NBS TECHNICAL NOTE 1137

U.S. DEPARTMENT OF COMMERCE / National Bureau of Standards

Evaluation of Methods For the Assay of Radium-228 in Water

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Issued February 1981

National Bureau of Standards Technical Note 1137
Nat. Bur. Stand. (U.S.), Tech. Note 1137, 18 pages (Feb. 1981)
CODEN: NBTNAL

U.S. GOVERNMENT PRINTING OFFICE
WASHINGTON: 1980

For sale by the Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402
Price $1.50
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EVALUATION OF METHODS FOR THE ASSAY OF RADIUM-228 IN WATER

by

J. R. Noyce

The technical literature from 1967 to May 1980 was searched for methods for assaying radium-228 in water. These methods were evaluated for their suitability as potential EPA reference methods for drinking water assays. We suggest the present EPA reference method (Krieger, 1976) be retained but improved, and a second method (McCurdy and Mellor, 1979), which employs β-γ coincidence counting, be added. Included in this report is a table that lists the principal features of 17 methods for radium-228 assays.

Key words: Analysis; assay; evaluation; radioactivity; radiochemistry; radium-228; reference method; water.

Introduction

Radium-228 is a radionuclide hazardous to human health because, like radium-226, it is incorporated into the skeleton and has a long biological residence time. Radium-228 is ingested primarily by the drinking of water. It has been found to occur in higher concentrations than ^{226}Ra in water in contact with some geologic formations in the United States (Johnson, 1971; Michel and Moore, 1980) and in Brazil (Hainberger, 1974). Radium-228 is difficult to assay accurately, especially at low concentrations, because it emits only very low-energy conversion electrons (less than 6 keV) and low-energy beta particles (10 keV average energy). Other isotopes of radium or their daughters cannot readily be added to samples as tracers for measuring the chemical yields of assays because: 1) they are also likely to be present in the water, particularly ^{224}Ra and ^{226}Ra, or 2) they are themselves inconvenient to prepare and assay, as in the cases of ^{223}Ra and ^{225}Ra.

The U. S. Environmental Protection Agency has regulations which set limits on the concentrations of radionuclides, including ^{228}Ra, in drinking water (EPA, 1976), and which specify approved assay methods. The EPA reference method for ^{228}Ra (Krieger, 1976) is based on a U.S. Geological Survey method (Johnson, 1971), which was also recommended by the World Health Organization (Lund, 1976). In addition, the U.S. Food and Drug Administration (FDA, 1979) has regulations pertaining to radionuclides in bottled water, modelled on the EPA regulations. Other methods for the low-level assay of ^{228}Ra are available, but each of them, as well as the present reference method, has one or more deficiencies for use in monitoring.

All assay methods for radionuclides should have sufficient and demonstrated precision, accuracy, and sensitivity to meet the purposes of the assay. With the possible exception of simple screening tests, such as "gross α" measurements, all radiochemical methods also should incorporate steps for determining the chemical yield.

There are additional desirable features that should be present in an ideal monitoring method. The method should be "rugged", which means it will perform well under less than ideal conditions. It should provide results without requiring a long wait for the ingrowth or decay of a radionuclide, and it should be suitable for use with large numbers of samples; that is, it should have relatively low labor and supply costs.

The Quality Assurance Division in the Environmental Monitoring Systems Laboratory of the EPA, Las Vegas, requested the NBS Radioactivity Group, Nuclear Radiation Division, to survey the literature for ^{228}Ra assay methods and evaluate their suitability as potential EPA reference methods.

Survey of Literature

Nuclear Science Abstracts was searched from 1967 until its termination in 1977. Then, the Energy Data Base was searched from 1977 through May 1980. Also, Chemical Abstracts was searched from 1968 through May 1980. References cited in the relevant papers found in these searches led to other papers dating from approximately 1960.

There are only a few papers giving methods for assaying ^{228}Ra in water and the results of such measurements. Many of the available papers deal with seawater rather than fresh water. The pertinent aspects of the methods are set forth in Table 1 in alphabetical order of author. Some of these publications also deal with ^{226}Ra assays, and salient features of these methods are included in Table 1. Papers concerned only with ^{226}Ra assays are not included.

Also in Table 1 are ^{228}Ra assay procedures found during the search that are for other types of samples, such as soil. The latter methods usually can be adapted to the assay of water samples, and some contain useful techniques not exploited in the water-assay procedures. Reports of investigations using previously published ^{228}Ra assay procedures are excluded.

Evaluation of Methods

Most of the following discussion further explains important points made in Table 1. Nearly all the assay methods separate and purify ^{228}Ac and count beta particles emitted in its decay. The method of McCurdy and Mellor (1979) is unique in that the ^{228}Ac is not separated from its ^{228}Ra parent for counting. Thus, ^{228}Ac can be counted as long as desired or recounted days later

without performing additional radiochemistry on the sample. A few procedures use the detection of radiations emitted by later members in the ^{228}Ac decay series, including ^{228}Th, ^{224}Ra, and even ^{212}Pb from de-emanated ^{220}Rn. Only the method of Tomza (1977) uses liquid-scintillation counting.

The assay methods listed in Table 1 appear to have the sensitivity required by the EPA drinking water regulations (EPA, 1976), with the apparent exceptions of those of Iyer et al. (1966) and Baretta and Feldman (1961). There is some evidence that at low ^{228}Ra concentrations a method for counting alpha particles from the decay of ^{228}Th has better precision than one for counting beta particles from the decay of ^{228}Ac (Knauss et al., 1978), but the time required for ^{228}Th ingrowth is much longer than that for ^{228}Ac.

The methods in Table 1 lacking a provision for determining the chemical yield for each sample processed are those of Humphrey et al. (1975), Johns et al. (1979), Kuchta et al. (1976), MacKenzie et al. (1979), Percival and Martin (1974), Petrow et al. (1964), Sakanoue et al. (1973), and Tomza (1977). This is also true of procedures of Barratta and Feldman (1961), and of Kahlos and Asikainen (1973), but both could readily be modified for gravimetric determination of chemical yields. Some of the methods which do provide a chemical yield, however, require long waiting times (see below).

Four of the methods use radioactive tracers to measure chemical yields: Kaufman et al. (1973), Koide and Bruland (1975), Michel and Moore (1980), and Smith and Mercer (1970). Because the present EPA reference methods for radionuclides other than tritium do not require radioactive tracers (Krieger, 1976), it was assumed they are to be avoided when another yield-measuring technique

is available. It should be noted that the chemical yield is incorrectly calculated in the reference method for ^{228}Ra (Krieger, 1976). Any loss of sample in steps 13 and 14 is counted twice. The $Ba(Ra)SO_4$ precipitate at the end of step 11 should be weighed, rather than in step 25 as stated.

The waiting periods in the methods given in Table 1 for ingrowth or decay of radionuclides during an assay can be divided into three groups: less than two full days, two days to 30 days, and greater than 30 days. Waiting periods include time for the ingrowth of ^{222}Rn for those procedures where assays of ^{226}Ra are an integral part of the ^{228}Ra assays. The short-time procedures in the first group are Baratta and Feldman (1961), Humphrey et al. (1975), Iyer et al. (1966), Krieger (1976), McCurdy and Mellor (1979), Percival and Martin (1974), and Petrow et al. (1964). Methods with waiting periods of between two and 30 days are Johns et al. (1979), Kahlos and Asikainen (1973), Koide and Bruland (1975), and Smith and Mercer (1970) (via ^{228}Ac). Methods requiring waiting times of longer than 30 days are Kaufman et al. (1973), Kuchta et al. (1976), MacKenzie et al. (1979), Michel and Moore (1980), Sakanoue et al. (1973), Smith and Mercer (1970) (via ^{228}Th), and Tomza (1977).

The methods of Kuchta et al. (1976) and Tomza (1977) would have waiting times of only two to 30 days if ^{224}Ra were known to be in equilibrium with ^{228}Ra, but this condition would probably be rare for drinking water. Finally, the waiting period in the procedure of MacKenzie et al. (1979) could also be reduced to between two and 30 days if the more volatile beta-particle-emitting progeny of ^{228}Ac could be flamed off a

source evaporated on a stainless steel planchet with no accompanying loss of ^{228}Ac (compare with Koide and Bruland (1975)).

A potential problem confronting the methods which assay ^{228}Ra *via* ^{228}Ac is the presence of radioisotopes of the rare-earth elements or those with similar chemical properties, especially ^{90}Y, daughter of ^{90}Sr. Some of the procedures have one or more steps designed to reduce such contamination, and the $\beta-\gamma$ coincidence counting technique of McCurdy and Mellor (1979) is not affected by ^{90}Y. A recent interlaboratory assay exercise, with a water test sample that had ^{90}Sr-^{90}Y added, indicated that the present EPA reference method does not adequately remove ^{90}Y from ^{228}Ac (Whittaker, 1980).

The Y or La compound chosen as the carrier for Ac should be checked for radiochemical purity before use, because it may be a significant source of beta activity (Johnson, 1971). Such a warning should be added to the reference method.

It is difficult to assess the "ruggedness" of an analytical method from just a literature survey. In general, the less complex an assay procedure is, the better chance of its being "rugged". However, some of the simpler procedures in Table 1 are without a means for measuring chemical yields.

The "special apparatus" columns in the table list items needed for each method that are judged not likely to be present in the usual water-analysis laboratory. Some of these items, such as the $\beta-\gamma$ coincidence-counting system of McCurdy and Mellor (1979), are a significant expense.

Conclusions

Most of the ^{228}Ra assay methods in Table 1 are eliminated from consideration as an EPA reference method by applying the criteria discussed earlier: adequate sensitivity, yield determination without radioactive tracers, and a waiting time for ingrowth or decay of radionuclides in the sample of no more than 30 days. The methods meeting these criteria, but with the reservation already noted about each, are Kahlos and Asikainen (1973), Krieger (1976) and McCurdy and Mellor (1979). The first of these three would be eliminated if the requirement of a waiting period for ingrowth or decay of no more than two days were imposed. It is possible that Iyer et al. (1966) and Barratta and Feldman (1961) are sufficiently sensitive, but multilaboratory studies would be necessary to demonstrate this.

It is suggested that the EPA keep Krieger (1976) as a reference method but improve its decontamination factor for ^{90}Y and correct its chemical yield calculation. It is also suggested that McCurdy and Mellor (1979) be added as a second reference method. The two procedures complement each other. The first uses radiation-detection systems which most laboratories that assay for radionuclides in water are likely to have, but it requires a considerable amount of labor to process samples. The second requires most laboratories to purchase a new radiation-detection system, but the labor to process samples is definitely less.

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The following papers were obtained too late to include in Table 1. They do not alter the conclusions of this report because none is suitable as a reference method for radium-228 assays. The radiochemical section of the Herment paper deals with the production of radium-228. Neither of the other two papers has a way to determine the chemical yield of an assay.

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Table 1. Comparison of Major Features of Methods for the Assay of Radium-228 in Water

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Reference	Intended Use	Sample Size	Preliminaries	Ra Separation	Purification	Final Form	Yielded By	Counted By
Baratta and Feldman (1961)	Assay of Ac in U mill effluents and natural waters	1 liter	Ac pptd. with La (12 mg) by HF					
Humphrey et al. (1975)	^{226}Ra and ^{228}Ra in effluent solutions with high ion concentration	250 ml	None	On $Ba(NO_3)_2$ in 70% HNO_3. Sulfate ion interferes	By AG50W-X8 ion exchange column operated at 60°. Eluted with 3 M HNO_3	Solution evap. on stainless steel planchet, planchet flamed	Nothing	Frisch-grid ionization chamber of 4.78 MeV α with PHA
Iyer et al. (1966)	Estimation of ^{228}Ra in monazite and Th samples	2 to 5 g	Digest with H_2SO_4 in Pt dish and fuse with Na_2CO_3 and K_2CO_3. Dissolve in HNO_3	On 100 mg of Ba pptd. as $BaCO_3$. Ba weighed before and after Ra separation				
Johns et al. (1979)	^{226}Ra and ^{228}Ra in water, soil, air, and biological samples	1500 g water, 1 g soil, up to 500 cm^2 of air filter, biological not specified	Water, none; digest soil in Paar bomb, then fuse; acid dissolution of air filters	Water, air filters: coppt. on $PbSO_4$; soil: coppt. on $BaSO_4$	Water and Microsorban filters: none; soil and glass-fiber filters: reppt. on $BaSO_4$	Water and Microsorban filters: OPTA complex in acetic acid; others: chloride solution	Nothing	Rn de-emanated into scintillation chamber after 30 days ingrowth. Counting begun 4.5 h after transfer
Kahlos and Asikainen (1973)	Assay of natural radioactivity in ground water	1 liter	Citric acid and NH_4OH added to filtered sample, then Ba and Pb carriers	Mixed sulfates pptd. with H_2SO_4	Dissolve ppt. in alkaline EOTA solution, reppt. sulfates at pH 4.5 with acetic acid	$Ba(Ra)SO_4$ on planchet	Weighing of $Ba(Ra)SO_4$ ppt.	ZnS(Ag)-activated α-partlcle scintillation counter
Kaufman et al. (1973), Broecker et al. (1973)	^{228}Ra and ^{228}Th in sea water	600 to 800 l	Acidify with HCl, add Fe and Ba carriers, ^{230}Th; ppt. $Fe(OH)_3$ and $Ba(Ra)SO_4$ with NH_4OH	Hydroxides dissolved in HCl; sulfates transposed to carbonates, dissolved in HCl	None	Rn de-emanated by He gas. Rn frozen out of He in liquid air-cooled trap	Nothing. Moore (1969b) found initial sulfate pptn. to be 60% to 90% efficient	α-scintillation detector
Koide and Bruland (1975)	^{226}Ra, ^{228}Ra and other natural emitters in sea water and marine sediments	20 l water, 1 to 5 g sediment	Add Pb carrier, ^{223}Ra tracer. Sediment: ignite and leach with HCl	Water: coppt. on $AlPO_4$, dissolve in 4 M HNO_3. Sediment: coppt. on $Pb(NO_3)_2$ with 75% HNO_3	Separate Ra and Pb on anion exchange column, alkaline earths on cation exchange column	Electroplated on Pt planchet after at least two weeks wait for unsupported ^{224}Ra to decay	^{223}Ra tracer. ^{227}Ac "cow" is required	α-spectrometry with semiconductor detector
Krieger (1975), based on Johnson (1971)	^{226}Ra and ^{228}Ra in drinking water	At least 1 liter	Add citric acid, Pb, Ba, and Y carriers; heat	Add NH_4OH, ppt. mixed sulfates with H_2SO_4, then add $(NH_4)_2SO_4$	Multiple coppton. on $Ba(NO_3)_2$ and on $BaSO_4$	In aklaline EOTA solution, or in $BaSO_4$ ppt. on stainless steel planchet	$BaSO_4$ gravimetrically	Rn de-emanation into scintillator cell
Kuchta et al. (1976)	Assay of skeletal ^{228}Ra and ^{228}Th in presence of gross amounts of ^{226}Ra	Not specified	Dry-ash bone sample, dissolve ash in HNO_3	None				

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Special Items	Purification	Final Form	Yielded By	Waiting Period	Counted By	Special Items	Comments
	Liquid-liquid extractions using TTA in benzene; ion exchange columns	La(Ac) oxalate. Iyer et al. (1966) think Y is better than La as carrier	Not mentioned, but could be gravimetrically	None stated, but sample should be two days old before starting assay	G-M tube or proportional counter, with Al_2 absorber when ^{227}Ac is present	None	Sekine et al. (1967) say TTA equimolar with TBP in CCl_4 is superior to TTA only
Heated ion exchange columns (60° C)	2nd ion exchange column at 60°C. Ac eluted with 6 M HNO_3	Solution evap. on stainless steel planchet	Nothing	24 h for ^{228}Ac ingrowth	Gas-flow proportional counter with 13 mg/cm^2 absorber	Same as for ^{226}Ra	Ba initially present in samples. Na, K, Ca, Mg, and Al each more than 100 mg/l in typical sample
	Ac copptd. on $Fe(OH)_3$, impurities scavenged on Pb and Bi sulfides	$Fe(Ac)(OH)_3$ dissolved in HNO_3. Known fraction evap. on planchet	Weighings of ppt. containing Ra and Ac, or spectrophotometric assay of Fe	24 h for ^{228}Ac ingrowth	End-window, gas-flow proportional counter	Pt crucible; spectrophotometer (Fe line at 304 nm used)	Method not tried on very low activity samples
Paar bomb; Pt dish for glass-fiber filter and soil samples	Reppt. Ba(Ra)SO_4, dissolve, extract Ac into OEHPA, back extract into 1 M HNO_3	Evap. acid solution to dryness on 5-cm diameter planchet	Nothing	About 30 d for ^{222}Rn ingrowth; at least 30 h for ^{228}Ac ingrowth	Low-background proportional counter. Time T_3 is ambiguously specified	None besides those for ^{226}Ra	Method could be changed to assay ^{228}Ra first. No specifics given for assaying biologicals. Section 11 wrongly states no ^{228}Ra std. are available
None. Method based on that of Goldin (1961)	Oissolve ppt., add Ce carrier, transfer to deposition cell, add Te; Pb, Bi, and Po plate out.	Ppt. Ce with NaOH, dissolve ppt. in HNO_3, add Ba holdback carrier, reppt. Ac on $Ce(OH)_3$	Nothing, but final ppt. could be Ce oxalate, which is weighable for yield	About 30 d for ^{222}Rn ingrowth	Low-background β counter, type unspecified	Constant temperature bath, for the spontaneous electrodeposition step	^{226}Ra could be counted by de-emanation of ^{222}Rn
Liquid air-cooled trap for Rn	Th copptd on $Fe(OH)_3$; ^{230}Th spike added, solution stored; Th copptd. on $Fe(OH)_3$	Th purified on cation and anion exchange columns, extracted into TTA in benzene, evap.	^{226}Ra assays of sample initially and during ^{228}Th ingrowth plus α-spectrometry of ^{230}Th tracer	4 to 11 months for ^{228}Th ingrowth	α spectrometry; counting system not specified	None additional	Method is based in part on Moore (1969a). Knauss et al. (1978) found precision of assays better by counting ^{228}Th than by counting ^{228}Ac
^{227}Ac "cow". Ultrasonic agitator	See under ^{226}Ra; none additional	On same planchet as ^{226}Ra	α counting of ^{223}Ra	At least 2 weeks for ^{224}Ra and progeny to decay	^{228}Ac in proportional counter with Al absorber. Other emitters must be flamed off planchet	None additional	$^{228}Ra/^{226}Ra$ activity ratio must be at least 0.1. As check, ingrown ^{228}Th can be counted later. Cochran (1979) completely dissolved sediments.
None	As for ^{226}Ra plus copptn. of ingrown Ac on $Y(OH)_3$	In Y oxalate ppt.	Weighing of Y oxalate	At least 36 h for ingrowth of ^{228}Ac	^{228}Ac in gas-flow proportional counter	None	Chemical yield of Ac is incorrectly calculated — see text
	None except by Rn de-emanation using a flow-through collection chamber	Solution is de-emanated, ^{220}Rn and progeny collected on negatively charged electrode	Nothing	2 counts about 1 year apart for samples without equilibrium between ^{228}Ra and ^{228}Th	Electrode placed in holder with ZnS-coated Mylar disc on phototube. ^{212}Pb and progeny counted	Flow-through chamber with collection electrode; holder for electrode on phototube	Data are analyzed by computer. Method designed to have poor sensitivity for ^{226}Ra

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Reference	Intended Use	Sample Size	Preliminaries	Ra Separation	Purification	Final Form	Yielded By	Counted By
MacKenzie et al. (1979)	Assay of ^{228}Ra and other radionuclides in sea water and marine sediments	Water: 10 to 20 l for ^{226}Ra, 1000 l for ^{228}Ra. Sediment: 5 to 10 g	Water: filter, acidify. Sediment: complete dissolution in Teflon with mixture of acids	^{226}Ra: none. ^{228}Ra: absorb on MnO_2, desorb with HCl, coppt. on $BaSO_4$	None	^{222}Rn gas	Nothing, but authors say assay is quantitative	α-scintillation in modified Lucas cell
McCurdy and Mellor (1979)	^{226}Ra and ^{228}Ra in drinking water	2 l	Add citric acid, adjust pH to 9.5, add Ba carrier (known amount)	On $BaSO_4$ pptd. with H_2SO_4	Dissolve ppt. in alkaline EDTA solution, reppt. $BaSO_4$ with acetic acid	$BaSO_4$ ppt. mixed with ZnS powder. ZnS/$BaSO_4$ ratio is approximately 2.4	Weighing of $BaSO_4$ ppt.	α-γ coincidence counting of ^{226}Ra
Michel and Moore (1980)	Assays of ^{226}Ra and ^{228}Ra in ground waters	^{226}Ra: 0.8 l; ^{228}Ra: 10 to 20 l	None (waters assayed were clear)	^{226}Ra: none. ^{228}Ra: absorb on MnO_2-impregnated acrylic fibers, desorb with HCl, ppt. on $BaSO_4$	None	^{222}Rn gas	Nothing, but each sample run at least twice	α-scintillation chamber
Percival and Martin (1974)	^{226}Ra and ^{228}Ra in soils, water, ores, and mill tailings and effluents	5 g of solids, 0.1 to 1 liter of liquid	Liquids: acidify, wait overnight, coppt. on $PbSO_4$, do sulfate fusion	Add acetic acid, coppt. Ra with $BaCl_2$ on $BaSO_4$. Dissolve ppt. in DTPA solution	None	^{222}Rn gas	Nothing	Pulse-ionization chamber or α-scintillation cell
Petrow et al. (1964)	Assay of ^{228}Ra and ^{228}Th in biological and mineral samples	Up to 50 g of bone ash; up to 200 g of food or plant ash; 5 g of minerals	Minerals: fuse with Na_2CO_3, digest in NaOH. All types: dissolve in acid	Liquid-liquid extractions with Aliquat 336 and DEHPA to remove Th. Coppt. Ra on $PbSO_4$				
Sakanoue et al. (1973)	^{228}Ra in sea water and ^{231}Pa in sea sediments	800 l	$BaCl_2$ and $FeCl_3$ added, then NH_3. $Fe(OH)_3$ and $BaSO_4$ ppt.	$Fe(OH)_3$ ppt. dissolved with HCl. $Ba(Ra)SO_4$ converted to carbonates by fusion, dissolved in HCl				^{222}Rn de-emanation (no specifics given)
Smith and Mercer (1970)	Assay of ^{226}Ra and ^{228}Ra in soils and plants	Up to 100 g of plant ash and soil	Repeated evap. with $HClO_4$ and HF, dissolve residue in HCl	Coppt. Ra on Ba and Pb sulfates. Ppt. dissolved in alkaline EDTA, sulfates repptd. with acetic acid	Cation exchange separation of Ra and Ba; Ra coppt. on $Pb(ND_2)_2$, separated by anion exchange at 60°	Electroplated on stainless steel planchet	^{133}Ba for dissolution only, ^{225}Ra for entire assay, from ^{229}Th "cow"	α-spectrometry with Frisch-grid pulse-ionization chamber
Tomza (1977)	Assay of low Ra concentrations in mineralized waters	10 l	Add citric acid and NH_4OH, then Pb and Ba carriers	Mixed sulfates pptd. with H_2SO_4	Ppt. dissolved in alkaline EDTA then repptd. by acetic acid at pH 4.5	$Ba(Ra)SO_4$ ppt. dispersed in toluene containing PPD, POPOP, and colloidal silica	Nothing	Liquid-scintillation, two counts several weeks apart

ABBREVIATIONS:
α = α particle
β = β particle
coppt. = coprecipitate
DEHPA = bis(2-ethylhexyl)phosphoric acid
DTPA = sodium diethylenetriaminepenta acetate
EDTA = sodium ethylenediaminetetraacetate
evap. = evaporate, evaporated
G-M = Geiger-Müller
PHA = pulse height analyzer
POPOP = 1,4-di-(2-(5-phenyloxazoyl))-benzene
PPD = 2,5-diphenyloxazole
ppt. = precipitate (as verb and as noun)

RADIUM-228

Special Items	Purification	Final Form	Yielded By	Waiting Period	Counted By	Special Items	Comments
25-liter glass vessel for equilibration of Rn in water samples, liquid N_2-cooled trap for ^{220}Rn	Sr removal step; extractions of Ac by TTA in benzene; store 5 weeks; more extractions of Ac	Evap. organic phases on heated stainless steel planchet	After ^{228}Ac assays, sample also assayed for ^{226}Ra. Extraction of Ac is not yielded	12 d for ^{222}Rn ingrowth, at least 5 weeks for decay of ^{228}Th progeny	Tracerlab Omniguard (gas-flow G-M counter with anticoincidence)	MnO_2-impregnated acrylic fibers, Pt crucible	Method does not discuss assay of ^{228}Ra in sediments
4 timing SCA, 2 coincidence analyzers, special detector assembly, PHA	None additional. See under ^{226}Ra	Same as ^{226}Ra	Same as ^{226}Ra	^{228}Ac ingrowth time not specified, assumed to be 1 to 2 days	β-γ coincidences counting of ^{228}Ac	Same as for ^{226}Ra	Considerable instrumentation is required, but amount of radiochemistry is reduced
Apparently none	Carbonate fusion, dissolve mixed ppt. in HCl, coppt. Th on Fe, store, add ^{230}Th, coppt. Th on Fe	Purify Th by ion exchange, mount on planchet for thin source α counting	α spectrometry of Th, ^{228}Ra assay of concentrated sample. Separate ^{226}Ra assay also needed	4 to 10 d for ^{222}Rn ingrowth, 2 to 4 months for ^{228}Th ingrowth	α spectrometry of ^{228}Th and ^{230}Th; counting system not specified	MnO_2-impregnated acrylic fibers. See Moore (1976) for preparation	Ra extraction by fibers based on Krishnaswami et al. (1972) and on Moore and Reid (1973)
Pt dish	Remove Ba as sulfate, extract Ac from supernate into 15% DEHPA, strip into HNO_3	Add Ce, coppt. Ac on Ce exalate	Nothing	At least 20 h for ^{228}Ac ingrowth	Gas-flow proportional counter	None additional	
	Metathesize ppt. to carbonates, dissolve, coppt. Ra on $Pb(NO_3)_2$; extract Ac with DEHPA, Aliquat	Evap. final acid solution on 5-cm diameter stainless steel planchet	Nothing	At least 36 h for ^{228}Ac ingrowth	β from decay of ^{228}Ac. Instrument unspecified	Pt crucible	This method is extention of similar method by Petrow and Allen (1961) for U mill effluents which also is unyielded
	Extract Ac into DEHPA, back extract into HNO_3, load on Doxex 50 ion exchange column, elute with 6 M HNO_3	Evap. on stainless steel plate	^{226}Ra assay on initial sample and on $Ba(Ra)Cl_2$ solution just before Ac extraction. Extraction is not yielded	Unspecified. Assume 27 d for each of 2 ^{222}Rn ingrowths	β counting of ^{228}Ac in guarded, gas-flow proportional counter, or in Fijitsu "pico beta" β spectrometer	None	
Heated ion exchange column; ^{229}Th "cow"; ^{133}Ba tracer	Coppt. Ac on Fe, solution stored, Ac reppt. on Fe, Ac separated on anion exchange column from Fe	Solution evap. on stainless steel planchet	β counting of ^{225}Ac and progeny. ^{133}Ba not needed if method used with water sample	7 to 10 d for growth of ^{225}Ac	β counting of ^{228}Ac and ^{225}Ac in gas-flow proportional counter with 13 mg/cm^2 absorber	None additional	Alternate ^{228}Ra method: α spectrometry after several months wait of ^{216}Po and ^{225}Ra on the Ra planchet, without further chemistry
None	Simultaneous with ^{226}Ra	Same as for ^{226}Ra	Nothing	Equilibrium between ^{228}Ra and ^{228}Th assumed. If not so, must reassay several months later	Liquid-scintillation counting of ^{224}Ra and progeny. β pulses of ^{228}Ra discriminated against	None	Use of moderately complex equations of ingrowth and decay is required

reppt. = reprecipitate
SCA = single channel analyzer
std. = standard
TBP = tributylphosphate
TTA = 2-theonyltrifluoroacetone

S-114A (REV. 2-8C)

U.S. DEPT. OF COMM. BIBLIOGRAPHIC DATA SHEET (See instructions)	1. PUBLICATION OR REPORT NO. NBS TN 1137	2. Performing Organ. Report No.	3. Publication Date February 1981

4. TITLE AND SUBTITLE

Evaluation of Methods for the Assay of ^{228}Ra in Water

5. AUTHOR(S)

J.R. Noyce

6. PERFORMING ORGANIZATION (If joint or other than NBS, see instructions)

NATIONAL BUREAU OF STANDARDS
DEPARTMENT OF COMMERCE
WASHINGTON, D.C. 20234

7. Contract/Grant No.

8. Type of Report & Period Covered

Final

9. SPONSORING ORGANIZATION NAME AND COMPLETE ADDRESS (Street, City, State, ZIP)

Quality Assurance Division
Environmental Monitoring Systems Laboratory
U.S. Environmental Protection Agency, P.O. Box 15027
Las Vegas, NV 89114

10. SUPPLEMENTARY NOTES

☐ Document describes a computer program; SF-185, FIPS Software Summary, is attached.

11. ABSTRACT (A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here)

The technical literature from 1967 to May 1980 was searched for methods for assaying radium-228 in water. These methods were evaluated for their suitability as potential EPA reference methods for drinking water assays. We suggest the present EPA reference method (Krieger, 1976) be retained but improved, and a second method (McCurdy and Mellor, 1979), which employs β-γ coincidence counting, be added. Included in this report is a table that lists the principal features of 17 methods for radium-228 assays.

12. KEY WORDS (Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons)

Analysis; assay; evaluation; radioactivity; radiochemistry; radium-228; reference method; water.

13. AVAILABILITY

☒ Unlimited

☐ For Official Distribution. Do Not Release to NTIS

☒ Order From Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402.

☐ Order From National Technical Information Service (NTIS), Springfield, VA. 22161

14. NO. OF PRINTED PAGES

18

15. Price

$1.50

USCOMM-DC 6043-P80

☆ U.S. GOVERNMENT PRINTING OFFICE : 1981 O—340-997 (1511)

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