

# Reassessment of Internal Radiation Dose from Sources at the Rocky Flats Plant Critical Mass Laboratory

White Paper  
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## **PURPOSE**

This report reevaluates prior assumptions used to assess upper bounds on personnel doses from mixed fission and activation products (MFAP) at the Rocky Flats Plant (RFP) Critical Mass Laboratory (CML). A publicly released document citing a maximum reactor operating power of 10 milliwatts (mW) and a typical experiment duration of one hour were previously used to estimate the MFAP inventory built up over time in high-enriched uranium (HEU) solution fuel used in CML experiments. External radiation dose was monitored by radiation dosimeters assigned to each individual and has not been considered herein.

In the earlier assessment, internal doses from the estimated MFAP inventory were bounded by assuming removable surface contamination from dried solution spills to be at the limit for a posted Contamination Area. The average air concentration of respirable particles was then estimated by applying a resuspension factor. Recently captured documents contain air monitoring results, surface contamination measurements, more-accurate power estimates based on carefully measured gamma photon emissions from the irradiated fuel, records of experiment duration, and neutron flux profiles. This information provides data against which the prior assumptions can be re-evaluated.

## **MFAP PERSONNEL EXPOSURE CONCERNS**

Fission products in irradiated fuels, and activation products in both the fuel and containment materials, are sources of external radiation dose to personnel using or working around the fuels, and they present an internal dose potential for personnel who might ingest or inhale them. Personnel dosimeters assigned to RFP radiation workers document the external exposures. Internal exposures might result: (1) during operations, from resuspension of contamination on surfaces, or (2) during facility demolition from airborne dust. CML staff was provided routine bioassay (urinalysis and whole-body counts) to detect intakes of plutonium, uranium, or americium, but MFAP were not routinely monitored.

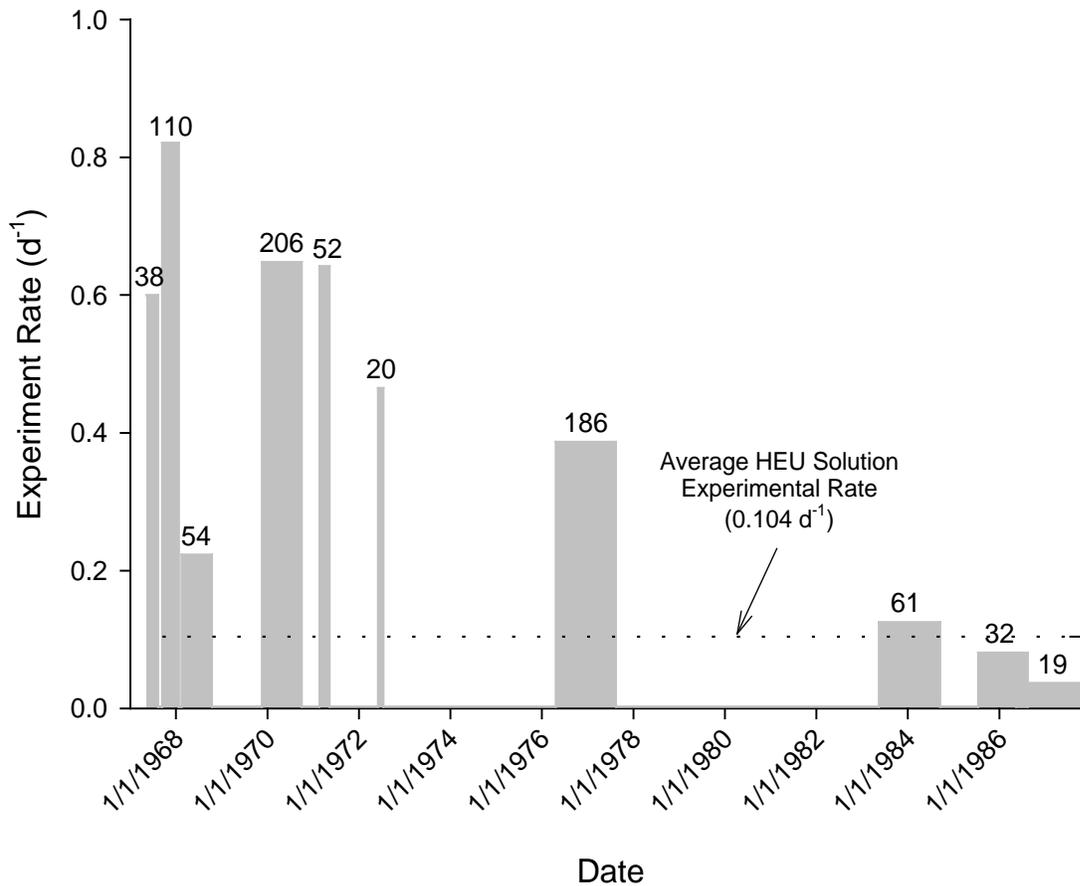
Surface contamination in CML experimental areas was extensive, predominantly due to high-enriched uranyl nitrate (HEU) solution spills<sup>1</sup> over the course of the facility's history (Rothe, 2005, PDF pp. 447, 449, 452, 454-458, 464, 467-471, 479, 486, 498, 500-502). Thus, MFAP important from either an acute or chronic personnel internal exposure perspective are those generated by CML criticality experiments involving HEU solution. MFAP atom ratios on CML contaminated surfaces and resuspended in air from these surfaces are presumed to be the same as those in solution.

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<sup>1</sup> Eleven spills involved solutions or dried salts from solution experiments, compared with two contamination events involving other solid fuels (one spill each of low-enriched uranium and plutonium powders).

## HISTORY OF UNH CRITICALITY EXPERIMENTS

Criticality experiments involving uranyl nitrate solution were conducted from the beginning of CML operations until the last experiment in 1987. These experiments are documented in some detail in a published history of the facility (Rothe, 2005, PDF pp. 376-394). Figure 1 shows the distribution of these experiments over time. Heights of the bars represent the average number of experiments per day during the Program period discussed in the published history; numbers above the bars are the total numbers of experiments. The dotted line shows the overall average number of HEU solution critical experiments over the history of CML operations.



Source: Rothe, 2005, PDF pp. 376-394

**Figure 1: Time Distribution of CML Criticality Experiments Involving HEU Solution**

The 778 experiments performed at the CML with HEU solution or systems of HEU solution plus HEU metal took place in 10 campaigns between May 1967 and October 1987. Table 1 summarizes the 10 campaigns, where the “Downtime (d)” for each campaign equals the number of days between the last experiment of the specified campaign and the first experiment of the subsequent campaign.

**Table 1: CML Experimental Campaigns Involving HEU Solution**

Campaign	No. of Experiments	Downtime (d)
1	38	32
2	110	23
3	54	398
4	206	111
5	52	387
6	20	1,377
7	186	2,069
8	61	274
9	32	182
10	19	N/A

### **REEVALUATED FISSION AND ACTIVATION PRODUCT LEVEL ESTIMATES**

NIOSH calculated fission and activation product build-up in uranium solution experiments at the CML using ORIGEN-S, a computer code system for calculating time-dependent concentrations of radionuclides that are simultaneously generated or depleted by processes such as fission, neutron absorption/transmutation, and radioactive decay. Initial assumptions about power levels and experiment durations were taken from a U.S. Department of Energy (DOE) public document that stated that the experiments conducted in Building 886 (housing the CML) generally involved power levels of no more than 10 mW for no more than one hour. It also stated that approximately half of the experiments conducted in Building 886 actually achieved criticality, and only rarely were the radiation levels such that it was not possible to directly touch the fissile material and testing apparatus immediately after the experiments (Building 886, 2011, PDF p. 3). Some of these assumptions were challenged during an oral interview with a former CML Associate Research Scientist, who asserted: (1) that the neutron flux for a CML experiment could not be bounded; and (2) that the best one could say is that the power level was probably less than 50 kW [the DOE Category B reactor limit] (Personal Communication, 2015a, PDF p. 9).

Documents subsequently captured by NIOSH contain neutron flux and thermal power estimates by CML staff using analytical measurements made during or after criticality experiments. These calculations were performed for at least three different enriched uranium fuel configurations: (1) bare HEU metal (Oy) spheres; (2) Oy spheres immersed in uranyl nitrate solution (UNH); and (3) UNH only.

### **Thermal Power and Fission Rate Estimates in Bare Metal Experiments**

Measurements of surface gamma photon fluence rates from a 7-cm-radius Oy sphere used in a 1967 one-hour criticality experiment led to an estimate of  $1.9508 \times 10^{-9}$  megawatts (MW) ( $1.9508 \text{ mW}$ )<sup>2</sup> thermal power, corresponding with  $2.18 \times 10^{11}$  total fissions<sup>3</sup> (Log, 1967, PDF p. 6).

### **Thermal Power and Fission Rate Estimates in Metal-Plus-Solution Experiments**

CML staff made two estimates of total fissions in experiments involving Oy immersed in UNH. One 1976 estimate used the residual gamma photon flux from the Oy + UNH to estimate an upper bound of  $1.8 \times 10^{18}$  total fissions over the 12 years of CML operation from 1965-1976<sup>4</sup> and an average power of 25 W (Log, 1976, PDF pp. 2-4). However, the senior scientist who performed the measurements and derived the estimate noted that the gamma photon background from a new unirradiated Oy part was the same as the gamma flux measured from irradiated parts used for the analysis. This means that measurements, made six years after the last criticality experiment, were unable to detect additional gammas from prior experiments because of radioactive decay of the fission and activation products. The original estimate did not take into account the intrinsic gamma photon emissions from unirradiated enriched uranium and was, therefore, invalid<sup>5</sup> (Log, 1976, PDF p. 2).

A second estimate of  $3.44 \times 10^{10}$  fissions in an average Oy + UNH experiment with a 20-minute run time was derived by estimating the number of neutrons produced ( $8.4 \times 10^{10}$ ) using the current generated by a neutron detector associated with an April 1977 experiment (Experiments at RFNSF, 1977b, PDF p. 3). This fission rate corresponds with a thermal power level of 0.92 mW.

### **Thermal Power and Fission Rate Estimates in Solution-Only Experiments**

The CML staff's most rigorous estimates of fission rates were derived for UNH (solution-only) fuel by measuring the rate of gamma decay in the fuel of Experiment 2-8-170. This experiment was configured as a 2x2 array of 8-in-diameter tanks and was performed on May 4, 1977 (Experiments at RFNSF, 1977a). The gamma photon count rate of a 2-mL aliquot withdrawn from the fuel after a 70.5-minute run was determined at 2- to 3-minute intervals using a "well crystal" calibrated to Cs-137 photons. The total number of fissions in the reactor was calculated

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<sup>2</sup> The conversion from MW to mW in the reference document incorporated a factor of ten error, incorrectly concluding that the power level was 19.508 mW; the error is corrected here.

<sup>3</sup> Using  $1 \text{ W} = 3.1 \times 10^{10}$  fissions per second.

<sup>4</sup> The estimate of total fissions is based on 27 days of continuous run time occurring in the middle of the 12-year period and corresponds with  $7.7 \times 10^{11}$  fissions per second.

<sup>5</sup> The laboratory notebook entry reads: "*All  $\gamma$  due to fission 6 years ago. (No believable background available.) Note: Part no. 80 never used and yet had same background/kg as others. [Therefore] Feel fission too low to detect by this method.*"

from the measured photon emission rate (Analysis Report, 1977, PDF p. 7) using Equation 1 below:

$$\dot{\Phi}_{\gamma} \cong 1.9 \times 10^{-6} \tau^{-1.2} \text{ photons/(second)(fission)}$$

Where:

$\dot{\Phi}_{\gamma}$  = Reactor gamma photon emission rate at time  $\tau$

$\tau$  = Days after shutdown

An initial evaluation<sup>6</sup> of the data (Introducing Nuclear Reactor Theory, 1977) arrived at a power estimate of 38 mW during the last 10 minutes of the experiment (when most of the corresponding  $7.1 \times 10^{11}$  fissions occurred), giving an average power of 5.4 mW for the 70.5-minute duration of the experiment.

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<sup>6</sup> Reactor power was evaluated using the integral of  $\dot{\Phi}_{\gamma}$  over the operating time of the reactor, giving the gamma energy emission rate as a function of reactor power and time after shutdown.

A subsequent June 7, 1977 estimate from Experiment 2-8-170 employed more precise values for the aliquot and reactor volumes (Analysis Report, 1977, PDF p. 4). The estimate gave 25 mW at the end of the experiment, corresponding with  $4.73 \times 10^{11}$  fissions (or 3.6 mW, average power). These results were within 2% of those obtained by the method of source multiplication (Summary of All Nuclear Safety Experiments, 1977; Experiment Run 2-8-170, Undated), using Equation 2 below:

$$f = s\bar{\mu}t$$

Where:

$f$  = Total number of fissions over time  $t$

$t$  = Time

$\bar{\mu}$  = Average multiplication from the neutron detector response during the experiment

$s$  = Beginning neutron flux from the Cf-252 seed source

On June 3, 1977, before the more precise June 7 calculation was completed, CML staff communicated officially to the Energy Research and Development Administration (ERDA). They provided an estimate of  $8.8 \times 10^{11}$  fissions and an average power of 6.7 mW for a typical (70.5 minute) experiment (Tuck, 1977). There is no indication why the values reported to ERDA were almost 25% higher than results of the May 4, 1977 calculation.

## Conclusions about Reactor Power and Fission Rates in CML Experiments

Table 2 provides a summary of the different CML criticality experiments for which thermal power and fission rate were estimated from measurements.

**Table 2: Average Reactor Power and Fission Rates Derived from Measurements at CML**

Configuration	Duration	Average Power (mW)	Total Fissions	Fission Rate (s <sup>-1</sup> )	Comment
Oy	60 min	1.9508	2.18E+11	6.06E+07	Based on exposure rate at the surface of an Oy sphere converted to $\gamma$ photon flux.
Oy + UNH (a)	27 d	25,000	7.7E+11	3.30E+05	Estimate is invalid; measurement results were indistinguishable from background.
Oy + UNH (b)	20 min	0.92	3.44E+10	2.83E+07	From neutron detector current.
UNH (a)	70.5 min	5.4	7.1 $\times$ 10 <sup>11</sup>	1.68E+08*	Initial calculation on May 4, 1977, for Experiment 2-8-170
UNH (b)	70.5 min	6.7	8.8E+11	2.08E+08*	Values reported to ERDA on June 3, 1977, based on results of Experiment 2-8-170.
UNH (c)	70.5 min	3.6	4.73E+11	1.12E+08	Recalculated values from Experiment 2-8-170 on June 7, 1977. Result is within 2% of that obtained from evaluation of source multiplication.

Source: Log, 1967, PDF p. 6 (Oy); Log, 1976 [Oy + UNH (a)]; Experiments at RFNSF, 1977b [Oy + UNH (b)]; Analysis Report, 1977, PDF p. 7 [UNH (a)]; Tuck, 1977 [UNH (b)]; Analysis Report, 1977, PDF p. 4 [UNH (c)].

\*These values are not reported in the original reference, but are calculated from total fissions and duration.

CML staff described Experiment 2-8-170 to ERDA as having a higher-than-normal power level (Tuck, 1977, PDF p. 2). A thermal power estimate of 3.6 mW averaged over 70.5 minutes (corresponding with  $4.73 \times 10^{11}$  total fissions) is based on careful evaluation of measurements made on Experiment 2-8-170. NIOSH considers this the most precise estimate available for this relatively high-power experiment. CML staff, however, reported to ERDA an average thermal power of 6.7 mW over 70.5 minutes ( $8.8 \times 10^{11}$  total fissions) based on the same experiment. NIOSH has concluded that the value reported to ERDA is the most appropriate value to represent the typical CML criticality experiments for the purpose of estimating doses from fission and activation products. This value is less than the earlier assumption that power averaged 10 mW for experiments lasting an hour ( $1.1 \times 10^{12}$  total fissions). However, the value is still claimant-favorable because it exceeds the most careful estimate of power for the experiment, and because not all CML experiments achieved criticality (Building 886, 2011, PDF p. 3).

## CML SURFACE CONTAMINATION

### Radiological survey results

Internal doses from the estimated MFAP inventory were bounded in the earlier estimate by assuming that removable surface contamination from dried solution spills was evenly dispersed over CML surfaces at the DOE limit (1,000 dpm/100 cm<sup>2</sup>) for a posted Contamination Area. The average air concentration of respirable particles was then estimated by applying a

resuspension factor. Recently captured documents contain both air monitoring and surface contamination measurement results against which the prior assumptions can be reevaluated.

Radiological survey results for Building 886 and 875 over the period January 1981 – December 1990 are captured in the following 105 files:

Radiological Surveys, 1980a-1980l; 1981a-1981m; 1982a-1982l; 1983a-1983k; 1984a-1984m; 1985a-1985m; 1986a-1986l; 1987a-1987e; 1988a-1988e; 1989a-1989g; 1990a, 1990b.

Information in these files includes results of daily removable contamination surveys in the office and experimental areas, and on equipment, as well as weekly penetrating-dose measurements in the nuclear material storage and experimental areas of Building 886.

### **Maximum Reported Values**

Based on the data in the files listed in the subsection above, Table 3 shows the maximum reported values of removable alpha surface contamination for each year.

**Table 3: Maximum Removable Alpha Contamination (dpm/100 cm<sup>2</sup>)**

<b>Year</b>	<b>Bldg. 886 Offices*</b>	<b>Bldg. 886 Experimental Area**</b>	<b>Bldg. 886 Equipment</b>	<b>Bldg. 875 Outside Plenum</b>	<b>Bldg. 875 Inside Plenum</b>
1981	30	366	156	12	11196
1982	18	174 (3930)	--	12	14346
1983***	<20	576	--	--	--
1984	<20	54000	--	<20	15660
1985	<20	174	--	<20	4788
1986	18	186 (2784)	3954	12	5394
1987	18 (24)	564	--	--	--
1988	12	1683	--	12	15240
1989****	18	18707	--	--	--
1990****	18	16212	--	6	--

Source: See the list of 105 files in the preceding subsection.

NOTE: Values in parentheses are singular outliers and not representative.

\* “Offices” includes control points (corridors and common areas) typically surveyed daily and individual offices surveyed weekly.

\*\*The experimental area (Rooms 101, 102 and 103) were surveyed weekly in 1981, but the frequency gradually diminished, apparently in response to decreased activities in these areas. Surveys were made approximately monthly by 1990.

\*\*\*Data in 1983 are available only for November and December.

\*\*\*\*Elevated values of this order were observed in weekly surveys of the Room 101 walk-in hood from December 1989 through April 1990, as discussed in the text.

Based on the data in the files listed in the subsection above, Table 4 shows maximum gamma and neutron penetrating dose rates.

**Table 4: Maximum Dose Rates in CML Experimental and Material Storage Areas**

<b>Year</b>	<b>Gamma* (mR/h): UNH Tank Room</b>	<b>Gamma* (mR/h): Solid Fuel Storage</b>	<b>Gamma* (mR/h): Experiment Room</b>	<b>Neutron** (mrem/h): UNH Tank Room</b>	<b>Neutron** (mrem/h): Solid Fuel Storage</b>	<b>Neutron** (mrem/h): Experiment Room</b>
1981	2.0	2.9	--	0.3	12.9	--
1982	2.0	2.5	3.7	0.2	14.7	5.4
1983	--	--	--	--	--	--
1984	5.0	3.2	--	0.2	0.6	--
1985	2.0	2.8	--	0.1	0.5	--
1986	2.0	6.0	0.4	1.0	2.6	0.2
1987	3.0	2.0	3.0	1.7	2.0	3.4
1988	2.4	1.5	1.8	2.0	0.8	0.6
1989***	2.8	4.2 (250)	0.0	1.0	1.8 (6.0)	0.6
1990	2.53	3.0	--	0.3	0.5	--

Source: See the list of 105 files in the preceding subsection.

NOTE: Values in parentheses are singular outliers and not representative.

\*Gamma dose rate surveys were made at least weekly during the entire 10-year period.

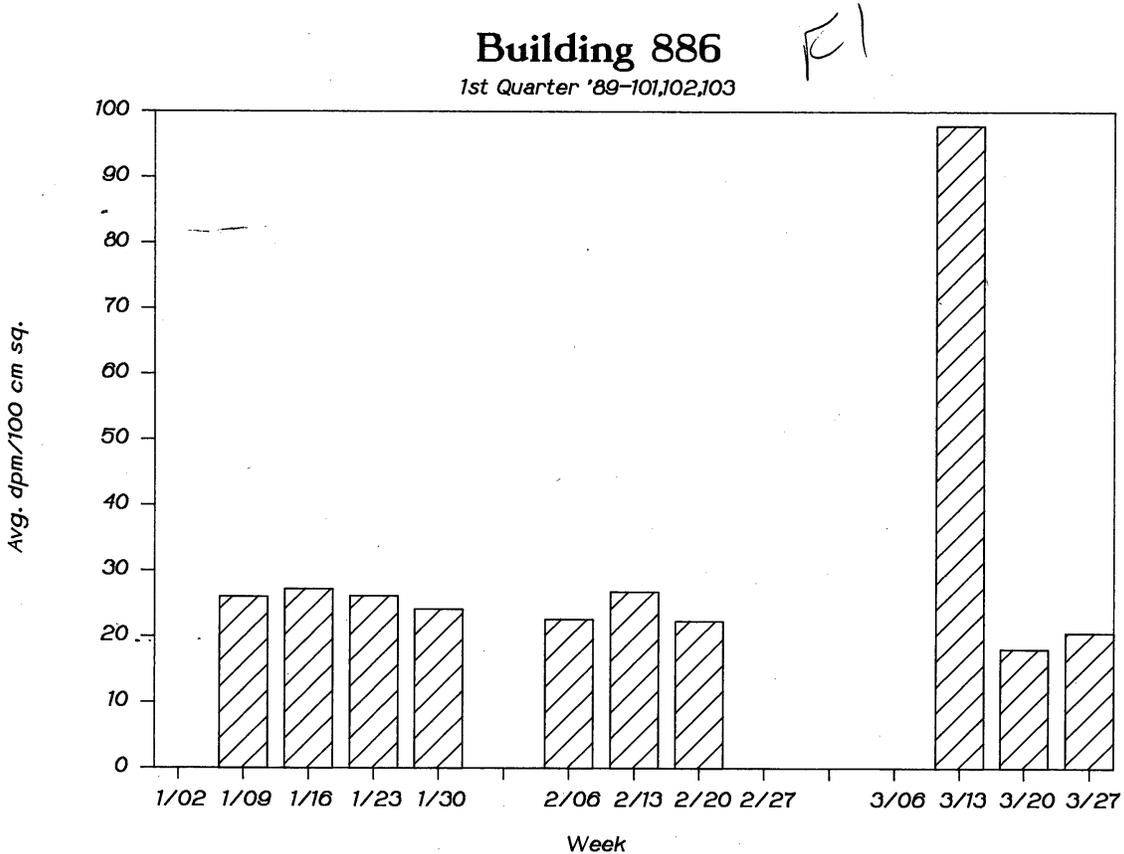
\*\* Neutron dose rate surveys were made weekly, along with gamma surveys, until 1990 when the frequency was reduced to monthly.

\*\*\*Atypically-elevated dose-rate readings reported for July 6, 1989 are associated with a survey location within an area marked off inside the solid storage area.

### **Evaluation of Removable Contamination Survey Results**

Contamination surveys were conducted daily at control points (hallways, doorways to offices), and weekly in offices within the unrestricted-access portion of Building 886. Surveys were conducted weekly in the experimental areas. Results of periodic contamination surveys in Building 875 and its air filtration plenums are also documented. Survey results for the office area are captured through December 20, 1990 (Radiological Surveys, 1990a, PDF p. 778), and for the experimental and material storage areas through April 1990 (Radiological Surveys, 1990a, PDF p. 330). No average values were found, except in 1989 documents, which contained a few graphs showing average contamination values in different areas for limited time periods (Radiological Surveys, 1989b, PDF pp. 3-6). These graphs are shown in Figures 2 through 5 below.

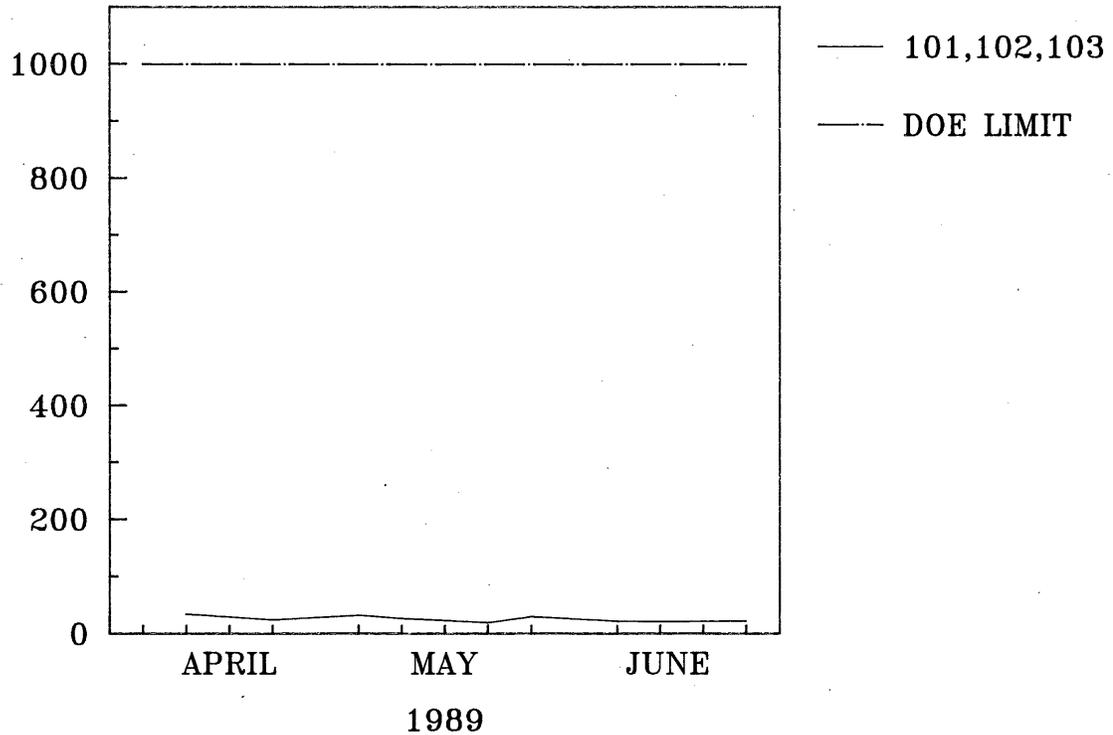
Table 3 above, showing maximum values for the removable alpha survey results in both the office and experimental areas, demonstrates that radiological containment of the contaminated areas was quite effective. Excursions of removable contamination above the 20 dpm/100cm<sup>2</sup> DOE limit in “cold” (office) areas is seen to occur only a few times (and only at low levels) in 1981 and once in 1984. Survey results in offices and hallways were typically well below 20 dpm/100 cm<sup>2</sup>, particularly toward the end of the period from 1981-1990 (e.g., see Figures 4 and 5).



Source: Radiological Surveys, 1989b, PDF p. 3

**Figure 2: Weekly Averages of Removable Alpha Contamination in the Building 886 Experimental and Material Storage Areas - Q1 1989**

# BUILDING 886 – CONTROLLED AREA CONTAMINATION SMEAR SURVEYS AVG. DPM/100 CM SQ.



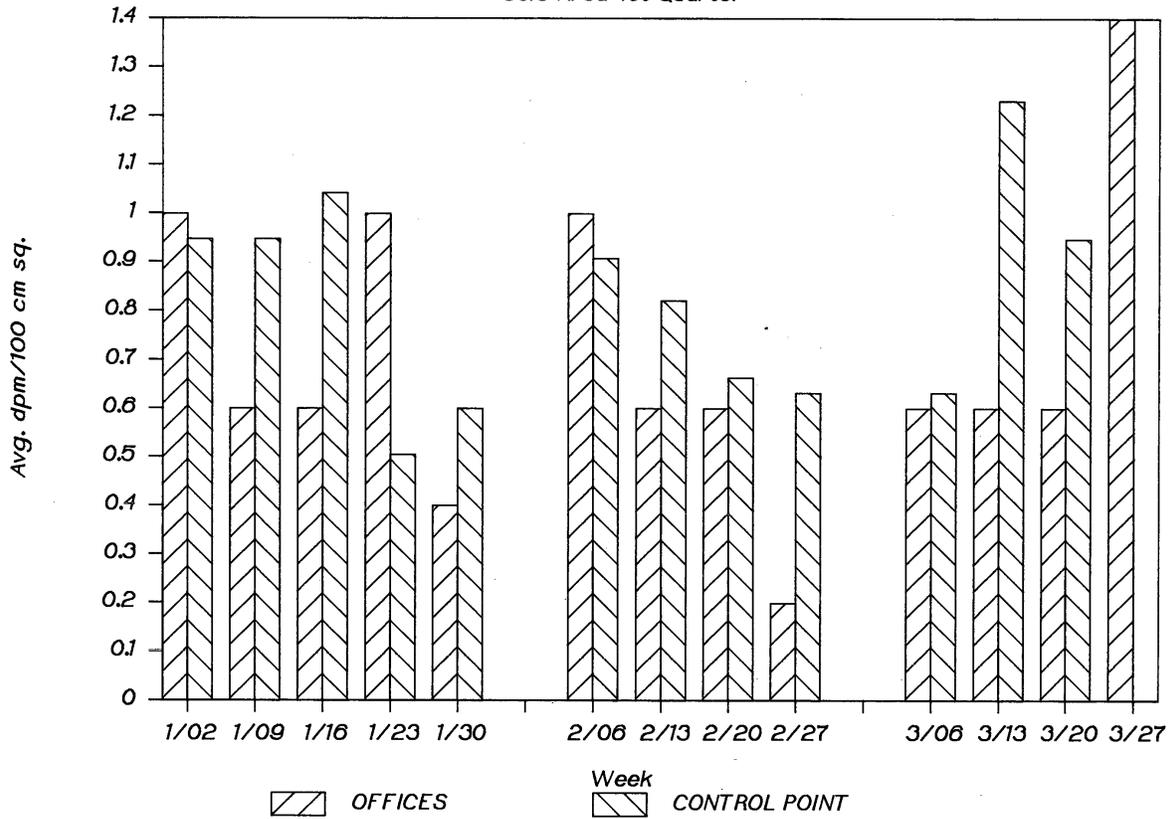
Source: Radiological Surveys, 1989b, PDF p. 5

Note: The solid line represents removable alpha contamination; the dashed line represents the DOE limit for uranium.

**Figure 3: Average Removable Alpha Contamination in the Building 886 Experimental and Material Storage Areas – Q2 1989**

## Building 886

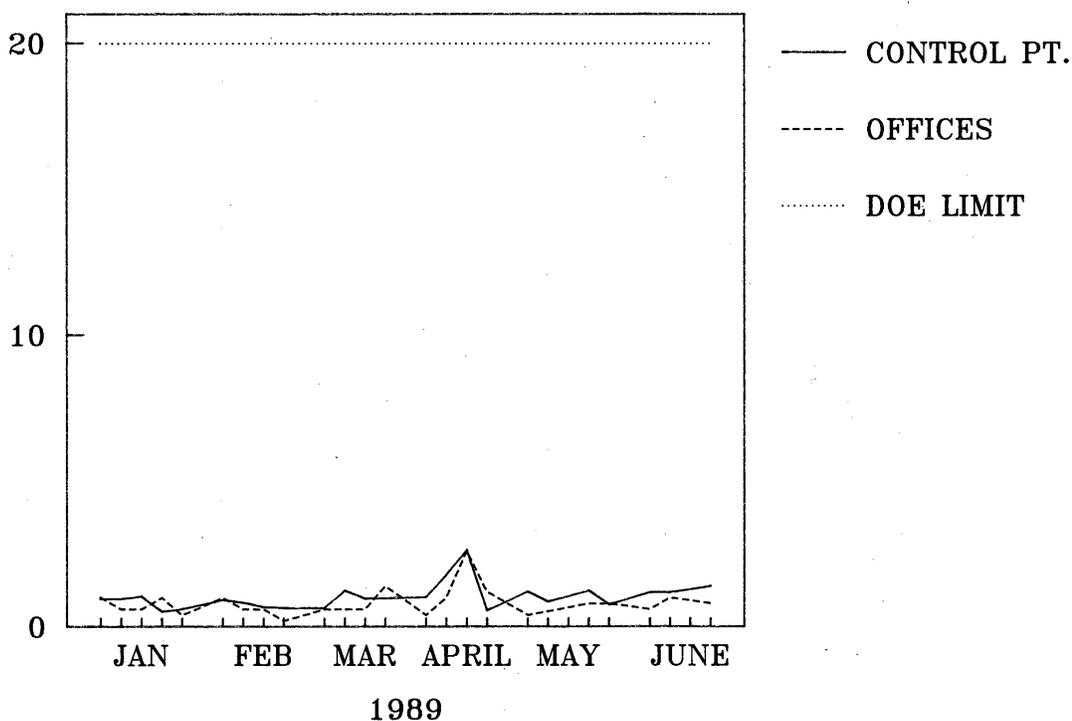
Cold Area-1st Quarter



Source: Radiological Surveys, 1989b, PDF p. 6

**Figure 4: Weekly Averages of Removable Alpha Contamination in the Building 886 Offices and Control Points (Hallways and Conference Rooms) – Q1 1989**

## BUILDING 886 – UNCONTROLLED AREA CONTAMINATION SMEAR SURVEY AVG. DPM/100 CM SQ.



Source: Radiological Surveys, 1989b, PDF p. 4

Figure 5: Average Removable Alpha Contamination in the Building 886 Office Area for January-June 1989

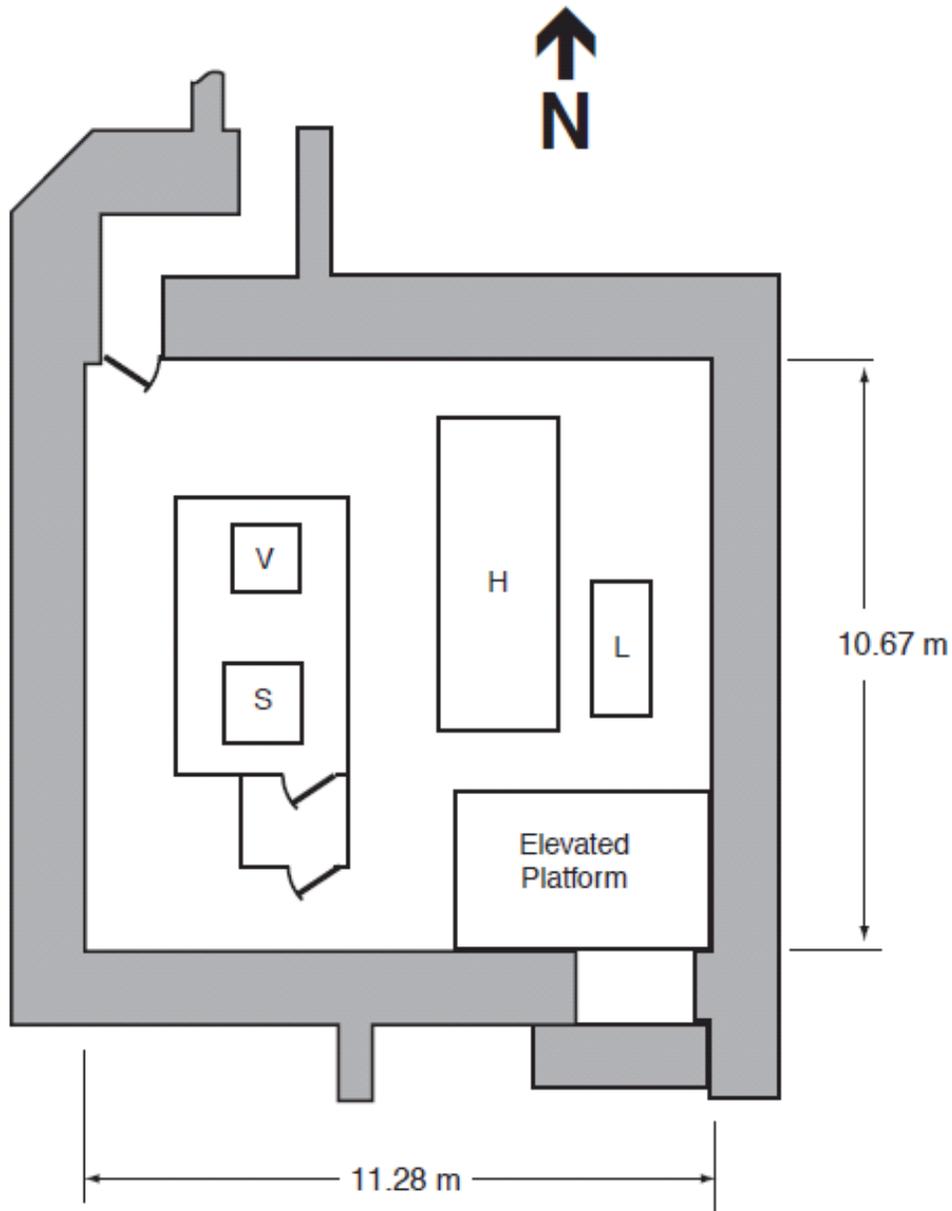
Review of the survey documents indicates that excursions of removable alpha contamination above the Contamination Area limit in the experimental area (Room 101) and the material storage area (Rooms 102, and 103) were usually reduced below 1,000 dpm/100 cm<sup>2</sup> within a few days, as noted below for entries in Table 3.

- The survey sheet showing 3,930 dpm/100 cm<sup>2</sup> for removable contamination in Room 103 (the Mixing Room where uranium solution was housed) on September 10, 1982, for instance, has notation showing that the area was decontaminated to 12 dpm/100 cm<sup>2</sup> on the same day (Radiological Surveys, 1982c, PDF p. 23).
- A survey result of 54,000 dpm/100 cm<sup>2</sup> on February 27, 1984 (the highest found in any of the captured documents), was located in the walk-in hood of Room 101 (Radiological Surveys, 1984c, PDF p. 13). A second survey sheet with the same date shows <20 dpm/100 cm<sup>2</sup> for all

survey locations in Rooms 101, 102 and 103, and bears the hand-written note, “Retake of survey after [name redacted] and [name redacted] cleaned all of Rm 101.”

- The high value of 2,784 dpm/100 cm<sup>2</sup> in 1986 is reported on a survey sheet dated June 17, along with the handwritten notation, “Table Top 3954 d/m/100 cm<sup>2</sup>; Rest of Table 25.8 d/m/100 cm<sup>2</sup>; deconned 6/18/86” (Radiological Surveys, 1986c, PDF p. 8). The survey sheet for June 18, 1986 indicates that all results in the controlled area were ≤ 24 dpm/100 cm<sup>2</sup> (Radiological Surveys, 1986c, PDF p. 9).
- The 1988 high result of 1,683 dpm/100 cm<sup>2</sup> was found just outside the walk-in hood of Room 101 on October 7, 1988 (Radiological Surveys, 1988d, PDF p. 37). A similar elevated result (1,422 dpm/100 cm<sup>2</sup>) was found inside the hood. Contamination at the location outside the hood was reduced to 60 dpm/100 cm<sup>2</sup> a week later, on October 14 (Radiological Surveys, 1988d, PDF p. 38), but the elevated value inside the walk-in hood persisted. It was reduced to 126 dpm/100 cm<sup>2</sup> in the October 21 survey report (Radiological Surveys, 1988d, PDF p. 39).

Prompt decontamination practices were evident during most of the period from 1981-1990; however, elevated contamination levels in the Room 101 walk-in hood were recorded in December 1989 and persisted through April 1990. The persistent contamination is reflected in the maximum-recorded values of 1989 and 1990 shown in Table 3 above, and is detailed in the weekly contamination survey results presented in Table 5 below. The FBI raid of Rocky Flats in June 1989 resulted in the curtailment of plutonium operations. Subsequently, the plant’s resources were redirected toward remediation of the issues resulting from the raid. According to the document, *A Technically Useful History of the Critical Mass Laboratory at Rocky Flats*, “Manpower was not available to decontaminate Room 101 in the late 1980s because Maintenance personnel had all been dedicated to solving the plant’s larger problems” (Rothe, 2005, PDF p. 395). Handwritten notes on the survey sheets sometime indicate that full-face respirator protection was required in Room 101 when contamination levels exceeded the DOE limit (e.g., Radiological Surveys, 1989b, PDF p. 109). Figure 6 shows the floor plan for Room 101, where criticality experiments were performed; the walk-in hood is the enclosure left of center.



Source: Rothe, 2005, PDF p. 120

**Figure 6: Floor Plan of the Assembly Room (Room 101), Showing the Walk-in Hood Containing the Vertical Split Table (V) and Solution Base (S) Experimental Locations**

**Table 5: Room 101 Walk-In Hood Contamination Results, November 1989 – April 1990**

Date	Location 20* (dpm/100 cm <sup>2</sup> )	Location 21* (dpm/100 cm <sup>2</sup> )	Location 22* (dpm/100 cm <sup>2</sup> )	Reference	PDF p.
11/21/1989	27	642	--	Radiological Surveys, 1989b	92
11/30/1989	239	--	--	Radiological Surveys, 1989b	116
12/7/1989	897	12579	18707	Radiological Surveys, 1989b	160
12/14/1989	9	100	90	Radiological Surveys, 1989b	183
12/21/1989	72	51	5235	Radiological Surveys, 1989b	196
1/18/1990	168	693	6990	Radiological Surveys, 1990a	60
1/25/1990	84	672	6963	Radiological Surveys, 1990a	81
2/1/1990	93	1230	12219	Radiological Surveys, 1990a	99
2/8/1990	105	1290	13452	Radiological Surveys, 1990a	117
2/15/1990	255	15099	10839	Radiological Surveys, 1990a	135
2/22/1990	717	12369	9978	Radiological Surveys, 1990a	156
3/1/1990	639	14163	6933	Radiological Surveys, 1990a	171
3/8/1990	616	14193	6987	Radiological Surveys, 1990a	183
3/15/1990	468	12423	7923	Radiological Surveys, 1990a	207
3/22/1990	1035	16212	5403	Radiological Surveys, 1990a	234
3/29/1990	1893	15708	7842	Radiological Surveys, 1990a	252
4/5/1990	2748	12951	8967	Radiological Surveys, 1990a	273
4/19/1990	2556	14967	13683	Radiological Surveys, 1990a	291
4/12/1990	1572	13593	10353	Radiological Surveys, 1990a	300
4/26/1990	2037	14463	11973	Radiological Surveys, 1990a	330

\*Locations 20, 21, and 22 correspond with the entry portal, Solution Base (S), and Vertical Split Table (V), respectively, as shown on the floor plan in Figure 6 above.

Air flow through Room 101 came from outside through the walk-in hood to the exhaust plenum in Building 875, and through HEPA filters before being exhausted to the outside air. The walk-in hood was, therefore, at a lower pressure than Room 101, drawing resuspended contamination to the exhaust plenum and not into the rest of Room 101. This engineered feature reduced exposure to Room 101 personnel and, along with workforce priorities redirected as a result of the FBI raid, probably explains why the hood was not decontaminated in late 1989 and 1990. The last CML criticality experiment concluded in October 1987 (Rothe, 2005, PDF p. 393); no routine work was performed in Room 101 after this date, further reducing the potential for personnel exposure as the result of contamination in the walk-in hood in 1989 and 1990.

### **Additional CML Personnel Identified for 1989**

The 1989 files were different from those in other years because the forms (Radiological Surveys, 1989b) were signed by the RCTs who were making the measurements. Based on the additional research performed for this review, NIOSH has identified no NOCTS claims associated with the seven individuals signing the survey forms.

### **Discovery of Sealed Sr-90 Sources**

The file of survey results for June 1982 contains a copy of a note detailing radiological characteristics of three sealed Sr-90 sources (Radiological Surveys, 1982k, PDF p. 3). The note is reproduced as Figure 7 below. It is the only documentation in hand indicating the presence of Sr-90 at CML. There is no information about why the sources were at CML, how long they might have been there, or their disposition. Integrity of the source encapsulations was confirmed by removable contamination surveys of the sources and the cabinet in which they were found.

1564 886 6/2/84

3 - Sr-90 sulfate sealed 6/56

Each container reads @ surface - Victoreen - 25 mcp/h

All three containers @ 3 ft. - Victoreen - < 0.5

Each container reads @ surface - Ludlum 31 - 200 K

All three containers @ 3 ft - Ludlum 31 - 1 K

Outside reading when placed in cabinet

w/Ludlum 31 750 cpm w/Victoreen < 0.5

No smear count from either container

Cabinet had no smear count

Source: Radiological Surveys, 1982k, PDF p. 3

**Figure 7: Memo on  $\beta$ - $\gamma$  Penetrating and Removable Surface Contamination Measurements Made upon Discovering Three Sealed Sr-90 Sources in the Building 886 Material Storage Area**

## **Conclusions About Surface Contamination at CML**

Surveys for removable alpha contamination at CML were conducted regularly. Excursions above the applicable DOE limits (20 dpm/100 cm<sup>2</sup> in uncontrolled office areas and 1,000 dpm/100 cm<sup>2</sup> in controlled-access experimental areas) were confined to discrete areas and were quickly decontaminated below the limits. Values above the limit in uncontrolled areas were only rarely observed. The largest measured value for removable contamination was 54,000 dpm/100 cm<sup>2</sup>, found at sampling location 21 in the walk-in hood (Radiological Surveys, 1984c, PDF pp. 13, 14). This amount of contamination, if spread uniformly over the entire surface area<sup>7</sup> of the controlled area, would be 1,800 dpm/100 cm<sup>2</sup>; it was decontaminated on the same day it was found. All other values for removable contamination captured by NIOSH correspond to <1,000 dpm/100 cm<sup>2</sup>, if distributed uniformly. All were quickly decontaminated, except for that in the walk-in hood from November 1989 – April 1990, which was contained and ventilated during a time of little or no personnel activity in Room 101.

In light of the above information, the assumption that average removable contamination available for resuspension in the experimental and material storage areas (Rooms 101, 102, and 103) was equal to or less than the Contamination Area limit of 1,000 dpm/100 cm<sup>2</sup> is claimant-favorable.

## **CML WORKPLACE AIR MONITORING**

Bounds on internal dose from MFAP were previously based on airborne concentrations calculated by applying a resuspension factor to surface contamination limits posted for the facility. This approach was based on interview comments from a former Radiation Safety Supervisor, who said that no routine air monitoring was performed at the CML prior to 1990, when this individual was assigned responsibility for Building 886 (Personal Communication, 2015b, PDF pp. 4, 7). NIOSH has since captured formal plant-wide procedures describing a particulate air monitoring program during the period from 1980-1989 for alpha-particle emissions from uranium, plutonium, and americium at sampling locations selected by process knowledge or professional judgment. Additional captured documents indicate that these procedures appear to have been followed and that routine alpha air monitoring was performed at the CML during the period 1980-1989.

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<sup>7</sup> Dimensions of the walk-in hood (Rothe, 2005, PDF p. 129) in Room 101 were 3.0 m × 4.9 m. Sampling location 21 was located in a part of the hood comprising about half this surface area, or 7.35 m<sup>2</sup>. Estimated floor areas for the individual rooms are 120 m<sup>2</sup> for Room 101 (Rothe, 2005, PDF p. 120); 40 m<sup>2</sup> for Room 103 (Rothe, 2005, PDF p. 167); and 60 m<sup>2</sup> for Rooms 102 and 108 (connecting hallway) combined, assuming this area to be about half that of Room 101 (Rothe, 2005, PDF p. 110). The total estimated floor area is the sum of these values, or 220 m<sup>2</sup>. Distributing the contamination at sampling location 21 over the entire surface area would result in an average contamination level reduced by the ratio 7.35/220.

## **Rocky Flats Plant Workplace Air Monitoring Requirements**

Formal procedures describe monitoring requirements and practices for routinely evaluating concentrations of alpha-emitting particulates (U, Pu, and Am) in Rocky Flats workplace air over the period from 1979 through 1990 (Radiation Monitoring Technical Support Programs, 1976, PDF pp. 19-136). The earliest captured procedure (issued November 1979) and its subsequent revisions through December 1990 are listed below:

- *Routine Air Sampling*, HS-RM-4.1, November 13, 1979 (Radiation Monitoring Technical Support Programs, 1976, PDF pp. 19-29)
- *Routine Air Sampling*, RMPM 4.1, August 2, 1982 (Radiation Monitoring Technical Support Programs, 1976, PDF pp. 30-43)
- *Routine Air Sampling*, RMPM 4.1, January 13, 1989, Replaces: August 2, 1982 (Radiation Monitoring Technical Support Programs, 1976, PDF pp. 65-81)
- *Routine Air Sampling*, RMPM 4.1, June 1989, Replaces: January 13, 1982 (Radiation Monitoring Technical Support Programs, 1976, PDF pp. 44-64)
- *Routine Air Sampling*, ROI 4.1, December 18, 1989, Replaces: June 15, 1989 (Radiation Monitoring Technical Support Programs, 1976, PDF pp. 102-118)
- *Routine Air Sampling*, ROI 4.1, Rev. 3, December 20, 1990, Replaces: December 19, 1989 (Radiation Monitoring Technical Support Programs, 1976, PDF pp. 119-136)

Review of the procedures shows consistent requirements for calibrating monthly both Gross Alpha (Continuous) Air Monitors (CAMs) and Selective Alpha Air Monitors (SAAMs). CAM filters were collected and sent for analysis each weekday (Monday – Friday), except holidays, until at least January 1989, when RMPM 4.1 specified that the exchange frequency was to be determined by Operational Health Physics personnel (Radiation Monitoring Technical Support Programs, 1976, PDF p. 67). Subsequent revisions assigned the responsibility for establishing sampling frequencies to Building 123 Count Room personnel (Radiation Monitoring Technical Support Programs, 1976, PDF p. 49), Operational Health Physics (Radiation Monitoring Technical Support Programs, 1976, PDF p. 104), or to Radiological Engineering personnel (Radiation Monitoring Technical Support Programs, 1976, PDF p. 121). When collected, filters were monitored with a hand-held alpha survey instrument. Filters with excessive alpha activity were held for specified periods before analysis to allow decay of the short-lived radon and thoron progeny; a time-dependent Koval factor was applied to the analytical results to account for undecayed progeny (Radiation Monitoring Technical Support Programs, 1976, PDF pp. 22-23, 36-37, 52, 72). Final results were reported as percentages of the radioactive concentration guide (RCG) airborne limit for the material in question, or 70 dpm/m<sup>3</sup> for uranium<sup>8</sup> (Radiation Monitoring Technical Support Programs, 1976, PDF pp. 25, 39, 73). In June 1989, the airborne limit for uranium (in any form) was changed to 44 dpm/m<sup>3</sup>, corresponding to the DOE Order 5480.11 Derived Air Concentration (Radiation Monitoring Technical Support Programs, 1976, PDF pp. 54, 104, 121).

### **Air Sampling Locations**

Air particulate samplers were located in Buildings 886 and 875, as marked on monthly air head calibration sheet maps (Calibrations, 1987; Calibrations, 1988; Calibrations, 1989a; Calibrations, 1989b); Figure 8 shows an example. The locations appear to be unchanged during the period from 1980-1989. Building 875 housed the air exhaust plenums from the Building 886 experimental (Room 101) and material storage (Rooms 102 and 103) areas. The air head samplers were numbered RR-1 through RR-11 and fixed in the locations listed below:

#### **Building 886**

- RR-1 through RR-4, Room 103 (material storage)
- RR-5, Room 102 (material storage)
- RR-6 and RR-7, Room 101 (experimental area)

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<sup>8</sup> The RCG used by Rocky Flats in its air-sampling program corresponds with the Maximum Allowable (air) Concentration (MAC) used by AEC contractors in referring to the AEC's "preferred level" of 50 µg/m<sup>3</sup> for all uranium compounds on the basis of chemical toxicity. The level was also stated as 70 dpm/m<sup>3</sup> for natural uranium. (ORAUT-OTIB-0004, 2006, PDF p. 8)

## **Building 875**

- RR-8 and RR-9, outside the plenum
- RR-10, inside the first plenum
- RR-11, inside the tunnel from Building 886

SAAM samplers also were located in Rooms 101-103 of Building 886 and in the second plenum of Building 775. The detectors on these samplers were equipped with high-voltage discriminators that registered only alpha energies above a certain threshold, providing a means to discriminate against lower-energy alpha emitters that would otherwise give false alarms. The filter media from SAAMs were normally discarded without analysis unless the instrument had alarmed or the filter failed a hand-held radiation monitor check (Radiation Monitoring Technical Support Programs, 1976, PDF pp. 26, 35, 51, 70, 88, 105-106, 123).

Other air sampling results from 1981-1988 for samples labeled “875-A”, “875-B” and “875-C” have also been captured but their locations in Building 875 have not been identified (Air Samples, 1981a, 1981b; 1982a-1982i; 1983a, 1983b; 1984a-1984d; 1985a-1985e; 1986a-1986m; 1987a; 1988a-1988g).

CML personnel did not routinely access Building 875, and consideration of air monitoring results for this building is limited to those in the “RR” series, with known locations and direct correlation with daily samples from Building 886.

AIRHEAD & SAAM CALIBRATION

BUILDING NUMBER 886-875  
 ROOM OR MODULE 101-102-103  
 DATE 7-29-87  
 TAKEN BY [Signature]



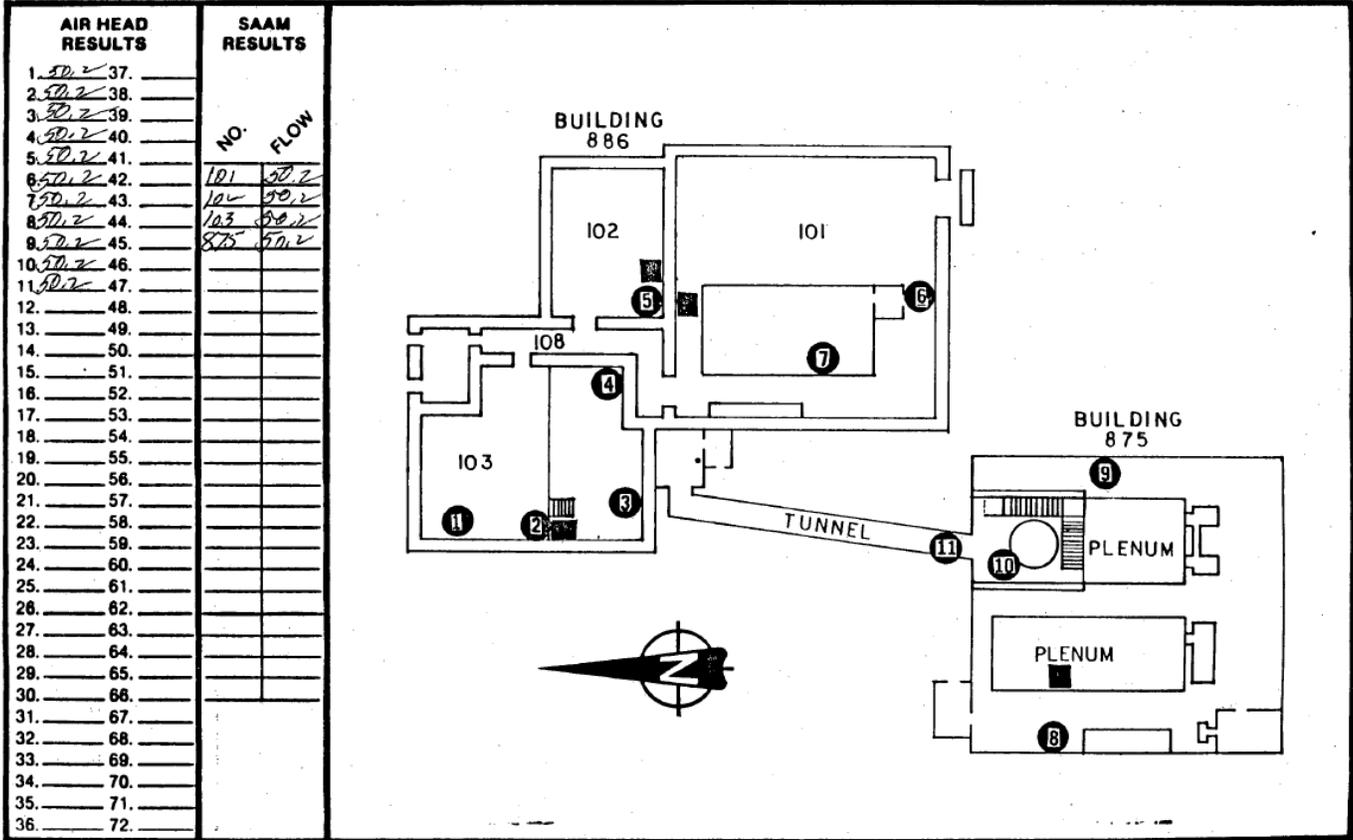
Rockwell International

ROCKY FLATS PLANT  
 886-20M RADIATION MONITORING

MASS FLOW METER

INSTRUMENT USED KURZ METER  
 RF NO. 78212  
 CALIBRATION DUE DATE 10-19-87  
 AIR ROUTE NO. RR

SAAM AIRHEAD



RMF 12 (3/84)

Source: Calibrations, 1987, PDF p. 6

Figure 8: Air Sampler Locations in the Building 886 Cluster

Air Sampling Results

NIOSH has captured air sample results in 41 of 120 months during the period 1980-1989. Results from samplers RR-1 through RR-7 (Building 886) and RR-8 through RR-11 (Building 875) are available for the period December 1980 to June 1989, although all but two of the results fall in the period from May 1983 to November 1988. The data, summarized in Table 6, indicate that procedural requirements for daily air monitoring appear to have been met at the CML. The number of analytical results per month for a particular sampler location is expected to be between 19 and 22, depending on the number of work days. In the captured data, daily sample collection is demonstrated with certainty (results available for 19 or more days) in 15 of the 41 months (37% of the time for which records are available, but only 1% of the period from

1980-1989). Sample collection on at least alternate days, on average (results available for 10-18 days), is demonstrated with certainty during an additional 10 months. The availability of air sampling data does not seem to correlate with known spills or criticality experiments. Only one sampling day's results were captured during operations prior to 1983, but 207 sampling days correspond with three operational periods after 1983. Four contamination incidents occurred from 1980-1989, but air monitoring results were only captured for the time period in which two incidents occurred closely together in 1987. Experiment periods, contamination incident dates, and associated records of airborne contamination results are summarized in Table 7.

**Table 6: Air Sample Results Summary for the CML Building 886 Nuclear Materials Storage and Experimental Areas**

Year	Month	References	Sample Days*	No. of Days 10%-99% RCG**	No. of Days ≥100% RCG**
1980	Dec	Air Samples, 1980	1	--	--
1983	May	Air Samples, 1983c	2	--	--
1983	Sep	Air Samples, 1983d	1	--	--
1983	Oct	Air Samples, 1983e	17	2	--
1984	Jul	Air Samples, 1984e	2	--	--
1984	Aug	Air Samples, 1984f	22	1	1
1984	Sep	Air Samples, 1984g	19	2	--
1984	Oct	Air Samples, 1984h	20	6	--
1984	Nov	Air Samples, 1984i	22	8	3***
1984	Dec	Air Samples, 1984j	15	5	--
1985	Jan	Air Samples, 1985f	19	4	--
1985	Feb	Air Samples, 1985g	19	8	2
1985	Mar	Air Samples, 1985h	22	1	--
1985	Apr	Air Samples, 1985i	21	--	--
1985	May	Air Samples, 1985j	22	3	--
1985	Jun	Air Samples, 1985k	22	--	--
1985	Jul	Air Samples, 1985l	23	2	--
1985	Aug	Air Samples, 1985m	20	2	--
1985	Sep	Air Samples, 1985n	15	--	--
1985	Oct	Air Samples, 1985o	9	--	--
1985	Nov	Air Samples, 1985p	19	1	--
1985	Dec	Air Samples, 1985q	15	--	--
1986	Jan	Air Samples, 1986n	19	--	--

White Paper: Reassessment of Internal Dose from Sources at the RFP CML

Year	Month	References	Sample Days*	No. of Days 10%-99% RCG**	No. of Days ≥100% RCG**
1986	Feb	Air Samples, 1986o	1	--	--
1986	Jun	Air Samples, 1986p	4	--	--
1986	Jul	Air Samples, 1986q	14	--	--
1986	Nov	Air Samples, 1986r	3	--	--
1986	Dec	Air Samples, 1986s	9	--	--
1987	Jan	Air Samples, 1987b	15	1	--
1987	Feb	Air Samples, 1987c	22	--	--
1987	Mar	Air Samples, 1987d	8	--	--
1987	May	Air Samples, 1987e	6	--	--
1987	Jul	Air Samples, 1987f	3	--	--
1987	Aug	Air Samples, 1987g	13	--	--
1987	Sep	Air Samples, 1987h	7	--	--
1987	Dec	Air Samples, 1987i	11	--	--
1988	Jan	Air Samples, 1988h	18	--	--
1988	Feb	Air Samples, 1988i	15	--	--
1988	Apr	Air Samples, 1988j	5	--	--
1988	Nov	Air Samples, 1988k; 1988l	5	--	--
1989	Jun	Air Samples, 1989	1	--	--
<b>TOTAL</b>	N/A	N/A	<b>526</b>	<b>46</b>	<b>6</b>

NOTE: Documents captured by NIOSH include results from only portions of years 1980 and 1983-1989, as shown in the table.

\* The number of days in the specified month for which captured air-monitoring data indicate that samples were collected. Procedures required that sample filters be changed and analyzed daily, except over weekends and holidays.

\*\* The number of days for which the specified fraction of the uranium Radiological Control Guide (RCG) of 70 dpm/m<sup>3</sup> alpha was met or exceeded.

\*\*\* Notations on the air results indicate that respirators were worn.

**Table 7: Experiment Periods, Contamination Incident Dates, and Associated Records of Airborne Contamination Results**

Experimental Campaigns*	Contamination Incident Dates**	Airborne Monitoring Days Captured
None	11/25/1980	0
02/1978 – 09/1981	None	1
Summer 1982 – 12/20/1982	None	0
05/1983 – 09/1984	None	64
None	07/07/1984 – 07/20/1984	0
07/1985 – 08/1986	None	139
None	02/14/ 1987 & 03/13/1987	30
04/1986 – 10/1987	None	104

\*Rothe, 2005, PDF pp. 389-393

\*\*Rothe, 2005, PDF pp. 486-487, 498, 500-501

Building 886/875 results appear in the same reports as those for Building 865, which housed the Metal Research and Development Laboratory<sup>9</sup> and which processed non-plutonium metals, including depleted uranium (DOE, 2011 , PDF p. 2). Reports are reviewed and initialed, and instructions on the report sheet specify that a copy is to be sent “TO RADIATION MONT 881” (to Radiation Monitoring, Building 881).

Individual sample results are occasionally lined out in the reports, with or without explanation. There are multiple instances when results are lined out with the notation “W/C” or “Wrong Color.” The meaning of this notation is not clear, and these samples are not included in evaluating the results.

A total of 526 days of results were evaluated against the Radiological Control Guide (RCG) of 70 dpm/m<sup>3</sup>. One or more results from the seven samplers in Building 886 exceeded the RCG on six days; results between 10% and 100% of the RCG were recorded on 46 additional days. All results were below 10% of the RCG for the remaining 473 days. Respirators were worn on three of the six days in which the RCG was exceeded (Air Samples, 1984e, PDF pp. 33, 36, 52). Results on the other three days in which the RCG was exceeded were: 522.02% RCG (Air Samples, 1984f, PDF p. 5); 111.72% RCG (Air Samples, 1985f, PDF p. 37); and 117.76% RCG (Air Samples, 1985f, PDF p. 41).

Results from Building 875, which housed the effluent air exhaust plenums from the Building 886 experiment room, exceeded the RCG on only one occasion<sup>10</sup> during the same 526 days; results between 10% and 100% of the RCG were recorded on two days.

<sup>9</sup> Each day’s results for Building 886 are labeled RR-1 through RR-7; those for Building 875 are labeled RR-8 through RR-11; Building 865 results begin with a “UU-” designator.

<sup>10</sup> The sample designation was “RR-12” for the single result exceeding the RCG, without explanation of the location or purpose for the sample. A location in Building 875 is assumed.

## Conclusions About Workplace Air Monitoring at the CML

NIOSH concludes that a robust and well-defined workplace air-monitoring program for alpha-emitting radioisotopes was required by Rocky Flats plant procedures on continuing basis during the period 1980-1989. In all air-monitoring records captured by NIOSH, air particulate samples from the CML, Building 886, and its air exhaust filtration plenums in Building 875, were routinely analyzed and reported along with those from the Metal Research and Development Laboratory, Building 865. Sampling results were evaluated for uranium alpha emissions and reported as a percentage of the radioactive concentration guide (RCG) airborne limit for uranium, 70 dpm/m<sup>3</sup>. The analytical reports were reviewed and initialed, and were to be sent to Radiation Monitoring in Building 881. Excursions in excess of the RCG were uncommon, occurring in Building 886 on six of 526 days for which monitoring results were captured, and in Building 875 only once.

A bounding value for activity concentrations in breathing air can be calculated as the weighted average  $\bar{C}$  of air results using recorded values for three results in excess of the RCG with no indication that respirators were worn, and by making the claimant-favorable assumptions that results were 70 dpm/m<sup>3</sup> for 46 results between 10% and 100% of RCG and 7 dpm/m<sup>3</sup> for the remaining 477 recorded samples, as shown in Equation 3 below:

$$\begin{aligned}\bar{C} &= \frac{[5.22 + 1.12 + 1.18 + (46)(1.0) + (477)(0.1)] \text{ RCG days}}{526 \text{ days}} = 0.1924 \text{ RCG} \\ &= 13.5 \text{ dpm/m}^3.\end{aligned}$$

Results for the 526 days of monitoring data captured for the period 1980-1989 are assumed to be representative of uncaptured data for that period for the following reasons:

- The same plant health physics procedures requiring air monitoring were in effect continuously (with revisions) for the entire period;
- NIOSH assumes that daily samples were collected and analyzed in accordance with plant procedures over the entire period; and
- The results are probably similar because CML operations were similar over the period.

These data were available for routine review by health physics personnel, who also had access to information about operations for making personnel monitoring decisions. It is therefore unlikely that an unrecorded intake of alpha contaminated airborne particulates occurred during this period because of a lack of relevant air monitoring data.

## **ASSESSMENT OF UNMONITORED RADIATION DOSE AT THE CML**

Radiation dose from intake of MFAP at CML could have occurred during clean up of numerous fuel spills, predominantly from enriched uranyl nitrate solution (UNH), or from inhalation of dried, resuspended contamination deposited on surfaces as the result of these spills (Rothe, 2005, PDF pp. 447-449, 452-462, 464-473, 486-487, 498, 500-501). Rocky Flats workers (including those assigned to the CML) with the potential for receiving intakes of plutonium, americium, or uranium were monitored by periodic urinalysis and body counts (NIOSH, 2006, PDF p. 30). However, NIOSH has found no indication that confirmatory bioassays were performed for employees involved in clean up of any of the accidental UNH spills. Fission and activation products, which decay primarily by beta/gamma emission, are not likely in any case to have been detected by bioassay intended to detect alpha particles emitted by uranium or transuranic radionuclides.

Maximum MFAP internal doses to CML workers were estimated by modeling a representative UNH experiment and calculating the MFAP inventory based on the historical record of CML experiments with UNH, and on the average thermal power and duration of CML UNH criticality experiments reported by CML researchers. Intakes of resuspended UNH contamination with the same MFAP-to-uranium atom ratio as the fuel were estimated from the weighted average of air monitoring results in the experimental and materials storage areas of the CML. Doses were calculated by applying ICRP 68 (ICRP, 1995) dose conversion factors for three solubility categories of dosimetrically significant radionuclides, using the method described in ORAUT-OTIB-0054 (2015).

### **Calculation of Fission and Activation Product Content of UNH Solution**

A series of experiments was performed at the CML during the mid-1970s to determine the critical height of UNH in suspended, cylindrical tanks. Experiments were performed using different uranium concentrations, different tanks, and with and without neutron reflectors. The same uranium enrichment was used in all experiments; only the concentration was varied.

NIOSH chose one of the unreflected suspended tank experiments from the mid-1970s to represent UNH experiments performed over the CML's history. There were ten unreflected suspended tank experiments in all. Two of them used a stainless-steel tank and the remainder used an aluminum tank.

NIOSH selected one of the experiments that used the stainless-steel tank so that the calculated fission and activation product content would include iron activation products. The tank had an inside diameter of 27.92 cm and an inside height of 41.6 cm. For the selected experiment, the tank was filled with UNH with a uranium enrichment of 93.172 weight percent U-235 at a concentration of 145.68 g/L. The other unreflected stainless steel tank experiment used UNH with the same enrichment, but at a concentration of 346.73 g/L. The lower concentration was selected because it represents the middle of the range of concentrations used across the ten unreflected tank experiments. The critical solution height for the selected experiment was found

to be 31.20 cm. The selected experiment is documented as Case Number 1 in “Minimally Reflected Cylinders of Highly Enriched Solutions of Uranyl Nitrate,” HEU-SOL-THERM-001, from the *International Handbook of Evaluated Criticality Safety Benchmark Experiments* (Palmer, 2004).

The fission and activation product composition for the selected experiment was calculated using SCALE. SCALE is a modular system of computer codes for nuclear- and radiological engineering-related analyses from the Oak Ridge National Laboratory. SCALE’s TRITON module was used to develop a case-specific cross-section library for the selected suspended tank experiment. The case-specific library was then used by the ORIGEN-S code to determine the time-dependent fission and activation product content of the UNH and the stainless-steel tank over the CML’s history.

The TRITON module performs neutron transport and depletion calculations for irradiated nuclear fuels. Neutron transport is performed using either discrete ordinates methods (via the NEWT code) or via Monte Carlo calculations (using the KENO –V or KENO-VI codes). TRITON’s capabilities include creating case-specific cross-section library files that can be used by other elements of the SCALE code system, notably the ORIGEN-S code. Among the numerous capabilities of ORIGEN-S is calculation of the inventory of activation products, actinides, and fission products in a composition as a function of time and burnup (as applicable).

TRITON was used to model the stainless-steel tank and UNH solution geometry of the selected suspended tank experiment as a system of cylinders representing the tank bottom, tank walls, and the UNH volume. The tank and solution heights were truncated to the critical solution height of 31.20 cm. Neutron transport was performed via the KENO-VI Monte Carlo code.

The case-specific cross-section library created by TRITON was subsequently used in an ORIGEN-S calculation to determine the fission and activation product content of the solution, and the tank volume at the end of the tenth campaign and thereafter. This reflects a modeling assumption that the same tank and solution were used for all 778 experiments involving HEU solutions performed at the CML over its operating history. The composition used for the ORIGEN-S calculations was a homogenized mixture of the HEU solution, the 304 stainless steel tank walls and bottom, and the associated impurities. The composition was irradiated and decayed in the ORIGEN-S case using the history provided in Table 7 above. The first campaign was represented by a 38-hour irradiation followed by 32 days of decay, and so on, through the final 19-hour irradiation representing the tenth campaign.

Each experiment was assumed to have lasted for 70.5 minutes and to have produced an average thermal power<sup>11</sup> of 6.7 mW (the average power and duration reported to ERDA in 1977) (Tuck, 1977, PDF p. 6).<sup>12</sup>

The ORIGEN-S calculation produced time-dependent fission and activation product inventories, with radioactive decay between each of the ten campaigns, for the HEU solution defined in the selected benchmark case, and using the timeline for all uranium solution experiments conducted over the CML's history. Radioactive decay corrections were also applied at the end of the final experiment, using a decay period of 180 days from those specified in ORAUT-OTIB 0054 (2015).

### **Calculation of Inhalation Intakes and Committed Organ Doses**

Solution spills resulting in surface (floor) contamination occurred throughout CML's operating history, as shown in Table 8. Most spills occurred in the late 1960s, but there were also several spills during the 1980s. NIOSH performed organ dose calculations after applying a decay interval of only 180 days to the ending fission and activation product inventory for the HEU solution. This is favorable to the claimant, given that the calculation of the fission and activation product inventory represents the entire operating history of the facility (i.e., includes the period after the last major spill).

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<sup>11</sup> The power in this context is an average value representing a given number of fissions over a given interval of time because nuclear criticality experiments are not steady-state and are not typically considered in terms of a power level. An average power was used for modeling purposes to account for any change in the composition of the fissile solution (i.e., for depletion effects) and to compute the ingrowth of fission products as the solution was used. In reality, most of the fissions would have occurred near the end of a given experiment when the system was alternately placed in slightly subcritical and slightly supercritical states.

<sup>12</sup> The 6.7 mW value is used, rather than the more precise 3.6 mW for 70.5 minutes estimated by CML staff after the communication with ERDA. There is no indication why 6.7 mW was reported instead of a lower value of 5.4 mW initially calculated by CML staff before refining their estimate. The higher value is adopted as more favorable for claimants.

**Table 8: High-enriched Uranium Solution Spills Over the CML’s History**

Date	Volume (L)	Uranium Mass (kg)	Contaminated Area	PDF Page(s)
7/2/1965	0.5	0.225	22 m <sup>2</sup> (Floor)	447
7/14/1965	---	---	10 m <sup>2</sup> (Floor)	448-449
7/22/1965	---	---	Small amount (Floor)	449
11/30/1967	---	9	(Inside large duct, filter housing, vent line)*	452-462
2/16/1968	---	1.14**	(Floor and cable trenches)	464-467
5/11/1968	0.06	---	(Workman’s knee)	467
5/9/1969	150. 1	16.1	20 m <sup>2</sup> (Mixing Room floor)***	467-473
11/25/1980	7	2.66	(Assembly Room hood)	486-487
7/7-20/1984	---	---	(Walk-in hood)****	498
2/14/1987	---	---	(Personnel, facility and fixtures)*****	500-501
3/13/1987	---	---	(Personnel, facility and fixtures)*****	501

Source: Rothe, 2005

\*Although contamination was confined to ducts and a filter housing, clean up of this incident resulted in a blowback of dried salts, resulting in facial contamination of a staff member.

\*\*Two conflicting accounts refer to this value as either the solution mass or uranium mass.

\*\*\*Standing HEU solution covering the floor was cleaned up by a staff member using a critically safe vacuum and wearing plastic booties and a half-face respirator.

\*\*\*\*Potentially-contaminated workmen repairing a leaking flange were required to evacuate when a criticality alarm was triggered by an electrician.

\*\*\*\*\*Two essentially identical events resulted in personnel, fixture, and facility contamination by resuspended high-enriched uranyl nitrate salts accidentally knocked from the surface of a large reactivity shim.

ORAUT-OTIB-0054 (2015) describes a method used to reduce the large number of fission and activation product isotopes in an ORIGEN-S result to a set of 36 dosimetrically significant nuclides. The same 36 nuclides were considered in the inhalation intake and committed organ dose calculations for the CML. Intakes were computed using the 180-day activity values for the HEU solution shown in Table 9, corrected for the average airborne concentration level, and assuming an intake period of 4,000 hours (two working years) at a breathing rate of 1.2 m<sup>3</sup>/h. The two-year intake period was selected to be consistent with ORAUT-OTIB-0054 (2015), which provides a basis for assigning internal dose from unknown inhalation of fission and activation product mixtures. Radioactive decay of the isotopic mixture over the two-year intake period was not considered, which is favorable to the claimant.

**Table 9: Accumulated Activities of Dosimetrically Significant Fission and Activation Products in UNH Fuel at the CML**

Isotope	Activity (Bq)
Mn-54	1.800E+02
Fe-55	1.664E+03
Co-58	1.359E+02
Co-60	3.077E+00
Sr-89	1.287E+04
Sr-90	2.506E+04
Y-90	2.507E+04
Y-91	1.984E+04
Zr-95	2.734E+04
Nb-95	5.025E+04
Mo-99	5.206E-14
Ru-103	4.732E+03
Ru-106	3.889E+03
Cd-113m	1.658E+00
Cd-115m	9.955E-01
Sb-125	3.236E+02
Te-129m	1.051E+02
Te-132	3.937E-11
I-131	8.936E-02
I-132	4.055E-11
Cs-134	7.004E-02
Cs-136	4.318E-02
Cs-137	2.567E+04
Ba-140	3.719E+01
La-140	4.281E+01
Ce-141	5.509E+03
Ce-144	5.417E+04
Pr-143	6.690E+01
Pr-144	5.417E+04
Nd-147	3.204E+00
Pm-147	2.171E+04
Pm-148m	0.000E+00
Sm-151	6.867E+02

Isotope	Activity (Bq)
Eu-154	1.691E-03
Eu-155	3.225E+02
Ta-182	0.000E+00

NOTE: Inventory in the UNH fuel was accumulated without radioactive decay until the final experiment, after which the activities of individual isotopes were decay-corrected for 180 days to yield these values.

Evaluation of air monitoring data in the earlier section, *CML Workplace Air Monitoring*, determined that a weighted-average concentration of 13.5 dpm/m<sup>3</sup> for airborne alpha activity was favorable to the claimant. Inhalation intakes were computed by assuming that the airborne alpha activity consisted entirely of HEU having a specific activity of 70 μCi/g. Therefore, the airborne mass concentration was 8.7 × 10<sup>-8</sup> g/m<sup>3</sup> and the total intake over the 4,000-hour period was 4.2×10<sup>-4</sup> g.

The UNH fuel, as modeled, contained 2,782.8 g HEU. The ratio of total inhalation intake over the two-year intake period and HEU solution mass gives an intake fraction  $f = 1.5 \times 10^{-7}$ . The inhalation intake  $I$  for the 36 dosimetrically-significant nuclides for the two-year period was then determined as the product of the ORIGEN-S result for each nuclide after 180 days of decay (activity  $A$ ) and the intake fraction, i.e.,  $I = fA$ .

Committed organ doses were computed (see Equation 4 below) as the sum of products of the 180-day intakes, computed for each of the 36 dosimetrically significant nuclides, and their corresponding inhalation dose conversion factors (DCF),  $e_{inh(50)}$ , from ICRP 68 (ICRP, 1995).

$$H(50) = \sum_{i=1}^{36} I_i e_{inh(50)}_i = f \sum_{i=1}^{36} A_i e_{inh(50)}_i$$

The DCFs were partitioned into three solubility categories to account for different absorption types, using the method described in ORAUT-OTIB-0054 (2015). Three committed dose values were therefore computed for each of the 25 individual organs defined in ICRP 68 (ICRP, 1995), corresponding to soluble, moderately soluble, and insoluble materials. Table 10 shows the maximum committed organ dose values for the three solubility categories.

**Table 10: Maximum Committed Organ Doses from Inhalation of Airborne Fission and Activation Products at CML**

Solubility	$H(50)$ (Sv)	Tissue
Soluble (Type F)	$2.5 \times 10^{-9}$	Bone surface
Moderately Soluble (Type M)	$2.3 \times 10^{-9}$	Bone surface
Insoluble (Type S)	$2.4 \times 10^{-9}$	Lung

These values are greatly reduced over previously calculated committed doses of  $3.7 \times 10^{-7}$  Sv (soluble, bone surface),  $4.0 \times 10^{-7}$  Sv (moderately soluble, lung), and  $6.1 \times 10^{-7}$  Sv (insoluble, lung). Earlier committed doses were calculated using airborne concentrations derived by applying a resuspension factor to the claimant-favorable DOE limit on removable surface contamination. The orders-of-magnitude difference in the two sets of dose values is due to a prior miscalculation in converting from  $100 \text{ cm}^2$  to  $\text{m}^2$ . The corrected calculation uses a resuspension factor of  $1.5 \times 10^{-4} \text{ m}^{-1}$  and 2,000 dpm/100  $\text{cm}^2$  removable alpha. This calculation is applied over the entire 220  $\text{m}^2$  footprint of the CML experimental and material storage areas, giving an estimated air concentration of 30 dpm/ $\text{m}^3$ . The claimant-favorable 13.5 dpm/ $\text{m}^3$  weighted average alpha air concentration from routine monitoring results, used in calculations for this analysis, further reduces the dose estimate from that obtained previously.

Previous estimates of fission and activation product inventories also assumed a thermal power of 10 mW for 60 minutes, compared with the lesser (but still claimant-favorable) value of 6.7 mW for 70.5 minutes used in this analysis to describe conditions for the typical CML criticality experiment.

## SUMMARY AND CONCLUSIONS

Re-evaluation of unmonitored personnel dose using recently captured air monitoring data and reactor performance estimates for the Rocky Flats Plant Critical Mass Laboratory has resulted in estimates on the order of a few nanosieverts for maximum organ doses due to inhalation of resuspended contamination containing mixed fission and activation products. These estimates are over two orders of magnitude lower than previous estimates. The greatest contributor to the large reduction in estimated doses is a correction in the calculation of a conversion factor. Lesser contributors to the reduction are lower estimates of reactor power in a typical criticality experiment at CML, and a lower value for respirable alpha air concentrations based on routine air monitoring results.

NIOSH computer-modeled criticality experiments using estimates of typical reactor power and corresponding neutron flux documented by CML research staff and alpha air concentrations from routine monitoring results. Based on its modeling, NIOSH concludes that no significant personnel dose to Rocky Flats workers or contractors resulted from the generation of fission or activation products in the uranyl nitrate fuel or resuspended contamination from fuel spills as a result of criticality experiments conducted at CML over its lifetime.

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