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Environmental Monitoring at the Lawrence Livermore National Laboratory

1982 Annual Report

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Foreword

This report is prepared for the U. S. Department of Energy by the Environmental Evaluations Group of the Hazards Control Department, Lawrence Livermore National Laboratory. Data are obtained through the combined efforts of the Nuclear Chemistry Division and the Hazards Control Department. In addition to the authors listed, the following personnel made significant contributions to this report.

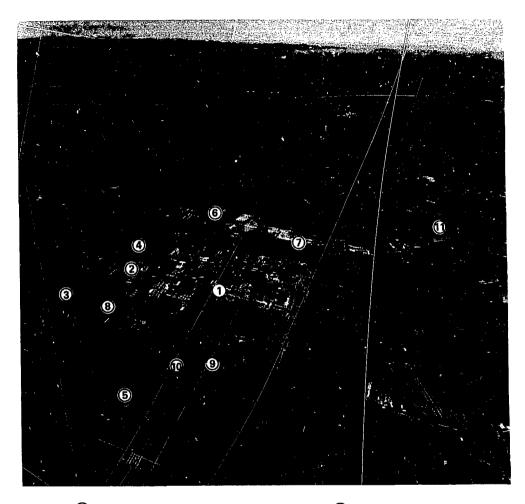
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- Sandia National Laboratories, Livermore
- South Bay Aqueduct
- Arroyo Seco
- Arroyo Las Positas
- 103466 Nearest residential area

- Vasco Road
- East Avenue
- Greenville Road
- Lupin Way
 - Interstate 580

Frontispiece. Lawrence Livermore National Laboratory (looking west).

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Introduction

The Laboratory

The Lawrence Livermore National Laboratory (LLNL) is located about 64 km east of San Francisco, Califernia, in the Livermore Valley of eastern Alameda County, approximately 5 km east of the city of Livermore. The site, which occupies an area of 2.54 km², is surrounded by open agricultural areas on the north, east, west, and part of the south side. Sandia Laboratories, Livermore, occupies a portion of the adjoining property on the south, and the nearest residential area is 0.8 km from the Laboratory's west perimeter. Of the nearly 4.8 million people who live within 80 km of the Laboratory (Fig. 1), 50,000 live in Livermore.

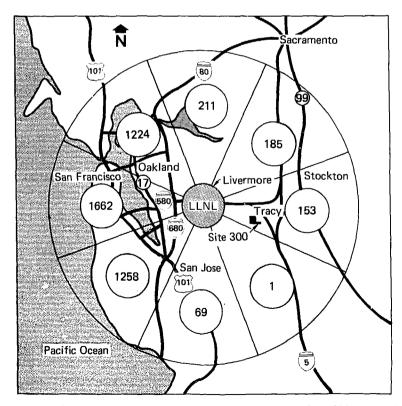


Figure 1. Estimated population distribution (in thousands) within 80 km of Livermore, by sectors.

The Laboratory, operated by the University of California for the United States Department of Energy, was established in 1952 to conduct nuclear weapons and controlled thermonuclear reaction research. Today, about 60% of its resources are devoted to defense programs, which include weapons and laser fusion research. Other major areas of focus are magnetic fusion energy; laser isotope separation; chemistry, engineering, and physics research; biomedical and environmental sciences; applied energy technology; and work for other federal agencies.

In 1982 the Laboratory employed approximately 7400 people.

Much of the Laboratory's materials testing and high-explosives diagnostic work is conducted at Site 300, 16 km southeast of Livermore. Located in the sparsely populated hills of the Diablo Range, Site 300 covers an area of 27 km². Figure 1 shows the location of LLNL and Site 300 with respect to the city of Livermore and surrounding areas.

The Site

The Livermore Valley has a climate characterized by mild, rainy winters and warm, dry summers. The highest and lowest annual rainfalls on record were 728 mm and 138 mm, with a 112-year average of 365 mm. Rainfall for the 1981-1982 water year (October 1, 1981, through September 30, 1982) was 538 mm. Surface water drainage from the Valley is from east to west through various arroyos; the outfall is near Sunol in the southwest corner of the Valley. Prevailing winds are from the west and southwest during April through September. During the remainder of the year, wind directions are variable, as shown by the wind rose in Fig. 2.

The Livermore site is situated on a northwesterly sloping alluvial flood plain bordering the low hills of the Livermore Uplands to the south. Groundwater is found at depths of 15 to 30 m below the LLNL site with a gradient indicating a generally westward flow. The lithology of the area consists of a series of unconsolidated marine and continental sedimentary units such as sandstones, gravels, silts, and clays overlying the interbedded sandstones of the Franciscan Formation. The hilly terrain surrounding the Valley is used for cattle and sheep pasture, and the principal agricultural products in the vicinity of LLNL are grapes and wine, cattle, and poultry. Water bodies adjacent to the Laboratory include the South Bay Aqueduct, which runs from the northeast to southwest, 1.8 km to the southeast; the Patterson Pass water treatment facility, about 2 km east of LLNL; and Frick Lake, 4 km north of LLNL, a sag pond that is dry most of the year. Aquatic recreation (boating, fishing, and swimming) is available at Lake Del Valle, about 8 km south of LLNL, and at the Shadow Cliffs Recreation Area, 11 km to the west.

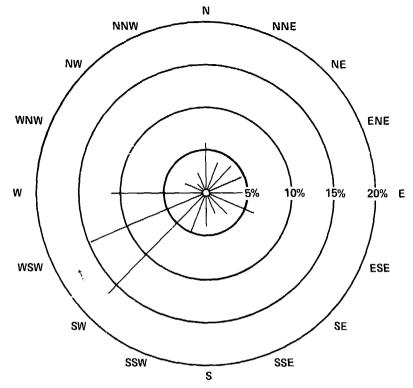
The Laboratory normally receives its treated water from the Hetch Hetchy Aqueduct (which supplies San Francisco), located 11 km southwest of Livermore.

Laboratory storm water is channeled through storm sewers designed to accommodate a 10-year flow. Open ditches are used in undeveloped areas of the site. Arroyo Seco crosses LLNL at the southwest corner. Arroyo Las Positas originally crossed the northeast section of the site. However, in 1965, as part of an erosion-control program, Arroyo Las Positas was channeled north to the northeast corner of the site, and then west along the north perimeter to an outlet at the northwest corner. This outlet, which also constitutes the main pathway for the Laboratory's surface drainage (storm and irrigation), runs north to the Western Pacific tracks, then west where it joins Arroyo Seco. The LLNL Master Site Plan calls for a small lake to be established in the center of the project. Provisions have been made for rerouting on-site water drainage and the Arroyo Las Positas to fill this lake during the rainy season.

Laboratory sewage is discharged into the City of Livermore's sanitary sewer system and processed at the Livermore Water Reclamation Plant (LWRP). As part of the Livermore-Arr ador Valley Wastewater Management program, the treated sanitary wastewater is transported out of the valley via a pipeline and discharged into the San Francisco Bay. The LWRP was connected to this pipeline on February 8, 1980. While the LWRP effluent is still used for summer irrigation of nearby Livermore city property, it is no longer discharged to Arroyo Las Positas, as was done during the wet season before construction of the pipeline.

Environmental Monitoring

A strict effluent-control program that emphasizes controlling effluents at the source has been in effect since the Laboratory began operation.



Average annual percent frequency of wind direction vs wind speed

					Spec	d (m/s)					
Direction	0-2	2-3	3-5	5-7	7-9	9-11	11-16	16-20	>20	Total	Average
N	2.1	1.0	1.7	0.5	0.3	0.1	0	0	0	5.6	3.1
NNE	1.2	0.8	1.4	0.2	0.1	0	0	0	0	3.8	3.0
NE	1.4	1.1	1.4	0.3	0	0	0	0	0	4.2	2.8
ENE	2.2	1.1	1.0	0.2	0	0	0	0	0	4.5	2.4
E	5.1	1.1	0.4	0	0	0	0	0	0	6.6	1.6
ESE	5.0	0.7	0.3	0	0	G	0	0	0	6.0	1.5
SE	2.9	0.3	0.2	0	0	0	0	0	0	3.4	1.4
SSE	2.0	0.2	0.2	0.1	0	0	0	r	0	2.5	1.6
S	2.7	0.5	0.4	0.1	0	0	0		0	3.8	1.8
SSW	2.2	1.0	1.1	0.4	0.1	0	0	0	0	5.0	2.9
SW	2.0	2.7	7.4	3.7	0.5	0.1	0	0	0	16.4	4.1
WSW	2.3	3.2	6.7	2,3	0.3	0	0	0	0	14.8	3.6
w	1.7	1.8	4.7	2.6	0.2	0	0	0	0	11.0	3.9
WNW	1.0	0.4	0.8	0.5	0	0	0	0	0	2.7	3.1
NW	1.2	0.1	0.1	0	0	0	0	0	0	1.4	1.4
NNW	1.5	0.4	0.3	0.2	0.1	0	0	0	0	2.5	2.4
Calm										5.9	
Total	36.4	16.5	28.0	11.3	1.6	0.3	0.1	0	0	100.0	2.8

Figure 2. Wind rose showing average annual wind direction and speed during 1982 (measurements are made at 10 m above ground).

The environmental monitoring program is maintained to evaluate the effectiveness of these measures, to document whether effluents from the Laboratory and Site 300 operations are within applicable standards, and to estimate the impact of these operations on the environment. Sensitive monitoring equipment is used that can detect radioactive and nonradioactive pollutants at environmental background levels. The program includes the collection and analysis of air, soil, water, sewer effluent, vegetation, foodstuffs, and milk samples. Also, environmental background radiation is measured at numerous locations in the vicinity of the Laboratory using gamma and neutron dosimeters.

Each spring, the Laboratory reports the results of environmental monitoring for the previous year, noting significant changes in either the scope of the program or the levels of effluents. This report is prepared in compliance with the DOE Manual 5484.1, *Environmental Protection*,

Safety, and Health Protection Information Reporting Requirements.

Appendix A is a tabulation of 1982 environmental monitoring data. Graphics have been used in the body of the report to aid in interpretation. When appropriate, the tabulated data contain maximum, minimum, and average values. Radioactivity values are tabulated with the associated counting uncertainties at the 2σ (95% confidence) level. Unless otherwise stated, the minimum detection limit of these measurements is assumed to have been reached when the 2σ error is $\pm 100\%$. In the case of radioactivity measurements, an attempt has been made to assess the potential impact on man of the observed environmental levels of artificially produced radionuclides. This assessment is made by calculating the whole-body or critical-organ doses delivered to man and comparing these with the much larger radiation dose received locally from natural sources.

Summary

Air Samples

In 1982, the annual average airborne gross beta activity in Livermore Valley air samples was $1.9 \times 10^{-14} \,\mu \text{Ci/ml}$, which was less than the level observed in 1981. Airborne 238U concentrations at Site 300 were higher than those at Livermore because of the "depleted" uranium (a byproduct of ²³⁵U enrichment) used in high-explosive tests at the Site. However, these concentrations were well below the standards set by DOE. The average gross alpha activity shows no significant change from 1981 data. The average annual beryllium concentrations were less than 1% of the local air pollution standard at both the Laboratory perimeter and Site 300. The beryllium concentrations can be accounted for by the natural beryllium in typical airborne dust. The total tritium released to the atmosphere in 1982 was 2014 Ci, the lowest since 1974.

Water Samples

Water samples collected in the Livermore Valley and at Site 300 exhibit gross beta and tritium activities within the ranges previously observed in these areas. Two water samples from Site 300 showed an above-average level of gross alpha activity. This activity has been found to be naturally occurring uranium at concentrations well within those specified in DOE Manual 5480.1.

The discharge of tritiated water (HTO) into the sanitary sewer system in 1982 was 1.6 Ci, which was the lowest since 1972. Tritium analyses were also made on well-water samples collected near the Livermore Water Reclamation Plant (LWRP). These analyses, begun in 1977, were made to determine the extent to which low levels of tritium in the LWRP effluent may be migrating into groundwater. As was found during earlier surveys, the highest tritium values were detected in test wells west of the site near Arroyo Las Positas. Since the LWRP effluent is no longer discharged to the arroyo, the primary means of tritium movement to nearby groundwater has been eliminated. Consequently, tritium concentrations observed in local wells in 1982 were generally lower than previous annual measurements.

Vegetation and Foodstuff Samples

Tritium levels found in the Livermore Valley and Site 300 vegetation were comparable to those observed in 1981. As a means of evaluating the possible impact of Laboratory effluents on locally grown foodstuff, the tritium content of Livermore Valley wines was compared with values from other California and European wines. The tritium levels in Valley wines are within the range found to be present in both European wines and surface waters throughout the world, but somewhat higher than those in other California wines. Honey produced in the Livermore Valley contained tritium levels comparable to those found in honey from neighboring areas.

Doses

The 1982 median annual gamma-radiation dose at the Laboratory perimeter was 49 mrem, and the median off-site background dose was 50 mrem, less than the 1981 levels of 54 mrem and 57 mrem. A 14-MeV neutron generator (Bldg. 212) near the south perimeter continued to be a source of elevated radiation. However, the annual radiation dose at the Laboratory's south perimeter is within allowable standards in DOE Manual 5480.1.

Monitoring Data-Collection, Analysis, and Evaluation

Radioactive Monitoring

Airborne Radioactivity

Concentrations of various airborne radionuclides are measured at the Laboratory perimeter, off-site locations near the Laboratory, and Site 300. Sampling locations are shown in Figs. 3 through 14. The six samplers on the Laboratory perimeter (Fig. 3) and the nine samplers at Site 300 (Fig. 4) use 5.2×10^{-2} m² Whatman-41 cellulose filters. These samplers are operated at average flow rates of 400 to 700 liters/min. The filters

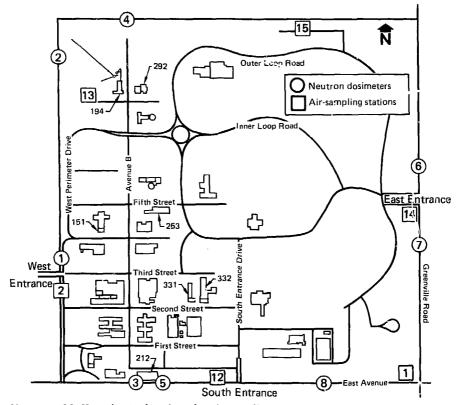


Figure 3. LLNL perimeter locations for air-sampling stations and neutron dosimeters.

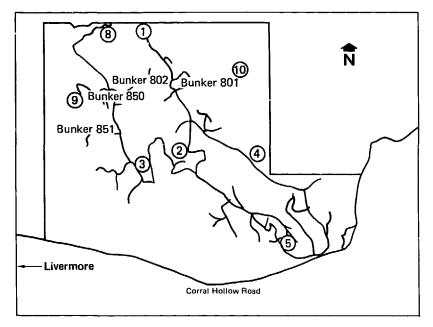


Figure 4. Site 300 on-site air-sampling locations.

are cut in half; one half of each filter is retained for berylltern analysis. An easily dissolved filter and a low trace-metal background are required for these analyses. Whatman 41 represents a balance between such requirements and particulatecollection efficiency.¹ Off-site samplers throughout the Livermore Valley (Fig. 5) use 4.56 \times 10^{-3} m² glass-fiber filters (Flanders F-700) and are operated at a flov rate of 80 liters/min. All air filters are changed weekly.

After a four-day delay for decay of the radon-thoron daughters, gross alpha and beta activities on the filters are determined with an automatic gas-flow proportional counter. Monthly composites of Laboratory perimeter and Site 300 filters are also counted for specific gamma-emitting radionuclides using low-background Ge(Li) detectors. Following gamma counting the Laboratory perimeter filters from in-dividue. locations and the Site 300 composites are analyzed for the presence of ²³⁹Pu, ²³⁸Pu, ²³⁵U, and ²³⁸U.

Tables 1 and 2 in Appendix A show airborne gross alpha and gross beta activities for the Livermore Valley samples, and Tables 3 and 4 show corresponding activities for Site 300. Average annual gross beta activities at Livermore and Site 300 were reduced by 80 and 85% from those observed in 1981. This decrease is attributed to reduction of the fallout from a nuclear test by the People's Republic of China in 1980. The average gross alpha activity showed no significant change.

Tables 5 and 6 list the activities of the principal radionuclides that contribute to the gross beta activity in Livermore and Site 300 samples. These activities are determined by gamma spectra analysis. The fission products were also reduced by 85-95% from the previous year. Table 7 shows the concentration of plutonium on air filter samples collected in Tracy and the Livermore Valley. Tables 8 and 9 show the concentrations of airborne ²³⁹Pu, ²³⁵U, and ²³⁸U in the Livermore perimeter and Site 300 air samples. The higher concentration of ²³⁸U at Site 300 is a result of the "depleted" uranium (a byproduct of ²³⁵U enrichment) used at the Site. The uranium concentrations are well below the concentration guides (CG) in DOE Manual 5480.1 (see Appendix B).

Concentrations of airborne tritiated water (HTO) were determined for each of the LLNL perimeter air-sampling locations (Fig. 3) and at off-site locations S1 and S2 (Fig. 5). Water vapor was collected on silica-gel samplers that operated at a flow rate of about 0.5 liters/min for a 2-week period. The collected water was recovered by vacuum drying the silica gel at 150°C, and the HTO was measured by liquid-scintillation counting. Table 10 shows average monthly HTO concentrations observed at each sampling location; the overall annual average concentration is $3.4 \times 10^{-11} \,\mu$ Ci/ml. The highest monthly concentration occurred in March and was observed at location 12. This concentration, $20 \times 10^{-11} \,\mu$ Ci/ml, is 0.1% of the DOE standard. Location 12 also showed the highest annual average concentration, probably because this location is so close to one of the tritium effluent sources (the 14-MeV neutron generator).

Radioactivity in Soil

An intensive soil-sampling program conducted in 1971 and 1972 provided a data base on the concentration ranges of various radionuclides in soil near the Laboratory and at Site 300. Since 1972, soil sampling in the vicinity of LLNL and at Site 300 has been part of a continuing LLNL surveillance program to document any changes in environmental levels of radioactivity that may have occurred and to evaluate any increase in radioactivity that might have resulted from Laboratory operations. As in previous years, a group of soil samples was collected at random from the listing of locations sampled in the 1971-1972 study. Figure 6 shows the 1982 soil-sampling locations in the Livermore Valley and Fig. 7 indicates the locations at Site 300 where annual samples have been collected since 1973.

All samples were collected to a depth of 5 cm. After collection, the samples were dried, ground, and blended. Plutonium-239 content was determined in each sample following an acid-leaching procedure. For gamma-spectra analysis, approximately 300-g aliquots of soil were sealed in 200-cm³ thin-walled cans of aluminum and counted with a Ge(Li) detector.

The data in Table 11 are in the range of those reported in the previous year.² Figure 8 is a distribution plot of these activities.

Table 12 shows the ²³⁹Pu, ¹³⁷Cs, and ²³⁸U activities in the Site 300 samples. There were negligible changes from activities observed in previous samples collected in the same locations.²

High-explosive tests at Site 300 often involve the use of depleted uranium. Accordingly, soil samples are taken annually to determine how these tests perturb the ²³⁵U/²³⁸U ratio of the soil. Isotopic uranium measurements were made with isotopic-dilution mass spectrometry. As in the past, the analyses indicated that isotopic perturbation is essentially limited to areas adjacent to the firing bunkers.^{3,4} The isotopic uranium data shown in Table 12 are comparable to those observed during 1981.

Radioactivity in Sewage

Radioactive wastewater is treated to reduce activity levels to the lowest levels practicable and well below standards in DOE Manual 5480.1. After treatment and analysis, the liquid effluents are released into the city of Livermore's sanitary

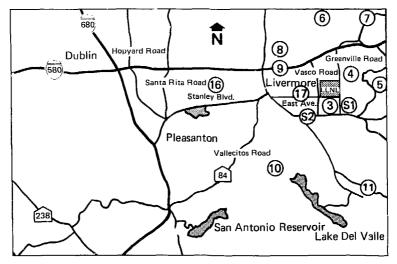


Figure 5. Livermore Valley air-sampling locations.

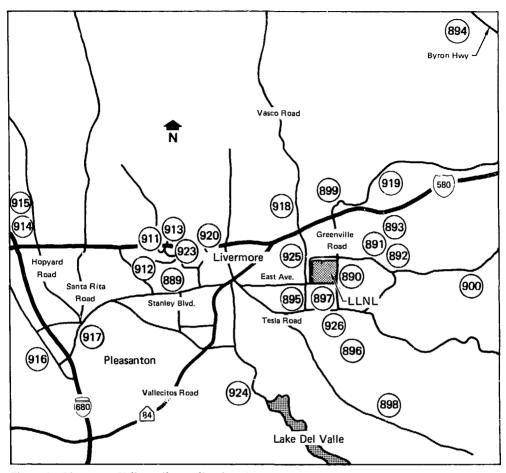


Figure 6. Livermore Valley soil-sampling locations.

sewer system at the outfall located at the northwest corner of the LLNL. The effluent is continuously monitored at the outfall for pH and radioactivity.

Liquid wastes from Livermore's sanitary sewer system are treated at the Livermore Water Reclamation Plant (LWRP), a tertiary sewagetreatment plant that serves residential, commercial, and industrial water users in Livermore. The salt content of the LWRP effluent presented a problem for groundwater quality locally and in the Niles Cone aquifers. Accordingly, as part of the Livermore-Amador Valley Wastewater Management program, a pipeline was constructed to transport wastewater out of the valley for discharge into the San Francisco Bay. The Livermore sewage plant was connected to this pipeline on February 8, 1980. Although the effluent will still be used for irrigation of municipal property during summer months, it will no longer be discharged into the arroyo, thus eliminating the primary means of tritium migration to local and downstream groundwater.

Representative San Francisco Bay water samples were collected before and after the LWRP's connection to the pipeline. Results of tritium analysis of these samples indicate no significant difference between the two sample sets.⁵

Samples of the LLNL and LWRP liquid effluents are collected daily. Table 13 compares the monthly composite activity levels of certain radionuclides in the LLNL effluent with those in

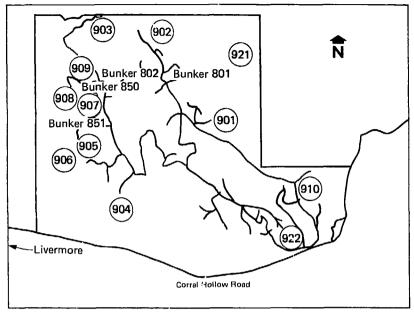


Figure 7. Site 300 soil-sampling locations.

the effluent from the LWRP. All concentrations are well below standards in DOE Manual 5480.1 for discharge into the sanitary sewer system.

Radioactivity in Water

Water samples are collected from the various Livermore Valley and Site 300 locations shown in

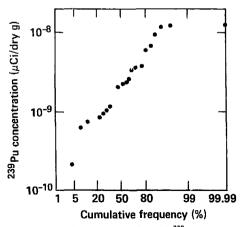


Figure 8. Distribution plot of ²³⁹Pu in soil samples collected in the Livermore Valley during 1982.

Figs. 9 and 10. These samples are analyzed for gross alpha and gross beta activity. This year a hydrogeologic investigation evaluated the effects that LLNL operations have on local groundwater. This study included a variety of analyses of samples taken from shallow water tables beneath the site: determinations of major cations and anions, total dissolved solids, total organic carbon, trace organic compounds, gamma spectra, and tritium. No concentration of contaminants was detected that would preclude the use of this water for drinking.⁶ Tables 14 and 15 show the gross alpha activities in Livermore Valley and Site 300 samples, respectively. Gross beta activities for Livermore and Site 300 samples are shown in Tables 16 and 17.

Livermore sampling locations 11, 16, 24, 26, and 29 (Fig. 9) are surface water sources (ponds, creeks, and reservoirs), and location 20 is the collection site of LLNL rainfall. The other locations are dornestic water sources. Gross alpha and beta activities in Livermore water samples collected in 1981 were below EPA and State of California standards for drinking water.⁷

Storm-drain samples taken during the rain season were analyzed for gross alpha, gross beta, and tritium. The highest concentrations of alpha and beta were 15% and 4% of the concentration

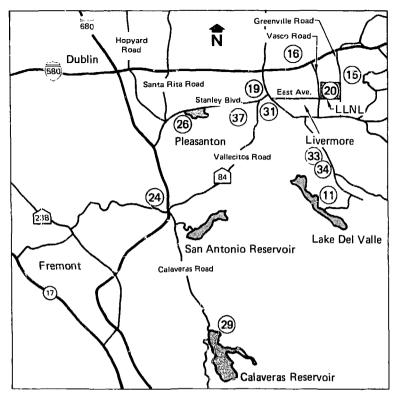


Figure 9. Livermore Valley water-sampling locations.

guide; the tritium concentration in the effluent was less than 0.2% of the concentration guide.

Site 300 water samples (Fig. 10) are collected from on-site wells (locations 1-6 and 22), off-site wells (locations 7, 11, and 23), and an off-site creek (location 14). Location 20 is Site 300 rainwater, and location 21 is a spring-fed pond near Bunker 812. Three locations showed high gross alpha concentrations which specific analyses have identified tc be natural uranium: location 4, a well near the Site 300 entrance, which has displayed these results in previous years; location 20, the March rain water; and location 21, the spring-fed pond, which was investigated during 1981. Neither the EPA⁷ nor the State of California⁸ specifies a limit for natural uranium in drinking water. In their limit for gross alpha activity, uranium is specifically excluded from the total. The uranium concentrations were well within that specified in DOE Manual 5480.1 (2 \times 10⁻⁵ μ Ci/ml).

The water samples from the Livermore Valley and Site 300 are also analyzed for tritium activity. Because of the low tritium levels typically found it is necessary to distill and electrolytically enrich the samples before liquid-scintillation counting. Tables 18 and 19 show the data for the Livermore and Site 300 samples, respectively. The samples have concentrations that are well below recommended concentration guide values. The maximum concentration of tritium in a rainwater sample was 19.6 $\times 10^{-7} \,\mu$ Ci/ml, which is 0.04% of the concentration guide for drinking water. Tables 18 and 19 also include an estimate of the annual dose that may be delivered to an adult who consumes water containing the listed tritium concentrations. These doses, which are all less than 0.2 mrem, are based on a water consumption of 2 liters/day and the dose-conversion factors contained in NRC Reg. Guide 1.109 (see Appendix C).

As noted previously, treated effluent from the LWRP is used to irrigate nearby municipal property. This effluent contains low levels of tritium which come from normal LLNL operational releases to the sanitary sewer system. As part of a

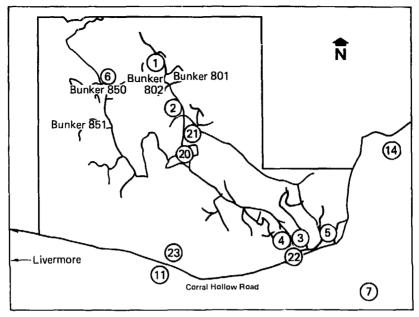


Figure 10. Site 300 water-sampling locations.

study begun in 1977,⁹ tritium measurements are now made annually on well-water samples collected from neighboring wells to determine the extent of tritium migration into groundwater. Many of the wells were in the immediate vicinity of the LWRP; however, additional samples were also collected from areas at some distance. Locations sampled during 1982 are indicated in Fig. 11,

and the tritium data are shown in Table 20. The highest tritium values appear in wells west of the LWRP. Tritium activities in all samples were well below the guidelines for water in uncontrolled areas as stated in DOE Manual 5480.1. The tritium levels in these wells are also lower than those observed in the 1981 samples, which is to be expected with diversion of effluent to the pipeline.

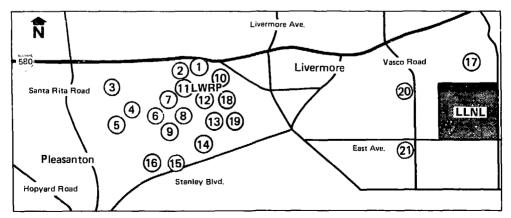


Figure 11. Livermore Valley groundwater-sampling locations.

Radioactivity in Vegetation and Foodstuffs

Vegetation samples (usually native grasses) are collected quarterly at locations in the Livermore Valley (Fig. 12) and at Site 300 (Fig. 13). These samples are freeze-dried and the tritium activity in the recovered water is determined by liquid-scintillation counting. Table 21 shows the tritium data for vegetation collected in the Livermore Valley. The whole-body radiation doses shown in Table 21 were derived using the doseconversion factors in NRC Reg. Guide, 1.109.¹⁰ Conservatively assuming that an adult's diet comprises vegetables with similar tritium concentrations and meat and milk derived from livestock fed on grasses of these concentrations, these potential doses are all less than 0.1 mrem/y.

Table 22 shows the 1982 tritium data for Site 300 vegetation. Location 6 is adjacent to an area that contains tritium-contaminated debris from a firing table. As a result of the seasonal rains, the tritium apparently entered an aquifer whose outflow is in the area where location 13 samples are routinely collected. This was also observed in 1981.

Beginning in 1977, as a means of evaluating the possible impact of Laboratory effluents on locally grown foodstuff, the tritium content of Livermore Valley wines was measured and compared with that found in other California wines and European wines.⁹ Wine samples collected in 1982 were catalytically oxidized to carbon dioxide and water, and the tritium content of the recovered water was measured by gas proportional counting. The data in Table 23 represent the analysis of samples purchased in 1982. As found since 1977, the tritium levels of the Valley wines were somewhat higher than those of California wines produced from grapes grown outside the Valley, but lower than those of the European wines sampled. The same European wines have been sampled in previous years.

Samples of honey produced from a variety of flower sources both in and outside the Livermore Valley were analyzed for tritium content. Following the oxidation of the samples in a Parr oxygen bomb, the tritium content of the water produced was determined by gas proportional counting. The data in Table 24 show that the tritium content of Livermore Valley honey samples is comparable to that of honey from neighboring areas.

Radioactivity in Milk

During 1982, goat-milk samples were obtained from three farms within about 5 km of the Laboratory. A portion of each sample was vacuum-distilled and the distillate analyzed for

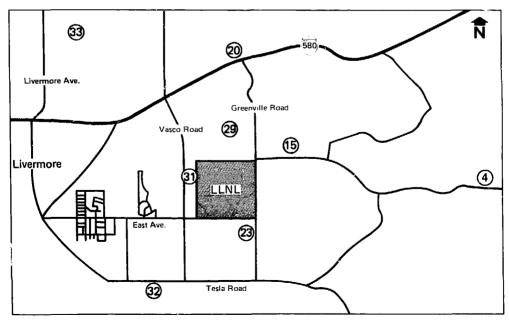


Figure 12. Livermore Valley vegetation-sampling locations.

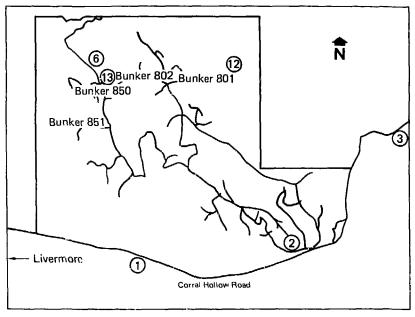


Figure 13. Site 300 vegetation-sampling locations.

tritium activity by liquid-scintillation counting. Another portion was oven-dried, and the residue blended and gamma counted in a Ge(Li) system. The activities of the ¹³⁷Cs, ⁴⁰K, and ³H in the samples are shown in Table 25. The annual average concentration of tritium in milk was 3.0×10^{-7} µCi/ml. Also shown are the calculated annual whole-body or critical-organ radiation doses that could be received from consuming this milk. These calculations are based on a milk intake of 310 liters/y and on the models previously referenced. The only dose above 1 mrem to an individual is from naturally occurring ⁴⁰K. The dose due to ⁴⁰K was calculated using dose factors from *ICRP* 30.¹¹

Environmental Radiation Measurements

Quarterly, environmental radiation is measured at the 22 LLNL perimeter locations (Fig. 14) and at the 47 off-site locations (Fig. 15). These measurements are obtained with thermoluminescence dosimeters using a previously published procedure.¹² Based on past measurements, ¹³ environmental terrestrial exposure rates in the Livermore Valley vary from 30 to 60 mR/y. Cosmic radiation, calculated from the local elevation and geomagnetic latitude according to the data of Lowder and Beck,¹⁴ is approximately 35 mR/y.

Table 26 shows the quarterly and annual radiation doses for the perimeter locations. The median annual dose of 49 mrem at the perimeter does not differ significantly from the 50 mrem offsite average. Both show decreases from the previous year's values of 54 and 57 mrem.

Figure 16 presents the 1982 annual frequency distribution of environmental dose rates observed at the 47 off-site locations. The dosimeter that recorded the highest dose (97 mrem) was near an off-site industrial plant where radiography is frequently performed. Figure 17 is a dosedistribution plot combining Laboratory perimeter and off-site measurements.

Environmental neutron dose rate measurements using ²³⁵U track-etch detectors are also made at eight locations on the LLNL site perimeter. A detailed description of the detector and the spark-counting procedure has been published.¹⁵ Neutron-monitoring locations are shown in Fig. 3 and 1982 quarterly measurements are shown in Table 27. Location 3 received the highest annual neutron dose, 36 mrem, because of its position near Bldg, 212, the 14-MeV neutron generator.

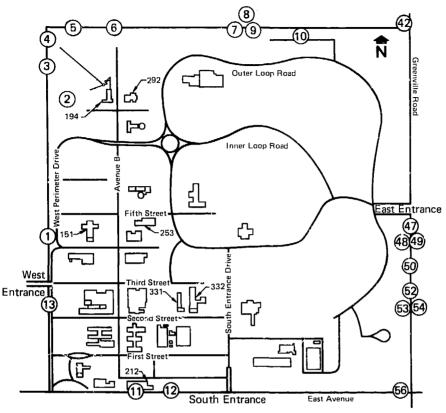


Figure 14. LLNL perimeter locations for gamma dosimeters.

Nonradioactive Monitoring

Beryllium in Air and Water

Beryllium monitoring of air, both at the source and near LLNL property boundaries. has always been a part of the Laboratory's safety program. Monthly, halves of the filters from the LLNL perimeter and Site 300 are composited by sampling location. Following sample preparation the beryllium content of the solutions is determined by atomic absorption analysis.

Tables 28 and 29 show average monthly concentrations of airborne beryllium for LLNL perimeter and Site 300 sampling locations, respectively. The concentrations, which average less than 1% of the ambient concentration limit established by the Bay Area Air Quality Management District, can be accounted for by resuspension of surface soil containing naturally occurring beryllium. Local soils contain approximately 1 ppm of beryllium, and Livermore's air typically contains 10–100 μ g of particulates per cubic meter. By using a value of 50 μ g/m³ for an average dust load and 1 ppm for beryllium content of this dust, an airborne beryllium concentration of 5.0 \times 10⁻⁵ μ g/m³ can be calculated. This value is in good agreement with the data in Tables 28 and 29. These concentrations are plotted in Figs. 18 and 19.

In compliance with the California Regional Water Quality Control Board (WQCB) Order 80-184, we began monthly sampling for beryllium (in addition to gross alpha, gross beta, and tritium) at Site 300 well-water monitoring locations and in surface water runoff at landfills during the rain season. The highest concentration of beryllium for the monitoring locations was less than 10 parts per billion (ppb); the highest runoff was 28 ppb. Neither of these values indicate beryllium contamination from landfills.

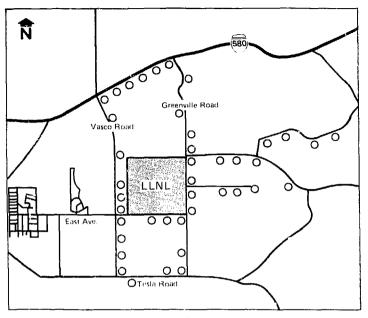


Figure 15. LLNL off-site locations for gamma dosimeters.

Nonradioactive Pollutants in Sewage

As noted previously, sanitary sewage from the Laboratory is treated at the LWRP. This effluent is continuously monitored for pH and radioactivity before it enters the Livermore sewer system.¹⁶ Sewage samples representative of daily flow are collected and composited monthly. These

50 Frequency (%) Median 40 dose 50 mrem 30 20 10 0 35 40 45 50 55 60 95 100 Annual radiation dose (mrem)

Figure 16. Annual off-site gamma radiation background for 1982.

composites are analyzed for the metals shown in Table 30.

In addition, samples of Laboratory sewage effluent are collected quarterly. These samples are analyzed for the parameters specified on LWRP's permit (National Permit Discharge Elimination System). Table 31 shows the data for 1982. All data demonstrate compliance with the City of Livermore's discharge limits listed in Appendix D.

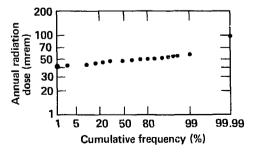


Figure 17. TLD dose-distribution plot for the LLNL perimeter and Livermore Valley during 1982.

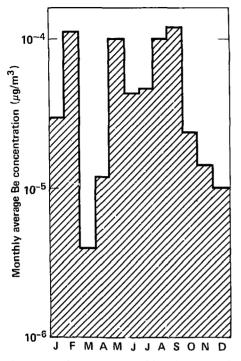


Figure 18. Concentration of beryllium on air filters at the LLNL perimeter.

Noise Pollution

As noted elsewhere, the Laboratory's highexplosive diagnostic work is conducted at Site 300. Because Site 300 is so remote, these experiments can be performed with minimal off-site impact from annoying noises or damaging overpressures. On the basis of meteorological measurements made twice each day, a limit is set on the weight of high explosives that can be detonated without impact in populated areas. To monitor these limits, four microbarograph sensors are maintained in or near the city of Tracy. The probability of overpressure is greatest in the Tracy area because of the direction of the prevailing winds. There were no complaints of overpressures attributed to LLNL operations at Site 300 during 1982.

Pollutants in Storm Runoff and Liquid-Discharge Sites

Beginning in 1975, the Laboratory's environmental surveillance program was expanded to in-

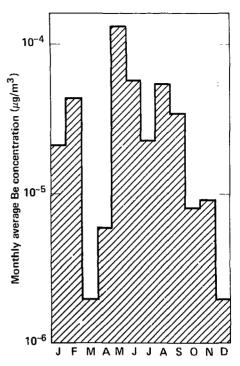


Figure 19. Concentration of beryllium on air filters at Site 300.

clude pesticide monitoring. Pesticides used at LLNL include herbicides, fungicides, and insecticides. The most probable way pesticides used at LLNL could be transported to the off-site environment is by entrainment in surface runoff water. Most of this surface drainage leaves the Laboratory via a ditch at the northwest corner of the property. A sample was collected from the ditch following the first major storm.

This sample was extracted with organic solvents, and the extracts were analyzed by gas-liquid chromatography (GLC) using a variety of detectors. Data obtained from these analyses were compared with the pesticides listed in Title 22 of the California Health and Safety Code.⁸ No materials were detected in the samples at concentrations exceeding the State-adopted standards for these organic chemicals.

Order 80-183 from the Water Quality Control Board requires the monthly sampling and analyses of wells which are down gradient from Site 300 liquid-discharge sites (locations 3, 4, 5, and 14). Any increase in total organic carbon, specific conductance, chlorides, or total hardness could indicate percolation of the liquid waste. The data shown in Table 32 do not indicate contamination.

Environmental Impact of LLNL Operations

Radioactive Airborne Effluents

In 1982 (see Table 33), radioactive airborne effluents consisted of a total of 2019 Ci of tritium from Bldg. 212 (14-MeV neutron generator), Bldg. 331 (tritium facility), and Bldg. 292 (rotating target neutron source), and 584 Ci of the short-lived radionuclides ${}^{15}O_2{}^{-13}N_2$ from Bldg. 194 (electron-positron linear accelerator). Comparative releases of radioactive effluents at Livermore during the 7-year period 1976 through 1982 are shown in Table 34. The Livermore reactor was shut down on March 31, 1980, because of a lack of programmatic need. Closure of this facility removed the local source of 41 Ar as a contributor to the site radiation dose to the public.

Table 33 also contains estimated radiation doses to the public from these radioactive airborne effluents. Three dose-reference points were used: (1) the "fence-post" dose at that location on the site boundary where maximum exposure rates exist, (2) the dose to the nearest resident, and (3) the man-rem dose within a radius of 80 km.

Dose calculations were made using a continuous-point-source computer code based on the Gaussian plume model.¹⁷ This code provides ratios of concentration to release rate (χ/Q) through sixteen 22.5° compass sectors, and distances from 0.1 to 100 km from potential release points. The average annual χ/Q values have been calculated using local meteorological data from an instrumented tower. This tower, located near the Laboratory's east site boundary, is equipped with sensors mounted at 10- and 40-m levels that measure wind direction, wind speed, and temperature. From records of these data, wind speed, wind direction, and atmospheric stability estimates were tabulated at 1/4-h intervals over the calendar year. Variance in the horizontal wind direction was used to estimate Pasquill-Gifford stability categories based on the method described by Slade.¹⁸ Lateral and vertical standard deviations, σ_v and $\sigma_{z'}$ are entered in the computer code as functions of these stability categories and the respective distances. From annual effluent data the release rate Q (in curies per second) was calculated for each of the principal radionuclides released to the atmosphere, and the concentrations at the site boundary and the nearest resident were calculated from appropriate χ/Q values. "Nearest resident" means that resident receiving the highest dose from each radioactivity release point, not necessarily the resident nearest to the site boundary. Dose estimates were based on the dose-conversion factor in the NRC Regulatory Guide 1.109.¹⁰ The results indicate that the maximum estimated dose to the nearest resident was less than 1 mrem.

Table 33 shows a combined population dose of less than 1 man-rem from ³H and neutron activation products in Laboratory airborne effluents. This dose is based on a population of 4.8×10^6 within 80 km of the Laboratory. Using 100 mrem/y as a typical average radiation dose from natural sources, the comparable natural radiation dose the same group receives is 4.8×10^5 man-rem. By comparison, the population dose from Laboratory operations is negligible.

Radioactive Liquid Effluents

Low-level radioactive wastewater is treated to reduce radioactivity to levels as low as reasonably achievable and well within applicable health and safety standards. The treated effluent is then discharged into Livermore's sanitary sewer system. During 1982 the quantity of principal radionuclides, 0.43×10^{-5} Ci $^{23^{\circ}}$ Pu and 1.6 Ci HTO, released into the sewer represented reductions of 95 and 73% from corresponding concentrations reported for 1981. Table 13 shows that the average annual concentration of these radionuclides represents 1.2×10^{-5} % and 5×10^{-3} % of the DOE standards.

Nonradioactive Liquid Effluents

The metals in sewage are listed in Table 30. The quantities of metals discharged did not prevent the LWRP from discharging effluents within limits set by the WQCB.

The first rains of the season, which were collected as storm drain runoff, showed no detectable level of specified total identifiable chlorinated hydrocarbons, and the concentrations of substances in the runoff were generally lower than in the influent. These data are displayed in Table 35.

Special Monitoring Assessments

In September 1982, a radiological survey of a former waste-storage area found soil contaminated with low-level radioactivity and organic solvents. Four separate pits were identified when 6 ft of soil was scraped away in 6-in. increments, followed by monitoring with a Field Instrument for Detection of Low-Level Radiation (FIDLER).

The California Regional Water Quality Control Board, Bay Region, and the California Department of Health Services, Hazardous Waste Mar.agement Branch, were informed of the situation end the corrective actions which LLNL had taken: soil was removed until radioactivity measu ements were equal to or less than twice backg ound, and organic solvents (principally 1,1,1,-trichloroethane, tetrachloroethylene, and trichloroethylene) were removed until their concentrations in soil were near or below the detection limit. Selected soil samples were submitted to an independent laboratory for analyses by EPA method 624 to ensure that clean-up was to acceptable concentrations.

Approximately 3000 cubic yards of soil were removed for disposal at a Class I landfill for organic solvents and at Nevada Test Site.¹⁹

At Site 300, three areas were found where trichloroethylene (TCE), used as a heat exchanger fluid, was released to the ground.

Of the seven on-site wells, one off-site well, and four on-site springs sampled monthly, partper-billion concentrations of TCE have been found in one on-site well and one spring downstream from the three contaminated areas. LLNL, with review by the DOE and the California Water Quality Control Board, Central Valley Region, has prepared an assessment plan to determine the ex-

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tent of contamination, evaluate consequences of the contamination, and undertake remedial action. 20

Quality Assurance

During 1982, the Laboratory participated in the Environmental Protection Agency's Environmental Radioactivity Laboratory Intercomparison Studies program.

Table 36 shows the comparison of analyses from LLNL with the known value and the grand average. The plus or minus values are calculated for 1σ .

A description of the sampling and analytical procedures used at LLNL is included as Appendix E.

As described in the Radiochemical Methods section of Appendix E, blank samples are routinely analyzed.

Air filter blanks consist of collection media which has been handled and processed in the same way as samples. The average values are: for ²³⁸U, 0.24 µg/blank \pm 25% SDM; for ²³⁵U, 0.0018 µg/blank \pm 39% SDM; for ²³⁹Pu, below the detection limit of 4 \times 10⁻⁹ µCi/sample.

Water blanks are tap water which has been analyzed just as the samples. The average values are: for ¹³⁷Cs, $3.4 \times 10^{-11} \mu$ Ci/ml ± 36% SDM; for ²³⁹Pu, $1.0 \times 10^{-12} \mu$ Ci/ml ± 44% SDM.

Washed and ignited sand is used for the soil blank. The ²¹⁹Pu content is below the detection limit of 4 \times 10⁻⁹ μ Ci/sample.

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Appendix A. Tables

The statistical values (2σ estimates) that accompany individual measurements of radioactivity in the following tables are the result of counting statistics. The minimum detection limit is assumed reached when the 2σ estimate is $\pm 100\%$. Statistical values for groups of data like annual averages are calculated standard deviations of the mean (average).

	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Average		
Location ⁴				-				Ci/ml)						% SDM ^b	% CG
Perimeter															
01	0.8	0.9	0.8	0.8	0.7	0.9	0.9	0.7	0.6	0.7	0,6	0.5	0.7	18	4
02	0.9	2.3	0.8	0.7	0.6	0.9	0.7	0.7	0.6	-0,6	1.5	0.6	0.9	56	4
12	0.8	0.8	0.7	0.7	0.6	0.9	0.7	0.7	0.8	1.1	0.9	0.6	0.8	18	4
13	0.4	0.4	0.3	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	8	2
14	0.8	0.7	0.7	0.7	0.7	0.9	0.7	0.8	0.7	0.8	0.7	0.5	0.7	14	4
15	0.4	0.4	0.4	0.4	0.5	0.5	0.4	0.5	0.6	0.6	0.4	0.3	0.4	20	2
Valley															
03	0.6	3.9	0.6	0.7	0.6	0.5	0.6	0.6	0.6	0. 6	0.9	0.6	0.9	106	4
04	0.6	0.5	0.5	0.5	0,4	0.5	0.9	0.7	0.6	0.7	0.7	0.5	0.6	24	3
05	0.7	0.2	0.7	0.8	0.7	0.6	0.6	0.7	0.6	0.6	0.7	0.6	0.6	24	3
06	0.9	2.4	0.7	0.7	0.6	0.6	0.7	0.7	0.6	0.6	0.8	0.7	0.6	60	4
07	0.6	2.8	0.6	0.7	0.7	0.5	0.7	0.6	0.7	0.7	0.8	0.4	0.8	77	4
08	0.8	0.8	0.8	0.8	0.7	0.7	0.9	0.8	0.7	0.6	0.5	0.4	0.7	20	4
09	0.9	2.8	0.7	1.0	1.0	0.8	0.6	0.8	0.9	0.9	0.9	0.9	1.0	56	5
10	0.8	0.8	0.8	0.8	0.7	0.7	0.8	0.7	1.0	0.7	0.8	0.5	0.8	16	4
11	0.7	2.4	0.6	0.9	0.8	ð.6	0.7	0.7	0.7	0.7	0.7	0.6	0.8	59	4
16	0.7	0.7	0.9	0.8	0.8	0.7	1.6	0.7	0.6	0.7	1.0	0.8	0.8	83	4
17	0.6	0.6	1.6	0.8	0.8	0.6	0.7	0.8	0.6	0.7	0.7	0.6	0.8	37	4
nnual average													0.7	22	4

Table 1. Gross alpha activity on air filters – LLNL perimeter and Livermore Valley.

* See Figs. 3 and 5.

^b % SDM = % standard deviation of mean at 1σ .

^c Concentration guide (CG) = $2 \times 10^{-14} \, \mu \text{Ci/ml}$.

Location	Jan.	Feb.	Mar.	Apr.	May			Aug. (Ci/ml)		Oct.	Nov.	Dec.	Average	% SDM	% CG ^b
Perimeter															
01	1.7	2.9	1.3	1.8	1.6	1.4	0.9	0.9	1.2	1.5	1.3	1.1	1.5	36	2
02	1.5	1.8	1.4	1.8	1.7	1.4	0.5	1.0	1.7	1.3	2.2	1.1	1.5	31	2
12	1.4	1.6	1.3	1.3	1.5	1.4	0.9	1.1	1.9	1.8	1.9	1.0	1.4	23	1
13	1.4	1.6	1.2	1.3	1.3	0.9	0.7	0.8	1.4	1.4	1.5	1.0	1.2	23	1
14	1.8	1.9	1.2	1.4	1.5	0.9	0.8	0.7	1.5	1.2	1.7	1.3	1.3	29	1
15	1.5	1.7	1.2	1.4	1.5	0.9	0.7	0.9	1.6	1.5	1.6	1.0	1.3	26	1
Valley															
03	2.2	13.9	2.3	2.5	2.1	1.6	1.2	1.5	2.0	2.3	1.9	1.8	2.9	118	3
04	1.3	1.9	1.3	1.5	1.6	0.6	0.5	0.5	0.9	0.9	1.4	1.3	1.1	40	1
05	2.5	8.6	2.3	2.5	2.3	1.7	2.0	1.6	2.3	2.2	2.3	2.0	2.7	70	3
06	2.6	9.1	2.5	2.4	2.3	2.2	1.5	1.4	2.4	2.7	2.8	2.5	2.9	70	3
07	2.1	9.6	2.0	2.3	2.2	1.2	1.6	1.6	2.4	2.8	2.5	1.8	2.7	83	3
08	1.9	2.0	1.4	1.7	1.7	1.1	1.0	1.1	2.1	2.0	2.2	1.0	1.6	29	2
09	3.6	11.7	3.0	3.4	3.2	2.8	1.2	2.3	3.3	2.4	3.7	2.9	3.6	73	4
10	1.5	1.8	1.2	1.4	1.4	0.5	0.7	0.8	1.3	1.4	2.0	1.3	1.3	34	1
11	2.6	9.4	2,5	3.4	3.0	2.3	2.0	2.2	2.7	2.9	2.6	2.3	3.2	63	3
16	1.7	1.3	1.5	1.5	1.5	0.9	1.0	0.9	1.3	1.4	2.3	1.1	1.4	29	1
17	1.3	1.6	1.4	1.5	1.6	1.0	0.7	0.8	1.6	1.6	1.8	1.1	1.3	27	1
nnual average													1.9	43	2

Table 2. Gross beta activity on air filters - LLNL perimeter and Livermore Valley.

* See Figs. 3 and 5.

^bCG = $1 \times 10^{-12} \mu \text{Ci/ml}$.

	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Average		
Location*	(10 ⁻¹⁵ μCi/ml)							_			% SDM	% CG⁵			
01	0.3	0.3	0.3	0.4	0.4	0.3	0.4	0.5	0.5	0.4	0.3	0.4	0.4	21	2
02	0.3	0.3	0.3	0.5	0.4	0.4	0.3	0.3	0.3	0.4	0.3	0.5	0.4	22	2
03	0.4	0.3	0.5	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.7	0.5	0.6	20	3
04	0.4	0.4	0.3	0.6	0.5	0.8	na'	0.5	0.4	0.5	0.6	0.4	0.5	29	2
05	0.3	0.3	0.3	2.3	0.3	0.3	4.0	0.4	0.3	0.3	0.4	0.5	0.8	142	4
06	1.1	0.9	0.6	1.0	0.7	0.7	1.2	0.7	0.7	0.8	0.7	0.5	0.8	25	4
08	0.3	0.3	0.3	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.3	0.4	14	2
09	0.3	0.2	0.4	0.4	0.5	0.5	0.4	0.3	0.4	0.4	0.4	0.4	0.4	21	2
10	0.4	0.3	0.3	0.4	0.5	0.5	0.5	0.6	0.5	0.4	0.4	na	0.4	21	2
nnual average													0.5	38	2

Table 3. Gross alpha activity on air filters – Site 300.

* See Fig. 4. Location 6 is in city of Tracy.

 b CG = 2 × 10⁻¹⁴ μ Ci/ml.

' Not available.

	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Average		
Location*	-					(1	10 ⁻¹⁴ д	Ci/ml)	-			_		% SDM	ም ርር _የ
01	2.0	1.7	1.6	1.6	2.0	1.3	1.1	1.4	1.6	1.3	1.8	1.4	1.6	18	2
02	1.9	2.0	1.6	1.4	1.9	1.4	1.3	1.3	1.5	1.7	1.8	1.7	1.6	15	2
03	2.6	2.3	2.0	1.9	2.7	1.0	1.8	1.6	2.1	1.7	2.6	2.0	2.0	24	2
04	2.2	1.6	2.1	1.6	1.1	1.5	na	0.7	1.3	1.8	1.6	1.7	1.6	27	2
05	2.1	1.9	1.5	1.2	0.9	0.3	1.9	0.4	0.6	1.3	2.1	1.7	1.3	50	1
06	2.2	2.7	1.5	1.8	1.9	1.1	1.0	1.0	1.4	1.3	2.2	1.5	1.6	33	2
08	1.6	1.5	1.2	1.2	1.6	1.2	0.5	1.1	1.3	1.2	1.6	1.2	1.3	24	1
09	1.8	1.8	1.7	1.6	2.1	1.3	1.2	1.4	1.ó	1.7	1.6	1.4	1.6	16	2
10	1.5	2.0	1.5	1.7	1.8	1.2	1.1	1.2	1.5	1.4	1.7	na	1.5	19	2
Annual average													1.6	12	2

Table 4. Gross beta activity on air filters - Site 300.

* See Fig. 4. Location 6 is in city of Tracy. ^b CG = $1 \times 10^{-12} \mu$ Ci/ml.

Table 5.	Gamma activity	on air filters–LLNL	perimeter.

	⁷ Be	⁴⁰ K	¹⁰⁶ Ru	125Sb	¹³⁷ Cs	144Ce	
Month	$[10^{-13} \mu \text{Ci/ml} \pm 2\sigma (\%)]$		[1 0 ⁻	¹⁶ μCi/ml ± 2σ	(%)]		
Jan.	0.73 ± 2	6.1 ± 57	5.6 ± 48	1.0 ± 47	1.9 ± 11	10.5 ± 12	
Feb.	1.3 ± 3	6.4 ± 97	10.7 ± 26	2.2 ± 36	4.4 ± 6	23.0 ± 13	
Mar.	0.79 ± 3	5.2 ± 42	6.9 ± 25	1.5 ± 29	2.9 ± 7	13.4 ± 13	
Apr.	1.2 ± 2	2.0 ± 100	9.0 ± 49	1.8 ± 21	3.9 ± 8	15.9 ± 9	
May	0.91 ± 2	57.6 ± 11	7.5 ± 51	1.4 ± 71	3.3 ± 10	11.0 ± 10	
June	0.53 ± 3	49.7 ± 25	3.3 ± 49	0.3 ± 100	1.6 ± 26	5.2 ± 29	
July	0.52 ± 4	53.4 ± 23	1.8 ± 100	0.5 ± 100	1.4 ± 35	3.9 ± 44	
Aug.	0.66 ± 4	10.1 ± 28	1.9 ± 82	0.2 ± 100	1.L ± 14	2.2 ± 26	
Sept.	1.0 ± 4	12.3 ± 28	0.7 ± 100	0.5 ± 100	1.2 ± 14	2.6 ± 100	
Oct.	0.7 ± 4	1.7 ± 100	0.7 ± 100	0.2 ± 100	0.4 ± 50	0.5 ± 100	
Nov.	0.74 ± 2	4.6 ± 68	0.6 ± 100	0.2 ± 100	0.7 40	1.1 ± 62	
Dec.	0.58 ± 2	$2.4~\pm~100$	0.1 ± 100	0.1 ± 100	0.7 ± 48	0.7 ± 100	
Annual average	0.80	17.6	3.8	0.8	2.0	7.5	
% SDM	31	125	101	88	68	96	
CG (µCi/ml)	4×10^{-8}	none	2×10^{-10}	9×10^{-10}	2×10^{-9}	2 × 10 ⁻¹⁰	
% CG	2×10^{-4}		2×10^{-4}	1×10^{-5}	1×10^{-5}	4×10^{-4}	

	⁷ Be	40K	¹⁰⁶ Ru	¹²⁵ Sb	¹³⁷ Cs	144Ce
Month	$[10^{-13} \mu \text{Ci/ml} \pm 2\sigma (\%)]$		[10	$^{-16} \mu Ci/ml \pm 2c$	7 (%)]	
Jan.	1.0 ± 3	36.0 ± 20	8.8 ± 21	1.6 ± 51	2.8 ± 12	15.9 ± 8
Feb.	1.2 ± 3	3.6 ± 37	12.2 ± 11	2.1 ± 19	4.3 ± 3	21.6 ± 8
Mar.	1.2 ± 2	0.8 ± 100	10.9 ± 9	2.1 ± 11	4.4 ± 4	19.9 ± 5
Apr.	1.1 ± 2	1.2 ± 100	8.4 ± 15	1.9 ± 15	3.7 ± 6	15.4 ± 7
May	1.6 ± 3	5.6 ± 64	13.2 ± 6	2.2 ± 19	5.8 ± 7	20.3 ± 8
June	0.90 ± 2	3.9 ± 58	4.8 ± 21	1.1 ± 27	2.4 ± 9	6.8 ± 17
July	0.9 ± 4	33.0 ± 25	4.4 ± 41	1.1 ± 72	2.4 ± 11	6.5 ± 12
Aug.	1.2 ± 2	5.5 ± 28	4.1 ± 24	0.8 ± 28	2.2 ± 6	5.6 ± 18
Sept.	1.4 ± 2	6.5 ± 20	3.2 ± 28	0.6 ± 26	1.7 ± 6	3.7 ± 20
Oct.	0.97 ± 2	3.2 ± 60	1.2 ± 46	0.1 ± 100	0.6 ± 22	1.3 ± 34
Nov.	0.79 ± 2	1.0 ± 100	0.4 ± 100	0.1 ± 100	0.8 ± 14	1.8 ± 44
Dec.	0.89 ± 2	0.9 ± 100	0.7 ± 76	0.1 ± 100	1.0 \pm 10	1.5 ± 28
Annual average	1.1	8.4	6.0	1.2	2.7	10.0
% SDM	22	146	75	71	60	80
CG (µCi/ml)	4×10^{-8}	none	2×10^{-10}	9 × 10 · 10	2×10^{-9}	2×10^{-10}
% CG	3×10^{-4}		3×10^{-4}	1×10^{-5}	1×10^{-5}	5 × 10 ⁻⁴

Table 6. Gamma activity on air filters - Site 300.

	Location*										
	090-04	090-08	090-10	090-16	090-17	040-06					
Month	$[10^{-17} \ \mu \text{Ci/ml} \pm 2\sigma \ (\%)]$										
Jan.	0.3 ± 27	0.3 ± 40	0.2 ± 38	0.3 ± 27	2.8 ± 35	0.4 ± 39					
Feb.	0.6 ± 21	0.5 ± 24	0.6 ± 24	0.4 ± 27	0.5 ± 24	0.5 ± 29					
Mar.	0.4 ± 18	0.5 ± 27	0.4 ± 25	0.5 ± 22	0.4 ± 35	0.5 ± 21					
Apr.	0.5 ± 16	0.5 ± 19	0.7 ± 22	0.7 ± 25	0.6 ± 22	0.6 ± 26					
May	0.5 ± 17	0.6 ± 22	0.5 ± 17	0.7 ± 17	0.5 ± 23	0.5 ± 23					
June	0.4 ± 30	0.3 ± 30	0.2 ± 46	па	0.3 ± 26	0.3 ± 31					
July	0.2 ± 37	0.2 ± 39	0.2 ± 44	0.3 ± 39	0.2 ± 37	0.2 ± 34					
Aug.	0.2 ± 54	0.2 ± 42	0.2 ± 46	0.6 ± 27	0.2 ± 56	0.2 ± 36					
Sept.	0.2 ± 47	0.1 ± 56	0.3 ± 42	0.2 ± 35	0.1 ± 45	0.2 ± 42					
Oct.	0.1 ± 73	0.1 ± 52	0.1 ± 52	0.2 ± 42	0.1 ± 61	0.1 ± 92					
Nov.	0.1 ± 42	0.1 ± 47	0.1 ± 46	0.2 ± 43	0.1 ± 52	0.2 ± 79					
Dec.	0.1 ± 49	па	na	0.06 ± 95	0.1 ± 65	0.1 ± 100					
Annual average	0.3	0.3	0.3	0.4	0.5	0.3					
% SDM	60	60	64	58	156	55					
% CG ^ኮ	5×10^{-3}	5×10^{-3}	5×10^{-3}	7×10^{-3}	8×10^{-3}	5×10^{-3}					

⁴ See Fig. 5. Location 040-06 is in city of Tracy. ^b CG = $6 \times 10^{-14} \mu$ Ci/ml.

		²³⁹ Pu ^b	²³⁵ U	²³⁸ U	²³⁵ U/ ²³⁸ U
Month	Location*	$[10^{-17} \ \mu \text{Ci/ml} \pm 2\sigma \ (\%)]$	$[10^{-7} \mu g/m^3 \pm 2\sigma (\%)]$	$[10^{-5} \mu g/m^3 \pm 2\sigma (\%)]$	(10 - 3)
Jan.	01	0.4 ± 33	1.9 ± 1	2.5 ± 1	7.1
	02	0.5 ± 29	2.8 ± 1	4.1 ± 1	7.0
	12	0.6 ± 26	4.6 ± 2	6.9 ± 1	6.7
	13	0.3 ± 27	1.5 ± 1	2.2 ± 1	7.0
	14	0.3 ± 30	1.7 ± 1	2.6 ± 1	6.7
	15	0.3 ± 24	1.5 ± 1	2.2 ± 1	7.0
Feb.	01	0.6 ± 20	2.8 ± 3	4.1 ± 3	6.9
	02	3.5 ± 11	4.5 x 3	6.5 ± 3	7.0
	12	0.7 ± 17	6.7 ± 3	10.1 ± 3	6.8
	13	0.4 ± 23	2.1 ± 3	3.0 ± 3	7.0
	14	0.5 ± 19	2.7 ± 3	4.0 ± 3	6.8
	15	0.5 ± 16	2.0 ± 3	2.9 ± 3	6.9
Mar.	01	0.5 ± 19	1.9 ± 3	3.0 ± 3	6.5
	02	0.5 ± 25	2.6 ± 3	4.0 ± 3	6.5
	12	0.4 ± 20	4.6 ± 3	7.0 ± 3	6.6
	13	0.3 ± 17	1.6 ± 3	2.4 ± 3	6.8
	14	0.4 ± 49	1.8 ± 3	2.6 ± 3	6.5
	15	0.4 ± 32	1.6 ± 3	2.4 ± 3	6.7
Apr.	01	0.6 ± 24	3.5 ± 3	5.2 ± 3	6.9
	02	0.7 ± 22	3.8 ± 3	5.0 ± 3	7.6
	12	0.5 ± 26	4.4 ± 3	6.2 ± 3	7.1
	13	0.5 ± 22	2.8 ± 3	3.9 ± 3	7.2
	14	0.6 ± 35	6.0 ± 3	8.5 ± 3	7.2
	15	0.5 ± 24	3.0 ± 3	4.3 ± 3	7.2
May	01	0.4 ± 32	6.3 ± 3	9.0 ± 3	7.1
	02	0.6 ± 23	4.8 ± 3	6.8 ± 3	7.1
	12	0.5 ± 30	8.7 ± 3	11.1 ± 3	6.9
	13	0.3 ± 25	5.4 ± 3	7.6 ± 3	7.2
	14	0.7 ± 20	6.1 ± 3	8.5 ± 3	7.2
	15	0.5 ± 21	7.7 ± 3	13.6 ± 3	7.2
}une	01	0.4 ± 25	6.9 ± 3	9.9 ± 3	7.0
	02	0.2 ± 33	4.4 ± 3	6.1 ± 3	7.2
	12	0.3 ± 30	7.6 ± 3	11.0 ± 3	6.9
	13	0.2 ± 22	3.7 ± 3	5.2 ± 3	7.1
	14	0.5 ± 20	6.4 ± 3	9.1 ± 3	7.0
	15	0.2 ± 23	7.4 ± 3	10.4 ± 3	7.1
July	01	0.2 ± 37	8.5 ± 3	12.3 ± 3	6.9
	02	0.1 ± 47	6.2 ± 3	8.6 ± 3	7.2
	12	0.3 ± 40	8.1 ± 3	11.7 ± 3	6.9
	13	0.2 ± 34	3.3 ± 3	4.6 ± 3	7.1
	14	0.3 ± 36	5.0 ± 3	7.4 ± 3	6.8
	15	0.2 ± 33	6.2 ± 3	8.8 ± 3	7.1

Table 8. Plutonium and uranium on air filters – LLNL perimeter.

		239 Pu ^b	²³⁵ U	²³⁸ U	²³⁵ U/ ²³⁸ U
Month	Location ^a	$[10^{-17} \mu \text{Ci/ml} \pm 2\sigma (\%)]$	$[10^{-7} \mu g/m^3 \pm 2\sigma (\%)]$	[10 ⁻⁵ µg/m ³ ± 2σ (%)]	(10-3)
Aug.	01	0.3 ± 34	6.7 ± 3	5.2 ± 3	6.8
	02	0.2 ± 42	5.4 ± 3	7.6 ± 3	7.1
	12	0.3 ± 34	10.0 ± 3	14.7 ± 3	6.8
	13	0.1 ± 31	3.9 ± 3	5.3 ± 3	7.3
	14	0.9 ± 16	6.2 ± 3	8.8 ± 3	7.1
	15	0.2 ± 27	10.5 ± 3	14.8 ± 3	7.1
Sept.	01	0.2 ± 81	7.5 ± 3	11.6 ± 3	6.5
	02	0.2 ± 40	7.2 ± 3	9.9 ± 3	7.3
	12	0.3 ± 30	11.4 ± 3	17.7 ± 3	6.4
	13	0.1 ± 37	4.7 ± 3	7.0 ± 3	6.7
	14	0.8 ± 21	6.9 ± 3	9.9 ± 3	7.0
	15	0.2 ± 37	15.2 ± 3	21.6 ± 3	7.0
Oct.	01	0.3 ± 36	6.3 ± 3	$9.0~\pm~3$	7.0
	02	0.1 ± 100	6.3 ± 3	9.0 ± 3	7.0
	12	0.2 ± 45	9.7 ± 3	14.7 ± 3	6.6
	13	0.1 ± 44	3.8 ± 3	5.4 ± 3	7.0
	14	0.2 ± 46	2.4 ± 3	3.5 ± 3	6.9
	15	0.2 ± 41	9.1 ± 3	12.8 ± 3	7.1
Nov.	01	0.1 ± 41	1.8 ± 3	2.6 ± 3	6.9
	02	U.2 ± 38	3.7 ± 3	5.4 ± 3	6.9
	12	1.3 ± 19	6.0 ± 3	9.0 ± 3	6.7
	13	J.1 ± 48	2.5 ± 3	3.5 ± 3	7.1
	14	1.5 ± 16	1.8 ± 3	2.7 ± 3	6.7
	15	0.1 ± 38	2.4 ± 3	3.6 ± 3	6.7
Dec.	01	0.2 ± 38	2.1 ± 3	3.2 ± 3	6.6
	02	0.4 ± 31	3.2 ± 3	4.7 ± 3	6.8
	12	1.1 ± 21	6.2 ± 3	9.8 ± 3	6.3
	13	0.3 ± 29	4.4 ± 3	6.2 ± 3	7.1
	14	0.5 ± 34	2.3 ± 3	3.6 ± 3	6.4
	15	0.2 ± 31	2.3 ± 3	3.3 ± 3	7.0

Annual averages:

	²³⁹ Pu ^b			²³⁵ U			²³⁸ U		
Location ^a	(10 ⁻¹⁷ µCi/ml)	% SDM	ኈ CG'	(10 ⁻⁷ µg/m ³)	% SDM	% CG ^d	(10 ⁻⁵ µg/m ³)	% SDM	% CG'
01	0.4	46	7 × 10 ⁻³	4.7	54	2 × 10 ⁻⁵	6.9	54	4 × 10 ⁻⁴
02	0.6	157	1×10^{-2}	4.6	32	2×10^{-5}	6.5	30	4×10^{-4}
12	0.5	63	8×10^{-3}	7.2	34	4×10^{-5}	10.8	32	7×10^{-4}
13	0.2	54	3×10^{-3}	3.3	38	2×10^{-5}	4.7	37	3×10^{-4}
14	0.6	58	1×10^{-2}	4.1	51	2×10^{-5}	6.0	50	4×10^{-4}
15	0.3	49	5×10^{-3}	5.9	76	3×10^{-5}	8.4	75	6×10^{-4}

* See Fig. 3.

^b In all tables activity listed as ²³⁹Pu includes activity due to ²⁴⁰Pu isotope. ^c CG = $6 \times 10^{-14} \,\mu$ Ci/ml for ²³⁹Pu (soluble) activity in air.

 ${}^{d}CG = 1.9 \ \mu g/m^3$ for ${}^{235}U$ (insoluble) activity in air. ${}^{e}CG = 15 \ \mu g/m^3$ for ${}^{238}U$ (insoluble) activity in air.

	²³⁹ Pu	¹³⁷ Cs		²³⁵ U	²³⁸ U	
	[10 ^{−17} µCi/ml	[10 ^{−15} µCi/ml	²³⁹ Pu/ ¹³⁷ Cs	[10 ⁻⁷ µg/m³	(10 ⁻⁵ µg/m³	²³⁵ U/ ²³⁸ U
Month	± 2a (%)]	± 2σ (%)]	(10 ⁻²)	± 2σ (%)]	± 2σ (%)]	(10 3)
Jan.	1.6 ± 8	0.28 ± 12	5.7	1.6 ± 1	4.1 ± 1	3.9
Feb.	0.6 ± 6	0.43 ± 3	1.4	4.1 ± 3	1.7 ± 3	2.5
Mar.	0.5 ± 7	0.44 ± 4	1.1	1.4 ± 3	3.4 ± 3	4.2
Apr.	0.6 ± 10	0.37 ± 6	1.6	1.8 ± 3	3.4 ± 3	5.5
May	0.6 ± 8	0.58 ± 7	1.0	4.8 ± 3	9.6 ± 3	5.1
June	0.6 ± 6	0.24 ± 9	2.5	4.1 ± 3	7.2 ± 3	5.7
July	0.3 ± 10	0.24 ± 11	1.2	2.4 ± 3	4.3 ± 3	5.5
Aug.	0.2 ± 14	0.22 ± 6	0.9	9.8 ± 3	37.5 ± 3	2.6
Sept.	0.2 ± 10	0.17 ± 6	1.2	6.0 ± 3	17.9 ± 3	3.4
Oct.	0.4 ± 9	0.06 ± 22	6.7	4.4 ± 3	6.7 ± 3	5.1
Nov.	0.1 ± 18	0.08 ± 14	1.3	6.7 ± 3	29.8 ± 3	2.2
Dec.	0.1 ± 13	0.10 ± 10	1.0	1.3 ± 3	3.0 ± 3	4.4
Annual sversge	0.5	0.27		4.0	10.9	
% SLM	83	60		64	107	
CG	$6 \times 10^{-14} \mu \mathrm{Ci/ml}$	$2 \times 10^{-9} \mu \text{Ci/ml}$		$1.9 \ \mu g/m^3$	15 μg/m ³	
% CG	8×10^{-3}	1×10^{-5}		2×10^{-5}	7 × 10 ⁻⁴	

Table 9. Plutonium, cesium, and uranium on air filters - Site 300.

	Perimeter location*						Valley location [*]			
	01	02	12	13	14	15	S:	S2	Average	
Month				(10 ⁻¹¹ µCi/ml)					% SDM
Jan.	1.0	2.7	2.5	9.3	2.3	1.8	1.3	1.3	2.8	98
Feb.	3.8	3.7	6.2	10.5	2.1	2.8	1.7	5.6	4.6	63
Mar.	7.4	13.3	20.4	11.4	11.6	5.5	3.2	7.7	10.1	53
Apr.	1.4	2.8	2.2	1.6	5.6	6.1	0.8	1.4	2.7	73
May	2.9	2.4	4.8	1.6	4.3	3.4	3.3	2.1	3.1	35
June	1.1	1.0	1.6	0.7	S.5	1.7	0.6	1.1	1.7	95
July	0.9	0.5	1.9	0.5	4.3	2.3	0.6	na	1.6	89
Aug.	1.3	1.2	1.9	0.6	2.7	1.5	2.5	Ũ.2	1.5	60
Sept.	1.6	2.2	3.8	1.3	3.1	2.4	1.2	0.6	2.1	49
Oct.	2.2	4.8	4.5	7.5	2.6	3.5	1.3	1.3	3.5	61
Nov.	2.5	6.2	5.6	4.3	2.5	3.0	2.3	2.5	3.6	44
Dec.	4.2	4.3	4.5	3.4	3.7	2.2	1.6	3.4	3.4	29
Average	2.5	3.8	5.0	4.4	4.2	3.0	1.7	2.5	3.4	
% SDM	74	91	102	94	63	48	55	92		
% C G ^b	1.2×10^{-2}	1.9×10^{-2}	2.5×10^{-2}	2.2×10^{-2}	2.1×10^{-2}	1.5×10^{-2}	0.8×10^{-2}	1.2×10^{-2}		
Calculated ^c adult whole-body dose										
(mrem)	3.2×10^{-2}	4.8×10^{-2}	6.3×10^{-2}	5.6×10^{-2}	5.3×10^{-2}	3.6×10^{-2}	2.2×10^{-2}	3.2×10^{-2}		

Table 10. Tritium (HTO) in air - LLNL perimeter and Livermore Valley.

* See Figs. 3 and 5.

 $^{b}CG = 2.0 \times 10^{-7} \,\mu Ci/ml.$

⁶ Doses are calculated using methods in U.S. NRC Regulatory Guide 1.109 unless otherwise stated.

		-	<u> </u>			
	²³⁹ Pu	40K	¹³⁷ Cs	232Th	238U	
Location ^a	$[10^{-9} \mu Ci/dry g \pm 2\sigma (\%)]$	$[10^{-5} \mu \text{Ci}/\text{dry g} \pm 2\sigma (\%)]$	[10 ⁻⁷ μCi/dry g ± 2σ (%)]	[µg/dry g	± 2σ (%)]	
889	0.8 ± 19	0.9 ± 5	0.6 ± 18	4.4 ± 8	3.2 ± 15	
890	11.8 ± 6	1.2 ± 3	2.2 ± 6	6.5 ± 5	6.6 ± 19	
891	9.4 ± 7	1.4 ± 4	2.7 ± 6	6.6 ± 7	4.0 ± 17	
892	12.2 ± 6	1.3 ± 6	3.2 ± 5	6.0 ± 11	3.9 ± 32	
893	6.7 ± 9	1.1 ± 4	3.7 ± 5	5.3 ± 5	3.0 ± 25	
894	0.7 ± 20	1.5 ± 4	0.4 ± 25	7.1 ± 6	4.1 ± 25	
895	3.5 ± 11	1.3 ± 4	1.2 ± 9	5,9 ± 6	3.3 ± 29	
896	12.7 ± 6	0.7 ± 14	6.4 ± 6	4.1 ± 19	2.7 ± 42	
897	3.5 ± 11	1.3 ± 6	1.5 ± 11	7.1 ± 10	4.0 ± 18	
898	3.7 ± 9	1.0 ± 4	2.5 ± 7	4.3 ± 8	4.5 ± 16	
899	2.0 ± 13	1.5 ± 3	1.4 ± 12	6.9 ± 2	2.5 ± 26	
900	2.2 ± 15	1.3 ± 6	1.2 ± 10	6.0 ± 11	4.5 ± 12	
911	2.3 ± 13	1.2 ± 6	1.2 ± 13	5.7 ± 10	3.9 ± 25	
912	2.0 ± 13	1.2 ± 5	0.8 ± 22	5.8 ± 10	5.4 ± 16	
913	1.1 ± 18	1.1 ± 6	0.9 ± 21	6.0 ± 10	3.6 ± 36	
914	0.7 ± 28	1.3 ± 11	0.4 ± 64	7.9 ± 13	3.9 ± 24	
915	0.7 ± 23	1.3 ± 8	0.3 ± 40	7.1 ± 10	3.9 ± 35	
916	1.0 ± 19	1.1 ± 8	0.5 ± 34	6.4 ± 12	3.0 ± 80	
917	0.2 ± 52	0.8 ± 11	па	5.5 ± 17	5.0 ± 50	
918	1.5 ± 16	1.4 ± 7	0.7 ± 28	6.8 ± 11	4.7 ± 30	
919	2.5 ± 14	1.8 ± 4	1.3 ± 13	8.1 ± 5	4.9 ± 15	
920	1.1 ± 19	1.0 ± 4	0.6 ± 17	5.0 ± 7	5.1 ± 17	
923	0.9 ± 19	1.2 ± 4	0.4 ± 20	5.5 ± 6	5.3 ± 16	
924	5.9 ± 9	1.1 ± 4	3.4 ± 5	4.5 ± 10	3.0 ± 26	
925	0.6 ± 20	1.5 ± 2	0.3 ± 24	7.2 ± 3	4.7 ± 17	
926	3.3 ± 10	1.2 ± 3	2.3 ± 5	6.2 ± 5	5.5 ± 19	

Table 11. Various radionuclides in soil – Livermore Valley (sampling depth = 0-5 cm).

* See Fig. 6.

Table 12. Plutonium, cesium and uranium in soil-Site 300 (sampling depth = 0-5 cm).

Location*	^{239}Pu [10 ^{~9} μ Ci/dry g ± 2 σ (%)]	^{137}Cs [10 ⁻⁷ μ Ci/dry g ± 2 σ (%)]	²³⁸ U		
			[µg/dry g ± 20 (%)]		
901	4.3 ± 9	2.6 ± 6	278.0 ± 3		
902	3.2 ± 10	1.5 ± 11	5.9 ± 21		
903	5.5 ± 4	2.8 ± 6	4.3 ± 19		
904	0.9 ± 17	0.4 ± 22	3.4 ± 38		
905	3.7 ± 10	2.2 ± 6	4.1 ± 29		
906	3.7 ± 12	1.8 ± 13	6.1 ± 15		
907	2.1 ± 14	1.6 ± 18	63.0 ± 6		
908	4.5 ± 10	2.6 ± 8	3.6 ± 38		
909	0.6 ± 24	0.2 ± 52	3.9 ± 25		
910	4.0 ± 9	1.7 ± 9	3.0 ± 31		
921	5.6 ± 10	2.7 ± 6	3.9 ± 17		
922	3.3 ± 13	2.1 ± 7	4.6 ± 22		

^a See Fig. 7.

	н	ю	137	'Cs	239	Pu
	$[10^{-6} \mu \text{Ci/ml} \pm 2\sigma (\%)]$		[10 ^{−11} µCi/1	nl ± 2σ (%)]	$[10^{-12} \mu \text{Ci/ml} \pm 2\sigma (\%)]$	
Month	LLNL	LWRP	LLNL	LWRP	LLNL	LWRP
Jan.	1.2 ± 70	0.4 ± 88	2.8 ± 78	1.0 ± 100	11.0 ± 12	4.4 ± 18
Feb.	2.8 ± 55	0.6 ± 82	3.7 ± 86	1.4 ± 100	5.6 ± 21	1.8 ± 31
Mar.	10.7 ± 24	1.3 ± 77	3.4 ± 44	0.9 ± 100	26.0 ± 12	1.7 ± 29
Apr.	13.8 ± 29	1.3 ± 56	4.8 ± 50	1.3 ± 100	15.0 ± 10	1.1 ± 39
May	4.8 ± 26	0.8 ± 99	2.7 ± 58	1.0 ± 100	16.0 ± 10	1.0 ± 36
June	1.8 ± 63	0.3 ± 100	2.4 ± 64	1.3 ± 100	4.7 ± 24	0.9 ± 49
July	6.6 ± 60	0.5 ± 100	3.2 ± 60	2.6 ± 70	11.1 ± 14	1.4 ± 34
Aug.	3.4 ± 50	0.7 ± 100	1.1 ± 100	2.2 ± 68	9.8 ± 16	na
Sept.	1.7 ± 49	0.4 ± 93	4.4 ± 64	3.2 ± 82	4.0 ± 24	1.4 ± 41
Oct.	2.6 ± 43	0.8 ± 85	6.2 ± 34	1.2 ± 100	9.0 ± 14	0.7 ± 49
Nov.	1.5 ± 68	0.4 ± 86	11.3 ± 20	2.0 ± 98	8.5 ± 17	0.5 ± 65
Dec.	4.9 ± 51	0.6 ± 86	3.9 ± 44	1.2 ± 100	$26.5~\pm~10$	1.3 ± 37
Annual average	4.7	0.7	4.2	1.6	12.3	1.5
% SDM	85	49	62	45	61	71
CG (µCi/ml)	0.1	3×10^{-3}	4×10^{-4}	2×10^{-5}	1×10^{-4}	5 × 10 ⁻⁶
% CG	5×10^{-3}	2×10^{-2}	1×10^{-5}	8×10^{-5}	1.2×10^{-5}	3×10^{-5}

Table 13. Various radionuclides in effluents - LLNL and Livermore Water Reclamation Plant.

Table 14. Gross alpha activity in water – Livermore Valley.

	Number of	Maximum	Minimum	Average		
Location ^a	samples		[10 ⁻⁹ μCi/ml ± 2σ (%)	1	% SDM	% CG ^b
11	4	≤4.2	≤0.3	≤2.5	65	≲8
15	4	≤4.2	≤0.3	≤2.5	66	≤8
16	4	≤4.2	≤0.3	≤2.5	65	≤8
19	4	≤4.2	≤2.7	≤3.1	24	≤10
20	8	≤4.2	≤0.4	≤2.3	68	≤8
24	4	≤4.2	≤0.3	≤2.5	65	≤8
26	4	≤4.2	≤0.3	≤2.6	63	≤9
29	4	≤4.2	≤0.3	≤2.5	65	≤8
31	4	≤4.2	≤2.7	≤3.2	21	≤11
33	4	≤4.2	≤2.7	≤3.1	24	≤10
34	3	≤4.2	≤0.3	≤2.4	81	≤8
37	4	≤3.5	≤0.3	≤1.7	96	≤6

^a See Fig. 9.

 $^{\rm b}$ CG = 3 × 10⁻⁸ μ Ci/ml.

	Number of	Maximum	Minimum	Average		
Location [*]	samples	[10	⁻⁹ μCi/ml ± 2σ (%)]		% SDM	% CG ^b
01	4	≤4.2	≤3,3	≤3.7	11	12
02	4	≤4,2	≤3.3	≤3.7	11	12
04	9	37.0 ± 32	≲2.8	≤9.0	122	30
05	9	4.8 ± 46	≤2.8	≤3.8	17	13
06	4	≤4.2	≤3.3	≤3.7	11	12
07	4	≤4.2	≤3.3	≤3.7	11	12
14	12	≤8.4	≤0.3	≤3.8	59	13
20	9	44.0 ± 23	≤0,3	≤9.2	148	31
21	10	11.0 ± 53	2.3 ± 90	≤4.4	56	15
22	8	5.4 ± 42	≤2.8	≤3.8	22	13
23	3	≲3.5	≤0.3	≤2.3	76	8

Table 15. Gross alpha activity in water - Site 300.

^a See Fig. 10. ^b CG = 3 × 10⁻⁸ μCi/ml.

Table 16. Gross beta activit	y in water – Livermore Valley.
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	Number of	Maximum	Minimum	Average		
Location ⁴	samples	[10	⁻⁹ μCi/ml ± 2σ (%)		% SDM	% ር G ^ь
11	4	12.0 ± 34	≤8.2	≤10	21	3
15	4	15.0 ± 32	≤8.4	≤13	25	4
16	4	15.0 ± 33	≤8.2	≤11	30	4
19	4	≤12.0	≤8.2	<u>≤</u> 9	20	3
20	8	20.0 ± 38	≤1.5	≤11	67	4
24	4	19.0 ± 29	≤8.4	≤13	36	4
26	4	16.0 ± 31	≤8.4	≲12	26	4
29	4	27.0 ± 20	≤8.4	≤15	53	5
31	4	≤12.0	≤8.2	≤9	20	3
33	4	18.0 ± 44	≤12.0	≤14	20	5
34	3	22.0 ± 28	≤12.0	≤16	31	5
37	4	18.0 ± 24	≤8.2	≤13	36	4

* See Fig. 9.

^b CG = $3 \times 10^{-7} \,\mu$ Ci/ml.

	Number of	Maximum	Minimum	Average		
Location ^a	samples	[10	⁻⁹ μCi/ml ± 2σ (%)]		% SDM	% CG ^b
01	4	15.0 ± 60	≤8.9	≤12	23	4
02	4	$12.0~\pm~72$	≤8.9	≤11	14	4
04	9	29.0 ± 32	≤10.0	≤17	41	6
05	9	25.0 ± 13	≤8.3	≤12	43	4
06	4	16.0 ± 57	≤8.9	≤12	27	4
07	4	20.0 ± 61	≤8.9	≤12	43	4
14	12	≤24.0	6.7 ± 22	≤14	47	5
20	9	110.0 ± 12	≤8.3	≤23	141	8
21	10	44.0 ± 16	\leq 4.6 ± 28	≤19	71	6
22	8	21.0 ± 14	≤8.3	≤12	35	4
23	3	28.0 ± 26	≤8.3	≤15	70	5

Table 17. Gross beta activity in water -- Site 300.

^a See Fig. 10.

^b CG = $3 \times 10^{-7} \mu \text{Ci/ml}$.

	Number of	Maximum	Minimum	Average			Calculated adult whole-body dose	
Location*	samples	$\frac{10^{-7} \mu \text{Ci/ml} \pm 2\sigma (\%)}{[10^{-7} \mu \text{Ci/ml} \pm 2\sigma (\%)]}$			% SDM	% CG [▶]	(mrem)	
11	4	0.70 ± 9	0.43 ± 14	0.60	16	2×10^{-3}	5 × 10 ⁻³	
15	4	1.10 ± 8	0.62 ± 10	0.86	25	3×10^{-3}	7×10^{-3}	
16	2	5.60 ± 3	1.84 ± 5	3.52	46	12×10^{-3}	27×10^{-3}	
19	4	1.40 ± 12	0.75 ± 31	0.94	33	3×10^{-3}	7×10^{-3}	
20	9	19.6 ± 2	6.40 ± 6	11.6	46	39×10^{-3}	89×10^{-3}	
24	4	1.64 ± 5	0.72 ± 24	1.00	43	3×10^{-3}	8×10^{-3}	
26	4	2.01 ± 5	1.46 ± 15	1.82	13	6×10^{-3}	14×10^{-3}	
29	4	0.77 ± 9	0.53 ± 12	0.65	15	2×10^{-3}	5×10^{-3}	
30	4	0.65 ± 10	0.52 ± 14	0.55	15	2×10^{-3}	4×10^{-3}	
31	4	0.85 ± 10	0.35 ± 3	0.56	43	2×10^{-3}	4×10^{-3}	
32	1	0.66 ± 9		0.66		2×10^{-3}	5×10^{-3}	
33	4	2.22 ± 10	0.60 ± 9	1.27	58	4×10^{-3}	10×10^{-3}	
38	1	0.69 ± 10		0.69		2×10^{-3}	5×10^{-3}	

Table 18. Tritium (HTO) in water – Livermore Valley.

⁴ See Fig. 9.

 b CG = 3 \times 10 $^{-3}$ μ Ci/ml.

	Number of	Maximum	Minimum	Average			Calculated adult whole-body dose
Location ^a	samples	$[10^{-7} \mu \text{Ci/ml} \pm 2\sigma (\%)]$			% SDM	% CG ^b	(mrem)
01	4	0.68 ± 9	0.37 ± 17	0.56	24	2×10^{-3}	4 × 10 ⁻³
02	4	0.24 ± 23	0.18 ± 33	0.22	13	1×10^{-3}	2×10^{-3}
03	2	0.20 ± 29	0.07 ± 77	0.14	70	0.5×10^{-3}	1×10^{-3}
04	11	0.26 ± 5	0.06 ± 100	0.15	37	0.5×10^{-3}	1×10^{-3}
05	11	0.42 ± 4	0.09 ± 61	0.23	44	1×10^{-3}	2×10^{-3}
06	4	0.66 ± 10	0.05 ± 100	0.25	115	1×10^{-3}	2×10^{-3}
07	4	0.28 ± 20	0.16 ± 39	0.22	28	1×10^{-3}	2×10^{-3}
11	3	2.53 ± 4	0.64 ± 9	1.52	62	5×10^{-3}	12×10^{-3}
14	11	0.86 ± 18	0.46 ± 12	0.62	19	2×10^{-3}	5×10^{-3}
20	9	1.55 ± 12	0.26 ± 25	0.58	66	2×10^{-3}	4×10^{-3}
21	6	0.77 ± 10	0.08 ± 59	0.29	83	1×10^{-3}	2×10^{-3}
22	9	0.27 ± 54	0.08 ± 60	0.15	35	$0.5 imes 10^{-3}$	1×10^{-3}
23	3	0.16 ± 38	0.07 ± 100	0.12	40	0.5×10^{-3}	1×10^{-3}

Table 19. Tritium (HTO) in water-Site 300.

⁴ See Fig. 10.

 b CG = 3 × 10⁻³ µCi/ml.

Table 20. T	ritium (HTO) :	in :	groundwater	Livermore	Valley.
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Location ^a	Well identification	Well depth (m)	Activity $[10^{-7} \mu \text{Ci/ml} \pm 2\sigma (\%)]$	% CG ^ь
01	3S1E-1P2	15	15.2 ± 9	5 × 10 ⁻²
02	3S1E-2R1	10	9.9 ± 3	3×10^{-2}
03	3S1E-8H2	62	1.2 ± 6	4×10^{-3}
04	3S1E-9G1	49	3.3 ± 4	1 × 10 ⁻²
05	3S1E-9P5	32	1.8 ± 5	6 × 10 ⁻³
06	3S1E-10A2	27	8.5 ± 3	3×10^{-2}
07	3S1E-11B1	13	10.5 ± 12	4×10^{-2}
08	3S1E-11C1	70	0.8 ± 9	3×10^{-3}
09	3S1E-11H1	92	0.9 ± 8	3×10^{-3}
10	3S1E-12A2	23	2.2 ± 4	7×10^{-3}
11	3S1E-12D2	14	9.1 ± 3	3×10^{-2}
12	3S1E-12G1	27	11.9 ± 11	4×10^{-2}
13	3S1E-12J1	44	0.9 ± 8	3×10^{-3}
14	3S1E-12N1	93	0.8 ± 10	3×10^{-3}
15	3S1E-14A2	67	0.9 ± 7	3×10^{-3}
16	3S1E-16H2	29	0.7 ± 11	2×10^{-3}
17	3S2E-1P2	44	0.1 ± 42	3×10^{-4}
18	3S2E-7C2	15	1.9 ± 5	6×10^{-3}
19	3S2E-7N1	41	1.1 ± 7	4×10^{-3}
20	3S2E-11A1	20	1.1 ± 8	4×10^{-3}
21	3S2E-14B4	79	0.6 ± 11	2×10^{-3}

^a See Fig. 11. ^b CG = 3 × 10⁻³ μCi/ml.

	Number of	Maximum	Minimum	Average		Calculated adult whole-body dose
Location [*]	samples	[10 ⁻⁷ µCi/m]	± 2σ (%)] in wate	r recovered	% SDM	(mrem)
04	4	3.5 ± 31	2.8 ± 36	3.2	11	1 × 10 ⁻²
15	3	13.9 ± 8	11.1 ± 10	12.9	12	4×10^{-2}
20	4	17.5 ± 8	3.2 ± 36	9.7	77	3×10^{-2}
23	2	17.3 ± 7	16.8 ± 7	17.1	2	5×10^{-2}
29	4	50.1 ± 4	7.9 ± 15	22.7	94	7×10^{-2}
31	4	73.6 ± 3	10.1 ± 11	31.3	92	9×10^{-2}
32	4	11.5 ± 11	4.2 ± 24	7.3	42	2×10^{-2}
33	4	15.1 ± 9	1.1 ± 100	5,5	119	2×10^{-2}

Table 21. Tritium (HTO) in vegetation – Livermore Valley.

4 See Fig. 12.

	Number of	Maximum	Minimum	Average		Calculated adult whole-body dose
Location ⁴	samples	[10 ⁷ µCi/ml	± 2σ (%)] in wate	r recovered	% SDM	(mrem)
01	4	2.3 ± 50	1.1 ± 100	1.4	39	4 × 10 ⁻³
02	4	2.4 ± 47	0.9 ± 100	1.4	49	4×10^{-3}
03	4	2.6 ± 43	1.0 ± 99	1.8	37	6×10^{-3}
06	2	4.7 ± 25	2.4 ± 48	3.6	47	1.1×10^{-2}
12	4	2.4 ± 47	1.0 ± 98	1.5	44	5×10^{-3}
13	4	14.0 ± 9	1.5 ± 74	4.9	124	$1.5 imes10^{-2}$

Table 22. Tritium (HTO) in vegetation – Site 300.

* See Fig. 13.

Table 23. Tritium (HTO) in wine.

	Number of	Maximum	Minimum	Average	
Location	samples	[10 ⁻⁷ µCi/n	nl ± 2σ (%)] in water	recovered	% SDM
Livermore Valley	7	4.7 ± 7	0.4 ± 45	1.9	76
Other California areas	2	0.6 ± 33	0.5 ± 26	0.5	7
Europe	2	12.8 ± 3	5.3 ± 6	9.0	59

	Number of	Maximum	Minimum	Average	
Location	samples	[10 ⁻⁷ µCi/n	nl ± 2σ (%)] in water	recovered	% SDM
Livermore Valley	3	6.60 ± 5	1.90 ± 12	3.53	75
Other California areas	2	2.13 ± 11	1.92 ± 12	2.03	7

Table 24. Tritium (HTO) in honey.

Table 25. Various radionuclides in milk – Livermore Valley.

	HTO [10 ⁻⁷ μCi/ml ± 2σ (%)]	40 K [10 ⁻⁶ μ Ci/ml ± 2 σ (%)]	¹³⁷ Cs [10 ⁻⁴ μCi/ml ± 2σ (%)]
Number of samples	18	i8	18
Maximum	9.3 ± 13	1.9 ± 2	5.1 ± 61
Minimum	1.0 ± 100	1.1 ± 3	0.6 ± 100
Average	3.0	1.6	2.1
% SDM	77	12	53
Calculated adult whole-body dose (mrem)	1×10^{-2}	8.9	5×10^{-2}

Location*	Jan Mar.	AprJune	July-Sept. (mrem)	OctDec.	Annua
		• <u> </u>			
1	11	14	11	16	52
2	12	12	11	13	48
3	13	15	14	15	57
4	12	14	12	15	53
5	12	14	13	14	53
6	11	14	10	14	49
7	11	14	12	13	50
8	11	11	12	12	46
9	12	14	12	1.2	50
10	10	11	10	12	43
11	9	11	12	10	42
12	10	12	9	12	43
13	12	13	12	13	50
42	11	12	11	13	47
47	11	12	10	12	45
48	12	12	10	12	46
49	12	12	10	12	46
50	12	13	11	12	48
5 2	12	12	10	15	47
53	12	13	11	13	49
54	12	13	11	13	49
56	13	lost	11	13	49
Average	12	13	11	13	49

Table 26. Environmental radiation measurements (TLD) - LLNL perimeter.

* See Fig. 14.

Location ⁴	JanMar.	AprJune	July-Sept. (mrem)	OctDec.	Annual
1	1.2	1.3	1.1	1.4	5.0
2	1.3	1.0	0.9	1.2	4.4
3	3.0	7.5	8.1	17.4	36.0
4	1.1	1.2	1.0	1.4	4.7
5	1.0	1.2	1.5	2.9	6.6
6	1.0	1.0	0.7	1.4	4.1
7	1.3	1.4	1.1	1.9	5.7
8	1.5	1.1	1.8	1.6	6.0
Average	1.4	2.0	2.0	3.7	9.1

Table 27. Environmental neutron monitoring - LLNL perimeter.

⁴ See Fig. 3.

			Locati	on'					
	01	02	12	13	14	15	Average		
Month			(10 ⁵ μg/π.	3)			% SDM	% Standard ^b
Jan.	2.2	3.6	5.3	2.5	2.3	2.1	3.0	42	0.3
Feb.	11.0	12.0	18.0	8.6	8.4	7.9	11.0	35	1.1
Mar.	0.3	<0.2	0.8	0.4	<0.2	0.2	< 0.4	66	<0.1
Apr.	1.3	1.1	1.2	1.7	0.6	1.6	1.2	31	0.1
May	12.1	18.0	7.7	8.5	6.3	10.0	10.0	40	1.0
June	8.9	0.5	5.8	0.6	3.3	6.9	4.3	79	0.4
July	9.1	3.4	6.1	0.8	1.9	6.2	4.6	68	0.5
Aug.	6.2	3.3	11.0	5.0	24.0	13.0	10.0	73	1.0
Sept.	15.0	9.5	18.0	8.7	11.0	па	12.0	32	1.2
Oct.	<0.3	<1.	3.0	1.7	0.4	8.8	<2.4	140	<0.2
Nov.	1.0	1.6	0,3	1.7	0.8	2.7	1.4	62	0.1
Dec.	0.4	0.2	0.2	4.4	0.2	0.4	1.0	174	0.1
Average	5.6	4.5	6.4	3.7	5.0	5.4	5.1		0.5
% SDM	95	125	98	88	141	79			

Table 28. Beryllium on air filters – LLNL perimeter.

⁴ See Fig. 3.

^b Ambient concentration limit set by the Bay Area Air Quality Management District is 0.01 µg/m³.

				Lo	Location*						
	01	02	03	04	5	8	99	10	Average		
Month					(10 ⁻⁵ μg/m ³)	/m³)			c	% SDM	% Standard ^b
Jan.	1.8	2.1	2.8	ž.6	1.9	2.2	1.4	1.9	21	22	0.2
Feb.	4.7	5.5	7.7	5.0	7.0	4.3	4.1	5.6	5.5	23	0.6
Mar.	0.3	0.2	0.6	<0.2	0.3	< 0.05	0.2	< 0.1	<0.2	70	< 0.1
Apr.	1.1	1.0	0.7	0.6	<0.1	0.4	0.6	0.6	<0.6	50	<0.1
May	15.	8.5	13.	11.	3.9	7.1	43.	23	13.	ûĥ	0.2
June	5.9	5.5	6.3	6.1	12.	5,6	1.3	4.1	5.8	51	0.4
July	0.3	1.6	1.7	3,9	0.5	4.9	<0.3	5.5	<2.3	91	0.6
Aug.	5.1	4.9	7.3	6.0	3.3	2.7	4.4	8.4	5.3	36	0.5
Sept.	4.0	4.0	7.3	5	0.9	3.9	1.4	4.9	3.5	60	0.5
Oct.	0.4	0.8	0.2	<0.2	1.4	1.4	0.8	1.5	< 0.8	64	0.2
Nov.	0.8	1.0	1.5	0.4	1.2	0.8	0.7	1.0	0.9	36	0.1
Dec.	0.1	0.1	0.6	<0.2	<0,1	0.1	<0,1	па	0.2	100	< 0.1
Average	3.3	2.9	4.1	3.2	2.7	2.8	4.9	3.3	3.4		0.3
% SDM	129	91	86	107	131	85	249	80			
See Fip	4										

Table 29. Beryllium on air filters-Site 300.

[•] See Fig. 4. [•] Ambient concentration limit set by the Bay Area Air Quality Management District is 0.01 μg/m³.

				•					
	Cł	ព្	Cu	æ	Fb	Ni	Ag	Zn	A
					(ppm)				
Jan.	0.003	0.56	0.64	0.88	0.01	0.14	0.007	0.37	0.78
Feb.	0.003	0.13	0.16	0.74	0.02	0.04	0.03	0.45	0,33
Mar.	0.003	0.05	0.10	0.75	0.01	0.01	0.01	0.37	1.47
Apr.	0.003	0.26	0.27	1.2	0.03	0.03	0.004	0.37	1.2
May	0.002	0.22	0.24	1.5	0.01	0.01	0.003	0.56	0,56
June	0.003	0.19	0.15	1.2	0.02	0.04	0.007	0.38	0.27
July	0.002	0.17	0.19	0.77	0.02	0.02	0.018	0.35	0.26
Aug.	0.002	0.08	0.09	0.55	0.02	0.02	0.002	0.14	0.01
Sept.	0.002	0.18	0.20	1.80	0.04	0.02	0.029	0.44	0.35
Oct.	0.004	0.19	0.16	0.80	0.03	0.02	0.023	0.24	0,41
Nov.	0.002	0.18	0.17	1.60	0.15	0.02	0.06	0.50	1.45
Dec.	0.003	0.16	0.49	0.74	0.01	0.01	0.005	0.14	0,30
Average	0.003	0.20	0.24	1.0	0.03	0.03	0.017	0.36	0,62
% SDM	38	65	69	39	126	114	103	36	81

Table 30. Various elements in LLNL liquid effluent.

	JanMar.	AprJune	July-Sept.	OctDec.	Average	
			(mg/liter)			% SDM
BOD	86	89	91	86	88	3
COD	230	362	178	159	237	43
Total nitrogen (as N)	36	34	29	24	31	17
Ammonia nitrogen (as N)	32	27	26	20	26	19
Nitrate nitrogen (as N)	< 0.01	0.01	0.17	< 0.01	< 0.05	160
Nitrite nitrogen (as N)	<0.01	0.02	0.01	< 0.01	< 0.01	80
Oil and grease (FEM)	14	12	8.5	10	11	21
Sulfate	131	62	55	52	75	50
Arsenic	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	
Boron	0.86	0.53	0.39	0.49	0.57	35
Cvanide	0.41	< 0.02	0.08	0.04	0.14	131
Mercury	0.0001	0.0025	0.019	0.0010	0.006	159
Selenium	< 0.001	<0,001	< 0.001	< 0.001	~ 0.001	
Total alkalinity	122	121	120	85	112	16
Total phosphorous (as P)	5.4	5.4	5.1	4.0	5.0	13
Chloride	56	84	4.4	150	74	82
Phenols	0.18	0.06	0.10	0.08	0.11	50
Calcium	12	9.0	15	44	20	81
Magnesium	1.6	4.8	1.3	40	12	157
Sodium	88	58	28	92	66	45
Chromium	0.22	0.06	0.12	0.075	0.12	61
Copper	0.080	0.10	0.089	0.022	0.073	48
Nickel	0.006	0.013	0.005	< 0.001	< 0.006	64
Potassium	22	18	18	18	19	11
Zinc	0.45	0.18	0.15	0.28	0.26	51
Sodium as 14 of cations	75	66	48	-10	57	28
Total solids	2000	285	232	429	736	115
Dissolved solids	346	245	221	378	298	26
Volatile solids as						
'? of total solids	7.1	44.9	42	20.5	28.6	63
Settleable solids						
(ml/liter/h)	0.8	0.7	2.5	3.5	1.9	73
Total suspended solids				15	15	
Identifiable chlorinated						
hydrocarbons	n one *	none	none	none		
РСВ	none	none	none	none		

Table 31. Physical and chemical examinations of LLNL liquid effluent.

* The following compounds would have been reported had they appeared at or above their respective detection limits as indicated below:

Organochlorine pesticides	µg/liter	Polychlorinated biphenyls	μg/liter
Aldrin	0.05	Aroclors 1016-1262	0.3
Chlordane	0.1		
Endrin	0.1		
Dieldrin	0.05		
BHC (mixed isomers)	0.05		
DDT (mixed isomers)	0.1		
DDE (mixed isomers)	0.05		
DDD (mixed isomers)	0.05		
Heptachlor	0.05		

		Feb.	Mar.	Apr.	May	Jue	July	Aug.	Sept.	Oct.	Nov.	Dec.	Average	% SDM
Specific condu (µmhos/cm at														
Location*:	4	1250	1320	1513	1220		1310	1240	1400	1290	1330	1340	1321	6
	5	1070	860	1173	960		1010	1000	820	980	1080	1040	999	10
	14	2110	1760	800	1440	1690	1750	1900	1980	1910	1800	1650	1708	20
	22	922	720	1016	916		923	930	1030	930	950	960	930	9
Total hardness as CaCO ₃ (mg														
Location:	4	30	24	20	22		25	23	23	31	22	22	24	15
	5	45	49	42	43		65	62	39	65	62	46	52	20
	14	592	510	237	400	420	470	527	531	555	390	440	461	22
	22	46	44	42	36		42	52	51	47	42	42	44	11
Chloride (mg/	1)													
Location:	4	168	153	210	152		16 6	150	158	160	189	167	167	11
	5	76	73	87	76		80	75	66	75	94	85	79	10
	14	172	141	35	104	120	128	150	160	150	142	125	130	28
	22	64	62	66	66		65	71	68	70	75	65	67	6
Total organic carbon (mg/1)														
Location:	4	17	14	30	49		15	11	8	25	17	17	20	59
	5	15	12	29	51		13	13	7	24	14	19	20	64
	14	32	23	30	71	31	63	25	30	38	28	25	36	44
	22	16	3	23	39		12	12	6	22	15	17	18	73

Table 32. Site 3	00 water sam	ples down	gradient from	liquid-discharge	e locations.
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* See Fig. 10.

Nuclide	Facility	Curies	Dose at sit. boundary (mrem)	Dose to nearest resident (mrem)	Dose within 80-km radius of LLNL (man-rem)
ч	Tritium facility	1914	0.09	0.07	0.78
	Insulating core accelerator	44	0.04	0.01	0.01
	Rotating target neutron source	56	0.01	0.01	0.01
¹³ N- ¹⁵ O	Linear accelerator	584	0.64	0.12	0.00

		Airborne effluents (Ci)	Liquid effluents (Ci)				
Year	⁴¹ Ar	³ Н	¹³ N- ¹⁵ O	НТО	239Pu			
1976	470	3991	1035	10	1.5×10^{-4}			
1977	380	5210	990	13	3.6×10^{-4}			
1978	766	5362	1445	9	8.6×10^{-4}			
1979	383	4517	829	7	9.8×10^{-4}			
1980	165	2305	1656	5	2.8×10^{-4}			
1981	0	2620	344	6	0.92×10^{-4}			
1982	0	2014	584	1.6	0.43×10^{-5}			

Table 34. Radioactive effluent releases from LLNL from 1976 through 1982.

Table 35. LLNL nonradioactive liquid effluent -- storm drain runoff.

	Influent	Effluent				
Analyses	(mg/liter)					
Nitrate nitrogen (as N)	4.3	0.82				
Total phosphorous (as P)	1.3	0.3				
Cyanide	<0.02	<0.02				
Phenols	< 0.005	< 0.005				
Oil & grease (freon extractable)	<5	<5				
Surfactants (MBAs)	0.21	0.08				
Sulfate	18	4.7				
Total identifiable chlorinated hydrocarbons	none*	none				

* The following compounds would have been reported had they appeared at or above their respective detection limits as indicated below:

Organochlorine pesticides	μg/liter	Polychlorinated biphenyl	µg/liter
Aldrin	0.05	Aroclors 1016-1262	0.3
Chlordane	0.1		
Endrin	0.1		
Dieldrin	0.05		
BHC (mixed isomers)	0.05		
DDT (mixed isomers)	0.1		
DDE (mixed isomers)	0.05		
DDD (mixed isomers)	0.05		
Heptachlor	0.05		

			Value reported (pCi/li	ter)
Analyses	Media	LLNL	Known	Grand average of all Laboratories
Gross alpha	water	11 ± 2	24 ± 6	21 ± 6
		21.3 ± 1.2	27.5 ± 7	24.7 ± 6.9
		14.0 ± 2	16 ± 5	16 ± 5
		15 ± 3	19 ± 5	17 ± 4
	air	23 ± 1	32 ± 8	28 ± 6
Gross beta	water	36 ± 1	32 ± 5	31 ± 6
		18 ± 2	29 ± 5	30 ± 6
		19 ± 1	23 ± 5	21 ± 5
		28 ± 1	24 ± 5	24 ± 3
	air	63 ± 1	67 ± 5	61 ± 8
Plutonium	water	7.6 ± 0.1	6.9 ± 0.7	7.3 ± 0.8
Tritium	water	2040 ± 50	1820 ± 590	na
		2970 ± 72	2860 ± 360	na
		1750 ± 56	1830 ± 340	1765 ± 229
		2837 ± 107	2890 ± 380	2847 ± 270
		2580 ± 72	2560 ± 350	2517 ± 250
		1907 ± 106	1990 ± 345	2009 ± 233

Table 36. Summary of analyses for the quality assurance program.

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Environmental Activity Concentration – Guide Levels Appendix B.

The standards for Radiation Protection (DOE Manual 5480.1 Chg 2, issued April 29, 1981) state that the average activity of a mixture of radionuclides (whose identities and concentrations are unknown) in air and water should not exceed the following values:

 $\begin{array}{c} 6 \times 10^{-13} \; \mu \text{Ci/ml} \\ 2 \times 10^{-14} \; \mu \text{Ci/ml} \end{array}$ 1. Air (controlled area) 2. Air (uncontrolled area)

- $4 \times 10^{-7} \,\mu \text{Ci/ml}$ Water (controlled area)
- 4. Water (uncontrolled area) $3 \times 10^{-8} \,\mu \text{Ci/ml}$

If alpha emitters and ²²⁷Ac are definitely not present, the following values may be used to determine permissible average activity:

 $3 \times 10^{-11} \,\mu \text{Ci/ml}$ 5. Air (controlled area) $1 \times 10^{-12} \,\mu \text{Ci/ml}$ 6. Air (uncontrolled area)

If ¹²⁹I, ²²⁶Ra, and ²²⁸Ra are definitely not present, the following values may be used:

- 7. Water (controlled area) $3 \times 10^{-6} \,\mu \text{Ci/ml}$ 8. Water (uncontrolled area) $1 \times 10^{-7} \,\mu \text{Ci/ml}$

Both air and water samples are subjected to gross alpha and gross beta measurements. The average annual alpha activities of samples may not exceed the activity values listed as 1-4 above. Since the alpha emitters have been accounted for in the gross alpha measurements and the assumption is made that ¹²⁹I, ²²⁷Ac, ²²⁶Ra, and ²²⁸Ra are not present in the samples, the average annual gross beta activities of the samples may not exceed the activities listed as 5-8 above. The assumption that ¹²⁹I, ²²⁷Ac, ²²⁶Ra, and ²²⁸Ra are not present in air and water samples is reasonable in view of the minute quantities of these radionuclides available at the Laboratory. This reference also states that average tritium activities in off-site water samples may not exceed $3 \times 10^{-3} \,\mu \text{Ci/ml}$.

The external whole-body radiation dose to workers in controlled areas may not exceed 5 rem/y, and the dose to an individual in an uncontrolled area may not exceed 500 mrem/y. Also, a group of individuals in an uncontrolled area may not receive an average annual dose of more than 170 mrem.

Appendix C. Method of Dose Calculations

The doses shown in this report have been calculated using the models and methods in the Nuclear Regulatory Commission Regulatory Guide 1.109, Calculation of Annual Doses to Man from Routine Releases of Reactor Effluent. Examples of these calculations and assumptions are shown in this appendix.

Annual Dose from Potable Water

Assuming that all water sampled is available as drinking water, the annual whole-body dose for tritium has been calculated using the following equation:

$$\mathbf{R}_{\text{total body}} = \mathbf{C}_{w} \mathbf{U}_{w} \mathbf{D}_{w} \quad . \tag{1}$$

where

 $C_w = \text{concentration in pCi/liter,}$ $U_w = \text{intake rate, liters/y,}$ = 730 liters/y for maximum exposed individual, $D_w = \text{dose factor, mrem/pCi,}$ $= 1.05 \times 10^{-7} \text{ mrem/pCi for the whole-body ingestion pathway for an adult.}$

 $R_{\text{total body}} = \text{annual dose in mrem to the total body from ingestion of 730 liters of potable water with concentration <math>C_w$.

Annual Dose from Forage-Cow-Milk Pathway for Tritium in Vegetation

Assuming that all feed for the cattle was pasture grass, the annual whole-body dose per μ Ci/ml HTO for the maximum exposed individual has been calculated using the following equation:

$$D_{\text{total body}} = D_{\text{veg}} + D_{\text{meat}} + D_{\text{milk}} , \qquad (2)$$

$$D_{veg}$$
 (leafy vegetables) = $U_{veg} \times C_{veg} \times D_{HTO}$, (2a)

where

 $U_{yeg} = intake rate, kg/y, 64 kg/y for maximum exposed individual ,$

$$C_{veg} = \text{concentration in } pCi/kg = 10^9 \frac{pCi/kg}{\mu Ci/ml} \times C_{veg} \mu Ci/ml \text{ (measured)}$$

 $D_{HTO} = \text{dose factor, mrem/pCi} = 1.05 \times 10^{-7} \text{ mrem/pCi for }^{3}\text{H}$ for the adult whole-body ingestion pathway.

 $D_{veg} (mrem/y) = 0.67 \times 10^4 C_{veg} \mu Ci/ml (measured)$.

$$\mathbf{D}_{\text{meat}} = \mathbf{U}_{\text{meat}} \times \mathbf{C}_{\text{meat}} \times \mathbf{D}_{\text{HTO}} \quad , \tag{2b}$$

where

$$U_{meat} = 110 \text{ kg/y},$$

 $D_{HTO} = 1.05 \times 10^{-7} \text{ mrem/pCi},$
 $C_{meat} = (F_{f}) (Q_{f}) (C_{veg}) \exp(-\lambda_{i}t_{s})$

 F_f = fraction of daily intake of nuclide per kg of animal/fish, days/kg,

 Q_f = amount of feed consumed, kg/day,

 $C_{veg} = same as above,$

 λ_i = radiological decay constant, day⁻¹,

 $t_s =$ time between slaughter to consumption, days.

$$C_{\text{meat}} = \left(1.2 \times 10^{-2} \frac{\text{day}}{\text{kg}}\right) \left(50 \frac{\text{kg}}{\text{day}}\right) \left(C_{\text{veg}} \frac{\mu \text{Ci}}{\text{ml}}\right) \left(10^9 \frac{\text{pCi/kg}}{\mu \text{Ci/ml}}\right)$$
$$\times \exp\left[-4.5 \times 10^3 (20)\right] = 0.6 \times 10^9 \frac{\text{pCi/kg}}{\mu \text{Ci/ml}} \times C_{\text{veg}} \frac{\mu \text{Ci}}{\text{ml}} (\text{measured})$$

 D_{meal} (mrem/y) = 0.69 $\times 10^4 \times C_{veg} \,\mu$ Ci/ml (measured) .

$$D_{milk} = U_{milk} \times C_{milk} \times D_{HTO} , \qquad (2c)$$

where

 $U_{milk} = 310$ liters/y,

 $D_{HTO} = 1.05 \times 10^{-7} \text{ mrem/pCi},$

 $C_{\text{railk}} = F_m Q_f C_{\text{veg}} \exp(-\lambda_i t_i),$

 F_m = fraction of daily intake of nuclide per liter of milk, day/liter,

 $Q_{t} = amount of feed consumed, kg/day,$

 $C_{veg} = same as above,$

 λ_i = radiological decay constant, days⁻¹,

 t_f = transport time from the feed to milk receptor,

$$C_{\text{milk}} = \left(1.0 \times 10^{-2} \frac{\text{day}}{\text{liter}}\right) \left(50 \frac{\text{kg}}{\text{d}}\right) \left(C_{\text{veg}} \frac{\mu \text{Ci}}{\text{ml}}\right) \left(10^9 \frac{\text{pCi/kg}}{\mu \text{Ci/ml}}\right)$$
$$\times \exp\left[-4.5 \times 10^3 (2)\right] = 0.5 \times 10^9 \frac{\text{pCi/liter}}{\mu \text{Ci/ml}} \times C_{\text{veg}} \frac{\mu \text{Ci}}{\text{ml}} \text{ (measured)}$$

 $D_{milk} (mrem/y) = 1.63 \times 10^4 \times C_{veg} \,\mu Ci/ml (measured)$.

$$\begin{split} D_{total} \mbox{ (mrem/y)} &= 0.67 \times 10^4 \mbox{ C}_{veg} \mbox{ μCi/ml (measured)} + 0.69 \times 10^4 \mbox{ C}_{veg} \mbox{ μCi/ml (measured)} \\ &+ 1.63 \times 10^4 \mbox{ C}_{veg} \mbox{ μCi/ml (measured)} = 2.99 \times 10^4 \mbox{ C}_{veg} \mbox{ μCi/ml (measured)} \quad . \end{split}$$

Annual Population Dose

To calculate the "fence-line" site boundary, nearest resident, and population doses, a continuouspoint-source computer code based on the Gaussian plume model was used.

$$C_{\rm downwind} = 3.17 \times 10^4 (\chi/Q)(Q) ,$$

where

$$\begin{split} \chi/Q &= \text{ diffusion parameter, sec/m}^3 \quad, \\ Q &= \text{ release rate, Ci/y,} \\ 3.17 \times 10^4 &= 1 \times 10^{12} \frac{\text{pCi}}{\text{Ci}}/3.15 \times 10^7 \frac{\text{sec}}{\text{y}} \quad, \\ C_{\text{downwind}} &= \text{ concentration downwind, pCi/m}^3 \quad, \\ D_{\text{max individual}} &= U \times C_{\text{downwind}} \times D \quad, \end{split}$$

where

 $U = intake \ rate, \ m^3/y = 8000 \ m^3/y, \ inhalation \ rate for adult maximum exposed individual \ ,$

 $C_{downwind} = 3.17 \times 10^4 (\chi/Q)(Q)$,

D = dose factor, mrem/pCi,

for ³H, D =
$$1.58 \times 10^{-7} \frac{\text{mrem}}{\text{pCi}}$$
,
for ⁴¹Ar, D = $8.84 \times 10^{-3} \frac{\text{mrem} - \text{m}^3}{\text{mrem}^3}$

$$r^{4}Ar, D = 8.84 \times 10^{\circ} \frac{10}{pCi-y}$$

(Inhalation rate is included in the dose factor)

$$\begin{split} &\text{for } {}^{13}\text{N},\text{D}=\text{D}\left({}^{41}\text{Ar}\right) \quad . \\ &\text{for } {}^{15}\text{O},\text{D}=\text{D}\left({}^{41}\text{Ar}\right)\times\frac{1.74}{1.2} \quad , \end{split}$$

 $D_{max individual} = mrem/y$.

For population dose, the diffusion factors and population figures for an area within 80 km of the Laboratory were summed over all directions.

.

Dose (man-rem) =
$$3.17 \times 10^4 \left[\sum_{i}^{n} (\chi/Q)_i P_i \right] Q \cup D$$
, (5)

where

$$\begin{bmatrix} \sum_{i}^{n} (\chi/Q)_{i} P_{i} \end{bmatrix} = \text{Summation of the } (\chi/Q) \text{ for region i} \\ \text{times the population in region i for all regions n, } \frac{\text{man-sec}}{\text{m}^{3}}$$

Q = release rate, Ci/y, U = inhalation rate, average individual (adult), D = dose factor, rem/pCi, Dose = man-rem. (3)

(4)

Appendix D. Discharge Limits to The Sanitary Sewer System Of the City of Livermore

Sections 18.63 and 18.66 of the Code of the City of Livermore (1959) state the discharge limits for Livermore's sanitary sewer system. These limits are as follows:

Section 18.63

No person shall discharge, or cause to be discharged, except for salt waste discharge from water softener units of any kind or description installed and in operation on or before January 31, 1966, which are regenerated by the owner thereof at the place of use of such units, any of the following described water or wastes to any public sewer unless the customer obtains a permit from the city in accordance with Section 18.65.

(a) Any liquor or vapor having a temperature higher than one hundred fifty degrees Fahrenheit.

(b) Any waters or wastes which contain more than two hundred ppm of fat, oil or grease that is petroleum-ether soluble.

(c) Any gasoline, benzene, naphtha, fuel oil or other inflammable or explosive liquid, solid or gas.

(d) Any garbage, except properly ground with a mechanical garbage grinder. Specifically excluded from the sewers are waste products resulting from the handling, storage, and sale of fruits and vegetables from other than retail produce establishments, or other foods not intended primarily for immediate consumption.

(e) Any ashes, cinders, sand, mud, straw, shavings, metal, glass, rags, feathers, tar, coal tar, asphalt, cement, plastics, woods, paunch manure or any other solid viscous substance capable of causing obstruction to the flow in sewers or other interferences with the proper economical operation of the sewage works.

(f) Any wastes or water with a pH lower than six and eight-tenths or higher than eight.

(g) Any waters or wastes containing total dissolved solid increments greater than three hundred and twenty-five ppm, nor chloride increments greater than seventy-five ppm, increase during a single cycle use of the water supply.

(h) Any water or wastes having a B.O.D. greater than three hundred ppm (the average B.O.D. for residential users).

(i) Any waters or wastes containing more than three hundred ppm of suspended solids (the average suspended solids for residential users).

(j) Any waters intended to be used or used to dilute waste discharge to avoid violation of the above limitation (Order No. 586, paragraph 1).

Section 18.66

No person shall discharge or cause to be discharged any radioactive wastes into any public sewers, except where:

A. The waste is discharged in strict conformity with current Atomic Energy Commission recommendations for safe disposal of radioactive wastes.

B. The discharging of radioactive waste will not cause injury to personnel or damage to the sewer works. Any person discharging a radioactive waste to a public sewer in accordance with the provisions of the preceding paragraph shall submit to the council such report as the council may deem necessary.

In the event of an accidental spill of any radioactive material into the public sewer, the person responsible shall (a) immediately notify the plant superintendent, and (b) render such technical or other assistance to the department of public works within his power to prevent the sewage works from becoming contaminated with radioactivity.

Appendix E. Sampling and Analytical Procedures for Environmental Monitoring at Lawrence Livermore National Laboratory

Described herein are sample collection and analysis procedures employed in environmental monitoring at the Lawrence Livermore National Laboratory (LLNL). An integral part of the quality assurance program developed for environmental monitoring at Livermore, these procedures provide a basis for verifying that sampling and analytical activities are being performed as specified.

The first report in this appendix describes the sampling procedures, analytical determinations, and preparation for radio-counting by the LLNL Hazards Control Department personnel. The second report (Manual M-122) describes the methods used by the LLNL Nuclear Chemistry Division.

SAMPLING AND ANALYTICAL PROCEDURES USED BY THE LLNL HAZARDS CONTROL DEPARTMENT

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INTRODUCTION

Written procedures have been prepared for each environmental monitoring activity at Livermore as part of the quality assurance program developed for environmental monitoring at LLNL. The sample collection and analysis procedures described herein have been reviewed and approved by those responsible for managing the monitoring program. During a program audit, such written documentation can be used to verify that sampling and analytical procedures meet program specifications.

The "Sampling" sections of this report describe the sampling procedures used for each sample medium monitored. A general description of each medium is usually provided. This is followed by the collection procedure used, provisions for sample identification, and a schedule for sample replication.

The "Analysis" sections of this report cover sample analysis. General comments on each sample medium analyzed are followed by specific descriptions of standards and calibrations, analytical procedures, and/or counting procedures as applicable for each medium.

AIR SAMPLING

Continuously operating air samplers are used at LLNL to measure the concentration of airborne particulate radioactivity, beryllium, and tritiated water. Samplers placed at six perimeter locations at LLNL and at nine locations at Site 300 measure particulate radioactivity and beryllium. Particulate radioactivity is also measured at 10 off-site locations near the Laboratory. Tritiated-water samplers are operated at the six LLNL perimeter locations and at two off-site locations. In all cases, samplers are so placed as to provide reasonable assurance that any significant concentration of effluents from Laboratory operations would be detected regardless of local meteorology.

Particulates

LLNL perimeter and Site 30° particulate samples are collected on 5.2 x 10^{-2} -m² Whatman filters, using average flow rates of 700 liters/min. Off-site samplers in the Livermore Valley use 4.6 x 10^{-3} -m² Flanders F-700 glassfiber filters operated at a flow rate of 80 liters/min. Particulate sample flow rates are calibrated with a spirometer that has been calibrated against an immersion unit certified by the National Bureau of Standards. Flow rates of Hi-vol air samplers are verified at monthly intervals with a portabi-field-calibration unit.

Tritiated Water

Samples of tritiated-water vapor are collected by drawing air through silica-gel-packed columns at flow rates of about 0.5 liters/min. Columns are exchanged weekly.

IDENTIFICATION

Particulates

Particulate samplers are run continuously, and sample filters are exchanged weekly. Each filter is identified by location, date on, date off, elapsed sampling time, and flow rate. Filter samples are placed in glassine envelopes and are transported to the laboratory, where this information is transferred to log sheets and each sample is given a serial number that accompanies it during analysis. Half of each LLNL perimeter and Site 300 filter is used for beryllium analysis; the other half is retained for radioactivity measurements.

Tritiated Water

Tritiated-water vapor samplers are run continuously, and silica-gel holders are exchanged biweekly. Each sample is identified by location, date

on, date off, elapsed sampling time, and flow rate. This information is transferred to log sheets in the laboratory, and each sample is given a serial number that accompanies it during analysis.

REPLICATION

Particulates

At bimonthly intervals, a particulate air sampler is operated in parallel with the permanent sampler at one of the six LLNL perimeter locations. This parallel operation is continued for one month, filters are changed weekly, and both filters are submitted for analysis in the usual manner. Bimonthly rotation of the extra sampler among the six locations ensures samplers are checked annually. The replicate samples are then compared with respect to gross alpha and beta activities and to ²³⁹Pu content.

Tritiated Water

Parallel samples of tritiated-water vapor are collected at LLNL perimeter locations during alternate months from those locations in which particulate replicates are being collected.

SOIL SAMPLING

Soil samples are collected annually within the Livermore Valley and at Site 300. Radiochemical analysis of these samples serves to document any changes that may have occurred during the year. Increases in radioactivity that may result from Laboratory operations are brought to the public's attention through annual monitoring reports.

COLLECTION

Sampling sites are selected in reasonably level areas that represent undisturbed soil. A 2-m square is marked off in each such area, and an

8.25-cm-diam coring tool designed at LLNL is used to collect 5-cm-deep samples at the corners and center of the square.

IDENTIFICATION

The five samples of soil collected from each 2-m square are placed in plastic bags and are identified by number. This number is entered in a log book, together with the sampling data and a description of the sample location. The number is used to identify the sample throughout the laboratory analysis and in the annual monitoring report.

REPLICATION

During each annual collection of soil samples, at least 10% of the sites are sampled in duplicate. In such collections, two adjacent 5-cm-deep cores are taken from the corners and center of each 2-m square. Separate composites of the five cores are then made, and the composites are identified by successive numbers.

WATER SAMPLING

At quarterly intervals, water samples from various sources in the Livermore Valley and at Site 300 are analyzed for gross alpha and beta radioactivity and for tritium content. These analyses are performed to determine if detectable changes in radioactivity have occurred.

COLLECTION

Grab samples of such surface sources as ponds, creeks, and reservoirs are obtained with a tethered plastic pail. The samples are transferred to 4-liter plastic containers to which 6.5 ml of concentrated HCl has been added. For tritium analyses, a 1-liter sample is collected in an argon-flushed glass container fitted with a ground-glass stopper.

IDENTIFICATION

As samples are collected in the field, they are tagged with sampling location and date. In the laboratory, each sample is assigned the number that accompanies it during analysis.

REPLICATION

During each sample collection period, at least 10% of the water samples are duplicates.

SEWAGE SAMPLING

Sewage from the Laboratory is discharged into the City of Livermore's sanitary sewer system. Radioactive or otherwise hazardous liquid wastes are first treated to reduce concentration levels to within applicable standards before they are released into the sanitary sewer. Sewage samples are collected from LLNL effluent and from the treated effluent of the Livermore Water Reclamation Plant (LWRP).

COLLECTION

LLNL Effluent

A 24-hr composite of LLNL sewage effluent is collected daily by a motorized proportional sampler. Aliquots of this composite are transported to the laboratory in 500-ml wide-mouthed polyethylene bottles.

LWRP Effluent

Daily composites of treated effluent from the LWRP are also collected by a proportional sampler. Aliquots of these composites are placed in 500-ml polyethylene bottles and are retained for weekly collection by LLNL.

IDENTIFICATION

As each sewage sample is collected in the field, it is tagged with the sampling location and date of sampling. In the laboratory, each sample is assigned a number that accompanies it during analysis.

REPLICATION

LLNL Effluent

Every Wednesday, one replicate sample of LLNL effluent is taken for gross alpha, beta, and tritium. For one month each year, duplicate samples of LLNL effluent are taken each day to permit preparation of duplicate monthly composites.

LWRP Effluent

One replicate sample of LWRP effluent is taken every Wednesday and is analyzed for gross alpha, beta, and tritium. For one month each year, duplicate samples are taken each day to permit preparation of duplicate monthly composites.

VEGETATION SAMPLING

At quarterly intervals, samples of vegetation (usually native grasses) are collected throughout the Livermore Valley, at Site 300, and at the off-site locations near Site 300. After these samples are freeze-dried, the tritium content of the recovered water is determined with liquid-scintillation counting.

COLLECTION

Samples of vegetation are routinely collected from the same locations each sampling period. Grass samples are pulled up or cut close to the ground, and any soil is discarded. The bulk volume of these samples is usually between

0.5 and 1 liter. As each sample is collected, it is placed in a plastic bag and frozen in a dry-ice chest to minimize water loss on the bag surface. The duplicate samples collected at each location are later transferred to a freezer in the laboratory.

IDENTIFICATION

Samples of vegetation collected in the field are tagged according to sample type and location. In the laboratory, each sample is assigned a number that accompanies it until the measurement is reported. Measurement data are permanently retained on computer cards.

REPLICATION

Two samples of vegetation are collected at each location in case the first sample is lost during analysis or the measurement is in question. At least one set of each group of samples collected is analyzed in duplicate to furnish replicate measurement data.

MILK SAMPLING

Milk samples are obtained at monthly intervals from two goat farms located about 5 km south and 5 km southeast of the Laboratory. A 3-liter portion of each milk sample is vacuum-distilled, and the tritium content of the distilled water is determined by liquid scintillation. Each 3-liter sample is then concentrated by evaporation and gamma-counted.

AIR ANALYSIS

Environmental air samples are analyzed for gross alpha and beta activity and for various gamma-emitting radionuclides, plutonium isotopes, uranium isotopes, and beryllium.

GROSS ALPHA AND BETA COUNTING

After a four-day delay to allow for decay of the radon-thoron daughters, all environmental-radioactivity air filters are counted for alpha and beta activity. Counting is done in a Sharp gas-proportional counter using 20-min counting periods. Normally, this counter is used only for environmental samples since a duplicate counting system is provided for routine samples.

Standards and Calibration

Separate ²³⁹Pu and ⁹⁰Sr standards on 5-cm-diam stainless steel convoluted planchets are used for determining alpha and beta counting efficiencies, respectively. These standards are traceable to NBS or to equivalent certified sources. Counting-efficiency measurements are made with each set of filters counted, and a background count is taken at the beginning of each run and between each set of 10 samples. Records are kept of background and counting-efficiency variations in the Sharp counter.

BERYLLIUM

Every month, half of each weekly LLNL perimeter and Site 300 paper air filter is composited by sampling location. Two paper filters at a time are wet-ashed with a mixture of 80% nitric and 20% perchloric acid. The solutions are then evaporated to a few ml, taking care not to allow samples to bake dry. After the samples are diluted with water, they are filtered, any residues are discarded, and the filtrates are diluted to 25 ml with deionized water.

Any fine particulate matter passing through the filter is removed by centrifugation. The analysis is performed in an atomic emission spectrometer with a plasma source. Blanks and known spikes are subjected to the same procedure for quality control.

Standards and Calibration

Beryllium standards are prepared with 0.5 and 1 μ g of beryllium per 25 ml of solution. The recorder readout of the Model 306 Perkin Elmer atomic

absorption spectrophotometer is set at $30 \times absorbance$ expansion, and the spectrophotometer is zeroed while aspirating deionized water into the flame. Instrument responses from the 0.5- and 1-µg beryllium standards are then recorded. Following these calculations, the response of each monthly composite is recorded and its beryllium content is calculated from the calibration data.

WATER ANALYSIS

Environmental water samples are analyzed for gross alpha and beta activity and for tritium content.

GROSS ALPHA AND BETA ACTIVITY

A 100-ml sample or less, depending on the solid's content, is digested with a mixture of 80% HNO_3 and 20% $HClO_4$, and taken to dryness. The residue is taken up with HNO_3 and a few milliliters of H_2O_2 (5%). The solution is evaporated to about 5 ml and transferred to a 5-cm-diam serrated stainless steel planchet. After drying and flaming the planchet, the sample is counted for 30 min in a Sharp 3-1/4-in proportional counter.

Standards and Calibration

The ²³⁹Pu and ⁹⁰Sr standards are used with every sample set (see the "Soil Analysis" section on standards and calibration). Instrument background measurements are made between every 10 sample-counting measurements.

SEWAGE ANALYSIS

Except for tritium analyses, all sewage samples are wet-digested with a mixture of 20% $HClO_4$ and 80% HNO_3 . Those sewage samples for tritium analysis are distilled to reduce self-absorption before counting.

GROSS ALPHA AND BETA RADIOACTIVITY

About 25 ml of an 80/20 mixture of HNO_3 and $HClO_4$ is added to a 500-ml aliquot (100 ml of LWRP effluent), and the mixture is heated to dryness on a hotplate. The residue is taken up in 10 ml HNO_3 and 5 ml H_2O_2 (5%). The volume is reduced to 3-5 ml on a hotplate and transferred to 5-cm-diam planchets and counted.

TRITIUM

A 5-ml aliquot of sewage sample is added to 15 ml of Packard Insta-Gel in a 25-ml screw-cap polyethylene bottle. Then, 5 ml of distilled water is used to prepare a background for the sample, and both are placed in the refrigerated detection chamber of the Packard scintillation counter for a 1-hr temperature-equilibration period prior to counting. The sample and background are each counted for 100 min.

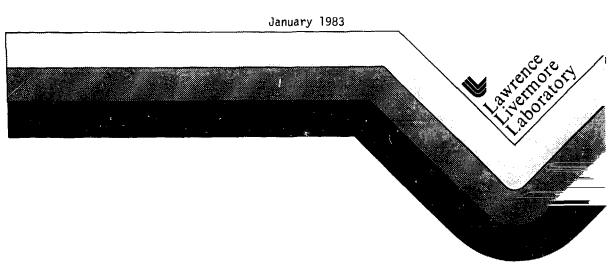
HEAVY METALS

Monthly composites of LLNL sewage effluent are analyzed for cadmium, chromium, copper, iron, lead, nickel, and silver.

A composite is prepared by combining daily samples of 500 ml, as follows: nitric acid (80%) and $HClO_4$ (20%) are added to the first 500-ml daily sample of the month, and the mixture is reduced in volume to about 100 ml. Each subsequent day the daily sample is added and the total volume reduced to about 100 ml. After the last 500 ml from the last day is added, the combined volume is reduced to about 50 ml. When cool, about 50 ml deionized water is added and the liquid is filtered through No. 40 Whatman paper. The paper is then placed in a platinum crucible and digested with about 5 ml HF, 16 ml HNO₃, and 4 ml HClO₄ to a residue of about 4-5 ml. Water is added and the liquid is refiltered. The residue is discarded. The filtrates are combined, transferred to a 100-ml volumetric flask, and brought to volume with deionized water. The metals are determined with a Perkin-Elmer Model 306 atomic absorption spectrophotometer. The required standards are prepared by dilution of 1000-ppm NBS-traceable stock solution.

RADIOCHEMICAL METHODS OF THE SITE ENVIRONMENTAL MONITORING PROGRAM

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ABSTRACT

This methods manual describes the facilities and methods used by the Site Environmental Monitoring Program of LLNL's Nuclear Chemistry Division. Included in this report are descriptions of (1) the low-level environmental laboratory and counting facilities, (2) types of samples collected and analyzed, (3) chemical and counting methods used, and (4) quality assurance procedures. The purpose of this report is to describe the facilities and methods used by Lawrence Livermore National Laboratory's Nuclear Chemistry Division in support of the Laboratory's Site Environmental Monitoring Program. This will provide a single, easily updatable description of the current program and environmental radio-analytical techniques.

Part I describes the types of samples analyzed for the Site Environmental Program and the laboratory and counting facilities used for low-level radiochemistry. Part II describes the specific methods of analysis, and Part III describes the quality assurance program.

I. OVERVIEW OF THE SITE ENVIRONMENTAL MONITORING PROGRAM

A. Sample Types

The Lawrence Livermore National Laboratory's Site Environmental Monitoring Program (SEMP) routinely analyzes five types of samples for heavy elements and/or tritium: air, water, soil, vegetation, and agricultural products. (Flow charts illustrating sample analyses for heavy elements and tritium are to be found in Figures 2 and 7, respectively.)

Weekly air filter samples are collected from the laboratory perimeter, Site 300, and other locations near the laboratory¹ and composited monthly. Perimeter and Site 300 air filters undergo gamma analysis followed by analysis for 239,240 Pu, 238 Pu, 235 U, and 238 U. Filters from other stations are analyzed only for 239,240 Pu and 238 Pu. Air moisture is collected from eight sites around the laboratory perimeter by pulling a constant air flow through a reservoir of silica gel desiccant. These weekly samples are processed to release the water which is subsequently analyzed for tritium.

Daily water samples are collected from the laboratory's main sewage line and also from Livermore's Liquid Waste Reprocessing Plant (LWRP). These samples are composited monthly and analyzed for ^{239,240}Pu, ²³⁸Pu and ¹³⁷Cs. A third water sample, composed of weekly, one-gallon Laboratory tap water aliquots, is analyzed monthly for these same nuclides. Tritium analysis is done on water samples collected quarterly from Site 300 and the Livermore Valley. Samples may originate from wells, creeks, or ponds. Monthly rain water samples are analyzed when available. Tap water is analyzed on a quarterly basis. Annual soil samples originate from Site 300 and locations selected at random throughout the Livermore Valley. Each sample is given a gamma scan, then analyzed for ^{239,240}Pu and ²³⁸Pu.

Monthly vegetation samples originating from Site 300 and the Livermore Valley are composited quarterly and freeze dried to extract their water for tritium analysis. Goat milk is collected from local farms each month, concentrated by evaporation, and analyzed by gamma spectrometry; an aliquot of the same goat milk sample is vacuum distilled for subsequent tritium analysis of the extracted water. Locally harvested honey is combusted in a Parr bomb and the resulting water is analyzed for ³H. Wines from Livermore Valley vineyards, plus controls from other areas, are processed in a Petersen furnace and analyzed for ³H.

B. Laboratory Facilities

A primary concern when maintaining an environmental-level radiochemical facility is the elimination of contaminants. Several methods are employed to achieve this. The Heavy Elements and Tritium Laboratories are located in a section of the building that is

dedicated exclusively to low-level work. Entry is controlled, and visitors don disposable plastic shoe covers. Hood and counter surfaces are rountinely cleaned and the floor is periodically mopped.

Because of tritium's rapid environmental exchange rate, special precautions are taken to maintain a low background in the Tritium laboratory. Luminous dial watches are excluded from the facility because they can release levels of tritium much higher than levels measured in routine environmental samples. For handling samples with very low tritium concentrations, argon-atmosphere glove boxes are used. All glassware used for tritium analyses is scrupulously cleaned, dried in an oven at 105°C, and then cooled in an argon atmosphere. Room air moisture is analyzed daily for tritium.

Pieces of equipment such as drying ovens, ashing furnaces, and balances are maintained and reserved for low-level samples. The Heavy Elements Laboratory is provided with an acid fume scrubber where fumes evolved from perchloric and hydrofluoric acids are washed with a sodium hydoxide solution and then passed through an electrostatic precipitator before being exhausted. Finally, all chemicals used are analytical reagent grade, and all water is distilled and deionized.

C. Counting Facilities

1. Gamma Analysis

Samples requiring low-level gamma-spectral analysis are counted either on a 15% relative efficiency Ge(Li) spectrometer equipped with a compton suppression system² or on a 20% relative efficiency Ge(Li) spectrometer. Both spectrometers are constructed of materials especially selected for minimal radioactivity in order to attain the lowest possible background (Table 1). Data are accumulated by an

LSI-11-controlled Canberra 80 multichannel analyzer with individual amplifiers and ADCs. Spectra are computer analyzed on a CDC-7600 with GAMANAL,³ a program designed for analysis of high-resolution gamma-ray spectra. The counting efficiencies of these systems vary as a function of the gamma-ray energies as can be seen in Fig. 1. Typical detection limits for nuclides can be determined from the information given in Fig. 1 and Table 1.

2. Alpha Analysis

Alpha pulse height analysis is used to detemine 239,240 Pu and 238 Pu in environmental samples. We use twelve Ge(Li) surface barrier detectors with 450 mm² area and 20% counting efficiency with three Nuclear Data (Model 130) 512 channel analyzers. Samples are normally counted for about 5000 min. which gives an optimal detection limit of approximately 4 x 10⁻³ pCi per sample. The accumulated spectral data is transferred from the analyzers to paper tape which is read into a LSI 11 computer system for analysis. ALPHAQ, a program designed for plutonium alpha spectra, deciphers the spectra.

3. Beta Analysis

Beta counting of ¹³⁷Cs is performed on CsPtCl₆ precipitates deposited in a 1.8-cm² circular area on 2.5-cm-diameter glass fiber filter paper discs.

Energy (KeV)	Probable Source	<u>Compton Supp</u> Suppressed (counts/min)	ression System Unsuppressed (counts/min)	20% Detector System (counts/min)
77.1	228 _{Th}		0.27	··
87.4	²²⁸ Th	0.006	-	-
186.1	226 _{Ra}	0.005	-	-
238.6	228 _{Th}	0.039	0.031	0.020
295.2	226 _{Ra}	0.007	0.009	0.068
351.9	226 _{Ra}	0.016	0.017	0.111
511.0	Annihilation	0.023	0.101	0.419
583.1	²²⁸ Th	0.003	0.013	0.011
609.3	226 _{Ra}	0.005	0.016	0.086
661.6	137 _{Cs}	0.005	0.004	-
727.3	228 _{Th}	-	0.003	-
860.5	228 _{rb}	-	0.002	-
911.1	228 ₀	0.003	0.006	0.008
968.9	228 ₄	0.003	0.004	-
1119.5	228 _{Ac}	0.002	0.005	0.017
1460.8	40,	0.041	0.045	0.045
1764.5	226 _{Ra}	0.005	0.005	0.015

TABLE 1. Backgrounds for the compton suppression and 20% detector system.

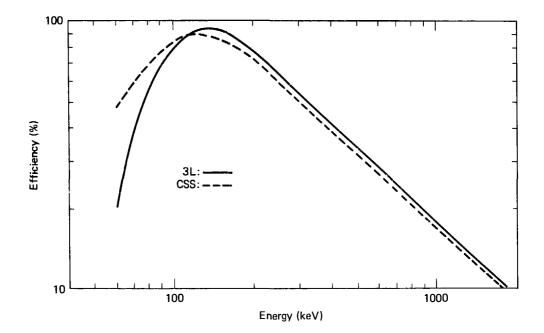


Figure 1. Counting efficiency vs energy for the compton suppression system (CSS) and the 20% system (3L).

Counting efficiencies decrease from about 31% to about 21% as the sample weight increases from 1 to 50 mg. The counting system, a low-background proportional counter with an Amperex Cosmic Ray Counter as anticoincidence shield,⁴ is equipped with an automated sample changer. Each sample is counted for 14 90-min. intervals. Data are analyzed on a CDC-7600 computer using the beta reduction program BRUNHILDE.⁵

4. Mass Spectrometry Analysis

Uranium is analyzed on a VG 354 mass spectrometer, which is a 90^{0} single magnetic-sector instrument that has two directional focusing and focal planes with an effective radius of 54 cm. Ions are detected with an electron multiplier of poly design, which is in turn read by a high-resolution digital voltmeter. Analyses are conducted in an automatic mode using a HP 9845 calculator resulting in an output of the isotopic ratios. This system is able to assess quantitatively the uranium content and isotopic composition of a 10-ng sample of uranium.

5. Tritium Analysis

Routine tritium samples are counted by liquid scintillation spectrometry. Low-level samples are electrolytically enriched⁶ prior to counting, and a single 5-ml aliquot of the enriched water is analyzed. For unenriched samples, either two 7-ml or three 5-m aliquots are used. Each counting set includes a total of eight standards (diluted to 5, 6, and 7 ml) to establish guench corrections,

and eight blanks for background determination. Sample, standard, and background aliquots are pipetted into low-potassium liquid scintillation vials. Under controlled (red/yellow) light, 10 ml of Packard Instagel scintillation cocktail are added to each vial and mixed thoroughly. The set is loaded into the spectrometer with blanks and standards interspersed among the samples and is allowed to reach temperature equilibrium. The set is then counted for three cycles of 50 min. per vial. Data from the spectrometer are analyzed on a CDC-7600 computer by the program SCNTCT.⁷

For low-volume or low-activity samples, or where radid 3 H analysis is required, a gas proportional counter is used.⁸ Water samples are converted to hydrogen gas by reaction with hot magnesium turnings under vacuum, and the hydrogen is collected in a charcoal trap at liquid nitrogen temperature. When the sample has been warmed to room temperature, it may be transferred into the proportional counter.⁹ Samples with activity of less than 3 counts/min. are counted overnight, 3-8 counts/min. samples are counted all day, and samples with more than 8 counts/min. are counted only long enough to accumulate 1000 counts. The efficiency of the system is updated at least once a month with a tritium standard, and the background (< 0.7 counts/min.) is measured over weekends. Sample results are calculated on the CDC-7600 computer with the program GASCT; the file GASLIB contains standard and background information used in the calculation and is updated periodically. Routine detection limits are approximately 2 TU (6.5 x 10^{-3} pCi/ml) enriched and 50 TU (1.6 x 10^{-1} pCi/ml) unenriched for liquid scintillation counting, and 0.2 TU (6.5 x 10-4 pCi/ml) enriched and 5 TU (1.6 x 10^{-2} pCi/ml) unenriched for gas proportional counting.

II. METHODS OF SAMPLE PREPARATION ANALYSIS

A. Heavy Elements Analysis

Flow charts illustrating procedures used in heavy elements analysis are shown in Figs. 2A and 2B.

1. Tracers

As a convenience, tracers and carriers used in plutonium, uranium, and cesium analyses are combined into a single mixed tracer. The mixed tracer is diluted with 2 \underline{M} HNO₃ to provide the following in a standard 10-ml aliquot:

- a) 1.3 pCi ²⁴²Pu
- b) 4.36 x 10^{13} atoms 233
- c) 25 mg CsCl For ¹³⁷Cs gamma-analysis a 12.3-dpm/ml ¹³⁴Cs tracer is used.

2. Air Filter Samples: Counting and Preliminary Chemistry

- a. Preliminary Treatment and Counting
 - Composite the month's air filters from the laboratory perimeter stations and seal in a 216-cm³ aluminum can for gamma analysis. Upon completion of a 1500-min. count, separate the air filters, by station, into platinum crucibles. Care should be taken to minimize cross-contamination.
 - 2) Composite the month's air filters from the Site 300 stations and seal in a 216-cm³ aluminum can for gamma analysis. Upon completion of a 1500-min. count, place all of the Site 300 air filters into a single Pyrex beaker.

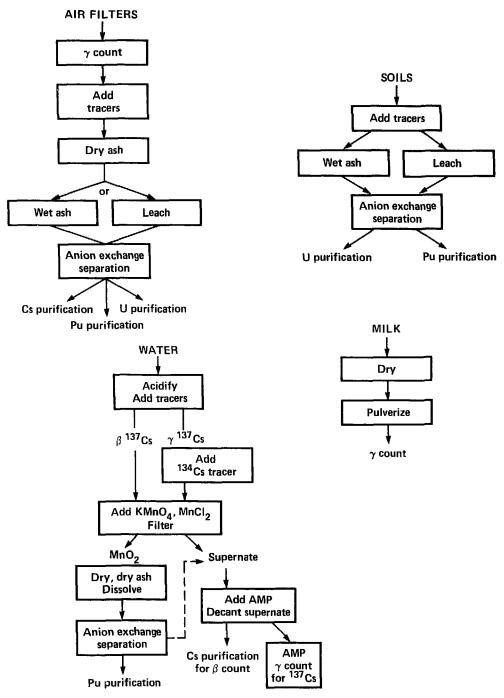


Figure 2A. Flow chart of the heavy elements separation procedures.

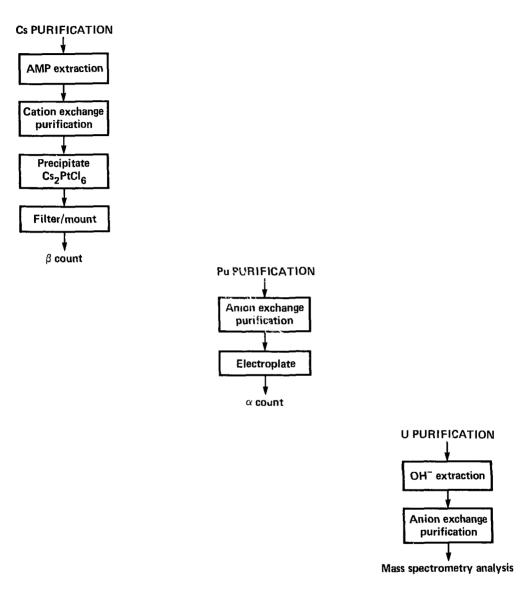


Figure 2B. Flow chart of the heavy elements purification procedures.

- Place other non-gamma counted Livermore Valley air filters into individual Pyrex beakers by station.
- 4) Add 10 ml of mixed tracer to each sample.
- Dry the air filters in a drying oven at 110°C for at least four hours.
- 6) Ash the air filters at 475°C for at least two days.
- b. Uranium Separation
 - For samples that require uranium analysis (LLNL perimeter, Site 300, standards, and blanks):
 - Transfer ashed samples from Pyrex beakers into platinum beakers with minimum conc nitric acid (HNO₃).
 Add conc HNO₃ to a total volume of 30 ml to all platinum beakers.
 - 3) Add 30 ml of conc hydrofluoric acid (HF).
 - 4) Heat the sample to dryness in the acid fume scrubber.
 - 5) Add 30 ml each of conc HNO_3 and conc perchloric acid $(HC1O_4)$.
 - 6) Heat until fuming ceases, in the acid fume scrubber.
 - 7) Dissolve as much of the residue as possible by heating with 20 ml of 8 \underline{M} HNO₃ and a few drops of 30% hydrogen peroxide (H₂O₂). Cool.
 - 8) Transfer the solution to a 250-ml plastic bottle using minimal 8 \underline{M} HNO₃.

c. Plutonium Separation

For samples that require only plutonium analysis (Livermore Valley):

- 1) Treat the ashed filter with 60 ml of conc HNO_3 and 20 ml of conc hydrochloric acid (HCl).
- Evaporate the sample to a final volume of 50 ml.
- 3) Transfer the sample to a 250-ml plastic bottle using minimal 8 \underline{M} HNO₂.
- 4) Make the sample approximately 8 \underline{M} HNO₃ by doubling the volume with 0.5 \underline{M} HNO₃.
- d. Preparation for Anion Exchange Separation
 - Treat all samples with 0.5 g of sodium nitrite (NaNO₂) to adjust the plutonium valence to +4 for anion exchange, and wait at least four hours before further processing.
 - Proceed to Section IIA-6 for anion exchange separation and heavy element purification procedures.

3. Water Samples

- a. Preliminary Chemistry
 - 1) For laboratory sewage and LWRP samples: add 5 ml of 3 <u>M</u> HCl to each 500-ml alliquot before combining these for the monthly composite. Add 10 ml of mixed tracer to the composite sample. If yielding 137 Cs by $_{\rm Y}$ counting add l ml 134 Cs tracer.
 - 2) For tap water samples: add 150 ml of 3 <u>M</u> HCl to the sample vessel followed by 10 ml of mixed tracer. If yielding 137Cs by $_{\rm Y}$ counting add 1 ml 134Cs tracer. Combine the weekly tap water aliquots in the vessel with stirring.

- b. <u>Plutonium</u> Separation¹¹
 - 1) Add 10 ml of saturated potassium permanganate ($KMnO_4$) to the sample.
 - Add 70 ml of 10 <u>M</u> NaOH and check the pH to make sure that it is basic.
 - 3) Coprecipitate plutonium with manganese dioxide (MnO₂) by slowly adding 150 ml of 0.1 <u>M</u> manganous chloride (MnCl₂). As MnCl₂ is added, maintain a basic pH by addition of small amounts of 3 M NaOH.
 - 4) Pump the sample through a $1-\mu$ cotton filter cartridge to remove the MnO₂ from solution. Save the filtrate. In the laboratory sewage sample, solid material is present before separation. This matter has been found to contain a significant percentage of the 137Cs in the sample. Therefore, cesium is later separated from this filter, by anion exchange (see Section IIA-6C).
 - Separate the cotton filter from the filter's plastic core and place it in a one liter beaker.
 - 6) Dry the filter in an oven at 110°C for two days.
 - 7) Ash the filter at 475°C for two days.
 - 8) Add 100 ml of conc $HN\dot{v}_3$ to the ashed $Mn\ddot{v}_2$ cartridge and heat for at least an hour.
 - 9) Add 30% H₂O₂ until the MnO₂ has dissolved.
 - Evaporate the sample to about 50 ml. Transfer the sample to a 250-ml plastic bottle.
 - 11) Make the sample approximately 8 \underline{M} HNO₃ by doubling the sample volume with 0.5 \underline{M} HNO₃.

- 12) Add approximately 0.5 g of NaNO₂ to prepare for anion exchange. Wait at least four hours before continuing processing.
- 13) Proceed to Section IIA-6 for anion exchange separation and heavy element purification procedures.
- c. <u>Cesium Separation</u>¹²
 - 1) For laboratory sewage samples:
 - a) Obtain the cesium eluate from the anion exchange separation performed on the MnO₂ cartridge fraction.
 - b) Add 10 M NaOH until basic.
 - c) Centrifuge, wash twice, combine supernatant liquids, and discard the solids.
 - d) Add the supernatant liquid to the plutonium separation filtrate.
 - Add 150 ml of 3 <u>M</u> HCl to the plutonium separation filtrate (from step 4 in plutonium separation above) and verify that the pH is 1-3.
 - 3) Add 5 g of AMP as a distilled $H_{2}O$ slurry.
 - 4) Allow the AMP to settle overnight.
 - 5) Pump the supernant liquid out and discard, being careful not to remove any AMP.
 - 6) Transfer the AMP with pH 1-3 HCl into a one-liter breaker.
 - Allow the AMP to settle overnight and discard the supernate.
 - a) For gamma analysis of ¹³⁷Cs:
 - Transfer the AMP with pH 1-3 HCl to a 40-ml glass centrifuge tube, centrifuge, and discard the supernant liquid.

2) Slurry the AMP with minimum pH 1-3 HCl,

transferring with a micro pipet to a prindle vial.

- 3) Bring the total volume to 20 ml with pH 1-3 HC1.
- 4) Proceed with gamma counting.
- b) For ß counting:
 - 1) Dissolve the AMP with 10 M NaOH.
 - Centrifuge, wash twice, combine supernatant liquids and discard the solids.
 - Acidify the supernatant liquid to pH 1-3 with 3 M HCl.

Note: If a white silicate precipitate appears, adjust the pH to 5-8 with 1 \underline{M} NaOH, centrifuge to remove the precipitate, and repeat step 3.

- 4) Dilute the sample to 500 ml with pH 1-3 HC1.
- 5) Add one (1) gram of AMP as a slurry and stir.
- 6) Allow the AMP to settle overnight.
- Proceed to Section IIA-6e for cesium purification and mounting procedures starting with step 13.
- 4. Soil Samples
 - a. Preliminary Treatment and Counting
 - 1) Dry the soil sample at 110°C for at least two days.
 - Remove rocks and pebbles with diameters greater than 0.5 cm.
 - 3) Pulverize the remaining material in a grinding mill.
 - Pass the sample through a 32-mesh sieve. Discard the fraction that is greater than 32 mesh.
 - 5) Thoroughly blend the sample.

- Pack a 216-cm³ tared aluminum can with the sieved soil for gamma analysis.
- b. <u>Plutonium Separation</u>

(Acid leach method¹³)

- 1) Weigh a 100-g aliquot of the prepared soil.
- 2) Add 10 ml of the mixed tracer.
- Leach the sample by heating for four hours with 300 ml of conc HNO₃ and 100 ml of conc HCl. Control foaming with a few drops of n-octanol.
- 4) Vacuum filter the hot sample through a buchner funnel fitted with 934 AH glass fiber filter paper. The supernatant liquid should be clear. Wash the sample twice with approximately 50 ml hot 8 <u>M</u> HNO₃ each wash.
- 5) Determine the molarity of the supernatant liquid by titrating a 0.5-ml aliquot with standardized 1 \underline{M} NaOH to a phenolphthalein end point.
- 6) Adjust the molarity of the solution to 8 \underline{M} HNO₃ with appropriate volume of 0.5 \underline{M} HNO₃.
- Add about 1 gram of NaNO₂ to the sample to prepare for anion exchange. Wait at least four hours before continuing processing.
- Proceed to Section IIA-6 for anion exchange separation and Pu purification procedures.
- c. Uranium Separation
 - 1) Weigh 1 gram of prepared soil into a platinum beaker.
 - 2) Add 10 ml of the mixed tracer.
 - Add 10 ml of conc HNO₂.
 - 4) Add 10 ml of conc HF.

- 5) Heat the sample to near dryness in the acid fume scrubber.
- Reconstitute the sample with an additional 10 ml of conc HF.
- 7) Evaporate the sample to dryness.
- 8) Treat the residue with 20 ml of conc HNO3.
- 9) Add 20 ml of conc $HC10_A$.
- 10) Heat the sample until the white fumes of HClO₄ are no longer evolved in the acid fume scrubber.
- 11) Dissolve the residue with a minimum volume of 8 \underline{M} HNO_3 and 2-4 ml of 30% $\rm H_2O_2$ and heat.
- 12) Transfer the sample to a 40 ml Pyrex centrifuge tube with minimum $\rm H_2C$.
- 13) Proceed to Section IIA-6A for anion exchange separation (using air filter resin and wash amounts), Pu, and U purification procedure.

5. Milk Analysis

- 1) Pour 3 liters of milk into a tared 4-liter beaker.
- Evaporate the milk sample slowly, with continuous stirring, to approximately 0.5 liter.
- 3) Dry the milk for two days at 110°C.
- 4) Weigh the dried milk solids.
- 5) Pulverize the milk solids in a blender.
- Fill a tared 216-cm³ aluminum can with the pulverized sample.
- 7) Weigh the can for gamma analysis, and gamma count.

6. <u>Anion Exchange Separation and Heavy Element Purification</u> Procedures

It is evident from flow charts of the various purification procedures (Fig. 2A & 2B) that, excluding the milk chemistry, each purification involves an ion exchange separation. Four different column sizes are used for ion exchange (Fig. 3). The techniques of column preparation and ion exchange separations are similar for all procedures, differing only in the size of the columns used and in the amounts of necessary reagents.

a. Resin Preconditioning

- Precondition AG 1-X8, 50-100 mesh (C1⁻ form) and AG
 1-X8, 100-200 (C1⁻ form) mesh anion exchange resins by putting the resin in a 2-liter buchner funnel fitted into a 4-liter filter flask, and
 - a) washing with 8 \underline{M} HNO₃ (2 liters acid per liter resin).
 - b) washing with conc HCl (2 liters acid per liter resin).
 - c) Repeatedly washing with H₂O until the eluate has a neutral pH.
- Prepare anion exchange resin for uranium purification as follows¹³:
 - a) Grade AG 1-X8, 100-200 mesh resin in a counterflow fluid grader, collecting the 30-70 mm/min fraction.
 - b) Wash about 250 ml of the resin with 2 M NaOH.
 - c) Rinse the resin with 8 liters of $H_{2}O$.

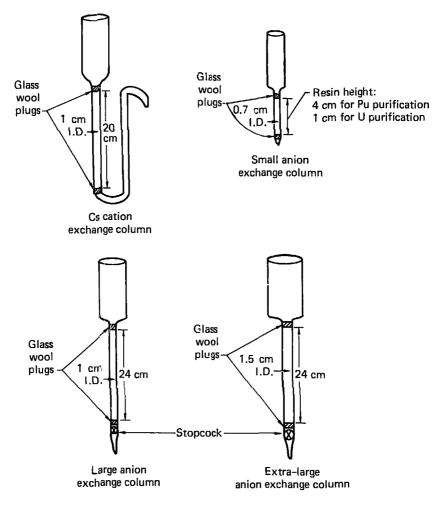


Figure 3. Columns used for ion exchange.

- d) Convert the resin to sulfate form with 1 \underline{M} H₂SO₄.
- e) Wash the resin with 4 liter of 0.06 \underline{M} H₂SO₄.
- Precondition Bio-Rex 40, 20-50 mesh cation exchange resin for cesium purification with repeated 5% NaCl washes until the eluate is clear.

b. <u>Column Loading</u>

- 1) Fill the column with $H_{2}O$.
- Insert a glass wool plug into the bottom of the column to prevent resin loss. Eliminate any air bubbles from this plug.
- 3) Load the required amount of ion exchange resin as an H_2O slurry and allow the resin to settle.
- Insert a glass wool plug into the top of the column.
 Eliminate any air bubbles from the plug before pressing it onto the top of the resin.
- 5) Rinse out the column with H_2^0 to remove any loose resin.

c. Anion Exchange Separation

See Table 2 for the appropriate volumes of reagents to be used for each particular sample type.

 Extra-large columns should be used for soil samples, while large columns should be used for water and air filter samples. Load the appropriate size column with preconditioned 1-X8, 50-100 mesh (C1⁻ form) anion exchange resin to a height of about 24 cm, followed by 1 cm of preconditioned AG1-X8 100-200 mesh (C1⁻ form) anion exchange resin.

 Condition the column with the appropriate volume of 8 <u>M</u> HNO₃.

- 3) Filter the sample through 934 AH glass fiber filter paper onto the column and, if cesium analysis is desired, collect the eluate. The eluate from this step and that from the next step comprise the cesium fraction (proceed to Section IIA-6e), for β-counted cesium purification and mounting).
- 4) When the liquid has run through the column, rinse the column reservoir twice with 8 \underline{M} HNO₃; then wash the resin with the appropriate volume of 8 \underline{M} HNO₃.
- 5) Rinse the column reservoir with conc HCl, then wash the resin with the appropriate volume of conc HCl to remove thorium contamination.
- 6) Elute plutonium with the appropriate volume of freshly prepared NH₄I-HCl solution (1.5 g NH₄I per 100 ml conc HCl). Add several ml conc HNO₃ and evaporate to dryness. Proceed to Section IIA-6d for plutonium purification processing.

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7) Elute uranium with the appropriate volume of 0.1 \underline{M} HCl add \sim 5 ml of conc HNO₃, evaporate to dryness. Proceed to Section IIA-6f for uranium purification processing.

TABLE 2. Reagent volumes used in anion exchange separations.

Sample type	<u>Column size</u>	8M HNO ₃ conditions <u>step (ml)</u>	8M HNO ₃ wash_(m1)	Conc HCl wash (ml)	NH4I/ HC1 (m1)	0.1M HCl (ml)
air filter	large	50	50	100	80	80
water	large	50	50	100	80	
soil	x-large	100	300	200	150	

d. <u>Plutonium Purification and Plating</u>¹⁴

Purification:

- 1) Dissolve the residue by reating with a few ml of conc HCl and several drops of 30% H_2O_2 . Make sure that excess H_2O_2 has been evaporated before proceeding.
- Transfer the solution to a 40-ml plastic centrifuge tube with a minimum amount of conc HCl.
- 3) Add 0.5 g of NaNO₂.
- Cap the tube and centrifuge to remove the insoluble sodium chloride (NaCl) formed by the NaNO₂ addition.
- 5) Load a small ion exchange column to a height of about 4 cm with preconditioned Dowex 50-100 mesh anion exchange resin (C1⁻ form).
- 6) Condition the resin with 20 ml of conc HCl.
- Load the sample onto the column. Repeat the centrifuging and loading procedures twice.
- 8) When the liquid has run through the column, rinse the sides of the column reservoir with conc HCl, then wash the resin with 20 ml of conc HCl.

- 9) Rinse the column reservoir with 8 \underline{M} HNO₃ and then wash the resin with 20 ml of 8 \underline{M} NHO₃.
- Re-rinse the column reservoir with conc HCl and wash the resin with another 20 ml of conc HCl.
- 11) Elute the plutonium with 20 ml of freshly prepared NH_4I-HCl_4 solution (1.5 g NH_4I per 100 ml conc HCl).
- 12) Add several ml conc HNO_3 to the eluate from step ll and evaporate to dryness.

Plating:

- 13) Assemble the electroplating cell 14,15 (Fig. 4).
 - a) Clean a polished 2.54-cm-diam. x 0.05-cm-thick stainless steel disc, engraved with the sample identification number.
 - b) Place the disc at the bottom of a stainless steel base to serve as the cathode.
 - c) Place a teflon washer on top of the disc and secure a labelled glass chimney in place with a teflon support, stainless steel ring, and brass wing nuts to form a watertight seal around disc.
 - d) Fill the cell with water to test for leakage.
- 14) Add 1 ml conc sulfuric acid (H_2SO_4) to the residual salts of plutonium eluate.
- Heat the sample until copious white fumes are evolved.
- 16) When the sample has cooled, dilute with 2 ml of l \underline{M} H₂SO₄.

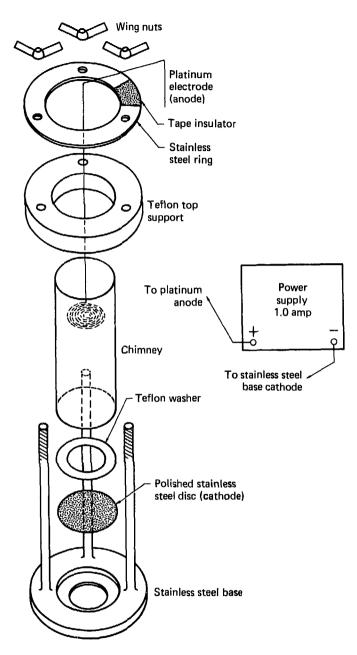


Figure 4. The plutonium electroplating cell.

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- 17) Add two drops of 0.1 M methyl red indicator.
- 18) Transfer the sample to the electroplating cell with a minimum volume of $1 \text{ M} \text{ H}_2\text{SO}_4$.
- 19) Titrate the sample to a yellow end point (pH 6.2) with conc NH₀OH.
- 20) Back titrate five drops past the red end point (pH 4.4) with 1 \underline{M} H₂SO₄.
- Use a 0.064-cm-thick platinum wire, coiled at the bottom, as the anode.
- 22) Electroplate at 1.0 amp for 70-90 min.
- 23) Before completion, add 1 ml of conc NH_4OH to the cell and continue plating for one min.
- 24) As quickly as possible:
 - a) disengage the power supply;
 - b) remove the anode;
 - c) discard the solution;
 - d) rinse the cell with 0.1 M NH₄OH;
 - e) disassemble the cell;
 - f) rinse the disc with 0.1 <u>M</u> NH_AOH;
 - g) rinse the disc with acetone containing \sim 5 ml 0.1 <u>M</u> NH_AOH.
- 25) Determine ^{239,240}Pu and ²³⁸Pu on the disc by alpha spectrometry.
- e. Cesium Purification and Mounting

Purification:

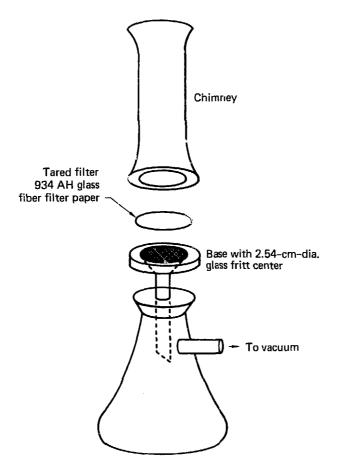
 Add sufficient amount of 10 <u>M</u> NaOH to the cesium fraction from the anion exchange separation to make the solution pH 10-12.

- 2) Centrifuge to remove any Fe(OH), precipitate.
- 3) Decant the supernatant liquid into a large beaker.
- Dissolve the Fe(OH)₃ in a minimum amount of 8 <u>M</u> HNO₃.
- 5) Dilute the solution with about 10 ml of H_20 .
- 6) Re-precipitate $Fe(OH)_3$ with 10 <u>M</u> NaOH.
- Centrifuge and combine the supernatant liquid.
- Repeat steps 4 through 7 once.
- Acidify the supernatant liquid with 3 M HCl to pH 1-3.
- 10) Dilute the sample to 600 ml with H_20 .
- 11) Add 1 g of ammonium molybdophosphate (AMP) as a slurry with $H_2^{0.12}$ Cesium in the solution will complex with the AMP.
- 12) Allow the sample to equilibrate and the AMP to settle overnight.
- Decant and discard the supernatant liquid.
- 14) Transfer the AMP complex to a 40-ml Pyrex centrifuge tube with a minimum amount of pH 2-3 HCl.
- Centrifuge and discard the supernatant liquid.
- 16) Dissolve the AMP complex with a combination of 1 ml H₂O, 1 ml of a 0.75 <u>M</u> NaOH - 2% EDTA solution, and 1 ml 10 <u>M</u> NaOH.
- 17) Load preconditioned Bio-Rex 40, 20-50-mesh cation exchange resin into a cesium ion-exchange column to a height of approximately 20 cm.
- Prime the cation exchange resin with 250 ml of 5% NaCl.
- 19) Slowly load the sample by micropipet.
- 20) After the liquid has drained from the reservoir, rinse the column sides with three 2-ml aliquots of H₂O.

- 21) Wash the resin with 45 ml of $H_{2}0$.
- 22) Wash the column with 200 ml of 0.75 <u>M</u> HCl administered in 50-ml aliquots.
- 23) Elute the cesium with 125 ml of 3.0 M HCl.
- 24) To permit reuse of the cation exchange resin, wash the resin with 250 ml of H_2O . This resin can be reused up to five times.
- 25) Evaporate the cesium eluate to dryness.

Mounting:

- 26) Prepare a tared filter disc:
 - a) Cut a 2.54-cm-diam 934 AH glass fiber filter paper disc.
 - b) Wash the disc with H_20 , then acetone.
 - c) Dry the disc under infrared light.
 - d) Weigh the disc to + 0.1 mg.
- 27) Dissolve the residual salts in the cesium eluate with 2-3 ml of 3 M HCl and heat.
- 28) Transfer the solution to a 40-ml plastic centrifuge tube with minimum $H_{2}O$.
- 29) Make the solution basic by adding 3 M NaOH.
- 30) Add 4 ml 0.12 <u>M</u> chloroplatinic acid (H_2PtCl_6) . Cesium will be precipitated as Cs_2PtCl_6 . If this does not occur instantly, place the centrifuge tube in an ultrasonic bath for about 30 s.
- 31) Cool the sample in an ice bath for 30-40 min.
- 32) Assemble the cesium filtering apparatus (Fig. 5).
- 33) Slowly filter the sample through the tared disc depositing the Cs₂PtCl₆.



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Figure 5. The cesium filtering apparatus.

- 34) Remove the chimney from the apparatus.
- 35) Rinse the disc (under vacuum) with 2 ml of cold H₂O.
- 36) Rinse the disc with 2 ml of cold acetone.
- 37) Dry the filter under infrared light.
- Weigh the sample.
- 39) Mount the disc on a plastic base with a plastic ring holding a mylar cover.
- Determine the cesium by beta spectrometry with a weight yield correction.
- f. <u>Uranium Purification</u>¹⁶
 - Dissolve the sample in 1-2 ml conc HNO₃ with heat and transfer with water to a 50-ml glass centrifuge tube.
 - Add 1 ml (5 mg/ml) lanthanum carrier to all uranium samples except those that may have hydroxides present.
 - 3) Make the sample basic with conc NH₄OH. Uranium is coprecipitated with the La(OH)₃. Soil samples may contain enough iron to coprecipitate uranium with Fe(OH)₃ in those samples.
 - Heat the sample in an 80°C water bath for at least five minutes.
 - 5) Centrifuge and discard the supernatant liquid.
 - 6) Dissolve the La(OH)₃ (or Fe(OH)₃) with a minimum of $1 \frac{M}{2} H_2 SO_4$.
 - 7) Dilute the solution to 5 ml with H₂O.
 - 8) Repeat steps 3 through 7 twice.
 - 9) After dissolving the La(OH)₃ and diluting to 5 ml a third time, dilute to 15 ml with 0.06 \underline{M} H₂SO₄.

- 10) Load a small ion exchange column to a height of 2 cm with preconditioned AG1-X8, 100-200 mesh uranium anion exchange resin (sulfate form).
- 11) Condition the resin with 10 ml of 0.06 \underline{M} H₂SO₄.
- 12) Load the sample into the column.
- 13) When the liquid has run through the column, rinse the column reservoir four times with 5 ml of $0.06 \text{ M} \text{ H}_2\text{SO}_4$, allowing the reservoir to drain completely between rinses.
- 14) Wash the resin with 5 ml of conc HCl.
- 15) Wash the resin with 5 ml of 6 M HCl.
- 16) Elute the uranium into a labeled 12-ml conical centrifuge tube with 10 ml of 0.1 M HCl.
- Determine uranium in the ion exchange eluate by mass spectrometry.

B. <u>Tritium Analysis</u>

A flow chart illustrating sample processing procedures used in tritium analysis is shown in Fig. 7.

- 1. Air Moisture
 - a. Reservoir Preparation
 - Bake silica gel desiccant in a vacuum oven at 100°C for at least 24 hours, then cool and store in an argon atmosphere.
 - Fill a clean reservoir vessel (Fig. 6, below) with silica gel in an argon atmosphere.
 - Plug the neck with glass wool and seal the reservoir with a stopper.

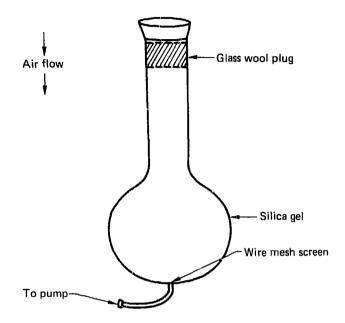


Figure 6. The silica gel reservoir.

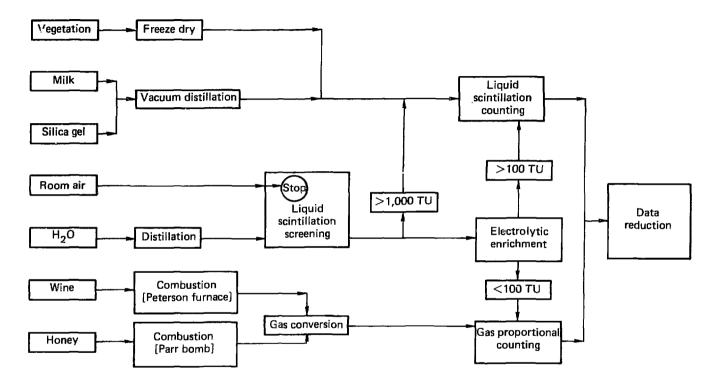


Figure 7. Low-level tritium determination.

b. Air Moisture Sample Processing

- After the silica gel has been used to sample atmospheric moisture, determine the weight of the silica gel sample that is contained in the reservoir.
- 2) Mix the silica gel thoroughly to achieve homogeneity.
- Remove an aliquot of about 300 cc and determine its precise weight.
- 4) Transfer the silica gel aliquot to a freeze dry jar.
- Construct the freeze dry assembly (Fig. 8): Caution: Vacuum grease should not be used.
 - Attach the trap to the vacuum port with a triangle seal and three wing nuts.
 - ii) Plug the other trap opening with a rubber stopper.
 - iii) Evacuate the trap and check for leaks.
 - iv) Fill the trap's reservoir 1/3 full with Dowanol (or equivalent).
 - v) Slowly add dry ice (CO_2) chunks to the reservoir until full.
 - vi) Insert a coarse 11.0-cm filter paper and a 9-cm screen into the bottom of the adapter.
 - vii) Attach o-rings to the neck of the adapter and the jar.
 - viii) Backfill the trap with argon and remove the stopper.
 - ix) Insert the adapter into the trap while simultaneously opening the vacuum valve. Vacuum will hold the apparatus together.

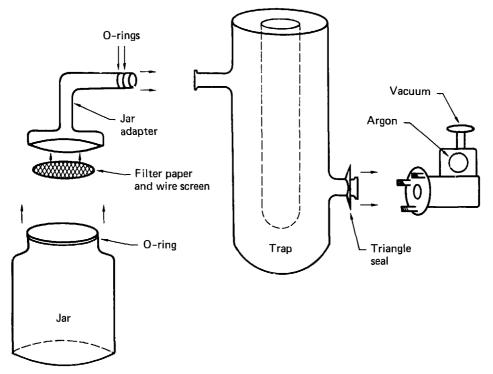


Figure 8. The freeze dry assembly.

- 6) Pump the assembly down, valve off, and check for leaks.
- After the pressure has stabilized, envelop the jar in a heating mantle set for 100-110°C.
- 8) Continue to heat the sample for at least 24 hours. Add dry ice as needed and evacuate the freeze dry assemblies individually every two hours until the pressure stabilizes.
- Backfill the trap with argon, remove the jar, and seal the opened trap end with a paraffin sheet.
- 10) Allow the silica gel to cool in a closed, tared container, and then reweigh to determine the amount of water lost.
- Remove the Dowanol from the trap and collect the melt water from the ice that has formed on the cold finger.
- Determine the tritium concentration in this water by liquid scintillation spectrometry.

2. Water Samples

The tritium concentration in water is normally too low to permit a direct liquid scintillation or proportional beta analysis. These samples must instead undergo a preliminary distillation to remove any physical or chemical contaminants followed by an electrolytic enrichment to increar 'he amount of tritium relative to the sample volume.

- a. Distillation
 - Pour approximately 500 ml of the water sample into a doublenecked, round-bottom boiling flask.

- Through a distillation top, attach an argon line to the side neck of the flask and assemble the distillation apparatus (Fig. 9).
- Attach the drain and water line to the condenser and check for leaks.
- Turn on the heating mantle (set for 100-110°C) and distill the entire sample.
- 5) Transfer the distilled sample from the receiving flask to a clean bottle and seal.

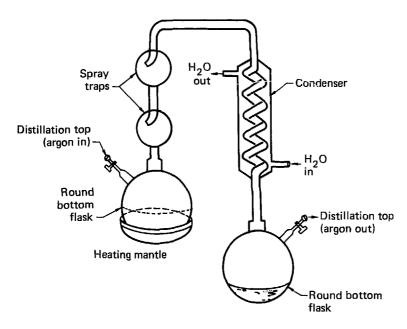


Figure 9. The water distillation apparatus.

b. Enrichment⁶

The enrichment assembly (Fig. 10), consists of an iron cathode-nickel anode unit supported in a Pyrex cell by teflon baffle plates. Additional sample is entered into the cell from a top-mounted 250-ml delivery flask reservoir. During enrichment, the sample reservoir is under an argon atmosphere while the enrichment cell is maintained at a constant 6.0°C in a water bath. An enrichment set consists of two standards, a blank, and seven to nine samples.

- 1) Insert the electrode unit into the enrichment cell.
- Add 1.5 x 10¹² moles of sodium hydroxide (as 1.6 ml of 9.4 M NaOH) to the cell.

NOTE: To avoid the tritium contamination of commercial reagent-grade sodium hydroxide, tritium-free NaOH is synthesized by dissolving 1/4 lb of sodium peroxide (Na₂O₂) in 313 ml of tritium-free water. The NaOH is stored in an argon atmosphere and periodically checked via gas conversion/gas proportional counting for its tritium level.

- Measure a 250-ml aliquot of the water sample.
- 4) Fill the enrichment cell to the 50-ml line and pour the remaining 200 ml of sample into the delivery flask.
- 5) Complete the enrichment cell assembly pulling the electrode wires through the cell head sidearm. Do not use grease around any ground glass joints.

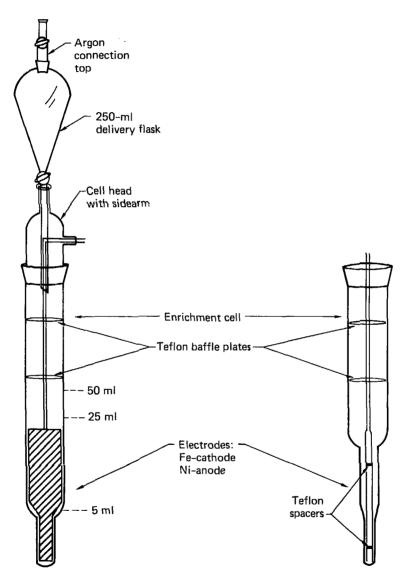


Figure 10. The enrichment cell assembly.

- 6) Immerse the enrichment cell in the 6.0°C water bath.
- Connect the electrodes to a 3-amp constant current power supply.

NOTE: The electrolysis process at 3 amps evaporates about 1 ml of solution every hour. It is therefore necessary to refill the cell to 50 ml daily. When filling the cell, connect the argon supply to the argon connection top to exclude contaminated air from the assembly. If the cell cannot be filled every day (over the weekend for example), turn the power supply down to 1 amp.

- 8) When the sample reservoir is empty (8-12 days) and the cell volume is 25 ml, electrolyze at 0.3 amp to a volume of 5-7 ml (about 10 days).
- 9) When the 5-7 ml volume is attained, disconnect the power supply, remove the enrichment assembly from the water bath, and remove the delivery flask and cell head from the enrichment cell.
- 10) Neutralize the sample by bubbling carbon dioxide through the sample. This is done to release the hydrogen that is bound in the sodium hydroxide.
- Attach the enrichment cell to a vacuum through a distillation head and a preweighed receiving bulb (Fig. 11).
- Cool the bulb with dry ice/methanol and evacuate the system.
- 13) Using a heating mantle, distill the sample in the enrichment cell, periodically relieving the pressure buildup in the system by opening the vacuum valve.

- 14) At completion, allow the receiving bulb to warm to room temperature. Reweigh to determine the final volume of water.
- 15) Enriched samples are analyzed for tritium by liquid scintillation spectrometry. A computer program, ENL184, is used to compute the enrichment factors which are entered into the final calculations.

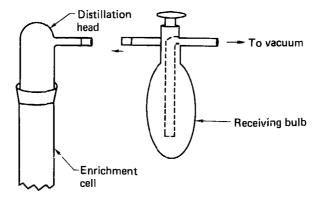


Figure 11. Enrichment cell head and receiving bulb for vacuum distillation.

3. Vegetation

Monthly vegetation samples are composited and analyzed quarterly. They are stored in plastic bags below 0°C to prevent loss or contamination of their water content.

- Construct the freeze dry assembly (Figure 7), according to instructions in Section IIB1-b.
- 2) As quickly as possible:
 - Remove sample from the freezer, cut top of the bag open, and weigh.
 - ii. Pack the bagged sample into the freeze dry jar.
 - iii. Attach the adapter to the jar.
 - iv. Backfill the trap with argon and remove the stopper.
 - v. Attach the jar and adapter to the trap, evacuate and valve off from the vacuum pump.
 - vi. Repeat for each sample.

The vegetation freeze drying process takes about four days to complete. During this time dry ice must be added to the cold fingers when needed and each sample assembly must be evacuated periodically and checked for leaks.

- Upon completion, remove the jar by backfilling the trap with argon and seal the trap opening with a paraffin sheet.
- Remove the Dowanol and dry ice from the trap. Allow the ice sample to melt.
- Reweigh the bagged vegetation sample to determine the volume of water removed.
- Transfer the water sample from the trap to an appropriate sample container.

- 8) If the volume of water in the sample exceeds 10 ml, tritium can be determined by liquid scintillation spectrometry. Otherwise, it must be determined by gas proportional counting.
- 4. Milk
 - 1) Place 25 ml of milk into a 50-ml round-bottom boiling flask.
 - Connect the boiling flask to an adapter and a receiving bulb (Fig. 12).

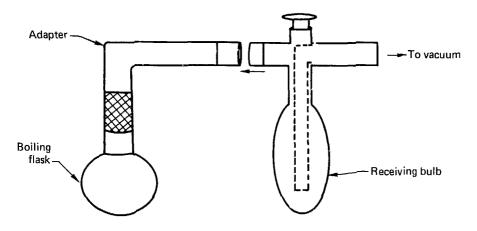


Figure 12. The milk distillation apparatus.

- Cool the bulb to -78°C with dry ice/methanol and slowly evacuate the system, taking care not to splatter milk into the receiving bulb.
- When the system is completely evacuated, warm the boiling flask in a heating mantle.
- 5) When the milk is nearly dry, remove the heating mantle and isolate the receiving bulb.
- Allow the bulb to warm to room temperature and transfer the water sample to a container.
- Analyze the water for tritium by liquid scintillation spectrometry.

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III. QUALITY ASSURANCE PROCEDURES

A. Purpose

The quality assurance procedures of the Site Environmental Monitoring Group are designed to ensure that results of sample analyses are both accurate and reproducible. An additional objective of SEMP's quality assurance program is to be able to trace results to either stored samples or stored records. Included in "QA procedures," then, are storage of samples and sample data, processing of duplicate blank and standard samples and verification of results.

B. Storage of Sample Data and Results

1. Heavy Elements Analyses

a. Sample Sheets

Laboratory worksheets are maintained for all samples. When a sample is received, initial information about the sample is recorded on the sheet. Sample number, date, type, and volume or weight, as well as the date received are recorded. Any additional relevant information about the sample is also recorded. As the sample is processed, the chemical and counting procedures used are documented on the sample sheet, as are the chemist's initials and the date of the processing. Finally, any special information or unusual circumstances concerning the sample are recorded. All sample sheets for the current year are stored in the laboratory, while those from previous years are archived.

b. Analysis Results

Results of gamma analysis are stored on magnetic tapes. Computer printouts of gamma results generated by the gamma analysis program GAMANAL are also stored.

Results from plutonium alpha analysis are stored on paper tape. Paper tape output containing alpha data and computer printouts containing both counting results and actual alpha spectra, all of which are generated by the alpha analysis program ALPHAQ, are stored permanently. Computer printouts containing beta analysis results or uranium analysis by mass spectrometry are stored permanently. Note that multiple methods of storage are used for all sample data. In addition, all samples with the exception of those analyzed for uranium are stored permanently and may be re-counted (see Section C, for information on sample storage).

2. Tritium Analysis

a. Sample Sheets

As with heavy elements analysis, laboratory worksheets are maintained for all samples analyzed for tritium. Upon receipt of a sample, sequentially numbered worksheets are filled out with preliminary sample information. Type of sample, date of receipt, originator's identification number, and any additional relevant information concerning the sample are recorded on the sample sheet. All sample sheets are filled out in duplicate; one copy remains with the sample while it is being processed, while a carbon copy is kept separately in a worksheet notebook. As the sample is processed, procedural steps are noted on the worksheets. After processing, the original copy of the sample

sheet replaces the carbon copy in the worksheet notebook. Original sample sheets are stored permanently.

b. Storage of Results

Results of all samples analyzed, either by gas proportional counting or liquid scintillation counting, are stored in a computer file called LOG. Scintillation counting results are also stored in a separate SEMP file called SCNSTR. Computer printouts of results determined by gas counting are stored with the actual sample sheets, while two different types of printouts containing liquid scintillation results are stored separately.

C. Sample Storage

1. Heavy Elements

Whenever possible, samples that have been processed and counted are stored in their final form. This ensures that samples may be re-counted and results re-calculated if there is any doubt as to the validity of original results.

- a. Gamma samples, including milk solids, soil, and special samples, are stored indefinitely in their gamma cans.
- b. Plated plutonium samples that have been alpha-counted are stored indefinitely.
- c. Cesium samples are stored on beta-counted planchets or prindle vial.
- d. Samples analyzed for uranium cannot be stored since the samples are evaporated completely in mass spectrometry analysis.

2. Tritium

Samples analyzed for tritium are stored at least until analytical results have been obtained and verified, and for as

long as is practical thereafter. Some samples may be stored in their original forms (e.g., wine, honey), while others cannot be and must be stored in some processed form (e.g., vegetation). When possible, a duplicate sample is taken initially to ensure that at a later point, a "duplicate" result may be obtained if neces_ary (see Section D3, for more information on duplication of samples).

a. Vegetation and Silica Gel Samples

Water collected from these samples is stored five to six months for routine samples and one year for special samples.

b. Milk Samples

Water collected from the distillation of routine milk samples is also stored for five to six months.

c. Water Samples

Low-tritium water (< 1000 TU) is enriched prior to liquid scintillation counting, and may therefore be stored in either the unenriched and enriched forms. Unenriched water samples are stored up to six months (the limitations here are space and equipment), while enriched samples are stored up to one year. Water that is higher in tritium content (> 1000 TU), which is not enriched, is stored up to six months after analysis is completed.

d. Wine and Honey Samples

Wine and honey samples are stored in their original form for at least six months.

D. "QA" Samples

1. EML Samples

Samples are received from the EML (U.S. Department of Energy Environmental Measurements Laboratory, New York, NY) approximately every six months and are analyzed for a variety of nuclides. Types of samples analyzed include soil, water, air filters, and vegetation. The procedures used for analyzing the soil samples are identical to those described in Section II, Methods of Sample Preparation and Analysis, for routine soil samples. Procedures used in the analysis of the EML air filters, vegetation, and water samples do not, however, exactly follow the routine procedures given in Section II, but require special processing as follows.

a. EML Air Filters

Air filters are first gamma counted in a plastic petri dish. One half of the dish is nested inside the other half, enclosing the air filter. Additional glass fiber filters are enclosed on the non-counter side to ensure that the filter to be counted is held in tightly. The entire apparatus is then taped and gamma counted. After counting, the chemical procedures used follow those used for routine samples (see Section IIA-2).

b. EML Vegetation Samples

The procedures used in processing vegetation are basically the same as those used in processing routine LLNL perimeter air filters. First, a pre-determined amount of sample is weighed out; it should contain approximately the same amount of plutonium as the tracer to be added. The approximate amount of plutonium in the sample is indicated on the

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information sheet accompanying the EML samples. Tracer is added, and the sample is dried at 105°C for at least four hours and then ashed at 475°C for at least two days. The "wet ash" procedure using HNO_3/HF followed by $HNO_3/HC1O_4$ (see Section IIA-2), is then performed twice in order to fully digest the sample.

c. EML Water Samples

Three 7-ml aliquots are removed from the 1-liter water sample and analyzed for tritium by liquid scintillation counting. The remaining water is analyzed for heavy elements, ordinarily only plutonium and cesium. The sample is diluted to four gallons total with distilled water and then analyzed according to routine procedure (see Section IIA-3). A blank, containing four gallons of distilled water, is analyzed concurrently.

d. Storage of EML Results

Computer printouts of all EML results are stored indefinitely. EML results are also entered into the data base ENV, in a table designated specifically for EML data.

2. EPA Samples

EPA water samples (U.S. Environmental Protection Agency Environmental Monitoring Systems Laboratory, Las Vegas, NV) are received every two months. They are analyzed only for tritium, by a direct liquid scintillation count; no distilling or enriching is involved. Results are stored in the SEMP computer files LOG (containing all tritium analysis results) and SCNSTR (containing all scintillation count results). Computer printouts containing all scintillation count results include EPA results.

3. Duplication of Samples

All types of samples are duplicated periodically to ensure reproducibility of results. Some samples (i.e., vegetation and other samples analyzed by freeze-drying) are always duplicated so that an alternate sample may be run in case of a processing error.

- a. Heavy Elements
 - 1) Air Filters

A duplicate sample is taken at one air filtering station every other month. Since there are six perimeter air filtering stations, and the extra sampler is moved to a different site bimonthly, all sampling stations are checked once a year.

2) Water Samples

Daily water samples are duplicated once weekly. These duplicate samples are analyzed separately.

3) Soil Samples

At least ten percent of the annual soil samples received are duplicate samples. These duplicates are analyzed with the original soil samples.

- b. Tritium
 - 1) Vegetation

Vegetation is sampled quarterly, and two vegetation samples are taken at each sampling location. Duplicate samples are not routinely analyzed, although one such sample is analyzed quarterly as a OA "check."

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2) Silica Gel Samples

A duplicate sample is collected and analyzed from a different station each month.

3) Water Samples

At least 10% of all water samples submitted for SEMP analysis are duplicate samples.

- 4. Internal Standards and Blanks
 - a. Heavy Elements
 - <u>Air Filter</u> standards prepared with stock solutions of plutonium and uranium are analyzed with routine air filters every other month. Air filter blanks are included with every month's air filter samples, and are analyzed monthly with the routine samples.
 - <u>Water</u> blanks are normally analyzed every other month; water standards are analyzed biannually. EML water samples, which are analyzed along with a "blank" (see Section IIID-1), suffice as biannual standards.
 - 3) Soil samples analyzed annually are first gamma counted, then processed chemically. An NBS soil sample is run as a gamma standard, but is not analyzed chemically. A blank comprising "washed and ignited." sand (Mallinckrodt) is run with the routine soil samples being processed.
- b) Tritium

Tritium standard solutions of approximately 5.5 x 10^{-3} µCi/ml are made at various water dilutions and serve as standards for liquid scintillation counting. Another standard solution of approximately 8.3 x 10^{-6} µCi/ml is made up from a stock solution, and is run with each enrichment set. This

standard is also used as a gas proportional counter/liquid scintillation counter cross-check.

Dead water (< 3 TU) is used as a blank for enrichment sets. Hydrogen gas is used to determine background in the gas counter.

E. Verification of Results

Once processing of a sample is completed, the result is checked against past results for the same type and location of sample. Any significant discrepancy here may necessitate recounting or possibly even reprocessing of the sample. The flow of sample data is also checked to ensure that no mistakes were made in transferring information.

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