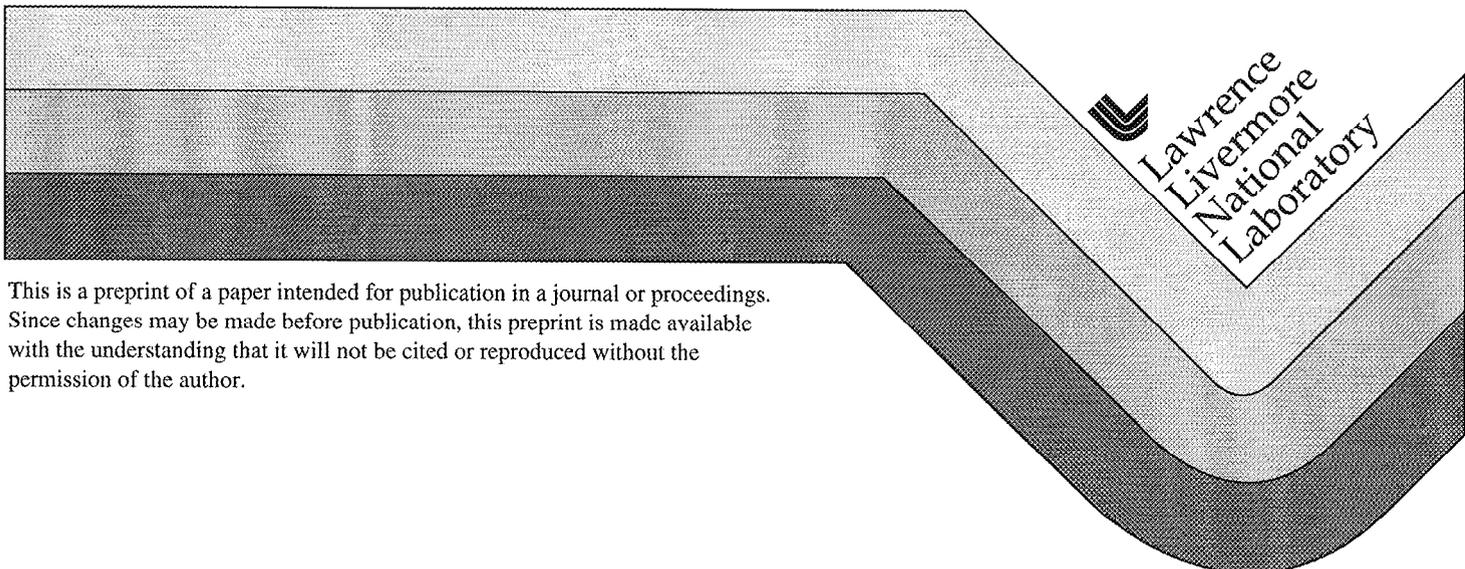


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Radiological Background Levels Found on Glass Fiber Filters used for Low-Level Environmental Surveillance Air Sampling

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Environmental surveillance of low-level radioactive particles in air requires a thorough understanding of low-level techniques and air sample collection media. High-volume air sampling for radioactive particles around Lawrence Livermore National Laboratory (LLNL) employs glass-fiber filters that are analyzed for gross alpha and gross beta activity and for specific isotopes.

This study was conducted to determine the activities of radionuclides contained in blank glass-fiber filters. Data from this study provided a partial explanation of differences between current reported concentrations of radionuclides in air and those reported historically when cellulose filters were used in the LLNL monitoring effort.

Introduction

For over 30 years, Lawrence Livermore National Laboratory has conducted routine surveillance of airborne radiological activity found in the local environs. Gross alpha, gross beta, and specific isotopic analysis are conducted. These data are reported in the annual LLNL site Environmental Report.¹

Historically, LLNL used cellulose filters for all environmental air particulate sampling. Cellulose filters have a low radiological background level,² in addition, state and local regulations require environmental monitoring for beryllium and clean blank cellulose filters have very little (if any) reported beryllium activity. In contrast, blank glass fiber filters have high and variable levels of beryllium.

The tradeoff between lower background levels of beryllium and radioactivity of the cellulose filters and the collection efficiency of the glass fiber filter was investigated in 1996 and the decision was made to implement dual air samplers. In April of 1997, LLNL purchased additional high-volume air sampling units, consequently some samplers were exclusively used to monitor airborne beryllium concentrations using cellulose filters.

This permitted the radiological monitoring media to be changed to the industry recommended, superior quality^{3,4} glass fiber filter. Glass fiber filter collection efficiencies have been measured at 99% efficiency compared to the cellulose, which varies from 64 to 95% depending on factors such as pore size, pressure drop, and face velocity.⁵ Beryllium monitoring is still conducted at LLNL using cellulose filters, due to the increased amount and variability of background levels of beryllium inherent in the composition of glass fiber filters.

A rise in the activity from the glass fiber filters for several radiological isotopes was expected because of the levels of radioactivity inherent in glass fiber filters.³ Upon receipt and preliminary analysis of the radiological environmental surveillance data using glass fiber filters, several routinely monitored analytes showed an immediate rise in activity over that which was previously reported. It was believed that the increase in activity for those isotopes was likely due to the composition of the glass fiber filters; however, the amount of the increase was not known for the filters used at LLNL and required investigation.

In order to accurately report the airborne environmental radiological activity, the average background concentrations and associated error detected on the blank glass fiber filter needed to be determined. If the standard deviation of the blank glass fiber samples was low for the various nuclides, then the average amount determined from the blanks could be subtracted from the total measured activities for the environmental samples.

A special study of blank glass fiber filters was conducted to determine the average activity on clean glass fiber filters for all analytes monitored in the LLNL air surveillance program. These analytes include gross alpha, gross beta, plutonium-238, plutonium-239, uranium-234, uranium-235, uranium-238, and a gamma scan suite which specifically identified isotopes of interest to LLNL. These isotopes include: sodium-22, potassium-40, cesium-137, radium-226, radium-228, thorium-228. Although not routinely monitored at LLNL, the curium-244 content on blank glass fiber filters was also evaluated.

Filter Paper and Equipment Statistics

The glass fiber filter paper and high-volume air sampling units used in this study were obtained from Hi-Q Environmental, San Diego, CA. The manufacturer of this paper was Hollingsworth & Vose Company, East Walpole, MA. Due to unavailability of different glass fiber manufacturers and different lot groupings, only one lot from one manufacturer was tested.

The filter is type FP-2063,⁶ made from 100% pure borosilicate glass with an acrylic binder, for collection of sub-particles larger than 0.8 micron. The filter thickness is 0.016. The glass fiber filters have a reported collection efficiency of 99%. The Environmental Surveillance Program at LLNL routinely collects the particulate for radiological analysis on glass fiber filters, using high-volume air samplers, which maintain a flow rate of 35 cubic feet per minute (CFM) using a brushless 2 stage centrifugal pump for 7 days.

Experimental

The 8" × 10" glass fiber filters were prepared for laboratory delivery and analysis using sample hygiene that reduced or eliminated the potential for accidental contamination from external sources. All filters were handled using clean gloves and a clean working surface. Because the data from this study were used in the routine LLNL Environmental Surveillance Program, filter preparation (where possible) closely matched the Standard Operating Procedures for Air Sampling at LLNL.⁷

Gross Alpha and Gross Beta

Routine analysis for gross alpha and gross beta are performed at an off-site laboratory; therefore, blank glass fiber filters were sent directly to the same laboratory for comparison. An unopened box of filters was placed inside a sealed box then shipped to the laboratory. The filters arrived with filter integrity intact. Gross alpha and beta analysis was performed on 12 separate glass fiber filters by Standard EPA Method 900 using a gas flow proportional counter. Americium-241 and cesium-137 were used as calibration sources to determine the counting efficiencies. The 12 different glass fiber blank filters were counted for 120 minutes, producing 12 separate data points identified as GFB1 through GFB12 in Table 2.

Gamma Scan

Filters for all other radiological analysis were prepared in the same manner as routine samples and analyzed by an on-site laboratory. See Figure 1 for a diagram showing the process for each data point. This process was performed eight times producing eight data points identified as GFB1 through GFB8.

With a gloved hand, six different filters were cut into one-quarter portions (approximately 4" × 5"). From each of the filters, a one quarter portion was removed, folded and placed into a composite. The composite then consisted of six one-quarter portions from six different filters (total mass equaling 24 one-quarter portions for a total of six 8" × 10" filters). This composite was sealed in a aluminum can (214 cm³) and counted using low-level background germanium (lithium) detectors for 168 hours (7 days).

Plutonium, Uranium, and Curium Isotopes

Upon completion of the gamma counting, the can was opened and from the 24 one-quarter portions, three sets of eight were removed, creating three new composite sets. One set was analyzed for plutonium, one set was analyzed for uranium and the third set was analyzed for curium.

The blank glass fiber filter set for plutonium-238, plutonium-239, 240 and uranium-234, 235, 235 analysis were dissolved by nitric acid. The plutonium and uranium was separated out through an ion-exchange process. The isotopes were electroplated onto a stainless steel disk then analyzed by alpha spectrometry.

The last set of blank glass fiber filters for curium-244 analysis were dry ashed then placed in a Teflon container where they were combined with an americium-243 tracer and wet ashed. The solution is eluted using a nitric solution; americium-241 and curium-244 were electroplated then counted by alpha spectrometry.

Results

Table 1. shows a summary of all analytes included in this study. Also shown are the achievable detection limits, methodology, activity and error supplied by the analytical laboratories. Only those analytes of specific concern to LLNL were available for this report.

Table 2. shows the reported raw values each of the samples for those analytes where the results were above the achievable detection limit. The values for each analysis are shown as well as their reported counting error. The averages and the standard error are provided.

Conclusions

Of the eight isotopes reported from the gamma data, potassium-40, radium-228, and thorium-228 were detected on glass fiber filter blanks. Additionally, uranium-234, uranium-235, uranium-238, and gross beta were also detected. These isotopes were also elevated in analytical results for routine samples collected by LLNL for the Environmental Surveillance Program. After applying the correction factor² that removed the contribution of the blank filters, the environmental surveillance data was normalized and only slightly higher than data collected in previous years using cellulose filters. Although the correction factor did reduce the overall airborne concentrations, by removing the naturally occurring radioactivity found in the filter material, it did not bring the data completely down to concentrations reported in previously years.

Further work is necessary to determine what contribution the improved collection efficiency of the glass fiber filters may play in the overall increase of airborne radiological activity seen at LLNL.

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ANALYTE	Detection Limit (pCi/filter)	Analytical Method	Activity/Filter	Standard Error
Gross Alpha	0.29	Gas Flow Proportional Counter	ND	NA
Gross Beta	0.041	Gas Flow Proportional Counter	55.9	2.30
Plutonium-238	0.017	Alpha Spectrometry	ND	NA
Plutonium-239,240	0.004	Alpha Spectrometry	ND	NA
Curium-244	0.06	Alpha Spectrometry	ND	NA
Uranium-234	4.26E-07 µg/filter ^(a)	Alpha Spectrometry	2.28E-04	3.9E-06
Uranium-235	1.40E-02 µg/filter ^(a)	Alpha Spectrometry	3.22E-02	1.00E-03
Uranium-238	2.00 µg/filter ^(a)	Alpha Spectrometry	4.12	0.068
Beryllium-7	6.47	Gamma Scan ^(b)	ND	NA
Sodium-22	0.43	Gamma Scan ^(b)	ND	NA
Cesium 137	0.41	Gamma Scan ^(b)	ND	NA
Radium-226	1.13	Gamma Scan ^(b)	ND	NA
Potassium-40	19.7	Gamma Scan ^(b)	52.3	0.874
Radium-228	2.15	Gamma Scan ^(b)	1.28	0.047
Thorium-228	0.900	Gamma Scan ^(b)	1.37	0.025

ND= Not Detected
NA= Not Applicable

^(a) All uranium activity is converted to mass for historical LLNL reporting purposes.
^(b) The isotopes listed within the gamma scan are of special concern to LLNL and called out specifically for historical purposes.

Table 1. Summary of detection limits, methodology, activity and standard errors for all analytes.

Potassium-40	Activity (pCi/filter)	Counting Error (pCi/filter)		Uranium-234	Activity (µg/filter)	Counting Error (µg/filter)
GFB1	54.8	4.3		GFB1	2.18E-04	1.59E-05
GFB2	53.9	4.3		GFB2	2.23E-04	1.65E-05
GFB3	48.7	4.3		GFB3	2.31E-04	1.66E-05
GFB4	49.1	2.2		GFB4	2.44E-04	1.76E-05
GFB5	53.5	3.4		GFB5	2.28E-04	1.64E-05
GFB6	54.0	2.5		GFB6	2.25E-04	1.62E-05
GFB7	50.5	3.8		GFB7	2.42E-04	1.74E-05
GFB8	54.1	3.0		GFB8	2.12E-04	1.54E-05
Average	52.3			Average	2.28E-04	
Standard Error	0.874			Standard Error	3.90E-06	
Radium-228	Activity (pCi/filter)	Counting Error (pCi/filter)		Uranium-235	Activity (µg/filter)	Counting Error (µg/filter)
GFB1	1.32	0.34		GFB1	3.13E-02	5.00E-03
GFB2	1.43	0.37		GFB2	3.17E-02	5.61E-03
GFB3	1.07	0.37		GFB3	3.15E-02	4.98E-03
GFB4	1.15	0.27		GFB4	3.40E-02	5.25E-03
GFB5	1.37	0.34		GFB5	3.27E-02	5.15E-03
GFB6	1.19	0.20		GFB6	3.75E-02	5.44E-03
GFB7	1.43	0.31		GFB7	2.75E-02	4.69E-03
GFB8	1.28	0.26		GFB8	3.11E-02	5.00E-03
Average	1.28			Average	3.22E-02	
Standard Error	0.047			Standard Error	1.00E-03	
Thorium-228	Activity (pCi/filter)	Counting Error (pCi/filter)		Uranium-238	Activity (µg/filter)	Counting Error (µg/filter)
GFB1	1.35	0.16		GFB1	3.99E+00	2.91E-01
GFB2	1.44	0.18		GFB2	4.05E+00	2.99E-01
GFB3	1.25	0.18		GFB3	4.22E+00	3.03E-01
GFB4	1.29	0.12		GFB4	4.44E+00	3.19E-01
GFB5	1.45	0.18		GFB5	4.11E+00	2.96E-01
GFB6	1.38	0.11		GFB6	4.30E+00	3.08E-01
GFB7	1.37	0.15		GFB7	3.99E+00	2.89E-01
GFB8	1.42	0.13		GFB8	3.85E+00	2.80E-01
Average	1.37			Average	4.12E+00	
Standard Error	0.025			Standard Error	6.77E-02	
Gross Beta	Activity (pCi/filter)	Error (pCi/filter)				
GFB1	56.1	8.13				
GFB2	49.2	8.13				
GFB3	52.4	8.13				
GFB4	54.9	8.13				
GFB5	58.9	8.13				
GFB6	55.3	8.13				
GFB7	41.9	8.13				
GFB8	56.5	8.13				
GFB9	76.4	8.13				
GFB10	55.7	8.13				
GFB11	54.5	8.13				
GFB12	58.9	8.13				
Average	55.9					
Standard Error	2.30					

Table 2. Activity of analytes tested above the reported detection limit.

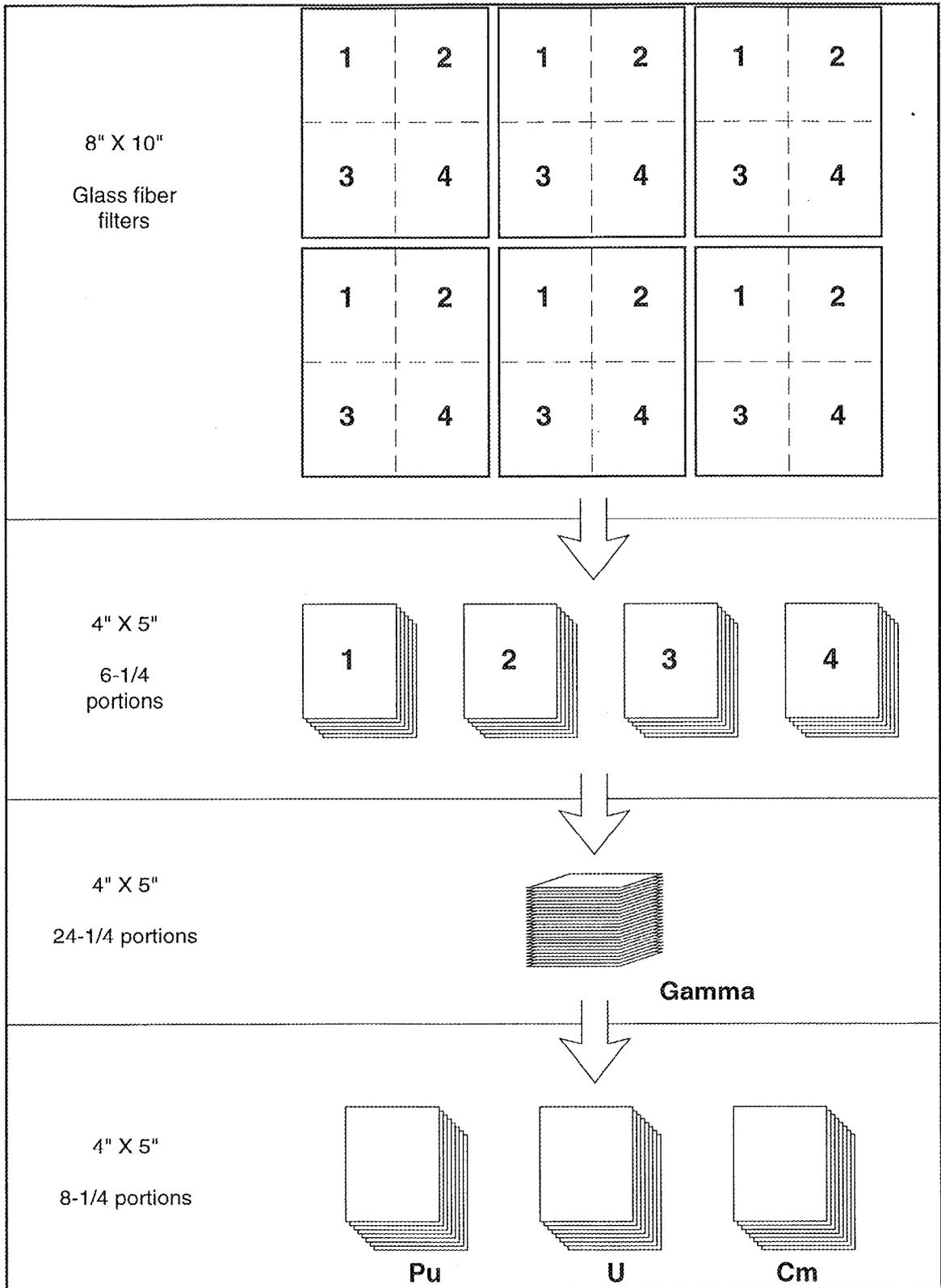


Figure 1. Flowsheet showing the preparation for glass filter analysis.