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SAMPLING AND ANALYTICAL PROCEDURES FOR
ENVIRONMENTAL MONITORING AT LAWRENCE
LIVERMORE LABORATORY

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SAMPLING AND ANALYTICAL PROCEDURES FOR ENVIRONMENTAL MONITORING AT LAWRENCE LIVERMORE LABORATORY

ABSTRACT

Described herein are sample collection and analysis procedures employed in environmental monitoring at the Lawrence Livermore Laboratory. 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.

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 Lawrence Livermore Laboratory (LLL). 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 LLL to measure the concentration of airborne particulate radioactivity, beryllium, and tritiated water. Samplers placed at 6 perimeter locations at LLL and at 10 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 LLL 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.

Collection

Particulates

LLL perimeter and Site 300 particulate samples are collected on $5.2 \times 10^{-2} \text{ m}^2$ Whatman filters, using average flow rates of 700 l/min. Off-site samplers in the Livermore Valley use $4.6 \times 10^{-3} \text{ m}^2$ Flanders F-700 glassfiber filters operated at a flow rate of 80 l/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 portable 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 l/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 LLL 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 LLL 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 ^{239}Pu content.

Tritiated Water

Parallel samples of tritiated water vapor are collected at LLL 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 with each such area, and an 8.25-cm-diam coring tool designed at LLL is used to collect 5-cm-deep samples at the corners and center of the square.¹

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-l plastic containers to which 6.5 ml of concentrated HCl has been added. For tritium analyses, a 1-l 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 LLL effluent and from the treated effluent of the Livermore Water Reclamation Plant (LWRP). Quarterly samples are also collected from the Site 300 sewage-oxidation pond.

Collection

LLL Effluent

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

Site 300 Sewage-Oxidation Pond

Grab samples of the Site 300 sewage-oxidation pond are collected quarterly in 3.78-l polyethylene containers.

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

LLL Effluent

Every Wednesday, one replicate sample of LLL effluent is taken for gross alpha, beta, and tritium. For one month each year, duplicate samples of LLL 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 monthly 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 l. 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 southeast of the Laboratory. A 3-l portion of each milk sample is vacuum distilled, and the tritium content of the distilled water is determined by liquid scintillation. Each 3-l 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 ^{239}Pu and ^{90}Sr 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.

Gamma Counting

Monthly composites of Laboratory perimeter and Site 300 filters are analyzed for specific gamma-emitting radionuclides. These filters are sealed in 210-cm³, thin-walled aluminum cans and are counted for nominal 1000-min periods. Counting is done with an 80-cm³, low-background Ge(Li) spectrometer that is equipped with a background-suppression system.²

Standards and Calibration

Low-level uranium-ore (New Brunswick) and NBS river-sediment standards are used periodically to check counting system response. Efficiency calibration checks are made with calibrated sources supplied by the International Association of Environmental Analysts (IAEA), the Environmental Monitoring Laboratory (EML), or NBS.

^{238}Pu , ^{239}Pu

Following gamma counting, the monthly composite of Laboratory perimeter filters is divided according to sampling location. This composite and that from Site 300 are dry ashed in a 450°C muffle furnace. Ashed samples are then dissolved with a HNO_3 -HF- H_2O_2 mixture and are equilibrated with a standardized cesium carrier and tracer amounts of ^{242}Pu and ^{233}U . This solution is made 8N with HNO_3 , the plutonium is oxidized to Pu^{+4} with NaNO_2 , and the plutonium, cesium, and uranium radionuclides are separated by anion-exchange techniques.

The ^{137}Cs is collected first in the 8N fraction. Then the plutonium is eluted with a mixture of concentrated HCl - NH_4I , and the uranium fraction is eluted with 0.1N HCl .

The plutonium fraction is purified by a second anion-exchange separation and is electrodeposited on a platinum disc. Then, ^{238}Pu and ^{239}Pu are determined by alpha spectrometry.

Standards and Calibration

The LLL Heavy Element Group purifies and calibrates internal tracer ^{242}Pu by means of mass spectrometry.³ Alpha spectrometry is also used to periodically calibrate ^{242}Pu against ^{236}Pu and ^{239}Pu standards from EML.

^{137}Cs

The cesium fraction collected in the plutonium separation is heated to evaporate excess acid. After it is diluted with water, sodium hydroxide (NaOH) is added to bring the pH to 2-3 and the cesium is extracted with ammonium molybdophosphate. The cesium is further purified by ion exchange with a Bio-Rex 40 cation-exchange resin and is then precipitated as Cs_2PtCl_6 for yield correction. The precipitate is mounted for ^{137}Cs determination and is counted in a low-background beta counter.

Standards and Calibration

Standard solutions of ^{137}Cs from EML, LLL, and commercial laboratories are used to prepare efficiency and self-absorption counting standards to calibrate low-background beta detectors.

^{235}U , ^{238}U

The uranium fraction from plutonium separation is converted to sulfate by evaporating to dryness and by fuming with H_2SO_4 . Further purification is obtained with anion exchange in sulfate form. Then ^{235}U and ^{238}U are determined with isotopic-dilution mass spectrometry.⁴

Standards and Calibration

The mass spectrometer is calibrated with NBS uranium sources containing certified quantities of ^{235}U , the balance being ^{238}U . Mass values are, as noted, based on the use, of ^{233}U , and semiannual alpha spectrometry measurements are employed to verify its purity.

HTO

Water vapor samples are collected biweekly at LLL perimeter locations by silica-gel samplers operated at flow rates of about 0.5 l/min. The collected water is recovered by vacuum drying at 150°C , and the HTO is measured by liquid scintillation counting.

Standards and Calibration

A primary HTO standard is obtained from NBS. The calibration curve for each counting set is made from eight replicate measurements of a dilution of this primary standard, and the background is determined from eight replicate measurements of a deep-well water sample.

Beryllium

Every month, half of each weekly LLL 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 1-2 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 demineralized water.

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 30x absorbance expansion, and the spectrophotometer is zeroed while aspirating deionized water into the flame. Instrument responses from the 0.5- and 1- μ g 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.

SOIL ANALYSIS

Each soil sample collected in accordance with the "Gamma Counting" section below is dried to constant weight at 110°C, is pulverized in a mechanical grinder, and is screened through a 32-mesh screen to remove pebbles and dry vegetation. The sample is then blended, and aliquots are taken for various analyses.

Gamma Counting

Approximately 300 g of the pulverized and blended soil sample is sealed in a 210-cm³, thin-walled aluminum can. It is then counted for a nominal 1000 min in the low-background gamma counter for gamma emitters and ²³⁸U.²

HTO

Samples collected in argon-flushed glass bottles are distilled under argon. A 250-ml aliquot of sample is electrolytically enriched, using the enrichment technique described by Ostlund.⁶ Following enrichment, the HTO content of the sample is determined with either liquid-scintillation or internal gas-proportional counting.

Standards and Calibration

Each enrichment set contains duplicate aliquots of the secondary HTO standard to verify the enrichment factors. See the "Standards and Calibration" section of "Air Analysis" on HTO for scintillation counter standards and calibration. For internal gas-proportional counter standardization, replicate samples of a secondary HTO standard are reduced to HT and are counted after 10 unknown samples are run.

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

A 100-ml quantity of LWRP effluent is wet digested, and the residue is transferred to 5-cm-diam planchets and is counted in accordance with the procedures in the "Water Analysis" section on HTO. Aliquots of 500 ml are taken for all other gross alpha and beta measurements.

Tritium

A 1-ml aliquot of sewage sample is added to 10 ml of Packard Insta-Gel in a 25-ml screw-cap glass bottle. Then, 1 ml of tap 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 2-hr temperature-equilibration period prior to counting. The sample and background are each counted for 100 min.

^{239}Pu , ^{137}Cs , and ^{90}Sr

Nuclear Chemistry analyzes monthly composites of Laboratory sewage effluent and treated effluent from LWRP for ^{239}Pu and ^{137}Cs . Duplicate composites are prepared by Hazards Control and are analyzed for ^{239}Pu and ^{90}Sr .

Procedure

Daily samples of sewage effluent are combined for the monthly composite, are acidified with 3N HCl, and ^{242}Pu and cesium carrier are added. Ten milliliters of saturated KMnO_4 are added to the mixture, and the pH is adjusted to 8-10 with 3N NaOH. Then, 150 mL of MnCl_2 are added, and the plutonium coprecipitates with the MnO_2 .⁷ Each sample is later filtered through a 1- μm cotton filter cartridge, which is then dried and ashed at 475°C. The filtrate is acidified to pH 1-3 with 3N HCl, and the cesium is extracted with 5 g of AMP. The sample is then refiltered, and the cartridge is dried and ashed.

The plutonium filter is heated with 100 mL of concentrated HNO_3 , and the MnO_2 is dissolved with 30% H_2O_2 . The volume of solution is then reduced to about 50 mL, and the sample is filtered through 934-AH glass-fiber filter paper. The plutonium is oxidized with NaNO_2 purified by anion exchange, is electroplated, and is counted by solid-state alpha spectrometry.

The ashed cesium filter is dissolved in acid, excess ammonia is evaporated with NaOH, and the cesium is reextracted with 1 g of AMP. Further purification is carried out as outlined in the "Air Analysis" section on ^{137}Cs .

Radiochemical Separations

The procedure outlined in the "Air Analysis" section on ^{238}Pu and ^{239}Pu is used to purify ^{239}Pu . The purified ^{239}Pu is then electro-deposited and is counted by solid-state alpha spectrometry.

The Sr-Y is washed from the anion column (used in plutonium separation) with 8N HNO_3 . The combined washings are then evaporated and are converted to chloride by repeated HCl additions followed by evaporation. Residue is taken up in 8N HCl, transferred to a freshly prepared Dowex 1 column, and the column is washed with 8N HCl. The washings are later collected and evaporated to near dryness. Water is added, and the solution is adjusted to pH 1.5 with NH_4OH . This solution is extracted twice with 20% 2,2-diethyl hexyl phosphoric acid, and the organic phase is washed twice with 0.5N HCl and 1N HCl. The Y is then back extracted with three 20-mL portions of HCl, the solution is evaporated to about 10 mL, and the Y is precipitated by adding HF. The YF_3 is dissolved and precipitated as hydroxide; then it

is redissolved with HCl. The Y is then precipitated as oxalate and is filtered and converted to oxide in a muffler furnace set at 900°C for 1 hr. The ash is weighed, dissolved in HNO₃, transferred to a 5-cm-diam planchet, and dried. After drying, the ash is beta counted.

Nonradioactive Metals and Fluorides

Monthly composites of LLL sewage effluent are analyzed for copper and chromium with a Model 306 Perkin Elmer atomic-absorption spectrophotometer. Aliquots of these composites are also analyzed for cadmium, iron, lead, nickel, silver, zinc, and fluoride.

A monthly composite of Laboratory sewage effluent is prepared by combining 500-ml daily samples. Following each 500-ml addition, the solution is evaporated with HNO₃ to approximately 100 ml. After compositing is complete and organic matter has been destroyed by HClO₄ and HNO₃ treatments, the sample is evaporated to near dryness. The residue is taken up in about 30 ml of deionized water, is filtered through a sintered glass funnel, and any filter residue is discarded. The filtrate is made up to 100 ml in a volumetric flask.

Standards and Calibration

The atomic-absorption spectrophotometer is calibrated with solutions of the metals or compounds of the metal of interest. This spectrophotometer is zeroed with deionized water and is calibrated with standards. Dilution of stock solutions and the monthly composite may be necessary to obtain readings within the spectrophotometer's linear response region. Following calibration, the monthly composite sample is aspirated into the flame, and concentrations are read out directly.

Precision Checks. The monthly composite sample is used to prepare four dilutions. The first dilution provides a concentration near the limit of sensitivity; two dilutions provide intermediate concentrations; and the fourth provides a concentration near the upper limit of the spectrophotometer's linear response region. To reflect maximum interference, they are analyzed in order of highest concentration, lowest concentration, and then by the two intermediate concentrations. This set of measurements is repeated seven times, and the average and standard deviation for each concentration are recorded.

Accuracy Checks. Using the monthly composite, accuracy checks are performed by adding known amounts of the metal of interest to the low and intermediate concentrations. The amount of metal added to the low concentration is designed to double in concentration, and the amount of metal added to the intermediate concentrations is sufficient to result in concentrations that are within 25% of the upper limit of the spectrophotometer's linear response region. As in the precision measurements, seven replicate determinations are made at each concentration, and accuracy is expressed as percent recovery based on the average recovery of the seven replicates.

Fluoride Measurements

Fluorides in LLL sewage effluent are determined with a specific ion-electrode measurement. Daily additions of 100-ml aliquots of sewage effluent are used to prepare a monthly composite sample. This composite sample is stored in a plastic bottle, and no chemical treatment or evaporation is employed. At the end of the month, an aliquot of the composite sample is analyzed for fluorides with an Orion Model 401 specific ion meter equipped with a lanthanum fluoride electrode. Precision and accuracy measurements are conducted quarterly in a manner similar to those described for metals in the "Nonradioactive Metals and Fluorides" section on precision and accuracy checks above.

VEGETATION ANALYSIS

Frozen vegetation is transferred to freeze-drying equipment, and a 10- μ m vacuum is established. Recovered water is collected in a receiver chilled with dry ice and Dowanol (dipropylene methyl ether). The vacuum is checked periodically, and dry ice is added as needed over a four-day period. Liquid scintillation counting is then used to analyze an aliquot of the recovered water for tritium content.

Standards and Calibration

See the "Air Analysis" section on standards and calibration for HTO.

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