APPENDIX A

AIRBORNE EFFLUENT PARTICLE SIZE STUDIES

APPENDIX A

AIRBORNE EFFLUENT PARTICLE SIZE STUDIES

Discussions of particle sizes rely on the use of the following terms:

- Aerodynamic equivalent diameter - the diameter of a unit density (1 g cm^{-3}) sphere with the same settling velocity as the particle in question. For PuO_2 particles, aerodynamic equivalent diameters are approximately three times actual particle diameters (Hayden, 1976).

- Mass median diameter - for a distribution of particles of various sizes, the mass median diameter identifies the size for which half the total *mass* of material collected is contributed by smaller particles and half by larger particles.

- Count median diameter - for a distribution of particles of various sizes, the count median diameter identifies the size for which half the total *number* of particles collected is contributed by smaller particles and half by larger particles.

- Activity median aerodynamic diameter - the median of the distribution of radioactivity or toxicological or biological activity with respect to aerodynamic diameter (Hinds, 1986). The AMAD is the diameter of a unit density sphere with the same terminal settling velocity in air as that of an aerosol particle whose activity is the median for the entire aerosol (USEPA, 1988).

The methods by which several of the above descriptors of particle size can be determined from sampling data are depicted in Figure A-1.

A number of studies of the particle size distribution of effluents from Rocky Flats were conducted in the early to mid-1970s. Several of these studies were authored by J. A. Hayden and are documented in Rocky Flats internal reports designated "Product and Health Physics Research Service Reports." A number of these studies are summarized in the following discussion and in Table A-1.

Building 776 exhaust was sampled in 1972 using 3 µm membrane filters, which were analyzed for particle size using the fission track method (Hayden *et al*., 1972a). The results of this study indicated that the count median diameter of particles observed in Building 776 effluent air was between 0.07 and 0.12 µm in general and between 0.12 and 0.15 µm in an exhaust system that appeared to be malfunctioning at the time. A recommendation was made that future collections be made with two filters arranged in tandem in order to allow for better quantitative estimates of material released.

Figure A-1

Table A-1

Samples of Building 776 effluent air were again obtained in 1972 over several 48-hour periods using 3 μm and 0.8 μm membrane filters (Hayden, 1972). The filters were again analyzed for particle size using the fission track method. The study concluded that the mass median diameter of particles observed in Building 776 effluent air was about 0.09 μm and that particle size distributions on the two types of filters were similar. However, a number of problems were noted with the sampling apparatus used in the study. For the 48-hour collection periods in this study, significant breakthrough of the millipore filter sampling media (millipore filters are not used for routine effluent sampling) occurred. An average of 50 percent of the total particles collected during 4 tests of the 3 μm filters were on the backup filter. During the one test of the 0.8 μm filter, 46 percent of the particles collected were found on the backup filter. No third filter assembly was used in these tests.

Samples of Building 771 effluent air were obtained in 1974 using membrane filters that were analyzed for particle size by the fission track method (Hayden, 1974). The results of this study indicated that

- The count median diameter of particles observed in Building 771 effluent air was between 0.09 and 0.19 μm.

- Certain upstream HEPA filters were leaking, resulting in a particle size distribution that was not lognormal. This indicated multiple sources for the sampled particles.

A review of Rocky Flats particle size data prepared in 1976 (Hayden, 1976) referred to the earlier particle sizing work done in Building 776 (Hayden, 1972) and stated some further conclusions:

- The measured mass median diameter of plutonium particles of 0.09 μm is equal to an aerodynamic equivalent diameter of 0.3 μm. This equivalency was based on a density of 11.45 g cm^{-3} for PuO_2 spheres. If the effluent was plutonium metal of density 19.8 g cm^{-3}, the aerodynamic equivalent diameter would have been about 0.4 μm.

- The observed aerodynamic equivalent diameter of 0.3 μm was considered to be consistent with the theory of operation of HEPA filters; that is, theory predicts that the filter media will be the least efficient for 0.3 μm particles. Prior to HEPA filtration, the effluent is expected to contain larger particles.

From the above studies, the best estimate of particle size distribution for particulate emissions that passed through HEPA filters (plutonium, uranium, etc.) is given by the size distribution that penetrates HEPA filters. Hayden (1976) reports that plutonium particles in Rocky Flats effluent air have a mean diameter of 0.09 micrometers and a very narrow size range (geometric standard deviation = 1.6). Taking 0.09 as a geometric mean particle size, and assuming a lognormal size distribution with geometric standard deviation 1.6, 99 percent of the particles are smaller than 0.27 micrometers.

Elsewhere, Hayden (1972) noted that virtually all of the particles collected in Building 776 air were less than 0.3 micrometers in size. Figure A-2 (5.2 from Attachment 1 to EG&G Report 93-RF-2657, "Determination of Particle Size Distribution and Composition of the Effluent Air Emissions from Building 559"; Nininger and Osborne, 1992) also shows a narrow, lognormal physical particle size distribution for particles passing through HEPA filters, with a median value of about 0.1 micrometer and a maximum size of about 0.3 micrometer.

Other studies were also identified that characterized the particle size of effluents at various points before they reached the final filtration step and were released to the environment. While this information is of limited interest in addressing environmental release and transport, it provides a more complete picture of the effluents generated by Rocky Flats processes prior to final stages of filtration. One of these studies involved particle size analyses on filters from the Building 707 continuous air monitoring (CAM) system after a plutonium oxide spill (Hayden *et al*., 1976). These analyses were conducted using fission track, alpha track, and optical and electron microscopy techniques. Although larger particles were present, most of the particles observed were less than 0.2 µm and were lognormally distributed.

Another study, conducted by Los Alamos researchers (Moss *et al*., 1961), looked at the particle size distributions of *unfiltered* plutonium aerosols resulting from various chemical, metal preparation, and fabrication processes. Results of this study indicated that

- Particle mass median diameters were quite small (on the order of 0.14 to 0.65 µm).
- Standard deviations of the observed distributions were very low (on the order of 1.3 to 1.9).
- Size distribution characteristics varied little from one operation to another.

Particle size analysis was also performed on samples of magnesium nitrate salts collected from between the fifth and sixth stages of the HEPA filtration system in Building 771 (Hayden, 1978). This study showed a count median diameter of 0.075 µm. For plutonium oxide, this is equal to a 0.25 µm aerodynamic equivalent diameter and is again consistent with the particle size distribution expected downstream of HEPA filters.

FIGURE A-2 Penetration measurement for a standard HEPA filter with no leakage

Another Los Alamos study looked at particle sizes of effluents from plutonium recovery activities in Building 771 and fabrication activities in Building 707 prior to final stages of HEPA filtration (Elder *et al*., 1974). The study found that particles from the fabrication operations (Building 707) were predominantly collected with activity median aerodynamic diameters in the range of 3.3 to 4.7 µm, which are larger than those from the previously discussed studies. However, significant quantities of much smaller particles were also present. The majority of the particles present in effluent from recovery activities in Building 771 had activity median aerodynamic diameters of less than 1.0 µm. The study made the observation that the recovery operations produced the highest activity and smallest aerosol size, presenting the most difficult air cleaning problem for a number of different facilities that were characterized in the study.

A 1992 particle sizing study examined particles in effluents from Building 559 (Nininger and Osborne, 1992). A laser-based airborne particle counter was used to count particles in a series of size ranges, the smallest being 0.3 to 0.5 μm. Results showed the particle size distribution to be strongly biased toward very small particles, particles smaller than the 0.3 μm size that could be detected with the laser particle counter. It is reported that well below 2 percent of the particles estimated to have been present would have had aerodynamic diameters larger than 5 μm. Alpha track analyses of filtered effluent revealed no significant alpha-emitting radioactivity, but did suggest the possible presence of some very small alpha-emitting particles. Isotopic analyses showed no quantifiable radioactivity present.

REFERENCES

Biermann and Bergman (1986). Biermann, A.M., and W. Bergman. "Filter Penetration Measurements Using the Condensation Nuclei Counter and the Aerosol Photometer." 1986 Conference on Chemical Defense Research, Aberdeen Provency Grounds, MD. November 15.

Elder *et al*. (1974). Elder, J. C., M. Gonzales, and H. J. Ettinger. "Plutonium Aerosol Size Characteristics." *Health Physics*, Vol. 27 (July), pp. 45-53, 1974. Repository Document RE-1178.

Hayden *et al*. (1972). Hayden, J. A., H. M. Baker, and R. A. Link. "Particle Size Analysis — PuO_2 in Building 776 Effluent Air Using the Fission Track Method." Report No. 317-72-165, July 17, 1972. Repository Document RE-1095.

Hayden (1972). Hayden, J. A. "Particle Size Analysis — Building 776 Effluent Air." Report No. 317-72-188, November 29, 1972. Repository Document RE-1094.

Hayden (1974). Hayden, J. A. "Particle Size Analysis, Building 771 Effluent Air." ES-376-74-118, May 17, 1974. Repository Document RE-1092.

Hayden (1976). Hayden, J. A., "Review of Particle Size Data Accumulated at Rocky Flats," ES-376-76-178, February 10, 1976. Repository Document RE-1091.

Hayden *et al*. (1976). Hayden, J. A., J. K. Fraser, and R. L. Murri. "Particle Size Analyses, Filters From 707 Building October 2, 1976." ES-376-76-194, November 2, 1976. Repository Document RE-1126.

Hayden (1978). Hayden, J. A. "Track Analysis, $Mg(NO_3)_2$ Salts Taken From HEPA Filter Plenum." ES-376-78-206, March 1, 1978. Repository Document RE-1090.

Hinds (1986). Hinds, W.C. "Data Analysis." In: *Cascade Impactor Sampling & Data Analysis*, J.P. Lodge Jr. and T.L. Chan, Eds. American Industrial Hygiene Association Monograph Series, AIHA, Akron, Ohio, 1986.

Moss *et al*. (1961). Moss, W. D., E. C. Hyatt, and H. F. Schulte. "Particle Size Studies on Plutonium Aerosols." *Health Physics*, Vol. 5, pp. 212-218, 1961.

Nininger and Osborne (1992). Nininger, R.C. and W.E. Osborne. "Determination of Particle Size Distribution and Composition of the Effluent Air Emissions from Building 559." Prepared by EG&G Rocky Flats for the USDOE for submittal to the USEPA. Report 93-RF-2657. November, 1992.

USEPA (1988). U.S. Environmental Protection Agency. "Limiting Values of Radionuclide Intake and Air Concentration and Dose Conversion Factors for Inhalation, Submersion, and Ingestion." Federal Guidance Report No. 11. Prepared by Oak Ridge National Laboratory for the USEPA Office of Radiation Programs.

APPENDIX B

SAMPLING LINE DEPOSITION LOSS CALCULATIONS

APPENDIX B
SAMPLING LINE DEPOSITION LOSS CALCULATIONS

Deposition losses for representative sampling lines at Rocky Flats were calculated using a computer program based on a method used by Voillequé *et al.* (1991). The following discussion outlines the approach used to estimate line deposition losses presented in Section 2.0.

The Reynolds number (Re) for a sampling line is defined as:

$$Re = \frac{v\ \delta\ \rho_a}{\eta}$$

where v is the exhaust gas velocity (cm sec^{-1}), δ is the inside diameter of the sampling line (cm), ρ_a is the density of the exhaust gas (g cm^{-3}), and η is the viscosity of the exhaust gas (dyne s cm^{-2}). The Reynolds numbers for the sampling lines included in this analysis exceed 4,000. Their flow is therefore classified as turbulent, and transport of particles from the sampled air stream to the sampling line wall by turbulent diffusion is much more important that transport by Brownian diffusion. Gravitational settling is not important because of the brief transport time through horizontal sections of the line.

Vincent (1989) summarized results from studies of deposition in lines under turbulent conditions. The equation for the transmission factor corresponding to deposition loss (TF_D) is:

$$TF_D = e^{\left[-4\ \frac{w}{v}\ \frac{L}{\delta}\right]}$$

where *w* is the deposition velocity (cm s^{-1}) for the particles in the sampling line, *L* is the length (cm) of the sampling line, *v* is again the stack gas velocity (assuming isokinetic sampling where sampling line velocity equals stack gas velocity), and δ is again the inside diameter of the sampling line.

Using a figure from Liu and Agarwal (1974), Vincent found satisfactory agreement among three theoretical approaches and experimental data by plotting normalized deposition velocity (*w**) against a normalized relaxation time (τ*) that reflects particle size. The dimensionless normalized parameters are:

$$w^* = \frac{w}{\sqrt{\frac{f}{2}}\ v}$$

$$\tau^* = \frac{\tau\ \rho_a \left(\frac{f}{2}\right)\ v^2}{\eta}$$

and

where τ is the relaxation time and f is the Fanning friction factor. The relaxation time is defined by:

$$\tau = \frac{\rho\ d^2\ C_c}{18\eta}$$

For Reynolds numbers less than 10^5, Perry *et al*. (1984) give the following expression for the friction factor:

$$f = 0.0791\ Re^{-0.25}$$

Approximations to the theoretical relationships between w^* and τ^* presented in Appendix G to the Draft Interim Task 2 and 3 Report for the Fernald Dosimetry Reconstruction Project (Voillequé *et al*., 1991) were used for these calculations. For τ^* between 0.1 and 10:

$$w^* = (5.40\times10^{-4})\ (\tau*)^{0.3178}$$

For τ^* between 10 and 300:

$$w^* = (2.45\times10^{-2})\ (\tau*)^{0.3178}$$

The normalized deposition velocity w^* is approximately constant at 0.15 for τ^* greater than 300.

The methods described above were used to estimate line deposition losses for:

- A 108-inch sampling line, with inside diameter of 0.43 inches and a flow rate of 688 cubic centimeters per second ($cm^3\ sec^{-1}$). This corresponds to the longest lines tested by Mossoni and Kittinger (1973).

- Lines with inside diameters of 0.402 inches, with lengths and flow rates of 31.75 inches and 950 $cm^3\ sec^{-1}$, 25.75 inches and 967 $cm^3\ sec^{-1}$, and 14 inches and 983 $cm^3\ sec^{-1}$. These parameters correspond to the lines where Mossoni and Kittinger measured the greatest deposition.

Particle deposition increases with particle size. Almost all of the particles in Rocky Flats filter plenum exhaust effluent have physical particle diameters smaller than 0.3 micrometers. As shown in Table B-1, calculated deposition losses of 0.3 micrometer particles in the sampling lines were less than 0.1 percent in all the above cases. This is fully consistent with the claim by Mossoni and Kittinger that errors due to sampling line losses were less than 10 percent of the measured concentrations.

REFERENCES

Liu and Agarwal (1974). Liu, B.Y.H., and J.K. Agarwal. "Experimental Observation of Aerosol Deposition in Turbulent Flow," *J. Aerosol Sci*. 5:145-155.

Mossoni and Kittinger (1973). Mossoni, N.J., and W.D. Kittinger. "The Deposition of Particulates in Air Sampling Tubes," HPO-73-001, April 30, 1973. Repository Document RE-1093.

Perry *et al*. (1984). Perry, R.H., D.W. Green, and J.O. Maloney (eds.). *Perry's Chemical Engineers' Handbook*. Sixth Edition. New York: McGraw Hill Book Company.

Vincent (1989). Vincent, J.H., *Aerosol Sampling Science and Practice,* John Wiley & Sons.

Voillequé *et al*. (1991). Voillequé, P.G., K.R. Meyer, D.W. Schmidt, G.G. Killough, R.E. Moore, V.I. Ichimura, S.K. Rope, B. Shleien, and J.E. Till. "The Fernald Dosimetry Reconstruction Project, Tasks 2 and 3, Radionuclide Source terms and Uncertainties— 1960-1962," Draft Interim Report for Comment. Radiological Assessments Corporation, Neeses, SC. December 1991.

TABLE B-1

SAMPLING LINE DEPOSITION LOSS CALCULATION RESULTS

Line Length (inches)	Inside Diameter (inches)	Volumetric Flow Rate (cm^3 sec^{-1})	Percent Loss of 0.3 μm Particles
108	.43	688	0.03
31.75	.402	950	0.04
25.75	.402	967	0.03
14	.402	983	0.02

APPENDIX C

ANALYSIS OF ANISOKINETIC SAMPLING ERRORS

APPENDIX C
ANALYSIS OF ANISOKINETIC SAMPLING ERRORS

Accurate sampling of airborne particles often requires that the fluid velocity in the sampling probe (u, cm s^{-1}) be the same as the velocity of the stack gas at the point of sampling (v, cm s^{-1}). When these velocities are matched, the sampling is termed "isokinetic." Deviations from this condition, anisokinetic sampling, can lead to bias in the sample. The bias may result in underestimation or overestimation of particle concentrations, depending upon whether the sampling flow rate yields a probe fluid velocity $u < v$ or $u > v$.

Errors introduced by anisokinetic sampling were estimated using a computer program based on the method described on pages G-1 and G-2 of Appendix G to the Draft Interim Task 2 and 3 Report for the Fernald Dosimetry Reconstruction Project (Voillequé *et al.*, 1991). The following outline of the approach closely follows the discussion in that report.

Durham and Lundgren (1980) developed a method to assess effects of deviations from isokinetic sampling conditions. The consequences of anisokinetic sampling depend on the isokinetic ratio (the ratio of the fluid velocities u/v), the size and densities of the particles sampled, the diameter of the sampling probe, and, to a much lesser extent, the air temperature.

Improper alignment of the sampling probe along the streamlines of flow in the stack can also lead to sampling biases. However, if the probe axis is within 15° of the proper position, the effects of misalignment are small, (about 5 percent or less, Durham and Lundgren, 1980). This analysis assumes that alignment of sampling probes was sufficiently accurate to make misalignment bias small compared with measurement uncertainties resulting from anisokinetic sampling.

For a properly aligned sampling probe, the ratio R of the sampled concentrations of particulates to the concentrations in the stack is given by:

$$R = 1 + \left[\frac{v}{u} - 1\right]\left[1 - \frac{1}{k}\right]$$

with

$$k = 1 + \left(2 + 0.62\frac{u}{v}\right) Stk \qquad \text{(C-1)}$$

where *u* and *v* are the velocities defined above and *Stk*, the Stokes number for the particles, is given by:

$$Stk = \frac{\rho\, d^2\, C_c\, v}{18\eta\delta} \qquad \text{(C-2)}$$

where ρ and *d* are the density (g cm^{-3}) and physical diameter (cm) of the particles, respectively, C_c is the dimensionless Cunningham slip correction factor for the particle, η is the viscosity of the exhaust air (dyne s cm^{-2}) and δ is the diameter (cm) of the probe opening. The factor C_c is calculated from the empirical equation given by Hinds (1982):

$$C_c = 1 + \frac{2\,[\,6.32 + 2.01\, e^{-0.1095\, P\, d\, 10^4}\,]}{P\, d\, 10^4} \qquad \text{(C-3)}$$

where *d* is the physical diameter of the particles, *P* is the absolute pressure (cm Hg), and the factor of 10^4 converts from cm to µm.

Conditions prevailing when deviations from isokinetic sampling occurred are unknown, so the following representative conditions were assumed:

Air temperature:	20°C
Air pressure (P):	76 cm Hg
Air viscosity (η):	1.81 x 10^{-4} dyne s cm^{-2}
Air density (ρ_a):	1.2 x 10^{-2} g cm^{-3}

Data on stack sampling systems at Rocky Flats for Analysis

Repository Document RE-1029 "Duct Measurements and Velocities; Sample System Diameters and Velocities" (Author unknown, date unknown) provides historical information on stack sampling systems for 51 exhaust vents at Rocky Flats. In addition, Rocky Flats submitted 37 Duct Assessment Reports (DARs) to the U.S. EPA on December 11, 1992. The DARs describe the present condition of the 63 radionuclide emission points at Rocky Flats. ChemRisk reviewed the DARs for the following representative effluent ducts:

771-MAI - the main exhaust from Building 771 through the 145-foot tall stack;

444-MAI - the main exhaust from Building 444;

444-DOS - the Building 444 beryllium shop exhaust;

447-MAI - the main exhaust from Building 447;

883-AAA - BBB and CCC - the Building 883 exhausts.

Each of the DARs reviewed says that "Effluent sample is extracted through each sample probe at a rate of 2 dry standard cubic feet per minute (dscfm)." Reynolds numbers in all of these ducts were in excess of 400,000, corresponding to turbulent flow, and the DARs claim this results in a homogeneous distribution of particles in the effluent stream. The DARs reviewed indicate that there is one sampling nozzle per sampling probe except in the 771-MAI duct. Duct flow rates and velocities given in the DARs are not the same as those given in Repository Document RE-1029, but this is to be expected because flow rates in the exhaust systems differed at different times in the history of Rocky Flats operations.

The largest flow rate listed in Repository Document RE-1029 is for exhaust system 771-MAI (the main exhaust through the Building 771 stack), with a flow rate of 203,344 cubic feet per minute. The total exhaust volume from all vents listed in Repository Document RE-1029 is about 1,466,000 cubic feet per minute, excluding the exhaust volume from Building 371 (lines 51 and 52) because that building never became operational. Therefore, the 771 stack, which released about 90 percent of the plutonium from normal operations at Rocky Flats, was responsible for about 15 percent of the total exhaust air volume (excepting Building 371) tabulated in Document RE-1029.

The "pitot diameter" tabulated in Repository Document RE-1029 for each exhaust vent is believed to be the sampling tube inside diameter (i.d.), because the listed sample velocity for all exhaust vents corresponds to a sampling rate of 2 cubic feet per minute if the pitot diameter is used as the sampling tube inside diameter. The sample velocity of 1956 ft min^{-1} for the 771-MAI system is 1/3 of the sampling line velocity of 5867 ft min^{-1} calculated for the listed 0.25-inch sampling line for sampling at a rate of 2 cubic feet per minute. This is consistent if three 0.25-inch i.d. sampling nozzles were feeding each of the three sampling lines on the 771 MAI system, and the sampling lines also had an i.d. of 0.25 inch. Otherwise, if 2 cubic feet per minute were drawn through each sampling nozzle, the sample velocity in each nozzle would be 5867 ft min^{-1}. The DAR for the 771-MAI system reports sample nozzle inside diameters of 0.125 inch each for the three sample nozzles on each of the three sampling lines. Mr. W. Osborne of Rocky Flats (telephone conversation on 5/13/93) stated that the sampling rate is 2 cubic feet per minute in each of the three nozzles on the sampling lines, consistent with the isokinetic ratio of 12.17 reported in the 771-MAI DAR.

All pitot diameters in Repository Document RE-1029 are multiples of 0.125 inch. However, Mossoni and Kittinger reported that the sampling lines they studied in Buildings 707, 771, and 776 were between .305 and .430 inches inside diameter, and sampling line inside diameters in six of the seven DARs reviewed were not multiples of 0.125 inch. Discrepancies between the DARs reviewed and Repository Document RE-1029, for exhaust ducts other than 771-MAI, were as follows:

444-MAI: sample probe i.d. = 0.456 inch compared to 0.5 inch in RE-1029; duct cross-sectional area = 72 ft^2 as compared to 64 ft^2 in RE-1029;

444-DOS: sample probe i.d. = 0.4 inch, compared to 0.5 inch in RE-1029;

447-MAI: sample probe i.d. = 0.456 inch, compared to 0.625 inch in RE-1029;

883-AAA: sample probe i.d. = 0.402 inch, compared to 0.5 inch in RE-1029; duct cross-sectional area = 35.4 ft^2, compared to 48 ft^2 in RE-1029.

883-BBB: sample probe i.d. = 0.402 inch, compared to 0.5 inch in RE-1029; and

883-CCC: sample probe i.d. = 0.313 inch, compared to 0.5 inch in RE-1029; duct cross-sectional area where stack samples taken = 17.4 ft^2 and exhaust stack area = 12.6 ft^2, compared to duct cross-sectional area 12 ft^2 in RE-1029.

Mossoni and Kittinger (1973) reported sampling rates of 1.4 cubic feet per minute in the Building 771 lines tested, 1.5 cubic feet per minute in the Building 707 lines tested, and between 1.9 and 2.1 cubic feet per minute in the Building 776 lines tested.

Effects of anisokinetic sampling at Rocky Flats

Hayden (1976) reports that plutonium particles in Rocky Flats effluent air have a mean diameter of 0.09 µm and a very narrow size range (geometric standard deviation = 1.6). Taking 0.09 as the geometric mean particle size, and assuming a long-normal size distribution with geometric standard deviation 1.6, 99 percent of the particles are smaller than 0.27 µm. Elsewhere, Hayden (1972) noted that virtually all of the particles collected in Building 776 effluent air were less than 0.3 µm in size. Figure 5.2 in Attachment 1 to EG&7G Report 93RF-2657 "Determination of Particle Size Distribution and Composition of the Effluent Air Emissions from Building 559" (Nininger and Osborne, 1992) also shows a narrow log-normal particle size distribution for particles passing through HEPA filters, with a median value of about 0.1 µm and a maximum size of about 0.3 µm.

Errors introduced by anisokinetic sampling were estimated using a particle density of 11.46 g cm^{-3}, corresponding to plutonium dioxide, and a computer program to perform the calculations described

above. About 90 percent of the plutonium released from normal operations in buildings at Rocky Flats came from the Building 771 stack (effluent duct 771-MAI), and the possible effects of anisokinetic sampling in duct 771-MAI can be summarized as follows:

- With duct velocity 2542 ft min^{-1} and sampling line velocity 1956 ft min^{-1}, as reported in Repository Document RE-1029, the isokinetic ratio is 0.77. This would occur if three 0.25-inch i.d. sample nozzles fed each sampling line, the sampling line i.d. was also 0.25 inch and the sample was drawn at 2 cubic feet per minute through the sampling line. Under these conditions, the measured sample concentration is less than 1 percent higher than the duct concentration for the maximum particle diameter of 0.3 µm, but only 0.2 percent higher than the duct concentration for the median particle diameter of 0.1 µm. If the sampling rate were to drop to 1.4 cubic feet per minute, the isokinetic ratio would be 0.54. The measured sample concentration would then be 2 percent higher than the duct concentration for the maximum particle diameter of 0.3 µm, but only 0.4 percent higher than the duct concentration for the median particle diameter of 0.1 µm.

- If the duct velocity were 2542 ft min^{-1}, as reported in Repository Document RE-1029, and 2 cubic feet per minute were drawn through each sample nozzle, the sampling velocity would be 5867 ft min^{-1}. Under these conditions, the isokinetic ratio is 2.3 and the measured sample concentration is less than 2 percent lower than the duct concentration for the maximum particle diameter of 0.3 µm, and less than 0.5 percent lower than the duct concentration for the median particle diameter of 0.1 µm. If the sampling rate were to drop to 1.4 cubic feet per minute, the isokinetic ratio would be 1.6, and the measured sample concentration would again be less than 2 percent lower than the duct concentration for the maximum particle diameter of 0.3 µm, and less than 0.5 percent lower than the duct concentration for the median particle diameter of 0.1 µm.

- The 771-MAI DAR says the exhaust velocity was 17.1 ft sec^{-1} at a point in the 771 stack where the cross-sectional area is 150.3 ft^2. Using the hydrodynamic continuity equation Q=AV, where Q is the flow rate, A is the cross-sectional area of the conduit and V is the flow velocity, the exhaust velocity was 32.1 ft sec^{-1} (1926 ft min^{-1}) at the stack sampling locations where the cross-sectional area of the exhaust duct is 80 ft^2 The sampling nozzle inside diameters are 0.125 inch and each nozzle samples at a rate of 2 cubic feet per minute (Osborne, 1993), resulting in the isokinetic ratio of 12.2 reported in the DAR. Under these conditions, the measured sample concentration is 12 percent lower than the duct concentration for the maximum particle diameter of 0.3 µm, but only 3 percent lower than the duct concentration for the median particle diameter of 0.1 µm. If the sampling rate were 1.4 cubic feet per minute, the isokinetic ratio is 8.5, the measured sample concentration is 9 percent lower than the duct concentration for the maximum particle diameter of 0.3 µm and 2 percent lower than the duct concentration for the median particle diameter of 0.1 µm.

- Historically, the maximum average flow in the main 771 exhaust duct was about 8.5 million cubic meters per day (3,500 cubic feet per second), corresponding to a duct velocity of 43.4 ft sec^{-1} (2604 ft min^{-1}) at the stack sampling location where the duct cross-sectional area is 80 ft^2. Assuming sample nozzle and sample line inside diameters of 0.125 inch and a sampling rate of 2 cubic feet per minute in each nozzle, the isokinetic ratio is 9. Under these conditions, the measured sample concentration is 12 percent lower than the duct concentration for the maximum particle diameter of 0.3 µm, and 3 percent lower than the duct concentration for the median particle diameters of 0.1 µm. If the sampling rate were 1.4 cubic feet per minute, the isokinetic ratio is 6.3, the measured sample concentration is 9 percent lower than the duct concentrations for the maximum particle diameter of 0.3 µm and 2 percent lower than the duct concentration for the median particle diameter of 0.1 µm.

Duct velocities and sampling probe inside diameters in the six DARs reviewed for buildings other than 771 indicated superisokinetic sampling with the sample concentration underestimating the stack concentration by less than 0.4 percent for 3 µm diameter particles, with a smaller error for 0.1 µm particles. The anisokinetic sampling errors for the duct velocities and sampling probe inside diameters for building other than Building 371 tabulated in Repository Document RE-1029 were 2 percent or less for 0.1 µm particles, the median particle size in Rocky Flats exhausts. For sampling rates between 1.4 and 2 cubic feet per minute, the maximum error for 0.3 µm particles, the approximate upper bound on the particles that pass through HEPA filters, was a measured sample concentration 8 percent higher than the duct concentration.

These results suggest that errors introduced by anisokinetic sampling in the estimation of radionuclide releases from Rocky Flats were of the order of 5 percent or less.

REFERENCES

Durham and Lundgren (1980). Durham, M.D., and D.A. Lundgren. "Evaluation of Aerosol Aspiration Efficiency as a Function of Stokes Number, Velocity Ratio, and Nozzle Angle." *J. Aerosol Sci.* 11:179-188.

Hayden (1972). Hayden, J.A., H.M. Baker, and R.A. Lind. "Particle Size Analysis — PuO_2 in Building 776 Effluent Air Using the Fission Track Methods." Report No. 317-72-165, July 17, 1972. Repository Document RE-1095.

Hayden (1976). Hayden, J.A.. "Review of Particle Size Data Accumulated at Rocky Flats." ES-376-76-178, February 10, 1976. Repository Document RE-1091.

Hinds (1982). Hinds, W.C., *Aerosol Technology, Properties, Behavior, and Measurement of Airborne Particles*. New York: John Wiley & Sons.

Mossoni and Kittinger (1973). Mossoni, N.J., and W.D Kittinger. "The Deposition of Particulates in Air Sampling Tubes." HPO-73-001, April 30, 1973. Repository Document RE-1093.

Nininger and Osborne (1992). Nininger, R.C., and W.E. Osborne. "Determination of Particle Size Distribution and Composition of the Effluent Air Emissions from Building 559." Prepared by EG&G Rocky Flats for the USDOE for submittal to the USEPA. Report 93-RF-2657. November, 1992.

Osborne (1993). Personal Communication with W. Osborne of the Rocky Flats Plant, May 13, 1993.

Voillequé *et al*. (1991). Voillequé, P.G., K.R. Meyer, D.W. Schmidt, G.G. Killough, R.E. Moore, V.I. Ichimura, S.K. Rope, B. Shleien, and J.E. Till. "The Fernald Dosimetry Reconstruction Project, Tasks 2 and 3, Radionuclide Source terms and Uncertainties— 1960-1962." Draft Interim Report for Comment. Radiological Assessments Corporation, Neeses, SC. December 1991.

APPENDIX D

ISOTOPIC COMPOSITION OF ALPHA-EMITTING EFFLUENTS

APPENDIX D

ISOTOPIC COMPOSITION OF ALPHA-EMITTING EFFLUENTS

This appendix discusses the composition of the primary radioactive metals that were used at the Rocky Flats Plant and methods that can be used to characterize the isotopic composition of historical radioactive effluents from the plant.

D.1 Rocky Flats Plutonium

The materials of concern identified in Task 2 of this project included individual isotopes of plutonium, that is, Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242, as well as Am-241. Rocky Flats plutonium has contained the those radionuclides in the proportions reflected in Table D-1. The mass percentages in Table D-1 are averages over a two-year period from July 1976 to July 1978 (USDOE, 1980). The ranges of percentages by mass indicate the variability of isotopic content (Del Pizzo *et al.*, 1970). The isotopic fractions of alpha activity are presented for total long-lived alpha-emitting radioactivity and for Pu-239/240, which are both quantities that have been measured for airborne emissions. As a result, these fractions are useful for translating nonspecific or multiple isotope effluent measurements to releases of specific radionuclides.

Pu-241, which emits beta particles but no alpha particles, typically comprises over 80 percent of the total (alpha plus beta) radioactivity of Rocky Flats plutonium (USDOE, 1980). The multiples of total alpha activity and Pu-239/240 alpha activity that Pu-241 typically equals are useful in estimating quantities of the beta-emitter released based on measured activities.

On weight and alpha activity bases, Pu-239 and Pu-240 would be expected to make up nearly all of the plutonium in Rocky Flats airborne effluents. While alpha spectroscopy to specifically identify alpha emitters was practiced on environmental samples as early as the late 1950s or 1960 (Ray and Hammond, 1960), routine isotopic analyses of effluent sample filters did not start until around 1973. Specific measurement of Pu-239 and Pu-240 in Rocky Flats airborne effluents began in July 1973 (Dow, 1974). The alpha particles emitted from Pu-239 and Pu-240 are so similar in energy that the isotopes cannot be separately quantified by the alpha spectrometric method used to analyze plutonium effluent samples. Because of this, plutonium emissions since 1973 have been reported as "radiochemically determined Pu-239 plus Pu-240."

The contributions of Pu-238 to Rocky Flats plutonium emissions were first reported around 1986; they have been included in DOE's Effluent Information System since 1986 and were first included in Rocky Flats annual environmental reports in 1990.

TABLE D-1

TYPICAL ISOTOPIC COMPOSITION OF ROCKY FLATS PLUTONIUM

Isotope	Percentage by Mass[1]	Range of Mass Percentage[2]	Specific Activity in Rocky Flats Plutonium[1] (Ci g^{-1})	Percentage of Total Alpha Activity[3]	Percentage of Pu-239/240 Alpha Activity[3]
Pu-238	0.01	0.03 - 0.05	0.00171	1.79 - 2.07	2.39
Pu-239	93.79	92.84 - 93.84	0.05834	61.1 - 70.7	81.5
Pu-240	5.80	5.5 - 6.5	0.01322	13.8 - 16.0	18.5
Pu-241	0.36	Not Given	0.37260* (beta activity)	390 - 451* (beta activity)	521** (beta activity)
Pu-242	0.03	Not Given	0.00000118	0.00124 - 0.00143	0.00165
Am-241	Not Given	Not Given	0.00930 - 0.0222[4]	11.3 - 23.2	13 - 31[6]
Plutonium-239 plus Plutonium-240[7]			0.0716	75.0 - 86.7	100
Total of Alpha-Emitting Plutonium Isotopes[7]			0.0733	76.8 - 88.7	102
Total of All Alpha-Emitting Radionuclides[7]			0.0826 - 0.0955	100	115 - 133

* Pu-241 is a beta emitter. It does not emit alpha particles. In Rocky Flats plutonium it emits beta particles at 3.82 to 4.51 times the rate that alpha-emitting nuclides present emit alpha particles.

** Pu-241 present in Rocky Flats plutonium emits beta particles at 5.21 times the rate that the Pu-239 and Pu-240 present emit alpha particles.

REFERENCES/SOURCES:

1 Rockwell, 1989.
2 Del Pizzo *et al*., 1970.
3 Calculated from the specific activities of the individual nuclides in Column 4.
4 Calculated based on the Column 6 range of Am-241 fraction of Pu-239/240 activity and the Pu-239 and Pu-240 specific activities in Column 4.
5 Calculated based on the Column 4 range of Am-241 specific activity and the sum of alpha-emitting plutonium isotope specific activities from Column 4.
6 Estimated based on Am-241 and Pu-239/240 measurements in Rocky Flats airborne effluents from 1985 through 1989.
7 Values in this row were calculated based on Column 4 specific activity values for the individual radionuclides.

Am-241 is a decay product of Pu-241, and as such has been present in the plutonium handled at Rocky Flats since the early 1950s. The americium to plutonium activity ratio has reportedly ranged from 10 percent to 20 percent (USDOE, 1980). As an emitter of alpha and gamma radiations, americium would have contributed to the total long-lived alpha measurements made of particulate filters from plutonium production areas. Although the alpha spectroscopy procedure used for airborne samples since 1973 has separated plutonium, uranium, and americium content into separate specimens for analysis, Am-241 emissions were not reported until 1985 because of problems with performance of the laboratory method (Hornbacher, 1975-1982). Am-241 release totals were reported in USDOE's Effluent Information System starting in 1985 and in Rocky Flats annual environmental reports since 1988.

In order to translate measurements of total long-lived alpha radioactivity from plutonium facilities and more recent combined Pu-239/240 measurements to emission totals for the specific radionuclides listed in Table D-1, a number of relationships must be characterized:

- The fractions of total long-lived alpha radioactivity from plutonium facilities that were actually Pu-238, Pu-239, Pu-240, Pu-242, and Am-241 must be estimated.

- The ratio of relatively short-lived beta-emitting Pu-241 (13-year half-life) to measured total alpha or Pu-239/240 activities must be determined.

- The relative quantities of Pu-239 and Pu-240 historically present must be estimated to translate combined alpha spectral measurements to individual release totals.

Each of these relationships can be described to some extent based on available site-specific information and generic information characterizing compositions of special nuclear materials historically encountered within the U.S. nuclear weapons complex. For this project, source terms are provided for Pu-241, Am-241, and plutonium alpha activity. Given the relatively similar physical and toxicological properties of the alpha emitting plutonium isotopes, taking the source term analyses to the level of individual alpha-emitting plutonium isotopes would result in little benefit in terms of dose assessment, at the cost of introducing additional uncertainty into the process.

Figures D-1 and D-2 present inhalation and ingestion dose coefficients for the plutonium isotopes and for Am-241 based on ICRP Report 56 (ICRP, 1989) and ICRP Report 30 methodology as applied in DOE/EH-0071 (USDOE, 1988). ICRP Publication 56 presents age-dependent dose coefficients that reflect dose delivered to age 70 from an intake at various ages, while

Insert Figure D-1 Inhalation Dose Factors For Plutonium and Americium

Insert Figure D-2 Ingestion Dose Factors for Plutonium and Americium

DOE/EH-0071 factors correspond to doses delivered over a 50-year period after intake by an adult. The doses that are calculated using these coefficients are called "committed" doses because they are estimated doses that a person will receive over a stated period (50 or 70 years) in the future after a one-time intake of a radionuclide. The quantity of radionuclide taken into the body is typically stated in units of becquerels (Bq) or curies (Ci), where 1 Bq is one disintegration per second, and 1 Ci equals 3.7×10^{10} disintegrations per second. The doses are also termed "effective" doses because they reflect the fact that a radionuclide taken into the body distributes to various organs, and weighting factors have been incorporated that reflect the relative importance of the impacted organs such that the result represents a total-body dose that represents equivalent risk. The values resulting from use of these coefficients are also called "dose equivalents" because they include application of quality factors that convert from estimates of the amount of energy absorbed in tissues (absorbed doses) to values that reflect the varying potential for damage associated with the different radiations (alpha particles, beta particles, gamma rays, etc.). Dose equivalents are stated in terms of sieverts (Sv) or rem, with one sievert equal to 100 rem.

The ICRP Task Group chose to continue to base their dose coefficients on the assumption that americium behaves like plutonium in adult humans. Figures D-1 and D-2 illustrate that there is relatively little variation in dose coefficients among the isotopes of plutonium and Am-241. While source terms used in the dose assessment phase of this project did not include specific release estimates for every radionuclide expected to be contained in Rocky Flats plutonium, the steps that can be taken to estimate historical emissions of the individual radionuclides listed in Table D-1 are described in this appendix.

D.2 Assignment of Plutonium Facility Emissions to Specific Radionuclides

An important step in the assignment of measured nonspecific radioactivity emissions (total long-lived alpha radioactivity) from plutonium facilities to the appropriate individual radionuclides is the characterization of Am-241 content. Am-241 is a decay product of plutonium and as such has been present at Rocky Flats since the early 1950s. It is an undesirable contaminant in weapons-grade plutonium and is present in increasing amounts as plutonium ages.

At Rocky Flats, americium was emitted in particulate form and was therefore collected by the exhaust sampling systems much like plutonium and uranium. The americium content of airborne effluents was not specifically measured for a large portion of the operational history of the Rocky Flats Plant. Am-241 release totals for airborne effluents were reported for calendar years 1985 through 1989 (EG&G, 1991).

Am-241 is formed when Pu-241 decays by emitting a beta particle. Plutonium is formed in a reactor when U-238 absorbs neutrons to form Pu-239, which in turn can absorb additional neutrons to form

the heavier "contaminant" isotopes of plutonium, including Pu-241. The extent to which these contaminant nuclides build up is determined by the length of time the fuel remains in the reactor.

Chemical purification of reactor fuel that took place at the plutonium production sites (Hanford and Savannah River) separated the plutonium from americium present at that point in time, as did the peroxide precipitation step of the Rocky Flats plutonium recovery process and the molten salt extraction process used at Rocky Flats.

Weapons-grade plutonium like that used at Rocky Flats contains about 0.3 mass percent Pu-241 (Rockwell, 1985) and initially contains about 0.0001 percent Am-241 (Krey *et al.*, 1976). The Pu-241, however, decays relatively quickly (with a 13-year half-life) to form Am-241 as time passes after purification.

The Final Environmental Impact Statement for the Rocky Flats Plant states that the americium to plutonium activity ratio has ranged from 10 percent to 20 percent, but also states that this ratio can change during processing that separates americium from plutonium. In a referenced study, when americium was measured in a facility where it was handled in the chemically separated form, the americium to plutonium ratio in effluents did not exceed 0.42 (USDOE, 1980).

Based on reported Rocky Flats airborne release totals for calendar years 1985 and 1987 through 1989 (EG&G, 1991) and independently reconstructed emission totals for 1986, the ratios of Am-241 released to Pu-239/240 released were as shown in Table D-2. It is therefore estimated that airborne Am-241 emissions for each year from 1953 to 1984 were between 13 and 31 percent of the plutonium-239/240 release total for the same year.

TABLE D-2

**ACTIVITY RATIOS OF AM-241 TO PU-239/240
IN MONITORED ROCKY FLATS EFFLUENTS**

Calendar Year	Ratio of Airborne Am-241 Released to Pu-239/240 Released
1985	22%
1986	31%
1987	21%
1988	13%
1989	24%
Average	22%

References: Independently reconstructed emissions for 1986, USDOE Effluent Information System (EG&G, 1991) for other years.

Long-lived alpha activity can be attributed to the various isotopes according to typical activity fractions such as those contained in Table D-1. The following steps can be taken to estimate the releases for the radionuclides listed in Table D-1:

For the period from 1953 through 1973 (based on nonspecific alpha measurements):

Emissions of the following radionuclides are estimated by multiplying the annual total long-lived alpha release total times the following values of Percentage of Total Alpha Activity from Table D-1. Because the americium content of total alpha-emitting radioactivity is specified as a range, all of the values derived from total alpha activity are also ranges.

Pu-238	0.0179 to 0.0207
Pu-239	0.611 to 0.707
Pu-240	0.138 to 0.160
Pu-241	3.90 to 4.51
Pu-242	0.0000124 to 0.0000143
Am-241	0.113 to 0.232

For the period from 1974 through 1989, except Am-241 1985-89 and Pu-238 1986-89 (based on Pu-239/240 measurements):

Emissions of the following radionuclides are estimated by multiplying the annual total long-lived alpha release total times the following values of Percentage of Pu-239/240 Alpha Activity from Table D-1. The americium content of Rocky Flats plutonium has been specified as a range of percentages of Pu-239/240 content. As a result, two multipliers are specified which define ranges of annual Am-241 emissions based on measured Pu-239/240 releases.

Pu-238	0.0239
Pu-239	0.815
Pu-240	0.185
Pu-241	5.21
Pu-242	0.0000165
Am-241	0.13 to 0.31

For the period 1985 through 1989 (based on specific Am-241 data):

Am-241 emissions are taken to be equal to the reported results of alpha spectrometric analyses of americium radiochemically separated from airborne particle samples.

For the period from 1986 through 1989 (based on specific Pu-238 data):

Pu-238 emissions are taken to be equal to the reported results of alpha spectrometric analyses of plutonium radiochemically separated from airborne particle samples.

The results of this process of assignment of activity released from plutonium facilities to specific radionuclides of concern are presented in Table D-3.

D.3 Rocky Flats Uranium

The materials of concern identified in Task 2 of this project included individual isotopes of uranium, that is, U-233, U-234, U-235, and U-238. Uranium has historically been processed and handled at Rocky Flats in two forms: enriched and depleted. Tables D-4 and D-5 show the reported activity fractions of the different radionuclides present in Rocky Flats enriched uranium and depleted uranium (USDOE, 1980).

TABLE D-3: ASSIGNMENT OF PLUTONIUM ACTIVITY TO SPECIFIC RADIONUCLIDES

Year	Plutonium-238 (μCi)	Plutonium-239 (μCi)	Plutonium-240 (μCi)	Plutonium-241 (μCi)	Plutonium-242 (μCi)	Americium-241 (μCi)
1953	2.9e-02 to 3.3e-02*	9.8e-01 to 1.1e+00	2.2e-01 to 2.6e-01	6.2e+00 to 7.2e+00	2.0e-05 to 2.3e-05	1.8e-01 to 3.7e-01
1954	9.5e-01 to 1.1e+00	3.2e+01 to 3.7e+01	7.3e+00 to 8.5e+00	2.1e+02 to 2.4e+02	6.6e-04 to 7.6e-04	6.0e+00 to 1.2e+01
1955	1.1e+00 to 1.2e+00	3.6e+01 to 4.2e+01	8.1e+00 to 9.4e+00	2.3e+02 to 2.7e+02	7.3e-04 to 8.4e-04	6.7e+00 to 1.4e+01
1956	3.4e+00 to 3.9e+00	1.2e+02 to 1.3e+02	2.6e+01 to 3.0e+01	7.4e+02 to 8.6e+02	2.4e-03 to 2.7e-03	2.1e+01 to 4.4e+01
1957	2.1e+02 to 2.5e+02	7.3e+03 to 8.5e+03	1.7e+03 to 1.9e+03	4.7e+04 to 5.4e+04	1.5e-01 to 1.7e-01	1.4e+03 to 2.8e+03
1958	4.5e+01 to 5.2e+01	1.5e+03 to 1.8e+03	3.5e+02 to 4.0e+02	9.8e+03 to 1.1e+04	3.1e-02 to 3.6e-02	2.8e+02 to 5.8e+02
1959	2.0e+01 to 2.3e+01	6.7e+02 to 7.8e+02	1.5e+02 to 1.8e+02	4.3e+03 to 5.0e+03	1.4e-02 to 1.6e-02	1.2e+02 to 2.6e+02
1960	2.0e+01 to 2.3e+01	6.7e+02 to 7.8e+02	1.5e+02 to 1.8e+02	4.3e+03 to 5.0e+03	1.4e-02 to 1.6e-02	1.2e+02 to 2.6e+02
1961	2.1e+01 to 2.5e+01	7.3e+02 to 8.5e+02	1.7e+02 to 1.9e+02	4.7e+03 to 5.4e+03	1.5e-02 to 1.7e-02	1.4e+02 to 2.8e+02
1962	4.5e+01 to 5.2e+01	1.5e+03 to 1.8e+03	3.5e+02 to 4.0e+02	9.8e+03 to 1.1e+04	3.1e-02 to 3.6e-02	2.8e+02 to 5.8e+02
1963	5.4e+01 to 6.2e+01	1.8e+03 to 2.1e+03	4.1e+02 to 4.8e+02	1.2e+04 to 1.4e+04	3.7e-02 to 4.3e-02	3.4e+02 to 7.0e+02
1964	4.1e+01 to 4.8e+01	1.4e+03 to 1.6e+03	3.2e+02 to 3.7e+02	9.0e+03 to 1.0e+04	2.9e-02 to 3.3e-02	2.6e+02 to 5.3e+02
1965	9.5e+01 to 1.1e+02	3.2e+03 to 3.7e+03	7.3e+02 to 8.5e+02	2.1e+04 to 2.4e+04	6.6e-02 to 7.6e-02	6.0e+02 to 1.2e+03
1966	4.7e+00 to 5.4e+00	1.6e+02 to 1.8e+02	3.6e+01 to 4.2e+01	1.0e+03 to 1.2e+03	3.2e-03 to 3.7e-03	2.9e+01 to 6.0e+01
1967	5.9e+00 to 6.8e+00	2.0e+02 to 2.3e+02	4.6e+01 to 5.3e+01	1.3e+03 to 1.5e+03	4.1e-03 to 4.7e-03	3.7e+01 to 7.7e+01
1968	7.2e+00 to 8.3e+00	2.4e+02 to 2.8e+02	5.5e+01 to 6.4e+01	1.6e+03 to 1.8e+03	5.0e-03 to 5.7e-03	4.5e+01 to 9.3e+01
1969	2.0e+01 to 2.3e+01	6.7e+02 to 7.8e+02	1.5e+02 to 1.8e+02	4.3e+03 to 5.0e+03	1.4e-02 to 1.6e-02	1.2e+02 to 2.6e+02
1970	5.5e+00 to 6.4e+00	1.9e+02 to 2.2e+02	4.3e+01 to 5.0e+01	1.2e+03 to 1.4e+03	3.8e-03 to 4.4e-03	3.5e+01 to 7.2e+01
1971	1.1e+00 to 1.3e+00	3.7e+01 to 4.3e+01	8.4e+00 to 9.8e+00	2.4e+02 to 2.8e+02	7.6e-04 to 8.7e-04	6.9e+00 to 1.4e+01
1972	9.0e-01 to 1.0e+00	3.1e+01 to 3.5e+01	6.9e+00 to 8.0e+00	2.0e+02 to 2.3e+02	6.2e-04 to 7.2e-04	5.7e+00 to 1.2e+01
1973	9.1e-01 to 1.1e+00	3.1e+01 to 3.6e+01	7.0e+00 to 8.2e+00	2.0e+02 to 2.3e+02	6.3e-04 to 7.3e-04	5.8e+00 to 1.2e+01
1974	2.3e+01	7.8e+02	1.8e+02	5.0e+03	1.6e-02	1.2e+02 to 3.0e+02
1975	2.4e-01	8.2e+00	1.9e+00	5.2e+01	1.7e-04	1.3e+00 to 3.1e+00
1976	9.6e-02	3.3e+00	7.4e-01	2.1e+01	6.6e-05	5.2e-01 to 1.2e+00
1977	9.6e-02	3.3e+00	7.4e-01	2.1e+01	6.6e-05	5.2e-01 to 1.2e+00
1978	6.7e-02	2.3e+00	5.2e-01	1.5e+01	4.6e-05	3.6e-01 to 8.7e-01
1979	1.3e-01	4.5e+00	1.0e+00	2.9e+01	9.1e-05	7.2e-01 to 1.7e+00
1980	2.9e-01	9.8e+00	2.2e+00	6.3e+01	2.0e-04	1.6e+00 to 3.7e+00
1981	2.0e-01	6.7e+00	1.5e+00	4.3e+01	1.4e-04	1.1e+00 to 2.5e+00
1982	4.8e-01	1.6e+01	3.7e+00	1.0e+02	3.3e-04	2.6e+00 to 6.2e+00
1983	1.9e+00	6.4e+01	1.4e+01	4.1e+02	1.3e-03	1.0e+01 to 2.4e+01
1984	1.9e+00	6.4e+01	1.4e+01	4.1e+02	1.3e-03	1.0e+01 to 2.4e+01
1985	2.2e-01	7.5e+00	1.7e+00	4.8e+01	1.5e-04	2.0e+00
1986	9.2e-01	2.4e+01	5.4e+00	1.5e+02	4.8e-04	4.8e+00
1987	5.6e-01	1.2e+01	2.8e+00	7.8e+01	2.5e-04	3.2e+00
1988	3.9e-01	1.2e+01	2.8e+00	7.8e+01	2.5e-04	2.0e+00
1989	2.0e-01	3.7e+00	8.3e-01	2.3e+01	7.4e-05	1.1e+00

* Note on Scientific Notation: 3.3e-02 equal 3.3×10^{-2} or 0.033.

TABLE D-4

ACTIVITY FRACTIONS OF ROCKY FLATS ENRICHED URANIUM

Nuclide	Percentage by Weight	Percentage of Alpha Activity	Percentage of Beta Activity
Th-231		-	99.1
Th-234		-	0.892
U-234		96.5	-
U-235	over 93	3.11	-
U-236		0.389	-
U-238		0.0280	-

TABLE D-5

ACTIVITY FRACTIONS OF ROCKY FLATS DEPLETED URANIUM

Nuclide	Percentage by Weight	Percentage of Alpha Activity	Percentage of Beta Activity
Th-231		-	1.42
Th-234		-	98.6
U-234		9.69	-
U-235	less than 0.7	1.28	-
U-238		89.0	-

The specific activity (curies per gram) of enriched uranium is approximately 6000 times lower than that of plutonium. Although enriched uranium is more than 93 percent U-235 by weight, U-234 constitutes over 96 percent of its alpha radioactivity. U-238 constitutes almost 90 percent of the alpha radioactivity of depleted uranium. Th-231 and Th-234 are beta-emitting decay products of U-235 and U-238, respectively. These thorium isotopes decay rather rapidly, with half-lives of 26 hours and 24 days, respectively.

On an alpha activity basis, U-234 and U-235 would be expected to make up nearly all of the uranium alpha activity in Rocky Flats airborne effluents from enriched uranium facilities. Likewise, U-238 and U-234 would be expected to comprise most of the uranium in effluents from depleted uranium facilities.

Routine isotopic analyses of effluent sample filters did not start until around 1973. However, reporting of total long-lived alpha radioactivity continued for uranium facilities until approximately 1978. Emissions from uranium facilities were "radiochemically determined as U-233, U-234, and U-238" for the first time in the 1978 Rocky Flats Plant annual environmental report (Rockwell, 1979). By calendar year 1979, there was reporting of both enriched and depleted uranium from essentially all uranium facilities (EG&G, 1991).

The analytical procedure used for chemical separation of uranium from other alpha emitters has historically involved addition of nonindigenous U-232 as an internal standard to gauge the yield of the separation process. Because U-232 emits alpha particles that interfere with the specific identification of U-235, the indistinguishable U-233/234 pair have been used to indicate the magnitude of enriched uranium releases in the same manner as U-238 has been used as an indicator of depleted uranium emissions. Although U-233 was indistinguishable from U-234 in measurement of uranium effluents, it was not a measurable constituent of the depleted or enriched uranium processed at Rocky Flats. All indications from review of historical operations at Rocky Flats are that no significant amounts of U-233 were processed or handled. As a result, no portion of measured emissions from the plant are attributed to U-233 in this analysis.

To translate measurements of total long-lived alpha radioactivity from uranium facilities and more recent U-233/234 and U-238 measurements to emission totals for the specific alpha-emitting radionuclides listed in Tables D-4 and D-5, a number of relationships must be characterized:

- The fractions of total long-lived alpha radioactivity from uranium facilities that were actually U-234, U-235, U-236, and U-238 must be estimated. The fractions might vary between periods of significant enriched uranium (oralloy line) recovery and machining compared to periods of large depleted uranium projects (e.g., M1A1 tank armor manufacturing in Building 883).

- The ratios of indicator isotope quantities (i.e., U-233/234 and U-238) to the quantities of other isotopes in enriched and depleted uranium (e.g., U-235 and U-236) must be characterized.

Each of these relationships can be described to some extent based on available site-specific information and generic information characterizing compositions of special nuclear materials historically encountered within the U.S. nuclear weapons complex. To support the dose assessment portions of this project, source terms were provided for enriched and depleted uranium. As with plutonium, given the relative similarities of the physical and toxicological properties of the uranium isotopes in question, taking the source term analyses to the level of individual radionuclides would result in little benefit in terms of dose assessment and would introduce additional uncertainty into the process. Figures D-3 and D-4 present inhalation and ingestion dose coefficients for the uranium isotopes of interest to this project from DOE/EH-0071 (USDOE, 1988). DOE/EH-0071 factors correspond to doses delivered over a 50-year period after intake by an adult. ICRP 56, which presents age-dependent dose coefficients, does not include uranium isotopes.

Figures D-3 and D-4 illustrate that there is relatively little variation in dose coefficients among the uranium isotopes. While the source terms used in the dose assessment phase of this project did not include specific release estimates for every radionuclide expected to be contained in Rocky Flats uranium, the steps that can be taken to estimate historical emissions of the individual radionuclides listed in Table D-4 and D-5 are described in this appendix.

D.4 Assignment of Uranium Facility Emissions to Specific Radionuclides

Assignment of alpha-emitting radioactivity released from facilities that processed and handled enriched and depleted uranium to specific radionuclides requires a number of approaches for different time periods because of the different forms of emission data that are available. The isotopic composition of the estimated release totals are based on observed measurements and the activity fractions presented in Tables D-4 and D-5.

Depleted Uranium from 1953-1977 (based on nonspecific alpha measurements):

Uranium isotopic releases are calculated by multiplying the measured total long-lived alpha activity released from depleted uranium facilities by the following, based on the fraction of depleted uranium alpha activity:

1.	U-234	0.0969
2.	U-235	0.0128
3.	U-238	0.890

Figure D-3 Inhalation Dose Factors for Uranium

Figure D-4 Ingestion Dose Factors for Uranium

Enriched Uranium from 1953-1977 (based on nonspecific alpha measurements):

Uranium isotopic releases are calculated by multiplying the measured total long-lived alpha activity released from enriched uranium facilities by the following, based on the fraction of enriched uranium alpha activity:

1.	U-234	0.965
2.	U-235	0.0311
3.	U-236	0.00389
4.	U-238	0.000280

Depleted Uranium Emissions for 1978-1980 and for 1985-1989 (based on alpha spectrometric measurements of U-238):

1. U-234 releases are calculated by multiplying the reported U-238 site emission total by 0.109, the ratio of U-234 alpha activity to U-238 alpha activity in Rocky Flats depleted uranium (see Table D-5).

2. U-235 releases are calculated by multiplying the reported U-238 site emission total by 0.014, the ratio of U-235 alpha activity to U-238 alpha activity in Rocky Flats depleted uranium (see Table D-5).

3. U-238 releases are taken to be equal to those calculated based on U-238 measurements.

Enriched Uranium Emissions for 1978-1980 and for 1985-1989 (based on alpha spectrometric measurements of U-233/234):

1. U-234 releases are taken to be equal to the measured U-233/234 emissions.

2. U-235, U-236, and U-238 releases are calculated by multiplying the reported U-233/234 site emission total by the following, based on ratios of their activities to U-234 alpha activity in Rocky Flats enriched uranium:

a.	U-235	0.0322
b.	U-236	0.00403
c.	U-238	0.000290

Uranium Emissions for 1981-1984
(based on alpha spectrometric measurements of U-233, U-234, U-238):

For calendar years 1981 through 1984, Rocky Flats uranium emissions were reported only as total uranium emissions, based on alpha spectral measurements of U-233, U-234, and U-238. Separate depleted (or U-238) and enriched (or U-233/234) results were not reported. To estimate releases of specific uranium isotopes for these years, the following calculations were performed:

1. Reported enriched and depleted uranium emission totals for the period from 1978 to 1980, reconstructed uranium releases from 1984 and 1986, and reported emissions for 1985 and 1987 through 1989 were used to calculate average fractions of Rocky Flats uranium emissions that were in the enriched and depleted forms. The historical fractions of airborne depleted and enriched uranium emissions are depicted in Figure D-17. These calculations yielded 0.6 as the average depleted fraction and 0.4 as the average enriched fraction of total uranium alpha activity.

2. Reported combined uranium emission totals based on U-233/234 and U-238 measurements were multiplied by the enriched and depleted fractions to estimate enriched and depleted uranium release totals for the years in question. Results are as follows:

Year	Reported Total Uranium Release (μCi)	Calculated Enriched Uranium Release (μCi)	Calculated Depleted Uranium Release (μCi)
1981	29.9	12	18
1982	30.9	12	19
1983	51.0	20	31
1984	34.5	14	21

Depleted Uranium Facilities:

3. U-234, U-235, and U-238 release estimates were calculated by multiplying the estimated depleted uranium component of the reported U-233/234 plus U-238 site emission total (from Step 2 above) by the following, based on the ratios of their alpha activities to U-234 plus U-238 alpha activity in Rocky Flats depleted uranium:

1.	U-234	0.0982
2.	U-235	0.0130
3.	U-238	0.902

Enriched Uranium Facilities:

4. U-234, U-235, U-236, and U-238 release estimates were calculated by multiplying the estimated enriched uranium component of the reported U-233/234 plus U-238 site emission total (from Step 2 above) by the following, based on the ratio of their alpha activities to U-234 plus U-238 alpha activity in Rocky Flats enriched uranium:

1.	U-234	0.9997
2.	U-235	0.0322
3.	U-236	0.00403
4.	U-238	0.000290

The results of this process for assignment of activity released from uranium facilities to specific radionuclides of concern are presented in Table D-6.

TABLE D-6: ASSIGNMENT OF URANIUM ACTIVITY TO SPECIFIC RADIONUCLIDES

Year	Depleted Uranium (μCi)			Enriched Uranium (μCi)				Total Uranium (Depleted plus Enriched; μCi)			
	U-234	U-235	U-238	U-234	U-235	U-236	U-238	U-234	U-235	U-236	U-238
1953	1.2e+02 *	1.5e+01	1.1e+03	8.8e+00	2.8e-01	3.5e-02	2.5e-03	1.3e+02	1.6e+01	3.5e-02	1.1e+03
1954	1.2e+02	1.5e+01	1.1e+03	8.8e+00	2.8e-01	3.5e-02	2.5e-03	1.3e+02	1.6e+01	3.5e-02	1.1e+03
1955	2.0e+02	2.7e+01	1.9e+03	7.1e+01	2.3e+00	2.9e-01	2.1e-02	2.7e+02	2.9e+01	2.9e-01	1.9e+03
1956	9.7e+01	1.3e+01	8.9e+02	1.1e+03	3.4e+01	4.3e+00	3.1e-01	1.2e+03	4.7e+01	4.3e+00	8.9e+02
1957	7.1e+01	9.3e+00	6.5e+02	3.5e+02	1.1e+01	1.4e+00	1.0e-01	4.2e+02	2.0e+01	1.4e+00	6.5e+02
1958	1.6e+02	2.0e+01	1.4e+03	3.0e+02	9.6e+00	1.2e+00	8.7e-02	4.5e+02	3.0e+01	1.2e+00	1.4e+03
1959	2.6e+01	3.5e+00	2.4e+02	5.2e+02	1.7e+01	2.1e+00	1.5e-01	5.5e+02	2.0e+01	2.1e+00	2.4e+02
1960	3.4e+01	4.5e+00	3.1e+02	8.3e+02	2.7e+01	3.3e+00	2.4e-01	8.6e+02	3.1e+01	3.3e+00	3.1e+02
1961	5.0e+01	6.7e+00	4.6e+02	4.6e+02	1.5e+01	1.9e+00	1.3e-01	5.1e+02	2.2e+01	1.9e+00	4.6e+02
1962	3.6e+01	4.7e+00	3.3e+02	2.4e+02	7.8e+00	9.7e-01	7.0e-02	2.8e+02	1.3e+01	9.7e-01	3.3e+02
1963	4.7e+01	6.3e+00	4.4e+02	3.2e+02	1.0e+01	1.3e+00	9.2e-02	3.7e+02	1.7e+01	1.3e+00	4.4e+02
1964	2.3e+01	3.1e+00	2.1e+02	1.8e+02	5.9e+00	7.4e-01	5.3e-02	2.1e+02	9.0e+00	7.4e-01	2.1e+02
1965	2.7e+01	3.6e+00	2.5e+02	1.8e+02	5.9e+00	7.4e-01	5.3e-02	2.1e+02	9.5e+00	7.4e-01	2.5e+02
1966	1.4e+01	1.8e+00	1.2e+02	2.2e+02	7.2e+00	8.9e-01	6.4e-02	2.4e+02	8.9e+00	8.9e-01	1.2e+02
1967	1.4e+01	1.8e+00	1.2e+02	1.1e+02	3.4e+00	4.3e-01	3.1e-02	1.2e+02	5.2e+00	4.3e-01	1.2e+02
1968	1.4e+01	1.8e+00	1.2e+02	1.5e+02	5.0e+00	6.2e-01	4.5e-02	1.7e+02	6.8e+00	6.2e-01	1.2e+02
1969	1.6e+01	2.0e+00	1.4e+02	4.8e+01	1.6e+00	1.9e-01	1.4e-02	6.4e+01	3.6e+00	1.9e-01	1.4e+02
1970	1.8e+01	2.4e+00	1.7e+02	6.2e+01	2.0e+00	2.5e-01	1.8e-02	8.0e+01	4.4e+00	2.5e-01	1.7e+02
1971	5.6e+00	7.4e-01	5.2e+01	4.0e+01	1.3e+00	1.6e-01	1.1e-02	4.5e+01	2.0e+00	1.6e-01	5.2e+01
1972	4.1e+00	5.4e-01	3.7e+01	3.9e+00	1.2e-01	1.6e-02	1.1e-03	7.9e+00	6.6e-01	1.6e-02	3.7e+01
1973	5.2e+00	6.9e-01	4.8e+01	1.2e+01	3.7e-01	4.7e-02	3.4e-03	1.7e+01	1.1e+00	4.7e-02	4.8e+01
1974	8.7e-01	1.2e-01	8.0e+00	2.6e+01	8.4e-01	1.1e-01	7.6e-03	2.7e+01	9.5e-01	1.1e-01	8.0e+00
1975	2.7e+00	3.6e-01	2.5e+01	2.7e+01	8.7e-01	1.1e-01	7.8e-03	3.0e+01	1.2e+00	1.1e-01	2.5e+01
1976	1.2e+00	1.5e-01	1.1e+01	1.5e+01	5.0e-01	6.2e-02	4.5e-03	1.7e+01	6.5e-01	6.2e-02	1.1e+01
1977	1.8e+00	2.4e-01	1.7e+01	2.0e+01	6.5e-01	8.2e-02	5.9e-03	2.2e+01	9.0e-01	8.2e-02	1.7e+01
1978	3.6e+00	4.6e-01	3.3e+01	2.1e+01	6.8e-01	8.5e-02	6.1e-03	2.5e+01	1.1e+00	8.5e-02	3.3e+01
1979	2.8e+00	3.6e-01	2.6e+01	9.2e+00	3.0e-01	3.7e-02	2.7e-03	1.2e+01	6.6e-01	3.7e-02	2.6e+01
1980	1.6e+00	2.1e-01	1.5e+01	1.5e+01	4.8e-01	6.0e-02	4.4e-03	1.7e+01	6.9e-01	6.0e-02	1.5e+01
1981	1.8e+00	2.3e-01	1.6e+01	1.2e+01	3.9e-01	4.8e-02	3.5e-03	1.4e+01	6.2e-01	4.8e-02	1.6e+01
1982	1.8e+00	2.4e-01	1.7e+01	1.2e+01	4.0e-01	5.0e-02	3.6e-03	1.4e+01	6.4e-01	5.0e-02	1.7e+01
1983	3.0e+00	4.0e-01	2.8e+01	2.0e+01	6.6e-01	8.2e-02	5.9e-03	2.3e+01	1.1e+00	8.2e-02	2.8e+01
1984	2.0e+00	2.7e-01	1.9e+01	1.4e+01	4.4e-01	5.6e-02	4.0e-03	1.6e+01	7.1e-01	5.6e-02	1.9e+01
1985	4.3e+00	5.5e-01	3.9e+01	7.9e+00	2.5e-01	3.2e-02	2.3e-03	1.2e+01	8.0e-01	3.2e-02	3.9e+01
1986	3.2e-01	4.1e-02	2.9e+00	1.1e+01	3.5e-01	4.4e-02	3.2e-03	1.1e+01	3.9e-01	4.4e-02	2.9e+00
1987	1.3e+00	1.7e-01	1.2e+01	4.6e+00	1.5e-01	1.9e-02	1.3e-03	5.9e+00	3.2e-01	1.9e-02	1.2e+01
1988	1.0e+00	1.3e-01	9.3e+00	2.6e+00	8.4e-02	1.0e-02	7.5e-04	3.6e+00	2.1e-01	1.0e-02	9.3e+00
1989	2.7e-01	3.5e-02	2.5e+00	5.2e+00	1.7e-01	2.1e-02	1.5e-03	5.5e+00	2.0e-01	2.1e-02	2.5e+00

* Note on Scientific Notation: 1.2e+02 equals 1.2 x 10^{-2} or 120.

REFERENCES

Del Pizzo *et al.* (1970). Del Pizzo, R. G., J. B. Owens, and E. A. Putzier, "Rapid Estimation of Am-241 Content in Plutonium," *Health Physics* 18(6), 725-727, 1970.

Dow (1974). Dow Chemical Company. 1973 Annual Environmental Monitoring Report, Rocky Flats Plant. Repository Document AC-260.

EG&G (1991). EG&G Idaho, Inc. Effluent Information System for Rocky Flats Plant, provided to ChemRisk in 1991. Repository Document RE-1196.

Hornbacher (1975-1982). Hornbacher, D. D., "Environmental Control Highlights," Monthly and weekly highlights for 1975 through 1982. Repository Documents OT-1130 through OT-1137.

ICRP (1989). International Commission on Radiological Protection, "Age-Dependent Doses to Members of the Public from Intake of Radionuclides," Annals of the ICRP, Vol. 20, No. 2, ICRP Publication 56, Pergamon Press, New York, 1989.

Krey *et al.* (1976). Krey, P., E. Hardy, H. Volchok, L. Toonkel, R. Knuth, M. Coppes, and T. Tamura, "Plutonium and Americium Contamination in Rocky Flats Soil — 1973," Health and Safety Laboratory, Energy Research and Development Administration, Report HASL-304, March 1976. Repository Document AC-192.

Ray and Hammond (1960). Ray, E. L., and S. E. Hammond, "Use of Pulse-Height Analyzer in Environmental Monitoring," Health Physics Department, Dow Chemical Company, Rocky Flats Plant. Circa 1960. Repository Document RE-1188.

Rockwell (1979). Rockwell International, "Annual Environmental Monitoring Report—January-December 1978," Report RFP-ENV-78, May 15, 1979. Repository Document AC-265.

Rockwell (1985). Rockwell International, "Rocky Flats Risk Assessment Guide," Safety Analysis Engineering, March 1985. Repository Document RF-5.

Rockwell (1989). Rockwell International. "Rocky Flats Plant Site Environmental Report for 1988." Repository Document AC-22.

USDOE (1980). U. S. Department of Energy, Final Environmental Impact Statement, Rocky Flats Plant Site, DOE/EIS-0064, April 1980. Repository Document EN-1.

USDOE (1988). U.S. Department of Energy. "Internal Dose Conversion Factors for Calculation of Dose to the Public." Report DOE/EH-0071, DE88-014297. July.

APPENDIX E

DETAILED DOCUMENT REVIEW OF ORGANIC SOLVENT EMISSIONS

APPENDIX E

DETAILED DOCUMENT REVIEW OF ORGANIC SOLVENT EMISSIONS

The material in this appendix describes the detailed review performed on documentation relating to the emission of organic solvents of concern. The review included the thorough evaluation of the Air Pollution Emission Notice (APEN) documents including various calculation checks and the review of other documentation relevant to estimating emissions from Rocky Flats. Each of the organic solvents of concern is discussed in the following sections.

E.1 Carbon Tetrachloride Emission Evaluation

Information sources relevant to development of source terms for carbon tetrachloride emissions includes four APEN reports and seven other technical reports pertaining to Rocky Flats emissions of the chemical. These information sources are discussed and evaluated.

E.1.1 APEN Resources

Four APEN reports were identified that document carbon tetrachloride use and emissions. These reports correspond to Buildings 707, 776/777, 460, and 881. Associated buildings that APEN reports did not identify as carbon tetrachloride users were not considered further unless some other resource(s) indicated differently. The APEN report for Building 774 was added to the list for evaluation of carbon tetrachloride emissions when non-APEN resources indicated emissions from Building 774. Building 774 handles carbon tetrachloride contained in wastes received from Buildings 707 and 776/777.

APEN reports with identified carbon tetrachloride uses were reviewed in detail for any significant flaws in their emission estimate determinations. In particular, the estimate bases were evaluated and calculations checked. A significant flaw was considered any flaw that may have resulted in an emission estimate error equal to one percent or more of the estimated site total emission for that material. No significant flaws were identified. Several minor calculation discrepancies were identified and recalculated, and revised estimates evaluated. The errors were found to have had insignificant overall effects on emission estimates (0.09 ton/yr), and the errors usually resulted in emission overestimates.

The emission estimate basis for each APEN report was usually a user estimate derived from inventory information or a derived evaporation rate. Mass balance information was usually not available. There was no chemical tracking system to document chemical use and movement. In several cases, estimates were based on standard emission factors published by USEPA in a document known as AP-42 (USEPA, 1985). However, in many cases, the AP-42 emission factors did not apply to Rocky Flats Plant processes.

The Building 774 APEN report stated that no carbon tetrachloride emissions need be accounted for from Building 774, because the APEN estimates for Building 707 and 776/777 assume complete

volatilization of used carbon tetrachloride. Review of the APEN reports for Building 707 and 776/777 confirmed uniform application of this conservative assumption and accounting. Therefore, monitoring data from Building 774 will be included in the process of comparing Building 707 and 776/777 emission estimates with available measurements.

The contribution of each building to the site carbon tetrachloride emission total was calculated, with the results shown in Table E-1.

TABLE E-1: CARBON TETRACHLORIDE EMISSIONS FROM RFP BUILDINGS BASED ON APENS

Building	Carbon Tetrachloride Emission Estimate (tons/yr)	Percent of Site Total Emission Estimate
707*	32.3	80
776/777*	8.1	20
460	0.00266	<1
881	0.000892	<1
Site Total	**40.4**	**100%**

* Building 774 emissions are included in emissions for these buildings.

All building emission estimates totaling less than one percent of the site total were not subject to a detailed review of the supporting APEN information. Detailed review of the supporting APEN information for minor chemical users is not expected to yield any significant difference in the final site-wide emission estimate even if the estimates are off by a factor of ten.

APEN reports for buildings contributing more than one percent to site-wide emissions were thoroughly evaluated. Emission estimate evaluations included but were not necessarily limited to:

- Checking calculations,
- Evaluating estimating methods,
- Checking assumptions,
- Looking for confirmatory information,
- Looking for overlooked emissions, and
- Addressing uncertainties or sources of error.

All calculations and mathematical extrapolations were checked. This usually just required converting the user quantity (gallons/year) to an emission estimate in tons/year. This calculation was generally found to be correct.

Each APEN accounted for building vents in detail to demonstrate inclusion of all emissions. However, none of the APENs for carbon tetrachloride users based the estimates on emission monitoring. The following is a summary of the major identified carbon tetrachloride users based on the APENs.

Building 707 Processes

Building 707 contained foundry and casting operations, and products assembly. Carbon tetrachloride was used as a cleaning agent. No emission controls for carbon tetrachloride were present in Building 707.

Module A - Casting Operations

Carbon tetrachloride was used to clean interior glove-box walls and furnaces in plutonium ingot casting furnace areas. A user estimate of 252 gallons per year was determined based upon inventory records and known routine cleaning practices. All of the carbon tetrachloride was assumed to evaporate. The calculated emission estimate is 1.68 tons per year.

Module J - Casting Operations

Plutonium ingots were cast in this area. Carbon tetrachloride was used to clean the glove boxes. The same basis and calculation as used for Module A were used here, yielding an emission estimate of 1.68 tons per year.

Module K - Casting Operations and Stacker Retriever

This operation stored and retrieved plutonium for distribution to other processes. Plutonium was weighed, melted in a furnace, and formed into ingots. Carbon tetrachloride was used to clean the glove boxes. The same basis and calculation as used for Module A were used here, yielding an emission estimate of 1.68 tons per year.

Module B - Rolling and Forming

This process involved the forming and thermal treatment of plutonium ingots. Carbon tetrachloride was used to clean the rollers. A user estimate of 3 gallons per day performed 220 days per year was the basis of an emission estimate of 4.4 tons per year.

Module C - Briquetting

Metal trimmings from Module C machining and Module B scrap cutters were placed into metal baskets and dipped into a series of five carbon tetrachloride open surface baths. Each bath was a steel tank containing approximately 4 gallons of carbon tetrachloride. Emissions were calculated based on the cold cleaner factors from AP-42, Section 4.6-1, "Solvent Degreasing" (USEPA, 1985). A common reduction factor was applied based on good operating procedures (such as keeping the lids closed when not in use), and the lowest allowable reduction of 28 percent was conservatively applied. Assumptions were found to be reasonable. The calculated emission estimate was found to be in error by a factor of 10 due to a math error, resulting in an overestimation of the emission estimate as 0.10 ton per year (rather than 0.01 ton/yr).

Module C - Machining Operations

Plutonium parts were machined, weighed, and then cleaned with carbon tetrachloride. A user estimate of 3,400 gallons per year was the basis of an emission estimate of 22.61 ton per year.

Modules C and D - Inspection

Carbon tetrachloride was periodically used to clean parts prior to inspection. User estimates based on inventories of 60 liters for Module C and 24 liters for Module D were used to calculate an emission estimate of 0.147 ton per year.

The Building 707 APEN reported carbon tetrachloride feed and waste transportation lines within the building, but no emissions were specifically identified from these lines. Consideration was given as to whether losses from these lines should have been accounted for. Separate transportation line emission accounting was not found to be necessary or appropriate because the APEN emission

estimates were based on the assumptions that all used carbon tetrachloride evaporates and is accounted for as an emission.

All waste carbon tetrachloride was pumped to the "C-pit," a partial basement under Module C. The pit consisted of two storage tanks and 16 "pencil" tanks for the storage of carbon tetrachloride. The waste carbon tetrachloride was pumped to Building 774 after being sampled. All potential emissions from the C-pit area were considered accounted for by assuming all of the carbon tetrachloride evaporated from each module source. This assumption was found to be reasonable with the exception of the Module C briquetting operation in which it is not clear if the waste materials had been assumed to evaporate.

The total Building 707 carbon tetrachloride emission estimate was 32.3 tons per year.

Building 776/777 Processes

Building 776 was originally a manufacturing building until operations were transferred to Building 707 in 1972. Building 776 served as a waste storage and waste reduction building after that time. Building 777 was an assembly building. No emission controls for carbon tetrachloride were present in either building. Building 776 and 777 share a common wall and ventilation system.

Building 776 - Baler

The baler was used to reduce the volume of low-level combustible waste. Carbon tetrachloride was a solvent found present in a maximum concentration of 750 pounds carbon tetrachloride per million pounds of waste. Assuming all of the carbon tetrachloride evaporated, the maximum concentration was used to determine an emission estimate of 2.32 tons per year (assuming 6,193,290 pounds of waste per year).

Building 777 - Briquetting

Machine turnings were placed in metal baskets and dipped into a series of four open surface carbon tetrachloride baths prior to placing in a hydraulic press for puck production. Emissions were calculated based on the cold cleaning factors from AP-42, Section 4.6-1, "Solvent Degreasing" (USEPA, 1985). A common reduction factor was applied based on good operating procedures (such as keeping the lids closed when not in use), and the lowest allowable reduction of 28 percent was conservatively applied. Assumptions were found to be reasonable and an emissions estimate of 7.9 x 10^{-2} ton per year was calculated.

Building 777 - Machining

Carbon tetrachloride was used as a cleaning agent for parts prior to machining. A user estimate of 850 gallons per year was used to quantify emissions assuming all used carbon tetrachloride evaporated. The resulting calculated emission estimate is 5.65 tons per year.

Building 777 - Inspection

Parts were cleaned with carbon tetrachloride prior to inspection. A user estimated of 31.5 liters per year was used to estimate emissions assuming all used carbon tetrachloride evaporated. The resulting calculated emission estimate is 5.5×10^{-2} ton per year.

Building 776/777 waste carbon tetrachloride was collected in a series of five "pencil" tanks. The waste carbon tetrachloride was pumped to Building 774 after being sampled. Potential emissions from the waste and fuel tanks as well as all associated transportation lines were considered to be addressed by the assumption that all used carbon tetrachloride evaporated. However, it is not clear if the waste materials from the briquetting operation were assumed to evaporate.

The total Building 776/777 carbon tetrachloride emission estimate was 8.1 tons per year.

E.1.2 Information Sources Other Than APEN Documents

The following resources were identified, evaluated, and found to contain carbon tetrachloride emission information to support a chemical emission source term.

Building 707

A Rockwell International report (Rockwell, 1988) estimated Building 707 carbon tetrachloride use of 10,000 gallons per year based upon an eight-week solvent use study. Assuming all of the carbon tetrachloride evaporated, this represents potential Building 707 carbon tetrachloride emissions of 66 tons per year.

The volatile organic emission report (Hamilton and Moser, 1990) prepared by Martin Marietta was found to contain detailed emission estimate information based on an air monitoring program. The methodology was studied to evaluate the reliability of reported results. General confidence in the reported methodology and associated results is high for reasons including:

- there were six sampling points,

- simulated normal operations were conducted during sampling, and worst-case scenarios were often assumed for calculation purposes,
- carbon tetrachloride was often the target chemical,
- three different sampling collection media were used (Tedlar bags, Tenax tubes, and Tenax/charcoal tubes),
- the highest concentration obtained with either bag or solid sorbent collection method was used to calculate the emission estimate, and
- air flow rates were determined based upon a multi-point transverse monitoring method rather than a single-point method.

Some uncertainty in sample result accuracy is present due to reported little net sampling time and small volumes. The overall sampling times were reasonable, but the actual net times were short and volumes were very small (8-20 minutes, 2-5 liters). An environmental team audit conducted in June 1989 (USDOE, 1989) also questioned the accuracy of Rocky Flats Plant air monitoring results.

The Hamilton and Moser (1990) emission estimates for 1989 were reported in pounds of carbon tetrachloride per hour. Extrapolation to estimated annual emissions was accomplished by assuming:

- 480 hours of operation per year for bimonthly cleaning (six 5-day inventory periods, two shifts per day, eight hours per shift).
- 3,520 hours of operation per year for normal operations (44 weeks per year with six one-week inventory periods and a two-week Christmas shutdown, two shifts per day, eight hours per shift).

These assumptions were derived from, and are consistent with, APEN assumptions. The total estimate extrapolated from the Hamilton and Moser (1990) report is 53 tons per year.

An internal Rocky Flats Plant report (Fruehauf and Richter, 1974) provided an estimated Building 707 carbon tetrachloride usage of approximately 1,000 gallons per month based on warehouse dispensing records. Assuming all of the dispensed carbon tetrachloride evaporated, a Building 707 carbon tetrachloride emission estimate of 80 tons per year is calculated.

An internal Rocky Flats Plant report (Hobbs, 1982) provided Building 707 carbon tetrachloride emission estimate information based on both monitoring and material balance bases. Monitoring data from 1974 and 1975 indicated an average and maximum Building 707 carbon tetrachloride emission

estimate of 46 tons per year and 120 tons per year, respectively, with wide concentration fluctuations. An attached material balance summary indicated a Building 707 carbon tetrachloride emission estimate of 20 tons per year. This report also provided some quantifiable associated uncertainty. Examples include:

- ± 10 percent error in the Building 707 maximum discharge rate due to observed rapid changes in carbon tetrachloride concentrations.

- Up to 12 percent error in the Building 707 monitoring data due to questionable air flow rates during monitoring.

A Rockwell International report (Rockwell, 1989) reported updated halogenated solvent usage in Building 707 for the period of July 1988 - July 1989. The monthly usage varied significantly due to inventory and production activities. Assuming all of the used carbon tetrachloride evaporated, an average and maximum Building 707 carbon tetrachloride emission estimate is 48 tons per year and 76 tons per year, respectively. This report also confirmed that the Rocky Flats Plant carbon tetrachloride uses as of 1988 were "exclusively in Buildings 707 and 776/777."

A Rockwell International report (Ferrera, 1988) provided weekly and monthly carbon tetrachloride usage rates for Building 707 during the period of June - November 1988. This report stated that the Rocky Flats Plant use of carbon tetrachloride was "almost exclusively in Building 707" and that use of halogenated solvents was close to the Rocky Flats Plant solvent use reduction goals, in part, because production rates had been down. Assuming all the used carbon tetrachloride evaporated, usage results indicated an average and maximum Building 707 carbon tetrachloride emission estimate of 50 tons per year and 80 tons per year, respectively. This report also referred to a baseline monthly carbon tetrachloride usage rate of 1,167 gallons based on 1987-1988 purchase records. Assuming all the purchased carbon tetrachloride evaporated, this baseline usage indicated a Building 707 carbon tetrachloride emission estimate of 93 tons per year.

Table E-2 summarizes the Building 707 carbon tetrachloride emission estimates including APEN and non-APEN resources.

insert TABLE E-2

Building 776/777

A Dow Chemical report (Dow Chemical, 1974) provided summary air monitoring data for Building 776/777 carbon tetrachloride emissions over a period of 46 days in 1973. Samples were collected every 15 minutes and carbon tetrachloride concentrations at exhaust points were found in the range of 0 to 270 pounds per day (24 hours). The detailed data table was missing, but average and maximum Building 776/777 carbon tetrachloride estimated emissions were calculated to be 10 and 34 tons per year, respectively.

The volatile organic emission report (Hamilton and Moser, 1990) provided detailed emission estimates information for Building 776/777 in 1989. The same assumptions were applied to the Hamilton and Moser emissions as were applied to Building 707 (described previously) to determine annual emission estimates. The total Building 776/777 carbon tetrachloride emission estimate extrapolated from the Hamilton and Moser (1990) report is 33 tons per year.

An environmental team audit conducted in June 1989 (USDOE, 1989) provided a Building 776/777 carbon tetrachloride eleven-month usage rate of 6,125 gallons. Assuming all used carbon tetrachloride evaporated, an emission estimate for Building 776/777 carbon tetrachloride is 44 tons per year.

An internal Rocky Flats Plant report (Hobbs, 1982) provided a material balance based carbon tetrachloride emission estimate for Building 776/777 of 15 tons per year. The report also provided a summary of air monitoring data for Building 776/777, but the data was found to be the same as that discussed above in the Dow Chemical 1974 report. Table E-3 summarizes the Building 776/777 carbon tetrachloride emission estimates including APEN and non-APEN resources.

Rocky Flats Plant - Total

The Final Environmental Impact Statement for Rocky Flats (USDOE, 1980) reported an estimated Rocky Flats Plant total carbon tetrachloride usage in 1977 of 5,334 gallons. Assuming all the carbon tetrachloride evaporated, a Rocky Flats Plant emission estimate is 36 tons per year. The same report listed a Rocky Flats Plant carbon tetrachloride effluent discharge rate of 4.73 grams per second for 1975. Applying the same operating assumptions as used in the APENs and as applied to the Hamilton and Moser (1990) report, an annual emission estimate of 66 tons per year was calculated.

insert Table E-3

An internal Rocky Flats Plant report (Fruehauf and Richter, 1974) provided an emission estimate for the Rocky Flats Plant for 1973 of 81 tons per year. This is based on the assumption that all used carbon tetrachloride evaporated and that the report authors considered all 700 area carbon tetrachloride users.

The volatile organics emission report for 1989 (Hamilton and Moser, 1990) provided detailed carbon tetrachloride emission estimates for Buildings 460, 707, 774, and 776/777. As discussed in the previous sections for Buildings 707 and 776/777, the Hamilton and Moser (1990) report provided emission monitoring results in pounds of carbon tetrachloride per hour that may be extrapolated to determine annual emission estimates. Applying the same assumptions, a total Rocky Flats Plant carbon tetrachloride emission estimate is 89 tons per year.

The environmental team audit conducted in June 1989 (USDOE, 1989) reported that the major use of carbon tetrachloride occurred in the 700 complex where it was estimated that 99 percent of the carbon tetrachloride was used. This supports the APEN evaluation results that indicated almost all (>99 percent) of the carbon tetrachloride use and emissions involved the 700 complex. The audit report contains good descriptions of identified carbon tetrachloride applications throughout the Rocky Flats Plant.

Agreement with APEN information was generally good with no major exceptions. The audit report did identify carbon tetrachloride users other than those identified in the APENs. These reported users were found to represent insignificant-quantity users.

The environmental team audit report also provided an estimated Rocky Flats Plant carbon tetrachloride usage of 186,816 pounds for 1988. No detail is available to defend the basis of this estimated usage. Assuming all of the used carbon tetrachloride evaporated, a Rocky Flats Plant carbon tetrachloride emission estimate is 93 tons per year.

An EG&G Rocky Flats air stack release tabulation (EG&G, circa 1990) indicated 1988 and 1989 air stack emissions from the Rocky Flats Plant of 130,000 and 48,212 pounds, respectively. Converting to tons per year, this corresponds to 65 tons per year (1988) and 24 tons per year (1989).

An internal Rocky Flats Plant report (Hobbs, 1982) provided carbon tetrachloride emission estimates, based on both monitoring and material balance approaches, that could be summed to estimate Rocky Flats Plant total carbon tetrachloride emissions. The monitoring summary yielded an average Rocky Flats Plant carbon tetrachloride emission estimate of 56 tons per year and a maximum Rocky Flats Plant carbon tetrachloride emission estimate of 153 tons per year. The material balance summary provided two different Rocky Flats Plant carbon tetrachloride emission estimates of 34 tons per year and 106 tons per year. No explanation for this difference was available.

An internal Rocky Flats Plant report (Rockwell, 1989) provided updated solvent use status from July 1988 to July 1989. Assuming all the carbon tetrachloride used evaporated, an average Rocky Flats Plant carbon tetrachloride emission estimate was 48 tons per year.

Table E-4 summarizes these Rocky Flats Plant total carbon tetrachloride emission estimates, including APEN and non-APEN resources.

E.2.2 Chloroform Emission Evaluation

Information sources relevant to chloroform emissions from Rocky Flats and the evaluation process used to develop source terms are described in this section. Information sources relevant to chloroform emissions at Rocky Flats include four APEN reports and six other resources, ranging from personnel interviews to hazardous material inventories and technical reports. The key information sources are described and evaluated in this section.

E.2.1 APEN Resources

Four APENs were initially identified documenting chloroform use and emissions. These APENs correspond to Buildings 559/561, 881, 374, and 460. Selected APENs with identified chloroform users were reviewed in detail for any significant flaws in the emission estimate determinations. No significant flaws were identified. The emission estimate basis for each APEN was a user estimate derived from inventory information. Mass balance information was not available, there was no chemical tracking system to document chemical use and movement, and the USEPA AP-42 emission factors did not apply to the Rocky Flats Plant chloroform uses.

Each building's percent emission relative to the total was calculated with the results shown in Table E-5.

The building emission estimates for Buildings 374 and 460 totaled less than one percent of the total Rocky Flats Plant emission estimate, and a detailed review of the supporting APEN information was not conducted.

The building emission estimates for Buildings 559/561 and 881 were thoroughly evaluated. Each APEN accounted for building vents in detail to demonstrate complete emission inclusion. However, none of the APENs for chloroform users based the estimates on emission monitoring. The following is a summary of the major identified chloroform users based on the APENs.

insert Table E-4

TABLE E-5: CHLOROFORM EMISSIONS FROM ROCKY FLATS BUILDINGS

Building	Chloroform Emission Estimate (ton/yr)	Percent of Site Total Emission Estimate
559/561	0.74	88
881	0.0968	11
374	0.0014	<1
460	0.00625	<1
Site Total	**0.84**	**100%**

Building 559/561 Processes

Building 559/561 contained laboratory facilities for conducting spectrochemical, chemical, and mass spectrometric analyses. No emission controls for chloroform were present in Building 559/561.

Gallium Determination

Chloroform was used to extract gallium oxide from plutonium metal samples. A user estimate of 60 milliliters of chloroform per sample was determined based on inventory records and process knowledge. All of the chloroform was assumed to evaporate even though some chloroform waste was known to have been disposed of and collected in Building 374. The calculated emission estimate is 0.74 ton per year.

Building 881 Processes

Building 881 contained laboratory, maintenance, and plant support facilities. No emission controls for chloroform were present in Building 881.

No explanation of Building 881 chloroform use was provided in the APEN. The source was identified in interviews to be joining of plastics by carpenters (see below). A user estimate of 15.5 gallons per year was the basis of an emission estimate of 9.68×10^{-2} ton per year.

E.2.2 Information Sources Other Than APEN Documents

A number of resources were identified, evaluated, and found to contain chloroform use or emission information to support a chemical emission source term.

Two Rocky Flats Plant personnel interviews indicated that chloroform had been used by carpenters (Building 881) to join plastics but is no longer used in this way (ChemRisk, 1991-1992).

A 1988-1989 inventory (EG&G, 1990d) identified two Building 881 chloroform operations for dissolving plastics and photoresists. This reference and the one above appear to identify the Building 881 use application, which was not provided in the APEN. The inventory also indicated a total Rocky Flats Plant chloroform inventory of 0.55 ton.

The Waste Stream and Residue Identification and Characterization report for Building 881 (Wastren Inc., 1991c) noted the use of chloroform to clean machines and metal parts in Process 881/18, "Special Assembly, Microshaping." There was no laboratory usage identified (possibly due to a discontinued operation).

A 1974 inventory (Barrick, 1974) indicated a total Rocky Flats Plant chloroform inventory of 9 tons. The significant inventory difference compared to the 1988-1989 inventory (EG&G, 1990d) is not currently explainable. The inventory change may be an indication of significant decreased use of chloroform from 1974 to 1988-1989.

An interim Rocky Flats Plant industrial hygiene department printout (Rocky Flats Plant, 1990) identified Buildings 559/561 and 881 as the only chloroform control areas, confirming the APEN evaluation of these two buildings as the major chloroform users.

The Waste Stream and Residue Identification and Characterization report for Building 559/561 (Wastren Inc., 1991d) confirmed the use of chloroform for the extraction of gallium oxide.

No comparable material balance or monitoring basis resources were identified.

E.3 Methylene Chloride Emission Evaluation

Information sources relevant to methylene chloride emissions from Rocky Flats and the evaluation process being used to develop associated source terms are described in this section. Information sources relevant to methylene chloride emissions at Rocky Flats include seven APEN reports and five other resources, ranging from personnel interviews to hazardous material inventories and technical reports. The key information sources are described and evaluated in this section.

E.3.1 APEN Resources

Seven APENs were initially identified documenting methylene chloride use and emissions. These APENs correspond to Buildings 776/777, 771, 881, 551, 460, 374, and 228 A/B. Selected APENs with identified methylene chloride uses were reviewed in detail for any significant flaws in the emission estimate determinations. No significant flaws were identified. The emission estimate basis for most of the APENs was a user estimate derived from inventory information or engineering data. One of the APENs (Building 228 A/B) used sludge monitoring data as the emission estimate basis. Mass balance information was not available, there was no chemical tracking system to document chemical use and movement, and the USEPA AP-42 emission factors did not apply to the Rocky Flats Plant methylene chloride uses.

Each building's percent emission relative to the total was calculated, with the results as indicated in Table E-6.

The emission estimates for Buildings 374, 460, and 551 totaled less than one percent of the total Rocky Flats Plant emission estimate, and a detailed review of the supporting APEN information was not conducted.

The building emission estimate for Buildings 776/777, 771, and 881 were thoroughly evaluated. Each APEN accounted for building vents in detail to demonstrate complete emission inclusion. However, none of the APENs for methylene chloride were based on emission monitoring.

The following is a summary of the major identified methylene chloride users based on the APENs.

Building 776/777 Processes

Building 776 was originally a manufacturing building until operations were transferred to Building 707 in 1972. Building 776 served as a waste storage and waste reduction building after that time. Building 777 was an assembly building. Buildings 776 and 777 share a common wall and ventilation system. No emission controls for methylene chloride were present.

TABLE E-6: METHYLENE CHLORIDE EMISSIONS FROM ROCKY FLATS PLANT BUILDINGS BASED ON APENS

Building	Methylene Chloride Emission Estimate (tons/yr)	Percent of Site Total Emission Estimate
776/777	2.32	70
771	0.70	21
881	0.28	8
374	0.025	<1
460	0.0014	<1
551	0.00053	<1
228 A/B	0.00012	<1
Site Total	**3.33**	**100%**

Baler

A baler was used to reduce the volume of low-level combustible waste. Methylene chloride was present in the wet, low-level waste. Methylene chloride was present in a maximum concentration of 750 pounds methylene chloride per million pounds of waste. Assuming all the methylene chloride evaporated, the maximum concentration was used to determine an emission estimate of 2.32 tons per year (assuming 6,193,290 pounds of waste per year).

Building 771 Processes

The principal operation in Building 771 was the recovery of plutonium from plutonium-bearing residues.

Maintenance

Maintenance personnel used chemical paint strippers consisting of approximately 85 percent methylene chloride (KS-3 paint remover). A user estimate of 165 gallons of paint stripper per year was used to determine an emission estimate of 0.70 ton per year. All of the methylene chloride was assumed to evaporate.

Building 881 Processes

Building 881 contained laboratories, maintenance shops, and plant support facilities.

Semivolatile Organics Analysis Laboratory

Methylene chloride was used to extract semivolatile organics from wastes using a Soxhlet extraction apparatus, or equivalent. Oil samples were first dissolved in methylene chloride, then prepared for gas chromatography/mass spectrometry (GC/MS) analysis. Laboratory personnel estimated that approximately 50 percent of the methylene chloride evaporated into the laboratory hood exhaust. The remaining methylene chloride was collected as waste in the satellite collection area. A user estimate of 100 gallons per year was used to determine an emission estimate of 0.28 ton per year.

E.3.2 Information Sources Other Than APEN Documents

The following resources were identified, evaluated, and found to contain methylene chloride use or emission information to support a chemical emission source term.

A 1974 inventory (Barrick, 1974) indicated a total Rocky Flats Plant methylene chloride inventory of 396.8 gallons (1,502 liters).

The Final Environmental Impact Statement for Rocky Flats (USDOE, 1980) did not list methylene chloride as one of the major chemicals used in 1977. This report did identify methylene chloride as having been used in "various" buildings for paint stripping, confirming the APEN reports of such use.

A 1989 inventory printout of Rocky Flats Plant methylene chloride use (Grocki, 1989a) indicated a total inventory of approximately 73.8 gallons.

A memorandum regarding hazardous wastes potentially generated at the Rocky Flats Plant (Roy F. Weston, 1985) reported an estimated 60.8 gallons of methylene chloride as waste in 1977.

The Waste Stream and Residue Identification and Characterization report for Building 881 (Wastren, 1991c) cited the generation of methylene chloride waste from the Building 881 laboratory extraction process, confirming the APEN reported use. One hundred gallons of waste methylene chloride per year were reported. This may indicate that more than 50 percent of the APEN reported methylene

chloride used (100 gallons per year) was recovered as waste, or it may indicate that the APEN estimated usage is low.

E.4 Tetrachloroethylene Emission Evaluation

Tetrachloroethylene is commonly known as percloroethylene (PCE). Information sources relevant to PCE emissions from Rocky Flats and the evaluation process used to develop an associated source term are described in this section.

Information sources relevant to PCE emissions at Rocky Flats include one APEN report and seven other comparable resources, ranging from personal interviews to hazardous material inventories and technical reports. The key information sources are described and evaluated in this section.

E.4.1 APEN Resources

One APEN was initially identified documenting PCE use and emissions. This APEN is for Building 374 and was reviewed in detail for any significant flaws in the emission estimate determinations.

No significant flaws were identified. The emission estimate basis was a user estimate derived from waste stream tests and waste generation records. Mass balance information was not available, there was no chemical use and movement tracking, and the USEPA AP-42 emission factors did not apply to the APEN-referenced emissions.

The Building 374 APEN was thoroughly evaluated. The APEN accounted for building vents in detail to demonstrate complete emission inclusion. However, the APEN did not base the emission estimate on monitoring. The following is a summary of the identified PCE use based on the APEN.

Building 374 Processes

Building 374 was activated in 1978 as the process waste treatment facility for many of the production buildings.

Process Waste Treatment

PCE was detected in a 1986 waste stream test for Building 889 waste. A 1987 user estimate (waste generation rate) was the basis of an emission estimate of 7.14×10^{-5} ton per year. All PCE was assumed to evaporate.

The Building 374 APEN did not identify how the PCE was used in Building 889. There is also no Building 889 APEN available to further explain the Building 889 PCE use.

E.4.2 Information Sources Other Than APEN Documents

The following resources were identified, evaluated, and found to contain PCE use or emission information to support development of a chemical emission source term.

A 1974 inventory (Barrick, 1974) indicated a total Rocky Flats Plant PCE inventory of 1,179 gallons (4,462.75 liters).

The Final Environmental Impact Statement for Rocky Flats (USDOE, 1980) did not list PCE as one of the major chemicals used in 1977.

The environmental team audit conducted in 1989 (USDOE, 1989) did not list PCE as a major or minor solvent in use.

Three ChemRisk investigation interviews (ChemRisk, 1991-1992, Interview Nos. 39, 48, 53) indicated significantly different historical PCE uses. PCE was reported to have been used in Building 881 at the rate of approximately 25 drums per month. Distillation was reported to have recovered approximately 10 percent of the used PCE in Building 881 during this period resulting in an estimated release of 100 tons per year. PCE was reported to have been used in Building 881 to clean scraps prior to briquetting and "lots of PCE" was reported to have been used in Buildings 444, 883, 771, 776, and 374. PCE was also reported to have been used in Building 886 during the approximate period of 1965-1975.

Industrial hygiene memorandums (Dow Chemical, 1965-1974) described air sampling efforts to evaluate potential occupational exposures in Buildings 776 (May 1966) and 444 (February 1974). No PCE use description or quantity was provided for the Building 776 application. The Building 444 application used PCE as a machine coolant.

Several resources described a four-month trial effort in 1966 to substitute PCE for carbon tetrachloride in "cold washing" applications. The PCE substitution was discontinued due to residue buildup on plutonium surfaces and inspection devices from inhibitors associated with the PCE.

A report describing potentially generated Rocky Flats Plant wastes (Roy F. Weston, 1985) reported PCE waste generation of 73.4 gallons (378 liters) in 1982, 109.6 gallons (415 liters) in 1983, and 121.5 gallons (460 liters) in 1984.

Table E-7 summarizes the Rocky Flats Plant PCE emission estimates including APEN and non-APEN resources.

TABLE E-7
ROCKY FLATS PLANT — TOTAL TETRACHLOROETHYLENE EMISSION ESTIMATES

Resource	Estimate Basis	Estimate Year	Tetrachloroethylene Emission Estimate (tons/yr)
Air Pollution Emission Notices, EG&G (1991d)	Use	1986-87	7.14×10^{-5}
ChemRisk (1991-1992; Interview No. 39)	Use	1958-62	100

E.5 1,1,1-Trichloroethane Emission Evaluation

Information sources relevant to 1,1,1-trichloroethane (TCA) emissions from Rocky Flats and the evaluation process used to develop associated source terms are described in this section.

Information sources relevant to TCA emissions at Rocky Flats include six APEN reports and twelve other resources, ranging from personnel interviews to hazardous material inventories and technical reports by Rocky Flats Plant personnel and outside groups. The key information sources are described and evaluated in this section.

E.5.1 APEN Resources

Six APENs were initially identified documenting TCA use and emissions. These APENs correspond to Buildings 776/777, 707, 774, 460, 374, and 881. In addition, trichloroethylene (TCE) use that was erroneously reported for Building 460 was determined to be TCA. The total TCA usage estimate for Building 460 reflects this correction. Associated buildings that APENs did not identify as TCA users were not considered further as TCA users unless some other resource indicated differently: The APEN for Building 444 was added to the list of APENs to be evaluated for TCA emissions when several non-APEN resources and monitored emissions from Building 444 indicated TCA use.

Selected APENs with identified TCA users were reviewed in detail for any significant flaws in the emission estimate determinations. No significant flaws were identified. The emission estimate basis for most of the APENs was a user estimate derived from inventory information or engineering data. Several Building 707 and 776/777 emission estimates were based on analogy to similar processes described in the USEPA AP-42 emission factors reference (USEPA, 1985). Mass balance information was not available, and there was no chemical tracking system to document chemical use and movement.

Each building's percent emission relative to the total was calculated, with the results as indicated in Table E-8.

TABLE E-8: 1,1,1-TRICHLOROETHANE EMISSIONS FROM ROCKY FLATS BUILDINGS BASED ON APEN REPORTS

Building	Estimate Basis	1,1,1-Trichloroethane Emission Estimate (tons/yr)	Percent of Site Total Emission Estimates
776/777	Use/Analogy	8.36	41
774	Use	8.3	40
707	Use/Analogy	3.8	18
881	Use	0.061	<1
460	Use	0.15*	<1
374	Use	0.0022	<1
Site Total		**20.7**	

* Corrected total.

The building emission estimates for Buildings 881, 460, and 374 totaled less than one percent of the total Rocky Flats Plant emission estimate, and a detailed review of the supporting APEN information was not conducted.

The emission estimates for Buildings 776/777, 774, and 707 were thoroughly evaluated. Each APEN accounted for building vents in detail to demonstrate complete emission inclusion. However, none of the APENs for TCA were based on emission monitoring. The following is a summary of the major identified TCA users based on the APENs.

Building 776/777 Processes

Building 776 was originally a manufacturing building until operations were transferred to Building 707 in 1972. Building 776 served as a waste storage and waste reduction building after that time. Building 777 was an assembly building. No emissions controls for TCA were present in either building. Building 776 and 777 share a common wall and ventilation system. All waste TCA was pumped to the waste TCA collection system.

Building 776 - Baler

The baler was used to reduce the volume of low-level combustible waste. TCA was a solvent present at a maximum concentration of 2,000 pounds of TCA per million pounds of waste. Assuming all of the TCA evaporated, the maximum concentration was used to determine an emission estimate of 6.19 tons per year (assuming 6,193,290 pounds of waste per year).

Building 777 - Foundry Coatings

Substrates to be coated with uranium or plutonium were cleaned with a heated combination vapor and ultrasonic degreaser containing TCA to remove oils. The TCA was changed out when dirty and at bimonthly inventories. Emissions were calculated based on the heated cleaner factors from AP-42, Section 4.6-1, "Solvent Degreasing" (USEPA, 1985). A common reduction factor was applied based on good operating practices (such as keeping the lids closed when not in use), and the lowest allowable reduction of 30 percent was conservatively applied. Assumptions were found to be reasonable. The calculated emission estimate was 0.20 ton per year.

Building 777 - Disassembly (Room 430)

Disassembled plutonium parts were cleaned with small quantities of TCA. A user estimate of one gallon per year was the basis of an emission estimate of 5.6×10^{-3} ton per year.

Building 777 - Assembly Superdry (Room 430)

Parts from inspection were cleaned in a 10-gallon TCA bath to remove oil, grime, and dirt prior to ultrasonic cleaning. The TCA was changed out when dirty or contaminated with water and during bimonthly inventories. Emissions were calculated based on the cold cleaner factors from AP-42, Section 4.6-1, "Solvent Degreasing" (USEPA, 1985). A common reduction factor was applied based on good operating practices, and the lowest allowable reduction of 28 percent was conservatively applied. Assumptions were found to be reasonable. The calculated emission estimate was 0.18 ton per year.

Building 777 - Ultrasonic Cleaner (Room 430)

A heated ultrasonic vapor degreaser containing TCA was used to clean parts. The emission estimate based on AP-42 heated cleaner emission factors previously described was 0.69 ton per year.

Building 777 - Ultrasonic Cleaner (Room 440)

A heated ultrasonic vapor degreaser containing TCA was used to clean metal filters. The emission estimate based on AP-42 heated cleaner factors previously described was 0.46 ton per year.

Building 777 - Downdraft Room 430, 432B, 433, and 440 Assembly and Cleaning

TCA was used to clean assembled parts using wetted lint-free wipes. A user estimate of 2 gallons per year was used. All used TCA was assumed to evaporate and the resulting calculated emission estimate was 1.1×10^{-2} ton per year.

Building 777 - Radiography

TCA was used for general cleaning. A user estimate of 5 liters per year was provided. Assuming all used TCA evaporated, the resulting calculated emission estimate was 7.4×10^{-3} ton per year.

Building 777 - Weighing

Parts were cleaned with TCA prior to weighing. A user estimate of 2 liters per year was used to estimate emissions. All TCA reportably evaporated into a glove box. The resulting calculated emission estimate was 3.0×10^{-3} ton per year.

Building 777 - Plutonium Metalography Laboratory

TCA was used as a cutting agent for grinding with a carbide tip. A user estimate of 24 gallons per year was provided. All TCA was assumed to evaporate, and the resulting calculated emission estimate was 0.13 ton per year.

Building 777 - Special Weapons Projects

TCA was used for general cleaning and in ultrasonic cleaners. A user estimate of 15 gallons per year for general cleaning was used, all TCA was assumed to evaporate, and the resultant emission estimate was 8.4×10^{-2} ton per year. Emission estimates for the ultrasonic cleaners based on AP-42 heated and cold cleaners (one of each assumed from the APEN calculations) emission factors previously described were 0.14 ton per year (cold cleaner) and 0.25 ton per year (heated cleaner).

Building 777 - TCA Collection and Filtration System

The TCA collection and filtration system collected TCA from buildings 707 and 776/777. Emission estimates for two tanks (T-1 and T-2) based on the AP-42, Section 4.3.1, "Storage of Organic Liquids" equation 2 were 4.94×10^{-3} ton per year (T-1) and 4.94×10^{-3} ton per year (T-2).

The total Building 776/777 TCA emission estimate was 8.36 tons per year.

Building 774 Processes

Building 774 handled TCA wastes received from Buildings 707 and 776/777. No emission controls for TCA were present in the building.

Organic and Sludge Immobilization System (OASIS)

OASIS received TCA and other organics from Buildings 707 and 776/777 and treated them by solidifying with gypsum cement. The treatment was performed in a glove box.

The OASIS emission estimate addressed any TCA not assumed to have evaporated from originating APENs (Buildings 707 and 776/777). Assuming all of the 1,478.4 gallons of TCA per year received evaporates, the emission estimate was 8.3 tons per year.

The total Building 774 TCA emission estimate was 8.3 tons per year.

Building 707 Processes

Building 707 contained foundry and casting operations and products assembly. No emission controls for TCA were present in Building 707. All waste TCA was gravity drained to tanks in the basement.

Assembly Operations - Modules D, E, and G

TCA was used for cleaning assembled parts in five ultrasonic cleaners (degreasers). There were four heated degreasers and one cold degreaser. Emission estimates for the degreasers based on AP-42 heated and cold cleaner emission factors (USEPA, 1985), as previously described, totaled 3.1 tons per year for all five degreasers. The degreaser emission estimates were considered conservative since the degreasers were used no more than 4 hours per day and the emission estimates assumed 16 hours of operation per day.

Assembly - Superdry - Module F

Parts were cleaned with TCA-wetted Kimwipes® prior to assembly. A user estimate of one gallon per month was provided. All used TCA was assumed to evaporate, and the resulting calculated emission estimate was 6.7×10^{-2} ton per year.

Assembly - Electron Bombardment Brazing/Scanning - Module G

TCA-wetted Kimwipes® were used to clean brazing operation bell jars. A user estimate of 24 gallons per year was provided. All used TCA was assumed to evaporate and the resulting calculated emission estimate was 0.13 ton per year.

Assembly Testing - Module H

Components were cleaned after quality control testing with TCA-wetted cheesecloth (infrequent use). A user estimate of less than a half gallon per year was provided. All used TCA was assumed to evaporate, and the resulting calculated emission estimate was 2.8×10^{-3} ton per year.

Radiography (Room 173)

TCA was used to clean parts prior to radiography. A user estimate of 5 liters per year was provided to estimate emissions. All used TCA was assumed to evaporate, and the resulting calculated emission estimate was 7.4×10^{-3} ton per year.

Weighing - Module D

TCA was used for general cleaning. A user estimate of 6 liters per year was provided. All used TCA was assumed to evaporate, and the resulting calculated emission estimate was 8.9×10^{-3} ton per year.

Eddy Current Testing - Module E

The same TCA use and quantity as Weighing — Module D were applied here with an emission estimate of 8.9×10^{-3} ton per year.

Weld Scanners and Fluorescent Penetrant Operations

TCA was used to clean dye from parts. A user estimate of 150 liters per year was used to estimate emissions. All used TCA was assumed to evaporate, and the resulting calculated emission estimate was 0.22 ton per year.

Production Control Operations - Module D

A 4-gallon capacity TCA dip tank was used to clean parts after grit blasting. A user estimate of 44 gallons per year based on estimated tank replenishment rates was used to estimate emissions. All replenished TCA was assumed to evaporate, and the resulting calculated emission estimate was 0.24 ton per year. This emission estimate did not account for bimonthly inventory refills or refills due to dirty TCA. Assuming 6 bimonthly inventory refills and 6 refills due to dirty TCA, a total emission estimate is 0.51 ton (versus the 0.24 ton per year APEN estimate). The larger emission estimate is more consistent with APEN assumptions, is more conservative, and will be used as an adjusted APEN emission estimate.

Calibration Laboratory - Modules D and G

TCA was used to clean gauges prior to performing precision measurements. A user estimate of 16 ounces per year was considered insignificant.

TCA Waste System

Waste TCA from ultrasonic cleaners (degreasers) was collected in a sump tank (V-100). The waste TCA was then pumped to Building 777 for subsequent pumping to Building 774 for treatment. Breathing losses were not calculated for the three feed tanks (V-36 A, B, and C) and the waste tank (V-100) due to the very small temperature change for indoor tanks and the fact that there were no day tanks involved. Working losses were calculated using AP-42, Section 4.3.1, "Storage of Organic Liquids" (USEPA, 1985) equation 2. The resulting calculated emission estimate for working losses from the three feed tanks was 7.6×10^{-3} ton per year based on a total 1989 TCA purchase for Buildings 707 and 777 of 2,450 gallons. The resulting calculated emission estimate for working losses from the waste tank was 9.0×10^{-4} ton per year based on an assumed waste tank throughput of 723 gallons per year.

The total Building 707 TCA emission estimate was 4.1 tons per year. This includes an adjustment for the higher emission estimate associated with the Production Control Operations — Module D TCA dip tanks.

E.5.2 Information Sources Other Than APEN Documents

The following resources were identified, evaluated, and found to contain TCA use or emission information to support a chemical emission source term.

A 1974 inventory (Barrick, 1974) indicated a total Rocky Flats Plant TCA inventory of 6,013 gallons (22,763 liters).

The Final Environmental Impact Statement for Rocky Flats (USDOE, 1980) listed a Rocky Flats Plant 1977 TCA consumption of 26 tons per year (4,675 gallons).

The volatile organics emissions report (Hamilton and Moser, 1990) provided detailed TCA emission estimates for Buildings 444, 460, 707, 774, and 776/777. The Hamilton and Moser report provides emission monitoring results in pounds per hour, which can be extrapolated using standard APEN operating assumptions to determine annual emission estimates. The resulting calculated emission estimates are summarized in Table E-9. All emission sampling was performed in July 1989. The Hamilton and Moser report was the first identified resource indicating significant TCA use and emissions from Building 444. Three other non-APEN resources were identified to confirm the use of TCA in Building 444 (USDOE, 1989; Ferrera, 1988; and Rockwell, 1976). The APEN for Building 444 (EG&G, 1991e) was reviewed, and although it did not identify any TCA emissions, it did list three TCA tanks as being out of service "due to plant program to minimize use of criteria, hazardous and toxic chemicals. . . ." Both the APEN report and the Hamilton and Moser report used 1989 as the basis year so the discrepancy cannot currently be explained. It is possible that the building TCA use was discontinued after the Hamilton and Moser monitoring and before the APEN evaluation. The documented use and emissions from Building 444 have been included in the source term evaluation.

The environmental team audit conducted in June 1989 (USDOE, 1989) identified "major" TCA users that generally supported the APEN information. Some APEN reported users were not identified by the environmental team audit report. The environmental team audit also reported a 1988 TCA usage of 47,630 pounds. Assuming all used TCA evaporated, a Rocky Flats Plant total emission estimate is 24 tons per year.

TABLE E-9: 1,1,1-TRICHLOROETHANE EMISSION ESTIMATES FROM MONITORING AND APENS

Building	Based on Monitoring* (tons/yr)	Based on APENs (tons/yr)	Operation/Comments
444	2.1	0	Intermittent part immersion in a TCA vapor degreaser vented only 40 minutes per week.
460	5.3	.0072	Submerging parts in Cee Bee cleaner followed by a TCA vapor degreaser bath followed by a TCA ultrasonic bath.
707	21.7	4.1**	Ultrasonic degreasers and general TCA use as a cleaning agent.
774	15.7	8.3	Organic waste solidification (OASIS).
776/777	1.2	8.36	Normal unspecified operations.
Total	**46**	**20.8**	

*Hamilton and Moser, 1990.
**Corrected total.

An internal Rockwell International chlorinated solvent usage report (Rockwell, 1988) confirmed APEN reported TCA uses for Building 707. The projected TCA usage in Building 707 was 3,500 gallons for 1988. Assuming all the used TCA evaporated, the resulting Building 707 TCA emission estimate is 20 tons per year.

A monthly status report on halogenated solvent use (Ferrara, 1988) documented planned efforts to reduce TCA use including reducing the Building 444 TCA use by 2,700 gallons per year. A "baseline" Rocky Flats Plant TCA use of 650 gallons per month was referenced based on 1987 and 1988 purchase records. Assuming all the used baseline TCA evaporated, a Rocky Flats Plant emission estimate is 44 tons per year. The Rocky Flats Plant solvent goal was to reduce the monthly TCA use from 650 gallons per month to 325 gallons per month.

An internal Rockwell International halogenated solvent usage update report (Rockwell, 1989) indicated an average TCA usage of 250 gallons per month for 1988-1989. Assuming all the used TCA evaporated, a Rocky Flats Plant emission estimate is 17 tons per year.

An EG&G Rocky Flats air stack release tabulation (EG&G, circa 1990) indicated 1988 and 1989 air stack emissions from the Rocky Flats Plant of 47,000 and 45,600 pounds, respectively. Converting to tons per year, this corresponds to 24 tons per year (1988) and 23 tons per year (1989).

A Rockwell International monitoring report (Rockwell, 1976) provided Building 444 TCA exhaust duct monitoring results in parts per million (ppm). An extrapolated emission estimate based on available exhaust duct flow rate and operating data (Hamilton and Moser, 1990) was calculated to be 5.0×10^{-5} ton per year. This result appears to contradict the Hamilton and Moser (1990) emission estimate of 2.1 tons per year; however, the discrepancy can likely be explained by the difference in time periods and monitoring methodologies.

The Waste Stream and Residue Identification and Characterization report for Building 776 (Wastren Inc., 1991e) did not cite any TCA waste generation. This may support the APEN assumption that all TCA associated with the Building 776 baler evaporated.

The Waste Stream and Residue Identification and Characterization report for Building 777 (Wastren Inc., 1991b) cited the generation of 2,802 gallons of TCA waste per year. The documented TCA waste sources generally supported the APEN reported users.

The Waste Stream and Residue Identification and Characterization report for Building 707 (Wastren Inc., 1991a) cited the generation of 3,854.5 gallons of TCA waste per year. The documented TCA waste sources generally supported the APEN reported users; however, the waste generation rate estimate is very high compared to the waste tank throughput reported in the Building 707 APEN.

Table E-10 summarizes the Rocky Flats Plant total TCA emission estimates including APEN and non-APEN resources.

E.6 Trichloroethylene Emission Evaluation

Information sources relevant to trichloroethylene (TCE) emissions from Rocky Flats and the evaluation process used to develop associated source terms are described in this section.

Information Sources relevant to TCE emissions at Rocky Flats include two APEN reports and twelve other comparable resources, ranging from personal interviews to hazardous material inventories and technical reports by Rocky Flats Plant and outside groups. The key information sources are described and evaluated in this section.

E.6.1 APEN Resources

Two APENs were initially identified documenting TCE use and emissions. These APENs are for Buildings 460 and 374. Other buildings that APENs did not identify as TCE users were not considered further unless some other resource indicated differently. The APEN for Building 444 was added to the list of APENs to be evaluated for TCE emissions when the Building 374 APEN (EG&G, 1991d) reported TCE in waste from Building 444.

Selected APENs with identified TCE users were reviewed in detail for any significant flaws in the emission estimate determinations. Given the plant's elimination of TCE usage for activities other than those related to research and analytic functions starting in 1975, the use of TCE in Building 460 reported in the APEN was questioned. A plant review, initiated at ChemRisk's request, revealed that TCE use reported for Building 460 was in fact TCA (Costain, 1992). TCE emission estimates for Building 460 have been reviewed and treated as TCA emissions.

The TCE emission estimate basis for the Building 374 APEN was a user estimate. Mass balance information was not available, there was no chemical tracking system to document chemical use and movement, and the USEPA AP-42 emission factors did not apply to the Rocky Flats Plant TCE uses.

The emission estimates for Building 374 were thoroughly evaluated. The APEN accounted for building vents in detail to demonstrate complete emission inclusion. The following is a summary of the major identified TCE users based on the APEN.

TABLE E-10

Building 374 Processes

Building 374 was activated in 1978 as the process waste treatment facility for many of the production buildings. Emission controls for TCE were not present.

Process Waste Treatment

TCE was detected in 1986 waste stream tests for Building 444, 779, and 889. A 1987 user estimate (waste generation rate) was the basis of an emission estimate of 1.5×10^{-3} ton per year. All TCE was assumed to evaporate. This emission estimate did not include the Building 444 emission estimate (4.95×10^{-5} ton per year), with the reasoning that solvent use in Building 444 had been eliminated since the 1986 data had been collected.

ChemRisk attempted to evaluate APENs for other buildings identified as waste sources of TCE in the Building 374 APEN. The Building 444 APEN was reviewed and no reference to TCE use or emission was identified. APENs are not available for Buildings 779 and 889.

E.6.2 Information Sources Other Than APEN Documents

The following resources were identified, evaluated, and found to contain information to support a chemical emission source term.

A 1974 inventory (Barrick, 1974) indicated a total Rocky Flats Plant TCE inventory of 4,4041.6 gallons (15,300 liters).

A 1989 inventory printout (Grocki, 1989b) indicated a total Rocky Flats Plant TCE inventory of 13 milliliters.

Stack emission monitoring results for Building 776/777 (Johnson, 1973b) indicated TCE emissions from Building 776/777 in 1973 as 5.0 tons per year (average) and 17.9 tons per year (maximum). A separate resource (Dow Chemical, 1974) described the Johnson, 1973b, sampling methodology and discussed the results. Sampling and analysis was performed with a portable gas chromatograph and a flame ionization detector. Samples were collected every 15 minutes from Booster No. 1 exhaust. The TCE source was ultrasonic cleaners and cleaning baths.
A Dow Chemical report on the annual use of TCE (Dow Chemical, 1972-1974) indicated a total Rocky Flats Plant TCE 1973 usage of 60 tons per year (9,790 gallons) (Table E-11).

TABLE E-11

An undated Rocky Flats Plant report (Musgrave, circa 1973) discussed possible replacements for TCE. TCE was described as the "workhorse solvent for metal degreasing over the past 20 years." Prior to 1963, Rocky Flats Plant personnel cleaned metal parts with acetone, isopropanol, and other solvents. Beginning in 1963, ultrasonic vapor degreasing with stabilized trichloroethylene was the prime degreasing process. TCA was considered the best choice replacement for TCE for degreasing beryllium, plutonium, and uranium. Fiscal year 1973 Rocky Flats Plant TCE use was reported to be 10,000 gallons. Assuming all used TCE evaporated, an emission estimate is 62 tons per year.

The Final Environmental Impact Statement for Rocky Flats (USDOE, 1980) listed a Rocky Flats Plant 1977 TCE consumption of 2 tons per year (330 gallons).

A Rocky Flats Plant memorandum to the USEPA (Rocky Flats Plant, 1975) described the Rocky Flats Plant schedule for compliance with USEPA regulations requiring the use reduction and/or control of TCE for degreasing operations. This memorandum stated that TCE was no longer used at the Rocky Flats Plant as of December 1, 1974 except in one plutonium operation, which used up all TCE inventory by February 10, 1975. Since then, the only TCE use was reported to have been for research and analytical purposes using "insignificant small quantities." TCE was replaced with detergent washes in non-plutonium areas and TCA in plutonium areas. Other resources confirming the historical (pre-1975) large-scale use of TCE for degreasing were identified including, Hamilton and Moser (1990), ChemRisk (1991-1992; Interview No. 35); Dow Chemical (1974), and Dow Chemical (1965-1974) industrial hygiene memoranda.

The Waste Stream and Residue Identification and Characterization reports for Buildings 460 and 364 (Wastren Inc., 1991f and 1991g) referenced TCE waste streams.

Table E-12 summarizes the Rocky Flats Plant total TCE emission estimates including APEN and non-APEN resources.

REFERENCES

Barrick (1974). Barrick, C.W. Harmful and Potentially Harmful Material Inventory, Memorandum from C. W. Barrick to J. B. Owen. Dow Chemical Company, January 25, 1974. Repository Document CH-93.

ChemRisk (1991-1992). Rocky Flats Historical Investigation Interviews. Repository Document RE-891. The specified number(s) identify the specific interviewees. Appendix A of the Task 3 & 4 report identifies the interviewees for each interview number.

TABLE E-12

Church (1989). Church, A. A. Halogenated Solvent Usage Update. Rockwell International, August 11, 1989. Repository Document CH-947.

Costain (1992). Letter from D. Costain of EG&G Rocky Flats to S. Ripple of ChemRisk Re: Verification of Chemical Use and Emissions, Cited-DBC-0019-92. August 6, 1992.

Dow Chemical (1965-1974). Industrial Hygiene Memorandums, J. Hill, S. Hammond, and J. Handschy. Repository Document CH-951.

Dow Chemical (1966). Manufacturing Technical Report, Oil Coolant Waste Disposal, The Dow Chemical Company, Rocky Flats Division, Anderson, D.M., MT-42-66, DPA 950087, December 22, 1966. Repository Document RE-932.

Dow Chemical (1972-1974). Trichloroethylene Folder, Dow Chemical, 1972-1974. Repository Document CH-896.

Dow Chemical (1973). Use of Trichloroethylene at Rocky Flats, The Dow Chemical Company, October 19, 1973.

Dow Chemical (1974). Hydrocarbon Emissions Booster No. 1, Building 776/777, CRDL 950373-104, C. M. Johnson, L. H. Faber, J. L. Lawless, Chemistry Research and Development, Dow Chemical U.S.A., Rocky Flats Division, August 12, 1974. Repository Document RE-822.

EG&G (circa 1990). Air Stack Release for 1988 and 1989, EG&G Rocky Flats, 1988 and 1989. Repository Document RE-76.

EG&G (1990a). Air Pollution Emission Notice — Building 707. October 12, 1990. Repository Document RE-245.

EG&G (1990b). Air Pollution Emission Notice — Building 776/777. October 24, 1990. Repository Document RE-224.

EG&G (1990c). Air Pollution Emission Notice — Building 559/561. August 24, 1990. Repository Document RE-242.

EG&G (1990d). Chemical Inventory Database File, EG&G Rocky Flats Plant, September 5, 1990. Repository Document CH-66.

EG&G (1991a). Air Pollution Emission Notice — Building 460. April 9, 1991, Repository Document RE-649.

EG&G (1991b). Air Pollution Emission Notice — Building 881. May 9, 1991. Repository Document RE-773.

EG&G (1991c). Air Pollution Emission Notice — Building 774. February 19, 1991. Repository Document RE-654.

EG&G (1991d). Air Pollution Emission Notice — Building 374. March 11, 1991. Repository Document RE-646.

EG&G (1991e). Air Pollution Emission Notice — Buildings 444, 445, 450, and 455. EG&G Rocky Flats, April 15, 1991. Repository Document RE-647.

Ferrera (1988b). Ferrera, K. P. Monthly Status — Halogenated Solvents. Rockwell International, December 8, 1988. Repository Document CH-948.

Fruehauf and Richter (1974). Fruehauf, V. and T. Richter. Handling and Monitoring of Carbon Tetrachloride. Memorandum, Safety Concern #73-56, January 8, 1974. Repository Document RE-495.

Grocki (1989a). Grocki, L. Rocky Flats Usage of Methylene Chloride and Tetrachloroethylene. October 3, 1989. Repository Document CH-844.

Grocki (1989b). Grocki, L. Rocky Flats Usage of Trichloroethylene. October 3, 1989. Repository Document CH-845.

Hamilton and Moser (1990). Hamilton, T.H. and L. V. Moser. "Volatile Organics Emission Report for the U.S. Department of Energy, Rocky Flats Plant." Martin Marietta Energy Systems Inc., March 1990. Repository Document RE-935.

Hobbs (1982). Hobbs, F. D. Carbon Tetrachloride Emissions from Production Area, Buildings 707 and 776/777. Rockwell International, October 14, 1982. Repository Document RE-281.

Johnson (1971). Johnson, T.C. Carbon Tetrachloride Vapor Emission from Building 774. Memorandum to B. L. Kelchner, March 1, 1971. Repository Document RE-301.

Johnson (1973a). Johnson, T. C. Monthly Environmental Progress Report-Stack Emissions Monitors. July 1973. Repository Document CH-850.

Johnson (1973b). Johnson, C. M. Stack Emissions Monitors. Report Index C-37, August 1973. Repository Document RE-544.

Kelchner (1971). Kelchner, B. L. Emissions of Carbon Tetrachloride Vapor From Building 774. Memorandum to W. C. Bright, February 5, 1971. Repository Document RE-303.

Musgrave (circa 1973). Musgrave, L. E. Use of Trichloroethane for Degreasing Beryllium, Plutonium, and Uranium. Rocky Flats Report RFP-223. Repository Document CH-905.

Porter (1992). Personal communication with Georgine Porter, January 1992.

Rocky Flats Plant (Date Unknown). Rocky Flats Plant Toxic Substances Inventory. Repository Document CH-836.

Rocky Flats Plant (1975). Schedule for Compliance with USEPA Regulations on Solvents for Degreasing Operations. E. W. Bean, Rocky Flats Plant Memorandum to I. L. Dickstein. (USEPA). March 17, 1975. Repository Document CH-852.

Rocky Flats Plant (1990). Master List of Carcinogen Control Areas, Internal Industrial Hygiene Printout. November 29, 1990. Repository Document CH-80.

Rockwell (1976). Miran Determination of 1,1,1-Trichloroethane Vapors in Exhaust Duct on Roof of Building 444 (Vapors Originating from Degreaser Bath in Plating Area). Rockwell International, P. A. Hymen. July 16, 1976. Repository Document CH-899.

Roy F. Weston (1985). Hazardous Waste Potentially Generated at Rocky Flats Plant. Roy F. Weston Memorandum to D. Hornbacher (Rocky Flats Plant), September 10, 1985. Repository Document CH-848.

USDOE (1980). U.S. Department of Energy. Final Environmental Impact Statement Rocky Flats Plant Site, Jefferson County, Colorado. Final Statement to ERDA 1545-D, Three Volumes, U.S. Department of Energy, DOE/EIS 0064, April 1980. Repository Document EN-1-3.

USDOE (1989). U.S. Department of Energy. Assessment of Environmental Conditions at the Rocky Flats Plant. U.S. Department of Energy, August 1989. Repository Document RE-888.

USEPA (1985). U.S. Environmental Protection Agency. Compilation of Air Pollutant Emissions Factors. USEPA Office of Air Quality Planning and Standards, Research Triangle Park, North Carolina, AP-42, September 1985 and supplements.

Wastren Inc. (1991a). Waste Stream and Residue Identification and Characterization, Building 707. Two Volumes, Version 3.0, January 1991. Repository Documents RE-668 and RE-673.

Wastren Inc. (1991b). Waste Stream and Residue Identification and Characterization, Building 777. Two Volumes, Version 3.0, January 1991. Repository Documents RE-672 and RE-673.

Wastren Inc. (1991c). Waste Stream and Residue Identification and Characterization, Building 881. Two Volumes, Version 3.0, January 1991. Repository Document RE-366.

Wastren Inc. (1991d). Waste Stream and Residue Identification and Characterization, Building 559. Version 3.1, January 1991. Repository Document RE-666.

Wastren Inc. (1991e). Waste Stream and Residue Identification and Characterization, Building 776. Version 3.0, January 1991. Repository Document RE-671.

Wastren Inc. (1991f). Waste Stream and Residue Identification and Characterization, Building 374. Version 3.0, January 1991. Repository Document RE-662.

Wastren Inc. (1991g). Waste Stream and Residue Identification and Characterization, Building 460. Version 3.0, January 1991. Repository Document RE-364.

Weis (1988). Weis, M. J. Chlorinated Solvent Usage — Building 707, Part 1, Part 2, and Final of the Chlorinated Solvent Study. Rockwell International, September 23, 1988. Repository Document CH-903.