
Evaluation of Instrumental Methods for the Measurement of Yellowcake Emissions

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Commission

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ABSTRACT

An evaluation of current sampling and analysis methods used for monitoring yellowcake emissions from uranium mill exhausts was performed by Pacific Northwest Laboratory. The representativeness of sampling once per quarter was felt to be questionable. A more representative sample would be obtained by a continuous sampling system. The analysis could be performed by relatively newer instrumental methods. Direct-spectrometric and isotopically excited x-ray fluorescence instrumental analysis methods were evaluated. Because of a redirection in funding, the evaluation was not completed in terms of identifying instrumental interferences and field testing of the chosen methods. However, in light of readily available technology, a preferred method for sampling and analysis of yellowcake from uranium mill exhausts is proposed. This method would sample the exhaust stacks continuously using a continuous, automatic, isokinetic stack sampler with deposition of the exhaust gas particulates onto filter paper. The deposited particulates would then be analyzed by x-ray fluorescence using ^{57}Co as an excitation source. It is also recommended that a paper-tape sampler that houses an isotopic excitation source and detector be interfaced to a continuous stack sampler. This system would require evaluation and field testing after development.

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EXECUTIVE SUMMARY

An evaluation of current sampling and analysis methods used for monitoring yellowcake emissions from uranium mill exhausts was performed by Pacific Northwest Laboratory (PNL). This evaluation was performed in support of the Nuclear Regulatory Commission's (NRC) mandate under 10 CFR 20, "Standards for Protection Against Radiation," and 10 CFR 40, "Domestic Licensing of Source Material." In particular, 10 CFR 40.65 ("Effluent Monitoring Reporting Requirements") requires semiannual reports to the NRC that will allow estimation of doses to the public based on effluent releases. Also, Regulatory Guide 4.14, Revision 1, stipulates that yellowcake drying and packaging exhaust stacks should be sampled at least quarterly so that stack release rates and concentrations of natural uranium, ^{230}Th , ^{226}Ra , and ^{210}Pb released to the environment may be estimated. The NRC requested PNL to evaluate current methods being used to monitor stack emissions of yellowcake and to suggest alternative or new procedures to obtain this information.

We determined that current mill practices follow the guidelines of Regulatory Guide 4.14, Revision 1 (i.e., quarterly sampling). The stack sampling should be isokinetic and representative, and adequate for the determination of release rates and concentrations of natural uranium in the stack gases. In addition, the sampling should be adequate for the determination of radionuclide release rates and concentrations of ^{230}Th , ^{226}Ra , and ^{210}Pb if these data cannot be obtained from other sources. Stack emission rates have been reported to vary with time by as much as a factor of 20. Also, the time required for stack sampling represented a small fraction of the total stack operating time. Thus, to obtain a more accurate estimate of the 'true' emission release rate, more samplings would need to be conducted. Ideally, continuous sampling of the stack gases and particulates would yield the 'true' emission release rates.

The proposed monitoring methods addressed two questions: 1) How is a representative sample collected? and 2) How is it analyzed? Three methods were investigated as to their feasibility for sample collecting. They were 1) a continuous, automatic, isokinetic, stack sampler; 2) an electrostatic precipitator system designed for continuous use; and 3) optical methods such as a single- or double-pass opacity or a lidar system. The analysis could then be performed in the laboratory by traditional means, isotopic excitation x-ray fluorescence (XRF), direct gamma-ray analysis using an intrinsic germanium detector (IG), or laser-excited kinetic phosphorimetry.

The following analytical methods were chosen for evaluation in the laboratory:

- 1) direct gamma-ray spectroscopy
- 2) isotopically excited x-ray fluorescence using either ^{109}Cd or ^{57}Co sources
- 3) laser-excited kinetic phosphorimetry.

Of these analytical methods, the ^{57}Co excited XRF system appears to be the most promising because of speed of analysis and low sensitivity to interferences.

Four main sampling-analysis methods were proposed:

- 1) electrostatic precipitator collection with kinetic phosphorimetry analysis
- 2) continuous, automatic, isokinetic stack sampling with kinetic phosphorimetry
- 3) electrostatic precipitation with isotopic excitation and spectrometric analysis using a ^{109}Cd source or ^{57}Co source
- 4) continuous, automatic, isokinetic stack sampler with isotopic source and spectrometric analysis using a ^{109}Cd source or ^{57}Co source.

Method 3 would be the lowest cost method. However, since electrostatic sample collection systems have not yet been proven reliable for continuous sampling, methods 1 and 3 will not be recommended at this time. Therefore, with current technology, method 4, the continuous, automatic, isokinetic stack sampler with isotopic source (^{57}Co) and spectrometric analysis, would be the preferred method. For the first year, this system would cost \sim \$46,000 in capital costs for two stacks. The manpower required for sampling and analysis would run \sim \$23,000, for a first year total cost of \$69,000. Each year thereafter the cost would be for manpower only.

Because of redirection in funding, the originally proposed work was not completed. Yet to be investigated were the possible interferences caused by water vapor in the chosen analysis method. The preferred method also needed to be field tested to determine reliability, ease of use, and any unexpected or unforeseen problems.

Under future consideration was the development of a combined sampling and analysis unit. Once installed and calibrated, this unit would only require the costs of manpower to check on the system and retrieve a hard copy output of the data being generated. This system would be a merger of the continuous, isokinetic, stack sampler with a moving paper-tape holder that has designed into it positions for the isotopic source and a room temperature detector. The capital cost for such a system to handle two stacks would be \sim \$103,000, with manpower of about \$8,000, for a total first year cost of \sim \$111,000. Thereafter, the manpower costs would be minimal.

1.0 INTRODUCTION

The U.S. Nuclear Regulatory Commission (NRC) is charged with regulating and licensing uranium mill operators in terms of their radiological effluent and environmental monitoring programs. These regulations are contained in 10 CFR 20, "Standards for Protection Against Radiation," and 10 CFR 40, "Domestic Licensing of Source Material" (NRC 1982a,b). Of particular interest to this study is 10 CFR 40.65, "Effluent Monitoring Reporting Requirements." This section requires uranium mill operators to submit a semiannual report to the NRC on information that will allow estimation of doses to the public based on effluent releases. The NRC has also issued Regulatory Guide 4.14, Revision 1 (NRC 1980a), which stipulates that yellowcake drying and packaging stacks should be sampled at least quarterly. Information obtained from the stack sampling will then be used to estimate stack release rates and concentrations of natural uranium, ^{230}Th , ^{226}Ra , and ^{210}Pb released to the environment.

The NRC has requested Pacific Northwest Laboratory (PNL) to evaluate current methods being used to monitor stack emissions of yellowcake and suggest alternative or new procedures to obtain this information. To accomplish this task, previously reported procedures were first reviewed. Then, a few of the mills were contacted to determine how they complied with the reporting requirements. With this information, the currently used sampling and analysis procedures were evaluated. Other alternative procedures were also proposed and evaluated. Based on this work, a preferred and a recommended procedure are proposed. These methods still need additional laboratory and field evaluation, which could not be completed within the available funding.

2.0 Literature Review

The release of yellowcake to the environment has been qualitatively addressed in discussions of the sources of radioactive materials in and around uranium mills by Glauberman and Breslin (1964) and Sears et al. (1975). They recognized that next to a tailings pile, the airborne release of yellowcake was the highest potential source for human exposure. Based on knowledgeable estimates and assumptions, they assigned quantitative values to represent the source terms for the release of yellowcake to the environment. Realizing that few real-world measurements had been made, the Las Vegas Facility of the U.S. Environmental Protection Agency (EPA) decided to obtain actual measurements from operating uranium mills. The EPA measured the release rates of yellowcake from the exhaust stacks of six operating mills (Fort et al. 1980a). These data were eventually incorporated into the Model Uranium Mill, a part of the uranium fuel cycle discussed in the Final Generic Environmental Impact Statement on Uranium Milling (FGEIS) (NRC 1980b). This EPA study by Fort et al. (1980a) demonstrated that the yellowcake exhaust emission rate is dependent upon a number of parameters, which includes the following:

- ore processing rate
- average ore grade
- process efficiency for recovery of uranium

- efficiency of air cleaning equipment
- assumptions about operating time
- nature of operational procedures and consistency between operators
- cleanliness of the yellowcake stack and processing system including ductwork.

The measured 'instantaneous' yellowcake emission rate measured from a single stack varied by a factor of two to five due to the variation in the above parameters. In addition, when a major problem occurred with the air cleaning equipment of a given stack, a variation of as much as a factor of 20 was observed in the measured emission rate. Fort et al. (1980a) reported that the major parameter affecting the measured rate was most likely whether the yellowcake was being dried or packaged during the stack sampling procedure. Typically, he found that the air cleaning equipment ran continuously while the packaging and drying operation ran intermittently.

Fort et al. (1980a) recommended a general stack testing and emission rate calculation procedure to give reasonable estimates of the yellowcake source term. Briefly, he suggested isokinetically sampling each stack several times by EPA methods under each of the mills' operating modes and then time weighting these various emission rates according to the proportion of time during the year the mill operated in that mode. The sum of the time-weighted emission rates during the various operational modes would then be the yearly emission rate. This rate would apply only to the uranium emission. The daughter radionuclides would be determined from a blended sample of yellowcake collected over each quarter of the year.

Fort et al. (1980a) noted that a shortcoming of the stack sampling was that it represented a very small percentage of the time the stacks operated. Thus, the more tests conducted, the more accurate an estimate for the 'true' emission rate would be obtained. Therefore, the ideal procedure would be continuous sampling of the stack gases and particulates.

In considering the health effects of exposure to yellowcake particulates, two parameters must be considered: respirable particulate size and solubility of the yellowcake. If the particles are respirable, they may be deposited in the respiratory tract where solubility becomes important. Additional work was performed by Fort et al. (1980b) to determine the particle-size distribution of yellowcake emissions from the drying and packaging stacks of the United Nuclear Corporation mill at Churchrock, New Mexico. The mass median diameter of particulates in the packaging and drying stacks was determined to be 1.62 and 1.19 μm , respectively. If respirable particulates are defined as those particles with aerodynamic diameter of $\leq 2.5 \mu\text{m}$, then 69% of the particles in the packaging stack and 90% of the particles in the drying stack were respirable.

The solubility of yellowcake influences the physiological uptake of uranium into the body. Kalkwarf (1979) investigated the solubility of yellowcake and found that it is dependent on the specific uranium compound present.

Momeni et al. (1979) measured the radioisotopic composition of yellowcake samples obtained from four mills. The yellowcake samples were blended samples that represented a year's production of yellowcake. The isotopes ^{238}U , ^{235}U , ^{234}U , ^{230}Th , ^{226}Ra , and ^{210}Pb were measured. Based on the previously determined stack release rate, the amount of yellowcake released was found to be 0.1% of the amount processed.

3.0 Reporting Requirements

Monitoring of yellowcake drying and packaging stacks is required at least quarterly by the NRC (1980a). The sampling should be isokinetic and representative, and adequate for the determination of the release rates and concentrations of natural uranium in the stack gases. The sampling should also be adequate for the determination of the radionuclide release rates and concentrations of ^{230}Th , ^{226}Ra , and ^{210}Pb if these data cannot be obtained from other sources such as isotopic analysis of the yellowcake product. At the time of sampling, the stack flow rates should also be measured.

A semiannual report to the NRC is required, which includes a listing for each yellowcake exhaust stack sample of 1) location of sample; 2) dates during which the sample was collected; 3) concentration of natural uranium, ^{230}Th , ^{226}Ra , and ^{210}Pb ; 4) the percentage of the appropriate concentration limit found in Table II of Appendix B to 10 CFR 20; 5) the estimated release rate of natural uranium, ^{230}Th , ^{226}Ra , and ^{210}Pb ; and 6) the flow rate of each stack. In addition, the report should include 1) a description of the sampling equipment, 2) a discussion of how the sampling location was chosen, 3) a description of sampling procedures, 4) a description of analytical procedures, 5) a description of calculational methods, 6) an error analysis, and 7) a description of the calibration procedures. The above extracted items are those that are pertinent to the monitoring of yellowcake emissions from the ventilation exhausts at uranium mills.

A number of mills were contacted to determine how they actually met these reporting requirements. They responded that 1 to 3 stacks generally serviced the yellowcake drying and packaging operations. These stacks usually had air cleaning equipment that were of the venturi scrubber type (which removes 99.8% of the particulates) or of the wet impingement scrubber type (which removes 98% of the particulates). The mills sampled the stacks once per quarter unless required to do so more often by state authorities, e.g., the state of Colorado requires once-a-month testing. The stack testing was normally performed under isokinetic conditions and followed the prescribed procedures for EPA Method 5 (EPA 1983). The measured stack gas velocity varied from $0.3 \text{ m}^3/\text{s}$ up to $6 \text{ m}^3/\text{s}$ with a measured emission rate of 0.009 to 0.14 kg/h of yellowcake (U_3O_8). The ventilation exhausts on the drying and packaging operations usually ran when the mills were operating. However, an operating mill did not imply the drying and packaging processes were online.

Therefore, to estimate the total emissions of yellowcake from the ventilation exhaust stacks, the information from the stack sampling performed each quarter was used. That is, the analysis of a sample taken for 0.5% of the total operating time was used to estimate the total emissions for a quarter. (It was assumed that the stack sampling took 8 hours and the total operation of the mill for a quarter was 13 weeks, 5 days per week, 24 hours per day.)

One question that might be raised is how representative the emissions monitored from the ventilation exhausts are to the actual emissions of the ventilation exhausts in light of the numerous parameters that can affect the emission rate. These parameters include the performance of the stack-gas cleaning equipment over time; whether the drying system or packaging systems are operating individually, together, or not at all when the ventilation exhaust is operating; possible variations in the stack gas velocity over time; variation in the uranium content of the yellowcake over time; and the variation in the amount of ore processed over time.

4.0 Proposed Monitoring Methods

The determination of the emission rate of yellowcake from ventilation exhausts at uranium mills poses two separate, but related questions: 1) How is a representative sample of yellowcake collected? and 2) How is it analyzed? To help answer these questions, a determination was made as to the current practices of various mills. The mills sample isokinetically, using commercially available-stack sampling equipment that meets EPA Method 5 requirements. The ventilation exhaust particulates are collected by deposition on pre-weighed filter paper. The filter paper is then sent to a laboratory for analysis of natural uranium, ^{226}Ra , ^{230}Th , and ^{210}Pb . The analysis of natural uranium is often done by fluorometry (Merritt 1971), ^{226}Ra is often analyzed by the radon-emanation procedure (Sill 1977), ^{230}Th is analyzed by wet chemical separations followed by alpha spectroscopy analysis (Sill 1977), and ^{210}Pb is analyzed by wet chemistry followed by beta-counting to measure the ^{210}Bi daughter of ^{210}Pb (Sill 1977).

The major criticism of the stack sampling methods currently being used is the lack of 'representativeness' of the samples in light of the short sampling time (2 to 8 hours) versus the total operational time (1560 hours) for a quarter.

4.1 Sampling Methods

We will discuss three ways in which a continuous sampling or analysis might be accomplished. The first method uses a continuous, automatic, isokinetic sampler^(a). This instrument constantly monitors the stack gas velocity, temperature, and pressure differentials to ensure a continuous, isokinetic particulate sample.

The second method uses an electrostatic precipitator designed for continuous use. Particulates may be collected isokinetically using electrostatic precipitation in a sampler such as the Isokinetic Dust Sampler^(b). This instrument collects an integrated particulate sample on the walls of an aluminum tube, which can be exchanged with a new tube. The sample weight can be determined by difference and analyzed individually or composited with other samples. Therefore, if the stack flow has been characterized, the fraction of the total daily release collected by this instrument is known. The total emission rate is easily calculated knowing the length of sample exposure. An advantage of this method is that only 110 V AC power or a 12 V battery is required to operate the sampler. Also this method does not require monitoring of the stack gas velocity.

a) Isokinetic sampler, Model 1275, is a product of KURZ Instruments, Inc., Carmel Valley, CA.

b) Isokinetic Dust Sampler, Model 3110, is a product of TSI Incorporated, St. Paul, MN.

The third method monitors the particulate output of the stacks using an optical method. This could be done by either single or double pass opacity methods or a ground-based lidar (laser-radar) system.

4.2 Analytical Methods

The method generally used to determine natural uranium is fluorometry, which is a labor-intensive procedure that requires constant oversight by a laboratory technician. Other methods used include radiochemical preparation followed by alpha spectroscopy or colorimetry, which are also labor intensive. Two more recently developed techniques are the determination of uranium by isotopic excitation x-ray fluorescence (XRF) analysis and direct gamma-ray analysis using an intrinsic germanium (IG) detector. The analytical procedure needed should require a minimum of time for analysis, since a sequentially integrating continuous sampling method will produce at least 2 to 3 samples per stack per day of mill operation. This could imply up to 195 samples per quarter per stack, which is a significant increase in sample load over current practices.

If the analytical work is performed in an onsite laboratory, the following three techniques are relatively fast and require a minimum of sample handling and preparation: 1) direct gamma-ray analysis using an IG detector, 2) isotopically excited XRF, and 3) laser excitation kinetic phosphorimetry. The particular technique chosen would partially depend on the sampling method used. For instance, if a continuous, automatic, isokinetic stack sampler were used, the sample would be deposited on a piece of filter paper, which could then be analyzed by direct gamma-ray spectroscopy or isotopically excited XRF without further preparation. However, the isokinetic electrostatic precipitator would require that the deposited particulates be washed out of the aluminum collection tube and prepared for analysis. The sample could then be analyzed by the kinetic phosphorimetry procedure, or the yellowcake material could be dried and collected until a sufficient amount of material was obtained for analysis by direct gamma-ray analysis or isotopically-excited XRF.

If a self-contained package that will perform both the sampling and analysis is preferred, one of the following two options may be used. Both options require interfacing to the continuous, automatic, isokinetic stack sampling system. The first option uses a sampler with paper-tape as the collection substrate and the subsequent analysis is done by isotopically excited XRF. The second option requires collection of the particulates in a liquid medium, dissolution of the particulates, and presentation of the liquid sample to a laser-excited kinetic phosphorimetry system.

5.0 Laboratory Evaluation of Several Methods for Yellowcake Analysis

A sample of yellowcake (76.3% uranium) was obtained for evaluation of the various analytical methods proposed. The natural uranium decay chain is shown in Figure 1. In this case, since the chemical product is purified yellowcake (U_3O_8), the decay products observed would essentially end at ^{234}U ($t_{1/2} = 2.446 \times 10^5$ yr). Table 1 lists the radioactive properties of these five radionuclides.

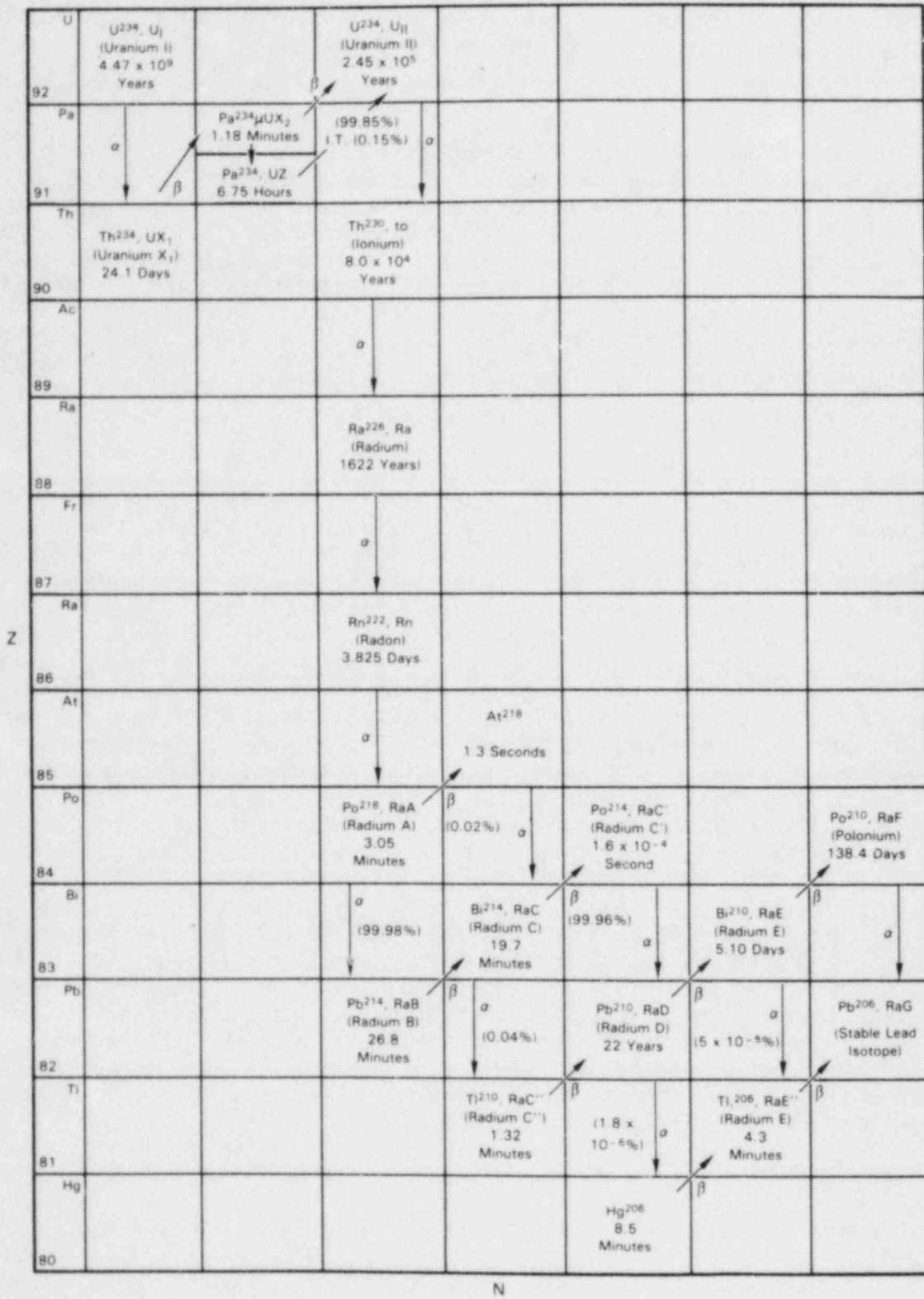


FIGURE 1. The Uranium Series

TABLE 1. Uranium Series Radionuclides of Interest
(after Leaderer et al. 1978)

Nuclide	Half-life	Major Radiation Energies and Intensities (MeV)		
		α	β	γ
238U	4.468 x 10 ⁹ yr	4.196 (77%)	---	0.04955 (0.32%)
		4.149 (23%)	---	---
234Th	24.101 day	---	0.1985 (72.5%)	0.06328 (3.6%)
		---	0.104 (20.7%)	0.09280 (3.2%)
		---	0.060 (5.4%)	---
		---	0.022 (1.3%)	---
234mpa	1.175 m	---	2.29 (98%)	1.001 (0.56%)
		---	---	0.766 (0.20%)
		---	---	0.258 (0.057%)
		---	---	0.742 (0.057%)
234pa	6.75 h	---	0.512 (63)	0.09844 (27%)
		---	0.680 (19%)	0.1312 (20%)
		---	0.280 (12%)	0.09466 (15%)
		---	1.19 (5%)	0.8808 (13.0%)
		---	---	0.8832 (12.0%)
		---	---	0.9461 (12.0%)
		---	---	0.9260 (11.0%)
		---	---	0.5695 (10.7%)
234U	2.446 x 10 ⁵ yr	4.774 (72%)	---	0.0532 (0.118%)
		4.723 (23%)	---	0.1210 (0.041%)
230Th	8.0 x 10 ⁴ yr	4.6875 (76.3%)	---	0.0677 (0.37%)
		4.6210 (23.4%)	---	0.1436 (0.044%)
		---	---	0.254 (0.011%)
		---	---	0.110 (0.0063%)
226Ra	1599 yr	4.7845 (94.45%)	---	0.6093 (46.1%)
		4.6015 (5.55%)	---	0.3519 (37.1%)
		---	---	0.2952 (19.2%)
		---	---	1.7645 (15.9%)
		---	---	1.120 (15.0%)
		---	---	1.2381 (5.92%)
		---	---	0.7684 (4.88%)
		---	---	1.3777 (4.02%)
210pb	22.26 yr	3.72 (1.7 x 10 ⁻⁶)	0.061 (19%)	0.04645 (4.05%)
		---	0.015 (81%)	---

5.1 Direct Gamma-Ray Spectroscopy

The spectroscopic methods rely on the inherent activity of natural uranium, ^{238}U , and its gamma-emitting daughters (^{234}Th , $^{234\text{m}}\text{Pa}$, ^{234}Pa , and ^{234}U) found in yellowcake (loosely defined as U_3O_8). Direct gamma-ray spectroscopy was used to collect spectra from a pellet (5.1 cm dia. x 0.4 cm high) made from 15.0 g of yellowcake and 3.0 g of cellulose binder. A lithium-drifted germanium detector (Ge(Li)) with a volume of 80 cm^3 , 16.0% efficiency, and a full width at half maximum (FWHM) of 1.68 keV resolution for the 1332 keV peak of ^{60}Co was used to collect the spectrum shown in Figure 2. The following isotopes, which are of interest in analyzing yellowcake, were observed: ^{234}Th , $^{234\text{m}}\text{Pa}$, ^{234}Pa , ^{235}U , Pb x-rays, and ^{234}U .

Since the main peaks of interest are at a fairly low energy, a planar IG detector was also used to collect a gamma-energy spectrum (Figure 3). To duplicate the kind of sample and mass of yellowcake expected from the stack sampling system, a filter sample was prepared by depositing 10 mg of yellowcake onto the filter surface. This detector has the advantage of better resolution and efficiency at low energies when compared to the Ge(Li) detector. The IG detector also is not susceptible to damage when liquid nitrogen is not available, whereas a Ge(Li) detector would be destroyed if allowed to come to room temperature. The following isotopes, which may be used in the analysis of the yellowcake product, were observed by the IG detector: ^{234}Th , ^{235}U , and Th L x-rays. A count of 2000 s on a 1900 mm^2 planar intrinsic germanium detector would have a detection limit (3 sigma above background) of 0.4 mg of uranium based on the 63.3 keV peak of ^{234}Th .

5.2 Isotopically Excited X-Ray Fluorescence

We evaluated the usefulness of two excitation sources for the analysis of uranium in yellowcake. The first was ^{109}Cd , which excites the L shell of the uranium nucleus. Relaxation to the ground state will result in the emission of L x-rays at 13.613 keV($L_{\alpha 1}$), 13.438 keV($L_{\alpha 2}$), 17.218 keV($L_{\beta 1}$), and 16.425 keV($L_{\beta 2}$). The second source was ^{57}Co , which will excite the K shell of the uranium nucleus and result in the emission of K x-rays at 98.428 keV ($K_{\alpha 1}$) and 94.648 keV($K_{\alpha 2}$).

In preparation for investigating the use of isotopic-excitation sources, a series of uranium standards were prepared to discern the self-absorption due to increased uranium content. These measurements were used to prepare a calibration curve that compared uranium concentration to instrument response.

5.2.1 Cadmium-109 Isotopic Source

The ^{109}Cd isotopic source has a photon emission line at 88 keV and silver x-rays at 22 keV (fluorescence yield of 75%), which was used for excitation of the L-shell electrons of the uranium atom. Uranium x-rays at 13.613 keV ($L_{\alpha 1}$), 13.438 keV($L_{\alpha 2}$), 17.218 keV($L_{\beta 1}$), and 16.424 keV($L_{\beta 2}$) are then detected by a lithium-drifted silicon detector [Si(Li)] used for low energy photon analysis. In our case, an 80 mm^2 planar Si(Li) detector (FWHM of 200 eV at the 6.4 keV line) was used to detect the L x-rays. This detector was coupled to a Canberra Series 80 multichannel analyzer that was part of a PDP 11/34

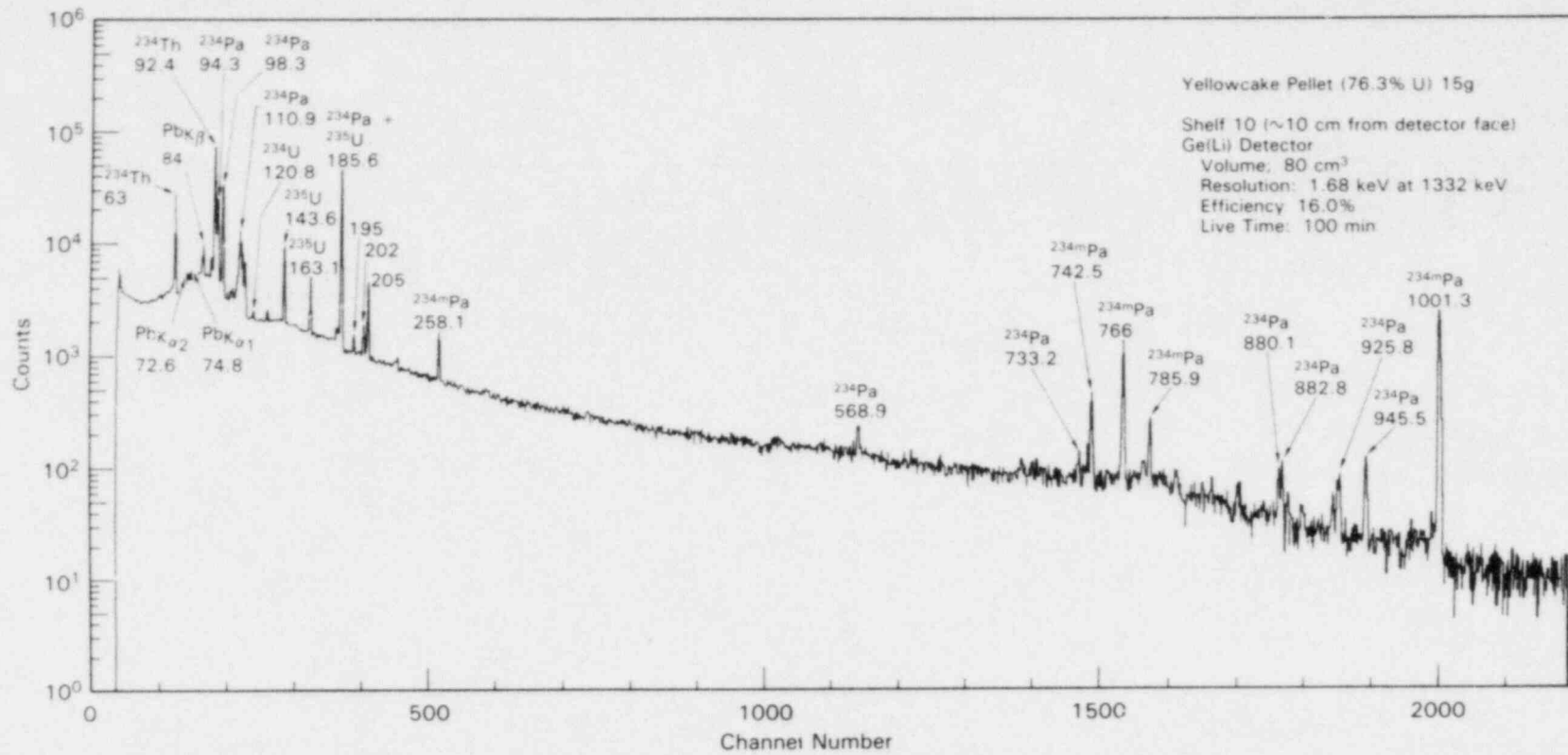


FIGURE 2. Spectrum of Yellowcake Pellet Collected by a Ge(Li) Detector

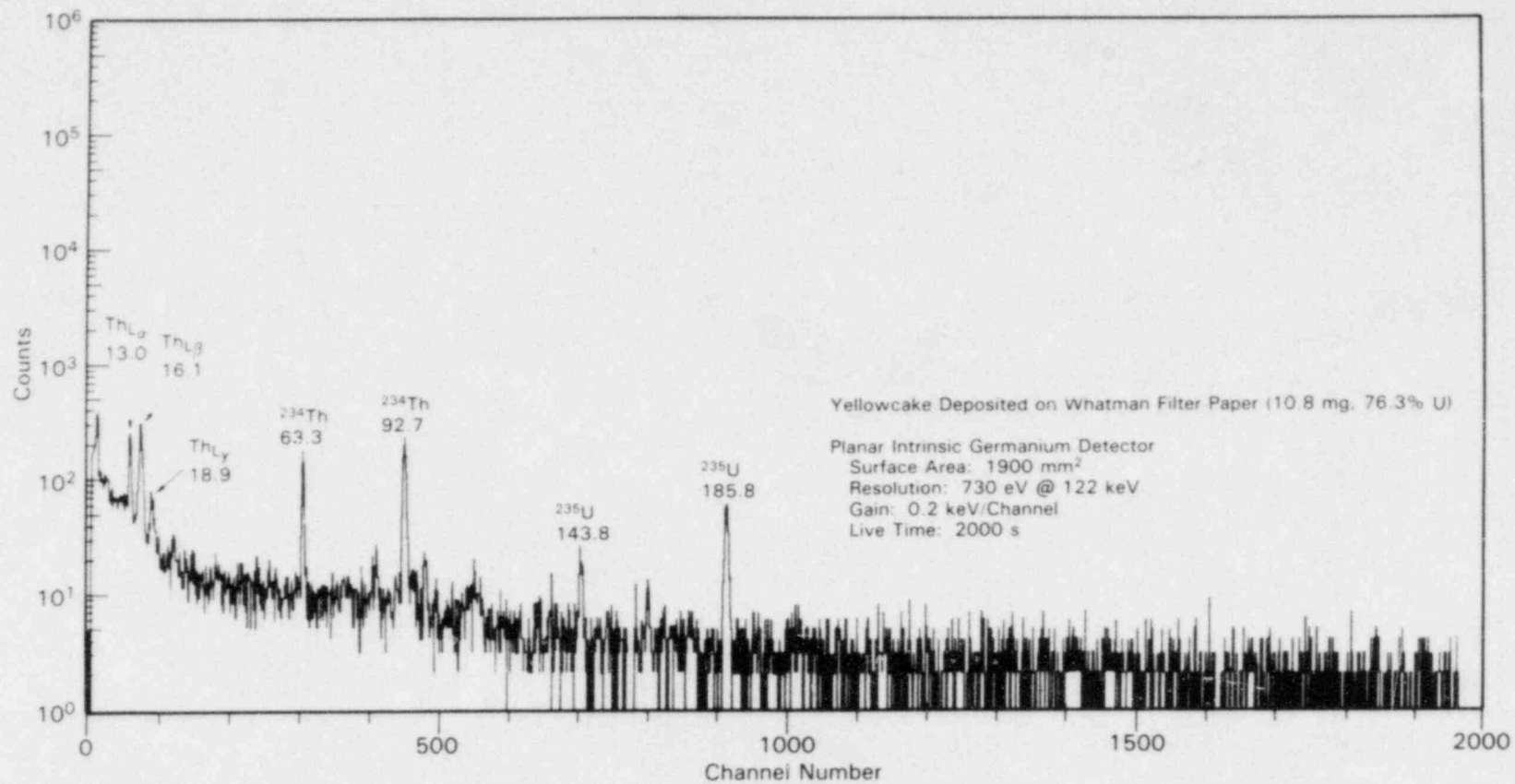


FIGURE 3. Spectrum of Yellowcake Deposited on Whatman Filter Paper and Counted with an Intrinsic Germanium Detector

computer based data-acquisition system^(a) used for data analysis. The ^{109}Cd ($T_{1/2}=462.3$ d) had an activity of 8.9 mCi as of November 29, 1983, and had been prepared as a ring source mounted on a lead holder.

The absorption of the emitted x-rays by Whatman filter substrate paper^(b) was investigated using a filter upon which yellowcake had been deposited. Spectra were collected with the deposited yellowcake material facing away from and then facing toward the source and detector. The results showed no significant absorption of the L_{α} line of uranium by the filter paper substrate.

Next, a series of standards were prepared by evaporating various concentrations of uranium onto cellulose filter material. This was done to determine the effect of particle loading on the observed response. A loading correction equation was then determined, which related response to concentration. A series of Whatman filters were then prepared by a puff technique (Giauque et al. 1972) and weighed. The filter deposition varied from 2.0 to 30 mg of yellowcake. A series of 22 counts were made, and the results were compared to the known amounts. A typical spectrum is shown in Figure 4. It was found that the average of the ratios of the experimental-to-known concentration was calculated to be 0.94 ± 0.15 . A three sigma detection limit was calculated to be 5 μg , based on a 500 s count and the ^{109}Cd source discussed earlier.

5.22 Cobalt-57 Isotopic Source

A 10 mCi (as of November 15, 1983) isotopic source of ^{57}Co was obtained and used for excitation of the K shell of uranium. The radioisotope, ^{57}Co , has an emission line at 122 keV which excites the K shell of uranium and results in the emission of x-rays at 98.428 keV($K_{\alpha 1}$) and 94.648 keV($K_{\alpha 2}$). These lines are easily detected and resolved by a planar IG detector used for low energy photon analysis (500 mm^2 area and a FWHM of 530 eV at the 122 keV line).

The cellulose uranium standards prepared earlier were then run to determine the effect of particle loading on the response curve. Figure 5 shows the response curves for the ^{57}Co source and for the ^{109}Cd source. As can be seen, the ^{57}Co response curve is linear and shows no loading effect. The ^{109}Cd response curve is also linear, but the lower slope indicates self-absorption effects as the mass increases.

Using the response curve obtained for the ^{57}Co source, the Whatman filters with yellowcake deposited on them were analyzed. A typical spectrum is shown in Figure 6. The experimentally determined mass was then compared to the known mass. For six filters, the average of the experimental to known ratio was found to be 0.97 ± 0.11 .

A three sigma detection limit was calculated to be 0.6 mg. This limit was based on the ^{57}Co (10 mCi) source, a live time of 600 s, and the IG detector discussed previously in Section 5.21.

a) The data-acquisition system is the Jupiter data-acquisition system, a product of Canberra Industries, Inc., Meriden, CT, 06450.

b) Whatman filter paper is a product of Whatman, Inc., Clifton, NJ, 07014.

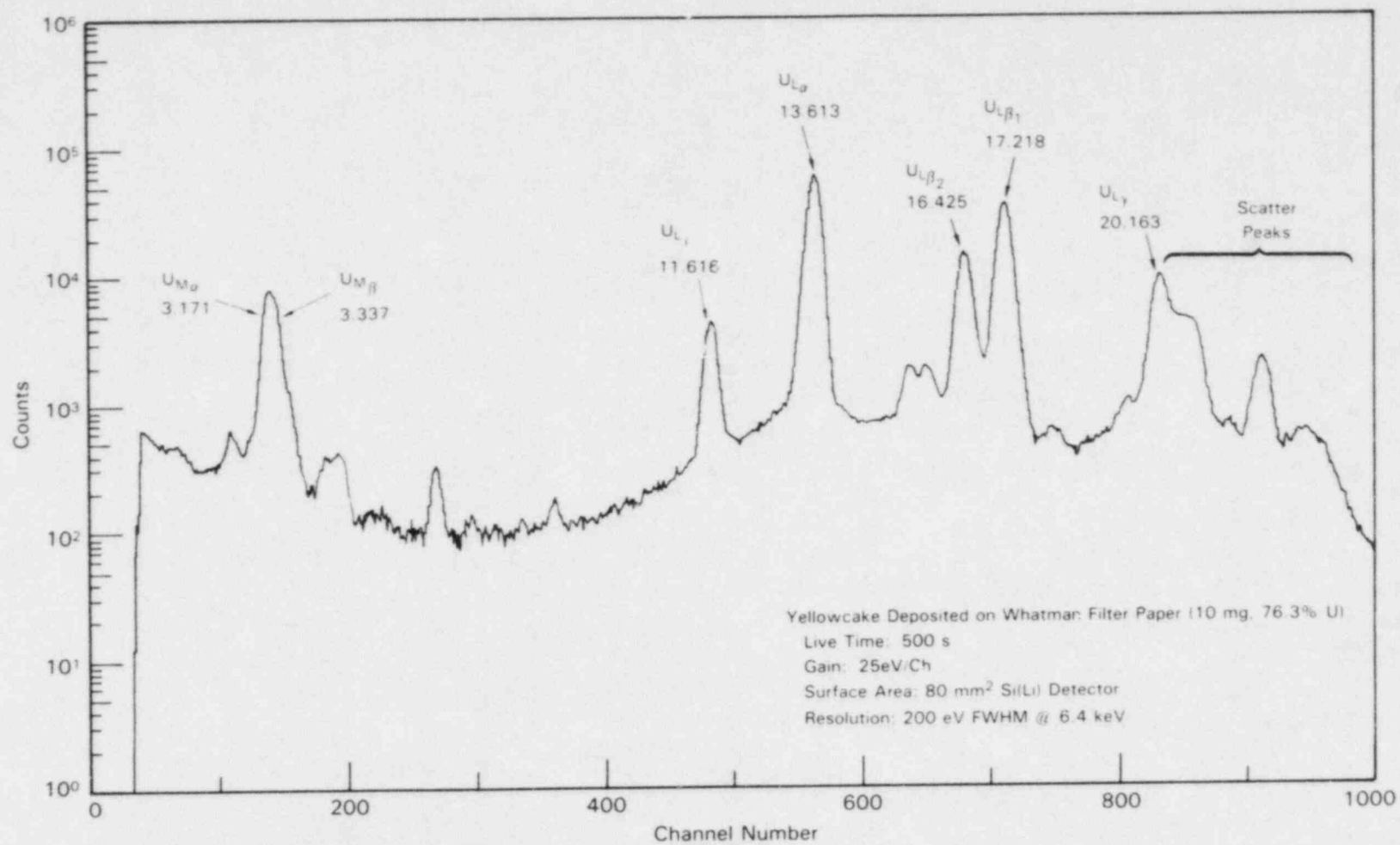


FIGURE 4. X-Ray Fluorescence Spectrum of Yellowcake Deposited on Whatman Filter Paper - ^{109}Cd Excitation Source

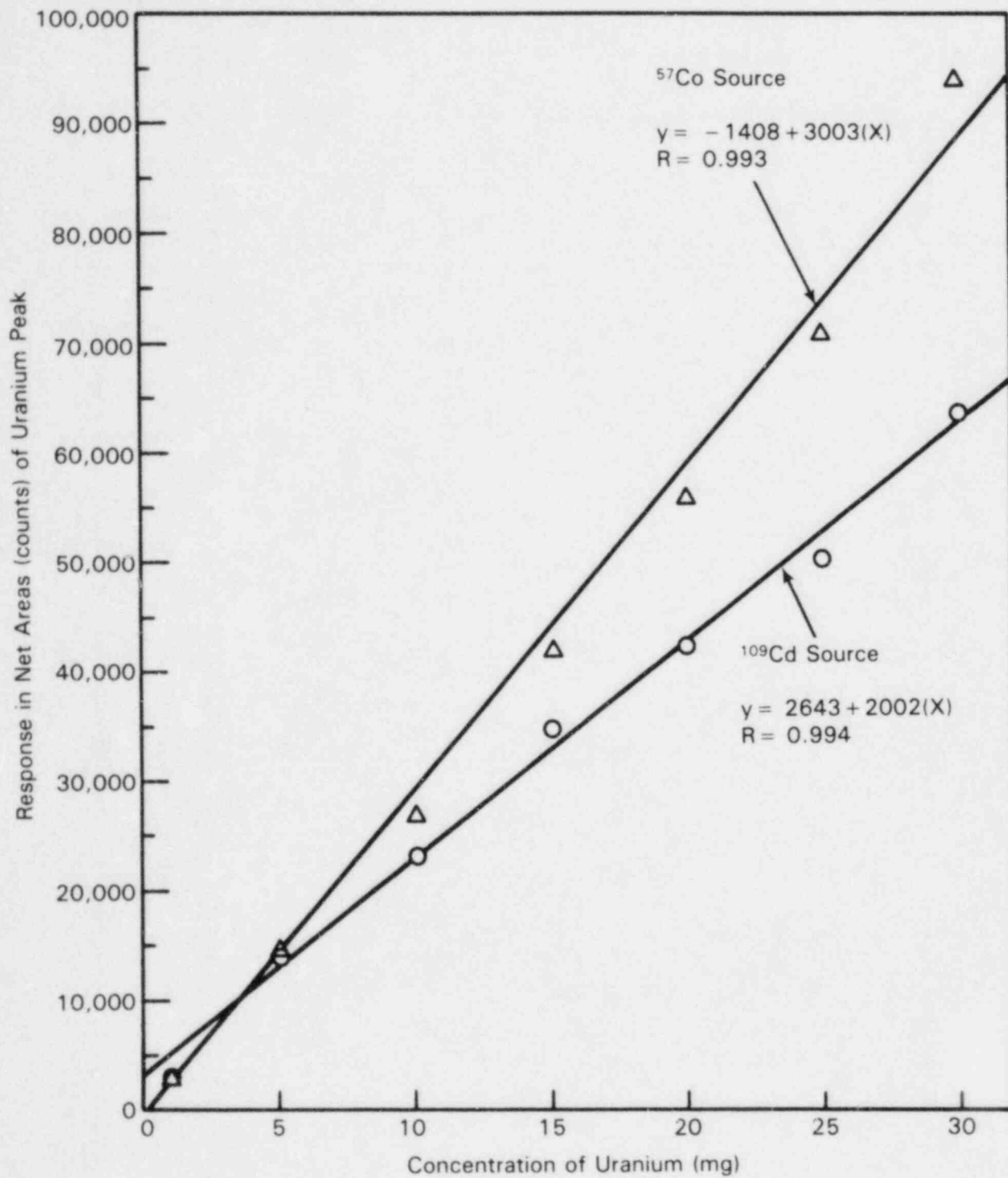


FIGURE 5. Instrument Response Versus Concentration for Uranium Cellulose Standards Exposed to Isotopic Sources

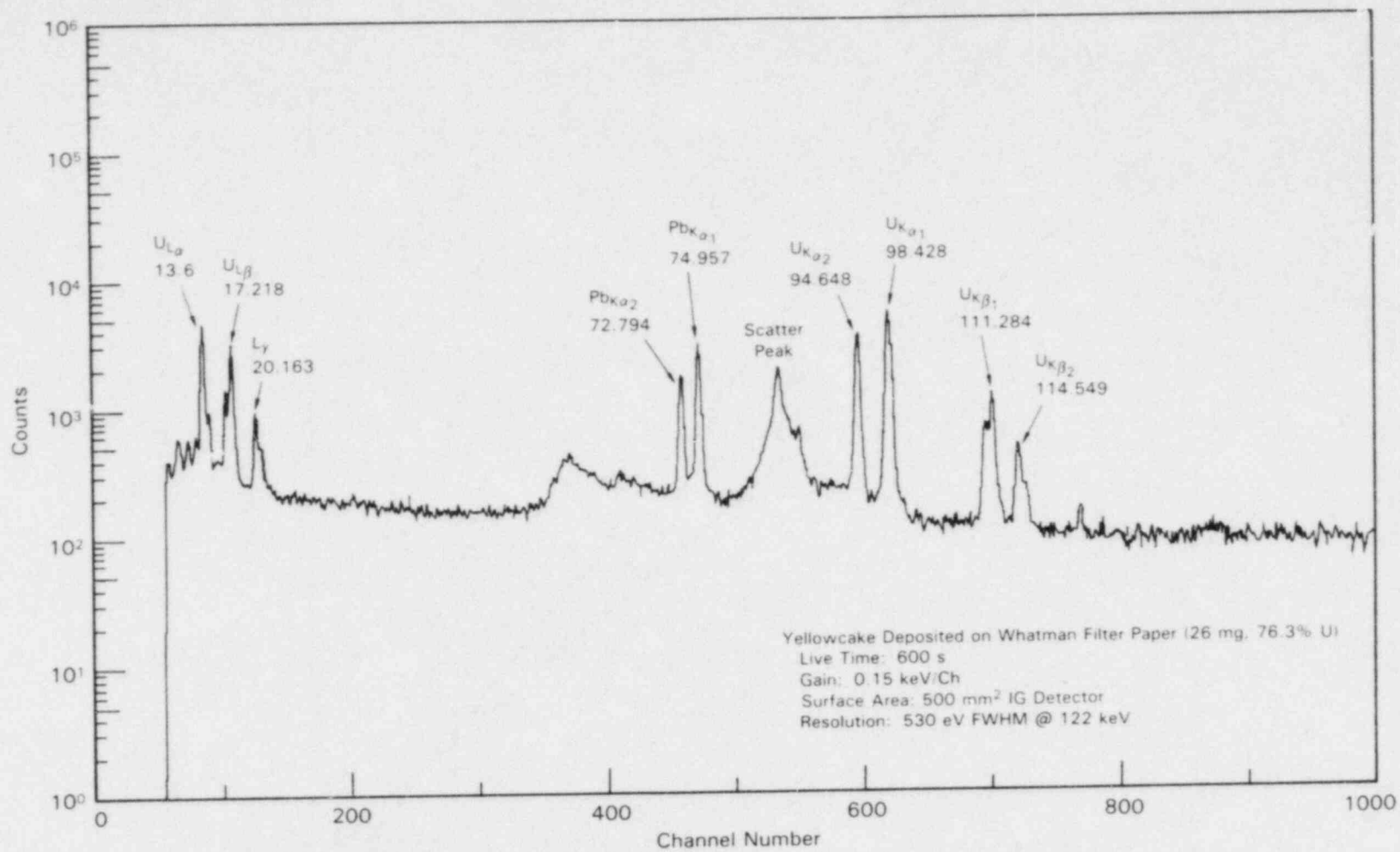


FIGURE 6. X-Ray Fluorescence Spectrum of Yellowcake Deposited on Whatman Filter Paper - ^{57}Co Excitation Source

5.3 Laser-Excited Kinetic Phosphorimetry

Laser-excited kinetic phosphorimetry is a new and powerful analytical technique that was applied to the analysis of uranium. This technique is sensitive to the ppt range (Bushaw 1983). The technique uses pulsed, dye-laser excitation with time-resolved photon counting to measure the concentration of uranium. Multi-channel scalar (MCS) photon counting helps correct for interferences caused by short-lived emitted species and scattered light. Kinetic analysis of the time-resolved spectra allows correction for possible phosphorescent quenching effects.

A minimal amount of sample preparation is required. The yellowcake particulates need to be dissolved in a nitric acid solution and then mixed into a dilute phosphoric-acid solution for measurement. Approximately 0.5 mL of sample solution is required for the analysis by the broadband dye laser-pumped system. The detection limit for uranium was estimated at 100 pg by this technique. The time for analysis by the laser system is less than 5 min per sample, with some additional time required for sample preparation and data reduction.

This laser-excited kinetic phosphorimetry technique might be used in two different ways depending on the method of sample collection. If the particulates are collected by the electrostatic precipitator system or by the stack sampling system, the samples could be returned to the laboratory for analysis. Otherwise, one could interface a continuous stack sampling system to a continuous sample collection and dissolution stream that feeds the laser-excitation analysis system. Using an expected emission rate of 0.001 lb/h of U_3O_8 and a stack gas flow of 2×10^5 DSCFH (Fort et al. 1980a), a uranium concentration of 0.1 ppb could be continuously measured. This assumes a water flow in the initial mixing chamber of 100 mL/min and the stack gas being sampled at 0.1 SCFM. A 0.05 mL aliquot of the water stream would be mixed with 0.05 mL of HNO_3 and 1.0 mL of 5% H_3PO_4 . This would result in a measurement of a concentration of 0.105 $\mu g/L$.

6.0 Comparison of Methods

A discussion of the stack sampling methods and sample analysis procedures discussed in previous sections resulted in the following list of methods. These methods have been further subdivided by the general analytical procedure to be used (Table 2).

Discussion of the possible methods in terms of their advantages and disadvantages are presented in the remainder of this section.

6.1 Laser Methods

The laser methods may be grouped into two categories. The first category is made of purely optical procedures such as lidar or in-stack opacity-type systems. These two methods suffer from a lack of specificity (i.e., they offer no speciation information), a dependence on the particle size distribution that could vary from mill to mill, and possible interference from water vapor in the air stream. Thus, they are not recommended at this time.

The second category incorporates kinetic phosphorimetry as the analysis method. There are two different isokinetic stack-sampling procedures that could be used to present the sample to the kinetic phosphorimetry system.

TABLE 2. Sampling and Analytical Methods for Yellowcake Analysis from Exhaust Vents

- A. Laser Methods
 - 1. In-stack and lidar
 - 2. Kinetic phosphorimetry
 - a. Electrostatic precipitator collection
 - b. Continuous, automatic, isokinetic stack sampler
- B. Direct-Spectrometric Methods
 - 1. Electrostatic precipitator and IG counting
 - 2. Continuous, automatic, isokinetic stack sampler and IG counting
- C. Isotopically Excited X-Ray Fluorescence
 - 1. Continuous stack sampler, ^{109}Cd source, and Si(Li) counting
 - 2. Continuous stack sampler, ^{57}Co source, and IG counting
 - 3. Continuous stack sampler, filter tape assembly, ^{57}Co source, and IG counting

If the electrostatic precipitator were used to collect the sample, the collection tube would be returned to the laboratory. Then, the material would be washed out of the sample collection tube by dilute nitric acid, an aliquot dissolved in phosphoric acid, and analyzed by the kinetic phosphorimetry system. If the continuous, automatic, isokinetic stack-sampler were used, the sample collected on the filter could be processed as above with the kinetic phosphorimetry system. It should also be possible to design a fully self-contained system that can sample the airstream isokinetically, dissolve the particulates in a dilute nitric acid solution, mix an aliquot with the phosphoric acid solution, and analyze by the laser-excited kinetic phosphorimetry technique.

The electrostatic precipitator system is fairly inexpensive, requires only 110 V, and is purported to be isokinetic. However, the reliability of the electrostatic precipitator has not been demonstrated for continuous use. The continuous, automatic, isokinetic stack-sampler provides an isokinetic

sample collected on a filter substrate. By sampling isokinetically, this system provides a representative sample. It has been field tested for continuous use and has an easily removable filter assembly. The stack-sampler requires 110 V AC power and consists of a couple of fairly heavy components that are not easily moved. It is also more complex both electronically and mechanically than the electrostatic precipitator. If filter collection of the particulates is used, kinetic phosphorimetry may be readily used in a central laboratory. If instead, an integrating sample-dissolution system feeding a kinetic-phosphorimetry system is chosen, a device will need to be designed. This device, although feasible, will require both clever adaptation of the currently available equipment and a complete system for each stack being sampled.

6.2 Direct-Spectrometric Methods

The two sampling methods associated with direct-spectrometric counting are differentiated by the stack-sampling method chosen. As mentioned previously, the electrostatic precipitator has not been proven under continuous sampling conditions. The yellowcake exhaust stacks would presumably be sampled for 8-h periods resulting in three samples per day per stack. The yellowcake could be washed out of the collection tube and deposited onto a piece of Whatman filter paper, dried, and then analyzed by gamma-ray spectroscopy. Depending on the amount of material collected and method sensitivity, it might be possible to analyze individual samples. If insufficient sample is collected during each sampling period, the daily filters may be combined before analysis to produce an integrated sample representing a day of operation or to reduce the number of samples that need to be analyzed.

The yellowcake samples obtained from the continuous, automatic, isokinetic stack sampler would be deposited on Whatman filter paper. The filter paper would be pressed into a pellet in the laboratory to attain a standard geometry. This pellet would provide a reproducible geometry for counting on the IG detectors. Sample preparation would then be fairly minimal. Analysis time would be approximately 40 min per sample, probably not fast enough to handle the sample load.

6.3 Isotopically Excited X-Ray Fluorescence

The isotopically excited XRF methods are based on sample collection by the electrostatic precipitator or the continuous, automatic, isokinetic stack sampler. Analysis is performed using either a ^{109}Cd or a ^{57}Co source. The advantages and disadvantages of both stack-sampling systems were discussed in Section 5.2. Since proper continuous sample collection by the electrostatic precipitator has not been proven, the electrostatic precipitation followed by isotopically excited XRF analysis is not presently favored.

Therefore, sample collection by the continuous, automatic, isokinetic stack sampler with isotopically excited XRF analysis will be discussed. Assuming the sample has been collected on a filter substrate that is returned to an on-site laboratory, the analysis may be performed using either a ^{109}Cd or a ^{57}Co isotopic source.

The ^{109}Cd isotopic source preferentially excites the uranium L x-rays, which are detected by a Si(Li) detector, so the method is specific for uranium.

However, this method exhibits self-absorption due to sample loading on the filter paper. This can be corrected by generating a calibration curve like that discussed in Section 5.2. The analysis time would be relatively short, a livetime of 10 min per sample. The half-life of ^{109}Cd is 453 days, 1.7 times longer than that of ^{57}Co . This means the ^{109}Cd source would be usable for almost twice as long as the ^{57}Co source if both contained the same original activity.

The ^{57}Co source is also specific for uranium by exciting the K x-rays of uranium, which are then detected by the IG detector. The peaks are quite clean and are at a fairly high energy (94 and 98 keV) so that self-absorption is not a significant problem. The analysis time would be relatively short, a livetime of 10 min per sample with 5 min of sample preparation.

The last method is another variation on both the stack sampling and ^{57}Co excitation with detection by an IG detector. The improvement in this case, is to incorporate a moving paper-tape sampler into the stack sampling system. After sample collection, the paper-tape is moved forward to a position where the sample can be analyzed by a ^{57}Co source and IG detector. A microprocessor would be used to control the data collection, serve as a multi-channel analyzer, and determine the uranium concentration present. This would essentially be a continuous, integrated sampling and analysis system. In one package, the sample collection and data analysis would be performed automatically with a hard copy readout of the results. This system would require minimal oversight on the part of the mill personnel after the system was set up. A disadvantage to the system is the continuous requirement for liquid nitrogen to cool the IG detector during operation.

7.0 Cost Comparison of Methods

Based on the discussion in Sections 5 and 6, the methods listed in Table 3 are deemed feasible. The estimated costs for the analysis by each method was broken down into capital and manpower costs. The capital costs are expressed in 1983 dollars. The manpower costs were determined by assuming a man hour charge of \$18 per hour and 5 hours per day required for sampling and analysis. Thus, over a period of a year, assuming 5 days of operation each week, 1300 man hours would be required. The total cost then reflects the capital and manpower costs for the sampling of two stacks for three periods each day and the analysis for total uranium content. After the initial capital outlay, the cost per year should principally be due to man-hour charges.

The electrostatic precipitator collection method coupled with isotopic excitation and spectrometric analysis appears to be the lowest cost technique. If the electrostatic sample collection method was proven to be reliable for continuous sampling, it probably would be the preferred method. However, since the reliability is unproven, methods 1 and 3 cannot be recommended at this time. Therefore, methods 2 and 4 are left, with method 4 being preferred because of cost and ease of implementation. In terms of total capital cost, the difference between the ^{109}Cd source and the ^{57}Co source is minimal (approximately \$400). The ^{109}Cd has a longer half-life, meaning it would not have to be replaced as often as the ^{57}Co . However, the ^{57}Co excites the K x-rays of uranium, which, because of their relatively high energy, show little self-absorption compared to the L x-ray lines of uranium excited by the ^{109}Cd . Also, the K x-ray lines of uranium are free of interferences, while the L_{α} lines are not as well resolved. Thus, the ^{57}Co source is the preferred isotopic source.

TABLE 3. Feasible Sampling Methods and Estimated Costs
(1983 dollars)

Sampling Methods	Capital Cost per 2 Stacks	Manpower Sampling and Analysis	Total Estimated Cost
1. Electrostatic precipitator collection with kinetic phosphorimetry analysis	\$36,400	\$23,400	\$59,800
2. Continuous, automatic, isokinetic stack sampler with kinetic phosphorimetry analysis	\$56,000	\$23,400	\$79,400
3. Electrostatic precipitator with isotopic excitation and spectrometric analysis			
a. ^{109}Cd source	\$26,300	\$23,400	\$49,700
b. ^{57}Co source	\$25,900	\$23,400	\$49,300
4. Continuous, automatic, isokinetic stack sampler with isotopic source and spectrometric analysis			
a. ^{109}Cd source	\$45,800	\$23,400	\$69,200
b. ^{57}Co source	\$45,400	\$23,400	\$68,800

8.0 Conclusions and Recommendations

The preferred method for continuous monitoring of the yellowcake ventilation exhausts with current technology is the use of a continuous, automatic, isokinetic stack sampler to collect particulates on a filter substrate. However, we recommend at least three samples per day per stack be collected at each mill. The filter samples would be brought to a laboratory at the mill site for analysis by isotopically excited XRF. A ^{57}Co radioisotopic source would be used to excite the K x-rays of the uranium nucleus. The photons would be detected by an intrinsic-germanium low-energy photon detector kept cool by liquid nitrogen. The detector signal would be fed to an MCA with the appropriate built-in electronics and data readout.

Future work should investigate the feasibility of a room temperature detector that could then be part of a complete, self-contained system. This system should also incorporate a moving paper-tape collection system and isotopic analysis. One such detector being developed is called a mercuric iodide detector (Hg_2I_2). It would be useful to measure the L x-ray lines of uranium using the ^{109}Cd source.

The originally proposed work in this project was not completed because of funding redirection by the sponsor. Yet to be investigated are possible interferences due to water vapor in the chosen analysis method. The chosen method was also to be tested in the field to determine reliability, ease of use, and any unexpected or unforeseen problems.

Future considerations should involve an evaluation of the feasibility of a room temperature detector (e.g., a mercuric-iodide detector) instead of a liquid nitrogen-cooled IG detector. This would be extremely valuable if sensitivity were not significantly reduced. The lack of dependence on liquid nitrogen cooling would reduce the maintenance cost and forestall the possibility of detector warm-up and/or possible loss. With proper design and engineering, the mercuric-iodide detector could be incorporated into a paper-tape sampler that is interfaced to the continuous, automatic, isokinetic stack sampling system. This incorporation would result in a system requiring little oversight once it was installed and operational.

The capital cost of such a system to handle two stacks would be ~\$103,000. This system would be entirely self-contained and require little operator intervention once it was set up and calibrated. A yearly man hour charge for maintenance, calibration, etc., would be ~\$8000. Thus, the total cost would be \$111,000 for the first year. This is contrasted to the cost of a continuous stack sampler, isotopic source, IG detector, and MCA to handle two stacks at a cost of ~\$46,000, with an additional cost of \$23,400 per year to cover manpower required for the filter changing, filter preparation, filter analysis, and data reduction.

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An evaluation of current sampling and analysis methods used for monitoring yellowcake emissions from uranium mill exhausts was performed by Pacific Northwest Laboratory. The representativeness of once per quarter sampling was felt to be questionable. A more representative sample would be obtained by a continuous sampling system. The analysis could be performed by relatively newer instrumental methods. Direct-spectrometric and isotopically excited x-ray fluorescence instrumental analysis methods were evaluated. Because of a redirection in funding, the evaluation was not completed in terms of identifying instrumental interferences and field testing of the chosen methods. However, in light of readily available technology, a preferred method for sampling and analysis of yellowcake from uranium mill exhausts is proposed. This method would sample the exhaust stacks continuously using a continuous, automatic, isokinetic stack sampler with deposition of the exhaust gas particulates onto filter paper. The deposited particulates would then be analyzed by x-ray fluorescence using ⁵⁷Co as an excitation source. It is also recommended that a paper-tape sampler that houses an isotopic excitation source and detector be interfaced to a continuous stack sampler. This system would require evaluation and field testing after development.

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