Title Page

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Novel Rapid Oxidizing Fusion Method to Determine Polonium-210 in Air Filters

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Highlights

- Polonium-210 analytical method needed for air filter samples following a radiological emergency such as the detonation of a radiological dispersive device (RDD)
- Polonium-210 can be difficult to measure due to its volatility during sample digestion
- New fusion method utilizing alkali metal nitrate and sodium hydroxide digests organic filters and solid particulates while minimizing Po volatility.
- Employs rapid, rigorous digestion, preconcentration steps, and extraction chromatography
- Polonium-210 was measured using alpha spectrometry, after purification and microprecipitation

Abstract

A rapid method for the determination of polonium-210 in air filters has been developed for use following a radiological event and for routine environmental monitoring. Air filter samples, including cellulose and glass fiber, are fused with a sodium or potassium nitrate/sodium hydroxide flux material. The alkaline matrix minimizes polonium volatility during sample digestion steps, while efficiently dissolving the air filter and inorganic matter implanted on the filter. The addition of nitrate to the flux destroys organic material which can decrease Polonium recovery during precipitation, extraction and source preparation steps. Polonium is concentrated and separated from the sample matrix using an iron hydroxide precipitation and extraction chromatography with DGA

Resin. Polonium-210 is subsequently measured by alpha spectrometry following rapid microprecipitation with bismuth phosphate. The new method employs extraction chromatographic purification to remove alpha emitting interferences and sample matrix constituents which can interfere with ^{210}Po measurements using alpha spectrometry. The novel blend of sodium hydroxide and alkali metal nitrate digests the air filters quickly and effectively.

Introduction

Polonium-210 is a naturally-occurring alpha emitting radionuclide that is present in the environment as a progeny in the ^{238}U decay series. Anthropogenic sources of ^{210}Po include fossil fuel burning power plants, phosphate fertilizers, cement manufacturing and fallout from nuclear weapons testing. (UNSCEAR, 2000) Polonium-210 is also used in industrial applications, such as static elimination and neutron activation, (Centers for Disease Control and Prevention, 2019) and is included in a list compiled by the US EPA of threat radionuclides that might be utilized in a Radiological Dispersive Device (RDD). (Griggs, 2012).

Pb-210 has a half-life of 22.3 years, decaying to the shorter-lived ^{210}Po ($t_{1/2}$=138.376 days). The environmental transport of ^{210}Pb therefore largely controls the distribution of ^{210}Po in the environment. For the purposes of estimating radiation doses to the population, the global-average atmospheric concentration of ^{210}Pb and ^{210}Po in surface air are estimated to be 500 and 50 mBqm^{-3}, respectively (UNSCEAR, 2000).

From aqueous samples, ^{210}Po may be spontaneously auto-deposited onto silver or other metal disks, efficiently separating Po from most interfering nuclides and simultaneously preparing sources for alpha spectrometry. (Matthews et al., 2007) However, for more complex solid matrices, additional sample preparation is required. The high temperature acid digestions and dry ashing typically employed to treat many solid samples will result in polonium loss due to volatilization, unless performed in specially designed vessels, such as Kjeldahl flasks. References suggest that significant polonium loss due to volatilization can begin to occur with temperatures as low as 100°C, with chlorides, organic compounds and chelates considered the most volatile. (Martin, 1969; Marbuchi, 1963; Matthews et al., 2007)

In addition to Po loss via volatilization, incomplete destruction of organic material and the presence of particulate matter and certain redox active metals can adversely impact both the recovery of polonium analytes and equilibration with chemical yield tracers. Therefore, a rugged digestion method that completely dissolve air filters and embedded material, destroys organic material and minimizes Po loss to volatility is required for efficient polonium analyses.

The Savannah River Environmental Laboratory (SREL) has developed many rapid, rugged methods for the determination of radionuclides in solid matrices by using rapid sodium hydroxide fusion. These methods employ preconcentration steps from the fused sample matrix and fast sequential purification using extraction chromatography. Recently, a new alkaline fusion digestion approach for ^{210}Po was reported for solid urban matrices. (Maxwell et al., 2019) Coupled with extraction chromatography, this new rapid approach eliminates the chance of degraded alpha spectra due to sample matrix effects and offers high chemical yields due to the rugged sample preparation. This is not only important for analyses performed following a radiological event, but for routine environmental monitoring as well.

The analysis of air filters for ^{210}Po is important for environmental monitoring (Khaing and Thakur, 2017) The alkaline fusion approach was studied for application to air filter samples. While glass fiber samples can be fused with sodium hydroxide without difficulty, successful digestion of cellulose air filter requires additional treatment to destroy the organic material. Initial scoping tests with sodium hydroxide and hydrogen peroxide to oxidize the cellulose showed promise. However, the chemical yields observed (<60%) suggested the presence of some undigested organic material, which led to Po losses during the extraction chromatography and precipitation steps. Additionally, the reaction with 30% H_2O_2 produced vigorous bubbling which could lead to the loss of sample from the digestion vessel. Therefore, a mixture of either sodium nitrate or potassium nitrate with sodium hydroxide was employed to digest the cellulose filters, while maintaining an alkaline environment to reduce polonium volatilization loss and mitigate safety concerns associated with a nitrate only fusion.

Recently, analytical methods for Po have been reported that utilize rapid microprecipitation techniques such as copper sulfide (Guerin and Dai, 2013 and 2014), bismuth phosphate (Maxwell et al., 2013) or tellurium (Song et al., 2017) instead of autodeposition for final source preparation. Microprecipitation methods can offer improved yields over autodeposition in less time without heating, but require chemical separation of Po from potentially interfering alpha emitting nuclides.

In this study, an innovative new method to recover and measure ^{210}Po on air filters with high chemical yields and effective removal of sample matrix interferences was developed. Following rapid sample fusion of the air filters with sodium or potassium nitrate and sodium hydroxide (1:1), polonium was isolated quickly using an iron hydroxide precipitation and separation with DGA Resin as reported previously (Maxwell et al., 2019). A bismuth phosphate ($BiPO_4$) microprecipitation step performed directly on the DGA Resin column eluate is used to quickly prepare a source for counting using alpha spectrometry.

Experimental

Reagents

DGA Resin, Normal (N,N,N',N'-tetraoctyldiglycolamide) was obtained from Eichrom Technologies, LLC, (Lisle, Illinois, USA) and is also available from Triskem International (Bruz, France). Nitric, hydrochloric and hydrofluoric acid solutions were prepared from reagent-grade acids (Fisher Scientific, Inc.). All water was obtained from a Milli-Q2™ water purification system. All other materials were ACS (American Chemical Society) reagent grade. The radiochemical isotopes ^{210}Pb, ^{209}Po and ^{210}Po were obtained from Eckert-Ziegler Analytics, Inc. (Atlanta, GA, USA) and diluted to the activity levels used for spiking samples. Additional ^{210}Po was also obtained by the chromatographic separation from ^{210}Pb (McAlister, 2011). Copper planchets (1 inch diameter x 0.016 inch thickness) were obtained from A.F. Murphy Die and Machine Co. (North Quincy, MA).

Procedures

Sample Preparation. Figure 1 shows the sample preparation flowchart for polonium in air filter samples. In this study, 90 mm cellulose (Whatman 42 ashless, GE Healthcare, Chicago, IL, USA)) and 76mm glass fiber filters (Pall Corporation, Port Washington, NY, USA) were tested.

Air filters were added to 250 mL zirconium crucibles and spiked with ^{209}Po tracer and ^{210}Po. The filters were soaked with 5 mL of 8.0M NaOH to prevent loss of polonium upon heating (since Po tracers were added in acid solution). Sodium hydroxide pellets (10 g) and sodium or potassium nitrate (10 g) were added, and the crucibles covered with zirconium lids. The crucibles were placed in a furnace at 200 ˚C and the temperature was increased to 350˚C to dry the samples and begin the digestion process. The temperature was increased to 400˚C and held for 30 minutes, and then ramped to 500 ˚C, where the samples were fused for about 30 minutes. The crucibles were removed from the furnace and allowed to cool for 10 minutes in a laboratory hood. Temperatures and heating times may need to be adjusted to prevent excessive bubbling and loss of the sample from the crucible during the drying process.

A small amount of water was added to each sample to ensure that the crucibles had cooled enough to prevent excessive splattering from boiling water. Then, 15 mL of DI water and 2 mL of 30% H_2O_2 were added to each crucible. The crucibles were heated on a hotplate set to 175-225˚C until the bubbling from the H_2O_2 stopped. The addition of DI water/H_2O_2 and heating was repeated 1-2 times until the entire fusion cake was dissolved and a clear solution with no visible solid particles was obtained. Dissolved samples were transferred to 225 mL centrifuge tubes, rinsing the crucible with DI water. The crucibles were rinsed further with 10 mL 3M HNO_3, and the nitric acid rinse was then carefully added to the alkaline matrix in the 225 mL centrifuge tube.

Preconcentration of Polonium. The ^{210}Po was preconcentrated using iron (150 mg) added to each tube, as iron nitrate, to coprecipitate polonium on iron hydroxide as previously reported.. In addition, 50 mg of calcium (as calcium nitrate) was added to enhance the chemical recovery of the polonium during the iron hydroxide precipitation step. Samples were mixed thoroughly, cooled in an ice bath for 10 minutes, and centrifuged for 5-10 minutes.

Following removal of the supernate, the precipitate was dissolved with a mixture of 5 mL 4M HCl-0.2M HF plus 10 mL 0.5M HCl-0.02M HF. After transferring the dissolved precipitate to a 50 mL plastic centrifuge tube, the 225 mL centrifuge tube was rinsed twice with 5 mL 0.5M HCl-0.2M HF. The rinses were added to the 50 mL tube. Hydrofluoric acid was included in the sample load solution to prevent column clogging due to the formation of colloidal silicates and facilitate removal of Th during the column separation. By keeping the HCl concentration in the column load solution relatively low (<1.5M HCl), significant competition from Fe(III) was prevented during Po retention on DGA Resin. Samples were centrifuged to remove any residual solid particulates, and clear solutions were processed using an extraction chromatographic separation with DGA Resin (Maxwell et.al., 2019)

Column separation for Po. In this study, DGA Resin was used to rapidly separate Po prior to measurement by alpha spectrometry, as previously reported. (Maxwell et al., 2019). Samples were loaded at 1-2 mL/min on to 2 mL cartridges of DGA, Normal Resin which had been preconditioned with 10 mL of 1M HCl. After loading, the cartridges were rinsed sequentially with 10 mL 1M HCl-0.25M boric acid, 20 mL 0.25M HCl (U and rare earth removal), 20 mL 0.5M HCl-0.02M HF (Th removal), and 10 mL 8.0M HNO_3 (conversion to nitrate). An optional rinse of 20 mL 9M HCl may also be added prior to the 0.5MHCl-0.02M HF to remove Bi. The polonium was recovered with 20 mL of 0.05M HNO_3-0.005M oxalic acid.

Sources for alpha counting were prepared using bismuth phosphate microprecipitation by adding 125 μg Bi, 0.05 mL of 30% H_2O_2, 1-2 drops of phenolphthalein indicator solution and 0.75 mL of 3.2M $(NH_4)_2HPO_4$. The pH was adjusted by the addition of 14.5M NH_4OH to the first pale pink of the phenolphthalein indicator. Samples were mixed, set aside for 15 minutes, and the microprecipitate collected onto 25 mm, 0.1 μm pore size polypropylene filters (Eichrom Resolve® filters). Filters were washed with DI water and ethanol, attached to stainless steel planchets with 2-sided tape, and dried under a heat lamp prior to measurement by alpha spectrometry.

Apparatus

Polonium isotopic measurements were performed using alpha spectrometry. A Canberra Alpha Analyst Integrated Alpha Spectrometer was employed in this study. The alpha system utilized Passivated Implanted Planar Silicon (PIPS) detectors (450 mm^2 active surface) with counting efficiencies of approximately 30% and a 3 mm distance between the sample test source and the detector surface. Liquid scintillation counting using Ultima Gold LLT (Perkin Elmer) on a Perkin Elmer Tri-Carb 4910 TR was used to measure ^{210}Po in some method development experiments (Table 1).

Results and Discussion

Experiments were performed without air filters to demonstrate the low volatility of Po from the alkaline fusion matrix. The results of these experiments are presented in Table 1. From fusion temperatures of 450°C to 600°C, nearly quantitative recovery of ^{210}Po was observed throughout the entire sample preparation method.

Early experiments showed that digestion of the $NaOH/NaNO_3$ fusion cake with 30% H_2O_2 was important for cellulose filters. Addition of 15 mL aliquots of DI water and 2 mL of 30% H_2O_2 facilitates the breakup and dissolution of the cooled, solidified fusion cake and destroys traces of organic material remaining following the fusion. Digestion with 2-3 aliquots of the DI/H_2O_2 on a hotplate at 200-250°C until the bubbling from the H_2O_2 stopped, led consistently clear yellow (not cloudy) solutions upon dissolution of the $Fe(OH)_3$ precipitate in HCl-HF and high recoveries of Po (80-95%). Without the H_2O_2 digestion, some samples produced cloudy solutions upon dissolution of the $Fe(OH)_3$ with residual solids and low Po recoveries (10-50%).

Table 2 shows the results for the determination of ^{210}Po in twelve cellulose air filter samples spiked with 367 mBq of ^{210}Po. Po-209 tracer was added to correct the analytical results for chemical yield. The average ^{210}Po result was 366 mBq filter^{-1}, with a -0.3% bias relative to the ^{210}Po known value, with a SD (standard deviation) of 14.8 mBq filter^{-1}. The average tracer yield for ^{209}Po was 87.2% ± 4.9% (SD). The high ^{209}Po tracer recoveries and very good agreement of the ^{210}Po results with the known value confirm the effectiveness of the sample preparation protocol applied. The chemical yields indicate that the preconcentration steps, rapid extraction chromatography purification using DGA Resin, coupled with steps employed to minimize Po volatilization worked well. The uncertainties for the individual ^{210}Po results were typically ± 5-7% (1 SD), with a 12 hour count time. The count times may be shortened for the analysis of radiological emergency samples, depending on measurement quality objectives for the radiological event.

Table 3 shows the individual results for the determination of ^{210}Po in five cellulose air filter samples spiked with 87.8 mBq of ^{210}Po. The average ^{210}Po result was 84.6 mBq filter^{-1}, with a –3.7% bias and SD of 6.3 mBq filter^{-1}. The average tracer recovery for ^{209}Po was 92.3% ± 8.4% (SD). The uncertainties for the individual ^{210}Po results were typically ± 5-7% (1 SD). A 12 hour count time was utilized for these polonium measurements. The additional of 10 ml 14.5 sodium hydroxide was utilized to completely cover the filter and minimize polonium loss during the heating steps

Table 4 shows the individual results for the determination of ^{210}Po in five glass air filter samples spiked with 367 mBq of ^{210}Po. The average ^{210}Po result was 363.9 mBq filter^{-1}, with a -0.91% bias and SD of 11.8 mBq g^{-1}. The average tracer recovery for ^{209}Po was 102.7% ± 4.7% (SD). The uncertainties for the individual ^{210}Po results were typically ± 5-7% (1 SD). A 12 hour count time was employed for these alpha spectrometry measurements. The higher chemical yields for the glass fiber air filters compared to the cellulose filters indicates there may be possible traces or organic material remaining for the cellulose filters that interferes. This also seems to demonstrate that if the cellulose filter material can be destroyed effectively that very high chemical yields can be realized.

The results of direct autodeposition of ^{210}Po onto copper planchets from the dissolved $Fe(OH)_3$ precipitate are shown in Table 5. The copper planchets were prepared by washing with 70% ethanol, 1M HCl, and DI water and then snapped into the caps of 50 mL centrifuge tubes. After performing the procedure outlined in Figure 1 with cellulose filters, the dissolved $Fe(OH)_3$ solutions were adjusted with 2 mL of 1M ascorbic acid to reduce Fe(III) to Fe(II). The addition of ascorbic acid has been shown to improve resolution in the alpha spectra of autodeposited Po sources. The 50 mL centrifuge tubes were assembled with the caps containing the copper planchets. The sample solutions were added to the inverted tubes through a hole drilled into the bottom of the centrifuge tube. The tubes were mixed on an orbital plate shaker and heated over night at 60-65°C with a heat lamp. The solution was decanted and the planchets rinsed with DI water. After drying the planchets under a heat lamp, ^{210}Po was measured by alpha spectrometry. Recoveries of ^{210}Po averaged 64 $\underline{+}$ 7% with full width half maxium values of 38 $\underline{+}$ 6 keV, which was nearly identical for ^{210}Po spiked directly into 25 mL 0.1M HCl with 2 mL 1M ascorbic acid (65 ± 7%, 38 ± 4 keV). This demonstrates the feasibility of autodepositon for routine analyses with this method. The use of autodeposition eliminates the need for the chromatographic separation, but may increase the overall analysis time. The autodeposition analysis time may be reduced and Po recovery increased through the use of silver planchets or further optimization of the autodeposition conditions.

The approach of blending alkaline fusion with an oxidizing flux material to destroy the organic material while minimizing polonium volatilization was effective. In addition to effective destruction of the cellulose filter material, the oxidizing flux material and subsequent hydrogen peroxide treatment facilitates the digestion of any organic materials collected on the air filter along with soil/dust particulates. The use of sodium nitrate or potassium nitrate in concert with sodium hydroxide fusion, coupled with rapid preconcentration and DGA Resin separation provides high chemical yields, effective removal of interferences and good alpha peak resolution. Following a radiological incident, rapid, high quality results are required to allow incident commanders to accurately assess the environmental impact of an event and implement measures to protect the public.

The MDA (Minimum Detectable Activity) for the polonium isotopes measured using this rapid alpha spectrometry method was calculated according to equations prescribed by Currie (1968):

$$\mathrm{MDA} = [2.71+4.65\sqrt{B}\,]/\,(CT*R*V*Eff*A*0.060)$$

where B = Total Background counts, = BKG (rate) * sample count time; CT = sample count time (min); R = Chemical Recovery; V = Sample aliquot (g) ; EFF = Detector Efficiency ; A = Isotopic abundance (in most cases this will be ~1); 0.060 = conversion from dpm to mBq.

The MDA for the alpha spectrometry results can be modified as needed, depending on the measurement quality objectives, by the count time employed. The method MDA for Po-210 with a 12 hour count time is ~700 uBq filter^{-1}, assuming a detector efficiency of ~28%, 1 count background per 12 hours and a chemical recovery of approximately 90%. Any changes in detector background, counting efficiency and yields will affect the MDA accordingly. Samples that are counted for only 4 hours, for example, are estimated to have an MDA of ~1.4 mBq filter^{-1}.

This new method for ^{210}Po determination in air filter matrices can be performed rapidly. The spiking, fusion and preconcentration steps take about 90-120 minutes, the extraction chromatography separation also takes about 90 minutes, and the sample source preparation using bismuth phosphate microprecipitation requires about 30 minutes. The alpha counting times are set based on the desired measurement quality objectives (typically 60-1000 minutes).

Conclusions

A novel method for the rapid determination of ^{210}Po in cellulose and glass fiber air filter samples has been developed that allows for the rapid separation and measurement of ^{210}Po with very good chemical yields and effective removal of sample matrix interferences. The sodium hydroxide/alkali metal; nitrate fusion technique used to digest the air filter samples is rapid and rugged. The DGA Resin extraction chromatography method employed in this work performed well, removing potential alpha emitting interferences. The test results show that ^{210}Po can be determined quickly in air filters and reliably following a radiological event.

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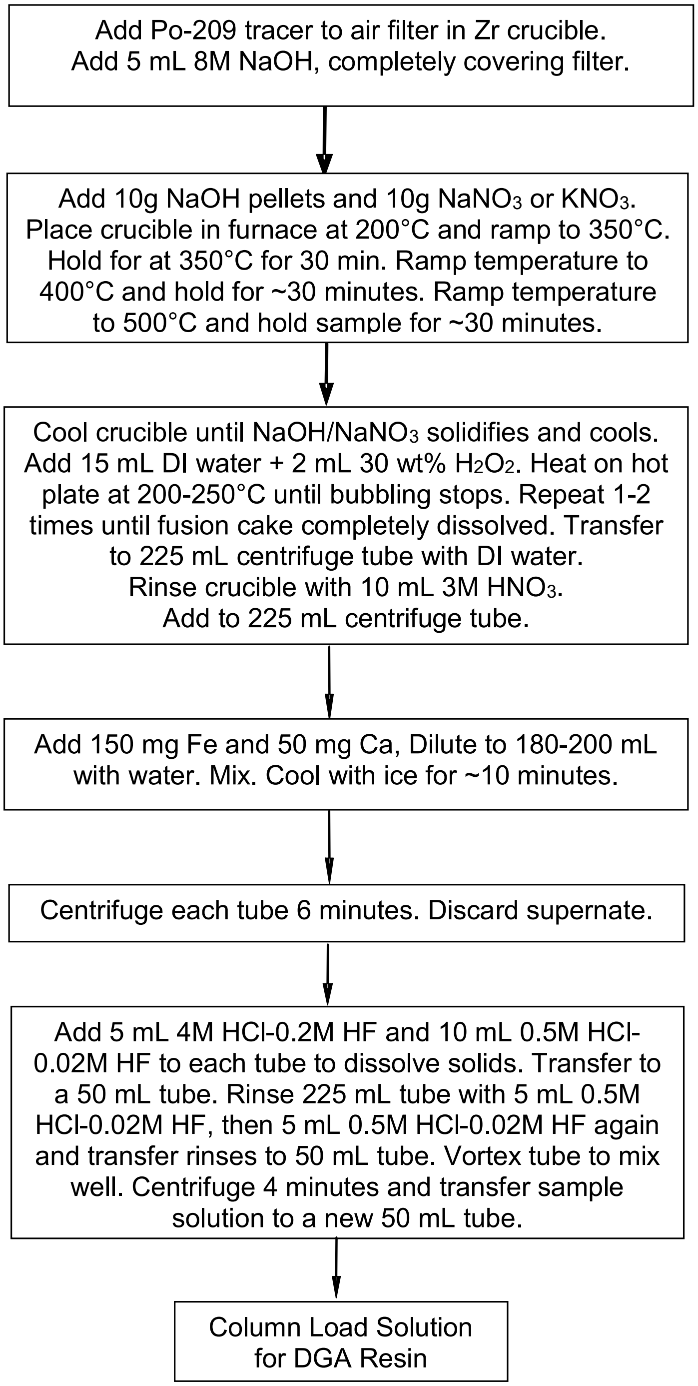
Wick, O. J., editor, 1980. Plutonium Handbook- A Guide to the Technology, Item ID: 250005|ISBN: 978-0-89448-023-2, 1st Edition|, 407-422.

Figure 1 Rapid Sample Preparation Method for ^{210}Po in Air Filters



Add Po-209 tracer to air filter in Zr crucible. Add 5 mL 8M NaOH, completely covering filter.

Add 10g NaOH pellets and 10g $NaNO_3$ or KNO_3. Place crucible in furnace at 200°C and ramp to 350°C. Hold for at 350°C for 30 min. Ramp temperature to 400°C and hold for ~30 minutes. Ramp temperature to 500°C and hold sample for ~30 minutes.

Cool crucible until NaOH/$NaNO_3$ solidifies and cools. Add 15 mL DI water + 2 mL 30 wt% H_2O_2. Heat on hot plate at 200-250°C until bubbling stops. Repeat 1-2 times until fusion cake completely dissolved. Transfer to 225 mL centrifuge tube with DI water. Rinse crucible with 10 mL 3M HNO_3. Add to 225 mL centrifuge tube.

Add 150 mg Fe and 50 mg Ca, Dilute to 180-200 mL with water. Mix. Cool with ice for ~10 minutes.

Centrifuge each tube 6 minutes. Discard supernate.

Add 5 mL 4M HCl-0.2M HF and 10 mL 0.5M HCl-0.02M HF to each tube to dissolve solids. Transfer to a 50 mL tube. Rinse 225 mL tube with 5 mL 0.5M HCl-0.02M HF, then 5 mL 0.5M HCl-0.02M HF again and transfer rinses to 50 mL tube. Vortex tube to mix well. Centrifuge 4 minutes and transfer sample solution to a new 50 mL tube.

Column Load Solution for DGA Resin

Table 1 Results for ^{210}Po (No Filters) vs. Fusion Temperature

Temperature (Celcius)	Replicates	^{210}Po Yield (%)	Range (%)
450	5	93 ± 2	91-95
500	5	97 ± 1	96-98
600	3	97 ± 4	94-102

86.2 Bq Po-210/sample

Table 2 Results for ^{210}Po Spiked into Cellulose Air Filters

Sample ID	^{209}Po Yield (%)	^{210}Po Reference Value (mBq filter^{-1})	^{210}Po Measured Value (mBq filter^{-1})	Difference (%)
1	88.4	367.2	359.2	-2.2
2	92.1	367.2	344.0	-6.3
3	82.9	367.2	374.4	2.0
4	90.7	367.2	382.9	4.3
6	93.3	367.2	356.5	-2.9
7	87.1	367.2	373.3	1.7
8	89.6	367.2	376.3	2.5
9	89.9	367.2	338.7	-7.8
10	79.8	367.2	367.5	0.1
11	87.6	367.2	381.8	4.0
12	78.3	367.2	373.3	1.7
Avg	87.2		366.2	-0.3
SD	4.9		14.8	4.0
% RSD	5.6			

Table 3 Results for ^{210}Po Spiked into Cellulose Air Filters (lower spike level)

Sample ID	^{209}Po Yield (%)	^{210}Po Reference Value (mBq filter^{-1})	^{210}Po Measured Value (mBq filter^{-1})	Difference (%)
1	92.8	87.8	80.1	-8.7
2	88.7	87.8	91.8	4.6
3	105.4	87.8	85.5	-2.6
4	92.2	87.8	87.3	-0.6
5	82.6	87.8	78.1	-11.0
Avg	92.3		84.6	-3.7
SD	8.4		5.5	6.3

Table 4 Results for ^{210}Po Spiked into Glass Fiber Air Filters

Sample ID	^{209}Po Yield (%)	^{210}Po Reference Value (mBq filter^{-1})	^{210}Po Measured Value (mBq filter^{-1})	Difference (%)
1	97.9	367.2	377.8	2.9
2	105.0	367.2	375.9	2.4
3	105.5	367.2	355.4	-3.2
4	97.6	367.2	356.0	-3.1
5	107.7	367.2	354.4	-3.5
Avg	102.7		363.9	-0.91
SD	4.7		11.8	3.23

Table 5 Results for ^{210}Po Spiked into Cellulose Air Filters (Autodeposition from dissolved Fe(OH)3 precipitate)

Sample ID	^{210}Po Recovery (%)	FWHM (keV)	Average Yield (%)	Average FWHM (keV)
Filter 1	66.9	45	64 ± 7	38 ± 6
Filter 2	74.5	43		
Filter 3	62.8	32		
Filter 4	57.3	38		
Filter 5	58.3	34		
Direct Spike 1	61.0	40	65 ± 7	38 ± 4
Direct Spike 2	60.6	38		
Direct Spike 3	63.9	40		
Direct Spike 4	77.4	31		
Direct Spike 5	60.4	40		

83.3 Bq Po-210/Sample, 10 minute count time