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Evaluation of the Anthropogenic Radionuclide Concentrations in Sediments and Fauna Collected in the Beaufort Sea and Northern Alaska

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Abstract

This study was performed to establish a quality controlled data set about the levels of radionuclide activity in the environment and in selected biota in the U.S. Arctic. Sediment and biota samples were collected by the National Oceanic and Atmospheric Administration (NOAA), the National Biological Service, and the North Slope Borough's Department of Wildlife Management to determine the impact of anthropogenic radionuclides in the Arctic. The results summarized in this report are derived from samples collected in northwest Alaska with emphasis on species harvested for subsistence in Barrow, Alaska. Samples were analyzed for the anthropogenic radionuclides ^{90}Sr, ^{137}Cs, ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Am. The naturally occurring radionuclides ^{40}K, ^{212}Pb and ^{214}Pb were also measured.

One goal of this study was to determine the amounts of anthropogenic radionuclides present in the Beaufort Sea. Sediment samples were isotopically fingerprinted to determine the sources of radionuclide activities. Biota samples of subsistence and ecological value were analyzed to search for evidence of bio-accumulation of radionuclides and to determine the radiation exposures associated with subsistence living in northern Alaska. The anthropogenic radionuclide content of sediments collected in the Beaufort Sea was predominantly the result of the deposition of global fallout. No other sources of anthropogenic radionuclides could be conclusively identified in the sediments. The anthropogenic radionuclide concentrations in fish, birds and mammals were very low. Assuming that ingestion of food is an important pathway leading to human contact with radioactive contaminants and given the dietary patterns in coastal Arctic communities, it can be surmised that marine food chains are presently not significantly affected.

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Introduction

This study was performed to establish a quality controlled data set about the levels of radionuclide activity in the environment and selected biota in the U.S. Arctic. This report provides results of radiochemical analyses of sediments and biological samples. The samples were collected by the National Oceanic and Atmospheric Administration (NOAA), the National Biological Service, and the North Slope Borough's Department of Wildlife Management. Samples were collected in northwestern Alaska with emphasis on species harvested for subsistence in Barrow, Alaska. Sediment samples were also collected and analyzed. Our objective was to characterize the types of radioactivities, and, if possible, determine their sources. Biota samples of subsistence and ecological value were analyzed to search for evidence of bio-accumulation and to determine the radiation exposures associated with subsistence living in northern Alaska.

Sample Collection and Preparation

Grab samples of sediments were collected during NOAA's 1993 sampling in the Beaufort Sea. Samples were collected by the United States Coast Guard (USCG) Polar Star on the 29 July - 16 August 1993 sampling cruise. Figure 1 illustrates the cruise track of the USCG Polar Star. Figure 2 illustrates the sampling locations selected for this study

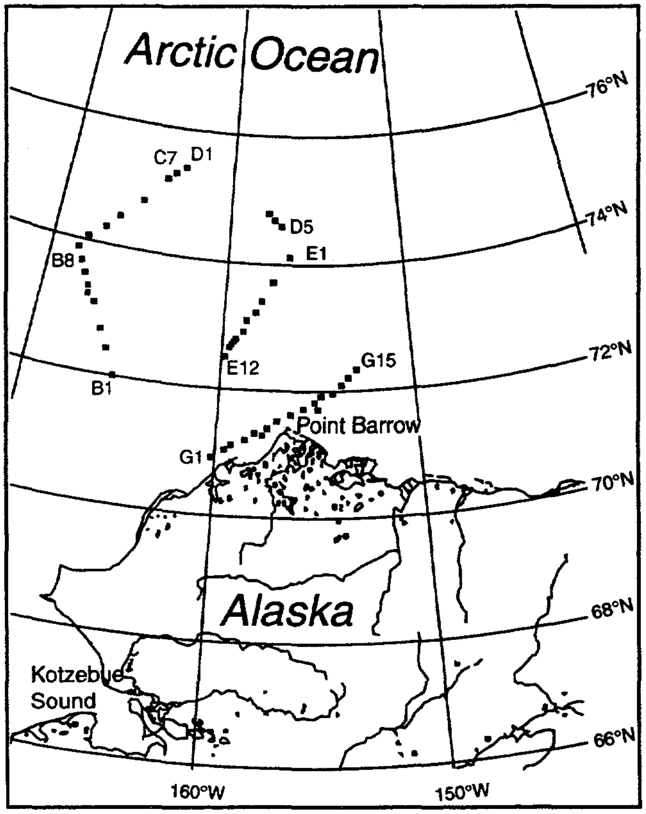

Figure 1 The Cruise Track of the USCG Polar Star

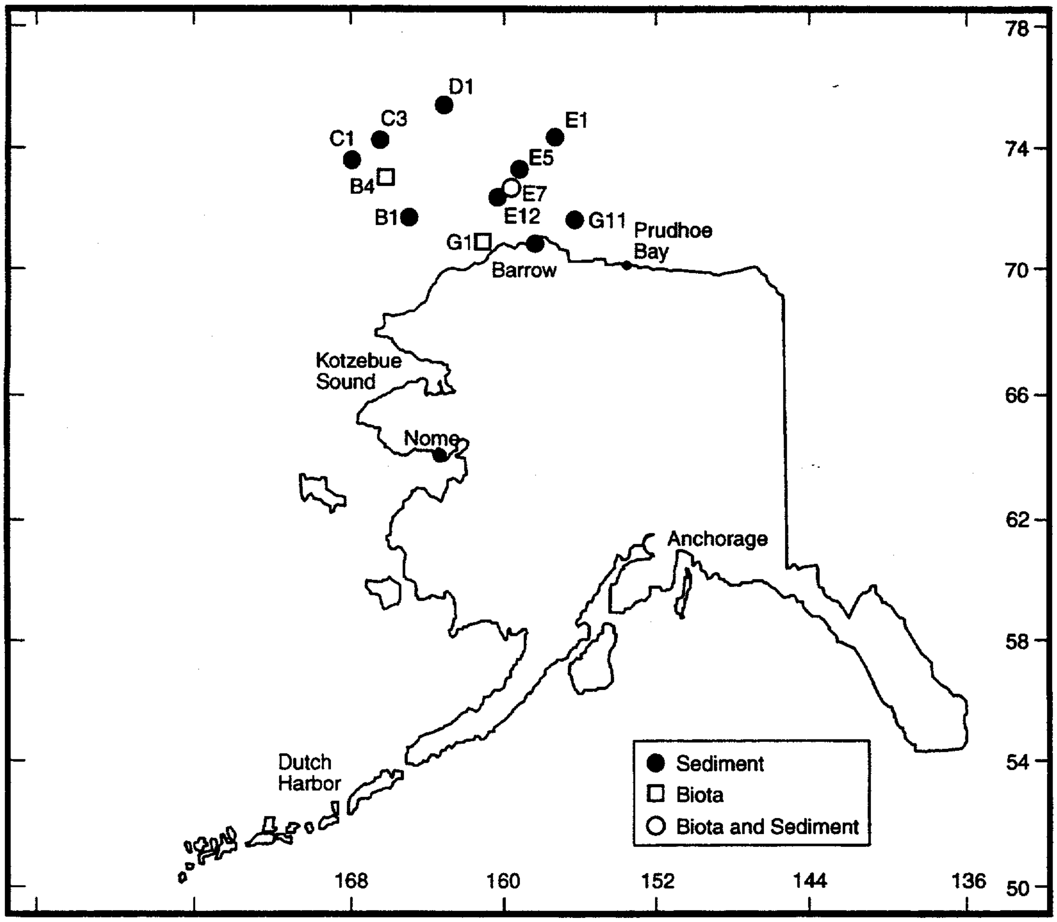

Figure 2 The Sampling Locations Selected During the Polar Star Cruise

The top 2-3 cm of each grab sample were reserved for radiochemical analyses. Table 1 lists the sediment ARCRAD 93 sampling locations assigned to the samples collected by personnel on the Polar Star. Two additional sediment samples, 1S1 and 1S2, were collected in the coastal waters off Barrow by personnel associated with the North Slope Borough's Department of Wildlife Management. Table 2 lists the sediment compositions. Whenever practical, sediments and biological samples were collected at the same locations. Representative specimens of the fauna collected at ARCRAD 93 sampling locations B-04, E-07 and G-01 were submitted for radiochemical analysis. Table 3 lists the biota collected at the ARCRAD 93 sampling locations. Additional fauna samples were collected as far east as Prudhoe Bay and as far south as Kotzebue Sound. Table 3 also lists these additional marine species and mammals collected for the study. Most of the mammals were collected in the Barrow vicinity. The one exception, the bearded seal, was collected in the vicinity of Nome, Alaska. Representative samples of tissue and organs from the mammals were analyzed for radioactivity content. The king eider specimens were divided into bone and muscle samples prior to radiochemical analyses. The fish and invertebrates were analyzed whole.

Table 1 - Sediment Sampling Locations in the Beaufort Sea and Barrow, Alaska

Station	Latitude	Longitude	Depth (m)	Date Sampled
E-12	72: 30.0N	159: 48.0W	40	30-Jul-93
E-07	72: 54.1N	158: 59.2W	113	31-Jul-93
E-05	73: 13.3N	158: 16.9W	1976	3-Aug-93
E-01	74: 24.0N	155: 43.1W	3813	5-Aug-93
D-01	75: 23.8N	163: 00.3W	2050	8-Aug-93
C-03	73: 18.1N	167: 05.8W	289	10-Aug-93
C-01	73: 53.8N	168: 29.7W	236	10-Aug-93
B-04	73: 05.8N	166: 59.8W	52	11-Aug-93
B-01	71: 59.7N	165: 21.2W	34	12-Aug-93
G-01	70: 54.1N	160: 10.7W	44	12-Aug-93
G-11	71: 54.3N	154: 46.0W	340	14-Aug-93
Barrow 1S1	71: 20 N	156: 39 W	< 5	29-Apr-95
Barrow 1S2	71: 20 N	156: 39 W	< 5	3-May-95

Table 2 - Composition of Sediments Collected in the Beaufort Sea

Station	% Gravel	% Sand	% Silt	% Clay	TOC (wt%)	TIC (wt%)
E-12	0.0	10.1	50.7	39.2	1.18	1.43
E-07	0.0	7.6	50.9	41.5	1.47	1.78
E-05	0.0	1.1	48.9	50.0	1.01	1.25
E-01	0.0	0.1	43.9	56.0	0.72	1.01
D-01	0.4	2.8	72.4	24.4	0.54	1.34
C-03	0.0	0.5	60.1	39.3	0.97	1.24
C-01	0.0	1.1	47.6	51.3	1.17	1.32
B-04	0.0	1.6	50.4	48.0	1.79	2.07
B-01	0.0	36.1	52.5	11.4	0.59	0.9
G-01	*100.0	**N/M	N/M	N/M	N/M	N/M
G-11	1.1	24.6	45.6	28.6	1.33	1.67
Barrow 1S1	N/M	N/M	N/M	N/M	N/M	N/M
Barrow 1S2	N/M	N/M	N/M	N/M	N/M	N/M

* The ocean floor at sampling station G-1 was covered with gravel. No grab sediment sample was collected.

** N/M = Sample not measured

Table 3 - Fauna Collected for Study

Species	Type of Tissue	Collection Location
Sipunculids	whole animal	ARCRAD 93 station B-4
Crinoids	whole animal	ARCRAD 93 station E-7
Sea urchin	whole animal	ARCRAD 93 station G-1
Spider crab	whole animal	ARCRAD 93 station G-1
Starfish	whole animal	ARCRAD 93 station G-1
Soft starfish	whole animal	ARCRAD 93 station G-1
Nudibranch	whole animal	ARCRAD 93 station G-1
Invertebrates	whole animals	Barrow vicinity
Broad Whitefish #1	whole fish	Prudhoe Bay
Broad Whitefish #2	2 whole fish	Prudhoe Bay
Arctic Char	whole fish	Noatak River
Arctic Cod	~ 30 whole fish	Chuckchee Sea
Arctic Cisco #1	whole fish	Prudhoe Bay
Arctic Cisco #2 (2 fish)	2 whole fish	Prudhoe Bay
Least Cisco (4 fish)	4 whole fish	Prudhoe Bay
Chum Salmon	whole fish	Kotzebue Sound
Dolly Varden (3 fish)	3 whole fish	Prudhoe Bay
Bowhead Whale #1	blubber	Barrow vicinity
Bowhead Whale #1	lung	Barrow vicinity
Bowhead Whale #1	liver	Barrow vicinity
Bowhead Whale #B8	blubber	Barrow vicinity
Bowhead Whale #B8	muscle	Barrow vicinity
Bowhead Whale #B8	kidney	Barrow vicinity
Bowhead Whale #B8	liver	Barrow vicinity
King Eider	skeleton	northern Alaska
King Eider	pectoral muscle	northern Alaska
Polar Bear #1	fat	Barrow vicinity
Polar Bear #1	muscle	Barrow vicinity
Polar Bear #1	kidney	Barrow vicinity
Polar Bear #1	liver	Barrow vicinity
Polar Bear #2	muscle	Barrow vicinity
Caribou #4	rumen	Barrow vicinity
Caribou #4	muscle	Barrow vicinity
Caribou #4	liver	Barrow vicinity
Caribou #4	kidney	Barrow vicinity
Bearded Seal	blubber	Nome vicinity
Bearded Seal	kidney	Nome vicinity

Samples were frozen and packed in dry ice for shipment to Los Alamos National Laboratory (LANL). The sample handling procedures used at LANL are documented in the Appendix. Table 4 and Table 5 list the wet weights, dry weights and ash weights of the sediments and fauna. The nomenclature for the sediment sample identification lists both the NOAA 93 station sample identifier assigned by NOAA prior to shipment of the sample to LANL and the ARCRAD 93 station sample identifier initially assigned during the cruise track of the USCG Polar Star between 29 July - 16 August 1993. Sample SR1A E12 identifies the sample as NOAA sediment sample SR1A collected at ARCRAD 93 station E12. Three sediments SR1A E12, SR1B E12 and SR1C E12 were collected at station E12. Analysis of these three samples from the same location was performed to assess the overall error associated with sample inhomogeneity and analytical measurement. Resources were limited and we could not obtain three samples from each sampling location.

Table 4 - Weight of Sediment Samples Delivered to LANL

Sample ID	Initial Weight (g)	Dry Weight (g)	Ash Weight (g)
SR1A E12	406.2	160.9	151.2
SR1B E12	476.8	183.7	170.9
SR1C E12	402.9	153.2	143.1
SR7 C1	274.2	99.7	89.9
SR2A E7	154.8	43.6	39.8
SR4 E1	217.1	71.5	67.9
SR3B, SO3A E5	311.3	101.3	90.6
SR6 C3	107.2	103.1	95.4
SR9 B1	539.8	282.0	271.8
SR10 G11	432.1	173.3	158.7
SR5A D1	446.4	207.1	193.3
SR8 B4	468.4	123.8	112.0
Barrow 1S1	N/M	32.8	31.6
Barrow 1S2	N/M	33.2	31.5

Table 5 - Weight of Biological Samples Delivered to LANL

Sample ID	Initial Weight (g)	Dry Weight (g)	Ash weight (g)
Sta B-4, Sipunculids	190.3	28.0	8.2
Sta E-7, Crinoids	119.5	31.2	10.7
Sta G-1, Sea Urchins	1575.6	602.3	507.7
Sta G-1, Spider Crab	711.4	259.7	133.3
Sta G-1, Regular Starfish	409.2	146.5	94.4
Sta G-1, Soft Starfish	94.6	32.7	12.4
Sta G-1, Nudibranchs	1453.1	439.6	271.8
Invertebrates	327.1	99.8	30.7

Table 5 - Weight of Biological Samples Delivered to LANL (cont'd)

Sample ID	Initial Weight (g)	Dry Weight (g)	Ash weight (g)
Broad Whitefish #1	1920.2	354.0	23.1
Broad Whitefish #2 (2 fish)	552.1	215.3	45.1
Arctic Char	230.0	95.0	10.3
Arctic Cod	936.0	487.0	18.2
Arctic Cisco #1	409.1	159.0	19.0
Arctic Cisco #2 (2 fish)	376.9	139.5	12.6
Least Cisco (4 fish)	463.0	134.3	12.8
Chum Salmon	3453.0	1204.0	24.8
Dolly Varden (3 fish)	1018.0	349.1	54.7
Bowhead Whale #1 blubber	826.0	798.5	6.5
Bowhead Whale #1 lung	952.3	295.8	8.7
Bowhead Whale #1 liver	1577.2	441.6	15.1
Bowhead Whale #B8 blubber	3197.4	2792.9	7.3
Bowhead Whale #B8 muscle	598.2	155.2	6.7
Bowhead Whale #B8 kidney	400.0	128.7	28.5
Bowhead Whale #B8 liver	410.1	118.0	21.8
King Eider bone	309.7	278.6	10.3
King Eider muscle	688.1	225.6	20.2
Polar Bear #1 fat	460.6	440.8	8.9
Polar Bear #1 muscle	286.3	105.9	7.4
Polar Bear #1 kidney	95.0	35.8	3.0
Polar Bear #1 liver	114.4	36.9	4.2
Polar Bear #2 muscle	1429.8	481.0	89.6
Caribou #4 rumen	99.4	16.7	2.1
Caribou #4 muscle	86.8	37.9	2.9
Caribou #4 liver	184.5	57.0	8.9
Caribou #4 kidney	156.7	38.0	4.2
Bearded Seal blubber	373.1	351.1	4.0
Bearded Seal kidneys	557.0	135.3	10.3

The ashed samples were analyzed by high-resolution gamma ray spectroscopy. After these analyses were completed, the ashed samples selected for radiochemical analyses were dissolved in strong mineral acids.[1,2] Plutonium-238, ^{239}Pu, ^{240}Pu, ^{137}Cs and ^{90}Sr were analyzed from a single aliquot of each sample. Plutonium was separated and purified by ion exchange chromatography.[3] The plutonium was electroplated on platinum and the ^{238}Pu and $^{239+240}$Pu concentrations were measured by alpha spectroscopy.[4,5] The plutonium was removed from the platinum disks and the ^{240}Pu/^{239}Pu atom ratios were measured by Thermal Ionization Mass Spectrometry (TIMS).[6,7] Next, iron was removed from the sediment samples by an isopropyl ether extraction. The small amount of iron present in biota was removed as the chloro complex by anion exchange. This removal was done to maximize the chemical yields for cesium and strontium. The cesium fractions were isolated by sorption on microcrystalline ammonium phosphomolybdate.[8] The cesium was measured by low-level beta counting and high-resolution gamma ray spectroscopy. The strontium fractions were isolated as carbonates and purified by cation exchange chromatography.[9] The ^{90}Sr activity levels were determined by measuring the ^{90}Y daughter using low-level beta counters. The ^{241}Am content was measured on separate ≈ 5 g aliquots of ash. Americium was separated by co-precipitation with lanthanum fluoride and by a HDEHP (Di-2-ethylhexylphosphoric acid in heptane) extraction.[10] Final purification was accomplished by anion exchange chromatography. The americium was quantified by solid state alpha spectrometry. The detailed chemical separation and purification procedures are listed in the Appendix.

Instrumentation

The high resolution gamma ray spectroscopy measurements were obtained by placing the ashed residues in counting vials 4 cm o.d. x 2.7 cm high and counting for 3000 minutes using an intrinsic germanium detector having 81% efficiency relative to a 3" x 3" sodium iodide crystal. The resolution of the intrinsic germanium detector is 1.7 keV Full Width Half Maximum (FWHM) at 1.33 MeV. The efficiency of the detector is dependent on energy. We measured the efficiency versus energy using gamma standards that emit gamma rays at various energies. These gamma standards are traceable to the National Institute of Standards and Technology (NIST). We also fit the data to a semi-empirical polynomial so that we could predict the efficiency at energies different from the calibration points.[11] The separated cesium samples were counted for 1000 minutes in a well-type intrinsic germanium detector having 26.5% efficiency relative to sodium iodide. The resolution of this detector is 2.4 keV FWHM at 1.33 MeV.

The plutonium and americium alpha activity levels were determined by counting the samples on 300 mm^2 solid state surface barrier detectors. The detectors have a resolution (FWHM ~20 keV) and a detection efficiency of 35%. The detectors had backgrounds of 0.5 - 1.2 c/hr in the 4-6 MeV range. Samples were counted for 6,000 to 10,000 minutes.

Strontium-90 and ^{137}Cs were also counted on low level beta counters. The detector system uses a titanium foil window that prevents sensitivity below 50 keV. The detector chambers are fabricated from high purity copper, placed inside an active background suppression umbrella, and surrounded by a few tons of lead. The counter background averages 0.2 cpm. Propane is used as the counting gas.

The TIMS used in this study to measure the ^{240}Pu/^{239}Pu atom ratios is a modified version of the 30.5-cm radius, 90° deflection, thermal ionization instrument developed at the National Bureau of Standards (now named the National Institute of Standards and Technology, or NIST).[12] The spectrometer is equipped with a 17-stage electron multiplier with a gain of 10^8, a discriminator-amplifier capable of supplying 1 V, 7 ns wide output pulses when triggered by input pulses between 6 mV and 2 V, and a 250

MHz scaler. The dark current of the counting system is 0.05 counts per second; its deadtime is 11 ± 1 ns. The system's overall linearity and deadtime were verified by analyzing NIST Standard Reference Materials (SRM): U-100, U-500 and U-900.[13] The data acquisition is automated with an IBM Personal Computer. The spectrometer is equipped with a sample turret that holds five Surface Ionization Diffusion (SID) controlled filaments.[14]

Quality Assurance/Quality Control

NIST SRM for environmental radioactivity were processed concurrently as part of our quality assurance program. These SRMs served as the matrix based quality assurance standards for the sediment analyses. Table 6 lists the NIST certified matrix based reference materials that we used during this investigation. Processing blanks were also analyzed as part of our QA/QC program.

Table 6 - NIST Matrix Based QA/QC Samples

SRM 4350B	Columbia River Sediment
SRM 4353	Rocky Flats Soil Number 1
SRM 4354	Freshwater Lake Sediment
SRM 4355	Peruvian Soil

We also have an extensive QA/QC program to verify that the instrument measurement systems are functioning properly. Each gamma counting system has one or more standards that are counted each day. Those data are computer processed to check energy calibration, energy resolution and overall detection efficiency. These data are archived as well as being plotted on running plots that are updated weekly for reference. Absolute detection efficiency checks are made whenever any out-of-normal incident occurs. Background counts are made whenever there are no samples being counted. The background data were also been analyzed to test for possible counter contamination.

Plutonium and americium samples submitted for alpha analyses are traced with ultra pure ^{242}Pu and ^{243}Am, respectively. The ^{242}Pu is standardized to gravimetrically prepared dilutions of SRM 949f plutonium. Weighed aliquots of SRM 949f are added to weighed amounts of the ^{242}Pu tracer and the ^{242}Pu/^{239}Pu atom ratio of the mixture is measured by TIMS. The amount of ^{239}Pu added from the SRM 949f is known. The ^{242}Pu/^{239}Pu atom ratio measured by TIMS is used to calculate the number of atoms of ^{242}Pu in the tracer. The ^{243}Am tracer is calibrated by stippling weighed amounts of the tracer on counting planchets and counting the samples on alpha detectors that have been calibrated for efficiency with standards traceable to NIST. The purity of the ^{243}Am tracer was verified by alpha spectrometry analyses. The energy calibration of the alpha spectrometers was done using a multipeak alpha standard. Background counts are made whenever there are no samples being counted. The background data were also been analyzed to test for possible counter contamination.

The proper operation of the low-level beta counter was verified by analyzing ^{90}Sr, ^{90}Y and ^{137}Cs samples traceable to NIST standards. Background counts are made whenever there are no samples being counted. The background data have also been analyzed to test for possible counter contamination.

The TIMS was calibrated using actinide standards certified by NIST, New Brunswick Laboratory and the Commission of the European Communities - Joint Research Centre - Institute for Reference Materials and Measurements (IRMM). The proper operation of the TIMS was verified by measuring Certified Reference Materials, CRM U500, CRM-128 and CBNM 072-7. CRM U500 has approximately equal atom amounts of ^{235}U and ^{238}U. This Certified Reference Materials list was originally issued in 1970 by the NIST as SRM U-500. CRM 128 has approximately equal atom amounts of ^{239}Pu and ^{242}Pu. This CRM was prepared at the New Brunswick Laboratory. CBNM 072-7 is a uranium isotopic standard certified by IRMM.

Potential Source Terms for Anthropogenic Radioactivity

Global fallout from nuclear weapons testing in the atmosphere, discharges from nuclear facilities in the Former Soviet Union (FSU) into Siberian rivers, undersea thermonuclear explosions at the Novaya Zemlya test site, the Chernobyl accident, aircraft and satellite accidents involving radioactive materials, and dumping of radioactive waste in the ocean are considered important sources of anthropogenic radioactivity in the Arctic. There are a number of excellent reviews documenting the release of anthropogenic radioactivity into the Arctic.[15,16,17,18] The estimates for the release of radioactivity vary from article to article, however. Information concerning these potential source terms has been summarized below. For completeness, information on the operation of nuclear process facilities in Europe and United States underground nuclear testing on Amchitka Island has been included.

Activities in the Former Soviet Union

For illustrative purposes, the review of the activities in the Former Soviet Union presented at the USA/FSU Workshop on Actinides in the Environment conducted in Dublin, California, August 15-18, 1995 has been summarized below.[18]

The Chernobyl Event

In April 1986, the N4 reactor at Chernobyl released 3.7 x 10^{18} Bq of radioactive materials into the environment. These radioactive materials included a variety of fission and activation products produced during the operation of the reactor. Radionuclides from the Chernobyl accident were distributed over wide geographical areas. The radioactive materials included a variety of fission and activation products produced during the operation of the reactor.

Altay

This area received radioactivity from the Semipalatinsk test area. The mean radioactivity level of Altay is (3.7-7.4) x 10^9 Bq/km^2 of ^{137}Cs, (3.7-18) x 10^8 Bq/km^2 of ^{90}Sr and (1.8-

7.4) x 10^8 Bq/km^2 of $^{239+240}$Pu. Two relatively small areas exist with higher levels of radioactivity.

Southern Ural

A source of radionuclides in the southern Ural is the "Mayak" (formerly Chelyabinsk-65, or Kyshtym, now known as Ozersk), facility. Radioactive contamination of the Ural region is partly due to three sources:

- liquid radioactive waste discharge into the Techa River,
- accidental discharge into the atmosphere resulting from the explosion of a tank holding high-level radioactive waste on September 29, 1957, and
- re-suspension of radioactive materials from the shores of Lake Karachay in 1967 due to drought conditions.

The majority of the radioactive waste is located in 257 ground surface storage tanks (3.7 x 10^{19} Bq). Other radioactive materials were also place near Ozersk in three locations.

- in water reservoirs (1.9 x 10^{19} Bq),
- in geological formations (3.7 x 10^{19} Bq), and
- in Lake Karachay (6.7 x 10^{18} Bq).

About 7.6 x 10^7 m^3 of waste containing 1 x 10^{17} Bq of radionuclides was discharged into the Techa River during the period 1949-1956. About 95% of the radioactivity was discharged from March, 1950 to November, 1951. Most of this material was adsorbed by sediments in the upper reaches of the Techa River. The average concentrations of ^{90}Sr, ^{137}Cs and ^{239}Pu in bottom sediments collected in 1993 at the Muslimovo settlement 78 km downstream from Ozersk were 0.08, 0.1 and 0.001 Bq/kg, respectively. The water from the Techa passes successively into the Iset, Tobol and Ob Rivers. The concentration of radionuclides in the Iset River was about an order of magnitude lower than that measured in the Techa River. Concentrations in the Tobol River are 100-1000 times lower than in the Techa River.

Western Siberia

A significant portion of the FSU nuclear program is located in the West Siberian Basin. Many caches of radioactive material (7.4 x 10^{19} Bq) are located in geological formations, injection sites and storage tanks. The Tomsk-7 and Krasnoyarsk-26 facilities are believed to be responsible for the majority of the radioactive contamination that is found in the major river systems that feed the Arctic Ocean through the Kara Sea. In 1991 the maximum plutonium levels measured in floodplains of the Yenisey River were estimated at (0.7-1.9) x 10^9 Bq $^{238+239+240}$Pu/km^2.

Arctic Areas Controlled by the FSU

A number of nuclear weapons tests were conducted in the Arctic:

- three tests in the Kol'skiy peninsula,

- 12 tests in Yakutiya,
- 12 tests in the northern part of Arkhangel'sk region and in western Siberia,
- 2 tests near Igarka and at Bol'shaya Kheta River, and
- some nuclear tests were also conducted in the Perm' region.

Eighty-three atmospheric, 3 ground surface, 3 underwater, and 1 low-altitude test were conducted at the Central Testing Ground for Nuclear Weapons at Novaya Zemlya Island from 1954-1962. These tests resulted in the release of 4.8 x 10^{17} Bq of ^{137}Cs into the atmosphere. The average concentration of plutonium in soils at Novaya Zemlya is 500-900 Bq/kg. Forty-eight underground tests were conducted from 1965-1991. These tests occasionally produced short-term increases of radioactive background in some areas.

A volume of 6000-7000 m^3 of liquid radioactive waste is generated annually by the Northern Fleet operations. Thirty percent of this material is associated with activities in the White Sea; the remaining 70% is associated with activities in the Barents Sea region. The annual amount of solid radioactive waste is estimated to be 45000 m^3. The FSU has also disposed of nuclear waste in the Kara Sea:

- at least 16 nuclear reactors (6 with fuel),
- over 10,000 containers of low-level radioactive waste, and
- liquid waste.

These disposals are estimated to contain 8.6 x 10^{16} Bq of radioactivity. Disposal of solid radioactive wastes in the Arctic Ocean was discontinued in 1992. The 16 nuclear submarine reactors (6 with fuel) and the reactor of the nuclear icebreaker Lenin (with fuel) were placed in the shallow areas of the Barents and Kara Seas during 1965-1988. The total activity of fission products inside these reactors is 8.5 x 10^{16} Bq. The average concentration of $^{238+239+240}$Pu in waters in the Barents and Kara Seas is 0.01 Bq/m^3.

Nuclear Facilities in NW Europe

The operation of nuclear facilities in NW Europe has resulted in the controlled release of low-level radioactive waste into coastal waters. The principal sources are due to nuclear fuel reprocessing. From 1952 to 1991 Sellafield (United Kingdom) discharged 2.9 x 10^{16} Bq ^{137}Cs, 4.2 x 10^{15} Bq ^{90}Sr and 6.9 x 10^{14} Bq $^{239+240}$Pu.[19] By 1985, La Hague (The Netherlands) discharged 7.6 x 10^{14} Bq ^{137}Cs, 6.75 x 10^{14} Bq ^{90}Sr and 3 x 10^{12} Bq of plutonium.[20] A significant portion of the radioactivity discharged from Sellafield was adsorbed onto particles and deposited in sediments. Sediments collected near Sellafield have $^{239+240}$Pu concentrations ranging from 260-4240 Bq/kg.[21] The transit time for ^{137}Cs from Sellafield to the Barents Sea is five years.[22]

U. S. Nuclear Testing on Amchitka Island

The United States conducted three underground nuclear tests on Amchitka Island, Alaska[23] during the period 1965-1971: Long Shot, 29 October 1965; Milrow, 2 October 1969; and Cannikin, 6 November 1971.

Global Fallout

The United States, the Former Soviet Union, the United Kingdom, France and China have conducted 511 atmospheric tests yielding a total equivalent to detonating 438 million tons of TNT.[24] Most of the global fallout was deposited in the Northern Hemisphere and the majority of the fallout was deposited in the middle latitudes.[25,26] The majority of the atmospheric testing was conducted prior to 1963. Approximately 25% of the total yield of atmospheric nuclear weapons tests occurred in 1956 and 75% occurred in 1962.[27] The total fission yield of atmospheric testing from 1963-1970 was about 5% of the yield of the pre-1963 testing.[28] The amount of fallout produced by atmospheric testing from 1970 to 1981 is insignificant compard with the global fallout produced prior to 1963. Essentially all of the global fallout has been deposited on the earth's surface. Table 7 lists the radionuclides remaining in global fallout.

Table 7 - List of Radionuclides Remaining in Global Fallout

Radionuclide	Characteristics	Half Life (Years)
^{3}H	G,C	12.3
^{14}C	G	5.7×10^3
^{36}Cl	C	3×10^5
^{85}Kr	G	10.8
^{90}Sr	G,C	29.1
^{99}Tc	C	2.1×10^5
^{129}I	G,C	1.6×10^7
^{133}Ba	NC	10.5
^{135}Cs	G,C,NC	2.3×10^6
^{137}Cs	G,C,NC	30.2
^{152}Eu	NC	13.5
^{154}Eu	NC	9.8
^{236}U	C	2.3×10^7
^{237}Np	C	2.1×10^6
^{238}Pu	NC	87.8
^{239}Pu	NC	2.4×10^4

Radionuclide	Characteristics	Half Life (Years)
^{240}Pu	NC	6.6 x 10^3
^{241}Pu	NC	14.4
^{241}Am	NC	437.2

- **C** = These radionuclides can be transported by water and they do not sorb significantly on sediments.
- **G** = These radionuclides are gases at or near ambient temperature or they have relatively long-lived precursors that can be transported by gaseous injections.
- **NC** = These radionuclides sorb onto sediments.

Cesium has gaseous precursors. It is soluble while in the water column. It also sorbs strongly on sediments

Today the ^{90}Sr, ^{137}Cs, ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Am radionuclides are the isotopes that produce the majority of the radiation dose associated with exposure to global fallout. The worldwide deposition of ^{90}Sr from global fallout was estimated to be 4.74 x 10^{17} Bq in 1970.[25] The $^{137}Cs/^{90}Sr$ activity ratio in global fallout is estimated to be 1.49.[29] Therefore, the worldwide deposition of ^{137}Cs from global fallout is 7 x 10^{17} Bq. Approximately 7.8 x 10^{14} Bq of ^{238}Pu was deposited on the surface of the earth.[30] Approximately 2.9 x 10^{14} Bq of this material was produced by atmospheric testing and the remainder resulted from the burning of the System for Nuclear Auxiliary Power (SNAP) generator. The SNAP 9A capsule re-entered the atmosphere in the Southern Hemisphere on April 21, 1964.[30] Approximately 1.20 x 10^{16} Bq of $^{239+240}Pu$ were distributed by atmospheric testing.[31] The quantity of ^{241}Am in global fallout is increasing. It will increase to 5.5 x 10^{15} Bq.[32] The increase is due to the fact that the ^{241}Pu in global fallout is undergoing radioactive decay to produce ^{241}Am.

The radiochemical analyses of the samples reported in this study were conducted in 1995 and 1996. The values for global fallout deposition listed above can be used to calculate the average activity ratio of one global fallout radionuclide relative to another global fallout radionuclide at the time the samples were analyzed. The average $^{239+240}Pu/^{90}Sr$ activity ratio in global fallout was 0.025 in 1970. Strontium-90 has a 29.1 year half life; therefore, the average $^{239+240}Pu/^{90}Sr$ activity in global fallout is calculated to be approximately 0.05 in 1995. ^{137}Cs has a half life of 30.17 years; therefore, the average $^{239+240}Pu/^{137}Cs$ activity ratio in global fallout should be approximately 0.07 in 1995. The $^{241}Am/^{239+240}Pu$ activity ratio in global fallout in 1995 is approximately 0.33. The average $^{238}Pu/^{239+240}Pu$ activity ratio in global fallout is 0.065. The $^{238}Pu/^{239+240}Pu$ activity ratio in the Northern Hemisphere was determined to be 0.036 $\pm$ 0.01.[30]

The activity ratios of one global fallout radionuclide relative to another global fallout radionuclide listed above were derived by analyzing a large number of samples collected from various locations throughout the world. They are representative of the average values for global fallout. However, they may not be representative of the global fallout that was deposited at a specific location. This phenemon is clearly illustrated by comparing the ratios of one radionuclide to another radionuclide at a specific location to the average values reported for global fallout. The activity ratio of $^{239+240}$Pu to ^{90}Sr and ^{137}Cs in global fallout in the 1970's was 0.025 and 0.038, respectively. The activity ratios of $^{239+240}$Pu to ^{90}Sr and ^{137}Cs were measured in the radioactive fallout collected in early 1970's in Tokyo to be 0.016 and 0.006, respectively. [33]

Global fallout is a complicated mixture whose isotopic composition was influenced by the type of nuclear device being tested, the location of the test, the mechanisms of atmospheric transport and diffusion processes coupled with various fractionation processes. Krey *et al.* measured the ^{240}Pu/^{239}Pu atom ratio in global fallout to be 0.176 with a standard deviation of 0.014 and ranges from 0.12 to 0.21 around the world. [34] The relative amount of strontium and cesium produced may vary from test to test. Table 8 summarizes the ^{137}Cs and ^{90}Sr fission yields of ^{235}U, ^{239}Pu and ^{238}U induced by thermal, fission spectrum and 14-MeV neutrons. [35]

Table 8 - ^{137}Cs and ^{90}Sr Fission Yields of ^{235}U, ^{239}Pu and ^{238}U Induced by Thermal, Fission Spectrum and 14-MeV neutrons

Fissioning Nuclide	Fission Product	Thermal Neutrons	Fission Spectrum Neutrons	14-MeV Neutrons
^{235}U	^{137}Cs	6.28%	6.09%	5.58%
^{235}U	^{90}Sr	5.86%	5.46%	4.32%
^{239}Pu	^{137}Cs	6.53%	6.19%	4.68%
^{239}Pu	^{90}Sr	2.12%	2.31%	2.17%
^{238}U	^{137}Cs	0%	5.46%	5.13%
^{238}U	^{90}Sr	0%	3.51%	3.34%

Cesium and strontium may fractionate from plutonium in the fireball condensation phase. ^{137}Cs and ^{90}Sr have gaseous precursors; plutonium does not. ^{90}Kr, a precursor of ^{90}Sr, has a half life of 32.3 seconds. ^{137}Xe, a precursor of ^{137}Cs, has a half life of 3.82 minutes. These half lives are significantly long relative to the cooling rate of the fireball produced by an atmospheric detonation. Therefore, the plutonium can condense in one segment of the fireball while the ^{137}Xe and ^{90}Kr remain in the gaseous phase. This effect can lead to chemical fractionation within the fallout cloud.

The global fallout may undergo additional fractionation after it is deposited on the surface of the earth. Cesium forms monovalent cations and it behaves predominantly as a conservative element, i. e., water soluble, while it is in the water column. [36] Once the cesium contacts clay minerals it behaves as a non-conservative element. Strontium is a divalent element that is more water soluble than plutonium. Plutonium can exist in the +3, +4, +5 and +6 valence states in nature. Silva and Nitsche calculate that the concentration of the plutonium species as a function of pH for a 10^{-6} M solution of plutonium in 0.01 M NaCl exposed to atmospheric CO_2 range from 10^{-10} - 10^{-13} M. [37] Americium is trivalent in natural environments. It is amphoteric and under certain conditions it is more soluble than plutonium.

Results and Discussions

Sediments

Aliquots of approximately 10-25 g of each sediment were placed in counting vials 4 cm o.d. x 2.7 cm high and counted for 3000 minutes using an intrinsic germanium detector having 81% efficiency relative to a 3" x 3" sodium iodide crystal. The results are reported in Table 9. Uncertainties are 1 sigma values derived from counting statistics of both the sample and background. Upper limits are values at which a distinct signal above background can be claimed with 95% confidence.

The matrix based quality assurance samples that were analyzed as part of our QA/QC program are also reported. The analytical results derived from the analyses of Irish Sea Sediment sample are also reported. Robert August, Jr., of the Naval Research Laboratory (NRL) provided a sample of Irish Sea sediment as a blind quality assurance sample. The Irish Sea sediment was collected in the sea off of the British Nuclear Fuels Sellafield facility in the United Kingdom.

Cesium-137 was the only anthropogenic radionuclide detected in the samples by high-resolution gamma ray spectroscopy. The ^{137}Cs contents of sediments SR1A E12, SR7 C1, SR4 E1, SR6 C3 and SR9 B1 were below our limits of detection. Naturally occurring ^{40}K, ^{212}Pb and ^{214}Pb were detected in the sediment samples. ^{212}Pb and ^{214}Pb are short-lived species in the ^{232}Th and ^{238}U decay series. For the sediments containing both ^{137}Cs and ^{40}K, there is almost 100 times more gamma and beta activity attributable to the decay of naturally occurring ^{40}K than there is gamma and beta activity attributable to the anthropogenic ^{137}Cs in the sediment samples. The average ^{40}K/^{137}Cs activity ratio for sediments containing both ^{40}K and ^{137}Cs was 90 ± 33. Hence, there is more beta and gamma activity in the sediment samples that results from naturally occurring uranium and thorium and their decay products than there is gamma and beta activity that originated from the introduction of anthropogenic radioactivity.

Table 9 - Radioactivity Detected In Sediment Samples By High Resolution Gamma Ray Spectroscopy

Sample ID	^{137}Cs Bq/kg	^{40}K Bq/kg	^{212}Pb Bq/kg	^{214}Pb Bq/kg
SR1A E12	≤ 4	593 ± 36	28.6 ± 3.4	20.7 ± 3.3
SR1B E12	6.1 ± 2.3	590 ± 41	33.8 ± 3.7	24.2 ± 3.6
SR1C E12	5.5 ± 1.6	582 ± 29	33.6 ± 3.0	26.0 ± 1.8
SR7 C1	≤ 2	671 ± 16	41.0 ± 2.5	16.7 ± 2.0
SR2A E7	11.4 ± 2.3	585 ± 41	43.9 ± 3.9	13.7 ± 3.4
SR4 E1	≤ 3	888 ± 36	62.0 ± 3.1	62.3 ± 4.4
SR3B, SO3A E5	8.2 ± 2.1	698 ± 35	41.3 ± 3.7	39.3 ± 3.9
SR6 C3	≤ 4	699 ± 42	57.0 ± 4.0	93.3 ± 5.6
SR9 B1	≤ 3	452 ± 27	22.3 ± 2.9	19.8 ± 2.8
SR10 G11	7.7 ± 1.5	500 ± 30	43.0 ± 2.6	21.5 ± 2.6
SR5A D1	3.1 ± 1.1	524 ± 21	44.1 ± 2.2	59.5 ± 3.6
SR8 B4	5.3 ± 1.8	567 ± 28	44.7 ± 3.6	22.6 ± 2.7
Barrow 1S1	7.2 ± 1.9	523 ± 55	27.4 ± 3.7	31.1 ± 5.2
Barrow 1S2	8.2 ± 1.8	507 ± 40	31.8 ± 4.9	29.5 ± 4.3
NIST SRM 4350B NIST SRM 4354	21.3 ± 1.5 39.2 ± 7.9	473 ± 24 293 ± 11	37.8 ± 2.6 33.0 ± 11.6	27.1 ± 3.3 ≤ 18
NIST SRM 4355	≤ 5.9	642 ± 58	44.0 ± 5.7	32.6 ± 5.6
Irish Sea Sediment	13.8 ± 1.0			

NIST SRM 4350B contained 21.7 Bq/Kg of ^{137}Cs on May 30, 1994. The NIST certified value was decay corrected using the 30.17 year half-life of ^{137}Cs. NIST SRM 4350B contains 554 Bq/kg ^{40}K (uncertified), 33.0 Bq/kg ^{232}Th/^{212}Pb (uncertified) and 30.8 Bq/kg ^{238}U/^{214}Pb (uncertified). All ^{212}Pb and ^{214}Pb concentrations reported in Table 3 were derived using the assumption that ^{232}Th and ^{238}U are in secular equilibrium with their decay products.

NIST SRM 4354 contained 49.9 Bq/Kg ^{137}Cs on May 30, 1994. The NIST-certified value was decay corrected using the 30.17 year half-life of ^{137}Cs. NIST SRM 4354 also contains 118 ± 15 Bq/kg of ^{60}Co. We measured 114 ± 9 Bq/kg of ^{60}Co.

NIST 4355 contained 0.25 Bq/Kg of ^{137}Cs on May 30, 1994. The NIST-certified value was decay corrected using the 30.17 year half life of ^{137}Cs. NIST SRM 4335 contains 585 Bq/kg of ^{40}K and 43 Bq/kg of ^{232}Th /^{212}Pb.

The Irish Sea Sediment datum is decay corrected March 1, 1995. The only radionuclide analysis requested for the Irish Sea sediment was ^{137}Cs.

In addition to direct counting by high-resolution gamma ray spectroscopy, aliquots of each sediment sample were dissolved and processed to produce purified cesium, strontium, plutonium and americium samples. The cesium and strontium results are reported in Table 10. The ^{238}Pu, $^{239+240}$Pu and ^{241}Am alpha activity levels measured in the sediment samples are listed in Table 11.

Table 10 - Cesium and Strontium Chemically Separated from Sediment Samples

NOAA Sample ID	^{137}Cs Bq/kg	^{90}Sr Bq/kg
SR1A E12	3.1 ± 0.5	≤ 1.2
SR1B E12	4.4 ± 0.3	≤ 0.6
SR1C E12	3.1 ± 0.2	≤ 0.6
SR7 C1	3.2 ± 0.3	≤ 0.7
SR2A E7	12.4 ± 0.9	≤ 1.2
SR4 E1	0.9 ± 0.1	≤ 0.6
SR3B, SO3A E5	9.7 ± 0.7	0.6 ± 0.3
SR6 C3	4.5 ± 0.4	≤ 0.7
SR9 B1	2.7 ± 0.3	≤ 0.7
SR10 G11	8.2 ± 1.1	≤ 1.2
SR5A D1	2.3 ± 0.2	≤ 0.5
SR8 B4	4.5 ± 0.4	0.2 ± 0.1
*NIST SRM 4354	42.5 ± 2.1	24.0 ± 1.5
***NIST SRM 4355	≤ 0.5	≤ 3.2
Reagent Blank	< 0.0002	≤ 0.1

Table 11 - Alpha Activity in Sediment Samples Measured by Alpha Spectroscopy

NOAA Sample ID	^{238}Pu Bq/kg	$^{239+240}$Pu Bq/kg	^{241}Am Bq/kg
SR1A E12	≤ 0.01	0.14 ± 0.01	0.07 ± 0.01
SR1B E12	≤ 0.06	0.27 ± 0.02	0.10 ± 0.01
SR1C E12	≤ 0.01	0.09 ± 0.01	0.09 ± 0.02
SR7 C1	≤ 0.02	0.05 ± 0.02	≤ 0.1
SR2A E7	≤ 0.03	0.52 ± 0.5	
SR4 E1	≤ 0.02	≤ 0.03	≤ 0.1
SR3B, SO3A E5	≤ 0.09	≤ 0.14	≤ 0.3
SR6 C3	≤ 0.01	0.06 ± 0.01	≤ 0.1
SR9 B1	≤ 0.02	0.14 ± 0.02	0.07 ± 0.01
SR10 G11	≤ 0.01	0.27 ± 0.02	≤ 0.3
SR5A D1	≤ 0.03	≤ 0.2	0.06 ± 0.01
SR8 B4	≤ 0.02	0.34 ± 0.03	0.11 ± 0.01
Barrow 1S1	≤ 0.02	0.41 ± 0.03	0.09 ± 0.04
Barrow 1S2	≤ 0.02	0.47 ± 0.03	0.12 ± 0.05
*NIST SRM 4354	0.32 ± 0.07	4.30 ± 0.30	1.56 ± 0.23
** NIST SRM 4355	N/A	< 0.01	≤ 0.08
Reagent Blank	≤ 0.005	≤ 0.004	≤ 0.007

* The certified values for NIST SRM 4354 are ^{238}Pu = 0.26 Bq/kg, $^{239+240}$Pu = 4.00 Bq/kg, and ^{241}Am = 1.1 Bq/kg.

** The certified values for NIST SRM 4355 are ^{238}Pu = uncertified, $^{239+240}$Pu = 0.001Bq/kg and ^{241}Am = 0.007 Bq/kg.

Plutonium-239 and ^{240}Pu emit alpha particles at essentially the same energies and cannot be resolved by alpha spectroscopy. Mass spectrometry measurement techniques measure ^{239}Pu and ^{240}Pu separately and provides an isotopic "fingerprint" that can be used to determine the origin of the plutonium. Table 12 reports the ^{240}Pu /^{239}Pu isotope ratios in sediment samples.

Table 12 - ^{240}Pu /^{239}Pu Isotope Ratios in Sediment Samples

NOAA Sample ID	**$^{240+239}$Pu Atom Ratio**
SR1A E12	0.17 + 0.02
SR1B E12	0.20 ± 0.02
SR1C E12	0.24 ± 0.07
SR7 C1	0.20 ± 0.02
SR2A E7	0.18 ± 0.02
SR4 E1	0.18 ± 0.05
SR3B, SO3A E5	0.19 + 0.05
SR6 C3	0.19 ± 0.02
SR9 B1	0.17 ± 0.04
SR10 G11	0.18 + 0.02
SR5A D1	0.19 ± 0.04
SR8 B4	0.18 + 0.02
Barrow 1S1	0.19 ± 0.03
Barrow 1S2	0.19 ± 0.02

Tables 9 - 12 provide information about the origins of the anthropogenic radionuclides detected in the Beaufort Sea sediments. It appears that the plutonium in the sediments originated as global fallout. The average $^{240+239}$Pu atom ratio measured in the Beaufort Sea sediments was 0.19 ± 0.02. Krey *et al.* measured the $^{240+239}$Pu atom ratio in global fallout to be 0.176 with a standard deviation of 0.014 and ranges from 0.12 to 0.21 around the world.[34] They measured the ^{240}Pu/^{239}Pu atom ratio in a soil sample collected at Barrow to be 0.18.[34] There is no evidence of weapons grade plutonium in the sediments. The ^{240}Pu/^{239}Pu atom ratios in samples collected at the Nevada Test Site and Semipalatinsk Nuclear Test Site range from 0.03-0.07.[38,39] The ^{240}Pu/^{239}Pu atom ratio in soil samples collected adjacent to the Rocky Flats Facility was ~0.06. [40,41] The ^{240}Pu/^{239}Pu atom ratio in a spent fuel having a burn-up of 44,400 Mega Watt Days/Ton (MWD/T) was measured to be 0.55; hence there is no evidence of plutonium that is associated with the processing of high-burn-up spent-fuel dissolver solutions.[42] The ^{238}Pu levels in the sediments were below our limits of detection. We were not able to detect the ^{238}Pu that resulted from the burn-up of the SNAP-9A capsule and deposition of global fallout.

Both ^{241}Am and $^{239+240}$Pu were detected in sediments SR1C E12, SR9 B1, SR1A E12, SR8 B4, Barrow 1S1 and Barrow 1S2. The average ratio of ^{241}Am to $^{239+240}$Pu alpha activity ratio in these sediments was 0.48 ± 0.23. This value is not statistically different than the ^{241}Am /$^{239+240}$Pu alpha activity ratio of 0.33 that is present in global fallout.

The cesium data are somewhat more difficult to interpret. Both $^{239+240}$Pu and ^{137}Cs were detected in sediment samples SR2A E7, SR1C E12, SR1B E12, SR10 G11, SR8 B4, Barrow 1S1 and Barrow 1S2. The average $^{239+240}$Pu/^{137}Cs activity ratio of these 7 sediment samples is 0.05 ± 0.02. This value could be interpreted to indicate that there was a secondary source of ^{137}Cs in addition to global fallout. However, the average

$^{239+240}$Pu/^{137}Cs activity ratio measured in the sediment samples cannot be used to conclusively prove that a second source of ^{137}Cs is present because the 0.05 ± 0.02 ratio measured in this study is not significantly different than the ~0.07 $^{239+240}$Pu/^{137}Cs ratio that is representative of present day global fallout.

The ^{137}Cs content of sediments SR2A E7, SR1C E12, SR3B, SR1B E12, SR10 G11, SR5A D1 and SR8 B4 was determined by direct counting of the sediments and by chemically separating the cesium fraction from the sediments. For these samples the direct counting of the sediment and the counting of the chemically separated cesium fraction produced comparable results. The ^{137}Cs direct count /^{137}Cs separated sample activity ratio was 1.20 ± 0.31. Chemically separating the cesium from the other radionuclides in the sediments reduces the background of the gamma ray spectrum. This process enabled us to obtain ^{137}Cs measurements on sediments SR7 C1, SR4 E1, SR6 C3, SR9 B1 and SR1A E12. Analyses of the chemically separated cesium samples enabled us to search for additional isotopes of cesium. The chemically separated and purified cesium samples were measured by both high-resolution gamma ray spectroscopy and beta counting. First, the cesium samples were analyzed by high-resolution gamma ray spectroscopy to determine the ^{134}Cs, ^{136}Cs and ^{137}Cs content. No ^{134}Cs or ^{136}Cs was detected in the samples. The ^{134}Cs and ^{136}Cs isotopes are shielded nuclides that have short half-lives. The half life of ^{134}Cs is 2.06 years. The half life of ^{136}Cs is 13.16 days. Their presence would indicate that the sediments contained materials that were recently irradiated. Next, the samples were beta counted to search for ^{135}Cs because the fission yields of ^{135}Cs and ^{137}Cs are similar. Cesium 135 does not emit gamma rays and must be detected by beta counting. No excess beta emissions were detected that could be attributed to the decay of ^{135}Cs. Review of the cesium data leads the authors to conclude that the major source of ^{137}Cs in the Beaufort Sea sediments is global fallout.

The ^{90}Sr concentrations appear to be lower than the ^{137}Cs concentrations. The amount of ^{90}Sr relative to ^{137}Cs and $^{239+240}$Pu in the samples is depleted relative to global fallout.

Fauna Samples

Ash from each biota sample was analyzed by high-resolution gamma ray spectrometry. The results are reported in Table 13.

Table 13 - Radioactivity Detected in Biological Samples by High Resolution Gamma Ray Spectroscopy

Sample ID	^{137}Cs Bq/kg	^{40}K Bq/kg	^{212}Pb Bq/kg	^{214}Pb Bq/kg
Sta B-4, Sipunculids	≤ 1.0	309 ± 6	10.4 ± 0.9	5.9 ± 0.8
Sta E-7, Crinoids	≤ 0.8	201 ± 4	≤ 0.6	≤ 0.9
Sta G-1, Sea Urchins	≤ 2.2	182 ± 20	5.6 ± 2.1	≤ 2.4
Sta G-1, Spider Crab	≤ 0.8	209 ± 25	2.5 ± 1.8	4.5 ± 0.7
Sta G-1, Regular Starfish	≤ 1.2	265 ± 8	9.9 ± 1.2	6.6 ± 1.1
Sta G-1, Soft Starfish	≤ 0.8	243 ± 5	3.9 ± 0.7	3.2 ± 0.6
Sta G-1, Nudibranchs	≤ 2.0	272 ± 19	≤ 2.5	4.3 ± 1.1
Invertebrates	≤ 1.0	295 ± 35	≤ 0.9	≤ 1.0
Broad Whitefish #1	0.3 ± 0.1	299 ± 21	2.1 ± 0.3	≤ 1.7
Broad Whitefish #2 (2 fish)	≤ 0.2	311 ± 31	≤ 2.0	≤ 2.0
Arctic Char	≤ 0.1	323 ± 19	≤ 1.9	≤ 0.9
Arctic Cod	2.6 ± 0.1	307 ± 12	≤ 1.7	≤ 0.9
Arctic Cisco #1	2.9 ± 0.2	294 ± 29	≤ 1.8	≤ 2.2
Arctic Cisco #2 (2 fish)	0.3 ± 0.1	328 ± 26	≤ 0.9	≤ 1.1
Least Cisco (4 fish)	0.9 ± 0.3	319 ± 26	≤ 1.9	≤ 2.4
Chum Salmon	0.3 ± 0.0	341 ± 19	≤ 1.4	≤ 1.0
Dolly Varden (3 fish)	0.5 ± 0.2	335 ± 28	≤ 2.0	≤ 2.5
Bowhead Whale #1 blubber	≤ 0.08	15 ± 2	0.3 ± 0.1	≤ 0.08
Bowhead Whale #1 lung	≤ 0.07	189 ± 10	≤ 0.08	≤ 0.07
Bowhead Whale #1 liver	0.4 ± 0.1	223 ± 5	0.5 ± 0.2	0.5 ± 0.3
Bowhead Whale #B8 blubber	≤ 0.01	≤ 29	≤ 0.02	≤ 0.02
Bowhead Whale #B8 muscle	≤ 0.05	370 ± 48	≤ 0.04	≤ 0.02
Bowhead Whale #B8 kidney	≤ 0.07	419 ± 36	≤ 0.08	≤ 0.04
Bowhead Whale #B8 liver	≤ 0.04	398 ± 20	≤ 0.4	≤ 0.06
King Eider bone	≤ 0.5	35 ± 5	≤ 0.5	≤ 0.5
King Eider muscle	≤ 0.4	305 ± 6	≤ 0.5	≤ 0.4
Polar Bear #1 fat	≤ 0.09	≤ 26	≤ 0.9	≤ 0.6
Polar Bear #1 muscle	≤ 0.14	341 ± 24	≤ 0.7	≤ 1.0
Polar Bear #1 kidney	≤ 0.3	329 ± 37	≤ 1.6	≤ 1.4
Polar Bear #1 liver	≤ 0.2	351 ± 50	≤ 1.1	≤ 1.0
Polar Bear #2 muscle	0.5 ± 0.4	339 ± 41	≤ 0.7	≤ 0.8
Caribou #4 rumen	24.3 ± 2.9	298 ± 31	≤ 1.5	≤ 1.4
Caribou #4 muscle	24.3 ± 2.9	355 ± 39	≤ 0.9	≤ 0.7
Caribou #4 liver	26.7 ± 2.8	321 ± 49	≤ 1.3	≤ 0.9
Caribou #4 kidney	36.2 ± 5.4	339 ± 42	≤ 1.1	≤ 1.1
Bearded Seal blubber	≤ 0.2	≤ 10	≤ 0.3	≤ 0.3
Bearded Seal kidneys	≤ 0.2	352 ± 51	0.8 ± 0.1	≤ 0.4

In addition to direct counting by high-resolution gamma ray spectroscopy, aliquots of each biota sample were dissolved and processed to produce purified cesium, strontium, plutonium and americium samples. The cesium and strontium results are reported in Table 14 and the plutonium and americium results are reported in Table 15.

Table 14 - Cesium and Strontium Chemically Separated from Biological Samples

Sample ID	^{137}Cs Bq/kg	^{90}Sr Bq/kg
Sta B-4, Sipunculids	1.1 ± 0.2	≤ 1.0
Sta E-7, Crinoids	0.7 ± 0.2	≤ 0.9
Sta G-1, Sea Urchins	0.6 ± 0.1	≤ 0.6
Sta G-1, Spider Crab	0.5 ± 0.1	≤ 0.3
Sta G-1, Regular Starfish	0.8 ± 0.1	≤ 0.5
Sta G-1, Soft Starfish	N/M	≤ 0.9
Sta G-1, Nudibranchs	0.3 ± 0.1	≤ 0.4
Broad Whitefish #1	N/M	0.6 ± 0.3
Invertebrates	0.03 ± 0.02	0.02 ± 0.01
Arctic Char	N/M	0.3 ± 0.2
Arctic Cod	N/M	≤ 0.1
Arctic Cisco	N/M	1.2 ± 0.6
Least Cisco (4 fish)	1.1 ± 0.1	1.5 ± 0.4
Chum Salmon	N/M	≤ 0.1
Dolly Varden (3 fish)	0.5 ± 0.1	0.4 ± 0.1
Bowhead Whale #1 blubber	≤ 0.00005	≤ 0.008
Bowhead Whale #1 lung	0.02 ± 0.02	0.05 ± 0.01
Bowhead Whale #1 liver	0.44 ± 0.04	0.05 ± 0.01
Polar Bear #2 muscle	0.2 ± 0.1	0.1 ± 0.1
King Eider bone	≤ 0.5	0.02 ± 0.01
King Eider muscle	0.3 ± 0.03	0.05 ± 0.02
Bearded Seal blubber	≤ 0.2	≤ 0.02
Bearded Seal kidneys	≤ 0.2	≤ 0.003

Table 15 - Alpha Activity in Biological Samples Measured by Alpha Spectroscopy

NOAA Sample ID	^{238}Pu Bq/kg	$^{239+240}$Pu Bq/kg	^{241}Am Bq/kg
Sta B-4, Sipunculids	≤ 0.01	≤ 0.03	≤ 0.08
Sta E-7, Crinoids	≤ 0.01	0.08 ± 0.02	≤ 0.06
Sta G-1, Sea Urchins	≤ 0.02	≤ 0.02	≤ 0.02
Sta G-1, Spider Crab	≤ 0.01	0.02 ± 0.01	≤ 0.02
Sta G-1, Regular Starfish	≤ 0.06	≤ 0.08	≤ 0.02
Sta G-1, Soft Starfish	≤ 0.01	≤ 0.01	≤ 0.05
Sta G-1, Nudibranchs	≤ 0.01	≤ 0.01	≤ 0.02
Invertebrates	≤ 0.005	≤ 0.005	*LIA
Broad Whitefish #1	≤ 0 0004	≤ 0.0003	≤ 0.005
Arctic Char	≤ 0.001	≤ 0.002	≤ 0.01
Arctic Cod	≤ 0.0005	≤ 0.0005	≤ 0.001
Arctic Cisco	≤ 0.0005	≤ 0.0005	≤ 0.009
Least Cisco (4 fish)	≤ 0.001	≤ 0.001	≤ 0.01
Chum Salmon	≤ 0.0001	≤ 0.0001	≤ 0.005
Dolly Varden (3 fish)	≤ 0.0007	≤ 0.0008	≤ 0.006
Bowhead Whale #1 blubber	≤ 0.004	≤ 0.003	≤ 0.0006
Bowhead Whale #1 lung	≤ 0.001	≤ 0.002	≤ 0.002
Bowhead Whale #1 liver	≤ 0.0001	≤ 0.001	≤ 0.004
Polar Bear #2 muscle	≤ 0.0005	≤ 0.0005	≤ 0.005
Caribou #4 liver	≤ 0.001	≤ 0.001	≤ 0.07
King Eider bone	≤ 0.02	≤ 0.03	LIA
King Eider muscle	≤ 0.0007	≤ 0.001	≤ 0.002
Bearded Seal blubber	≤ 0.004	≤ 0.005	≤ 0.004
Bearded Seal kidneys	≤ 0.004	≤ 0.008	≤ 0.01

* Sample lost in analysis

Very little anthropogenic radioactivity was detected in the biota samples. Cesium-137 was the radionuclide most often detected in the biota samples. Observed levels of radioactivity in animals and tissues are much lower than the screening value for human food according to U. S. Food and Drug Administration regulations. For example, the screening limit for ^{137}Cs is 370 Bq/kg.

Conclusions

The anthropogenic radionuclide content of sediments collected in the Beaufort Sea is predominantly the result of deposition of global fallout. No other sources of anthropogenic radionuclides could be conclusively identified in the sediments. Measured anthropogenic radionuclide concentrations in fish, birds and mammals were very low.

Assuming that ingestion of food is an important pathway leading to human contact with radioactive contaminants and given the dietary patterns in coastal Arctic communities, it can be surmised that marine food chains are presently not significantly affected.

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Appendix

Detailed Procedure for Isolation and Chemical Purification of Sr, Cs, Pu and Am from Biota and Sediments Collected in the Arctic

Introduction

A series of biota and sediment samples was collected by the National Oceanic and Atmospheric Agency (NOAA) as part of studies to determine the effects of releasing anthropogenic radionuclides into the Arctic environment. Samples were frozen and packed in dry ice for shipment to Los Alamos National Laboratory (LANL). The samples were dried to constant weight, ashed at 550 °C and analyzed by high-resolution gamma ray spectroscopy. Next, the samples were completely dissolved in strong mineral acids.[1,2] Plutonium, cesium and strontium were analyzed from a single aliquot of each sample. Plutonium was separated and purified by ion exchange chromatography.[3] The plutonium was electroplated on platinum and the ^{238}Pu and $^{239+240}$Pu concentrations were measured by alpha spectroscopy.[4,5] The plutonium was removed from the platinum disks and the ^{240}Pu/^{239}Pu atom ratio was measured by thermal ionization mass spectrometry.[6,7] Next, iron was removed from the sediment samples by an isopropyl ether extraction. The small amount of iron present in biota was removed as the chloro complex by anion exchange. This process was done to maximize the chemical yields for cesium and strontium. Cesium was isolated by sorption on microcrystalline ammonium phosphomolybdate.[8] The cesium was measured by low-level beta counting and high-resolution gamma ray spectroscopy. Strontium was isolated as a carbonate and purified by cation exchange chromatography.[9] The ^{90}Sr was determined by measuring the ^{90}Y daughter using low-level beta counters.

The ^{241}Am content was measured on separate ~ 5 g aliquots of ash. Americium was separated by co-precipitation with lanthanum fluoride and by HDEHP (Di-2-ethylhexylphosphoric acid in heptane) extraction.[10] Final purification was accomplished by anion exchange chromatography. The americium was quantified by solid state alpha spectrometery. Sample preparation, detailed chemical separations and purification procedures are described below.

Sample Preparation

Upon arrival at LANL the samples were thawed, transferred into tared quartz beakers and wet weights were determined to the nearest 0.01 g. Samples were dried at 110 °C until constant weights were obtained. Next, the samples were ashed in muffle furnaces. The samples were heated at 300 °C until they were charred. Then the temperature of the muffle furnaces was raised to 550 °C and the samples were ashed.

Whale blubber required a slightly different ashing procedure. The blubber decomposed into oils that were flammable. Approximately 500 g aliquots of whale blubber were placed in tared quartz beakers. The blubber was heated to 110 °C for 72 hours. The samples were cooled to room temperature and weighed. This weight was used as the dry weight for subsequent analyses. Ashing of the blubber was accomplished by raising the temperature in 50 °C increments daily until the sample was completely ashed at 550 °C.

The ashed samples were analyzed by high resolution gamma-ray spectroscopy to assess the presence of both naturally occurring and anthropogenic radionuclides. Aliquots of each sample were placed in counting vials 4 cm o.d. x 2 .7 cm high and counted for 3000 minutes on high-resolution gamma spectrometers.

Dissolution

1. Place a 15 g aliquot of the ashed biota sample in a 600 mL Teflon beaker and add 50 mL of conc. HNO_3, 50 mL of $HClO_4$, and slowly and carefully, 50 mL of HF.

   Place a 30 - 50 g aliquot of the sediment sample in a 1000 mL Teflon beaker and add 100 mL of conc. HNO_3, 100 mL of $HClO_4$, and slowly and carefully, 50 mL of HF.

2. Heat the sample on a hot-plate until strong fumes of $HClO_4$ are evolved. Cool to room temperature.

3. Add 50 mL of HF and heat again until $HClO_4$ fumes, then cool. Repeat until there is no unreacted siliceous material. After the last HF addition, fume strongly for at least 15 minutes.

4. Transfer the sample to a 1000 mL quartz beaker. Rinse the Teflon beaker with 25 mL of $HClO_4$ and combine with the sample. Add 25 mL of $HClO_4$ to the Teflon beaker and heat for 5 minutes after fumes of $HClO_4$ appear. Add the contents of the Teflon beaker to the sample. Heat the sample (in the quartz beaker) until strong fumes of $HClO_4$ appear and continue fuming for at least 30 minutes. Flame the walls of the beaker with a Meeker burner three times during the middle of the $HClO_4$ fume-down to remove any HF that has condensed on the sides of the beaker. Additional $HClO_4$ should be added if the sample becomes too thick and begins to spatter. Allow the sample solution to cool to room temperature.

5. To the cooled contents of the quartz beaker, add 2.5 mL cesium carrier, 3.0 mL strontium carrier, and 2 mL ^{242}Pu tracer and 1 mL of ^{85}Sr tracer. (For americium analysis, substitute 1 mL ^{243}Am tracer and 1 mL lanthanum carrier.)

6. Continue fuming the $HClO_4$ until sample is near dryness. Allow to cool. (For americium analysis proceed to step #62.)

Plutonium Removal

7. Add 200 mL of conc. HNO_3 to the cooled sample material. Boil gently for 10-15 minutes.

8. Cool the solution and add 200 mL of water. Warm to dissolve solids. Add 100 mg of sodium nitrite, $NaNO_2$, dissolved in 2 - 3 mL of water. Centrifuge the warm sample solution and collect any insoluble material in a 40 mL centrifuge tube. Add 3 mL of $HClO_4$ to the solids and fume to near dryness. Cool to room temperature and add 20 mL of 8 M HNO_3. The mixture is centrifuged and the leachate is added to the sample solution. Any solid residue still remaining may be discarded. At this point the solution concentration with respect to HNO_3 should be close to 8 M and the volume should not be over 600 mL.

9. Prepare a 20 mL ion-exchange column with AG MP-1 anion exchange resin and equilibrate with 75 mL of 8 M HNO_3. Pass the sample solution through the column at a rate of 3 - 4 mL/minute. Pass 20 mL of 8 M HNO_3 through the column and combine the eluate with that from the sample solution. Save the sample solution for step #37, "Iron Removal." (Note that iron must be removed to prevent the loss of strontium and cesium by co-precipitation.) Wash the column with an additional 20 mL of 8 M HNO_3 and discard the eluate.

10. Elute the plutonium into a 250 mL beaker with 80 mL of 0.5 M HCl followed by 50-60 mL of freshly prepared HI-HCl mixture.

11. Evaporate the plutonium eluate solution to near dryness. Cool, add 4 mL of conc. HNO_3 and take to near dryness again.

Plutonium Purification and Electroplating for Alpha Spectroscopy

12. Prepare a 2 mL ion-exchange column with AG MP-1 anion exchange resin and equilibrate with 15 mL of 8 M HNO_3.

13. Load the sample from step #11 onto the column using 5 mL of 8 M HNO_3. Use two additional 5 mL portions of HNO_3 to rinse the beaker. Pass the rinsings through the column and discard the eluate.

14. Elute the plutonium with 10 mL of 0.5 M HCl followed by 10 mL of 1:9 HI-HCl mixture. Evaporate the solution to dryness. Add 2 mL of HNO_3 and again evaporate to dryness. Add 1.5 mL HCl and evaporate until one drop of liquid remains.

15. Prepare a 0.8 mL ion-exchange column with AG MP-1 anion exchange resin and equilibrate with 5 mL of H_2O_2-HCl reagent.

16. Apply the sample to the column using 1.5 mL of H_2O_2-HCl reagent and wash with 6-7 mL of conc. HCl. Elute the plutonium with 6 mL of 1:9 HI-HCl mixture and evaporate to dryness. Add 1 mL of conc. HNO_3 and evaporate to dryness. Add 1.5 mL of 2 M HCl and heat to boiling. Cool and electroplate.

17. Add ~ 150 mg of high purity NH_4Cl, one drop of methyl red indicator, and the sample solution to a 3/8 inch diameter electroplating cell (containing a platinum disk which acts as both the base of the cell and the cathode). Rinse the sample container (from step #16) with 1 mL of 2 M HCl and add to the cell.

18. Adjust the pH of the electrolyte (sample solution) using NH_4OH and 2 M HCl until it is just on the acid side of the methyl red end-point. Using a rotating platinum anode, electroplate at a current density of 450 - 500 mA/cm^2 for 25 minutes. Prior to shutting off the current add 2 mL of conc. NH_4OH to the cell.

19. Wash the disk first with water, then acetone. Using a Meeker burner, flame the disk to red heat and submit for α-pulse height analysis.

Purification for TIMS Analyses

20. Place the platinum disks on top of inverted 50 mL Teflon beakers. Add 0.5 mL of HF to each disk and evaporate to dryness under a heat lamp.

21. Add 0.5 mL of conc. HNO_3 to the disk and evaporate to dryness under a heat lamp.

22. Rinse the plutonium from the disks with 8 M HNO_3 into 40 mL centrifuge tubes. Evaporate the nitric acid solutions containing the plutonium to dryness.

23. Dissolve the samples in 5 mL of 8 M HNO_3.

24. Pass the solutions through 2 mL ion-exchange columns which have been prepared with AG MP-1 anion exchange resin and equilibrated with 10 mL of 8 M HNO_3.

25. Rinse the columns with three separate 10 mL aliquots of 8 M HNO_3.

26. Elute the plutonium fractions into clean 40 mL glass centrifuge tubes by passing 10 mL of 0.5 M HCl and 10 mL of HI-HCl solution through the columns. Evaporate to dryness.

27. Add 1 mL of 0.1 M H_2SO_4 to each tube to dissolve the plutonium.

28. Load the samples onto 0.8 mL ion-exchange columns which have been prepared with AG MP-1 anion-exchange resin and pre-conditioned with two 1 mL aliquots of 0.1 M H_2SO_4.

29. Rinse the centrifuge tubes with 1 mL of 0.1 M H_2SO_4 and add the rinses to the columns.

30. Rinse the columns with two 1 mL portions of the H_2O_2-HCl reagent. Rinse the tips of the columns with a stream of deionized water. Discard the eluate.

31. Rinse the columns with two 1 mL portions of the 0.06 M HF-12 M HCl solution. Rinse the tips of the columns with a stream of deionized water. Discard the eluate.

32. Rinse the columns with three 1 mL portions of 12 M HCl. Discard the eluate.

33. Elute the plutonium into 10 mL quartz test tubes with three 0.5 mL additions of HI-HCl reagent. Evaporate to dryness.

34. Add 4 drops of 16 M HNO_3 and 4 drops of 12 M $HClO_4$ to each sample. Heat at 130° C for one hour. Raise the temperature to 180° C and continue evaporating to dryness.

35. Add 10 drops of 12 M HCl to each sample and slowly heat on a heat block until dry. Cool to room temperature.

36. Submit the samples for TIMS analyses.

Iron Removal

37. Transfer the column eluate from step #9 into the original quartz beaker and evaporate to as small a volume as possible without spattering (about 100 mL total volume).

38. Add 75 mL of conc. HCl and evaporate to near dryness.

39. Repeat step #38 twice more (or until absence of brown NO_2 gas evolution indicates complete removal of HNO_3).

40. Add 8 M HCl, as necessary, while warming to dissolve solids, and then cool to room temperature. (Up to 800 mL may be used, if necessary, for large sediment samples; 400 mL should be sufficient for smaller biota samples.) For samples containing less than 500 mg of iron proceed to step # 43a.

41. Add 175 mL of diisopropyl ether to a 500 mL separatory funnel. Add 100 mL of the sample solution to the separatory funnel and shake for 1 minute. Save the aqueous layer. Add 100 mL of water to the separatory funnel containing the diisopropyl ether solution and shake for 1 minute. Discard the aqueous iron containing layer.

42. Add another 100 mL portion of the sample solution to the funnel, equilibrate with the diisopropyl ether layer, and combine the aqueous layer with that from step #41. Again, wash the ether layer with 100 mL of water and discard the aqueous layer.

43. Repeat step #42 until the entire sample solution has been processed. If any sample remains in the original sample container (i.e. if any solid settled out prior to extraction) wash with 50 mL of 8 M HCl, extract with diisopropyl ether, and combine with the rest of the extracted sample solution. Discard any remaining solid material. Evaporate the sample solution to as small a volume as possible (50-75 mL). Go on to step #44.

43a. Prepare a 20 mL ion-exchange column with AG MP-1 anion exchange resin and equilibrate with 75 mL of 8 M HCl. Pass the sample solution through the column at the rate of 3 - 4 mL/minute. Pass 20 mL of 8 M HCl through the column and combine the eluate with that from the sample solution. Evaporate the sample solution to as small a volume as possible (50-75 mL).

Cesium Removal and Purification

44. Add 400 mL of water to the sample. Slowly add saturated Na_2CO_3 solution until CO_2 gas evolution ceases. Increase the volume to 750 mL and add more Na_2CO_3 until precipitation is complete. Warm and centrifuge, transferring the supernate to a clean 2 L beaker. Wash the carbonate precipitate once with warm 2% Na_2CO_3 solution and add to the original supernate. Set aside the precipitate for the strontium analysis (step #53).

45. Acidify the supernate pool from step #44 with HCl to pH=1 using pH test paper.

46. Add 1 g AMP-1 (microcrystalline ammonium phosphomolybdate) and stir with gentle warming for 30 minutes.

47. Allow the sample mixture to settle for one hour, decant the bulk of the supernate, and then centrifuge to collect the solid AMP-1. Discard the supernate; wash the AMP-1 with 10-20 mL of 0.1 M HCl. Centrifuge and discard the wash liquid.

48. Add 3 mL of 6 M NaOH to the solid AMP-1 and heat to dissolve. Dilute to 15 mL with water.

49. Add 9 mL of 3 M $CaCl_2$ to precipitate solid $CaMoO_4$ and adjust the pH to 7 using 6 M HCl (a little more than 1 mL will be required). Add the 6 M HCl dropwise until the precipitate adopts a very faint yellow tinge. (pH paper should indicate 5.0 - 7.5.)

50. Centrifuge and transfer the supernate (through a Whatman #541 filter paper, if cloudy) into a clean centrifuge tube. Wash the precipitate with 5 mL of water and add the wash to the sample solution.

51. Acidify the sample solution with 1 - 2 drops of 6 M HCl, add 3 mL of 0.1 M H_2PtCl_6 solution, and 5 mL of ethanol. Stir and cool in an ice bath for at least 10 minutes.

52. Centrifuge and discard the supernate. Wash the precipitate with 5 mL of 0.1 M HCl. Centrifuge and discard the supernate. Transfer the precipitate to a counting vial (rabbit) using a few mL of ethanol. Place the vial into a 40 mL centrifuge tube and centrifuge gently in order to pack the sample material into the bottom of the vial. Pour off the ethanol. Submit for γ-counting using a well-type Ge-Li detector.

Strontium Removal

53. Transfer the carbonate precipitate from step #44 to a large container (1 or 2 L beaker) with a minimum of water, and with heating, dissolve the solid by adding 3 M HCl in small increments. Adjust the acidity to not more than 0.1 M hydrogen ion concentration (pH should be in the range of 1-2). If any solid remains, warm the solution and centrifuge. Wash the solid with 10 mL of 0.02 M HCl and add the wash liquid to the sample solution. Discard any remaining solid.

54. Add 0.2 M tetrasodium EDTA to the sample solution at the rate of 50 mL for each 1 g of calcium carbonate (or equivalent: with sediment samples aluminum and manganese may be present and react in a manner similar to calcium) in the original sample. Add the EDTA solution gradually while monitoring the pH with a pH meter. The target pH is in the range of 4.8 -4.9. If the pH reaches that range and no precipitate has formed it may be assumed that sufficient EDTA has been added.

55. Prepare a 20 mL ion-exchange column with AG MP-50 cation exchange resin. Wash the column with 30 mL of water and discard the wash. (Precondition the column by passing through it 50 mL of a solution which is 1.0 M in ammonium hydroxide and 0.1 M in EDTA. Further condition the column by passing through it 30 mL of the above solution which has been adjusted to a pH of 5.) Pass the entire sample solution through the column at a rate of about 5 mL/min. Discard the eluate. Rinse the column with 50 mL of water and discard the rinse.

56. Elute the strontium from the column, with 150 mL of 5 M HCl, into a 250 mL beaker. Evaporate to about 5 - 10 mL. Add 20 mL of conc. HNO_3 and evaporate again to 20 mL.

57. Heat the sample on a hot-plate and evaporate until a precipitate just begins to form (or add water dropwise to just dissolve any precipitate when the solution is hot). Remove from heat, cool slightly, and add 30 mL of fuming (i.e. 90%) HNO_3. Cool in an ice bath and centrifuge; discard the supernate. Dissolve the precipitate in 2-3 mL of water and reprecipitate with another 20 mL of fuming HNO_3. Dissolve the precipitate again in 2-3 mL of water and γ-count for ^{85}Sr in order to determine percent recovery for strontium. Add 1 mL of standardized yttrium carrier to the sample. Allow 14 days for grow-in of ^{90}Y.

58. Dilute sample solution to 15 mL. Add 3 M NH_4OH (~3 mL) to precipitate $Y(OH)_3$, heat to near boiling to help coagulate the precipitate and centrifuge. Note the date and time as that of the yttrium/strontium separation.

59. Set the supernate aside and dissolve the precipitate in about 1 mL of 3 M HCl. Add 0.5 mL of strontium carrier, dilute to 10 mL, and reprecipitate the $Y(OH)_3$ using dilute NH_4OH. Centrifuge and combine the supernate with the solution from step #58, in case another "milking" of the strontium solution is required.

60. Wash the precipitate with 10 mL of water containing a slight excess of NH_4OH. Dissolve the precipitate in 0.5 mL of dilute HCl and dilute to a volume of 10 mL. Add a 5% solution of $(NH_4)_2C_2O_4$ in 1 mL increments until a persistent precipitate of hydrated $Y_2(C_2O_4)_3$ is obtained. Add an additional 1 mL of the $(NH_4)_2C_2O_4$ solution and heat to coagulate the precipitate. Cool and test for completeness of precipitation by adding a few additional drops of the $(NH_4)_2C_2O_4$ solution.

61. Centrifuge and discard the supernate. Resuspend the precipitate with 3 -5 mL of water and filter onto a tared 2.1 cm filter (Whatman #542 or equivalent). Wash twice with 2-3 mL portions of water and twice with 2-3 mL portions of ethanol. Dry at room temperature for 30 minutes, weigh to determine the yttrium recovery, cover with mylar film, and submit for β-counting. (Weight of yttrium oxalate obtained is typically about 33 mg, corresponding to the 9-hydrate.)

Americium

Americium analyses were simplified by dissolving separate 5 g aliquots of ash. Sample dissolution was accomplished in the same manner as for the larger Pu-Cs-Sr sample.

62. To the cooled sample resulting from dissolution step #6, add 150 mL of 10 M HCl and warm to dissolve the solid matter. Any remaining solid material may be removed by centrifugation and discarded.

63. Prepare a 20 mL ion-exchange column with AG MP-1 anion exchange resin, filled only to the 15 mL level, and equilibrate with 50 mL of 10 M HCl.

64. Pass the sample solution through the column. Rinse the column with 10 mL of 10 M HCl and add to the eluate (sample) solution. Evaporate the sample solution to 50 mL and add 100 mL of water. Warm to dissolve solids.

65. Remove undissolved material by centrifugation and transfer the solution to a 250 mL polypropylene beaker. Add 20 mL of 50% HF and let stand for 30 minutes.

66. Centrifuge and discard the supernate. Wash the precipitate with a solution of 3 M HCl / 3 M HF. Centrifuge and discard the wash.

67. Dissolve the fluoride precipitate using equal volumes of saturated H_3BO_3 solution and conc. HCl. Dilute to 20 mL and add 9 M NaOH to precipitate La/Am$(OH)_3$. The americium coprecipitates. Heat the suspension to near boiling, cool, and centrifuge.

68. Wash the precipitate with 15 mL of water, then dissolve it in 5 mL of 3 M HCl.

69. Dilute to 20 mL and add 15 M NH_4OH to reprecipitate La/Am$(OH)_3$. Centrifuge and discard the supernate. Wash the precipitate with water containing a few drops of 15 M NH_4OH. Again centrifuge and discard the supernate. Add 2 mL of HCl and evaporate to dryness in a dry-bath heating block.

70. Add about 45 mL 0.05 M HCl to the dry sample and warm to dissolve. Extract the La/Am from the sample solution into 10 mL of 0.5 M HDEHP (Di-2-Ethylhexylphosphoric acid in heptane). Wash the organic phase with 15 mL of 0.05 M HCl.

71. Back extract the La/Am using two 7.5 mL portions of 6 M HCl. Combine the two extracts and wash with 10 mL of heptane. Discard the heptane.

72. Carefully add 15 M NH_4OH to the sample solution (which is 6 M in HCl) in order to again precipitate La/Am$(OH)_3$. Centrifuge and discard the supernate. Wash the precipitate with 10 mL of water containing 2 drops of 15 M NH_4OH.

73. Prepare a 0.8 mL ion-exchange column with AG MP-1 anion exchange resin and equilibrate with 5 mL of conc. HCl. Dissolve the sample in 2 mL of conc. HCl and apply to the column. Use 6 - 7 mL conc. HCl to complete the transfer and wash the column. Save the eluate.

74. Evaporate the eluate from step #73 (or step #75) to soft dryness. Prepare a fresh 0.8 mL ion-exchange column with AG MP-1 anion exchange resin and equilibrate with 5 mL of 1:3 conc. HCl / acetone.

75. Dissolve the dry sample material in 1 - 2 mL of the 1:3 HCl/acetone mixture and pass through the column. Wash the column with 4 mL of the HCl/acetone mixture. Elute the americium with 6 mL of conc. HCl.

76. Repeat steps #74 and #75, again using a fresh column.

77. Evaporate the sample to dryness, add 1 mL of HNO_3 and evaporate to dryness. Add 1 mL of HCl and evaporate to soft dryness. Add 1.5 mL of 2 M HCl, heat to boiling, and cool.

78. Electroplate the americium in exactly the same manner as for plutonium (steps #17-#19).

79. Submit for α–spectrometric analysis.

ION-Exchange Columns

All anion exchange columns are filled with Bio-Rad macroporous anion exchange resin: AGMP-1, 50 to 100 mesh. This resin is pretreated by warming overnight in a mixture of 50% 12 M HCl and 50% Type 1 reagent grade water. It is washed 20 times with Type 1 reagent grade water and stored as an aqueous slurry in Teflon bottles.

Ion-exchange columns of three different sizes are used at various places in the procedure. Column specifications are:

- 20mL - Bio-Rad "Econo-Pac" polypropylene, filled with resin and conditioned as indicated.

- 2mL - Bio-Rad "Poly-Prep" polypropylene, filled with resin and conditioned as indicated.

- 0.8mL - Prepared by placing plugs of prewashed quartz wool into automatic pipettor tips that are 7-cm-length and approx. 7-mm-i.d., and adding resin to a depth of 2 cm.

Reagents

- Cesium carrier: 10 mg cesium/mL, added as CsCl in H_2O. Make up an aqueous solution containing 12.7 g of CsCl/L
- Strontium carrier: 10 mg strontium/mL added as $Sr(NO_3)_2$ in dilute HNO_3. Dissolve 24.15 g $Sr(NO_3)_2$ in H_2O, add 10 mL conc HNO_3, and dilute to 1L with water
- Lanthanum carrier: 1.5 mg lanthanum/mL, added as $La(NO_3)_3{\cdot}6H_2O$ in water

- Yttrium carrier: Add 2 mL of yttrium carrier. Dissolve 12.67 g of Y_2O_3 in a minimum of conc. HCl and make the solution up to a volume of 2 L with water
- ^{242}Pu tracer: 2 ng ^{242}Pu/mL in 2M HNO_3
- ^{243}Am tracer: 1 dpm ^{243}Am/mL in 2 M HNO_3
- ^{85}Sr tracer: 10 dpm ^{85}Sr/mL, add 1000 dpm of ^{85}Sr to 100 mL of water containing 2 drops of conc. HNO_3
- H_2SO_4: 0.1 M
- $HClO_4$: 12 M
- HNO_3: 16 M, 8 M
- Fuming nitric acid: 90% HNO_3
- HI: 48% (unstabilized)
- HF: 50% (29 M)
- HCl: 12 M, 10 M, 8 M, 6 M, 5 M, 3 M, 2 M, 0.5 M, 0.1 M, 0.05 M, 0.02 M
- HI-HCl mixture: 1:9 mixture, by volume, of 48% HI and 12 M HCl, freshly prepared
- H_2O_2-HCl reagent: 2 drops of 30% H_2O_2 to 10 mL 12 M HCl, freshly prepared
- HF-HCl solution: 0.06 M HF in 12 M HCl, 3M HF and 3M HCl
- NaOH: 9 M, 6 M
- $NaNO_2$: solid
- Na_2CO_3:saturated in water, 2% in water
- Diisopropyl ether: reagent grade
- Ammonium Molybdophosphate Microcrystals: Bio-Rad AMP-1, $(NH_4)_3PMo_{12}O_{40}$
- Anion exchange resin: Bio-Rad macroporous anion exchange resin: AGMP-1, 50 to 100 mesh
- Tetrasodium EDTA: 0.2 M
- NH_4Cl: solid
- Acetone: glass distilled
- Acetone: HCl mixture: 10 mL conc HCl and 30 mL acetone. Prepare fresh daily
- NH_4OH: 15 M, 6 M, 3 M
- Ethanol: 95%
- H_2PtCl_6: 0.1 M
- $(NH_4)_2C_2O_4$: 5% in water
- H_3BO_3: saturated in water
- HDEHP: (0.5 M Di-2-Ethylhexylphosphoric acid in heptane)
- Heptane:reagent grade
- $CaCl_2$: 3M

Grades used :

- For dissolution and initial separation processes nitric, hydrochloric, and perchloric acids were from GFS Co., redistilled (PPB grade). Hydrofluoric acid and other reagents were ACS grade.

- For subsequent separation, purification, and isolation steps mineral acids were from Seastar Co.; other reagents were of the highest purity generally available.