependence of daily concentration ratios ( $CR_{D4}$  and  $CR_{D6}$ ) upon measured centerline concentration for Building 771. The numbers of values available to compute the average ratios for 1963 are shown in parentheses.

Centerline concentration	Mean concentration rat	io and (range of values)
$C_2 (dpm m^{-3})$	CR <sub>D4</sub>	CR <sub>D6</sub>
$C_2 \le 0.005 \ (20)^a$	27 (1.0–172)	82 (1.0–786)
$0.005 < C_2 \le 0.1 \ (51)^a$	5.7 (0.050–71)	4.9 (0.050–57)
$0.1 < C_2 \le 0.2 \ (25)^a$	2.5 (0.036-8.1)	2.6 (0.26–11)
$0.2 < C_2 \le 0.5 \ (41)^a$	1.3 (0.013–6.8)	2.4 (0.015–16)
$0.5 < C_2 \le 1$ (40) <sup>a</sup>	1.2 (0.0070–7.2)	1.7 (0.0096–6.5)
$1 < C_2 \le 2 (27)^a$	0.99 (0.0031-3.8)	1.6 (0.029–6.2)
$2 < C_2 \le 5 (12)^a$	0.45 (0.13–1.1)	1.2 (0.13-4.3)
$C_2 > 5 (7)^a$	1.3 (0.20–1.9)	1.2 (0.21–1.5)
<sup>a</sup> Number of values in specifie	ed concentration range sho	wn in parentheses.

Table 3. Mean Concentration Ratios for Individual Samples, 1963

Table 4. Concentration Ratios for Weekly and Monthly Periods, 1963

Averaging	Mean concentration ratio and (range of values)		
period	$CR_{W4}$	$CR_{W6}$	
Weekly (30) <sup>a</sup>	1.5 (0.46–8.7)	1.8 (0.41–6.6)	
	$CR_{M4}$	$CR_{M6}$	
Monthly (7) <sup>a</sup>	1.2 (0.71–1.4)	1.9 (0.87–2.2)	

Number of periods of specified length shown in parentheses.

Although the ranges of ratios for the weekly and monthly averaging periods include values that are less than 1, the average values still indicate that the off-centerline concentrations exceeded those in the center of the duct. When the entire period covered by <u>Table A-1</u> (16 May–31 December) was considered, the mean concentration ratio for sector 4 was found to be 1.2 and that for sector 6 was found to be 1.5.

#### 3.2 Concentration Ratios Derived Using Measurements Made during 1969

A second set of data was compiled from plant sampling records (Dow 1969). The measurement results and the computed concentration ratios are tabulated in Appendix A, Table A-2. Monitoring data for the period 1 January through 15 October were evaluated. During this time, samples were collected daily except for weekend and holiday periods. Longer sampling periods were routinely employed after mid-October. Those results were not included in the analysis because the lengths of the sampling periods differed from those for 1963 and for the earlier part of 1969. A broad range of air concentrations was measured during the period. Only a few samples were below the detection limit of the analysis. The other centerline concentrations covered approximately the same concentration range observed in 1963, but there were more concentrations in the range 0.1-1 dpm m<sup>-3</sup> and fewer that were greater than 1 dpm m<sup>-3</sup>. For that reason, the centerline concentration categories in Table 5, which shows the concentration ratios for individual samples, differ from those in Table 3.

Centerline concentration	Mean concentration ratio and (range of values)		
$C_2 (dpm m^{-3})$	$CR_{D4}$	$CR_{D6}$	
$C_2 \le 0.005 \ (6)^a$	23 (1.0-60)	22 (1.0–54)	
$0.005 < C_2 \le 0.1 \ (30)^a$	2.4 (0.32–15)	4.4 (0.057–29)	
$0.1 < C_2 \le 0.2 \ (55)^a$	1.0 (0.16–3.1)	1.7 (0.11–7.7)	
$0.2 < C_2 \le 0.5 \ (75)^a$	0.78 (0.011-2.3)	1.2 (0.017–11)	
$0.5 < C_2 \le 1.0$ (23) <sup>a</sup>	0.92 (0.11-5.7)	0.74 (0.18–2.1)	
$C_2 > 1$ (12) <sup>a</sup>	0.78 (0.074–1.6)	0.95 (0.23–1.7)	
<sup>a</sup> Number of values in specif	ied concentration range sh	own in parentheses.	

 Table 5. Mean Concentration Ratios for Individual Samples, 1969

The same general behavior of the concentration ratios was observed for these data. When centerline concentrations were low, the mean daily concentration ratios were highest and quite variable. Substantial variability was also seen in samples for other centerline concentrations.

The concentration ratios for weekly and monthly averaging period are shown in <u>Table 6</u>. There were 38 weekly average values and 9 monthly averages. As for the previous data set, average concentration ratios for samples from sector 6 were higher than those for sector 4.

Averaging	Mean concentration ratio and (range of value		
period	$CR_{W4}$	$CR_{W6}$	
Weekly (38) <sup>a</sup>	0.85 (0.21–3.0)	1.4 (0.27–12)	
	$CR_{M4}$	$CR_{M6}$	
Monthly (9) <sup>a</sup>	0.85 (0.37–1.2)	1.1 (0.66–1.4)	

Table 6. Concentration Ratios for Weekly and Monthly Periods, 1969

#### 3.3 Distributions of Concentration Ratios for Duct Centerline Sampling

The two data sets discussed above cover a period of about 16 months during which more than 400 sets of measurements of concentrations C1, C2, and C3 were performed. We combined and analyzed the two data sets to develop distributions of estimates of the observed single sample concentration ratios. The computed concentration ratios were grouped according to the value of the centerline concentration  $(C_2)$  and the means and ranges of values were determined. Table 7 contains results for seven ranges of centerline concentrations (C2). Distributions of the concentration ratios ( $CR_4$  and  $CR_6$ ) for these centerline concentration ranges are shown in Figures 6 through 12. The concentration ratios typically cover a large range and have been plotted using a logarithmic scale for convenience.

Table 7. Mean C	Table 7. Mean Concentration Ratios for Individual Samples				
Centerline	Mean concentration ratio and (range of values)				
concentration (dpm m <sup>-3</sup> )	$CR_{D4}$	$CR_{D6}$			
$C_2 \le 0.005 \ (26)^a$	26 (1.0–172)	68 (1.0–786)			
$0.005 < C_2 \le 0.1 \ (81)^a$	4.4 (0.0050–71)	4.7 (0.050–57)			
$0.1 < C_2 \le 0.2 \ (80)^a$	1.5 (0.036-8.1)	1.9 (0.11–11)			
$0.2 < C_2 \le 0.5 \ (116)^a$	0.97 (0.011-6.8)	1.6 (0.015–16)			
$0.5 < C_2 \le 1 \ (63)^a$	1.1 (0.0070–7.2)	1.4 (0.0096–6.5)			
$1 < C_2 \le 2 (33)^a$	0.93 (0.0031-3.8)	1.5 (0.029–6.2)			
$C_2 > 2$ (25) <sup>a</sup> 0.79 (0.13–1.9) 1.1 (0.13–4.3)					
<sup>a</sup> Number of values in specified concentration range shown in parentheses.					

As can be seen from the plots, most of the distributions are irregular. A lognormal distribution would appear as a single straight line in these plots. Some distributions are approximately lognormal over most of the range (for example, see Figures 7 and 8). Histograms of the actual distributions of concentration ratios were developed for use in later calculations. Those results are included in Appendix B.



**Figure 6.** Distributions of concentration ratios ( $CR_4$  and  $CR_6$ ) for duct centerline concentrations ( $C_2$ ) that were below the detection level of 0.005 dpm m<sup>-3</sup>.



**Figure 7.** Distributions of concentration ratios ( $CR_4$  and  $CR_6$ ) for duct centerline concentrations ( $C_2$ ) that were in the range  $0.005 < C_2 \le 0.1$  dpm m<sup>-3</sup>.



**Figure 8.** Distributions of concentration ratios ( $CR_4$  and  $CR_6$ ) for duct centerline concentrations ( $C_2$ ) that were in the range  $0.1 < C_2 \le 0.2$  dpm m<sup>-3</sup>.



**Figure 9**. Distributions of concentration ratios ( $CR_4$  and  $CR_6$ ) for duct centerline concentrations ( $C_2$ ) that were in the range  $0.2 < C_2 \le 0.5$  dpm m<sup>-3</sup>.



**Figure 10**. Distributions of concentration ratios ( $CR_4$  and  $CR_6$ ) for duct centerline concentrations ( $C_2$ ) that were in the range  $0.5 < C_2 \le 1$  dpm m<sup>-3</sup>.



**Figure 11**. Distributions of concentration ratios ( $CR_4$  and  $CR_6$ ) for duct centerline concentrations ( $C_2$ ) that were in the range 1 <  $C_2 \le 2$  dpm m<sup>-3</sup>.



**Figure 12.** Distributions of concentration ratios ( $CR_4$  and  $CR_6$ ) for centerline concentrations ( $C_2$ ) that were > 2 dpm m<sup>-3</sup>.

#### 3.4. Average Duct Concentration Ratios for One-Month Periods Based Upon Duct Centerline Sampling

To assess the effects of nonuniform concentration distributions on the estimates of plutonium discharge concentration, it was necessary to consider the distribution of concentrations throughout the exhaust duct. Several steps are needed to complete the process. Figure 4 shows the division of the duct into nine sectors with concentration ratios defined for each sector. In the previous section, the observed distributions of values of  $CR_4$  and  $CR_6$  were described as functions of the measured duct centerline air concentration. The other concentration ratios were not measured, but they were estimated using knowledge of the physical arrangement of the fan exhaust ducts that entered the main exhaust duct (Figures 2 and 3). Development of relationships between measured concentration ratios ( $CR_4$  and  $CR_6$ ) and those that were not measured relies on professional judgment that is based upon the author's experience in measuring tracer concentration distributions in exhaust ducts at nuclear facilities.

<u>Table 8</u> shows the relationships that were developed and gives information about the distributions of uncertain parameters used in the calculations. Because the fan exhausts enter near the bottom of the duct, it is likely that the concentration ratios in the top three sectors of the duct ( $CR_1$ ,  $CR_2$ , and  $CR_3$ ) were lower than those for the central sectors. However, the possibility that they are higher is not excluded; this is shown by the range of values for parameters  $k_{41}$ ,  $k_{42}$ ,  $k_{62}$ , and  $k_{63}$ . Concentration ratios for the lower sectors of the duct ( $CR_7$ ,  $CR_8$ , and  $CR_9$ ) are expected to be higher than those in the center section. Sectors further from the entry of the fan exhausts are estimated to be less contaminated than sector 9, which is closest to the points of entry of contaminated air.

Concentration		Parameter			
ratio	Functional dependence	distributions			
$CR_5$	$CR_5 = 1$ , by definition	None			
$CR_4$	Calculated using $C_1$ and $C_2$	Empirical results for ranges of C <sub>2</sub>			
$CR_6$	Calculated using $C_3$ and $C_2$	Empirical results for ranges of C <sub>2</sub>			
$CR_1$	$= k_{41} CR_4$	k <sub>41</sub> : Uniform (0.3–1.5) <sup>a</sup>			
$CR_2$	$= 0.5 \ (k_{42} \ CR_4 + k_{62} \ CR_6)$	k <sub>42</sub> : Uniform (0.3–1.5) <sup>a</sup>			
		k <sub>62</sub> : Uniform (0.3–1.5) <sup>a</sup>			
$CR_3$	= k <sub>63</sub> CR <sub>6</sub>	k <sub>63</sub> : Uniform (0.3–1.5) <sup>a</sup>			
$CR_7$	$= k_{47} CR_4$	$k_{47}$ : Uniform (1–2) <sup>a</sup>			
$CR_8$	$= k_{68} CR_6$	k <sub>68</sub> : Uniform (1–3) <sup>a</sup>			
$CR_9$	= k <sub>69</sub> CR <sub>6</sub>	k <sub>69</sub> : Uniform (1–5) <sup>a</sup>			
<sup>a</sup> Values in parentheses are upper and lower bounds of the distribution.					

Table 8. Concentration Ratios Used to Estimate Effects of NonuniformExhaust Duct Concentration, Duct Centerline Sampling

During the early years of operation, monthly average release estimates were based upon average duct centerline concentrations ( $C_{2m}$ , dpm m<sup>-3</sup>) computed from sequences of measurement results from daily or daily and weekend sampling periods. That procedure was used during most of the years with plutonium releases exceeding 1000 µCi (>~14 mg). For a duct centerline concentration in a particular range ( $0.1 < C_2 \le 0.2$  dpm m<sup>-3</sup>, for example), the relationships in <u>Table 8</u> allow us to estimate an average concentration of plutonium in the exhaust duct that reflects the bias and uncertainties associated with the single point sampling approach that was used. First, we consider a single sampling period. During that sampling period (lasting 1 day in most cases and for 2–3 days for holidays and weekends), the average concentration of plutonium in the duct ( $C_a$ , dpm m<sup>-3</sup>) can be estimated from the single measured concentration ( $C_2$ ) using:

$$C_{\rm a} = C_{\rm g} \left(\frac{1}{\mathfrak{g}}\right) \sum_{i=1}^{\mathfrak{g}} C R_{\rm g}$$
<sup>(2)</sup>

Section 3.3 contains distributions for the concentration ratios that are applicable to each range of values of  $C_2$ . Those distributions were used to estimate  $CR_4$  and  $CR_6$ . The relationships in <u>Table 8</u> were used to estimate the other concentration ratios, and  $C_a$  was computed using <u>Equation (2)</u>. This procedure was repeated with different parameter values, chosen by Monte Carlo techniques, to perform the calculations many times for each sampling period in a month. The simulations (1000 repetitions) produced distributions of estimates of the monthly average concentration across the duct ( $C_{am}$ , dpm m<sup>-3</sup>) and distributions of estimates of the ratio ( $C_{am}$ / $C_{2m}$ ) for the month.

The simulations were performed for the several ranges of centerline concentrations of interest, yielding distributions of the ratio ( $C_{am} / C_{2m}$ ) for a broad range of monthly average centerline concentrations. This procedure assured that the higher and more variable concentration ratios, which apply to individual samples collected when effluent concentrations were low, were fully expressed in the calculation of the monthly average concentration ratios.

Figure 13 provides a perspective of the computed distributions of the estimated concentration ratio ( $C_{am} / C_{2m}$ ) for different values of the monthly average duct centerline concentration ( $C_{2m}$ ). Not surprisingly, the highest concentration ratios were found for the lowest values of  $C_{2m}$  and the lowest concentration ratios were associated with the highest average centerline concentrations. The spread of the distribution of values was also found to be greater for the lower concentrations. This is illustrated by the difference between the slopes of the two labeled distributions and is further shown by the following numerical values. For the lower concentration range (0.01 dpm m<sup>-3</sup>  $\leq C_{2m}$ ), a spread of a factor of 300 in the estimated values of ( $C_{am} / C_{2m}$ ) was found. For the highest concentration range ( $C_{2m} > 2$  dpm m<sup>-3</sup>), the spread in estimates was about a factor of 40.

Median values of the ratio ( $C_{am} / C_{2m}$ ) ranged from 10 for the lowest concentration range ( $C_{2m} \leq 0.01$  dpm m<sup>-3</sup>) to 1.3 for the highest centerline concentrations ( $C_{2m} > 2$  dpm m<sup>-3</sup>). Although only two other distributions of ( $C_{am} / C_{2m}$ ) are shown on the plot, all other distributions were found to lie between the labeled extreme distributions. The icosatiles (the 0<sup>th</sup>, 5<sup>th</sup>, 10<sup>th</sup>, 15<sup>th</sup>, ... 100<sup>th</sup> percentiles) of the distributions of the ratio ( $C_{am} / C_{2m}$ ) for various concentration ranges are given in the tables in <u>Appendix C</u>.



**Figure 13.** Distributions of concentration ratios ( $C_{am} / C_{2m}$ ) for different monthly average duct centerline concentrations ( $C_{2m}$ ). Although, for clarity, not all distributions are included in the figure, the labeled distributions show the range of observed distributions. The two intermediate distributions correspond to  $0.05 < C_{2m} \le 0.1$  dpm m<sup>-3</sup> (long dashes) and to  $1 < C_{2m} \le 2$  dpm m<sup>-3</sup> (short dashes).

#### 3.5 Distributions of Concentration Ratios for Three-Point Sampling

After the sampling approach was changed from the single duct centerline method to the collection of three samples (see Figure 1), the three measured concentrations ( $C_1$ ,  $C_2$ , and  $C_3$ ) were averaged by the plant staff and the mean value (termed  $C_{bar}$  here) was used to estimate releases. For the assessment of sampling bias during the years when the 3-point sampling technique was used, a different set of concentration ratios was developed. These concentration ratios are defined to be

$$CR_{4*} = \frac{C_1}{C_{\text{bar}}}, \quad CR_2 = \frac{C_2}{C_{\text{bar}}}, \quad \text{AND} \quad CR_2 = \frac{C_2}{C_{\text{bar}}}$$
(3)

Values of the three ratios have been grouped according to the magnitude of the mean concentration ( $C_{bar}$ , dpm m<sup>-3</sup>). The distributions of the ratios  $CR_{4*}$ ,  $CR_{5*}$ , and  $CR_{6*}$  are presented in Figures 14–19. These ratios are less variable than those based on comparisons with the duct centerline concentration alone. Most values of these concentration ratios are in the range between 0.1 and 3. The distributions are irregular, but the central portions of some of them are approximately lognormal. Histograms of the distributions of the actual concentration ratios were used in later calculations and have been tabulated in <u>Appendix B</u>.



**Figure 14.** Distributions of concentration ratios ( $CR_{4*}$ ,  $CR_{5*}$ , and  $CR_{6*}$ ) for mean measured concentrations ( $C_{bar}$ ) that were  $\leq 0.1$  dpm m<sup>-3</sup>.



**Figure 15.** Distributions of concentration ratios ( $CR_{4*}$ ,  $CR_{5*}$ , and  $CR_{6*}$ ) for mean measured concentrations ( $C_{bar}$ ) that were in the range  $0.1 < C_{bar} \le 0.2$  dpm m<sup>-3</sup>.



**Figure 16.** Distributions of concentration ratios ( $CR_{4*}$ ,  $CR_{5*}$ , and  $CR_{6*}$ ) for mean measured concentrations ( $C_{bar}$ ) that were in the range  $0.2 < C_{bar} \le 0.5$  dpm m<sup>-3</sup>.

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**Figure 17.** Distributions of concentration ratios ( $CR_{4*}$ ,  $CR_{5*}$ , and  $CR_{6*}$ ) for mean measured concentrations ( $C_{bar}$ ) that were in the range  $0.5 < C_{bar} \le 1$  dpm m<sup>-3</sup>.



**Figure 18.** Distributions of concentration ratios ( $CR_{4*}$ ,  $CR_{5*}$ , and  $CR_{6*}$ ) for mean measured concentrations ( $C_{bar}$ ) that were in the range  $1 < C_{bar} \le 2$  dpm m<sup>-3</sup>.



**Figure 19.** Distributions of concentration ratios ( $CR_{4*}$ ,  $CR_{5*}$ , and  $CR_{6*}$ ) for mean measured concentrations ( $C_{bar}$ ) that were > 2 dpm m<sup>-3</sup>.

#### 3.6. Average Duct Concentration Ratios for One-Month Periods Based Upon Three-Point Sampling

A procedure similar to that described in <u>Section 3.4</u> was used to develop average duct concentration ratios for 1-month periods that could be related to the results of three-point sampling of the discharge for the same interval. The breakdown of the exhaust duct into nine sectors (Figure 4) was used again. Distributions of the concentration ratios  $CR_{4*}$ ,  $CR_{5*}$ , and  $CR_{6*}$  found for various ranges of the measured average concentration ( $C_{bar}$ ) were employed in a manner similar to that described previously. <u>Table 9</u> shows the functional relationships that were used and summarizes the distributions for the parameters employed.

As before, Monte Carlo calculations were performed using distributions of the concentration ratios  $CR_{4*}$ ,  $CR_{5*}$ , and  $CR_{6*}$  (Figures <u>14–19</u>) and the other distributions given in <u>Table 9</u>. An equation similar to <u>Equation (2)</u> was used to estimate the average duct air concentration (with  $C_{bar}$  in place of  $C_2$  and the  $CR_{i*}$  in place of the  $CR_i$ ). Repeated calculations were performed to generate distributions of estimates of the monthly average plutonium concentration in the exhaust duct ( $C_{am}$ , dpm m<sup>-3</sup>) and distributions of estimates of the ratio ( $C_{am}$  /  $C_{barm}$ ) of that concentration to the monthly average concentration based upon 3-point measurements ( $C_{barm}$ ). Simulations were performed for six ranges of measured mean air concentrations. The ranges correspond to those shown in Figures <u>14–19</u> for the three primary concentration ratios. The icosatiles of the distributions of the ratio ( $C_{am}$  /  $C_{barm}$ ) for each concentration range are tabulated in <u>Appendix C</u>. The distributions of the ratios are not strongly dependent upon the measured mean concentration. It was found that all estimates of the ratio ( $C_{am}$  /  $C_{barm}$ ) were between 0.9 and 2.5.

	ust Duct Concentration, Infec	i onic Sumpring				
Concentration		Parameter				
ratio	Functional dependence	distributions				
$CR_{4*}$	Calculated using $C_1$ and $C_{bar}$	Empirical results for				
		ranges of C <sub>bar</sub>				
$CR_{5*}$	Calculated using C <sub>2</sub> and C <sub>bar</sub>	Empirical results for				
		ranges of C <sub>bar</sub>				
$CR_{6*}$	Calculated using C <sub>3</sub> and C <sub>bar</sub>	Empirical results for				
		ranges of C <sub>bar</sub>				
$CR_{1*}$	$= k_{41} CR_{4*}$	k <sub>41</sub> : Uniform (0.3–1.5) <sup>a</sup>				
$CR_{2*}$	$= 0.5 \; (k_{42} \; CR_{4*} + k_{62} \; CR_{6*})$	k <sub>42</sub> : Uniform (0.3–1.5) <sup>a</sup>				
		k <sub>62</sub> : Uniform (0.3–1.5) <sup>a</sup>				
$CR_{3*}$	$= k_{63} CR_{6*}$	k <sub>63</sub> : Uniform (0.3–1.5) <sup>a</sup>				
<i>CR</i> <sub>7*</sub>	$= k_{47} CR_{4*}$	k <sub>47</sub> : Uniform (1–2) <sup>a</sup>				
<i>CR</i> <sub>8*</sub>	$= k_{68} CR_{6*}$	k <sub>68</sub> : Uniform (1–3) <sup>a</sup>				
$CR_{9*}$	$= k_{69} CR_{6*}$	k <sub>69</sub> : Uniform (1–5) <sup>a</sup>				
<sup>a</sup> Values in parentheses are upper and lower bounds of the distribution.						

Table 9.	Concentration	<b>Ratios Used to</b>	Estimate	Effects of	Nonuniform
	<b>Exhaust Duct</b>	Concentration	, Three-Po	oint Sampl	ing

#### 4. UNCERTAINTY IN THE SELF-ABSORPTION CORRECTION FACTOR

Alpha particles are doubly charged and interact strongly with any medium. As a result, the distance that they travel before their energies have been dissipated is short. For example, the range of plutonium alpha-particles in air under standard conditions is about 3.6 cm. In a material whose density is higher, the plutonium alpha particle range is shorter (e.g., about 50  $\mu$ m in human tissue). When counting alpha particles emitted from samples, one must address the possibility that some alpha particles are absorbed before reaching the detector. Conversion from a measured gross alpha counting rate (counts per minute) to the activity (disintegrations per minute or other unit such as picocuries) of alpha-emitters on a filter involves estimating the absorption of alpha particles by the filter paper itself or by dust on the filter. Because such losses occur in the sample being counted, they are often termed losses due to self-absorption by the sample. For the period of greatest interest in this report, the Rocky Flats plutonium stack and vent effluent samples were collected using HV-70 filters. Filters submitted for counting would not have had heavy loadings of particulate material because the exhaust air had already passed through high-efficiency particulate air (HEPA) filters before sampling.

When converting from counts to activity, the plant staff applied an additional correction to reflect the counting efficiency for the alpha particles. The counting efficiency was based upon routine counts of a standard containing a known amount of plutonium and is not considered here.

The basis for the self-absorption correction factor of 0.7 that was used at Rocky Flats for many years has been elusive (<u>ChemRisk</u> 1994). A point estimate was used at Rocky Flats without consideration of the uncertainty in the correction factor. <u>ChemRisk</u> (1994) also used the point

estimate because the authors could not determine the basis for the traditional value. The choice of a correction factor of 0.7 implies that 30% of the alpha particles emitted by plutonium in the filter sample were not expected to escape from the surface of the filter. This fraction is sometimes called the burial loss. <u>Bokowski</u> (1973) indicated that unpublished data (collected by J. B. Owen at Rocky Flats) were supportive of the burial loss of 30% for HV-70 filters, but those data were not the original basis for selecting that value.

The self-absorption correction factor used at Rocky Flats was reportedly based upon measurements performed at Oak Ridge. Indeed, an early Oak Ridge report gives a factor of 70%, with a spread of -5% to +10% (Smith and McPherson 1945). The corresponding burial loss would be in the range 20–35%. Two more basic reports referenced by Smith and McPherson were not located. The measurements were made on filters (type unknown) that contained natural uranium. Natural uranium emits several alpha particles with somewhat lower alpha particle energies than plutonium (~4.5 MeV versus 5.15 MeV). The average range of the uranium alpha particles in air under standard conditions is about 3 cm.

The early Oak Ridge results are similar to those reported in more detail by <u>Alercio and Harley</u> (1952) of the U.S. Atomic Energy Commission's Health and Safety Laboratory (HASL). The HASL data were for Whatman 41 filters containing uranium. Their results for solid particles gave a mean burial loss of about 28% (98 samples) with relative standard deviations of 18–23%. An overall burial loss of 30% was recommended for routine use. A range of burial losses of 15–40% was reported for the HASL data groups of differing uranium mass loading. The lowest burial losses (15%) were found for filter loadings greater than 1 mg. The highest burial losses (41%) were measured for loadings of 0.2–12  $\mu$ g of uranium.

<u>Struxness</u> (1954) reported results of measurements for filters used to collect uranium dioxide particles generated in the laboratory. Estimated burial losses (32 determinations) for Whatman 41 filters ranged from 44–52% and increased with increasing flow rate through filters of the same diameter. This report noted that results for Whatman filters appeared to differ from those made for other filters, including the HV-70 filters that were employed at Rocky Flats. <u>Struxness</u> (1954) reported average burial losses of 14% for 9-mil thick HV-70 filters (12 determinations). Results for 11 measurements using the 18-mil thick filters were similar and yielded a mean burial loss of 12%. In both cases, great variability in the results was indicated by the reported standard errors of the mean. Filter mass loadings were not reported for these measurements.

Angleton and Barker (1954) estimated alpha particle absorption losses by comparing the alpha to beta counting rates for various filters to those for a very thin source with no losses. The alpha and beta particle emissions used came from the short-lived progeny of <sup>220</sup>Rn. Mass loadings for these experiments would have been quite low. Alpha particles emitted by those radionuclides have energies that exceed 6 MeV and have ranges that exceed those of plutonium alpha particles. Their report gives an absorption loss for Whatman 41 filters of 55%; this value appears to be based on a single measurement. However, the high burial losses for low mass loadings (less than 50 µg) of uranium in the same filters were also reported by Boback (1963). A mean loss of 51% was reported for 250 samples that contained small quantities of uranium. Measured absorption losses for various compounds of uranium were in the range of 45–53% under those measurement conditions.

The <u>Angleton and Barker</u> (1954) report also gave results for HV-70 filters. Losses in the range of 36–40% were reported for flow rates of 2, 4, 8, and 10 ft<sup>3</sup> min<sup>-1</sup>, all through filters of the same diameter. Changing the flow rate through the filter over this range did not appear to

affect the measured burial loss. An internal Los Alamos Scientific Laboratory memo (Vasilik 1976) also cites a Japanese result of 39% burial loss, apparently obtained using a similar method. When HV-70 filters were employed at Los Alamos, they estimated self-absorption losses to be  $37.6\pm2.8\%$ . The uncertainty is twice the standard deviation of the five measurements considered. One measurement by <u>Angleton and Barker</u> (1954) was not included in the average. For a flow rate of 1 ft<sup>3</sup> min<sup>-1</sup>, they reported a burial loss of about 63%. <u>Struxness</u> (1954) also reported higher burial losses for lower flow rates, but for the Whatman 41 filter.

Lindeken (1961) also used radon progeny to estimate burial loss for several filter types. He reported a burial loss of 18% for the 9-mil HV-70 filters and 28% for 20-mil moving filter material, also HV-70. The highest losses that he measured were for Whatman 41 filters (40%) and Microsorban filters (43%). The lowest burial loss that he found was 9% for the Gelman E glass fiber filter. In contrast, <u>Bokowski</u> (1973) found losses of nearly 50% for Gelman E filters that had been used to collect plutonium samples from the incinerator exhaust in Building 771.

When a different filter paper for effluent sample collection was introduced at Los Alamos (~1982), they determined the burial loss for particles collected on filters used to sample exhausts from both uranium and plutonium areas. Burial losses for the new filter paper were higher than those for the HV-70 filters that had been used, so the results are not directly applicable. However, data provided by Los Alamos National Laboratory (Miller 1998) were used to compare losses for uranium and plutonium and to estimate the uncertainty in individual determinations. The latter was found to be 15–20% of the reported burial loss estimate. For plutonium, burial losses were estimated to be 54% with a range of 36–68% for 11 samples. For uranium (14 samples) the mean burial loss was 61% with a range of 50–70%. Although the mean burial loss was somewhat higher for uranium, there was no statistical difference between the mean losses for plutonium and uranium.

It is desirable to base an estimate of the self-absorption factor for the Rocky Flats monitoring on measurements of burial losses for plutonium collected on HV-70 filters. None of the historic data meet those criteria. Nearly all of the early measurement results are for uranium and most are for collection of samples using Whatman 41 filters.

A lower bound for the HV-70 burial loss is the range of 12–14% reported in <u>Struxness</u> (1954) for those filters. Unfortunately, the data do not indicate whether this result was influenced by high mass loading of uranium in the samples. The upper bound on burial loss for HV-70 filters is the value of 63% reported in <u>Angleton and Barker</u> (1954) for a flow rate of 1 ft<sup>3</sup> min<sup>-1</sup>. Again, the reason for the result is not clear but there are no known reasons why it should not be considered. The other estimates of burial loss for HV-70 filters were in the range of 36–40% (<u>Angleton and Barker</u> 1954).

Although there are questions about the applicability of results for the Whatman 41 filters to situations where HV-70 filters were employed, it is interesting that the range of values for those filters is comparable (15–55%). Definitive studies of the effects of varying the filter loading and flow rate have not been found for either filter.

A broad range of burial losses for the HV-70 filters is necessary to encompass the range of values that have been reported. For the present work, we selected a triangular distribution of burial loss (*b*) with bounds of 13 and 63% and a mode of 38%. The mean burial loss obtained from this distribution is 38%, which differs from the historical value ( $b_o = 0.3$ ). The original self-absorption correction factor ( $f_o = 1 - b_o = 0.7$ ) was in the denominator of the equation used to estimate filter activity. To examine the effect of using a revised correction factor (f) (and its

associated uncertainty) on effluent measurements at Rocky Flats, we consider the ratio (R) given by

$$R = \frac{f_o}{f} = \frac{1 - b_o}{1 - b} \tag{4}$$

We used the triangular distribution for b given above to estimate the bias and uncertainty associated with the use of the historic value  $(b_o)$  for effluent measurements at Rocky Flats. A simple estimate of R for the mode of the distribution is 1.13, indicating that a relatively small correction in daily sample results occurs when the selected distribution is used. Monte Carlo calculations were performed to determine the effect on monthly average air concentrations of using the value for b, including its uncertainty, in place of the historic value. From these calculations we obtained a distribution of monthly average values of the ratio of the revised to the original concentration estimate. That distribution, shown in Figure 20 was used in calculations of revised release estimates that are discussed in Section 5.



**Figure 20.** Distribution of estimates of the ratio (R), which reflects the change in concentration estimates when the revised estimate of burial loss replaces the one used historically.

#### 5. CALCULATION OF REVISED ANNUAL RELEASE ESTIMATES

The calculations of revised estimates of annual releases reflect the effects of nonrepresentative sampling of the exhaust ducts (<u>Section 3</u>), the bias and uncertainty associated with the self-absorption correction factor (<u>Section 4</u>), and the bias and uncertainty factors identified in Phase I analysis. As noted in <u>Section 1</u>, the median bias factor estimated in Phase I

was taken to be the constant (1.3) over time, but the GSD of the bias factor was somewhat greater (1.6) for the period 1953–1973 from that (1.4) for later years (ChemRisk 1994; Ripple et al. 1996). These differences were considered in the calculations of revised annual releases for the appropriate years. We used the distribution of monthly average values of R (Figure 20) to incorporate the bias and uncertainty associated with the self-absorption factor. The correction for nonrepresentative sampling varied with the time period, depending on whether only duct centerline sampling or three-point sampling was used to obtain the plant's release estimates. For all periods, Monte Carlo calculations were used to obtain distributions of the product of the distributions of the original estimated release, the correction for nonrepresentative sampling, the correction for the self-absorption factor, and the bias factor identified in Phase I.

For the early years of operation, when only duct centerline data were collected, the monthly concentration ratio distributions for single-point sampling ( $C_{am} / C_{2m}$ ) were used to correct for nonrepresentative sampling. In these years, the estimated effluent concentrations ( $C_{2m}$ ) often varied substantially from month to month. Examination of the distribution of 10 years of monthly concentration data compared to the appropriate annual average values showed that fully one third of the monthly concentrations were  $\leq 40\%$  of the average for that year and more than one-half (56%) were  $\leq 80\%$  of the corresponding annual average. High concentrations in a single month could dominate the annual average value. In the calculations, the monthly concentration ratio ( $C_{am} / C_{2m}$ ) distribution varied according to the magnitude of  $C_{2m}$  for a particular month. This approach permitted expression of the larger and more variable concentration ratios in the majority of months when reported concentrations were low. For years after 1963, when 3-point sampling was employed, the monthly concentration ratios ( $C_{am} / C_{barm}$ ) were used to correct for nonrepresentative sampling. Although these concentration ratios did not vary greatly with the measured concentration ( $C_{barm}$ ), variation in monthly concentrations was considered in the calculations.

Results of the calculations of revised routine plutonium releases are summarized in Tables <u>10</u> and <u>11</u> for the Building 771 stack release and releases from the vents that discharge near roof level of other buildings, primarily Buildings 776-777. In each table, the 5<sup>th</sup>, 25<sup>th</sup>, 50<sup>th</sup>, 75<sup>th</sup>, and 95<sup>th</sup> percentiles of the distribution of release estimates for the year are provided. The two sets of release estimates are tabulated separately because the atmospheric dispersion of the roof vent releases differs from that used for the tall stack. In assessing the risks from the routine releases, dispersion of plutonium from the two sources are treated separately.

Table 10 shows that the highest routine plutonium releases from Building 771 occurred between 1957 and 1965 and that median release quantities after 1965 were below 1,500  $\mu$ Ci (~21 mg). After 1970, median annual releases were below 500  $\mu$ Ci (~7 mg). Figure 21 shows the distributions of plutonium release estimates for the Building 771 stack for the years 1957, 1964, and 1974. The distribution of plutonium release estimates for 1957 does not include releases during the fire in September of that year. That event is the subject of a separate analysis (Voillequé 1999). However, it does include other incidents and accidents (the peroxide explosion in Room 146) that occurred during that year. The plot shows that the releases in 1957 were both higher and more uncertain than those in 1964 or 1974.

	Percentiles of the distribution of release estimates (uCi) for the year				
Year	5 <sup>th</sup>	25 <sup>th</sup>	50 <sup>th</sup>	75 <sup>th</sup>	95 <sup>th</sup>
1953	30	54	84	150	450
1954	160	260	360	570	1,600
1955	170	270	370	580	1,100
1956	530	780	1,100	1,400	2,900
1957	16,000	30,000	45,000	71,000	150,000
1958	3,400	5,300	7,800	12,000	22,000
1959	1,300	2,200	2,900	3,700	5,800
1960	1,500	2,300	3,200	4,500	8,600
1961	1,200	2,400	3,200	4,200	6,400
1962	3,400	5,600	8,000	13,000	24,000
1963	2,700	5,900	8,100	11,000	17,000
1964	3,500	4,400	5,100	6,100	7,900
1965	8,400	11,00	13,000	15,000	19,000
1966	530	640	740	860	1,100
1967	560	670	780	900	1,100
1968	730	860	1,000	1,100	1,500
1969	1,000	1,300	1,500	1,700	2,100
1970	340	410	460	530	670
1971	49	61	72	84	100
1972	73	87	100	120	150
1973	30	38	45	55	68
1974	30	37	42	46	60
1975	11	14	15	17	22
1976	4.3	5.2	5.9	6.7	8.5
1977	4.5	5.3	6.1	7.0	8.6
1978	2.9	3.5	3.9	4.5	5.6
1979	6.5	7.7	8.7	10	13
1980	14	17	20	23	29
1981	9.7	11	13	15	19
1982	29	35	39	45	56
1983	140	160	190	210	260
1984	16	20	22	26	30
1985	6.0	8.4	11	13	19
1986	7.2	10	13	16	22
1987	10	14	19	23	31
1988	11	15	20	24	35
1989	0.052	0.072	0.090	0.11	0.16
<sup>a</sup> For weapons	grade plutoniur	n the activity to	mass ratio is 0.	072 μCi μg <sup>-1</sup> . Ε	xample median
(50 <sup>th</sup> percentile) mass releases are 630 mg in 1957 and 71 mg in 1964.					

#### Table 10. Distributions of Revised Release Estimates for the Building 771 Stack, 1953–1989

Radiological Assessments Corporation "Setting the standard in environmental health"

Vear	file	25 th	50 th	75 th	05 th
1057	5		<u> </u>	100	95
1957	5.5	19	45	100	410
1958	57	110	170	320	8/0
1959	99	170	270	440	1,100
1960	38	75	130	260	810
1961	7.7	24	45	100	440
1962	14	31	55	130	530
1963	220	320	460	720	1,700
1964	1,300	1,600	1,900	1,200	2,700
1965	2,500	3,200	3,700	4,400	5,500
1966	31	39	46	55	67
1967	150	190	210	240	290
1968	200	240	280	320	390
1969	1,500	1,900	2,100	2,400	3,100
1970	320	410	460	530	660
1971	80	99	110	130	170
1972	31	39	45	53	70
1973	74	92	110	130	160
1974	2,000	2,300	2,700	3,000	3,700
1975	9.6	11	13	15	19
1976	3.9	4.8	5.5	6.1	7.7
1977	3.9	4.6	5.2	6.0	7.5
1978	2.9	3.5	3.9	4.6	5.6
1979	5.1	5.9	7.1	8.0	9.7
1980	10	13	14	16	21
1981	8.0	9.5	11	13	15
1982	13	15	17	20	26
1983	26	30	35	41	50
1984	150	170	200	220	270
1985	8.1	9.6	11	13	15
1986	42	51	60	71	90
1987	13	15	17	20	24
1988	11	14	15	17	22
1989	9.7	11	13	15	19
or weapons	grade plutoniun	n the activity to	mass ratio is 0.	072 μCi μg <sup>-1</sup> . Ε	xample me

Table 11. Distributions of I	<b>Revised Release Estimates</b>
for Building Ve	nts, 1957–1989



**Figure 21**. Distributions of routine plutonium release estimates for the Building 771 stack for the years 1957, 1964, and 1974.

Table 11 shows that the median estimates of releases from building vents exceeded 500  $\mu$ Ci in the years 1964, 1965, 1969, and 1974 and were generally much lower than that value in other years. Greater uncertainties in these release estimates, as indicated by the spread of values for a particular year, reflect the greater uncertainties associated with measurements of plutonium concentrations in the earlier years of operation when single-point duct centerline sampling was employed to estimate releases.

Figure 22 shows the distributions of estimates of the total routine releases of plutonium for each decade of operation (1953–1959, 1960–1969, 1970–1979, and 1980–1989) and for the entire period (1953–1989). This figure clearly shows the importance of releases during the first two decades of operation. Releases during the 1950s and 1960s were more than 10 times greater than those during the 1970s, and releases during the 1980s were even lower.

The revised release estimates are higher and reflect a broader range of uncertainty than those developed in Phase I. It is believed that all prior biases in the estimates have now been addressed. The greatest uncertainties are for years before 1964 and reflect the correction for nonrepresentative sampling. The estimated median release of 0.12 Ci (1.7 g) for the entire period of operation is about 3 times greater than previous estimates. The uncertainty range (5<sup>th</sup>–95<sup>th</sup> percentiles of the distribution) associated with that value is 0.087–0.24 Ci and is relatively narrow as the result of summing of the distributions of both stack and vent releases over a period of many years.



**Figure 22**. Distribution of estimates of total routine plutonium releases from the stack and vents by decade and for the entire period of operation (1953–1989).

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#### APPENDIX A

## TABULATIONS OF DAILY EFFLUENT MEASUREMENTS TAKEN FROM PLANT LOGBOOKS

1963	Measured P	u concentrations	(dpm m <sup>-3</sup> )	Concentra	tion ratios
Date	Main 1 $(C_1)$	Main 2 $(C_2)$	Main 3 $(C_3)$	$CR_4 (C_1 / C_2)$	$CR_{6} (C_{3} / C_{2})$
16 May	1.33	1.90	3.09	0.70	1.6
17 May	0.86	1.62	4.48	0.53	2.8
18 May	2.33	1.29	2.95	1.8	2.3
19 May	1.00	0.43	1.81	2.3	4.2
20 May	0.86	0.95	1.71	0.91	1.8
21 May	0.86	0.86	1.24	1.0	1.4
22 May	0.95	1.29	4.10	0.74	3.2
23 May	4.50	1.20	7.40	3.8	6.2
16–23 May	1.59	1.19	3.35	1.3	2.8
24 May	2.20	0.90	2.90	2.4	3.2
25 May	1.33	2.10	1.71	0.63	0.81
26 May	0.86	2.33	2.19	0.37	0.94
27 May	1.95	2.33	3.48	0.84	1.5
28 May	0.71	1.00	2.00	0.71	2.0
29 May	2.00	1.31	6.48	1.5	4.9
30 May	1.90	1.57	2.29	1.2	1.5
31 May	1.14	0.95	5.43	1.2	5.7
24–31 May	1.51	1.56	3.31	1.0	2.1
1 Jun	0.19	0.005	2.48	38	500
2 Jun	0.86	0.71	2.33	1.2	3.3
3 Jun	0.90	1.57	3.10	0.57	2.0
4 Jun	1.71	3.76	2.33	0.45	0.62
5 Jun	0.86	1.29	1.76	0.67	1.4
6 Jun	1.29	2.38	2.67	0.54	1.1
7 Jun	2.57	0.38	1.48	6.8	3.9
1–7 June	1.20	1.44	2.31	0.83	1.6
8 Jun	2.24	1.24	3.52	1.8	2.8
9 Jun	0.33	0.10	1.00	3.3	10
10 Jun	0.38	0.81	1.10	0.47	1.4
11 Jun	1.00	0.90	2.00	1.1	2.2
12 Jun	2.33	1.10	0.76	2.1	0.69
13 Jun	0.38	0.48	0.33	0.79	0.69
14 Jun	1.19	0.05	1.38	24	28
15 Jun	0.29	0.43	3.10	0.67	7.2
8–15 June	1.02	0.64	1.65	1.6	2.6
16 Jun	0.57	0.38	3.62	1.5	9.5
17 Jun	0.95	0.81	2.70	1.2	3.3
18 Jun	0.71	0.52	2.33	1.4	4.5
19 Jun	1.48	1.05	2.19	1.4	2.1
20 Jun	0.10	0.52	0.005	0.19	0.010
21 Jun	0.24	0.14	1.52	1.7	
22 Jun	0.005	0.005	0.005	1.0	1.0
16–22 June	0.58	0.49	1.77	1.2	3.6

 Table A-1. Measured Effluent Concentrations and Concentration Ratios, 1963

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Table A-1. (Continued)					
1963	Measured	Pu concentration	ns (dpm m $^{-3}$ )	Concentra	tion ratios
Date	Main 1 ( $C_1$ )	Main 2 $(C_2)$	Main 3 $(C_3)$	$CR_4 (C_1 / C_2)$	$CR_{6} (C_{3} / C_{2})$
23 Jun	1.14	0.14	0.95	8.1	6.8
24 Jun	0.33	0.52	0.05	0.63	0.10
25 Jun	0.005	0.30	0.005	0.017	0.017
26 Jun	1.19	0.71	2.00	1.7	2.8
27 Jun	0.43	2.10	1.67	0.20	0.80
28 Jun	0.005	0.005	0.005	1.0	1.0
29 Jun	0.005	0.71	0.67	0.0070	0.9
30 Jun	0.48	0.71	0.71	0.68	1.0
23–30 June	0.45	0.65	0.76	0.69	1.2
1 July	1.14	4.57	7.05	0.25	1.5
2 July	1.00	0.90	1.24	1.1	1.4
3-4 July	0.38	NDa	0.57		
5 July	0.29	0.38	0.52	0.76	1.4
6 July	0.29	0.29	2.00	1.0	6.9
7-8 July	1.10	0.95	0.10	1.2	0.11
1, 2, 5–8 July	0.64	1.18	1.82	0.54	1.54
9 July	0.24	0.24	1.14	1.0	4.8
10 July	0.10	0.48	0.14	0.21	0.29
11 July	1.95	0.86	2.24	2.3	2.6
12 July	0.67	1.10	0.29	0.61	0.26
13 July	1.29	0.33	1.33	3.9	4.0
14 July	0.76	1.38	2.10	0.55	1.5
15 July	0.57	1.48	0.48	0.39	0.32
9–15 July	0.80	0.84	1.10	0.95	1.32
16 July	0.71	0.33	0.62	2.2	1.9
17 July	0.14	0.86	0.81	0.16	0.94
18 July	0.24	0.19	1.10	1.3	5.8
19 July	0.52	0.62	1.06	0.84	1.7
20 July	0.41	0.52	0.52	0.79	1.0
21 July	0.005	1.61	3.81	0.0031	2.4
22 July	0.38	2.00	0.81	0.19	0.41
23 July	0.57	0.38	0.90	1.5	2.4
16–23 July	0.37	0.81	1.20	0.46	1.5
24 July	0.19	0.29	0.33	0.66	1.1
25 July	0.19	0.67	0.33	0.28	0.49
26 July	0.81	0.14	0.33	5.8	2.4
27 July	2.29	1.00	1.0/	2.5	1./
28 July	1.25	0.33	ð.33 12 0	0.20	1.3
29 July	17.9	9.57	12.9	1.9	1.3
50 July 21 July	21.4 10.2	11.1 10 <i>4</i>	14.0 12 4	1.9	1.5
21 JULY 24 21 I.J.	17.2	10.0	13.4	1.0	1.3
∠ <del>4</del> –51 July	7.90	4.7/	0.49	1.0	1.5

Table A-1.	(Continu	ed)
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Measured P	u concentratio	$\frac{1}{ns}$ (dnm m <sup>-3</sup> )	Concentra	tion ratios
Main 1 (C <sub>4</sub> )	$\frac{d}{d} \frac{d}{d} \frac{d}$	$\frac{\text{IIS (upin III )}}{\text{Main 3 (C_2)}}$	$\frac{CR}{CR}$ (C <sub>1</sub> /C <sub>2</sub> )	$\frac{CR}{CR}$ (C <sub>2</sub> /C <sub>2</sub> )
13.0	8 33	$\frac{12.00}{12.00}$	$\frac{16}{16}$	$\frac{CR_6}{1.4}$
7 67	8.57	12.00	0.89	1.4
2 10	0.53	2 14	4 0	4.0
5.81	0.81	0.62	7.2	0.8
0.90	0.38	0.90	2.4	2.4
2.86	1.14	1.38	2.5	1.2
1.52	0.62	4.00	2.5	6.5
g 4.19	1.82	3.13	2.3	1.7
0.38	0.19	0.19	2.0	1.0
0.57	0.33	2.19	1.7	6.6
0.57	0.05	0.29	11	5.8
0.24	0.76	0.43	0.32	0.57
0.24	0.90	0.29	0.27	0.32
0.38	0.43	0.29	0.88	0.67
0.81	0.19	0.38	4.3	2.0
0.71	0.24	0.71	3.0	3.0
g 0.49	0.39	0.60	1.3	1.5
1.00	0.14	0.24	7.1	1.7
1.95	1.00	1.05	2.0	1.1
0.81	0.38	5.95	2.1	16
1.95	1.62	0.29	1.2	0.18
0.67	2.29	0.29	0.29	0.13
0.95	1.71	0.05	0.56	0.029
1.20	1.70	1.04	0.71	0.61
g 1.31	1.23	1.25	1.07	1.01
0.57	0.95	0.05	0.60	0.05
0.24	0.19	0.29	1.3	1.5
0.05	0.05	0.005	1.0	0.10
ND <sup>0</sup>	1.81	ND <sup>a</sup>	2.0	1.0
0.90	0.24	0.29	3.8	1.2
0.48	0.14	0.19	3.4	1.4
0.76	1.38	0.33	0.55	0.24
0.48	3.0/	1.57	0.13	0.43
0.30	0.93	0.09	1.2	0.015
0.38	0.55	0.003	1.2	0.015
1 1/	1 10	0.20	1.0	1.5
1.14	0.90	1 20	2.1	1 /
1.00	0.30	0.67	2.1	2.0
0.62	0.33	1 1/	0.69	2.0
0.02	1 24	1.14	0.57	0.92
0.85	0.77	0.86	1.10	1.11
	Measured P           Main 1 (C <sub>1</sub> )           13.0           7.67           2.10           5.81           0.90           2.86           1.52           4.19           0.38           0.57           0.24           0.24           0.24           0.24           0.38           0.81           0.71           5           0.67           0.95           1.20           1.31           0.57           0.24           0.38           0.81           0.71           0.49           1.00           1.95           0.67           0.95           1.20           1.31           0.57           0.24           0.05           NDb           0.90           0.48           0.76           0.48           0.50           0.38           0.19           1.14      1.86      1.05      0	Measured Pu concentratioMain 1 ( $C_1$ )Main 2 ( $C_2$ )13.08.337.678.572.100.535.810.810.900.382.861.141.520.624.191.820.380.190.570.330.570.050.240.760.240.900.380.430.810.190.710.2420.490.380.430.810.381.951.000.810.381.951.620.672.290.951.711.201.701.311.230.570.950.240.190.050.05NDb1.810.900.240.480.140.761.380.483.6730.520.500.950.510.900.520.330.620.900.711.240.850.77	Measured Pu concentrations (dpm m <sup>-3</sup> )           Main 1 (C <sub>1</sub> )         Main 2 (C <sub>2</sub> )         Main 3 (C <sub>3</sub> )           13.0         8.33         12.00           7.67         8.57         12.48           2.10         0.53         2.14           5.81         0.81         0.62           0.90         0.38         0.90           2.86         1.14         1.38           1.52         0.62         4.00           4.19         1.82         3.13           0.38         0.19         0.19           0.57         0.33         2.19           0.57         0.33         2.19           0.57         0.05         0.29           0.24         0.76         0.43           0.24         0.90         0.29           0.81         0.19         0.38           0.71         0.24         0.71           5         1.62         0.29           0.81         0.38         5.95           1.95         1.62         0.29           0.67         2.29         0.29           0.67         2.29         0.29           0.67         2.29	Measured Pu concentrations (dpm m <sup>-3</sup> )         Concentrations (dpm m <sup>-3</sup> )         Concentrations (dpm m <sup>-3</sup> )         CR4 $(C_1/C_2)$ 13.0         8.33         12.00         1.6           7.67         8.57         12.48         0.89           2.10         0.53         2.14         4.0           5.81         0.81         0.62         7.2           0.90         0.38         0.90         2.4           2.86         1.14         1.38         2.5           1.52         0.62         4.00         2.5           4.19         1.82         3.13         2.3           0.38         0.19         0.19         2.0           0.57         0.33         2.19         1.7           0.57         0.33         2.19         1.7           0.57         0.05         0.29         11           0.24         0.76         0.43         0.32           0.24         0.90         0.29         0.27           0.38         0.43         0.29         0.88           0.81         0.19         0.38         4.3           0.71         0.24         0.71         3.0           1.

Table A-1.	( <b>Continued</b> )
I GOIC II II I	Commuca,

			(Continucu)		
1963	Measured	Pu concentration	ns (dpm $m^{-3}$ )	Concentra	tion ratios
Date	Main 1 ( $C_1$ )	Main 2 $(C_2)$	Main 3 $(C_3)$	$\overline{CR}_4 (C_1 / C_2)$	$CR_6 (C_3 / C_2)$
8 Sept	0.62	1.00	0.24	0.62	0.24
9 Sept	0.67	0.33	0.81	2.0	2.5
10 Sept	0.19	0.19	0.33	1.0	1.7
11 Sept	3.67	3.24	3.52	1.1	1.1
12 Sept	0.29	0.33	0.38	0.88	1.2
13 Sept	0.67	0.52	0.43	1.3	0.83
14 Sept	0.85	0.29	0.43	2.9	1.5
15 Sept	0.33	1.10	0.19	0.30	0.17
8–15 Sept	0.91	0.88	0.79	1.0	0.90
16 Sept	0.38	1.90	0.76	0.20	0.40
17 Sept	0.24	0.10	0.29	2.4	2.9
18 Sept	0.81	0.14	0.19	5.8	1.4
19 Sept	0.24	0.62	2.76	0.4	4.5
20 Sept	0.33	0.10	0.10	3.3	1.0
21 Sept	0.19	0.14	0.62	1.4	4.4
22 Sept	0.33	0.10	1.05	3.3	11
16–22 Sept	0.36	0.44	0.82	0.81	1.9
23 Sept	0.24	0.67	0.14	0.36	0.21
24 Sept	0.10	0.05	0.19	2.0	3.8
25 Sept	0.43	0.62	0.38	0.69	0.61
26 Sept	0.38	0.38	0.67	1.0	1.8
27 Sept	0.24	0.01	0.57	24	57
28 Sept	0.005	0.10	0.33	0.050	3.3
29 Sept	0.67	0.10	0.48	6.7	4.8
30 Sept	0.14	0.48	0.71	0.29	1.5
23-30 Sept	0.28	0.30	0.43	0.91	1.4
1 Oct	0.38	0.29	1.24	1.3	4.3
2 Oct	0.19	0.14	0.52	1.4	3.7
3 Oct	0.38	0.05	0.24	7.6	4.8
4 Oct	0.67	0.14	0.10	4.8	0.7
5 Oct	0.05	0.76	0.10	0.066	0.13
6 Oct	0.14	0.33	0.14	0.42	0.42
7 Oct	0.43	0.19	0.40	2.3	2.1
1–7 Oct	0.32	0.27	0.39	1.2	1.4
8 Oct	0.14	0.10	0.10	1.4	1.0
9 Oct	0.05	0.05	0.38	1.0	7.6
10 Oct	0.10	0.05	0.19	2.0	3.8
11 Oct	0.10	0.04	0.14	2.5	3.5
12 Oct	0.24	0.19	0.24	1.3	1.3
13 Oct	0.10	0.05	0.05	2.0	1.0
14 Oct	0.10	0.05	0.05	2.0	1.0
15 Oct	0.10	0.10	0.14	1.0	1.4
8–15 Oct	0.12	0.08	0.16	1.5	2.0

Table A-1. (Continued)

		I able II I	(continued)		
1963	Measured l	Pu concentration	$(dpm m^{-3})$	Concentra	tion ratios
Date	Main 1 ( $C_1$ )	Main 2 $(C_2)$	Main 3 ( $C_3$ )	$CR_4 (C_1 / C_2)$	$CR_6 (C_3 / C_2)$
16 Oct	0.10	0.005	0.10	20	20
17 Oct	0.005	0.38	0.10	0.013	0.26
18 Oct	0.90	0.62	1.33	1.5	2.1
19 Oct	0.14	0.05	0.14	2.8	2.8
20 Oct	0.14	0.05	0.14	2.8	2.8
21 Oct	0.005	0.05	0.10	0.10	2.0
22 Oct	0.19	0.05	0.05	3.8	1.0
23 Oct	0.10	0.14	0.71	0.71	5.1
16–23 Oct	0.20	0.17	0.33	1.2	2.0
24 Oct	0.05	0.05	0.005	1.0	0.10
25 Oct	0.05	0.05	0.05	1.0	1.0
26 Oct	0.10	0.005	0.05	20	10
27 Oct	0.23	0.005	0.10	46	20
28 Oct	0.67	1.19	3.33	0.56	2.8
29 Oct	7.57	8.95	1.90	0.85	0.21
30 Oct	0.81	2.05	8.81	0.40	4.3
31 Oct	0.48	2.76	2.14	0.17	0.78
24–31 Oct	1.25	1.88	2.05	0.66	1.1
1 Nov	0.01	0.005	0.005	2.0	1.0
2 Nov	0.15	0.09	0.19	1.7	2.1
3 Nov	0.005	0.005	0.005	1.0	1.0
4 Nov	0.12	0.01	0.005	12	0.50
5 Nov	0.10	0.05	0.05	2.0	1.0
6 Nov	0.23	0.005	1.26	46	250
7 Nov	0.04	0.41	0.27	0.10	0.66
1–7 Nov	0.09	0.08	0.26	1.1	3.1
8 Nov	0.10	0.05	0.05	2.0	1.0
9 Nov	0.10	0.43	0.14	0.23	0.33
10 Nov	0.005	0.02	0.05	0.25	2.5
11 Nov	0.04	0.29	0.25	0.14	0.86
12 Nov	0.09	0.26	0.26	0.35	1.0
13 Nov	0.15	0.12	0.18	1.3	1.5
14 Nov	0.38	0.24	0.24	1.6	1.0
15 Nov	0.05	0.05	0.09	1.0	1.8
8–15 Nov	0.11	0.18	0.16	0.63	0.86
16 Nov	0.05	0.02	0.13	2.5	6.5
17 Nov	0.08	0.08	0.09	1.0	1.1
18 Nov	0.19	0.30	0.34	0.63	1.1
19 Nov	0.24	0.05	0.14	4.8	2.8
20 Nov	0.10	0.005	0.005	20	1.0
21 Nov	0.86	0.005	0.04	170	8.0
22 Nov	0.05	0.05	0.08	1.0	1.6
16–22 Nov	0.22	0.07	0.12	3.1	1.6

Table A-1. (Continued)

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1963	Measured	Pu concentration	$\frac{1}{18}$ (dpm m <sup>-3</sup> )	Concentra	tion ratios
Date	Main 1 ( $C_1$ )	Main 2 $(C_2)$	$\frac{1}{1} \frac{(C_1)}{Main 3 (C_3)}$	$CR_4 (C_1 / C_2)$	$\overline{CR_{6} (C_{3} / C_{2})}$
23 Nov	0.04	0.05	0.10	$\frac{4 \times 1}{0.80}$	2.0
24 Nov	0.08	0.005	0.005	16	1.0
25 Nov	0.07	0.46	0.01	0.15	0.022
26 Nov	0.09	0.04	0.56	2.3	14
27 Nov	0.10	0.33	0.005	0.30	0.015
28 Nov	0.04	0.01	0.06	4.0	6.0
29 Nov	0.05	0.02	0.55	2.5	28
30 Nov	0.12	0.005	0.04	24	8.0
23–30 Nov	0.07	0.12	0.17	0.64	1.4
1 Dec	0.16	0.005	3.93	32	790
2 Dec	0.02	0.02	0.10	1.0	5.0
3 Dec	0.06	0.35	0.08	0.17	0.23
4 Dec	0.03	0.05	0.04	0.60	0.80
5 Dec	0.83	0.02	0.04	42	2.0
6 Dec	0.005	0.14	0.43	0.036	3.1
7 Dec	0.09	0.14	0.14	0.64	1.0
1–7 Dec	0.17	0.10	0.68	1.6	6.6
8 Dec	0.14	0.19	0.14	0.74	0.74
9 Dec	0.14	0.10	0.02	1.4	0.20
10 Dec	0.52	0.10	0.10	5.2	1.0
11 Dec	0.005	0.10	0.005	0.05	0.050
12 Dec	0.24	0.005	0.005	48	1.0
13 Dec	0.14	0.43	0.10	0.33	0.23
14 Dec	0.10	0.05	0.19	2.0	3.8
15 Dec	0.24	0.48	0.10	0.50	0.21
8–15 Dec	0.19	0.18	0.08	1.0	0.45
16 Dec	0.10	0.14	0.14	0.71	1.0
17 Dec	0.005	0.005	0.06	1.0	12
18 Dec	4.95	0.07	0.005	71	0.071
19 Dec	0.07	0.19	0.14	0.37	0.74
20 Dec	0.005	0.14	0.24	0.036	1.7
21 Dec	0.33	0.05	0.005	6.6	0.10
22 Dec	0.10	0.005	0.01	20	2.0
23 Dec	0.02	0.04	0.05	0.5	1.3
16-23 Dec	0.70	0.08	0.08	8.7	1.0
24-26 Dec	0.005	0.005	0.03	1.0	6.0
27 Dec	0.14	0.005	0.04	28	8.0
28 Dec	0.05	0.03	0.10	1.7	3.3
29 Dec	0.38	0.05	0.14	7.6	2.8
30 Dec	0.96	0.19	0.05	5.1	0.26
31 Dec	0.05	0.005	0.05	10	10
24-31 Dec	0.20	0.04	0.06	5.4	1.6

Table A-1.	(Continued)
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<sup>a</sup> No data available; concentration ratios could not be computed. <sup>b</sup> Short-term sample; not included in weekly average.

1969	Measured Pu concentrations (dpm m <sup>-3</sup> ) Concentration ratios				
Date	Main 1 $(C_1)$	Main 2 $(C_2)$	Main 3 $(C_3)$	$CR_4 (C_1 / C_2)$	$CR_{6} (C_{3} / C_{2})$
2 Jan	0.639	0.314	0.417	2.04	1.33
3 Jan	0.182	0.214	0.246	0.85	1.15
4–6 Jan	0.512	0.790	0.665	0.65	0.84
7 Jan	0.184	0.214	0.526	0.86	2.46
8 Jan	0.156	0.332	0.202	0.47	0.61
2–8 Jan	0.39	0.49	0.48	0.78	0.98
9 Jan	0.352	0.387	0.410	0.91	1.06
10 Jan	0.078	0.098	0.092	0.80	0.94
11–13 Jan	1.462	1.882	1.462	0.78	0.78
14 Jan	0.250	0.407	0.391	0.61	0.96
15 Jan	0.569	0.637	0.609	0.89	0.96
16 Jan	0.739	0.995	0.544	0.74	0.55
9–16 Jan	0.80	1.02	0.80	0.78	0.79
17 Jan	0.094	0.053	0.119	1.77	2.25
18–20 Jan	0.087	0.243	0.090	0.36	0.37
21 Jan	0.107	0.154	0.080	0.69	0.52
22 Jan	0.213	0.804	0.369	0.26	0.46
23 Jan	0.114	0.132	0.107	0.86	0.81
24 Jan	0.116	0.691	0.352	0.17	0.51
16–24 Jan	0.11	0.32	0.16	0.35	0.51
25–27 Jan	0.158	0.474	0.650	0.33	1.37
28 Jan	0.099	0.102	0.021	0.97	0.21
29 Jan	0.041	0.069	0.036	0.59	0.52
30 Jan	0.078	0.182	0.196	0.43	1.08
31 Jan	0.173	0.092	0.168	1.88	1.83
25–31 Jan	0.12	0.27	0.34	0.46	1.27
1–3 Feb	0.121	0.149	0.177	0.81	1.19
4 Feb	0.089	0.005	0.010	17.80	2.00
5 Feb	0.041	0.179	0.135	0.23	0.75
6 Feb	0.005	0.005	0.029	1.00	5.80
7 Feb	0.083	1.116	0.357	0.07	0.32
1–7 Feb	0.08	0.25	0.15	0.33	0.61
8–10 Feb	0.014	0.044	1.297	0.32	29.48
11 Feb	0.065	0.088	0.005	0.74	0.06
12 Feb	0.063	0.049	0.046	1.29	0.94
13 Feb	0.005	0.014	0.230	0.36	16.43
14 Feb	0.080	0.063	0.149	1.27	2.37
8–14 Feb	0.04	0.05	0.62	0.74	12.49
15–17 Feb	0.135	0.341	0.278	0.40	0.82
18 Feb	4.865	3.282	4.478	1.48	1.36
19 Feb	4.723	2.874	2.938	1.64	1.02
20 Feb	1.588	1.213	2.044	1.31	1.69
21 Feb	0.251	0.198	0.340	1.27	1.72
15–21 Feb	1.69	1.23	1.52	1.38	1.24

Table A.2 Measured Effluent	Concentrations and	Concentration	Ratios 1969
Table A-2. Measureu Ennuent	Concenti ations and	Concenti ation	Nau05, 1707

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Table A-2. (Continued)						
1969	Measured	Pu concentration	ns (dpm $m^{-3}$ )	Concentration ratios		
Date	Main $1(C_1)$	Main 2 $(C_2)$	Main 3 $(C_3)$	$CR_4 (C_1 / C_2)$	$CR_6 (C_3 / C_2)$	
22–24 Feb	0.381	0.423	0.321	0.90	0.76	
25 Feb	0.467	0.458	0.831	1.02	1.81	
26 Feb	0.477	0.457	0.834	1.04	1.82	
27 Feb	0.346	0.507	0.627	0.68	1.24	
28 Feb	0.447	0.304	0.244	1.47	0.80	
22–28 Feb	0.41	0.43	0.50	0.96	1.17	
1–3 Mar	0.090	0.103	0.089	0.87	0.86	
4 Mar	0.259	0.283	0.475	0.92	1.68	
5 Mar	0.326	0.294	0.279	1.11	0.95	
6 Mar	0.128	0.246	0.175	0.52	0.71	
7 Mar	0.157	0.190	0.310	0.83	1.63	
1–7 Mar	0.16	0.19	0.22	0.86	1.14	
8–10 Mar	0.194	0.251	0.357	0.77	1.42	
11 Mar	0.190	0.190	0.094	1.00	0.49	
12 Mar	0.272	0.210	0.096	1.30	0.46	
13 Mar	0.931	0.907	0.918	1.03	1.01	
14 Mar	0.189	0.075	0.100	2.52	1.33	
8–14 Mar	0.31	0.31	0.33	1.01	1.07	
15–17 Mar	0.112	0.181	0.281	0.62	1.55	
18 Mar	0.204	0.220	0.389	0.93	1.77	
19 Mar	0.089	0.055	0.124	1.62	2.25	
20 Mar	0.202	0.079	0.148	2.56	1.87	
21 Mar	0.246	0.115	0.240	2.14	2.09	
15–21 Mar	0.15	0.14	0.25	1.06	1.72	
22–24 Mar	0.103	0.164	0.295	0.63	1.80	
25 Mar	0.163	0.128	0.209	1.27	1.63	
26 Mar	0.082	0.077	0.060	1.06	0.78	
27 Mar	0.005	0.005	0.085	1.00	17.00	
28 Mar	0.070	0.107	0.154	0.65	1.44	
28–31 Mar	0.071	0.146	0.305	0.49	2.09	
22–31 Mar	0.08	0.12	0.23	0.68	1.85	
1 Apr	0.224	0.317	0.239	0.71	0.75	
2 Apr	2.642	16.421	3.734	0.16	0.23	
3 Apr	0.099	0.910	0.313	0.11	0.34	
4–7 Apr	0.199	0.221	0.144	0.90	0.65	
8 Apr	0.152	0.172	0.194	0.88	1.13	
1–8 April	0.49	2.34	0.63	0.21	0.27	
9 Apr	0.397	0.489	0.619	0.81	1.27	
10 Apr	1.212	1.990	3.128	0.61	1.57	
11 Apr	0.191	0.400	0.141	0.48	0.35	
12–14 Apr	0.182	0.149	0.291	1.22	1.95	
15 Apr	0.243	0.078	0.106	3.12	1.36	
9–15 April	0.37	0.49	0.70	0.76	1.43	

Table A-2. (Continued)

1969	Measured	Pu concentration	as (dpm m <sup><math>-3</math></sup> )	Concentra	tion ratios
Date	Main 1 ( $C_1$ )	Main 2 ( $C_2$ )	Main 3 $(C_3)$	$CR_4 (C_1 / C_2)$	$CR_6 (C_3 / C_2)$
16 Apr	0.048	0.160	0.670	0.30	4.19
17 Apr	0.029	0.177	0.175	0.16	0.99
18 Apr	0.131	0.134	0.953	0.98	7.11
19–21 Apr	0.088	0.233	0.211	0.38	0.91
22 Apr	0.438	0.414	0.390	1.06	0.94
23 Apr	0.169	0.195	0.212	0.87	1.09
16–23 April	0.13	0.22	0.38	0.61	1.70
24 Apr	0.160	0.230	0.220	0.70	0.96
25 Apr	0.203	0.226	0.236	0.90	1.04
26–28 Apr	0.431	0.402	0.408	1.07	1.01
29 Apr	0.176	0.214	2.305	0.82	10.77
30 Apr	0.225	0.211	0.229	1.07	1.09
24–30 April	0.29	0.30	0.60	0.99	2.02
1 May	0.083	0.297	0.137	0.28	0.46
2 May	0.335	0.341	0.372	0.98	1.09
3–5 May	0.156	0.209	0.248	0.75	1.19
6 May	0.088	0.168	0.169	0.52	1.01
7 May	0.059	0.123	0.144	0.48	1.17
1–7 May	0.15	0.22	0.22	0.66	1.01
8–9 May	0.095	0.237	0.184	0.40	0.78
10-12 May	0.240	0.330	0.390	0.73	1.18
13 May	0.237	0.126	0.175	1.88	1.39
14 May	0.117	0.114	0.069	1.03	0.61
15 May	0.096	0.161	0.174	0.60	1.08
8–15 May	0.17	0.23	0.24	0.73	1.05
16 May	0.126	0.160	0.571	0.79	3.57
17–19 May	0.153	0.175	0.797	0.87	4.55
20 May	0.175	0.250	0.174	0.70	0.70
21-22 May	0.855	0.925	0.783	0.92	0.85
23 May	0.155	0.340	0.492	0.46	1.45
16–23 May	0.33	0.39	0.65	0.84	1.66
24–26 May	0.967	0.886	0.698	1.09	0.79
27 May	1.033	0.779	0.543	1.33	0.70
28 May	0.213	0.465	0.949	0.46	2.04
29 May	1.038	0.610	1.263	1.70	2.07
30–31 May	0.448	0.616	0.597	0.73	0.97
24–31 May	0.76	0.72	0.76	1.06	1.05
1–2 June	0.448	0.616	0.597	0.73	0.97
3 Jun	0.227	0.384	0.547	0.59	1.42
4 Jun	4.407	0.776	0.422	5.68	0.54
5 Jun	0.120	0.373	0.823	0.32	2.21
6 Jun	0.117	0.290	0.005	0.40	0.02
1–6 June	0.96	0.51	0.50	1.89	0.98

Table A-2. (Continued)

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Tuble II 2. (Continued)					
1969	Measured	Pu concentration	us (dpm m <sup>-3</sup> )	Concentra	tion ratios
Date	Main 1 ( $C_1$ )	Main 2 $(C_2)$	Main 3 $(C_3)$	$CR_4 (C_1 / C_2)$	$CR_{6} (C_{3} / C_{2})$
7–9 June	0.005	0.445	0.061	0.01	0.14
10 Jun	0.232	0.124	0.232	1.87	1.87
11 Jun	0.044	0.155	0.066	0.28	0.43
12 Jun	0.131	0.005	0.272	26.20	54.40
13 Jun	0.161	0.005	0.256	32.20	51.20
7–13 June	0.08	0.23	0.14	0.36	0.62
14–16 June	0.380	0.396	0.059	0.96	0.15
17 Jun	0.202	0.110	0.124	1.84	1.13
18 Jun	0.167	0.180	0.355	0.93	1.97
19 Jun	2.060	3.910	4.230	0.53	1.08
20 Jun	0.189	0.904	0.413	0.21	0.46
21–23 June	0.407	0.873	0.474	0.47	0.54
14–23 June	0.50	0.89	0.67	0.56	0.75
24 Jun	0.329	0.414	0.298	0.79	0.72
25 Jun	0.578	0.094	0.237	6.15	2.52
26 Jun	0.125	0.054	0.019	2.31	0.35
27 Jun	0.369	0.238	0.185	1.55	0.78
28–30 June	0.724	0.696	0.125	1.04	0.18
24–30 June	0.51	0.41	0.16	1.24	0.39
1 Jul	0.276	0.278	0.139	0.99	0.50
2 Jul	0.136	0.146	0.032	0.93	0.22
3 Jul	0.077	0.106	0.323	0.73	3.05
4–7 July	0.146	0.411	0.366	0.36	0.89
8 Jul	0.118	0.218	0.309	0.54	1.42
1–8 July	0.15	0.30	0.28	0.50	0.95
9 Jul	0.089	0.300	0.147	0.30	0.49
10 Jul	1.100	1.770	0.954	0.62	0.54
11 Jul	0.370	0.405	0.281	0.91	0.69
12-14 July	0.119	0.161	0.366	0.74	2.27
15 Jul	0.486	0.161	0.362	3.02	2.25
9–15 July	0.34	0.45	0.41	0.77	0.91
16 Jul	0.034	0.114	0.074	0.30	0.65
17 Jul	0.051	0.008	0.059	6.38	7.38
18 Jul	0.064	0.113	0.146	0.57	1.29
19–21 July	0.198	0.086	0.620	2.30	7.21
22 Jul	0.169	0.421	0.359	0.40	0.85
23 Jul	0.573	0.358	0.373	1.60	1.04
16–23 July	0.19	0.16	0.36	1.17	2.26
24 Jul	0.369	0.201	0.326	1.84	1.62
25 Jul	0.247	0.306	0.527	0.81	1.72
26–28 July	0.596	0.587	0.349	1.02	0.59
29 Jul	0.005	0.201	0.481	0.02	2.39
30 Jul	2.920	3.170	4.460	0.92	1.41
31 Jul	0.298	0.005	0.005	59.60	1.00
24–31 July	0.70	0.71	0.86	1.00	1.21

Table A-2. (Continued)

		1 4510 11 2	(continueu)		
1969	Measured	Measured Pu concentrations (dpm m <sup>-3</sup> ) Concentration ratios			tion ratios
Date	Main 1 ( $C_1$ )	Main 2 $(C_2)$	Main 3 $(C_3)$	$CR_4 (C_1 / C_2)$	$CR_6 (C_3 / C_2)$
1 Aug	0.641	1.023	0.745	0.63	0.73
2–4 Aug	0.602	0.257	0.379	2.34	1.47
5 Aug	0.296	0.264	0.568	1.12	2.15
6 Aug	0.272	0.190	0.367	1.43	1.93
7 Aug	0.032	0.187	0.155	0.17	0.83
8 Aug	0.222	0.229	0.396	0.97	1.73
1–8 Aug	0.41	0.33	0.42	1.23	1.26
9–11 Aug	0.271	0.498	0.229	0.54	0.46
12 Aug	0.135	0.132	0.487	1.02	3.69
13 Aug	0.090	0.351	0.203	0.26	0.58
14 Aug	0.087	0.094	0.472	0.93	5.02
15 Aug	0.42	0.134	0.39	3.11	2.88
9–15 Aug	0.22	0.32	0.32	0.70	1.01
16–18 Aug	0.322	0.106	0.169	3.04	1.59
19 Aug	0.522	0.076	0.604	6.87	7.95
20 Aug	0.116	0.040	0.283	2.90	7.08
21 Aug	0.145	0.075	0.034	1.93	0.45
22 Aug	0.059	0.084	0.051	0.70	0.61
16–22 Aug	0.26	0.08	0.21	3.05	2.49
23–25 Aug	0.342	0.196	0.197	1.74	1.01
26 Aug	0.204	0.289	0.175	0.71	0.61
27 Aug	0.106	0.297	0.145	0.36	0.49
28 Aug	0.203	0.309	0.465	0.66	1.50
29 Aug	0.268	0.114	0.111	2.35	0.97
30–31 Aug	0.362	0.368	0.303	0.98	0.82
23–31 Aug	0.28	0.26	0.23	1.08	0.90
1–2 Sept	0.362	0.368	0.303	0.98	0.82
3 Sep	0.410	0.625	0.546	0.66	0.87
4 Sep	0.221	0.128	0.991	1.73	7.74
5 Sep	0.083	0.120	0.080	0.69	0.67
6–8 Sept	0.242	0.188	0.128	1.29	0.68
1–8 Sept	0.27	0.27	0.33	1.00	1.20
9 Sep	0.037	0.123	0.072	0.30	0.59
10 Sep	0.331	0.370	0.461	0.89	1.25
11 Sep	0.156	0.834	0.238	0.19	0.29
12 Sep	0.358	0.559	0.275	0.64	0.49
13–15 Sept	0.231	0.391	0.560	0.59	1.43
9–15 Sept	0.23	0.44	0.39	0.51	0.89
16 Sep	0.136	0.296	0.402	0.46	1.36
17 Sep	0.150	0.509	0.438	0.29	0.86
18 Sep	0.447	0.029	0.662	15.41	22.83
19 Sep	0.250	0.219	0.662	1.14	3.02
20-22 Sept	0.088	0.090	0.146	0.98	1.62
16–22 Sept	0.18	0.19	0.37	0.94	1.97

 Table A-2. (Continued)

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1969	Measured	Pu concentration	ns (dpm $m^{-3}$ )	Concentra	tion ratios
Date	Main 1 (C <sub>1</sub> )	Main 2 $(C_2)$	Main 3 $(C_3)$	$CR_4 (C_1 / C_2)$	$CR_{6} (C_{3} / C_{2})$
23 Sep	0.187	0.248	0.005	0.75	0.02
24 Sep	0.282	0.482	0.312	0.59	0.65
25 Sep	0.278	0.188	0.021	1.48	0.11
26 Sep	0.065	0.119	0.210	0.55	1.76
27-29 Sept	0.106	0.329	0.243	0.32	0.74
30 Sep	0.065	0.114	0.071	0.57	0.62
23-30 Sept	0.149	0.267	0.169	0.56	0.63
1 Oct	0.152	0.101	0.094	1.50	0.93
2 Oct	0.066	0.076	0.067	0.87	0.88
3 Oct	0.065	0.078	0.108	0.83	1.38
4–6 Oct	0.100	0.347	0.139	0.29	0.40
7 Oct	0.042	0.097	0.084	0.43	0.87
8 Oct	0.195	0.217	0.196	0.90	0.90
1-8 Oct	t 0.10	0.20	0.12	0.51	0.60
9 Oct	0.055	0.063	0.044	0.87	0.70
10 Oct	0.077	0.100	0.127	0.77	1.27
11-13 Oct	0.151	0.240	0.194	0.63	0.81
14 Oct	0.034	0.117	0.038	0.29	0.32
15 Oct	1.5	2.6	1.8	0.58	0.69
9–15 Oct	t 0.30	0.51	0.37	0.59	0.72

Table A-2.	(Continu	ed)
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#### **APPENDIX B**

## CONCENTRATION RATIOS FOR DUCT CENTERLINE AND THREE-POINT EFFLUENT SAMPLING MEASUREMENTS

#### **B.1 CONCENTRATION RATIOS FOR DUCT CENTERLINE MEASUREMENTS**

Tables <u>B1</u> through <u>B7</u> contain data that describe the empirical distributions of concentration ratios ( $CR_4$  and  $CR_6$ ) for various ranges of the duct centerline plutonium concentration ( $C_2$ , dpm m<sup>-3</sup>). This information was used to construct histograms of the distributions for subsequent Monte Carlo calculations. Plots of the distributions of these concentration ratios were presented in <u>Section 3.3</u>.

Range of values for	Fraction of va	alues in range
concentration ratio	$CR_4$	$CR_6$
0–2	0.308	0.385
2–10	0.038	0.269
10–20	0.231	0.154
20-50	0.346	0.000
50-200	0.077	0.077
200-500	0.000	0.077
500-1000	0.000	0.038

Table B-1. Distribution of Concentration Ratios forDuct Centerline Concentrations C₂ ≤	Ξ
0.005 dpm m <sup>-3</sup>	

Table B2. Distribution of Concentration Ratios for Duct Centerline Concentrations 0.005 <  $$C_2 \le 0.1$ dpm m^{-3}$$ 

Range of values for	Fraction of values in range		
concentration ratio	$CR_4$	$CR_6$	
0-0.5	0.072	0.101	
0.5–1	0.159	0.145	
1–2	0.261	0.232	
2–4	0.290	0.261	
4–8	0.116	0.159	
8–16	0.043	0.014	
16–32	0.029	0.072	
32–64	0.014	0.014	
64–80	0.014	0.000	

Range of values for	Fraction of va	alues in range
concentration ratio	$CR_4$	$CR_6$
0–0.5	0.174	0.0978
0.5-1	0.315	0.185
1–2	0.283	0.424
2–4	0.130	0.163
4-8	0.087	0.0978
8–12	0.0109	0.0326

# Table B3. Distribution of Concentration Ratios for Duct Centerline Concentrations $0.1 < C_2 \leq 0.2 \ dpm \ m^{-3}$

Table B4. Distribution of Concentration Ratios for Duct Centerline Concentrations 0.2  $< C_2 \leq 0.5 \ dpm \ m^{-3}$ 

Range of values for	Fraction of values in range		
concentration ratio	$CR_4$	$CR_6$	
0-0.25	0.0862	0.0948	
0.25-0.5	0.216	0.103	
0.5-0.75	0.155	0.121	
0.75–1	0.241	0.155	
1–1.5	0.129	0.250	
1.5–3	0.138	0.172	
3–6	0.0259	0.0517	
6–8	0.00862	0.0259	
8–16	0.000	0.0259	

# Table B5. Distribution of Concentration Ratios for Duct Centerline Concentrations $0.5 < C_2 \leq 1 \ dpm \ m^{-3}$

Range of values for	Fraction of values in range		
concentration ratio	$CR_4$	CR <sub>6</sub>	
0–0.5	0.305	0.237	
0.5–1	0.288	0.339	
1–2	0.288	0.203	
2–4	0.0847	0.136	
4–8	0.0339	0.0847	

	2	•
Range of values for	Fraction of values in range	
concentration ratio	$CR_4$	CR <sub>6</sub>
0–0.5	0.139	0.250
0.5–1	0.500	0.194
1–2	0.250	0.278
2–4	0.111	0.222
4–8	0.000	0.0556

Table B6. Distribution of Concentration Ratios	for
Duct Centerline Concentrations $1 < C_2 \le 2$ dpm	m-3

Table B7. Distribution of Concentration 1	Ratios for
Duct Centerline Concentrations $C_2 > 2$ d	lpm m <sup>-3</sup>

Range of values for	Fraction of values in range	
concentration ratio	$CR_4$	CR <sub>6</sub>
0-0.25	0.20	0.12
0.25-0.5	0.20	0.04
0.5–1	0.32	0.24
1–1.5	0.08	0.52
1.5–2	0.20	0.04
2–6	0.00	0.04

#### **B.2 CONCENTRATION RATIOS FOR THREE-POINT MEASUREMENTS**

Tables <u>B-8</u> through <u>B-13</u> contain data that describe the empirical distributions of concentration ratios ( $CR_{4*}$ ,  $CR_{5*}$ , and  $CR_{6*}$ ) for various ranges of the mean measured plutonium concentration ( $C_{bar}$ , dpm m–3). This information was used to construct histograms of the distributions for subsequent Monte Carlo calculations. Plots of the distributions of the three concentration ratios for various ranges of  $C_{bar}$  were presented in <u>Section 3.5</u>.

Range of values for Fraction of values in range		ıge	
concentration ratio	$CR_{4*}$	<i>CR</i> <sub>5*</sub>	<i>CR</i> <sub>6*</sub>
0-0.33	0.111	0.361	0.250
0.33-0.50	0.0556	0.111	0.000
0.50-0.75	0.111	0.167	0.222
0.75–1.0	0.139	0.194	0.0833
1.0–1.33	0.0556	0.0833	0.0556
1.33-2.0	0.333	0.0556	0.0306
2.0-3.0	0.194	0.0278	0.0833

Table B-8. Distribution of Concentration Ratios for Mean Measured Concentrations  $C_{bar} \leq 0.1 \ dpm \ m^{-3}$ 

Table B-9. Distribution of Concentration Ratios for Mean Measured Concentrations  $0.1 < C_{bar} \leq 0.2 \ dpm \ m^{-3}$ 

Range of values for	Fraction of values in range		
concentration ratio	$CR_{4*}$	$CR_{5*}$	<i>CR</i> <sub>6*</sub>
0-0.33	0.250	0.107	0.143
0.33-0.50	0.0714	0.250	0.0357
0.50-0.75	0.107	0.143	0.107
0.75-1.0	0.250	0.107	0.143
1.0–1.33	0.143	0.143	0.321
1.33–2.0	0.107	0.0714	0.143
2.0–3.0	0.0714	0.179	0.107

Range of values for	Fraction of values in range		nge
concentration ratio	$CR_{4*}$	$CR_{5*}$	$CR_{6*}$
0-0.33	0.105	0.246	0.140
0.33-0.50	0.158	0.175	0.0877
0.50-0.75	0.175	0.0533	0.140
0.75-1.0	0.158	0.123	0.158
1.0-1.33	0.123	0.140	0.211
1.33-2.0	0.158	0.228	0.140
2.0-3.0	0.123	0.0351	0.123

Table B-10. Distribution of Concentration Ratios for Mean Measured Concentrations  $0.2 < C_{har} \le 0.5$  dpm m<sup>-3</sup>

Table B-11. Distribution of Concentration Ratios for Mean Measured Concentrations  $0.5 < C_{bar} \leq 1 \ dpm \ m^{-3}$ 

Range of values for	Fraction of values in range		
concentration ratio	$CR_{4*}$	$CR_{5*}$	$CR_{6^*}$
0–0.33	0.0909	0.121	0.0909
0.33-0.50	0.152	0.212	0.121
0.50-0.75	0.152	0.182	0.0303
0.75-1.0	0.212	0.0909	0.0909
1.0–1.33	0.242	0.152	0.212
1.33–2.0	0.152	0.212	0.303
2.0–3.0	0.000	0.0303	0.152

Table B-12. Distribution of Concentration Ratios for Mean Measured Concentrations  $1 < C_{bar} \leq 2 \ dpm \ m^{-3}$ 

Range of values for	Fraction of values in range		
concentration ratio	$CR_{4*}$	<i>CR</i> <sub>5*</sub>	$CR_{6*}$
0-0.33	0.163	0.140	0.0698
0.33-0.50	0.140	0.0930	0.000
0.50-0.75	0.233	0.279	0.0465
0.75-1.0	0.186	0.209	0.209
1.0–1.33	0.0930	0.140	0.186
1.33–2.0	0.163	0.116	0.349
2.0-3.0	0.0233	0.0233	0.140

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Range of values for	Fraction of values in range		
concentration ratio	$CR_{4*}$	$CR_{5*}$	$CR_{6*}$
0-0.33	0.125	0.125	0.0833
0.33-0.50	0.167	0.125	0.000
0.50-0.75	0.250	0.375	0.000
0.75-1.0	0.0833	0.167	0.167
1.0–1.33	0.292	0.125	0.167
1.33–2.0	0.0417	0.0833	0.458
2.0-3.0	0.0417	0.000	0.125

# Table B-13. Distribution of Concentration Ratios for Mean Measured Concentrations $C_{bar} > 2 \ dpm \ m^{-3}$

#### **APPENDIX C**

## DISTRIBUTIONS OF COMPUTED CONCENTRATION RATIOS FOR MONTHLY AVERAGE CONCENTRATIONS

#### C.1 DISTRIBUTIONS OF COMPUTED CONCENTRATION RATIOS FOR MONTHLY AVERAGE CENTERLINE CONCENTRATIONS

Tables <u>C-1</u> through <u>C-3</u> contain the icosatiles (the 0<sup>th</sup>, 5<sup>th</sup>, 10<sup>th</sup>, 15<sup>th</sup>... 100<sup>th</sup> percentiles) of distributions of monthly average concentration ratios ( $C_{am}$  /  $C_{2m}$ ) that were estimated for a variety of average duct centerline concentrations ( $C_{2m}$ , dpm m<sup>-3</sup>). The 0<sup>th</sup> and 100<sup>th</sup> percentile values are the lower and upper bounds of the distribution, respectively.

Concentration ratio ( $C_{am} / C_{2m}$ ) for specified range of $C_{2m}$ (dpm m <sup>-3</sup> )			
Percentile	$0.01 \leq C_{2m}$	$0.01 < C_{2m} \le 0.02$	$0.02 < C_{2m} \le 0.05$
Oth	0.87	0.28	0.18
5 <sup>th</sup>	2.9	0.94	0.89
10 <sup>th</sup>	3.7	1.2	1.2
15 <sup>th</sup>	4.4	1.4	1.5
20 <sup>th</sup>	5.0	1.7	1.8
25 <sup>th</sup>	5.7	1.9	2.0
30 <sup>th</sup>	6.5	2.1	2.2
35 <sup>th</sup>	7.4	2.4	2.4
40 <sup>th</sup>	8.1	2.7	2.6
45 <sup>th</sup>	8.9	2.9	2.9
50 <sup>th</sup>	10	3.2	3.3
55 <sup>th</sup>	11	3.6	3.7
60 <sup>th</sup>	13	3.9	4.1
65 <sup>th</sup>	15	4.4	4.5
70 <sup>th</sup>	18	5.1	5.1
75 <sup>th</sup>	23	5.8	6.0
80 <sup>th</sup>	30	7.3	7.1
85 <sup>th</sup>	38	10	9.3
90 <sup>th</sup>	53	16	16
95 <sup>th</sup>	79	25	25
100 <sup>th</sup>	260	56	77

Table C-1. Icosatiles of Distributions of Concentration Ratios (C<sub>am</sub> / C<sub>2m</sub>)

	Concentration ratio ( $C_{am} / C_{2m}$ ) for specified range of $C_{2m}$ (dpm m <sup>-3</sup> )			
Percentile	$0.05 < C_{2m} {\leq} 0.1$	$0.1 < C_{2m} \le 0.2$	$0.2 < C_{2m} \le 0.5$	
$0^{th}$	0.31	0.49	0.30	
$5^{\text{th}}$	1.2	1.0	0.72	
$10^{\text{th}}$	1.4	1.2	0.86	
15 <sup>th</sup>	1.6	1.3	0.98	
$20^{ ext{th}}$	1.9	1.4	1.1	
$25^{\text{th}}$	2.1	1.6	1.1	
$30^{\text{th}}$	2.3	1.7	1.2	
35 <sup>th</sup>	2.5	1.8	1.3	
$40^{ ext{th}}$	2.7	1.9	1.4	
$45^{\text{th}}$	2.9	2.0	1.5	
$50^{\text{th}}$	3.1	2.2	1.6	
55 <sup>th</sup>	3.5	2.4	1.7	
$60^{\text{th}}$	3.8	2.6	1.8	
$65^{\text{th}}$	4.2	2.8	1.9	
$70^{\text{th}}$	4.8	3.1	2.0	
75 <sup>th</sup>	5.5	3.5	2.2	
$80^{ ext{th}}$	6.3	3.9	2.4	
$85^{\text{th}}$	7.9	4.3	2.8	
$90^{\text{th}}$	13	5.0	3.3	
95 <sup>th</sup>	20	6.3	4.5	
100 <sup>th</sup>	48	10	14	

Table C-2. Icosatiles of Distributions of Concentration Ratios  $(C_{am}\,/\,C_{2m})$ 

Concentration ratio ( $C_{am} / C_{2m}$ ) for specified range of $C_{2m}$ (dpm m <sup>-3</sup> )			
Percentile	$0.5 < C_{2m} \leq 1$	$1 < C_{2m} \le 2$	$C_{2m} > 2$
$0^{th}$	0.29	0.23	0.15
$5^{\text{th}}$	0.71	0.71	0.43
$10^{\text{th}}$	0.85	0.82	0.61
$15^{\text{th}}$	0.95	0.92	0.72
$20^{\text{th}}$	1.01	1.01	0.87
25 <sup>th</sup>	1.11	1.08	0.96
$30^{\text{th}}$	1.18	1.16	1.04
35 <sup>th</sup>	1.25	1.25	1.10
$40^{\text{th}}$	1.34	1.34	1.16
$45^{\text{th}}$	1.44	1.40	1.22
$50^{\text{th}}$	1.54	1.51	1.28
55 <sup>th</sup>	1.66	1.62	1.36
$60^{ ext{th}}$	1.79	1.73	1.42
65 <sup>th</sup>	1.93	1.87	1.48
$70^{ ext{th}}$	2.10	2.00	1.56
75 <sup>th</sup>	2.31	2.20	1.63
$80^{ ext{th}}$	2.59	2.43	1.72
85 <sup>th</sup>	2.93	2.65	1.82
$90^{\text{th}}$	3.45	3.07	1.95
95 <sup>th</sup>	4.28	3.85	2.18
$100^{\text{th}}$	7.6	7.7	6.2

Table C-3. Icosatiles of Distributions of Concentration Ratios  $(C_{am}\!/C_{2m})$ 

#### C.2 DISTRIBUTIONS OF COMPUTED CONCENTRATION RATIOS FOR MONTHLY AVERAGES OF THREE-POINT CONCENTRATION MEASUREMENTS

Tables <u>C-4</u> and <u>C-5</u> contain the icosatiles (the 0<sup>th</sup>, 5<sup>th</sup>, 10<sup>th</sup>, 15<sup>th</sup> . . . 100<sup>th</sup> percentiles) of distributions of monthly average concentration ratios ( $C_{am} / C_{barm}$ ) that were estimated for a variety of average duct centerline concentrations ( $C_{barm}$  dpm m<sup>-3</sup>). The 0<sup>th</sup> and 100<sup>th</sup> percentile values are the lower and upper bounds of the distribution, respectively.

	Concentration ratio ( $C_{am} / C_{barm}$ ) for specified range of $C_{barm}$ (dpm m <sup>-3</sup> )		
Percentile	$C_{barm} \leq 0.1$	$0.1 < C_{barm} \leq 0.2$	$0.2 < C_{barm} \leq 0.5$
$0^{^{th}}$	0.97	0.94	0.91
5 <sup>th</sup>	1.11	1.08	1.10
$10^{\text{th}}$	1.16	1.14	1.15
$15^{\text{th}}$	1.19	1.19	1.18
$20^{\text{th}}$	1.23	1.22	1.21
$25^{\text{th}}$	1.25	1.25	1.24
$30^{\text{th}}$	1.28	1.28	1.27
$35^{\text{th}}$	1.30	1.30	1.30
$40^{\text{th}}$	1.32	1.33	1.33
$45^{\text{th}}$	1.35	1.35	1.35
$50^{\text{th}}$	1.38	1.38	1.38
$55^{\text{th}}$	1.40	1.41	1.41
$60^{\text{th}}$	1.42	1.44	1.43
$65^{\text{th}}$	1.45	1.47	1.46
$70^{\text{th}}$	1.48	1.50	1.49
$75^{\text{th}}$	1.52	1.55	1.52
$80^{ ext{th}}$	1.57	1.59	1.56
$85^{\text{th}}$	1.62	1.63	1.61
$90^{\text{th}}$	1.69	1.69	1.70
$95^{\text{th}}$	1.79	1.82	1.80
$100^{\text{th}}$	2.25	2.33	2.23

#### Table C-4. Icosatiles of Distributions of Concentration Ratios (C<sub>am</sub> / C<sub>barm</sub>)

	Concentration ratio ( $C_{am} / C_{barm}$ ) for specified range of $C_{barm}$ (dpm m <sup>-3</sup> )		
Percentile	$0.5 < C_{barm} \leq 1$	$1 < C_{barm} \le 2$	C <sub>barm</sub> > 2
$0^{ ext{th}}$	0.95	0.95	0.94
$5^{\text{th}}$	1.12	1.11	1.12
$10^{\text{th}}$	1.16	1.17	1.17
15 <sup>th</sup>	1.20	1.21	1.20
$20^{ ext{th}}$	1.23	1.25	1.23
25 <sup>th</sup>	1.26	1.28	1.26
30 <sup>th</sup>	1.28	1.31	1.29
35 <sup>th</sup>	1.31	1.33	1.32
$40^{\text{th}}$	1.34	1.36	1.34
$45^{\text{th}}$	1.37	1.39	1.38
$50^{\text{th}}$	1.40	1.43	1.41
55 <sup>th</sup>	1.44	1.45	1.45
$60^{\text{th}}$	1.46	1.49	1.48
65 <sup>th</sup>	1.50	1.52	1.52
$70^{\text{th}}$	1.54	1.56	1.56
75 <sup>th</sup>	1.58	1.60	1.60
$80^{ ext{th}}$	1.62	1.63	1.65
85 <sup>th</sup>	1.67	1.69	1.71
$90^{\text{th}}$	1.74	1.75	1.78
95 <sup>th</sup>	1.88	1.84	1.90
$100^{\text{th}}$	2.27	2.18	2.35

Table C-5. Icosatiles of Distributions of Concentration Ratios (C  $_{\rm am}$  / C  $_{\rm barm})$