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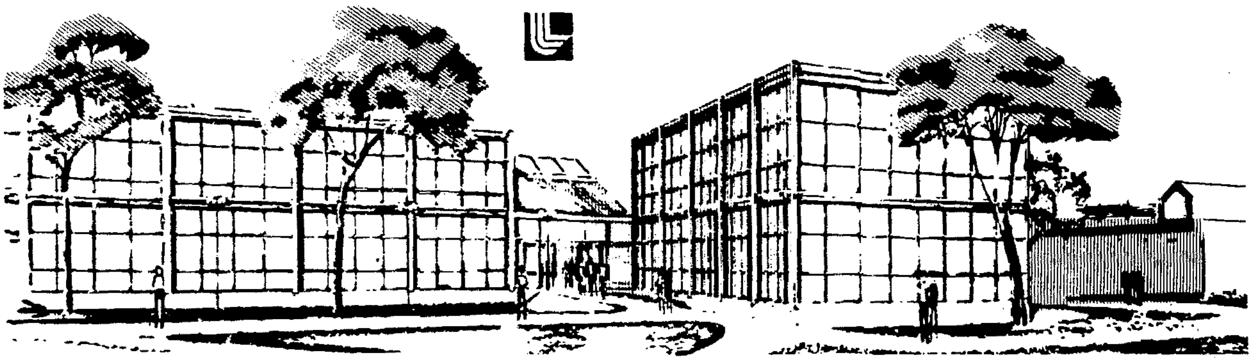

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Quality Assurance in Environmental Monitoring at the Lawrence Livermore Laboratory*

C. L. Lindeken, J. H. White, and W. J. Silver

Abstract

The quality assurance program for environmental monitoring that has been developed at the Lawrence Livermore Laboratory (LLL) consists of procedure documentation, replicate field-sample analysis, and participation in intercomparison measurements. Sampling, analytical, data processing and record keeping procedures are described. A replicate-sample collection schedule has been established for all media sampled at LLL. At present, blind-spiked samples are not utilized. Flow rates of air samplers are verified at monthly intervals using a portable, field calibration unit. Intercomparison measurements are made on samples supplied by the Quality Assurance Branch of the Environmental Protection Agency-Environmental Monitoring and Support Laboratory and the Department of Energy-Environmental Measurements Laboratory. Replicate sampling currently accounts for approximately 8% of both the total samples collected and the analyses performed. Including standard, in-house, quality-control checks and the intercomparison measurements, it is estimated that during 1978 quality assurance will represent about 15% of the total environmental-monitoring effort at LLL.

Introduction

As a result of growing interest in the environmental movement, public attention is increasingly being focused on environmental monitoring programs carried out by both government agencies and private industry. To establish confidence in the data being generated, quality assurance (QA) measures are being incorporated in many of these monitoring programs. At the Lawrence Livermore Laboratory (LLL), an environmental monitoring program is maintained to assess the effectiveness of the effluent-containment procedures used here. Because LLL is a contractor of the U.S. Department of Energy (DOE), monitoring is conducted in accordance with the requirements of DOE IMD 0513 "Effluent and Environmental Monitoring and Reporting" (DO74).

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These monitoring results are published annually in a document available to the public. To provide confidence in the data contained in these reports, a QA program has been developed, which is now an integral part of the LLL environmental monitoring program.

Organization and Responsibilities

At LLL, environmental monitoring is conducted through the combined efforts of the Nuclear Chemistry Division, the Environmental Sciences Division, and the Hazards Control Department. The overall program, including QA, is directed by the Environmental Evaluations Group of the Hazards Control Department. This group has the responsibility for sample collection, compiling and review of analytical data, and preparation of reports. Most analyses for specific radionuclides are made by Nuclear Chemistry and Environmental Sciences. Hazards Control performs all in-house stable-element analyses, gross alpha- and beta-activity measurements, as well as analyses for selected specific radionuclides. Infrequent measurements that require specialized equipment or techniques and are not normally associated with the Laboratory's mission are contracted to outside laboratories.

The supervisor of the Environmental Evaluations Group is responsible for directing the QA program, which developed from the format outlined in the LLL Quality Assurance Manual (L178). Annual audits are made by the LLL Quality Assurance Office to verify compliance with the procedures specified for the program. Because the primary purpose of the program is to provide confidence in the analytical data, the scope of the QA plan was limited to the elements of sample collection and analysis, and data processing and recordkeeping.

Procedure Development

Our first undertaking at LLL was to prepare a document to accurately describe the environmental sampling and analytical procedures currently in use. Much of this information was obtained from internal memoranda and reports, but there was a general need for formalization and updating. In a description of the procedures in the draft document, each activity is outlined in sufficient detail for the knowledgeable reader to judge whether the procedures are appropriate in principle. References are provided when the methods or procedures in use have been previously published.

Records and Calibrations

Throughout the program, emphasis has been placed on maintaining records that cover each activity. Sample flow through the system is shown in Figure 1. After collection, the sample description and identification are entered in a field log book. When the sample is delivered to the appropriate laboratory for analysis, the sample is recorded and assigned a laboratory number, which is used for identification until the analytical data are reported.

Each laboratory participating in sample analyses maintains its own records on instrument backgrounds, analytical blanks, standardizations, and data reduction. Data furnished the Environmental Evaluations Group for interpretation and subsequent reporting are permanently retained by the originating laboratory, either in laboratory notebooks or work sheets, or on magnetic tapes or similar computer-storage records.

Periodic internal audits are made of each activity to ensure that adequate records are maintained and that data appearing in past, LLL annual environmental reports can be verified. In some cases, such as soil samples, the sample itself is retained so the analysis can be repeated if desired. In other cases, such as air filters, the sample is destroyed during analysis. However, the laboratory data pertaining to these measurements is permanently retained. As noted, our program is also regularly reviewed by the Laboratory's Quality Assurance Office.

Air sampling is the major sampling activity in which collection depends on a continually operating mechanical device that lends itself to calibration. Figure 2 shows the portable unit designed for field checking the calibration of Hi-Vol* air samplers. The unit consists of a Venturi meter equipped with a tapered adaptor to fit a 0.052-m^2 filter. The Venturi meter is connected to a Magnehelic gauge. Each month this field calibrator is itself calibrated in the laboratory; then the flow rates indicated by samplers in the field are compared with those of the portable unit.

Table 1 shows the type of records being maintained for the particulate air samplers at the LLL perimeter area. The data generally indicate

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*Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Department of Energy to the exclusion of others that may be suitable.

Figure 2. Field check of flow-rate calibration of particulate air samplers.

consistent sampler to calibrator ratios. Any sampler that differs from the calibrator by more than 20% in two successive checks will be replaced and recalibrated in the laboratory.

Quality Control Samples

Documenting accuracy and reproducibility of measurements is implicit in the QA objectives of any environmental monitoring program. Accuracy demonstrations are usually based on analyses of samples that contain known concentrations of the constituent of interest. Typical of such samples are those regularly distributed in the intercomparison programs of the Environmental Protection Agency (EPA)-Environmental Monitoring and Support Laboratory (EMSL) and the DOE-Environmental Measurements Laboratory (EML). For in-house checks on accuracy, a known quantity of the radioactive or stable constituent is added to the sample. These "spiked" samples are submitted for analysis along with regular samples. If the analyst is unaware that spiking has occurred, the sample is termed a "blind spike".

As previously noted, the analyses of LLL environmental samples are divided among different groups, and at present these samples do not pass through a central receiving or process area. Without such centralization, it is logistically difficult to maintain the "blind" element on a program-wide basis. As a result, blind spiking is not employed in the LLL program, and we depend on performance in intercomparison analyses to demonstrate accuracy.

Replicate Sampling

The analysis of replicate field samples is a recognized means of evaluating the reproducibility of measurements. In the LLL program, because all sample collections are made by one group, it was relatively easy to introduce replicate samples into the system. Accordingly, a replicate-sampling schedule was adopted that included all the media routinely monitored at LLL. Table 2 shows annual, routine and replicate schedules, and indicates the number of samples collected and the number of analyses performed.

In general, replicate samples account for about 10% of the samples collected. Air samples run considerably less than this, because here replication involves operation of samplers in parallel, and such checks are at present restricted to on-site sampling locations.

At bimonthly intervals, a particulate air sampler is operated in parallel with the permanent sampler at one of the six perimeter locations at the Laboratory. This parallel operation is continued for a one-month period; both filters are changed on the regular weekly basis and are submitted for analysis in the usual manner. The extra sampler is rotated among the six locations, so that all samplers are checked annually. Replicate samples are compared with respect to gross alpha and beta activities and ^{239}Pu content.

Typical gross alpha and beta activity data are shown in Table 3. The reason for the poor agreement in the alpha data might be that all activities are near the limit of detection sensitivity. However, if this were the explanation, we would expect the ratios to be better distributed around 1.0, rather than being biased toward lower values. Inspection of the sampling records showed that sampler 2A consistently operated at a higher flow rate. As a result, the dust loading on the Whatman-41 filter on this sampler was greater, and larger burial losses of alpha activity would be expected. The better agreement shown for beta activities implies that the flow rates indicated by these samplers were probably correct.

Table 4 shows the replicate measurements for plutonium in air. Note that, with the exception of Location 14, the variations between the results of replication runs are comparable to the variation between results for the other perimeter stations. This indicates that the plutonium source is global fallout and that all five samplers may be considered as replicate collections. Location 14 is normally downwind from an area where now discontinued waste-processing operations resulted in low-level plutonium contamination (Si 74).

Parallel air samples for tritiated water analyses are collected at the perimeter locations during alternate months from those in which replicate samples for particulate analyses are collected. Water vapor is collected on silica gel, using air flow rates of about 0.5 litre/m over collection periods of 2 weeks. The air mover is a positive-displacement pump, filtered with an in-line critical orifice to provide flow control. Figure 3 shows a pair of tritiated water-vapor collectors in operation. Table 5 illustrates the variability in the measured tritium content, using these perimeter samplers. Because the counting errors associated with these measurements are typically under 5%, these data suggest that replicate measurements provide a more realistic indication of overall reproducibility.

Table 4. Replicate Data for Plutonium Activity in Air at LLL-Perimeter Stations — 1977.

Location	^{239}Pu Activity × 10^{-17}, μCi/ml	
	May	November
15	1.9 ± 9%	2.0 ± 10%
15A	2.3 ± 7%	2.2 ± 8%
Average of all stations except location 14	2.2	2.4

Table 5. Replicate Data for Airborne Tritiated Water — 1978.

Sampling period	Location	Activity, μCi/ml	Ratio: 15/15A
2/8–2/22	15	4.4×10^{-11} ± 3%	1.33
	15A	3.3×10^{-11} ± 6%	
2/22–3/8	15	3.4×10^{-11} ± 5%	0.92
	15A	3.7×10^{-11} ± 4%	

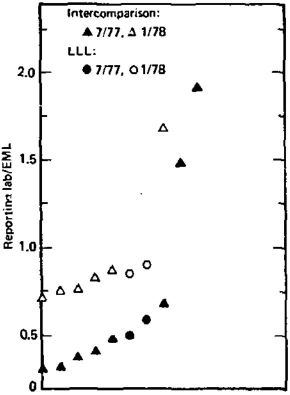

Figure 4. Plutonium-in-water intercomparison data from DOE Quality Assurance Program. (Data points arranged in order of increasing values.)

Table 6. Replicate TLD (CaF_2 phosphor) Measurements at LLL-perimeter Stations – 1977.

1977 Quarter	Measurements,[a] mrads ± 2σ (%)		
	Group 1	Group 2	Group 3
1	19.1 ± 12.9	18.4 ± 9.8	19.3 ± 5.9
2	19.0 ± 3.7	17.2 ± 9.1	17.6 ± 6.2
3	17.0 ± 13.4	14.9 ± 8.6	17.3 ± 14.8
4	19.3 ± 15.3	18.0 ± 9.5	19.3 ± 5.1

[a]Each dose value represents an average of 9 measurements.

Table 7. Relationship Between Numbers of QA and Routine Samples Processed and Analyzed in LLL Environmental Monitoring Program - 1978.

	Analyses				Samples			
Medium	No. Routine	No. QA	Total No.	QA/total, %	No. Routine	No. QA	Total No.	QA/total, %
Air	3416	124	3540	3.5	1560	48	1608	3.0
Water	276	58	334	17.4	82	18	100	18.0
Soil	62	18	80	22.5	31	7	38	18.4
Vegetation	216	32	248	12.9	216	32	248	12.9
Sewage	2786	383	3169	12.1	966	160	1126	14.2
Milk	24	2	26	7.7	12	1	13	7.7
TLD	216	72	288	25.0	216	72	288	25.0
Totals	6996	689	7685	9.0	3183	338	3421	9.9

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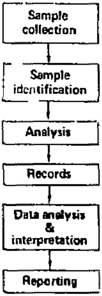

Figure 1. Sample and data flow in the LLL Environmental Monitoring Program.

Table 1. Field Checks of Air-flow Calibrations — 1978.

Location	Sampler/calibrator ratio [(a)]			
	Jan.	Feb.	March	April
01	1.05	1.04	1.01	1.12
02	0.88	0.83	0.89	0.81
12	0.84	0.82	1.48	0.83
13	1.09	1.06	1.14	1.06
14	0.82	1.11	0.95	—
15	1.05	1.07	1.06	0.92

(a) Ratios of flow indicated by sampler compared with flow indicated by calibrator.

Table 2. Number of Samples Collected and Number of Analyses Performed for Environmental Monitoring Program — 1978.

Medium	No. of samples		No. of analyses	
	Routine	Replicate	Routine	Replicate
Air	1560	44	3416	116
Water	182	8	276	24
Soil	31	3	62	6
Vegetation	216	24	216	24
Sewage	966	160	2786	383
Milk	12	1	24	2
TLD[a]	216	48	216	48
Totals	3183	288	6996	603

[a]Thermoluminescence dosimetry (TLD).

Table 3. Gross Alpha and Beta Activities of Parallel Air Samplers.

	Alph activity × 10^{-15}, µCi/ml			Beta activity × 10^{-15}, µCi/ml		
	Sampler		Ratio:	Sampler		Ratio:
Test	2	2A	2A/2	2	2A	2A/2
1	1.55	1.23	0.79	9.68	9.83	1.02
2	1.74	1.16	0.67	23.68	17.45	0.75
3	3.14	2.82	0.90	22.33	22.83	1.02
4	0.81	0.94	1.16	16.02	14.36	0.90
5	0.97	0.56	0.58	10.55	11.66	1.12
6	0.98	0.43	0.44	10.17	10.70	1.05
7	1.42	1.01	0.71	15.23	15.40	1.01
8	1.89	1.50	0.80	22.67	21.98	0.97
9	1.24	0.54	0.44	20.03	17.01	0.85
10	0.83	0.47	0.57	11.31	16.97	1.50
11	0.50	0.43	0.86	10.01	8.32	0.83
12	1.92	2.00	1.04	14.54	13.43	0.92
13	0.73	0.56	0.77	6.47	5.90	0.91
14	0.49	0.41	0.84	7.82	6.69	0.86
Average ratios			0.76			0.98

Figure 3. Replicate sampling of tritiated water vapor.

In 1975, we began taking replicate measurements of environmental gamma radiation by exposing multiple thermoluminescence dosimeters at each of three locations on the LLL-perimeter fence. These measurements were initially undertaken to determine the precision that could be expected from thermoluminescence dosimetry (TLD) under field conditions. However, they are now part of the QA program. Each location is provided with three dosimeters. Each dosimeter contains three TLD-700 chips (LiF phosphor) and three TLD-200 chips (CaF_2 phosphor) so reproducibility data can be based on nine measurements of each phosphor type. Replicate dosimeters along with the other environmental dosimeters are changed quarterly and read as part of the routine set, using a procedure developed for environmental background measurements (Li76). Results of this replication study were reported by White et al. (Wh78). Table 6, which shows the CaF_2-phosphor data obtained in 1977, indicates that overall reproducibility of the technique is within 15%, well within the performance recommended by ANSI N545-1975 (An75).

Intercomparison Analyses

Lawrence Livermore Laboratory participates in both the EPA Cross Check Program conducted by EMSL-Las Vegas and the DOE Quality Assurance Sample Analysis Program operated by EML. In the EPA program, samples are distributed bimonthly, quarterly, semi-annually, or annually — the schedule being set by the interests of program users. At present, all DOE samples are distributed quarterly. Both programs offer a variety of radionuclides in all media normally encountered in environmental monitoring. The EPA program is the older and larger effort, having at present over a hundred laboratories participating on a regular basis. The DOE program, started in 1976, is limited to about thirty DOE-contractor laboratories.

At LLL, the Nuclear Chemistry and Environmental Science Divisions participate in both programs; at present, Hazards Control participates in only the EPA Cross Check analyses. All intercomparison samples are received by and distributed internally by the Environmental Evaluations Group, which also is responsible for reporting the results of all analyses. No attempt is made to conceal the presence of the QA samples in the system. In many cases the QA samples are analyzed along with other routine samples, but this has never been considered mandatory.

When the results of each intercomparison are complete, both EPA and DOE send participants a coded report that lists all data submitted. Whereas, EPA compares these data with the grand average as well as with the known value based on the EPA measurement, DOE compares each participant's data only with that obtained by the EML. These reports are made an integral part of the records of our QA program.

The Laboratory also has begun a quarterly TLD intercomparison with EML. Dosimeters from LLL are exposed at EML's monitoring station at Chester, New Jersey, along with EML dosimeters. Because LLL dosimeters must be air shipped to and from the east coast, corrections must be made for transit doses. These corrections are based on data from dosimeters shipped in the same package as the others but kept in a radiation shield while the field exposures are in progress.

Early in the DOE intercomparisons, it was found that the LLL values for plutonium in water were about half those found by EML. According to the report, the values reported by other laboratories were also quite low compared with those of EML. In general, our results agreed closely with those of EML if plutonium-spiked filter papers were used, so the trouble did not appear to be in the plutonium procedures. Discussions with EML revealed that they used the whole sample for plutonium analysis, whereas LLL and several other laboratories used an aliquot. Samples were shipped in plastic bottles and, although the samples were acidified, plutonium was apparently adsorbed on the container walls. After we adopted the EML practice, much closer agreement was obtained, as indicated in Figure 4.

We have also observed differences between the LLL gamma-count data and the EML reference data for counts of spiked air filters. Both EML and LLL count the papers in a plane configuration. Although the discrepancy is not yet resolved, it may be related to differences in the counting geometries assumed for the measurements. Plans for special intercalibrations are being discussed.

Quality Assurance Effort

Table 7 shows the relationship between the numbers of QA and routine samples processed and analyzed at LLL. Quality assurance accounts for about 10% of the total on the basis of the number of samples and 9% based on analyses. At present, the major emphasis is on replicate sampling, which currently accounts for approximately 8% of both the total samples and the number of analyses performed. Including in-house quality control checks and the intercomparisons, QA is estimated to represent about 15% of the total environmental monitoring effort at LLL.

Conclusions

We believe that this present level of effort adequately meets the need for QA in our environmental monitoring program. One of the immediate benefits of implementing the QA plan has been the introduction of a formal mechanism of continual review of the entire LLL program. The emphasis on replicate sample analysis is also providing valuable field data that may provide a more realistic base for estimating the reproducibility of environmental measurements. Because the objective of the QA program is to provide confidence in environmental monitoring data, both field and laboratory personnel have been cooperative in implementing the plan and been personally interested in the results. In theory, at least, much of what is being done in the program has always been done. The difference is that we now have a name for it — quality assurance.