Radium

Environmental Measurements Laboratory
U.S. Department of Energy

Ra-01-RC

RADIUM-226 IN BONE ASH

Contact Person(s) : Isabel M. Fisenne

APPLICATION

This procedure is applicable to bone ash only and has been adapted from Hallden et al. (1963).

The separated sample is measured by radon emanation. Only ^{226}Ra yields a radon progeny with suitable characteristics, so the method is specific. Most of the calcium, the alkali metals and phosphates are separated by coprecipitation of the radium as sulfate with barium carrier in a buffered solution. Further purification is obtained by repeating this step in the presence of undissolved $BaSO_4$. The sulfate precipitate is dissolved in alkaline EDTA to prepare the emanating solution. The chemical yield of barium is determined with the γ-emitting tracer ^{133}Ba.

SPECIAL APPARATUS

Radon bubblers - see Specification 7.7.

SPECIAL REAGENTS

1. Barium-133 tracer solution - about 50 counts sec $^{-1}$ per 0.1-g aliquot, prepared in 1:99 HCl.
2. Barium carrier solution (20 mg mL^{-1}) - 30.4 g $BaCl_2$ L^{-1} of 1:99 HCl.
3. Ammonium acetate solution - 15 g NH_4 $C_2H_3O_2$ (NH_4OAc) L^{-1} of water.
4. Acetic acid solution - 20 mL glacial CH_3 CO_2H (HOAC)L^{-1} of water.

5. Ammonium sulfate solution - 100 g $(NH_4)_2SO_4$ L^{-1} of water.

6. Aerosol OT solution - 0.1%.

7. EDTA solution - 300 g tetrasodium salt of EDTA L^{-1} of water.

8. EDTA wash solution - 1:9 dilution of EDTA solution.

9. Monoethanolamine - 1:1 (v/v) with water.

DETERMINATION

1. Weigh 10 g of bone ash into a 90-mL glass or 100-mL polycarbonate centrifuge tube. Add a weighed aliquot (about 0.1 g) of ^{133}Ba tracer solution, and 1 mL of barium carrier solution.

2. Add 20 mL of HCl (slowly at first to prevent foaming). Stir and warm in an 85°C water bath for about 5 min.

3. Add 10 mL of water. Add NH_4OH until a dense white permanent hydroxide flock forms (about 8-10 mL).

4. Dissolve the flock in HCl (about 4 mL is required).

5. Add 2 mL of NH_4OAc solution and 1 mL of HOAc solution. Cool in a water bath to room temperature.

6. Add 1 mL of $(NH_4)_2SO_4$ solution. Stir and let stand for 0.5 h.

7. Centrifuge at 2000 rpm for 1 h. Add one drop of 0.1% Aerosol OT. Decant carefully and discard the supernate.

8. Add 5 mL of HCl and warm in an 85°C water bath for 5 min. (Most of the $BaSO_4$ remains as a precipitate.)

9. Add 10 mL of water and then add NH_4OH until a permanent flock forms.

10. Dissolve the flock in HCl.

11. Add 2 mL of NH_4OAc solution and 1 mL of HOAc solution. Cool in a water bath to room temperature.

12. Add 1 mL of $(NH_4)_2SO_4$ solution. Stir and let stand for 0.5 h.

13. Centrifuge at 2000 rpm for 1 h. Add one drop of 0.1% Aerosol OT. Decant carefully and discard the supernate.

14. Heat a solution of EDTA (300 g L^{-1}) and an EDTA wash solution (30 g L^{-1}) in an 85°C water bath.

15. Break up the $BaSO_4$ precipitate with a stirring rod. Add 1 mL of 1:1 monoethanolamine and 5 mL of the hot EDTA solution and stir. Let stand for 5 min.

16. Wash down the sides of the tube with about 10 mL of the hot EDTA wash solution. Let the tube remain in the water bath for 15 min, stirring occasionally.

17. Gravity filter the hot solution through a 12.5 cm Whatman No. 41 filter paper into a 30 mL polyethylene bottle.

18. Wash the centrifuge tube and the filter paper with hot EDTA wash solution. Discard the paper and residue.

19. Dilute the sample to the same liquid level as the ^{133}Ba standard. The standard prepared by diluting a known aliquot (about 0.1 g) of ^{133}Ba solution to 25 mL in a 30 mL polyethylene bottle. Gamma count on a flat crystal to determine the chemical yield of barium.

20. Transfer the sample solution to a ^{222}Rn bubbler.

21. De-emanate ^{222}Rn by bubbling with forming gas for about 10 min at 100 cm^3 min^{-1} as described in Radium - Emanation Procedure (see Ra-02-RC). Record the time as the starting time for ^{222}Rn buildup. Continue the analysis using the emanation technique.

LOWER LIMIT OF DETECTION (LLD)

Counter Efficiency	(%)	57.5
Counter Background	(cps)	0.0028
Yield	(%)	90
Blank	(cps)	0.0012
LLD (400 min)	(mBq)	3.3
LLD (1000 min)	(mBq)	1.7

*Reagent blanks must be analyzed with each set of samples.

REFERENCE

Hallden, N. A., I. M. Fisenne and J. H. Harley
"The Determination of Radium-226 in Human Bone"
Talanta, 10, 1223-1227 (1963)

Ra-02-RC

RADIUM-226 - EMANATION PROCEDURE

Contact Person(s) : Isabel M. Fisenne

APPLICATION

This procedure is specific and may be applied to almost any matrix which can be converted to a homogeneous solution.

Radium-226 in solution can be determined by de-emanating its ^{222}Rn into an ionization chamber or scintillation cell for measurement. The ^{222}Rn can be de-emanated by bubbling an inert gas through the solution, either after equilibrium has been established or after any known time period. Two half-lives, 7.65 days for example, give 75% of the maximum buildup.

SPECIAL APPARATUS

1. Radon measuring equipment are described in Radon-222 in Air and Breath Samples, Procedure Rn-01-RC, and in Fisenne and Keller (1985).
2. Radon bubblers - see Specification 7.7.

SAMPLE PREPARATION

1. The sample with barium carrier should be in homogeneous solution in a volume of 15-25 mL. The solution should be acidic with perchloric acid, neutral or in basic EDTA solution. Hydrochloric acid, NH_3 or other volatile materials must be absent.
2. Detailed preparation procedures for separation of radium from sample matrices are presented in other radium procedures in this Manual.

3. Almost all types of samples require simultaneous analysis of reagent blanks, since radium appears in many chemical compounds. Using distilled or organic reagents reduces possible contamination.

DETERMINATION

1. Transfer the sample solution to a ^{222}Rn bubbler.
2. Clamp the bubbler in place to the ^{222}Rn system.
3. Open the forming gas (85% N_2, 15% H_2) tank valve and adjust the line pressure to 70 kPa (10 psi) gauge.
4. Bleed the gas line, the rotometer, and 9.5 mm diameter rubber tubing.
5. Adjust the flow through the rotometer to a rate of 20 cm^3 min^{-1}.
6. Open both stopcocks on the ^{222}Rn bubbler.
7. Attach the rubber tubing on the exit of the rotometer to the inlet of the ^{222}Rn bubbler. Adjust the forming gas flow rate through the bubbler to 100 cm^3 min^{-1}.
8. Flush the ^{222}Rn from the solution (de-emanate) for 10 min.
9. Close both stopcocks simultaneously. Record the date and time as the beginning of the ^{222}Rn build-up period.
10. Shut off the forming gas and remove the rubber tubing from the bubbler.
11. Place the ^{222}Rn bubbler in a refrigerator for a suitable build-up period, usually 1 week.
12. To emanate ^{222}Rn into a pulse ionization chamber, repeat Steps 2-4.
13. Attach the rubber tubing from the rotometer to the inlet of the measurement system.

14. Open the valve to the ionization chamber and close the vacuum valves. Partially fill the chamber with forming gas to a pressure of -2.7 (-50 cm of Hg) gauge. Close the chamber valve and system entry valve. Open the vacuum valves and detach the rubber tubing from the measurement system.

15. Attach a small drying tube containing about 5 g of a color indicating drying agent and 4 cm lengths of 9.5 mm diameter rubber tubing at both ends to the outlet of the bubbler and the inlet of the measurement system.

16. Open the system entry valve to evacuate the drying tube for 3 min.

17. Adjust the forming gas flow rate to 20 cm^3 min^{-1} and attach the tubing outlet of the rotometer to the inlet of the bubbler.

18. Close the vacuum valves and open the ionization chamber valve.

19. Open the outlet stopcock of the bubbler and allow the bubbling to subside. Record the date and time as the end of the ^{222}Rn build-up period.

20. Carefully open the bubbler inlet stopcock and adjust the forming gas flow through the bubbler to about 100 cm^3 min^{-1}.

21. When the system gauge indicates that atmospheric pressure has been reached, close both bubbler stopcocks simultaneously. Close the chamber, system entry and forming gas valves. Open the vacuum valves.

22. Remove the bubbler and drying tube from the system. Record the date and time of the end of the emanation as the beginning of the next ^{222}Rn build-up period.

23. Turn on the ionization chamber's high voltage. A red LED will light when the chamber is at or above atmospheric pressure.

24. Measure the sample for at least 17 h.

Notes for Proper Operation:

1. If the Teflon stopcocks of the bubbler are snug fitting, it is not necessary to grease them. If they do leak, however, a silicone stopcock grease may be used.

2. The glass-to-glass ground joint at the top of the bubbler should also be lightly greased with silicone.

3. During the de-emanation, the gas bubbling should be started slowly to prevent popping of the bubbler top or stopcocks.

4. A better transfer of ^{222}Rn from acid or neutral solutions is obtained if the bubbler is first chilled in a refrigerator. This apparently reduces the bubble size and improves the transfer efficiency. For samples dissolved in alkaline EDTA solution, a drop of octyl alcohol is added to reduce foaming.

STANDARDIZATION

The chambers are standardized by de-emanating aliquots of a National Institute of Standards and Technology SRM ^{226}Ra solution contained in ^{222}Rn bubblers.

DATA PROCESSING AND ANALYSES

1. For routine work, discard the first 5 h of counting data (equilibrium for ^{222}Rn and progeny).

2. Determine the total count over the remaining counting period.

3. Calculate the net counts per hour per sample by determining the total sample counts per hour and subtracting the background counts per hour.

4. Divide the net counts per hour by the chamber standardization value of net counts per hour per Bq of ^{222}Rn.

5. Using the midpoint of the counting interval as the time of counting, extrapolate the value obtained to the time noted for the end of ^{222}Rn buildup. Figure 1 may be used for this calculation. This correction is only a few percent. The decay during the counting period is small and is not corrected for.

6. Correct the ^{222}Rn measurement at the end of buildup for the fractional buildup. The figure in Procedure Rn-01-RC may be used to obtain the build-up factor.

7. Subtract the value obtained by carrying a blank determination through the full procedure.

8. Samples are measured twice and the mean and standard deviation of the duplicate emanation results are reported.

9. A computational data sheet is attached for handling routine calculations on a step-by-step basis.

LOWER LIMIT OF DETECTION (LLD)*

Counter Efficiency	(%)	57.5
Counter Background	(cps)	0.0028
Yield	(%)	90
Blank	(cps)	0.0012
LLD (400 min)	(mBq)	3.3
LLD (1000 min)	(mBq)	1.7

*Reagent blank must be analyzed with each set of samples.

REFERENCE

Fisenne, I. M. and H. W. Keller
"The EML Pulse Ionization Chamber Systems for the Measurement of Radon-222"
USDOE Report EML-437, March (1985)

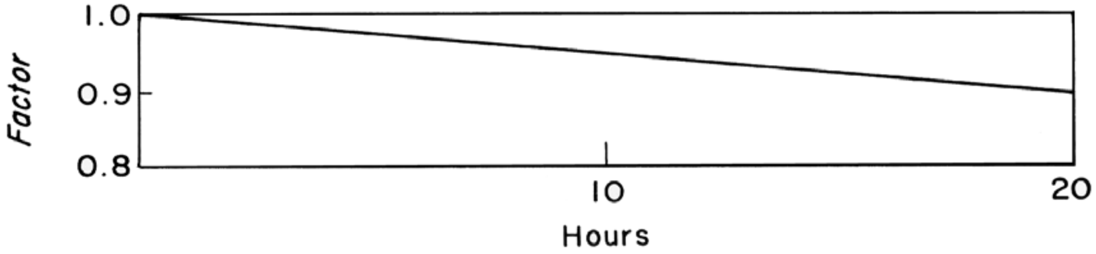

Figure 1. Correction factor from midpoint to the counting interval to the start of radon measurement interval.

Date _________________ Sample Number

		Operation						
1	Chamber number	-						
	BACKGROUND							
2	Total count	-						
3	Count interval (h)	-						
4	Background (cph)	2/3						
	STANDARDIZATION							
5	Chamber factor cph Bq^{-1}	-						
	SAMPLE COUNT							
6	Total count	-						
7	Count interval (h)	-						
8	Sample at count time (cph)	6/7						
	CORRECTIONS							
9	Time for midpoint of count	-						
10	Time for end of buildup	-						
11	Difference (days)	9-10						
12	Factor (from graph)	-						
13	Sample at end of buildup	8/12						

Date _________________ Sample Number

		Operation						
14	Time for start of buildup	-						
15	Time for end of buildup	10						
16	Difference	15-14						
17	Factor (from graph)	-						
18	Sample with full buildup	13/17						
19	Bq in sample	18/5						
20	Bq in blank[*]							
21	Net Bq in sample	19-20						

*This sheet is suitable for calculating the blank value.

Ra-03-RC

RADIUM-226 IN SOIL, VEGETATION ASH, AND ION EXCHANGE RESIN

Contact Person(s) : Isabel M. Fisenne

APPLICATION

This procedure is applicable to 5 g samples of soil and 10 g of vegetation ash or to ion exchange resin from sampling columns.

Soil, vegetation ash or ion exchange resin are prepared for ^{222}Rn emanation measurement. The sample is pretreated with HNO_3-HF, fused with KF and transposed to pyrosulfate (Sill, 1961). The cake is dissolved in dilute HCl. $RaBaSO_4$ is precipitated, filtered, and dissolved in alkaline EDTA. The chemical yield is determined with the γ-emitting tracer ^{133}Ba.

SPECIAL APPARATUS

1. Radon bubblers - see Specification 7.7.
2. 100 mL platinum dishes or 250 mL platinum crucibles.
3. Millipore filter setup - 47 mm diameter.
4. Millipore filters - 47 mm diameter, 0.45 µm pore size.

SPECIAL REAGENTS

1. Barium-133 tracer solution - about 50 cps per 0.1-g aliquot, prepared in 1:99 HCl.

2. Barium carrier solution (20 mg mL^{-1}) - 30.4 g $BaCl_2$ L^{-1} in 1:99 HCl.

3. EDTA solution - 300 g tetrasodium salt of EDTA L^{-1} of water.

4. Triethanolamine - 1:1 in water.

SAMPLE PREPARATION

A. Soil and vegetation.

1. Weigh 5 g of soil or 10 g of vegetation ash into a 100-mL platinum dish. Add a weighed aliquot (about 0.1 g) of ^{133}Ba tracer solution and 1 mL of Ba carrier solution.

2. Slowly add 10 mL of HNO_3 and 10 mL of HF to the sample and evaporate on a hot plate to near dryness.

3. Continue the analysis as described below.

B. Ion exchange resin.

1. Transfer the resin and paper pulp from the collection column to a 250-mL platinum crucible. Dry under a heat lamp and ash at 500°C in a muffle furnace for 48 h.

2. To the cooled crucible, add a weighed aliquot (about 0.1 g) of ^{133}Ba tracer solution and 1 mL of barium carrier solution.

3. Continue the analysis as described below.

DETERMINATION

1. Weigh out 15 g of KF and add to the sample. Press the KF into the sample with a plastic spatula.

   Caution - wear rubber gloves and safety glasses during Steps 2-5.

2. Fuse the sample over an air fed Meker burner, gradually increasing the temperature until a clear melt is obtained. Cool the melt.

3. Using a burette, slowly add 17.5 mL of H_2SO_4 to the melt. Heat the dish on a hot plate until a clear melt accompanied by dense fumes is obtained. Cool the melt.

4. Weigh out 10 g of Na_2SO_4, add to the dish and fuse over the Meker burner until a clear melt accompanied by dense fumes is obtained. Cool the melt.

5. Transfer the cake to a 600 mL beaker containing 350 mL of hot water and 25 mL of HCl. Stir the solution to dissolve the cake. Cool for 1 h.

6. Filter the precipitate onto a 47 mm diameter, 0.45 μm pore size Millipore filter into a 1 L sidearm flask, police the beaker, and wash with water. Add the washing to the funnel. Discard the filtrate.

7. Using a strong stream of water from a wash bottle, transfer the precipitate from the filter into a 150 mL beaker. Discard the filter.

8. Add 5 mL of EDTA solution and 1 mL of 1:1 triethanolamine to the beaker. Heat on a warm hot plate for about 15 min, stirring occasionally. Reduce the sample volume to ~15 mL.

9. Gravity filter the warm solution through a Whatman No. 41 filter paper into a 30-mL polyethylene bottle.

10. Wash the beaker dish and filter with hot water. Discard the filter.

11. Dilute the sample to the same liquid level as a known aliquot (about 0.1 g) of the ^{133}Ba tracer solution and dilute to 25 mL in a 30 mL polyethylene bottle.

12. Gamma count the samples and standard to determine the chemical yield of barium.

13. Transfer the sample to a ^{222}Rn bubbler with water.

14. De-emanate ^{222}Rn by bubbling with forming gas for about 10 min at 100 mL min^{-1} as described in ^{226}Radium - Emanation Procedure (see Ra-03-RC). Record the time as the starting time for ^{222}Rn buildup. Continue the analysis by the emanation technique.

LOWER LIMIT OF DETECTION (LLD)*

Counter Efficiency	(%)	57.5
Counter Background	(cps)	0.0028
Yield	(%)	90
Blank	(cps)	0.0020
LLD (400 min)	(mBq)	3.3
LLD (1000 min)	(mBq)	1.7

*Reagent blanks must be analyzed with each set of samples.

REFERENCE

Sill, C. W.
"Decomposition of Refractory Silicates in Ultramicro Analysis"
Anal. Chem., 33, 1684 (1961)

*EPA APPROVED[*]

Ra-04-RC

RADIUM-226 IN TAP WATER, URINE, AND FECES

Contact Person(s) : Isabel M. Fisenne

APPLICATION

The procedure presented has been applied to tap water, ashed urine, and feces samples. Only ^{226}Ra yields ^{222}Rn progeny that has suitable characteristics for detection by an emanation technique; therefore, the procedure is specific.

After sample preparation, radium is isolated from most other elements by coprecipitation with barium sulfate. Further purification is obtained by the removal of silica with HF and reprecipitation of the sulfate. The sulfate precipitate is dissolved in alkaline EDTA to prepare the emanating solution. The chemical yield of barium is determined with the γ-emitting tracer ^{133}Ba.

SPECIAL APPARATUS

Radon bubblers - see Specification 7.7.

SPECIAL REAGENT

1. Barium carrier solution - 20 mg mL^{-1} - 30.4 g $BaC1_2$ L^{-1} in 1:99 HCl.

2. Ammonium sulfate solution - 100 g $(NH_4)_2SO_4$ L^{-1} in water.

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[*] *Environmental Protection Agency - Guidelines Establishing Test Procedures for the Analysis of Pollutants, Under the Clean Water Act; National Primary Water Regulations and National Secondary Drinking Water Regulations; Methods Update, tentatively slated for approval, 66FR3466-3497, January 16, 2001.*

3. Aerosol OT solution - 0.1%.

4. EDTA solution - 300 g tetrasodium salt of EDTA L^{-1} in water.

5. EDTA wash solution - 1:9 dilution of EDTA solution.

6. Ammonium acetate solution - 15 g L^{-1} in water.

7. Acetic acid solution - 20 mL glacial acetic L^{-1} in water.

8. Triethanolamine - 1:1 in water.

SAMPLE PREPARATION

A. Tap water.

1. Transfer two 0.5-L of tap water to a 3 L beaker.

2. Add 25 mL of HCl and 1 mL of barium carrier solution. Add a weighed aliquot (about 0.1 g) of the ^{133}Ba tracer solution.

3. Evaporate and add an additional two 0.5-L aliquots of tap water until a 10-L collection has been obtained. Evaporate gently to about 100 mL.

4. Transfer to a 400-mL beaker with water, policing the sides of the 3-L beaker thoroughly. Evaporate gently to about 100-mL.

5. Adjust the pH to 4 with 1:1 NH_4OH and proceed with **Determination**.

B. Urine.

1. Weigh an aliquot of ^{133}Ba tracer solution (about 0.1 g) into a 2-L beaker containing a small amount of water and 1 mL of barium carrier solution.

2. Add 1500 mL of urine, then 100 mL of HNO_3 and evaporate to about 1-L.

3. Slowly add 100-mL of HNO_3.

4. Repeat the evaporation and addition of acid until a total of 500-mL of HNO_3 has been added.

5. Evaporate to about 20 mL. Transfer to a 400-mL beaker with water.

6. Adjust the pH to 4 with 1:1 NH_4OH and proceed with Determination.

C. Feces ash.

1. Weigh 1 g of fecal ash into a 150-mL beaker. Add a weighed aliquot (about 0.1 g) of ^{133}Ba tracer solution and 1 mL of barium carrier solution.

2. Cover the ash with a small quantity of water and slowly add 10 mL of HNO_3. Evaporate to a small volume on a medium hot plate.

3. Add about 25 mL of water. Add NH_4OH until a permanent hydroxide flock forms (3-5 mL).

4. Dissolve the flock in a few drops of HNO_3.

5. Add 2 mL of NH_4Ac solution and 1 mL of acetic acid solution. Dilute to about 100-mL and proceed with **Determination**.

DETERMINATION

1. Add 1 mL of $(NH_4)_2SO_4$ solution and allow the sample to digest for 1 h at room temperature.

2. Filter by gravity on a 9 cm Whatman No. 42 filter paper. Wash the paper thoroughly with water. Discard the filtrate and washings.

3. Transfer the paper to a platinum dish. Dry the paper and then ash at 500°C for about 1 h.

4. Add 1 mL of H_2SO_4 and 2 mL of HF. Evaporate to SO_3 fumes.

5. Cool and transfer to a 90 mL centrifuge tube with water.

6. Police the dish and add the washings to the centrifuge tube.

7. Stir and let stand for 0.5 h.

8. Centrifuge at 2000 rpm for 1 h. Add one drop of 0.1% Aerosol OT. Decant carefully and discard the supernate.

9. Heat a solution of EDTA (300 g L^{-1} EDTA) in an 85°C water bath.

10. Break up the $BaSO_4$ precipitate with a stirring rod.

11. Add 1 mL of 1:1 triethanolamine and 5 mL of the hot EDTA solution, and stir. Wash down the sides of the tube with water.

12. Digest in the steam bath for 15 min, stirring occasionally.

13. Transfer the solution to a 30-mL polyethylene bottle.

14. Dilute the sample to the same liquid level as a known aliquot (about 0.1 g) of ^{133}Ba solution diluted to 25 mL in a 30 mL polyethylene bottle.

15. Gamma count samples and standard on a flat crystal to determine the chemical yield of barium.

16. Transfer the sample solution to a radon bubbler.

17. De-emanate radon by bubbling with forming gas for about 10 min at 100 mL^{-1} min as described in Procedure Ra-03-RC. Record the time as the starting time for the radon build-up period. Continue the analysis by the emanation technique.

LOWER LIMIT OF DETECTION (LLD)*

		A	B	C
Counter Efficiency	(%)	57.5	57.5	57.5
Counter Background	(cps)	0.0028	0.0028	0.0028
Yield	(%)	90	80	85
Blank	(cps)	0.0012	0.0020	0.0012
LLD (400 min)	(mBq)	0.33	0.45	0.39
LLD (1000 min)	(mBq)	0.17	0.29	0.15

*Reagent blanks must be analyzed with each set of samples.

Pulse ionization chamber:
A = Tap water
B = Urine
C = Feces

Ra-05-RC

RADIUM-224 IN URINE

(*see Volume II*)

Ra-06-RC

RADIUM-226 IN URINE AND WATER

Contact Person(s) : Isabel M. Fisenne

APPLICATION

This procedure is applicable to samples of urine and water. Small amounts of thorium are carried by the $BaSO_4$ and the method fails in the presence of ^{230}Th. Natural thorium does not interfere. Other α-emitting isotopes of radium also interfere.

Radium is initially separated from untreated water or urine by coprecipitation with calcium phosphate. Calcium and most other elements are removed by coprecipitation of radium on barium sulfate. Organic material is removed by ignition, silica is removed by volatilization with HF, and the radium-barium sulfate is reprecipitated.

Radium-226 in the barium sulfate precipitate is stored to allow the buildup of ^{222}Rn, ^{218}Po, and ^{214}Po. The equilibrated α activities are then measured with a scintillation counter. The instrument response is converted to Bq by application of corrections for counter background and efficiency and for self-absorption. Chemical recovery is measured gravimetrically and the ^{226}Ra activity is equivalent to one-fourth of the total Bq measured.

SPECIAL APPARATUS

1. Rings and discs - see Specification 7.2.
2. Teflon filter holders and filter funnels and sample mounts - see Specification 7.12.
3. Mylar film - see Specification 7.10.

SPECIAL REAGENTS

1. Barium carrier solution: 5 mg Ba mL^{-1} - 9.51 g $Ba(NO_3)_2$ L^{-1} of 1:99 HCl.

SAMPLE PREPARATION

A. Water.

1. Transfer 4 L of the sample to a 5 L stainless steel pot.

2. Add 1 mL of barium carrier solution and 4 mL of HNO_3.

3. Evaporate to near dryness, add additional aliquots of water and HNO_3, and continue until 50 L of sample have been treated.

4. Evaporate to about 100 mL, add 3-4 g of NH_4Ac, and adjust the pH to 5 with 1:1 NH_4OH.

B. Urine.

1. Transfer a measured volume of urine to a 250-mL centrifuge bottle and add 1 mL of barium carrier solution.

2. Add 2 mL of H_3PO_4. Adjust the pH to 9 with 1:1 NH_4OH. Stir, centrifuge, and discard the supernate.

3. Dissolve the precipitate with 10 mL of HNO_3. Dilute to about 100 mL with water.

4. Add 1 mL of H_3PO_4. Adjust the pH to 9 with 1:1 NH_4OH. Stir, centrifuge, and discard the supernate.

5. Dissolve the precipitate in a few mL of acetic acid and dilute to 100 mL with water. Adjust the pH to 5 with 1:1 NH_4OH.

DETERMINATION

1. Add 3-4 g of $(NH_4)_2SO_4$ with stirring. Digest for 12 h. Cool and filter by gravity on a 9 cm Whatman No. 42 filter paper. Wash with 0.5% H_2SO_4. Discard the filtrate and washings.

2. Transfer the precipitate and filter to a platinum dish. Dry and ignite at 900°C.

3. Cool and add 2 mL of 1:1 H_2SO_4 and 5 mL of HF. Evaporate on a sand bath until SO_3 fumes appear.

4. Transfer the solution to a 40 mL, heavy walled centrifuge tube with 30 mL of H_2O and digest for 12 h. Centrifuge and discard the supernate.

5. Wash the precipitate with 10 mL of water. Centrifuge and discard the washings.

6. Slurry the precipitate with water and filter on a tared Whatman No. 42 filter paper.

7. Dry in an oven at about 150°C for at least 2 h.

8. Cool, weigh, and mount on a ring and disc with an α phosphor and Mylar.

9. Store for 30 days and count on an α-scintillation counter.

DATA PROCESSING AND ANALYSES

The ^{226}Ra disintegration rate is obtained from the counting rate of the equilibrated radium fraction through the following calculation:

$$\text{Bq} = R_s\ YTCE$$

where R_s is the net counting rate of the sample, Y is the recovery factor, T is the self-absorption correction, E is the counter efficiency factor, and C is a theoretical conversion from total α activity to ^{226}Ra α activity.

Chemical recovery, Y, is obtained by weighing the final Ba sulfate precipitate. The calculation of the recovery factor is:

$$Y = \frac{x}{(w - t)\, f}$$

where x is the weight of barium added as carrier, w is the total weight of the final precipitate and filter paper, t is the weight of the filter paper, and f is the gravimetric factor which equals 0.588 mg barium per mg of barium sulfate.

Self-absorption correction, T, is used to normalize the self-absorption of the ^{226}Ra, ^{222}Rn, ^{218}Po, and ^{214}Po α activities in samples and standards to a common thickness (mg cm^{-2}). T is obtained by counting representative α emitters over the energy range of 4.8-8.8 MeV in precipitates of varying sample thicknesses. The correction is taken as the ratio of counting rates at an arbitrary minimum thickness to other thicknesses over the range of probable sample recoveries. Figure 1 is a typical composite plot of the correction obtained for ^{230}Th, ^{212}Pb, and ^{226}Ra activities for varying thicknesses of their oxalate, chromate, and sulfate derivatives, respectively.

Theoretical activity conversion factor, C, equals 0.25 after 30 days of buildup, when the three α-emitting progeny are in secular equilibrium with ^{226}Ra. However, the ratio of ^{226}Ra to total α activity at any time after the separation of radium may be calculated from the Bateman equation for buildup of ^{222}Rn.

Efficiency factor, E, is determined by α counting a known quantity of ^{226}Ra under sample conditions. The correction is taken as the ratio of the theoretical activity to the observed counting rate. The calculation is expressed as:

$$E = \frac{A}{R_S' T' C'}$$

where A is the theoretical activity of the standard, and R_S' is the net counting rate of the standard, T' is the self-absorption correction for the standard, and C' is the theoretical ratio of ^{226}Ra to total α activity in the standard.

An accompanying computational data sheet illustrates a procedure for routine calculation of ^{226}Ra activity rates. Experimental data are tabulated as they are derived and the calculations are performed on a step-by-step basis.

LOWER LIMIT OF DETECTION (LLD)*

Counter efficiency	(%)	50
Counter background	(cps)	1.67×10^{-5}
Yield	(%)	85
Blank	(cps)	1.67×10^{3}
LLD (400 min)	(mBq)	3
LLD (1000 min)	(mBq)	2

*Reagent blank must be analyzed with each set of samples.

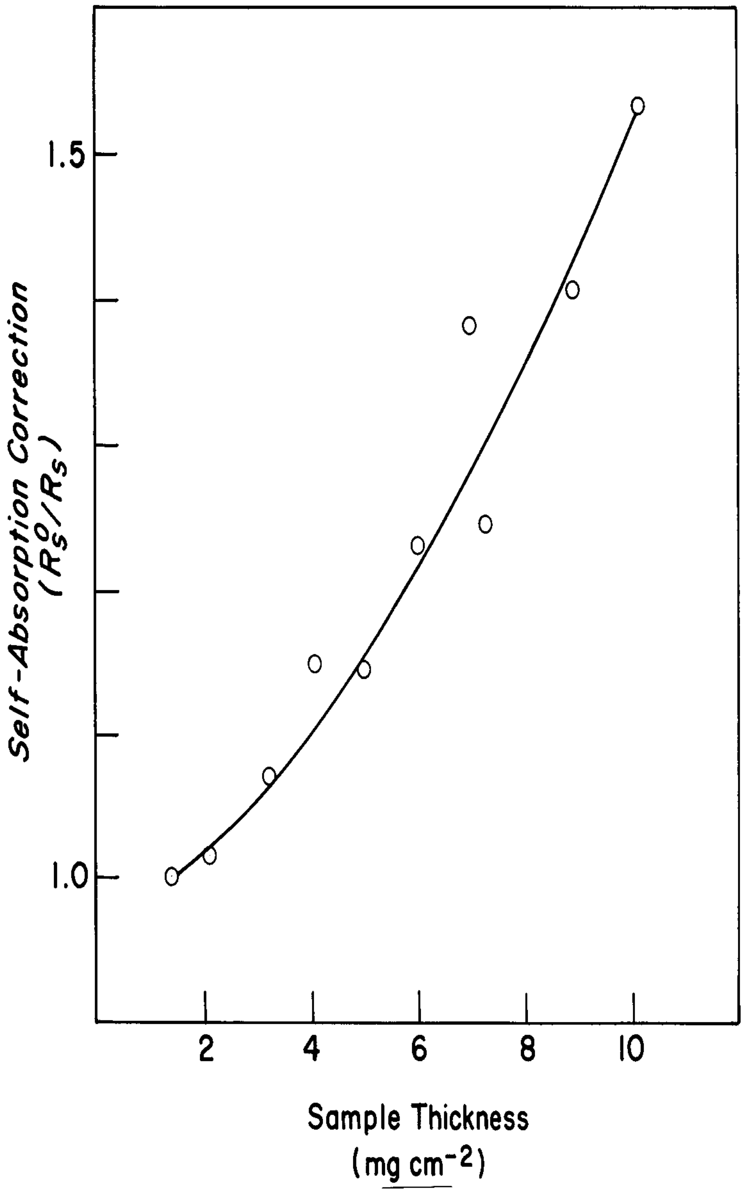

Figure 1. Self-absorption for α activities (4.8-8.9 MeV).

Date _________________ Sample Number

		Operation						
1	Counter number	-						
	STANDARD COUNTING							
2	Gross background count	-						
3	Count interval (s)	-						
4	Background (cps)	2/3						
5	Gross standard count	-						
6	Count time	-						
7	Standard (cps)	5/6						
8	Standard (net cps)	7-4						
	CORRECTIONS							
9	Gross weight (mg)	-						
10	Tare weight	-						
11	Net weight	9-10						
12	Weight of barium	11x0.59						
13	Carrier added	-						
14	Chemical yield factor (Y)	13/12						

Date _________________ Sample Number

		Operation						
11	Net weight	Copy						
15	Thickness correction (T)	Graph						
16	Corrected standard (cps)	8x14x15						
17	Added x 4[*] (Bq)	-						
18	Efficiency factor (E)	17/16						

*To include progenys

Date _________________ Sample Number

		Operation						
1	Counter number	-						
3	Count interval (s)	Copy						
4	Background (cps)	Copy						
18	Efficiency factor (E)	Copy						
	SAMPLE COUNTING							
19	Gross sample count	-						
20	Count time	-						
21	Sample (cps)	19/20						
22	Sample (net cps)	21-4						
23	Sample (Bq)	22x18						
	CORRECTIONS							
24	Gross weight (mg)	-						
25	Tare weight	-						
26	Net weight	24-25						
27	Weight of barium	26x0.59						
28	Carrier added	-						
29	Chemical yield factor (Y)	28/27						
26	Net weight	Copy						
30	Thickness correction (T)	Graph						

Date _________________ Sample Number

		Operation						
31	Corrected sample (Bq)	23x29x30						
	STANDARD DEVIATION							
32	Background/count interval	4/3						
33	Sample/count intervals	21/20						
34	S^2	32+33						
35	S for sample (net cps)	$\sqrt{}$34						
36	Total correction factor	18x29x30						
37	S for sample (Bq)	35x36						