Environmental Science & Technology

Article

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A Method for Continuous ^{239}Pu Determinations in Arctic and Antarctic Ice Cores

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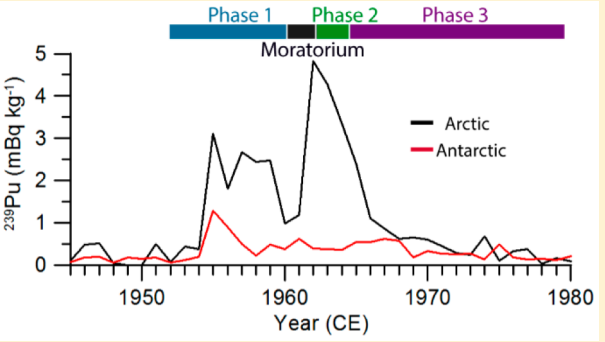
ABSTRACT: Atmospheric nuclear weapons testing (NWT) resulted in the injection of plutonium (Pu) into the atmosphere and subsequent global deposition. We present a new method for continuous semiquantitative measurement of ^{239}Pu in ice cores, which was used to develop annual records of fallout from NWT in ten ice cores from Greenland and Antarctica. The ^{239}Pu was measured directly using an inductively coupled plasma−sector field mass spectrometer, thereby reducing analysis time and increasing depth-resolution with respect to previous methods. To validate this method, we compared our one year averaged results to published ^{239}Pu records and other records of NWT. The ^{239}Pu profiles from the Arctic ice cores reflected global trends in NWT and were in agreement with discrete Pu profiles from lower latitude ice cores. The ^{239}Pu measurements in the Antarctic ice cores tracked low latitude NWT, consistent with previously published discrete records from Antarctica. Advantages of the continuous ^{239}Pu measurement method are (1) reduced sample preparation and analysis time; (2) no requirement for additional ice samples for NWT fallout determinations; (3) measurements are exactly coregistered with all other chemical, elemental, isotopic, and gas measurements from the continuous analytical system; and (4) the long half-life means the ^{239}Pu record is stable through time.



1. INTRODUCTION

The transuranic radioactive chemical element plutonium (Pu), first artificially produced in 1940 Common Era (CE), is present in the environment as a result of nuclear weapons testing (NWT) conducted from 1945 to 1980 CE.[1] Plutonium primarily exists as six isotopes: ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{244}Pu, with ^{239}Pu being the most abundant in the environment and ^{244}Pu having the longest half-life. It is estimated that 6.5 PBq of ^{239}Pu was released globally as a result of atmospheric NWT.[1]

Atmospheric nuclear weapons tests were primarily conducted in three major phases. Phase one occurred from 1952 to 1959 CE and was dominated by United States (U.S.) testing in the low latitude Pacific (Bikini, Eniwetok, and Johnston Islands) and in Nevada[1] (Figure 1). One of the largest tests conducted during this time was the Bravo test in February 1954 at Bikini Atoll, with a total yield of 15 Mt.[1] Other testing during this first period took place in the Pacific (Malden and Christmas Islands) and Australia by the United Kingdom (U.K.).[1] This period was followed by the Partial Test Ban moratorium from 1959 to 1961 CE. Phase two occurred from 1961 to 1963 CE and was dominated by testing conducted by the former Soviet Union (USSR) at Novaya Zemlya (Russian Arctic) and Semipalatinsk (Kazakhstan) (Figure 1). The largest Northern Hemisphere (NH) testing occurred over the Russian Arctic during this period, with the yield accounting for ∼57% of all atmospheric NWT.[1,2] Additional testing was conducted at the U.S. Pacific sites. In 1963 CE, the USSR and U.S. signed the Limited Test Ban Treaty in which the two countries stopped all

Received: March 3, 2016
Revised: May 26, 2016
Accepted: May 31, 2016

DOI: 10.1021/acs.est.6b01108
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

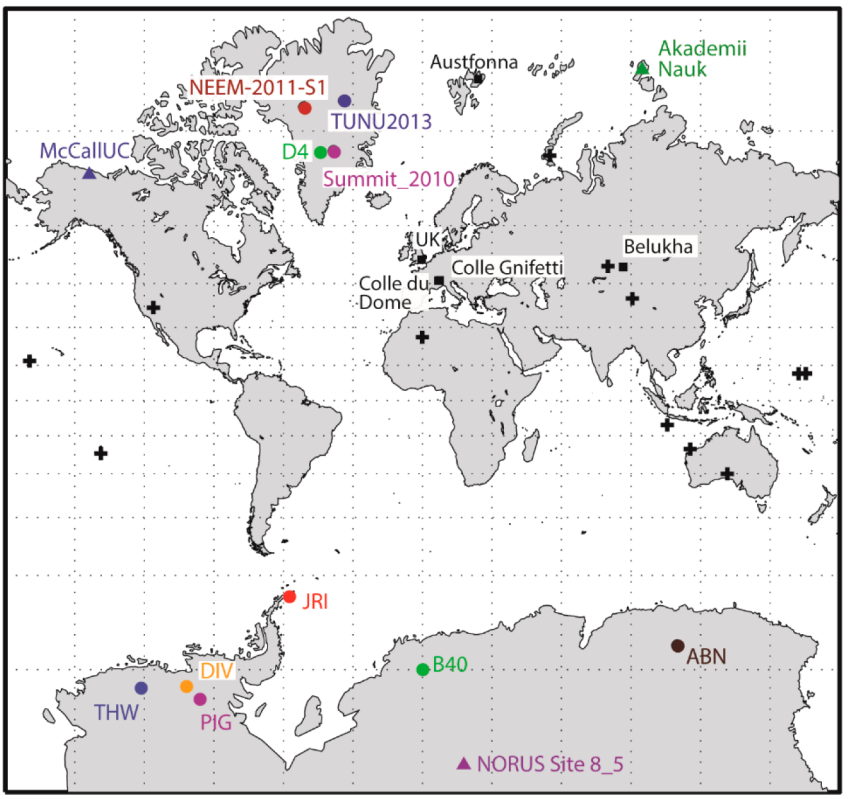


Figure 1. Ice cores analyzed in this study with well-constrained ages are shown as circles; ice cores with less constrained ages are shown as triangles. Black squares are ^{239}Pu records previously published from U.K. herbarium samples,[4] and ice cores from Austfonna,[2] Colle du Dome near Mont Blanc,[4] Colle Gnifetti,[15] and Belukha Glacier.[16] Crosses indicate sites with significant NWT.[1]

aboveground testing. Phase three was dominated by 64 aboveground tests from 1960 to 1980 CE largely conducted by France and China. French testing was conducted in the Algerian Sahara and French Polynesia (Mururoa and Fangataufa Atolls) while Chinese testing primarily was conducted in Lop Nor, western China[1] (Figure 1). Radionuclide aerosols additionally were released during the Chernobyl accident in 1986 CE.[2]

Aerosols from NWT were dispersed on local, regional (tropospheric), or global (stratospheric) scales. Aerosols emitted by NWT were partitioned depending on the altitude and size of the test as well as the local meteorology,[1] and fallout occurred during periods ranging from minutes to five years following the atmospheric tests.[3] Aerosols injected into the stratosphere, which is thermally stratified from the troposphere, had the longest residence times. Radionuclides released from testing sites were transported to high latitude ice core sites primarily in the stratosphere.[4] Radionuclides were transferred from the stratosphere to the troposphere seasonally, which in the NH occurred during the late winter to spring.[4] Removal of Pu from the atmosphere occurred either through wet (precipitation) or dry deposition,[2] and the greatest surface deposition of radionuclide aerosols was in the NH temperate latitudes with only 20% of the total fallout in the Southern Hemisphere (SH).[5]

Various chemical tracers have been utilized to reconstruct the transport and deposition of radionuclides associated with NWT (i.e., ^{3}H, ^{14}C, ^{36}Cl, ^{90}Sr, ^{137}Cs, ^{210}Pb, ^{239}Pu, ^{240}Pu/^{239}Pu, total-beta). Records of NWT have been developed from archives including vegetation and soil samples,[4,6] corals,[7,8] air filters,[3,9] lake sediments,[6,10] polar ice cores,[2,11−14] and midlatitude ice cores.[15−19] Proxies such as corals, lake sediments, and soils may exhibit postdepositional alteration, low accumulation, and mixing,[6] while ice cores typically exhibit higher annual accumulation rates and minimal postdepositional alteration or mixing. Ice cores have been successfully used to reconstruct atmospheric transport and fallout of NWT.[2,20] Measurements of ^{239}Pu also have the potential to provide specific age tie points between various ice-core and other environmental records.[11]

The chemical content of ice cores is a proxy for atmospheric aerosol composition and therefore historical changes. Because of the long half-life of ^{239}Pu (24.2 ky), the records of ^{239}Pu will be stable in ice cores through time, unlike beta-radiation-based methods. Due to the low concentration of Pu in the atmosphere and ice cores, sensitive instrumentation or large sample size is required for measurement. Traditional methods for analyzing Pu in ice cores include accelerator mass spectrometry (AMS) which requires large dedicated sample sizes (hence reduced depth and temporal resolution, typically ~3 years) and is time-consuming both for sample preparation and analysis.[16] Gabrieli et al.[15] achieved higher resolution using semiquantitative inductively coupled plasma − sector field mass spectrometry (ICP-SFMS) equipped with a desolvation nebulizer for measurements of ^{239}Pu in discrete samples from an ice core from the Swiss/Italian Alps. These measurements yielded a time resolution of 0.5−1.5 years while greatly reducing the time required for analysis.[15] Here we extend the ice core ICP-SFMS method from discrete to continuous, melter-based measurements using ICP-SFMS[21] − with the aim of minimizing sample requirements, sample handling, and decontamination efforts while maximizing depth resolution and measurement sensitivity, as well as ensuring exact depth

DOI: 10.1021/acs.est.6b01108
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registration with all other chemical, elemental, isotopic, and gas measurements. We applied this new method to an array of ten annually dated ice cores from widely spaced sites both in Antarctica and Greenland (Figure 1) to develop an annual, semiquantitative record of ^{239}Pu deposition throughout the high latitudes, and evaluate this new method through comparison to previously published discrete ^{239}Pu records. We also demonstrated the usefulness of this new method as a dating tool by applying the method to three additional ice cores from Alaska, the Russian Arctic, and Antarctica with lower confidence depth-age scales.

2. MATERIALS AND METHODS

2.1. Samples. Four Arctic and six Antarctic ice cores were analyzed for semiquantitative ^{239}Pu concentrations (Table 1, Figure 1). All 10 cores previously had been dated using annual layer counting of multiple seasonal chemical cycles in the ice, and the dating was constrained with volcanic synchronization to the time scale of Sigl et al.[22,23] The Arctic cores include D4,[24] Summit_2010, Tunu2013,[22] and NEEM-2011-S1[22] (Figure 1). The Antarctic cores are Aurora Basin North (ABN) and B40[25] from East Antarctica, James Ross Island[26] (JRI) from the Antarctic Peninsula, and Pine Island Glacier (PIG),[27] Thwaites Glacier (THW),[27] and the divide between Pine Island and Thwaites Glaciers (DIV)[27] from West Antarctica (Figure 1). Three additional ice cores from Alaska, the Russian Arctic, and Antarctica also were analyzed for ^{239}Pu and these samples, discussed in detail in section 4.3, were from sites where dating of the ice core records was confounded by very low snowfall rates or by extensive surface melt and percolation.

Table 1. Arctic and Antarctic Sites Used in This Study

site	latitude (deg)	longitude (deg)	recent accumulation kg m^{-2} y^{-1}
D4	71°24′ N	43°54′ W	414
NEEM-2011-S1	77° 26′ 56″ N	51° 03′ 22″ W	204
Summit_2010	72° 36′ N	38° 18′ W	221
Tunu2013	78° 2′ N	33° 52′ W	112
Akademii Nauk[a]	80° 31′ N	94° 49′ E	423
McCallUC[a]	69° 18′ N	143° 48′ W	546
ABN	72° 00′ S	110° 00′ E	109
B40	75° 0′ S	0°3′36″ E	68
DIV	76° 46′ 13″ S	101° 44′ 15″ W	372
JRI	64° 12′ S	57° 42′ W	595
PIG	77° 57′ 25″ S	95° 57′ 42″ W	400
THW	76° 57′ 9″ S	121° 13′ 13″ W	274
NUS_8_5[a]	82° 38′ S	17° 52′ E	35

[a]Indicates records with lower confidence depth-age scales.

2.2. Analytical Methods. ^{239}Pu and a broad range of more than 20 elements and chemical species were analyzed using the Desert Research Institute's (DRI's) continuous melter system (adapted from McConnell et al.[21]) (Figure 2). For this study, methods and results will focus on ^{238}U and ^{239}Pu. Prior to analysis, longitudinal samples with a cross section of ~0.032 by ~0.032 m from all ice cores were cut and the ends decontaminated by scraping with a precleaned ceramic knife.[21,28] The ice cores were melted continuously from bottom to top and a portion of the meltwater from the uncontaminated center of the longitudinal sample was introduced to a Thermo-Finnigan Element2 (Thermo Scientific, Bremen, Germany) ICP-SFMS approximately 4 min after melting. The continuous sample stream was acidified inline to 1% HNO_3 immediately after the melter and then to 2% HNO_3 prior to injection into the ICP-SFMS with ^{89}Y and ^{115}In added to the sample stream as external and internal standards, respectively (Figure 2). The ICP-SFMS was housed in a class-100 clean room, and the instrument outfitted with a cyclonic spray chamber and a Teflon PFA self-aspirating nebulizer (Elemental Scientific, Omaha, NE) for stable sample introduction. The entire flow system from the melter into the ICP-SFMS was acid cleaned (1% HNO_3) at least twice daily.

The ICP-SFMS instrument measured a suite of elements continuously in low resolution (M/ΔM = 300)[21] (Figure 2). There is an inherent trade-off between temporal resolution of the analyses (i.e., ice core depth resolution) and measurement sensitivity (i.e., dwell time spent on each element) (Figure 2). With increased dwell time, the number of measurements per element increases but the depth resolution decreases. For ^{239}Pu measurements, the magnet was fixed at mass 238.050 with electric scanning (E-scan) between ^{238}U and ^{239}Pu. The ^{239}Pu

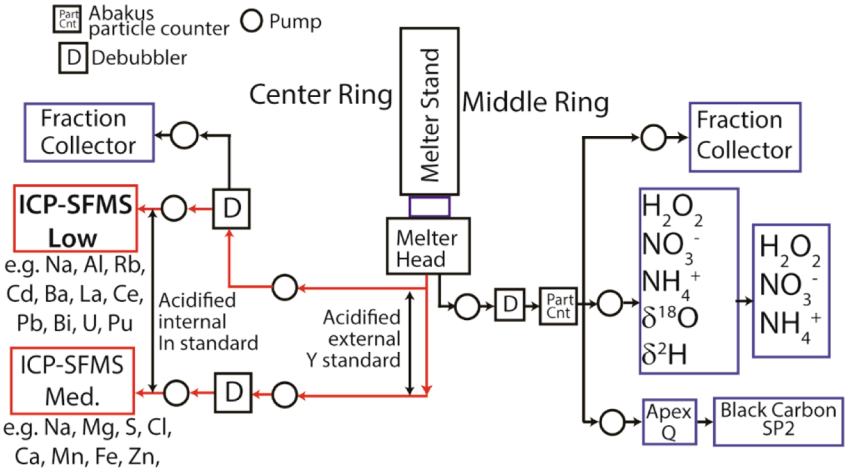

Figure 2. Schematic of the ice-core melter, ICP-SFMS (left) and continuous flow analysis[21] (CFA) (right) systems, with examples of the types of elements and chemical species analyzed. The water pumped to the ICP-SFMS is from the center of the ice, and the flow path to both ICP-SFMS instruments is highlighted in red.

DOI: 10.1021/acs.est.6b01108
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sample time was 0.4 s with 50 samples per peak (for 4 s total), and the ^{238}U sample time was 0.02 s with 50 samples per peak. Overall, instrument measurement of all elements consisted of an effective sample rate of approximately 8−10 s (~6 mm sample depth). This compromise allows for enough measurement time to be spent on ^{239}Pu to acquire robust measurements while maintaining the overall ice core depth resolution. Ice core samples were not filtered since continental dust and other particulate concentrations generally are very low in polar ice cores. Every ~2.5 h during routine pauses in the continuous ice-core analyses, procedural blanks were analyzed.

Similar to Gabrieli et al.,[15] we conducted an indirect calibration of ^{239}Pu utilizing ^{238}U so all ^{239}Pu values reported here are semiquantitative. Five standards ranging in U concentration from 0.01 to 8.0 pg g^{-1} were measured at the beginning of each analysis day with quality control standards analyzed at the beginning and end of the day. Standards were prepared from a 0.2 μg g^{-1} multielemental stock solution (Inorganic Ventures, Christiansburg, VA) in ultrapure 1% HNO_3. Using the diluted standards, we acquired a linear calibration curve and matrix matched the standards to the samples. As demonstrated by Gabrieli et al.,[15] this method provides a first approximation since the ^{239}Pu and ^{238}U ions behave similarly in the ICP-SFMS. From the semiquantitative calibration, the ^{239}Pu results were expressed in concentration and activity units, using the ^{239}Pu specific activity value from Baglan et al.[29] of 2.29×10^9 Bq g^{-1}. Depositional flux of ^{239}Pu was calculated from ^{239}Pu activity multiplied by each year's water-equivalent accumulation derived from annual-layer counting.

One potential source of interference for ^{239}Pu is $^{238}UH^+$.[15] As shown by Gabrieli et al.,[15] at low U concentrations (<40 pg g^{-1}) the $^{238}UH^+$ interferences were minimal and when interferences were detected, the interferences were much greater than ^{239}Pu measurements. In the present study, the 1940−1985 CE average U concentrations were ~0.25 pg g^{-1} for the Greenland ice cores and ~0.05 pg g^{-1} for the Antarctic ice cores. Additionally, covariability between Pu and U measurements was not observed for the Greenland or Antarctic ice cores between 1940 and 1985 CE. Even though U concentrations were very low, the individual measurements were averaged to one year averages (~40 Pu measurements $year^{-1}$) to minimize potential $^{238}UH^+$ interferences. The method detection limit of ~0.24 fg g^{-1} was derived from the blank water measurements periodically analyzed throughout the continuous analyses. The blank measurements were averaged to ~100 s intervals (~10 Pu measurements) and the detection limit calculated as three times the standard deviation. With a relatively rapid response times (>90% washout after ~60 s), memory effects for the continuous method are minimal. Blank correction was made by subtraction of the average 1900 to 1940 CE measured ^{239}Pu concentrations in the ice. The average ^{239}Pu for the ten ice cores from 1900 to 1940 CE was 0.28 fg g^{-1}.

3. RESULTS

Here we report continuous measurements of ^{239}Pu from four Arctic and six Antarctic ice cores. All cores were previously dated with annual-layer counting with age uncertainties typically ≤1 year. The ^{239}Pu data are presented as yearly averages. Composite records for activities and fluxes were calculated from the geometric mean of the annual averages.

In the Arctic, ^{239}Pu was first detected in the ice cores in 1953 CE, followed by a peak in 1955 CE, a small decline in 1956 CE, and increased values to 1959 CE (Figure 3). The 1955 to 1959

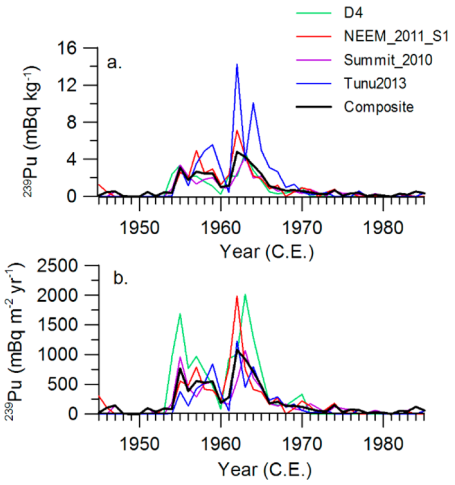

Figure 3. Annual average ^{239}Pu results from the Arctic ice cores with well-constrained ages. (a) Semiquantitative ^{239}Pu activities and (b) semiquantitative ^{239}Pu activity fluxes for each of the ice cores, with the composite geometric mean shown in black.

CE period consisted of an average ^{239}Pu semiquantitative activity of 2.5 mBq kg^{-1}. All ice cores exhibited a minimum from 1960 to 1961 CE, with an average activity of 1.1 mBq kg^{-1}. This was followed by a rapid increase in ^{239}Pu from 1962 to 1965 CE, with average ^{239}Pu values of 3.7 mBq kg^{-1}, and the greatest ^{239}Pu value was observed in the Tunu2013 ice core in 1962 CE. The ^{239}Pu levels significantly declined by 1968 CE, with values returning to background by ~1980 CE. The average standard error of the measurement from 1945 to 1985 CE was 0.6 mBq kg^{-1}. Concentrations varied between sites because wet and dry deposition processes may have differed with accumulation rates and other depositional processes, therefore the ^{239}Pu activity was converted to flux (Figure 3b). The D4 ice core had a greater accumulation rate and hence greater ^{239}Pu activity flux than the other Arctic ice cores, with an average value of 996 mBq m^{-2} yr^{-1} from 1953 to 1965 CE (Figure 3b). The average ^{239}Pu activity flux for the four Arctic ice cores from 1953 to 1965 CE was 500 mBq m^{-2} yr^{-1}.

The semiquantitative ^{239}Pu activity measurements from six Antarctic ice cores are shown in Figure 4a. Overall, ^{239}Pu levels were lower than those observed in the Arctic. Increased activities were observed from 1955 to 1957 CE (Figure 4a) with an average activity of 0.9 mBq kg^{-1} and the greatest value measured in the THW core in 1955 CE. After 1957 CE, ^{239}Pu values declined followed by a peak in 1961 CE and a second peak from 1967 to 1968 CE and a return to background by ~1975 CE. The average standard error of the measurement from 1945 to 1985 CE was 0.1 mBq kg^{-1} for the Antarctic ice cores. When accounting for accumulation rate variations, the greatest ^{239}Pu flux was observed in the DIV, PIG, and THW cores, likely because of higher accumulation rate at these sites (Table 1, Figure 4b) with an average ^{239}Pu flux of 250 mBq m^{-2} yr^{-1} from 1953 to 1965 CE. The average Antarctic ^{239}Pu flux for the six ice cores was 120 mBq m^{-2} yr^{-1} from 1953 to 1965 CE.

DOI: 10.1021/acs.est.6b01108
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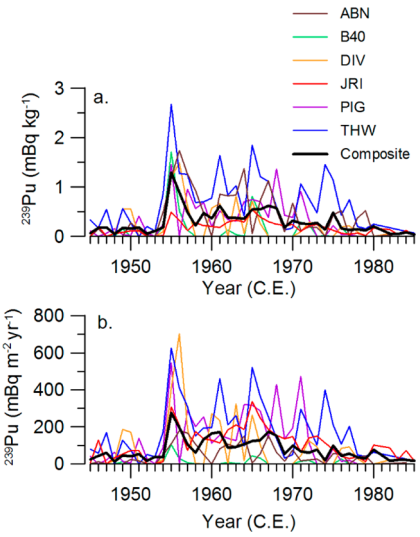

Figure 4. Annual average ^{239}Pu measurements from the Antarctic ice cores with well-constrained ages. (a) Semiquantitative ^{239}Pu activities and (b) semiquantitative ^{239}Pu activity fluxes for each of the ice cores with the composite geometric mean shown in black.

4. DISCUSSION

4.1. Comparison to Published NWT Records. To evaluate the ^{239}Pu measurements, we compared composite ice-core records of ^{239}Pu activity to published NWT fission yields[1] (Figure 5). The first significant atmospheric tests were conducted in 1952 CE and included the Mike test in Eniwetok Atoll and the 1955 CE Bravo test.[1] These tests were reflected in both the Arctic and Antarctic ice cores with the first detection of ^{239}Pu in 1953 CE and increased ^{239}Pu from 1955 to 1959 CE dominated by the U.S. tests in the low-latitude Pacific. The largest tests were conducted from 1952 to 1958 CE.[1] The Partial Test Ban moratorium resulted in a decline in ^{239}Pu, but values remained above baseline. This has been shown in other ice cores[15] and is thought to be due to the longer residence time of ^{239}Pu in the atmosphere. Postmoratorium in the fall of 1961 CE, the USSR resumed tests corresponding to a period of the most powerful testing, particularly at the Novaya Zemlya site with a test in October 1961 CE with a total release of 50 Mt and a fission release of 1.5 Mt.[1] This increase in testing clearly was reflected in the Arctic record, with the greatest measured values during the postmoratorium (post-1961 CE) period. Unlike the Arctic, where the peak ^{239}Pu concentration measurement occurred during the early 1960s, after 1958 CE the Antarctic ^{239}Pu record remained relatively low, with only a slight increase in the early and late 1960s. Although the tests conducted in the 1960s were large, there was minimal transport from the Russian Arctic to Antarctica, resulting in the low ^{239}Pu in the postmoratorium period (discussed further in section 4.2). In 1963 CE, the Limited Test Ban Treaty was signed and ^{239}Pu activity in both the Arctic and Antarctic records began to decline. Activity remained above baseline, however, as French and Chinese testing continued into the late 1970s. French testing in the South Pacific Ocean in Fangataufa and Mururoa Islands peaked in 1968 CE, which was reflected in our Antarctic records (Figure 5).

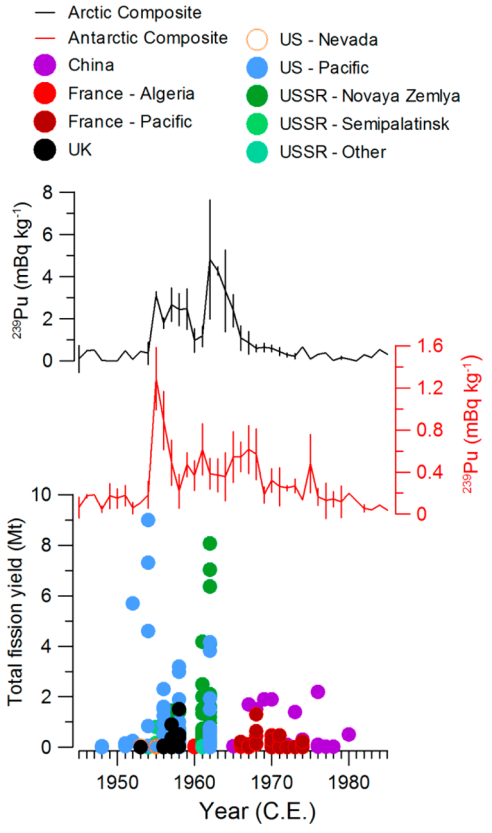

Figure 5. Arctic (black) and Antarctic (red) composite semi-quantitative ^{239}Pu activity compared to total NWT fission yields.[1] The NWT fission yields are divided by country and location of testing. Error bars represent the standard error of the mean.

From 1953 to 1980 CE, more than 500 aboveground nuclear weapons tests resulted in global fallout of ^{239}Pu. With most of the testing conducted in the NH, the NH to SH ratio of ^{239}Pu fallout[4] was ~3:1, and similarly the average ^{239}Pu activity for the Arctic and Antarctic ice cores was 1.3 and 0.4 mBq kg^{-1}, respectively.

4.2. Comparison to Previously Published Records of Fallout. We performed further evaluation of the continuous ^{239}Pu method by comparing the Antarctic and Arctic composite records to previously published discrete ^{239}Pu records. When comparing to various ice-core records, overall good agreement was observed—expected given that NWT aerosols were globally distributed—and provided greater confidence in the method (Figure 6). Results from three Greenland sites (South Dome, Camp Century, and Dye-3) showed increased ^{239}Pu from 1955 to 1960 CE with greater values from 1963 to 1965 CE.[11,30,31] The 1965 CE ^{239}Pu activity from South Dome[30,32] was 9 ± 0.3 d.p.h. kg^{-1}, similar to the average activity of 2.4 mBq kg^{-1} observed in Greenland from this study. The average value at Camp Century[32] for the 1965 CE stratum, however, was higher at 11.3 ± 0.3 d.p.h. kg^{-1}, potentially because of variations in flux. The ^{239}Pu postmoratorium (1962 to 1965 CE) to premoratorium (1955 to 1959 CE) ratio was 59:41% for Dye-3, 56:44% for South Dome,[31] and 60:40% for this study. These ratios were offset from the 70:30% determined from the total atmospheric NWT, possibly due to variations in the type of weapons tested,[31] transport, or depositional processes.

With respect to lower latitude records, the Colle Gnifetti and Colle du Dome from the Alps both show two ^{239}Pu peaks in the premoratorium period (1955 to 1959 CE) with a minimum in 1957 CE,[15] similar to the observations in the Arctic records (Figure 6). The records from UK herbarium specimens,[4] and ice cores from Belukha Glacier,[16] Colle Gnifetti,[15] and Colle du

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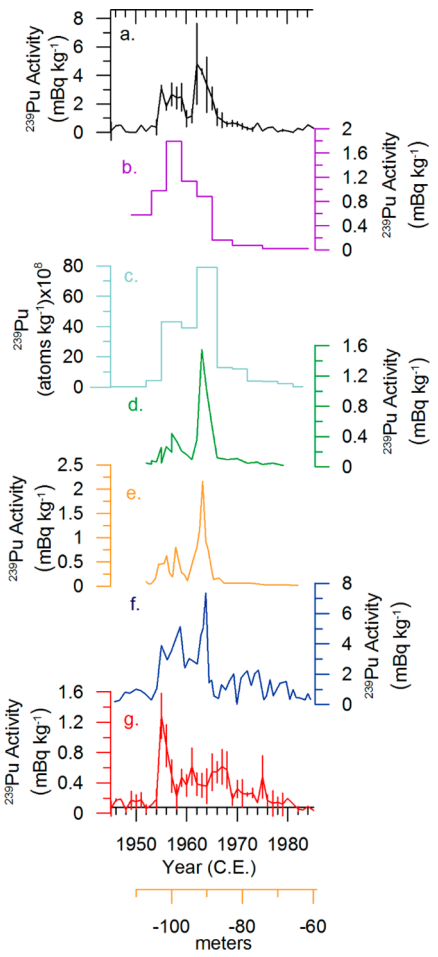

Figure 6. Comparison to previously published ^{239}Pu records. (a) Arctic composite mean (this study) (black), (b) ^{239}Pu activity from Austfonna ice core (purple),[2] (c) Belukha Glacier ice core (light blue),[16] (d) herbarium samples collected in the U.K. (green),[4] (e) Colle du Dome ice core (orange),[4] (f) Colle Gnifetti ice core (blue),[15] and (g) Antarctic composite mean (this study) (red). Note the Colle du Dome ^{239}Pu activity record is plotted on its own depth scale. Error bars are standard error of the mean.

Dome[4] all demonstrate increased ^{239}Pu activity postmoratorium (post-1961 CE).

Few studies have been conducted on Antarctic ice cores, but we observed generally favorable agreement with discrete ^{239}Pu records from Antarctica. The ^{239}Pu record from the Ross Ice Shelf showed a similar trend to the Antarctic ice-core records presented here, with the greatest ^{239}Pu values observed from 1952 to 1955 CE (~8 d.p.h. kg^{-1}),[33] slightly higher than the peak values observed in this study. This was followed by a ^{239}Pu activity peak of about half the size from 1962 to 1966 CE attributed to USSR and U.S. testing and an increase in the early 1970s attributed to French low-latitude testing.[33] Similar observations were made at Dome C with a large increase in ^{239}Pu observed in 1956 CE (34 ± 0.8 d.p.h. kg^{-1}) and significantly lower levels in the 1960s.[34] The ^{239}Pu activities from Dome C were greater than those observed here, but displayed a very similar overall trend.[32−34] The postmoratorium (1962 to 1965 CE) to premoratorium (1955 to 1959 CE) ratio for ^{239}Pu was 36:64% for Dome C, 57:43% for the Ross Ice Sheet,[31] and for this study was 38:62% (Figure 6).

4.3. Application of the Continuous ^{239}Pu Method.

Considering the favorable comparison between the well-dated ice cores to previously published discrete records, we applied our method to ice cores with lower confidence depth-age scales and compared the measurements to the Arctic and Antarctic ^{239}Pu composite records (Figure 7). Three additional cores

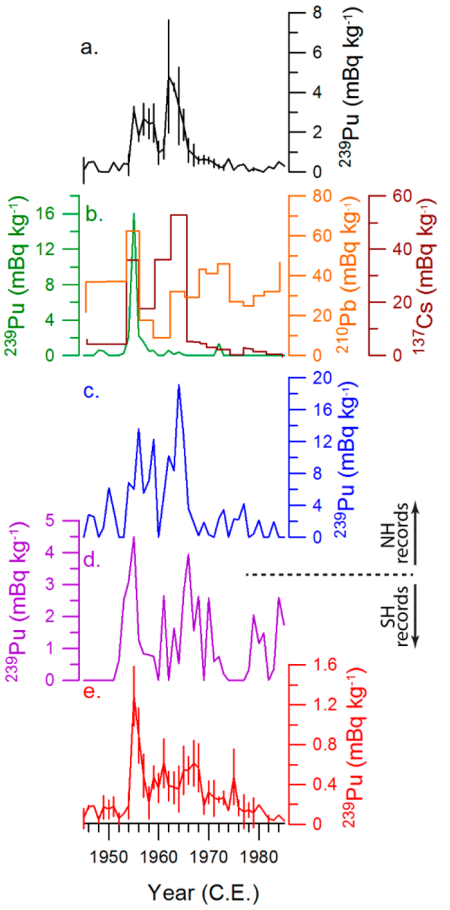

Figure 7. Comparison between ^{239}Pu activity records from well-dated ice cores and ice cores with less constrained age scales. (a) Arctic composite record, (b) Akademii Nauk ice core ^{239}Pu activity (green), (c) McCallUC ice core (blue), (d) the Antarctic NORUS site NUS_8_5 (purple), and (e) the Antarctic composite. Also shown are the Akademii Nauk ^{210}Pb activity (orange) and ^{137}Cs activity (brown) measurements from Pinglot et al.[12] Error bars are standard error of the mean.

were analyzed for ^{239}Pu from sites where very low snow accumulation and/or surface melting and percolation result in less distinct annual chemical cycles and so lower confidence depth-age scales (Table 1). These additional cores included McCall Glacier (McCallUC) from the Brooks Range, Alaska,[35] Akademii Nauk[36] from the Russian Arctic, and a Norwegian/U.S. (NORUS) traverse core site NUS_8_5 from East Antarctica[37] (Figure 1).

The Akademii Nauk results from Severnaya Zemlya (Russian Arctic), located in close proximity to the Russian Novaya Zemlya test site (Figure 1), showed increased ^{239}Pu from 1953 to 1958 CE, with a peak value of 16 mBq kg^{-1} in 1955 CE and no ^{239}Pu increase in the postmoratorium period (Figure 7b). This was similar to the ^{210}Pb measurements on the same ice core (Figure 7b) which showed a peak from 1953 to 1956 CE.[12] This is in contrast to the ^{137}Cs activity measurements

DOI: 10.1021/acs.est.6b01108
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from Akademii Nauk which peaked from 1962 to 1965 CE with a smaller increase from 1953 to 1955 CE, in agreement with NWT records[12] (Figure 7b). The Austfonna ice core record from Svalbard, sampled at a 3−5 year resolution, also contained one significant ^{239}Pu peak from 1956 to 1959 CE (Figure 6b). Considering the low Pu sampling resolution of the Austfonna record, care must be taken in the interpretation of this core. However, previous studies proposed that the deeper ^{239}Pu from Austfonna[2] and the deeper ^{210}Pb peak in Akademii Nauk[12] are due to the percolation and migration of ^{239}Pu and ^{210}Pb during surface melt periods.[2,12,36] This interpretation potentially is supported by the ^{239}Pu measurements presented here. Alternatively, the electrical conductivity and sulfate records for Akademii Nauk exhibited a sharp increase at 1956 CE thought to be associated with the Bezymianny volcanic eruption.[36] This suggests that the ^{210}Pb and ^{239}Pu records in Akademii Nauk may be impacted by tephra and other particles deposited during the volcanic event.[36] These results suggest that pronounced surface melt and percolation, as well as high amounts of volcanic fallout may impact the ^{239}Pu record.

The ^{239}Pu record from McCallUC consisted of the greatest ^{239}Pu activity measured (Figure 7c). ^{239}Pu was initially detected in 1946 CE and steadily increased to a peak in 1956 CE of 13.6 mBq kg^{-1}. This was followed by a decline in 1957 CE and a second peak in 1959 CE. Post-1961, ^{239}Pu activities increased to 19 mBq kg^{-1} in 1964 CE. Values in the McCallUC record remained elevated until 1980 CE, when values returned to baseline. While the ^{239}Pu activity was much greater in McCallUC than found in the other cores analyzed here, the overall pattern was similar to observed Greenland records, verifying the depth-age scale (Figure 7). The postmoratorium (1962 to 1965 CE) to premoratorium (1955 to 1959 CE) ratio for ^{239}Pu was 59:41% for McCallUC, also similar to the Greenland records. The McCallUC site is a high dust site potentially influenced by high northern latitude mining operations. Therefore, the greater ^{239}Pu activities measured in the McCallUC record may be impacted by the deposition of crustal dust material contaminated with ^{239}Pu or ^{238}U, suggesting that care must be taken when applying this method in high dust localities.[15]

The NORUS site NUS_8_5 is a site of very low accumulation, however the ^{239}Pu results agreed well with the composite Antarctic record providing confidence in the age dating of this core. The ^{239}Pu record showed increased semiquantitative ^{239}Pu activity from 1953 to 1956 CE and lower ^{239}Pu activity postmoratorium (Figure 7d). The semiquantitative ^{239}Pu activity was much greater than that measured at the other Antarctic sites due to the low accumulation rate (Table 1). When accounting for variations in snowfall rates, the 1955 CE ^{239}Pu activity flux was 130 mBq m^{-2} yr^{-1} for Site 8_5, lower than the average 277 mBq m^{-2} yr^{-1} observed in Antarctica.

4.4. Environmental Application. These results demonstrate the capabilities of the continuous ICP-SFMS ^{239}Pu method when applied to ice cores. Here we produced two high latitude composite records of ^{239}Pu applicable to the future evaluation and synchronization of ice cores chronologies, particularly for hard to date ice cores. While this method should be used with caution in high dust regions because of isobaric interferences from high U levels, our method for semiquantitative ^{239}Pu determinations provides an age constraint without the need for additional ice analyses. The continued application of this new method to a wide range of ice cores from varying localities may additionally shed light on lower latitude atmospheric aerosol sources and transport processes to the high latitudes, for example the transport of sulfur from volcanic eruptions. Assuming a similar distribution pattern between the radioactivity from nuclear bomb tests and sulfate injected into the stratosphere by explosive volcanic eruptions[38] a more comprehensive array of coregistered measurements of Pu and sulfur will also benefit efforts to estimate stratospheric aerosol loading in Earth's past atmosphere, necessary information to reconstruct global volcanic forcing from polar ice-core records.[22,39,40]

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Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

We thank those who aided with the organization, drilling, processing, and analysis in the field and laboratory. We acknowledge L. Layman for his initial laboratory work and R. Kreidberg for his editorial advice. We also thank the three anonymous reviewers for their constructive feedback. The following US NSF PLR grants supported collection and analyses of the cores as well as interpretation of the measurements: 0538416, 0856845, 0968391, 0909541, 1023672, 1023318, and 1204176. Ice-core data (^{239}Pu) for all ice cores are accessible at the NSF Arctic Data Center (https://arcticdata.io/).

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DOI: 10.1021/acs.est.6b01108
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