Environmental Protection Department

Environmental Monitoring and Analysis Division

Plutonium Discharges to the Sanitary Sewer: Health Impacts at the Livermore Water Reclamation Plant

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Plutonium Discharges to the Sanitary Sewer: Health Impacts at the Livermore Water Reclamation Plant

I. Introduction

The Lawrence Livermore National Laboratory (LLNL) is the largest discharger of sewage treated by the Livermore Water Reclamation (LWRP), contributing approximately 7% by volume of the LWRP influent. LLNL operations, as potential sources both of industrial pollutants and radioactivity, are therefore of particular concern to the LWRP. For this reason, LLNL has maintained vigorous wastewater discharge control and monitoring programs. In particular, the monitoring program has demonstrated that, except in a few rare instances, the concentration of contaminants in LLNL effluent have always remained below the appropriate regulatory standards. The exceptions have generally been due to inadvertent discharges of metals-bearing solutions produced by metal plating or cleaning operations.

As regards plutonium discharges to the sanitary sewer, the monitoring program has demonstrated that levels have always been far below the DOE discharge guidelines. The data do, however, confirm that LLNL's effluent contains plutonium, and have revealed instances of acute, as well as chronic, discharges.

In order to assess the impact of those discharges on LWRP operations, LLNL performs monitoring to obtain a complete analysis of the environmental fate of the plutonium. The concentration of plutonium in LLNL effluent is measured on a monthly basis. At the LWRP, the sewage is separated into two phases. Most of the heavy radioisotopes in the sewage are precipitated with sludge that is dried and disposed of at a sanitary landfill. The remainder is discharged to the San Francisco Bay with the water purified by the treatment process. To cover the solid disposal pathway, LLNL analyzes both dried sludge, and soil from areas where sludge affected by acute releases has been spread for drying. The liquid pathway is monitored through analysis of a monthly composite of LWRP's effluent.

For the quantities of radiation contained in LLNL sewage (on the order of a microcurie released each month in roughly 30 million liters of effluent), the main exposure pathway is via inhalation of resuspended sludge and soil during drying operations at the LWRP. Due to the relative insolubility of plutonium, skin contact and inadvertent ingestion represent lesser hazards.

Using recent monitoring data and work schedules obtained from the LWRP, calculations are performed that demonstrate that past and current discharges of plutonium to the LWRP pose no significant risk to the health and safety of either LWRP personnel or the public.

II. Monitoring Data

Table I summarizes recent monitoring data (from 1988 to 1990, Reference 1) used in performing the dose assessments described in section V below. As described in Reference 1, sludge samples are acquired directly from activated sludge tanks at the LWRP, then dried and analyzed for plutonium. The soil sampling procedure calls for 8.25 cm diameter by 5 cm deep cores to be collected at each of the corners of a 2-meter wide square, and at the center of the square. The five subsamples are mixed and then analyzed.

Based upon monitoring data collected at locations remote from the LLNL site, global fallout has deposited 0.005 pCi/dry g of plutonium on Livermore Valley soils. The concentrations in Table I are therefore primarily due to Laboratory operations. The soil contamination is principally due to sludge spreading operations following a 1967 release to the sewer of 32 mCi of plutonium. The sludge contamination is assumed to be due to the slow leaching of plutonium remaining in the sewer pipes from past discharges.

Table I. Summary of plutonium activity in LWRP soil and sludge (1988-1990)

	²³⁹ Pu [pCi/dry g]	
	average	maximum
Sludge	0.07	0.23
Soil	0.10	0.57

Along with the soil monitoring data, LLNL maintains an air particulate sampling station on the east boundary of the LWRP, downwind from the soil affected by the 1967 discharge. This is part of an air particulate sampling network that includes locations remote from the LLNL site. Air plutonium concentrations at the LWRP sampler, and two remote sampling locations (the Livermore Veterans Administration hospital, and the Tracy Fire Department), are shown in Figure 1 and Table II (Ref. 1 and 2). As seen in Figure 1, the results at all locations show a large decrease in the reported plutonium concentrations since 1979, when the analysis was first performed, followed by fairly stable readings from 1984 onward. The decrease is most probably due to improvements in the analytical method, since the actual

plutonium concentration at the remote locations, which is due primarily to global fallout introduced into the atmosphere prior to 1965, was stable throughout this period. The average air concentrations shown in Table II, therefore, are based only on the data gathered starting in 1984.

The data show only a slight indication that levels at the LWRP are higher than those at the other sites. Such an increase could be attributed to the elevated soil plutonium concentrations presented in Table I.

Table II. Plutonium activity in air at locations remote from the Livermore site.

Location	Pu Concentration [10 ⁻¹² pCi/mL air]
VA Hospital	0.52 ± 0.13
Tracy Fire Dept.	0.57 ± 0.12
LWRP	0.72 ± 0.12

III. Exposure Models

In the case of contamination levels as low as those presented in Tables I and II, the primary dose is received through inhalation of respirable dust. The dose received is controlled by the plutonium activity on respired dust particles and the length of time the individual breathes affected air. Plutonium in dust which remains in the lung cavities can cause radiation damage to cells in the lungs. Furthermore, the plutonium will partially solubilize in the bloodstream, and be captured in other body tissues. It is the exposure of lung and bone that dominates the lifetime dose to the individual.

Two populations may be exposed to plutonium at the LWRP site: the public and LWRP workers. In estimating the dose received by the public, the downwind air monitoring data presented in Table II can be used to estimate the plutonium activity in the respired air. Use of those data will overestimate the exposure, as the sampling technique captures air particulates up to 50 μm in size, while "respirable dust" is generally only particles up to 10 μm . For the exposure time, we maximize the calculated dose to the individual, assuming that exposure occurs continuously throughout the year (8766 hours).

Calculation of the dose that might potentially be received by workers is more difficult. Without benefit of a detailed study of ambient conditions at the LWRP, it is not possible to make a precise estimate of worker exposure. However, using public health standards for ambient dust levels, and data

obtained from LWRP supervisors concerning work practices, very conservative upper bounds on possible worker doses can be calculated. As these doses are extremely small compared to regulatory standards (Section V), LLNL proposes to forego the development of a more detailed model.

The first step in performing the worker dose assessment is to estimate the ambient concentration in the air of plutonium-contaminated dust. When contamination sites are old, such as the soil at the LWRP, it is generally assumed that the loose surface soil has been blown off, and the remaining contamination is fairly stable. During sludge drying operations at the LWRP, however, this assumption cannot be applied, since a tractor is used to sift and turn the sludge.

If no assumptions can be made about the stability of the sludge, we may rely on occupational and public health exposure guidelines for ambient respirable dust. The standard, 0.005 g/m^3 , is a level at which prolonged exposure results in labored breathing (Ref. 3). Workers exposed at or above this level for extended periods must be issued protective equipment. In general, dust levels will be reduced during sludge spreading due to the high moisture content of the sludge: the sludge is typically dried only to 50% moisture content prior to removal to the sanitary landfill. However, using this upper bound for exposure to respirable dust will maximize the calculated dose. We therefore use 0.005 g/m^3 for the dust loading in order to derive an upper limit on worker exposure.

Given the above upper limit on respirable dust (in grams per cubic meter), we can calculate the maximum amount of plutonium activity in the air breathed by workers, simply by multiplying by the plutonium activity in the sludge (in curies per gram). To estimate the exposure time, we use work schedules provided by the LWRP supervisor on work schedules. As reported by the LWRP, sludge spreading with a tractor requires roughly 20 man-hours every other week. Total exposure time is therefore 520 man-hours each year. If we assume that a single individual carried out all the sludge drying operations during the year, these parameters are sufficient to allow a calculation of the maximum dose that might be received by an LWRP worker.

IV. Exposure Calculations

Using the above exposure parameters, we determined the maximum possible effective dose that could be received over fifty years by LWRP workers and the public. The actual calculation was performed using the GENII exposure and dose assessment code (Ref. 4). This code implements the exposure assessment criteria established by the ICRP in 1977, including transport of inhaled contamination to different tissues of the body. Recent revisions to the ICRP

recommendations, which are not incorporated in the model, would result in slightly lower estimates of the total dose.

Three different calculations were performed. First, the GENII code was run to calculate the dose that would be received by LWRP workers exposed to sludge at the maximum concentrations shown in Table I. Next, the calculated dose due to worker exposure at the average sludge concentrations was obtained by scaling by the ratio of the concentrations.

Finally, the calculated maximum dose to the public was obtained by scaling by the ratio of the exposure times, and by scaling by the ratio of airborne plutonium activity.

Using the assumptions for the dust level and the maximum sludge plutonium concentration, the airborne plutonium activity in the worker scenario is

$$0.005 \text{ g/m}^3 \times 0.23 \text{ pCi/g} \times (1 \text{ m}^3)/(10^6 \text{ mL}) = 1,200 \times 10^{-12} \text{ pCi/mL},$$

which is more than 1,600 times larger than the air concentrations shown in Table II. In making the conversion from the worker exposure to public exposure, the decreased concentration outweighs the longer exposure time: the two factors combine to yield an overall decrease in the dose by a factor of 100.

We note that, in principle, we should subtract out the contribution of global fallout (as obtained from the non-LWRP data shown in Table II) to the plutonium activity measured in air down-wind of the LWRP. This would reduce the dose by a further factor of six. In order to obtain an unquestionably conservative calculation of the maximum dose, we did not make that subtraction. Once again, this assumption leads us to over-estimate the maximum dose.

V. Results and Comparison to Regulatory Standards

DOE and proposed EPA standards limit the annual effective committed dose to, respectively, 100 mrem (DOE Order 5400.5, section IV.3.a) and 10 mrem (Ref. 5). The "committed" refers to the total dose received over 50 years due to a single years exposure to a radioisotope. The calculation of section IV shows that, in the maximum worker exposure scenario, the annual effective committed dose equivalent dose would be 0.17 mrem. The total dose received by any one individual over a fifty-year period would be 5.5 mrem (Attachment A).

From those results, the remaining scenarios were assessed by scaling of the exposure times and contamination concentrations, as discussed above. The results are presented in Table III.

Table III. Summary of calculated upper bounds for radiation exposure of the public and LWRP workers due to LLNL-originated plutonium contamination of LWRP soil and sludge. As explained in the text, doses actually received will be far lower than these calculated values. The public health exposure limits, which should be compared to the numbers in the first column, range from 10 to 100 in these units.

	Calculated Maximum Effective Dose Equivalent [mrem]	
	Annual 50-year dose commitment	Total exposure over 50 years
Public	0.0017	0.055
Workers		
Max. sludge	0.17	5.5
Ave. sludge	0.05	1.6
Exposure Standard	10-100	N/A

VI. Conclusions

LLNL concludes that residual plutonium contamination in LWRP soil and in LLNL sewage represents no significant health risk to the public. This conclusion is based upon highly health-conservative calculations of the maximum possible dose to the public and LWRP workers. Under the extreme assumptions of the model, the calculated doses are all less than 1.7% of the most restrictive (proposed) standard for protection of the public health, which limits exposure to 10 mrem per year. Actual doses received by the public and LWRP workers are lower than the calculated numbers.

REFERENCES

1. Environmental Report for 1988, 1989, and 1990, Lawrence Livermore National Laboratory publications UCRL-50027-88, 89 and 90
2. Environmental Monitoring at the Lawrence Livermore National Laboratory, Annual Report for 1979 through 1987, LLNL publications UCRL-50027-79 through -87.
3. "Particulates not otherwise regulated" in Table A-1-A of Title 29 Code of Federal Regulations, Part 1910.1000, OSHA report 3112
4. "GENII - The Hanford Environmental Radiation Dosimetry System," Napier, B.A., *et al.*, Pacific Northwest Laboratory publication PNL-6584, Vol. 1-3.
5. EPA Interim Guidance: Dose Limits For Persons Exposed to Transuranium Elements in the General Environment, March 20, 1987, page 12

ATTACHMENT A
Sample Output of the GENII Program (4 pages)
Maximized Calculation of Exposure of LWRP Workers

GENII Dose Calculation Program
(Version 1.473 27-Jun-90)

e title: LWRP Worker Exposure to Pu-containing Sludge

ecuted on: 01/13/92 at 20:11:29

Page A. 1

This is a near field (narrowly-focused, single site) scenario.
Release is chronic
Individual dose

THE FOLLOWING EXPOSURE PATHS ARE CONSIDERED:
Inhalation uptake

THE FOLLOWING TIMES ARE USED:
Intake ends after (yr): 50.0
Dose calculations ends after (yr): 50.0

===== FILENAMES AND TITLES OF FILES/LIBRARIES USED =====

Input file name: \GENII\WRPSL.in
GENII Default Parameter Values (28-Mar-90 RAP)
ADLIB - Radionuclide Master Library (8-May-90 RAP)
External Dose Factors for GENII in person Sv/yr per Bq/n (8-May-90 R
Internal Yearly Dose Increments (Sv/Bq) 29-Aug-88 RAP

Surface soil input unit: (1-m2, 2-m3, 3-kg)

-----Basic Concentrations-----					
Release	Air	Surface	Deep	Ground	Surface
Radio-		Soil	Soil	Water	Water
nuclide	pCi/L	pCi/kg	pCi/m3	pCi/L	pCi/L
PU239	0.0E+00	2.3E+02	0.0E+00	0.0E+00	0.0E+00

===== NEAR-FIELD PARAMETERS =====

24.0 Inventory disposed n years prior to beginning of intake period
0 LOIC occurred n years prior to beginning of intake period
0E+00 Fraction of roots in upper soil (top 15 cm)
0E+00 Fraction of roots in deep soil
0E+00 Manual redistribution: deep soil/surface soil dilution factor

===== WASTE FORM AVAILABILITY =====

0E+00 Waste form/package half life, yr
0E+00 Thickness of buried waste, m
0E+00 Depth of soil overburden, m

===== INHALATION =====

5.2E+02 Hours of inhalation exposure per year
1 Resuspension model: 1-Mass Loading, 2-Anspaugh
5.0E-03 Mass loading factor (g/m3)

Input prepared by:

Brian Balke

Date:

1/13/92

ut checked by: _____

Date: _____

=====

GENII Dose Calculation Program
(Version 1.473 27-Jun-90)

e title: LWRP Worker Exposure to Pu-containing Sludge

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Page C. 1

Uptake/exposure period: 50.0
Dose commitment period: 50.0
Dose units: Rem

Organ	Committed Dose Equivalent	Weighting Factors	Weighted Dose Equivalent
Gonads	2.2E-05	2.5E-01	5.5E-06
Breast	1.8E-10	1.5E-01	2.6E-11
R Marrow	1.4E-04	1.2E-01	1.7E-05
Lung	6.5E-04	1.2E-01	7.8E-05
Thyroid	1.7E-10	3.0E-02	5.1E-12
Bone Sur	1.8E-03	3.0E-02	5.3E-05
Liver	3.2E-04	6.0E-02	1.9E-05
LL Int.	5.7E-08	6.0E-02	3.4E-09
UL Int.	1.9E-08	6.0E-02	1.1E-09
S Int.	3.4E-09	6.0E-02	2.0E-10
Stomach	1.5E-09	6.0E-02	9.0E-11
Internal Effective Dose Equivalent			1.7E-04
External Dose			0.0E+00
Annual Effective Dose Equivalent			1.7E-04

Controlling Organ: Bone Sur
Controlling Pathway: Inh
Controlling Radionuclide: PU239

Total Inhalation EDE: 1.7E-04
Total Ingestion EDE: 0.0E+00

 GENII Dose Calculation Program
 (Version 1.473 27-Jun-90)

title: LWRP Worker Exposure to Pu-containing Sludge

executed on: 01/13/92 at 20:11:44

Page C. 2

intake/exposure period: 50.0
 dose commitment period: 50.0
 dose units: Rem

	Dose Commitment Year				
	1	2	3	...	
Internal :					
Intake :					
Year:	3		1.7E-05	...	
			+		
	2	1.7E-05	1.2E-05	...	Internal
		+	+		Effective
	1	1.7E-05 + 1.2E-05 + 9.0E-06 + ...	=	1.7E-04	Dose
					Equivalent
Internal					Cumulative
Annual		1.7E-05 + 2.9E-05 + 3.8E-05 + ...	=	5.5E-03	Internal
Dose		+	+	+	Dose
External					
Annual		0.0E+00	0.0E+00	0.0E+00	...
Dose					
Annual		1.7E-05 + 2.9E-05 + 3.8E-05 + ...	=	5.5E-03	Cumulative
Dose					Dose
				1.6E-04	Maximum
					Annual
					Dose Occurred
					In Year 50

Figure 1

Pu 239,240 in Air Particulate