Assessment of Sealed Radioactive Sources, and Fission and Activation Products as Radiological Exposure Sources in the Rocky Flats Plant Critical Mass Laboratory (Building 886 Cluster)

White Paper
Rev. 0

National Institute for Occupational Safety and Health

June 9, 2015

James S. Bogard, Daniel Stempfley, Bob Burns
Oak Ridge Associated Universities Team

-

Reviewed by LaVon Rutherford and James W. Neton
Division of Compensation Analysis and Support

PURPOSE

This white paper describes the Critical Mass Laboratory (CML) at the Rocky Flats Plant and the programs and experiments conducted there. It discusses the radioactive materials present and their amounts and assesses these materials as radiological sources.

Radioactive materials at the CML included the nuclear fuels and sealed radioactive sources used in criticality experiments. Fission and activation products generated in the fuels, building materials, and fixtures as a result of the nuclear criticality experiments conducted there are an additional source of radiological exposure. Several liquid fuel spills, especially early in the facility's history, resulted in residual contamination in the rooms where experiments were performed and in which the nuclear fuels were stored. The ability to estimate contributions to personnel dose from these sources is assessed from detailed accounts of the facility's history, results from surveys and assessments conducted in association with facility decommissioning and demolition, and computer-modeled predictions of specific radioisotopes and quantities generated as fission and activation products.

HISTORY OF THE CRITICAL MASS LABORATORY

Construction of Building 886 (originally called Building 86), the Critical Mass Laboratory (CML), was completed in 1963. The Building 886 complex ultimately consisted of Building 886 (Critical Mass Laboratory), Building 880 (storage facility), Building 875 (filter plenum facility), and an underground tunnel containing ventilation ducts that connected Building 886 to Building 875 (SRDB 104452, PDF p. 3). Highly enriched uranium was introduced into Building 886 in the summer of 1965; the first criticality experiments were performed in September 1965. The building was used for experiments on enriched-uranium metal and solution, plutonium metal, low-enriched uranium oxide, and for several special applications.

Experiments after 1983 were conducted primarily with uranyl nitrate solutions and did not involve solid materials (SRDB 104452, PDF p. 3). The RFP Nuclear Safety Group conducted 1,600 (SRDB 21358, PDF p. 69) to 1,700 (SRDB 104452, PDF p. 3) critical mass experiments using uranium and plutonium in solutions (900 tests), compacted powder (300), and metallic forms (500) (SRDB 104452, PDF p. 3). Testing programs at the CML were temporarily suspended in 1987 for routine equipment modifications, and for contamination control and ventilation repairs; however, the modifications and repairs had not been completed in 1989 when all operations at RFP were curtailed as a result of an FBI raid. Criticality research at the CML never resumed after the raid (SRDB 21358, PDF p. 69). All the fissile materials were removed by 1997 for reprocessing (SRDB 136853, PDF p. 4). Internal fixtures were removed by 2000,

with the exception of the Room 101 walk-in hood and the heavily contaminated annular tank on its elevated platform in a corner of the Assembly Room (SRDB 21358, PDF p. 411). These were eventually also removed, and the CML building was demolished in April 2002 (SRDB 21358, PDF p. 411).

RADIOACTIVE MATERIALS USED IN THE CML

A variety of sealed radioactive sources and nuclear fuel (uranium and plutonium) was housed at the CML. The types and amounts of fuel varied with time, but ranged up to the amounts shown in Table 1.

Table 1: Types and Amounts of Fissile Fuels Housed at the Critical Mass Laboratory

Type of Fissile Fuel	Amount (kg)	SRDB 21358, PDF page(s)
High-enriched uranium hemishells and rods	280	162, 289-295
Low-enriched (4.46% U-235) uranium oxide compacted into briquettes and tightly packaged in cubical aluminum cans	2,100	162, 289, 311-314
Plutonium ingots	800	289
Highly-enriched (93% U-235) uranium (as uranyl nitrate solution)	570	289, 295-303
Plutonium in the form of metal hemishells	Unknown	303-305
Machined plutonium metal cylinders sealed in double containers	375	162, 305-310

Sealed sources of ^{210}Po-Be, Co-60, and Cf-252 were used in the CML for instrument testing and for use in experiments (SRDB 21358, PDF p. 162). The ^{210}Po-Be and Cf-252 sources were used in criticality experiments involving uranium to provide initiating neutrons that allowed a smooth transition toward criticality, giving better experimental results and preventing the inadvertent creation of a supercritical geometry. A ^{210}Po-Be source was used before 1970, after which Cf-252 was used (SRDB 21358, PDF pp. 315-316). Gamma photons from Co-60 were used in testing a gamma-sensitive radiation detector mounted on the north wall of the Assembly Room that was used to monitor the gamma-ray field during critical experiments (SRDB 21358, PDF p. 316).

Sealed sources in use at the facility in 1989 are shown in Table 2 (SRDB 21358, PDF p. 355). The relatively short half-lives of Co-60 and Cf-252 made it necessary to replace these sources periodically (SRDB 21358, PDF p. 316). The listed sources and their activities are taken as typical for the CML. Approximate initial exposure rates[1] from the Co-60 sources (from lowest activity to highest on the specified date) are 13.6 mR m^2 h^{-1} and 18.5 mR m^2 h^{-1}. Initial dose equivalent rates (neutron + photon) from the Cf-252 sources[2] are 1.0 mrem m^2 h^{-1}, 26.4 mrem m^2 h^{-1}, 31.3 mrem m^2 h^{-1}, and 32.8 mrem m^2 h^{-1}.

Table 2: Sealed Radioactive Sources in Use at the Critical Mass Laboratory in July 1989

Registry No.	Manufacturer's No.	Isotope	Radiation	Source Strength		
				Strength at (Date)	n/sec (10^7)	Half-life (years)
S-082	---	Co-60	gamma	14.0 mCi (1965)	---	5.27
S-083	NS-28	Cf-252	neutron	5.9 mCi (3/71)	3.3	2.6
S-297	SRCF-134	Cf-252	neutron	7.0 mCi (11/76)	3.9	2.6
S-363	SN-112	Cf-252	neutron	0.225 mCi (~1977)	0.13	2.6
S-475	F-621	Co-60	gamma	10.28 mCi (8/84)	---	5.27
S-507	SRCF-147	Cf-252	neutron	7.32 mCi (7/85)	4.16	2.6

Source: SRDB 21358, PDF p. 355, Table VIII
NOTE: Encapsulation - All sources used for critical mass measurements are doubly-encapsulated stainless steel enclosures except for: (1) source S-082, whose encapsulation is not known; and (2) source S-363, which was used for another program and whose encapsulation is not known.

GENERATION OF FISSION AND ACTIVATION PRODUCTS

The amounts and types of fission and activation products produced in a criticality (or near-criticality) vary according to the nuclear fuel and geometry of the event. The products of a criticality are radioactive and contribute to radiation dose from the fuel. An assessment of experimental parameters used at the CML is necessary to identify the most likely distribution of fission and activation products adding to the radiation environment over time. Most of the experiments conducted in the CML are described in some detail in the document that presents the comprehensive history of the facility (SRDB 21358, PDF pp. 376-394). Table 3 provides summary information about the date ranges, materials involved, and number of experiments for each program. Table 4 further consolidates the information to show the number of experiments using each chemical and physical form of nuclear material.

[1] Exposure rates for Co-60 photons are estimated using Γ =13.2 R cm^2 h^{-1} mCi^{-1} (SRDB 75017, PDF p. 144).
[2] The specific activity of Cf-252 is 0.536 mCi/µg, and the dose equivalent rate from 1 µg of Cf -252 at 1 m in air is 0.0221 mSv/h (2.21 mremlh) from fast neutrons plus 0.0019 mSv/h (0.19 mrem/h) from gamma rays (SRDB 83312, PDF p. 4).

Table 3: Timeline of Experiments at the Rocky Flats Critical Mass Laboratory				
SRDB 21358 PDF Page(s)	**Dates**	**Material***	***n*****	**Program Description**
380	09/10/1965-04/05/1967	HEU Metal	235	Uranium Spherical Assemblies
380	08/19/1966-03/08/1967	HEU Metal	61	Massive Subcritical Uranium Spheres
381	04/20/1966	Weapon Component***	1	Weapons Study
381	ca. 1966	Not Specified	13	Special Materials
381-382	05/09/1967-08/11/1967	U solution	38	Poison Plate I
382	05/31/1967-09/12/1969	Pu metal	167	Oil-Reflected Plutonium
382-383	09/12/1967-01/24/1968	U solution	110	The “Christmas Tree”
383	02/16/1968-10/15/1968	U solution	48	Slab/Cylinder
383	05/24/1968-06/20/1968	U solution	6	Partially-Reflected Solution Slab
383-384	02/03/1968-02/07/1968	Pu Oxide	9	Plutonium Calorimeter
384	11/17/1969-04/16/1970, 10/1970	U metal in U solution****	206	Coupled Assembly
384-385	late 1960s	Pu metal	---	Plutonium Ingots
385	02/19/1971-05/11/1971	U metal in U solution****	52	Uncoupled Coupled Assembly
386	06/01/1972-07/14/1972	U solution*****	20	Poison Plate II
387	late 1973-02/17/1976, with 11-mo shutdown in 1975	Pu metal	76	Plutonium Cylinders I
388-389	04/21/1976-05/26/1976, 08/1977	U solution	186	Uranium Solution Cylinder Benchmark
389-390	02/1978-09/1981	LEU oxide	116	Uranium Oxide Contract
390-391	Summer 1982-12/20/1982	Pu metal	22	Pu Metal Cylinders II
391-392	05/1983-09/1984	U solution	61	Poisoned Tube Tank
392	07/1985-08/1986	U solution	32	Assorted Fuels Geometry
392-393	Spring 1986-10/1987	U solution	19	Shielded Annular Tank

*HEU (high-enriched uranium) is uranium enriched to 92% U-235 (PDF p. 288); LEU is low-enriched uranium, 4.5% U-235; U solution is high-enriched uranyl nitrate with a concentration of ~450 gU/L (PDF pp. 289, 381) ; Pu refers to ‘weapons grade’ plutonium (PDF p. 288) consisting of 93.56% Pu-239, 5.97% Pu-240, <0.5% Pu-241 (which decays to Am-241 with a 13.2-y half-life), and 0.01% Pu-242 (PDF p. 307).
**The symbol *n* is the number of criticality experiments in this program. The designation ‘---’ indicates that the number is unknown.
***The weapon component was returned to production and was not part of the permanent Building 886 inventory.
****Three different (unspecified) concentrations of uranyl nitrate were used in this study.
*****Uranyl nitrate in two concentrations: 450 gU/L and 52 gU/L.

Table 4: Number of Experiments for Each Material Physical and Chemical Form	
Material	**Experiments**
HEU Metal	296
HEU Solution	520
HEU Solution + HEU Metal	258
LEU Oxide	116
Pu Metal	274
Unknown	>15
Total	>1,479

Source: SRDB 21358, PDF pp. 376-394

Fission products in the fuels, and activation products in both the fuel and building materials, are sources of external radiation dose to personnel using the fuels and occupying the building; they present an internal dose potential for personnel that might ingest or inhale them. Personnel dosimeters assigned to RFP radiation workers document the external exposures. Internal exposures might result: (1) during operations from re-suspension of contamination on surfaces; or (2) during facility demolition from airborne dust. CML staff were provided routine bioassay (urinalysis and whole-body counts) to detect intakes of radioactive material. No follow-up bioassay was performed subsequent to any of several accidental spills of nuclear fuel, however, according to a senior staff member involved with the clean-up activities (SRDB 138605, PDF pp. 5, 10). NIOSH has found no incident reports or related bioassay results, although many documents were lost when RFP was decommissioned (e.g., SRDB 142480, PDF pp. 5, 8).

All the nuclear materials at the CML, except for HEU solution, were treated in some way to reduce contamination. Plutonium metal cylinders (SRDB 21358, PDF pp. 288, 305) and hemishells (SRDB21358, PDF p. 305) were coated with a light film of grease or oil, as were the HEU hemishells (SRDB 21358, PDF pp. 268, 295). LEU oxide was compacted into briquettes on receipt, sealed in plastic, and then sealed in an aluminum can with small holes drilled for injection of water (SRDB 21358, PDF p. 312). Plutonium ingots, borrowed from RFP production and returned immediately after conclusion of the experiments, were contained in thin-walled film cans (SRDB 21358, PDF p. 385). The HEU solution represents the most likely source of internal contamination by fuel and associated fission and activation products because it was handled in open tanks, and because several spills occurred requiring clean-up and recovery of the spilled fuel, leaving residual contamination that could dry and be re-suspended from surfaces (SRDB 21358, PDF pp. 447, 449, 452, 454-458, 464, 467-468, 486, 498, 500).

Four incidents involving facility contamination by solid material (powder) are reported; two of these incidents involved dried uranyl nitrate salt (from HEU solution) on reactor components:

- Plutonium metal sealed in a can reacted with water in the can, oxidizing the plutonium to powder and pushing off the can lid (SRDB 21358, PDF pp. 488-471).

- A can containing compressed low-enriched uranium powder dropped from a table to the floor, rupturing the can and spilling the powder (SRDB 21358, PDF p. 479).

- Two almost identical incidents involved dried uranyl nitrate salt (from an HEU solution experiment) on the surface of a suspended metal "reactivity shim" being knocked to the floor, contaminating the facility and workers (SRDB 21358, PDF pp. 500-502).

The fission and activation products most important from either an acute or chronic personnel exposure perspective are clearly those generated in criticality experiments involving HEU solution because of the predominance of solution spills over the course of CML's history (11 spills involving solutions or dried salts from solution experiments, compared with two contamination events involving solid fuels).

The experiments conducted in Building 886 generally involved power levels of no more than 10 milliwatts for no more than one hour (SRDB-20170, PDF p. 16). Approximately half of the experiments conducted in Building 886 actually achieved criticality. 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 (SRDB 104452, PDF p. 3). RFP never had a criticality accident, or even a close call, according to a Senior Research Scientist and author of a history of the Critical Mass Laboratory (SRDB 21358, PDF p. 79); CML operating specifications were designed to prevent such an accident from criticality experiments intended to approach (but never achieve) prompt criticality or nuclear excursion. A discussion of the adopted safety parameters is found in the *Technical Specifications for the Rocky Flats Critical Mass Laboratory* is provided in Attachment 2. The Site Description portion of the *RFP Technical Basis Document* includes this summary of criticality experiments at Building 886:

> *Approximately half of the 1,600 criticality experiments in Building 886 achieved criticality. Experiments in the RFP laboratory generally involved power levels of no more than 10 milliwatts, for no more than an hour. Approximately six high-power experiments were taken to between 10 and 100 times the power of typical tests. Using a conversion factor of $3x10^{16}$ fissions per megawatt-second, this power level and duration corresponds to a maximum of $1.01x10^{12}$ fissions from a typical RFP criticality experiment and a maximum of $1x10^{14}$ fissions from a high-power experiment. Records indicate that there were no incidents at Rocky Flats in which the power level of fissionable material became uncontrollable. The experiments were controlled by bringing the materials slowly to near criticality, observing the neutron flux to observe the reaction state. There is no indication in the available records*

> *that gamma exposure or exposure to created fission products was a worker exposure problem during these experiments.* (SRDB 20170, PDF p. 16)

The same document cites the ChemRisk study in concluding that some short-lived fission products were generated, but resulted in no personnel exposures (SRDB 20170, PDF p. 42):

> *Materials used in the experiments (uranyl nitrate metal powder)* [sic] *were re-used. Short-lived fission products were produced and none were indicated as having been released to the work or outdoor environment. The isotopes decayed rapidly and were contained until stable.* (ChemRisk, 1991)."

NIOSH simulated 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 and transmutation, and radioactive decay. The code was used to estimate the production of fission and activation products in an unreflected stainless steel tank containing a solution of high-enriched uranyl nitrate. The composition used for the ORIGEN-S calculations was a homogenized mixture of the uranyl nitrate solution, the 304 stainless steel tank, and the associated impurities. All input data were derived from the published criticality benchmark case for this series of experiments (SRDB 142464, PDF pp. 3-10). The calculations used a case-specific cross section library created using the TRITON analysis sequence of the SCALE code system. The TRITON case used the heterogeneous geometry with the height of the tank truncated to the solution height of 31.2 cm (the solution height of the benchmark case). KENO-VI was used to model neutron transport.

Analysis of the number and duration of experiments at CML involving uranium solution (from Table 4) yields the results shown graphically in Figure 1, which is a time distribution of HEU solution critical experiments over the history of CML. Dates correspond to Program dates in Table 3. Heights of the bars represent the average number of experiments per day during the Program period, and numbers above the bars are the total number of experiments. The dotted line shows the overall average number of HEU solution critical experiments over the history of operations at CML.

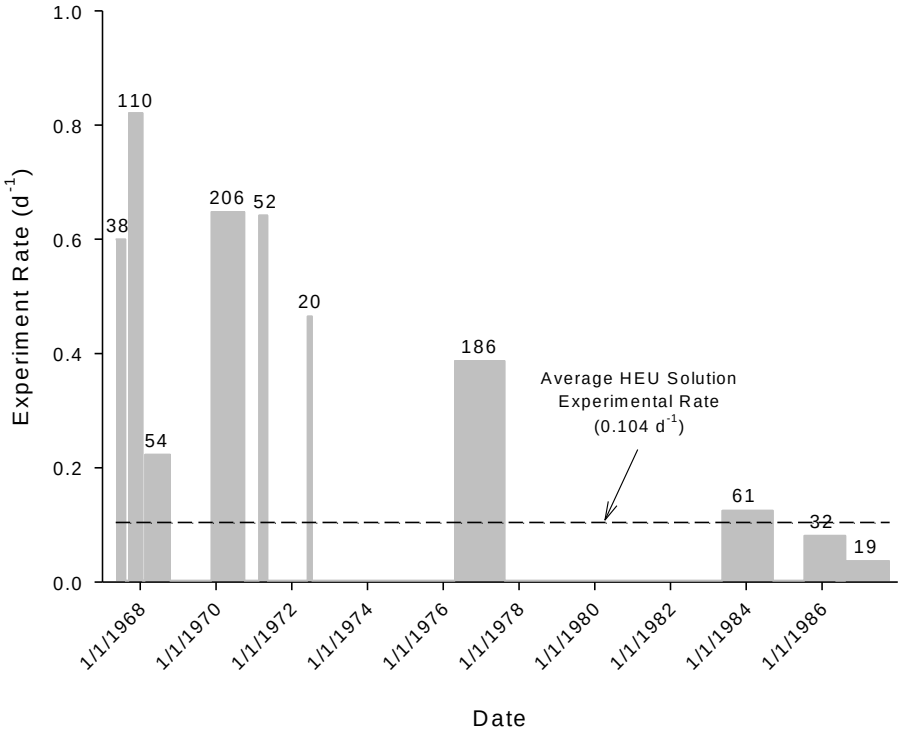


Figure 1: Time Distribution of HEU Solution Critical Experiments Over the History of CML

Most of the solution experiments (480 of 778 total) were conducted in the first six years of the facility's history, from 1967 through 1972. Another 186 experiments were conducted in 1976 and 1977, and the remainder (112) from 1983 to October 1987. The rate of experimentation ranged from just over 0.8 per day (1967-1968) to less than 0.04 per day (1986-1987), with an overall rate of 0.104 per day.

The ORIGEN-S calculations assumed that each experiment took one hour and was conducted at a power level of 10 mW. All experiments during a Program period were assumed to have been performed serially and continuously at the end of the period, and then the resulting reaction products were allowed to decay until the start of the next Program period.[3] The fuel was

[3] For example, there were three Program periods involving HEU solution from 05/09/1967 to 06/20/1968, consisting of 38, 110, and 54 experiments. The analysis considers continuous runs (at a power level of 10 mW) of 38 h and 110 h, followed respectively by decay over 32 d and 23 d between periods, and then a 54-h run followed by decay over the idle time between this Program and the next in 1976.

assumed to have an HEU concentration of 145.68 g U/L in each experiment. Total activities of the dosimetrically important fission and activation products[4] in the fuel at the end of CML operations, plus 10 days' decay time (the shortest decay time allowed by ORAUT-OTIB-0054 [SRDB 140847] in assessing worker exposures) are provided in Table 5.

Table 5: Activity of Dosimetrically Important Fission and Activation Products 10 Days After the End of CML Operations

Radioisotope	(Ci)	(Bq)
Mn-54	7.825E-09	2.895E+02
Fe-55	6.860E-08	2.538E+03
Co-58	2.131E-08	7.885E+02
Co-60	9.777E-11	3.617E+00
Sr-89	4.585E-06	1.696E+05
Sr-90	8.719E-07	3.226E+04
Y-90	8.703E-07	3.220E+04
Y-91	5.120E-06	1.894E+05
Zr-95	6.017E-06	2.226E+05
Nb-95	3.110E-06	1.151E+05
Mo-99	7.765E-06	2.873E+05
Ru-103	3.277E-06	1.212E+05
Ru-106	1.835E-07	6.790E+03
Cd-113m	5.792E-11	2.143E+00
Cd-115m	4.752E-10	1.758E+01
Sb-125	1.236E-08	4.573E+02
Te-129m	1.212E-07	4.484E+03
Te-132	7.020E-06	2.597E+05
I-131	7.149E-06	2.645E+05
I-132	7.231E-06	2.675E+05
Cs-134	2.810E-12	1.040E-01
Cs-136	1.152E-08	4.262E+02
Cs-137	8.927E-07	3.303E+04
Ba-140	1.325E-05	4.903E+05
La-140	1.490E-05	5.513E+05
Ce-141	7.126E-06	2.637E+05
Ce-144	2.816E-06	1.042E+05
Pr-143	1.349E-05	4.991E+05
Pr-144	2.817E-06	1.042E+05
Nd-147	5.067E-06	1.875E+05
Pm-147	7.897E-07	2.922E+04
Pm-148m	8.407E-15	3.111E-04
Sm-151	2.361E-08	8.736E+02
Eu-154	6.034E-14	2.233E-03

[4] Radionuclides contributing ≥ 1% of the committed organ dose are considered to be dosimetrically important, as specified in ORAUT-OTIB-0054 (SRDB 140847).

Table 5: Activity of Dosimetrically Important Fission and Activation Products 10 Days After the End of CML Operations		
Radioisotope	**(Ci)**	**(Bq)**
Eu-155	1.175E-08	4.348E+02
Ta-182	0.000E+00	0.000E+00

These inventories would have been fairly uniformly distributed within the total volume of uranium solution at the CML because the solution used in experiments was eventually transferred back to the tank farm for storage. These back-and-forth transfers would tend to result in fairly uniform mixing; in addition, transfers between storage tanks were sometimes performed intentionally for homogenizing the solutions (SRDB 21358, PDF p. 449).

Organ doses for CML staff from intakes of these radionuclides are discussed in the later section, *Assessment of Unmonitored Radiation Dose at the CML*.

DECOMMISSIONING AND DEMOLITION

The potential for fission and activation product contamination in the nuclear materials used as fuel, in building fixtures, and in facility construction materials was recognized and considered in planning the Building 886 Cluster decommissioning and demolition. However, based on survey results, personnel interviews, and the historical record, the isotopes of concern were instead determined to be components of the fuels themselves used in the experiments (U-235, U-234, U-238, Pu-239 and Am-241). High Contamination Areas were posted in Buildings 886, 875 (filter plenum), and 880 (storage shed)[5] and controlled to the transuranic limits of 10 CFR 835 (SRDB 104734, PDF pp. 22-23).

DCAS has captured copies of original neutron, gamma, removable alpha, and removable beta/gamma survey results forming the basis of this radiological characterization (SRDB 104734, PDF pp. 32-265). A summary of the dose rate survey results is shown in Table 6. Penetrating dose rates from gamma-emitting radionuclides ranged from 0.03 mrem/h to 3.4 mrem/h in Building 886, Rm 103 on 07/18/1994 (SRDB 104734, PDF pp. 83-85). Dose rates associated with the tank farm, which housed both the enriched uranium solution inventory and a set of metal plutonium hemishells in Room 103, ranged from 0.25 mrem/h – 3.4 mrem/h, whereas dose rates around the glove box, cabinets, and hood varied from 0.03 mrem/h – 0.5 mrem/h. Surveys

[5] Parts and equipment used in experiments were stored in Building 880. Contaminated items were sealed in plastic bags, but these tended to degrade with time (SRDB 21358, PDF p. 263).

of Building 886, Rooms 101, 102, and 103 in November and December 1997 document greatly-reduced gamma exposure rates of <0.5 mrem/h (SRDB 104734, PDF pp. 33-35 and 48-51).

These surveys bracket the beginning and end of the project to remove enriched-uranium solution (the most difficult and time-consuming part of decommissioning prior to demolition), that spanned the 2-year period from June 1995 to June 1997 (SRDB 136853, PDF p. 4). Removal of the uranyl nitrate solution concluded by the end of 1996 (SRDB 21358, PDF p. 407; 22018, PDF p. 5. The results suggest that the source for personnel dose from penetrating radiations was predominantly the radioactive fuel used in experiments. Any dose contributions from activation products in the building materials and fixtures after removal of the fuel in late 1996 were too small to be detected by instruments used for the 1997 radiological surveys.

Table 6: Dose Rate Survey Results for Building 886, Rocky Flats Plant Critical Mass Lab

Survey Date	Area	n*	Dose Rate Range of Values (mrem/h)			
			Gamma	Neutron	(Gamma + Neutron)	Beta/Gamma
07/18/1994**	B886, Rm 103 – Glovebox, Cabinets, Hood	15	0.03-0.5	0.0-0.1	0.03-0.5	0.05-0.3
07/18/1994**	B886, Rm 103 – Tank Farm	18	0.25-3.4	0.0-0.2	0.25-3.5	0.18-2.40
11/04/1997***	B886, Rms 101, 102 & 103	15	<0.5	<1.0	<1.5	---
12/02/1997***	B886, Rms 101, 102 & 103	15	<0.5	<1.0	<1.5	---

Source: SRDB 104734, Appendix A, PDF pp. 33-35, 48-51 and 83-85

* n = number of measurements

**Survey instruments used on 07/18/1994 were the Victoreen 450B for γ and β-γ measurements and the Ludlum 12 4 for neutrons, with reported backgrounds of 0.01 mrem/h and 0.0 mrem/h, respectively (SRDB 104734, PDF p. 83).

***Survey instruments used on 11/04/1997 and 12/02/1997 were the Eberline RO-20 for γ and β-γ, and the Ludlum 12-4 for neutrons, with reported backgrounds of <0.05 mrem/h and <1.0 mrem/h, respectively (SRDB 104734, PDF pp. 33, 48).

Contamination levels determined for specific areas in the Building 886 Cluster are described in a pre-demolition characterization study (SRDB 22002, PDF pp. 22-25). 10 CFR 835 unrestricted release criteria for surface radiological contamination on equipment and building materials were specified for post-deactivation decontamination and decommissioning activities (SRDB 22002, PDF p. 38). Neutron activation of U-238, with subsequent beta decay of the short-lived Np-239 product to Pu-239, was considered as a source of plutonium contamination in the Building 886 Cluster; however, it was not believed to be a source of significant plutonium production because of the low power and short criticality times historically attained during experiments at CML. An RFP technical basis document (TBD) on building release criteria contains an analysis of the

neutron activation scenario and notes that, due to the low power levels and short criticality times involved:

> *Analysis for low levels of plutonium within the solutions (or hold-up) has never been deemed necessary.* (SRDB 103869, PDF pp. 3-4)

The TBD containing this analysis also states:

> *In spite of the fact that low-level criticality experiments were performed in Room 101 of Building 886, residual radioactivity resulting from the production of fission products is not expected to exist within the facility, refer to Process Knowledge Characterization Building 886 Highly Enriched Uranyl Nitrate Solutions, TD 95-012, June 28, 1995.*[6] (SRDB 103869, PDF p. 5)

A Los Alamos Technical Associates Project Manager responsible for removing the weapons-grade enriched uranium from the CML from 1995 to 1997 indicated in a NIOSH interview that baseline contamination surveys were performed in support of the facility deactivation project; he does not recall any measurements that were out of the ordinary (SRDB 136853, PDF pp. 4-5).

The Final Project Closeout Report for the Building 886 Cluster demolition and closure summarizes actions taken and the final condition of the site. Uranium was the predominant contaminant addressed in the Building 886 Cluster closure project because of multiple enriched uranium solution spills over the facility's history. Uranium release limits based on DOE Order 5400.5 were adopted[7] for property and waste release, and for final building disposition (SRDB 103869, PDF pp. 4-5). The *Final Project Closeout Report* includes a breakdown of waste types generated by the closure project, which is reproduced in Table 7 (SRDB 22018, PDF p. 14). Over 90% of the waste (by volume) from the closure project was disposed of as follows: in a sanitary landfill (52%), or recycled and used as onsite backfill (39%). Less than 3.5% of the waste and debris from closure of the Building 886 Cluster included a radiological component broken down as follows: low-level radiological waste (3.3%), low-level mixed waste (0.02%) and transuranic waste (0.04%).

[6] TD 95-012 was prepared by Environmental Technologies Technology Development, E&WM EG&G Rocky Flats, Inc. (Kathy Swan-Bogard and Angelo E. Hodges, III). NIOSH has been unable to capture this document.

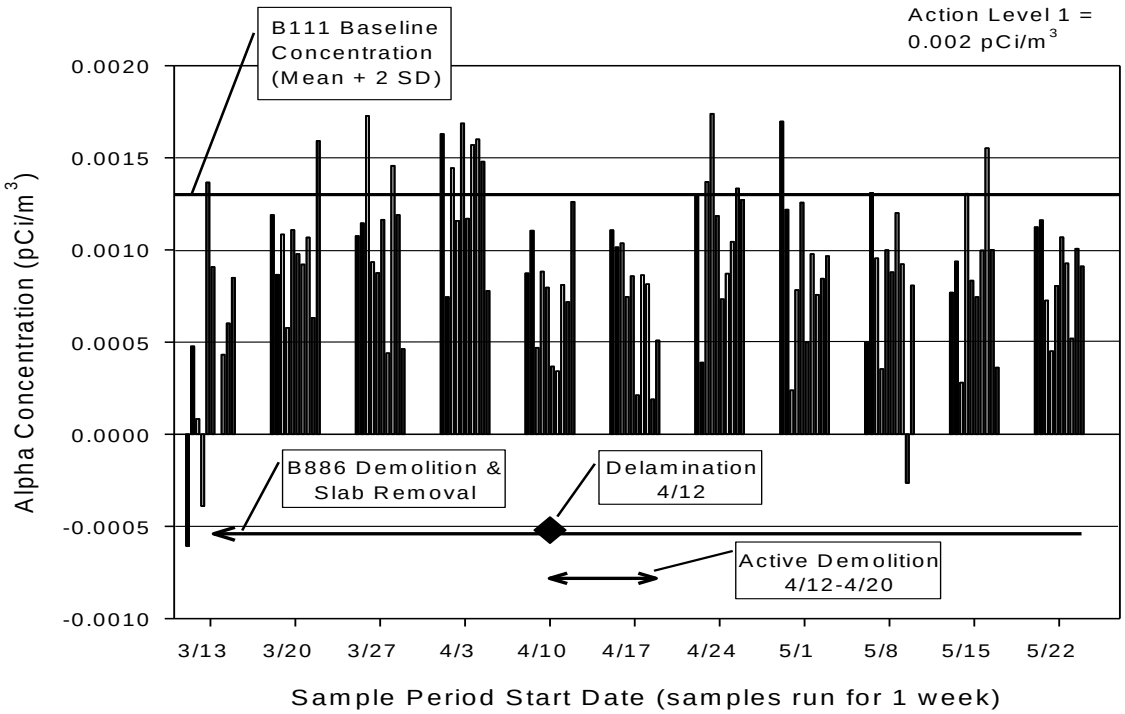
[7] Release limits are the same in DOE Order 5400.5 and 10 CFR 835 (specified in the earlier *Interim Measure/Interim Remedial Action Plan for the 886 Cluster* (SRDB 22002, PDF p. 38), except that the guideline values of DOE Order 5400.5 specify limits on alpha activity "for uranium, associated decay products and alpha-emitters, and beta/gamma activity for these emitters."

Table 7: Disposition of B886 Demolition Wastes		
Waste Type	**Volume**	**Description**
Sanitary	3,835 yd^3 (2,420 tons)*	Above-grade building debris
Hazardous	Minor amounts	Electronic circuit boards, thermostats, exit signs, batteries, fluorescent light bulbs, and any other RCRA hazardous components were removed and taken for combination with like waste streams for disposal.
TSCA Waste	0.132 m^3	PCB ballasts were removed and packaged for disposal.
Asbestos	265 m^3	Friable asbestos, including the hydrolasing waste from removal of the skimcoat.
Low-Level Radiological	187 m^3	Miscellaneous decommissioning and demolition debris
Low-Level Mixed	0.865 m^3	Miscellaneous decommissioning and demolition waste
Low-Level TSCA	71 m^3	Low-Level PCB Bulk Product waste
TRU	2.32 m^3	Glove box and associated waste
Recycled Material	2,171 m^3	Concrete for onsite backfilling

Source: SRDB 22018, PDF p. 14

* 3,835 yd^3 = 2,932 m^3

The Final Project Closeout Report also provides results of air monitoring for alpha-emitting radionuclides during the Building 886 Cluster demolition activities, as performed by a 10-sampler network within and around the RFP perimeter, and outside the perimeter toward the Denver metropolitan area (SRDB 22018, PDF pp. 13, 40-41). Filters were analyzed weekly. Figure 2 shows these results graphically. Results are compared with the mean plus two standard deviations value from demolition of an uncontaminated structure (B111). The Building 886 Cluster results were consistently below the 0.002 pCi/m^3 Action Level 1 (corresponding to a 1 mrem annual dose to a public receptor).



Source: SRDB 22018, PDF pp. 13,40

Figure 2: Results of Continuous Alpha Air Monitoring During Demolition of the CML Building 886 Cluster

ASSESSMENT OF UNMONITORED RADIATION DOSE AT THE CML

From 1964 until the early 1990s, all Rocky Flats workers (including those at CML) were assigned dosimeters that monitored external radiation dose; this time span includes the entire period of CML operations (SRDB 20175, PDF pp. 19-20). The available RFP internal monitoring information indicates that RFP workers with the potential to receive intakes primarily of plutonium, americium, enriched uranium, or depleted uranium were monitored accordingly (SRDB 132776, PDF p. 30). A principal CML scientist indicated in a NIOSH interview that he participated in urinalysis and body count bioassays on an approximately annual basis, and that he believed everyone working with uranium or plutonium was similarly monitored (SRDB 138605, PDF pp. 5, 10). These personnel dosimetry and bioassay records are available to NIOSH. Doses from external penetrating radiation sources (nuclear fuel, sealed radioactive sources, fission and activation products) and intakes of uranium or plutonium are accounted for by the RFP radiation dosimetry monitoring programs. Sufficient detail exists about both routine operations and

incidents, as well as about the ambient radiation dose environment prior to decommissioning, that external doses to, and intakes of radioactive materials by, individual claimants could be reconstructed with sufficient accuracy if dosimetry records are unavailable.

Possible exceptions to the ability to reconstruct dose with sufficient accuracy lie in the reported absence of confirmatory bioassays after any of the contamination incidents or spills, and in the likelihood that routine bioassays (designed to detect primarily plutonium and uranium) could fail to recognize the presence of fission or activation products. *In-vivo* gamma spectrometry would likely detect gamma-emitting nuclides, but would be insensitive to pure beta emitters, like Sr-90. Urinalysis by alpha-counting or spectrometry is unlikely to detect any of the fission products, which tend to be beta-gamma emitters and require different chemical separation techniques. However, sufficient detail about contamination incidents exists to provide a basis for deriving bounding estimates of resulting acute intakes. The possibility that fission and activation products might have been missed in routine radio-bioassays is addressed below by evaluating the bounding chronic intake of these radionuclides and their contribution to internal dose.

Radiation dose from intake of fission and activation products could have occurred during the 22 years of CML operation or from the generation of dust during facility demolition. The vast majority of CML building debris was buried in sanitary landfills or used as backfill for onsite projects. These means of disposal, coupled with the low alpha concentrations in air around the facility, indicate that personnel dose from uranium, transuranic, or fission and activation product contamination in the CML during demolition was unlikely. Dose to personnel during operations might have resulted from intakes occurring during clean-up of fuel spills (mostly from spills of uranyl nitrate solution) or from re-suspension of contamination deposited on surfaces as a result of these spills.

Information about reported HEU solution spills is provided in Table 8. Most of the large solution spills occurred in the first four years of facility operation and involved up to 16.1 kg of uranium. One spill in 1980 involved 2.66 kg of solution. Other spills were small enough that the volume of solution or mass of uranium involved were not mentioned. Considerable effort was spent to recover as much uranium as possible from the larger spills because of monetary, radiological health, and accidental criticality considerations. All the spills were confined within either the Assembly/Experiment Room (Room 101) or the Mixing Room (Room 103), except for the November 30, 1967 event, which involved an exhaust duct and filter plenum. These rooms were located in the "Hot Area," to which access was controlled (in contrast to the "Cold Area" where offices were located) (SRDB 21358, PDF p. 110).

Table 8: High-enriched Uranium Solution Spills Over the History of the CML				
Date	**Volume (L)**	**Uranium Mass (kg)**	**Contaminated Area**	**PDF Page(s)**
7/2/1965	0.5	0.225	22 m^2 (Floor)	447
7/14/1965	---	---	10 m^2 (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^2(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: SRDB 21358

*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 re-suspended high-enriched uranyl nitrate salts accidentally knocked from the surface of a large reactivity shim.

As shown in Table 8, the recorded amount of high-enriched uranyl nitrate solution spilled is just under 30 kg. Almost all of this spilled fuel was recovered and the surfaces decontaminated.

Radiological characterization of the CML prior to demolition (SRDB 22002, PDF p. 25) noted that Rooms 101 (Experiment/Assembly Room), 102 (dry HEU storage), 103 (HEU solution tank farm and Pu hemishell storage), and 108 (hallway) were all Contamination Areas (<2,000 dpm/100 cm^2 alpha).

The only High Contamination Areas found prior to demolition were in the Assembly hood (Room 101) and the downdraft table with attached glove box (Room 103). The exhaust air duct from Building 886 to the exhaust plenum in Building 875 was listed as an area of radiological concern. The Assembly hood, downdraft table with attached glove box, and exhaust air duct and

plenum are locations designed to prevent exposure and move air out of the building; therefore, they would not contribute to re-suspended radiological activity in the CML.

The Contamination Area limit of 2,000 dpm/100 cm^2 alpha corresponds[8] to 9×10^{-4} µCi/100 cm^2 or 9 µCi/m^2 HEU. The total floor area of these rooms[9] is approximately 220 m^2, giving the total amount of contamination to be 1,980 µCi HEU.

Bounding estimates for intake of the fission and activation products from re-suspended contamination during routine operations at CML are derived using Equation 1 below:

(Equation 1)

$$I = \frac{C f_S f_R R t}{A}$$

Where:

- I = Intake for each of the dosimetrically-important radionuclides
- C = Radionuclide concentration
- f_S = Spill fraction evaluated for this assessment to be 0.002917 (1,980 µCi divided by 70 µCi/g for HEU, divided by total HEU mass in the ORIGEN-S model) (SRDB 144472)
- f_R = Re-suspension factor of 1.5×10^{-4} m^{-1}, corresponding to the 95th percentile value in the distribution of re-suspension factors reported in Figure 3-2 of ORAUT-OTIB-0070 (SRDB 108851, PDF p. 9).
- R = Breathing rate of 1.2 m^3 h^{-1} from ORAUT-OTIB-0054 (SRDB 140847)
- t = Intake period of 2 y = 4,000 h from ORAUT-OTIB-0054 (SRDB 140847)
- A = Combined surface area of 220 m^2 of Building 886 Rooms 101, 102, 103, and 108

[8] The specific activity of HEU, given by the empirical formula $S = 0.4 + 0.38\,E + 0.0034\,E^2$, is approximately 70 µCi/g for E = 93% enrichment in ^{235}U (SRDB 1781, PDF p. 25).

[9] Estimated floor areas for the individual rooms (SRDB 21358) are 120 m^2 for Room 101 (PDF p. 120); 40 m^2 for Room 103 (PDF p. 167); and 60 m^2 for Rooms 102 and 108 combined, assuming this area to be about half that of Room 101 (PDF p. 110).

The total activity of each radionuclide in the 1,980 µCi of HEU contamination is given as the product of the total radionuclide inventory and spill fraction minus the ratio of HEU in contamination to HEU in the total fuel inventory used in the ORIGEN-S calculations. The intake *I* for each of the dosimetrically important radionuclides in Table 5 was estimated and the resulting values are shown in Table 6.

Table 6: Intakes of Dosimetrically Important Fission and Activation Products from Re-suspension of Contaminants in CML

Radioisotope	Total Inventory (Bq)	Intake (Bq)
Mn-54	2.895E+02	2.764E-03
Fe-55	2.538E+03	2.423E-02
Co-58	7.885E+02	7.528E-03
Co-60	3.617E+00	3.454E-05
Sr-89	1.696E+05	1.620E+00
Sr-90	3.226E+04	3.080E-01
Y-90	3.220E+04	3.074E-01
Y-91	1.894E+05	1.809E+00
Zr-95	2.226E+05	2.125E+00
Nb-95	1.151E+05	1.099E+00
Mo-99	2.873E+05	2.743E+00
Ru-103	1.212E+05	1.158E+00
Ru-106	6.790E+03	6.482E-02
Cd-113m	2.143E+00	2.046E-05
Cd-115m	1.758E+01	1.679E-04
Sb-125	4.573E+02	4.366E-03
Te-129m	4.484E+03	4.281E-02
Te-132	2.597E+05	2.480E+00
I-131	2.645E+05	2.525E+00
I-132	2.675E+05	2.554E+00
Cs-134	1.040E-01	9.926E-07
Cs-136	4.262E+02	4.069E-03
Cs-137	3.303E+04	3.153E-01
Ba-140	4.903E+05	4.681E+00
La-140	5.513E+05	5.263E+00
Ce-141	2.637E+05	2.517E+00
Ce-144	1.042E+05	9.947E-01
Pr-143	4.991E+05	4.765E+00
Pr-144	1.042E+05	9.951E-01
Nd-147	1.875E+05	1.790E+00

Table 6: Intakes of Dosimetrically Important Fission and Activation Products from Re-suspension of Contaminants in CML		
Radioisotope	**Total Inventory (Bq)**	**Intake (Bq)**
Pm-147	2.922E+04	2.790E-01
Pm-148m	3.111E-04	2.970E-09
Sm-151	8.736E+02	8.340E-03
Eu-154	2.233E-03	2.131E-08
Eu-155	4.348E+02	4.151E-03
Ta-182	0.000E+00	0.000E+00

Maximum organ doses from these intakes are shown in Table 7. The largest total organ dose, if radioiodines are included[10], is 1.1×10^{-6} Sv to the thyroid, primarily from soluble I-131. The largest total organ dose if radioiodines are excluded is 6.1×10^{-7} Sv to lungs. Dose conversion factors and individual organ doses are provided in an Excel spreadsheet (SRDB 144472). All individual organ doses are less than 1×10^{-6} Sv. Therefore, NIOSH concludes that doses to CML staff from re-suspension of contamination containing fission and activation products are negligible.

Table 7: Maximum Committed Organ Doses from Inhalation of Fission and Activation Products in Re-suspended Contamination				
Category	**Organ**	**Committed Dose (Sv)**	**Excluding Radioiodines**	
			Organ	**Committed Dose (Sv)**
Soluble	Thyroid	1.1×10^{-6}	Bone Surface	3.7×10^{-7}
Moderately Soluble	Thyroid	1.0×10^{-6}	Lungs	4.0×10^{-7}
Insoluble	Thyroid	1.0×10^{-6}	Lungs	6.1×10^{-7}

[10] Iodines are volatile and vaporize from the matrix quickly; therefore, including them in the analysis is very claimant-favorable.

SUMMARY AND CONCLUSIONS

NIOSH concludes that external radiation exposure to CML workers and staff is accounted for by Rocky Flats' personnel dosimetry program, which assigned radiation dosimeters to all workers. The personnel dosimetry program also included periodic bioassay (urinalysis and body counts) that focused primarily on identifying uranium and plutonium intakes. The *in-vivo* bioassay, using gamma spectrometry, would be expected to easily detect most fission and activation products present in any significant amount, except for radioisotopes like Sr-90, which emit beta radiation not detectable in a routine body count or in a urinalysis evaluated for alpha-emitters.

No monitoring results or theoretical assessments suggest that fission products from the criticality experiments in the Critical Mass Laboratory, or activation products from neutrons produced in the controlled low-level criticalities, resulted in the production of other than low levels of radioactive materials in the fuels. The fuel itself was removed by the end of 1996 (SRDB 21358, PDF p. 407; 22018, PDF p. 5) for reprocessing (SRDB 136853, PDF p. 4) prior to facility demolition. The possibility of activation products in Building 886 construction materials and fixtures was considered in pre-demolition characterization studies, but monitoring results failed to detect beta or gamma emissions characteristic of these radionuclides (SRDB 104734, Appendix A, PDF pp. 33-35, 48-51, and 83-85). Release criteria based on requirements of DOE Order 5480.5 were adopted for property, waste release, and final building disposition (SRDB 103869, PDF pp. 4-5). Over 90% of waste (by volume) from the closure project was disposed of in a sanitary landfill or recycled. Less than 3.5% of the waste and debris from closure of the Building 886 Cluster included a radiological component sufficient to require disposal in a low-level radiological, low-level mixed waste, or TRU waste site (SRDB 22018, PDF p. 14).

Estimates of the total activity in fission and activation products from irradiation of high-enriched uranyl nitrate solution fuel (the fuel contributing most to contamination within the facility) indicate that only inconsequential amounts of these products were available to contribute to radiation doses from re-suspension of residual contamination. Organ doses to individual radionuclides from inhalation of re-suspended contamination at CML are less than 10^{-6} Sv, with the largest total organ dose being 1.1×10^{-6} Sv to the thyroid, if radioiodines are included (a very claimant-favorable assumption).

It is possible that some personnel could have inhaled or ingested dosimetrically-significant amounts of fuel (particularly high-enriched uranyl nitrate) during clean-up of several accidental spills. NIOSH has found no record of follow-up bioassay related to these accidents; and one interviewee closely associated with the facility and involved in the clean-up activities stated that no follow-up bioassay was performed. However, the number of individuals who might have been so exposed is limited to a single senior staff member and a few workers and radiation monitors. Considerable information about the spills and clean-up activities is provided in the published facility history; therefore, individual assessment of their exposure potential is possible. The resulting dose, in any case, would be dominated by uranium (or, in one case, plutonium) because of the very low ratio of fission and activation products to fuel. All the CML staff members are believed to have been included in routine bioassay programs for uranium and plutonium.

NIOSH concludes, based on the weight of evidence from the detailed history of the CML, computer modeling of criticality experiments, and radiological measurements after operations ceased, that no significant personnel dose to Rocky Flats workers or contractors resulted from the generation of fission or activation products in the building materials and fixtures of the Building 886 Cluster as a result of the criticality experiments conducted there over its lifetime.

REFERENCES

SRDB 20170: *Technical Basis Document for the Rocky Flats Plant – Site Description* (ORAUT-TKBS-0011-2)

SRDB 20175: *Technical Basis Document for the Rocky Flats Plant – Occupational External Dosimetry* (ORAUT-TKBS-0011-6)

SRDB 21358: *A Technically Useful History of the Critical Mass Laboratory at Rocky Flats*

SRDB 22002: *Interim Measure/Interim Remedial Action Plan for the 886 Cluster*

SRDB 22018: *Final Project Closeout Report for the 886 Cluster Closure Project*

SRDB 22731: *Annals of the ICRP – Dose Coefficients for Intakes of Radionuclides by Workers*

SRDB 103869: *Methodology for Establishing Property, Waste, and Building Release Criteria for the 886 Cluster*

SRDB 104452: *Historic American Engineering Record – Rocky Flats Site Building 886*

SRDB 104734: *Reconnaissance Level Characterization Report for the 886 Cluster Decommissioning Project with Appendices*

SRDB 132776: *SEC Petition Evaluation Report, Petition SEC-00030*

SRDB 136853: *Rocky Flats Interview – Radiological Exposure Sources at the Critical Mass Laboratory*

SRDB 138605: *Rocky Flats Interview – Anomalous Events at the Critical Mass Laboratory*

SRDB 140847: *Fission and Activation Product Assignment for Internal Dose-Related Gross Beta and Gross Gamma Analyses* (ORAUT-OTIB-0054)

SRDB 142464: *Minimally Reflected Cylinders of Highly Enriched Solutions of Uranyl Nitrate*

SRDB 142480: *Preservation and Dissemination of the Hardcopy Documentation Portion of the NCSP Nuclear Criticality Bibliographic Database*

SRDB 108851: *Dose Reconstruction During Residual Radioactivity Periods at Atomic Weapons Employer Facilities* (ORAUT-OTIB-0070)

SRDB 144472: *Rocky Flats CML – DCFs and Organ Doses from Resuspension of CML F&AP*