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A new ground-level fallout record of uranium and plutonium isotopes for northern temperate latitudes

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Abstract

Plutonium and uranium isotope ratios can be used to differentiate the sources of nuclear contamination from nuclear weapon establishments (Environ. Sci. Technol. 34 (2000) 4496; Internal Report for AWRE Aldermaston, UK (1961)), weapon fallout (Geochim. Cosmochim. Acta 51 (1987) 2623; Earth Planet. Sci. Lett. 63 (1983) 202; Earth Planet. Sci. Lett. 22 (1974) 111; Geochim. Cosmochim. Acta 64 (2000) 989), reprocessing plants, reactor or satellite accidents (Science 105 (1979) 583; Science 238 (1987) 512) and in addition they provide markers for post-1952 geochronology of environmental systems. A good record of plutonium and uranium isotope ratios of the background resulting from atmospheric nuclear testing is essential for source characterisation studies. Using recently developed mass spectrometric techniques (J. Anal. At. Spectrom. 16 (2001) 279) we present here the first complete records between 1952 and the present day of northern temperate latitude $^{240}Pu/^{239}Pu$ and $^{238}U/^{235}U$ atom ratios for atmospheric deposition. Such information was not derived directly during the period of atmospheric testing because suitable mass spectrometric capability was not available. The currently derived records are based on an annual herbage archive and a core from an Alpine glacier. These studies reveal hitherto unseen fluctuations in the $^{238}U/^{235}U$ atmospheric fallout record, some of which are directly related to nuclear testing. In addition, they also provide the first evidence that plutonium contamination originating from Nevada Desert atmospheric weapon tests in 1952 and 1953 extended eastwards as far as northwestern Europe. The results presented here demonstrate that we now have the capability to detect and precisely identify sources of plutonium in the environment with implications for the development of atmospheric transport models, recent geochronology and environmental studies.

Keywords: plutonium; uranium; isotope ratios; weapon fallout; multi-collector ICP-mass spectrometry; Rothamsted grass archive

1. Introduction

The radioactive debris from nuclear explosions (Figs. 1 and 2) was partitioned into the troposphere and stratosphere according to its particle size and the power of the explosion. The subsequent fallout occurred on a time scale of minutes through to years. The amount of debris produced and its particle size distribution depended on the explosive yield of the device and its height above ground when detonated [9]. Fine debris from small yield tests (<100 kt TNT equivalent) produced fallout that was expected to have a maxi-

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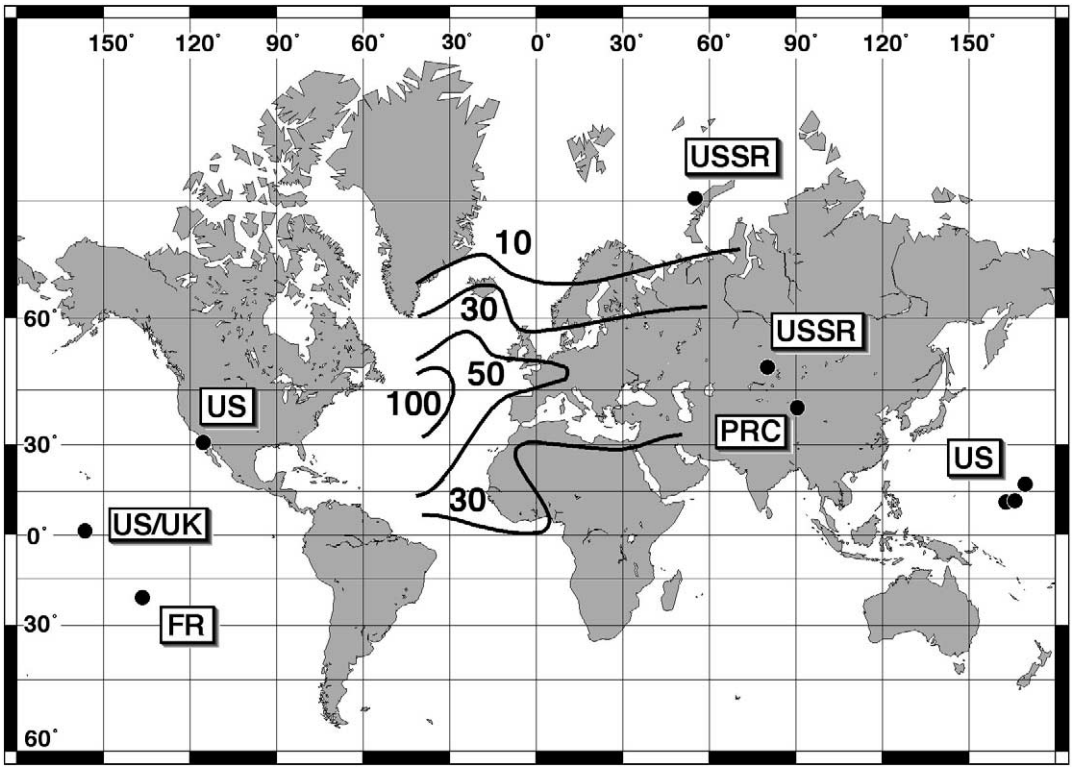

Fig. 1. Locations of the main atmospheric test sites. The contour lines show the radioactivity distribution of the Nevada Test Site fallout in 1953 in relative units [9].

mum residence time of 70 days in the troposphere [10]. Fallout from large yield tests (> 500 kt TNT equivalent), however, was almost wholly derived from material injected into the stratosphere and, though falling mostly in the hemisphere of injection, was distributed globally and deposited over several years. A typical residence time for aerosols in the stratosphere is 15–18 months [11]. The transfer of fallout from the stratosphere to the troposphere is seasonally modulated and for the

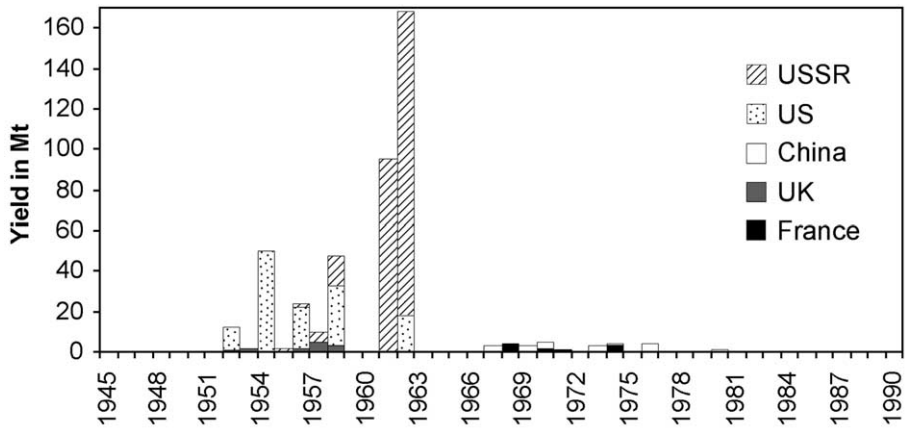

Fig. 2. Annual yields of atmospheric tests greater than 500 kt [21,22] for all years of testing (1945–1980).

Northern Hemisphere occurs mostly in the late winter and spring with little fallout occurring during summer and autumn. At ground level the maximum fallout, termed the Spring Peak or Spring Maximum [11] occurs between March–June with the main stratosphere–troposphere exchange occurring roughly 1 month before. This input of stratospheric material into the troposphere occurs by seasonal adjustment of the tropopause at temperate to high latitudes, by the Hadley circulation at around 30° and by the Jetstreams at around 30° and 60°. The first process accounts for the exchange of about 10% of the total stratospheric mass per year and the Jetstreams and Hadley circulation account for about 50–60% [11]. Existing records of nuclear fallout, particularly for plutonium, were either derived from sampling of stratospheric air by aircraft or from Polar ice cores and therefore may not represent the ground level fallout seen in temperate latitudes. Detailed measurements of other radionuclides such as ^{90}Sr were made during the testing period and in New York, for example, determinations were made in rainwater going back to 1983 [12–15]

The two primordial isotopes of uranium are ^{235}U and ^{238}U with $^{238}U/^{235}U_{PRESENT} = 137.88$ [16] and this atomic abundance ratio has no significant variation in nature except in fossil natural reactors [17]. Deviations from this ratio in environmental materials can therefore only be explained by the addition of technologically modified uranium (Table 1) [1]. Although uranium was a common component of many nuclear weapons there are no published accounts of $^{238}U/^{235}U$ in weapon fallout.

Plutonium, unlike uranium, is virtually entirely anthropogenic [18] in origin and its main isotopes found in the environment, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu, are derived from civil and military sources (Table 2). Approximately 6 tonnes of ^{239}Pu were introduced into the environment from 541 atmospheric weapon tests [19] with a total explosive yield of 440 megatons (TNT equivalent), with 25 tests accounting for two thirds of the yield. Fallout was distributed globally at a ~3:1 ratio between the Northern Hemisphere and the Southern Hemisphere.

Table 1
$^{238}U/^{235}U$ (atom ratio) for different sources

Type of uranium	$^{238}U/^{235}U$ atom ratio
Natural	137.88
Reactor	15–32 before burn-up
Weapon grade	<0.1
Depleted	250–500

Table 2
$^{240}Pu/^{239}Pu$ atom ratios for different sources [28]

Source	$^{240}Pu/^{239}Pu$
Integrated weapon test fallout	0.18
Weapon production	0.01–0.07
Chernobyl accident	0.40
MAGNOX reactor (GCR)	0.23[a]
Pressurised heavy water reactor (PHWR)	0.41[a]
Advanced gas-cooled reactor (AGR)	0.57[a]
Pressure tube boiling water reactor (RBMK)	0.67[a]
Boiling water reactor (BWR)	0.40[a]
Pressurised water reactor (PWR)	0.43[a]

[a] After fuel burn-up.

The current work, though now of wider interest and importance, was initiated by a recent investigation of the alleged nuclear accident at the former USAF Greenham Common airbase in Berkshire, UK in 1958 [1,7]. A confidential Cold War study [2] was leaked to the UK media in 1996 that reported a small excess of ^{235}U in environmental samples collected around the Greenham Common airbase. One aspect of the study by Croudace et al. [1] was to determine if the small amounts of enriched U reported by Cripps and Stimson could have originated from weapon fallout rather than from a weapon accident. To do this 30 years after the incident would have required knowledge of $^{238}U/^{235}U$ in 1957–58 weapon fallout in southern England. The results found in this study now make this assessment possible.

2. Methods

The IACR Rothamsted (Harpenden, UK) archive is a unique herbage collection with continuity from 1843 until now of some 20 000 samples of crops and soil taken from fully documented field experiments. The Rothamsted archive includes summer and autumn harvested herbage samples

Table 3
Measurement reproducibilities for $^{240}Pu/^{239}Pu$ atom ratios [28]

Pu concentration	$^{240}Pu/^{239}Pu$	±	2 S.D. %	$n=$	Average in-run 2 S.E. %
100 fg ml^{-1}	0.2265	0.0031	1.36	12	1.44
500 fg ml^{-1}	0.2263	0.0021	0.94	13	0.66
1–5 pg ml^{-1}	0.2262	0.0010	0.45	22	0.32
5–10 pg ml^{-1}	0.2262	0.0007	0.32	7	0.18
100 pg ml^{-1}	0.2262	0.0003	0.11	10	0.12

A test sample was purified from Irish Sea Sediment.

and the existence of samples from both harvests is important because they provided an ideal means of identifying the source and timing of some nuclear tests due to the 'Spring Peak'. The first harvest samples are expected to include stratospheric fallout (from the 'Spring Peak') while the second harvest should mostly include tropospheric fallout from summer tests or residual stratospheric fallout in the same year. Fifty-gram samples of herbage were made available from the Rothamsted archive from 1945 through 1990 and were used for determinations of ^{137}Cs and $^{239+240}Pu$ specific activities (Bq/kg) and $^{238}U/^{235}U$ and $^{240}Pu/^{239}Pu$ atom ratios.

The samples were first ashed then γ counted for ^{137}Cs specific activities using an HPGe well-type γ spectrometer. After γ counting a ^{242}Pu spike was added to the ashed samples followed by a double *aqua regia* acid digest. The separated liquid from the two leaches was evaporated and redissolved in 5–10 ml 8 M HNO_3 with one drop of concentrated HCl. Two ion exchange columns were prepared for each sample. The first was a 6×0.7 cm i.d. column of Eichrom 1-X8 anion exchange resin (100–200 mesh) and the second a 2×0.7 cm i.d. column of Eichrom UTEVA™ resin. The UTEVA™ column was placed immediately below the anion exchange column and both columns were pre-conditioned with 10 ml 8 M HNO_3. The sample was transferred to the anion exchange column and the eluent passed directly onto the UTEVA™ column. The two columns were washed with 20 ml 8 M HNO_3 followed by 30 ml of 3 M HNO_3, and then separated. The anion column was washed with 30 ml 3 M HNO_3 followed by 25 ml 9 M HCl to remove thorium. Plutonium was then eluted with 50 ml of freshly prepared 1.2 M HCl/H_2O_2 (50:1). The UTEVA™ column was washed with 10 ml 3 M HNO_3 followed by 10 ml 9 M HCl. The uranium was eluted with 10 ml 0.02 M HCl. Since the ^{238}U forms a hydride that interferes with the mass spectrometric measurement of ^{239}Pu the following additional separation of the plutonium is required. The eluent from the first Pu column was concentrated to 2 ml 9 M HCl, one drop H_2O_2 and 7 ml concentrated HCl were added to the concentrate before loading it onto an 4×0.7 cm i.d. anion exchange pre-conditioned with 9 M HCl. The column was washed with 10 ml 9 M HCl, 40 ml 7.2 M HNO_3 and 15 ml 9 M HCl. Plutonium was then eluted with 25 ml of freshly prepared ammonium iodide reagent (0.1 M NH_4I/9 M HCl). The eluent was evaporated to dryness and the residue was treated with *aqua regia* and concentrated HNO_3, before dissolving the residue in 2% HNO_3 for mass spec-

Table 4
Accuracy for $^{240}Pu/^{239}Pu$ atom ratios

	Certified $^{240}Pu/^{239}Pu$	Measured $^{240}Pu/^{239}Pu$	
		0.5 ppt	5 ppt
NBL122	0.1320	0.1318 ± 0.001 ($n=4$)	0.1321 ± 0.0001 ($n=3$)
NBL 126	0.0209	0.0211 ($n=1$)	0.0204 ($n=1$)
NBL 128	0.0007	–	0.0007 ($n=1$)
UK-Pu-5	0.9662 ± 0.0011	–	0.9645 ± 0.0013 ($n=7$)

NBL, New Brunswick National Laboratory; UK-Pu-5, AEA Technology.

Table 5
Variations in $^{240}Pu/^{239}Pu$ for the twin Rothamsted grass harvests (1945–1957)[a]

Year	Stratospheric input	Potential tropospheric input		Jun–Jul Rothamsted harvest			Sep–Nov Rothamsted harvest		
	Global fallout[b] $^{240}Pu/^{239}Pu$	Kazakhstan Tests USSR (yield)	Nevada Desert Tests USA (yield)	Date of harvest	$^{239,240}Pu$ (Bq/kg)	$^{240}Pu/^{239}Pu$	Date of harvest	$^{239,240}Pu$ (Bq/kg)	$^{240}Pu/^{239}Pu$
1945	–	–	16 Jul (19 kt)	Spring	–	–	Autumn	–	–
1951	–	24 Sep (38 kt)	–	21–23 Jun	–	–	2–5 Oct	–	–
1952	–	–	1 Apr–5 Jun (104 kt)	19–23 Jun	0.05	0.060	22–23 Sep	–	–
1953	0.33–0.34	12 Aug–10 Sep (440 kt)	17 Mar–4 Jun (253 kt)	21–23 Jul	0.04	0.154	19–23 Nov	0.08	0.135
1955	0.25–0.27	29 Jul–5 Aug (15 kt)	18 Feb–15 May (163 kt)	28–30 Jun	0.26	0.273	15–20 Sep	0.05	0.295
1957	0.19–0.22	8 Mar–16 Apr (1142 kt) 22 Aug–13 Sep (526 kt)	28 May–7 Oct (340 kt)	18–19 Jun	0.19	0.165	24–26 Sep	0.44	0.113

[a] Only for the years where samples from both cuts were available.
[b] Based on Arctic and Antarctic ice core measurements [23].

trometry. A Micromass IsoProbe™ multi-collector ICP-mass spectrometer was used to measure $^{242}Pu/^{239}Pu$, $^{240}Pu/^{239}Pu$ and $^{238}U/^{235}U$ atom ratios. $^{238}U/^{235}U$ can be measured with a reproducibility of $<0.1\%$ (2 S.D.) on the Isoprobe. For the more demanding case of Pu measurements a precision of better than 1% is achievable for 500 fg Pu (equivalent to an activity of 1.5 mBq; Tables 3 and 4). The Pu solutions generally had concentrations between 0.2 and 15 pg/ml and Pu specific activities were calculated from the mass spectrometer 242/239, using the measured 240/239 [8].

Also investigated was a 116-m ice core from Dome du Gouter (4300 m), Mont Blanc, France, which was shown by Vincent et al. [19] to have a good ^{137}Cs fallout record. Alpine Ice sample preconcentrates were received as ion exchange filter papers [20] which were first ashed and then leached with *aqua regia*. The same chemical procedure described earlier was then used to extract and purify uranium and plutonium prior to mass spectrometry. The chronology for the ice core was established by fitting the presented plutonium data to the Rothamsted grass and previously reported data. A linear age–depth relation was assumed for the ice core.

3. Results and discussion

Results of the current study (Tables 6 and 7) were compared with records of device testing and contemporary radiochemical monitoring of fallout [21–25]. The profiles for ^{137}Cs and $^{239+240}Pu$ activities in the Rothamsted grass (Fig. 3A,B) are very similar to these records of fallout and reflect the yields of atmospheric testing (Fig. 2), if a 1-year transit time for stratospheric deposition is assumed. The data variations reveal clear evidence for the earliest US thermonuclear tests at Eniwetok atoll in 1952, the cessation of atmospheric testing by the USA, UK and the USSR in 1963 and the 1986 Chernobyl accident (Fig. 3A).

The first measurable $^{240}Pu/^{239}Pu$ in the Mont Blanc ice core at 103.65–100.52 m agrees with the grass ratio in 1955 (Fig. 4B). Using a linear age–depth relation in the ice core the $^{240}Pu/^{239}Pu$

atom ratios in the Alpine ice and the Rothamsted grass are almost identical for the period 1955–1970. The good agreement in the data for both areas indicates that the $^{240}Pu/^{239}Pu$ record is representative of northern temperate latitudes. This conclusion may not hold when particular areas have relatively significant components of non-stratospheric fallout (e.g. Rothamsted in 1952).

Table 6
Radionuclide data for grass samples from the Rothamsted archive

Year	Sampling	^{137}Cs (Bq/kg)	$^{239,240}Pu$ (Bq/kg)	$^{240}Pu/^{239}Pu$ atom ratio	$^{238}U/^{235}U$ atom ratio
1945	n.r.	b.d.	b.d.	b.d.	137.78
1945	n.r.	12.9	b.d.	b.d.	n.a.
1946	n.r.	b.d.	b.d.	b.d.	n.a.
1950	13–15 Jun	b.d.	b.d.	b.d.	137.78
1951	21–23 Jun	b.d.	b.d.	b.d.	137.78
1951	2–5 Oct	b.d.	b.d.	b.d.	n.a.
1952	19–23 Jun	b.d.	0.05	0.060	137.26
1952	22–23 Sep	b.d.	b.d.	b.d.	n.a.
1953	21–23 Jul	9.2	0.04	0.154	137.46
1953	19–23 Nov	24.3	0.08	0.135	n.a.
1954	24–26 Jun	17.7	0.07	0.306	137.98
1955	28–30 Jun	36.3	0.26	0.273	137.78
1955	15–20 Sep	48.4	0.05	0.295	n.a.
1956	11–12 Jul	40.2	0.27	0.241	137.70
1957	18–19 Jun	32.1	0.19	0.165	137.77
1957	24–26 Sep	55.6	0.44	0.113	n.a.
1958	7–8 Jul	70.7	0.34	0.166	137.14
1959	15–16 Jun	63.3	0.21	0.175	137.34
1960	16 Jun	39.6	0.16	0.183	137.56
1961	20 Jun	22.7	0.10	0.198	137.68
1962	13 Jun	81.5	0.36	0.162	137.15
1963	27 Jun	257.0	1.55	0.221	137.50
1964	26–30 Jun	209.0	1.00	0.201	n.a.
1965	28–29 Jun	122.6	0.53	0.188	n.a.
1966	7 Jun	66.1	0.12	0.202	137.72
1967	12 Jun	33.6	n.a.	n.a.	n.a.
1968	11 Jun	34.2	0.10	0.208	137.79
1969	9–10 Jun	18.6	n.a.	n.a.	n.a.
1970	15 Jun	28.3	0.11	0.205	n.a.
1971	24 Jun	31.2	n.a.	n.a.	n.a.
1972	15–16 Jun	25.5	0.05	0.169	137.92
1973	13 Jun	12.9	b.d.	b.d.	n.a.
1974	20–21 Jun	16.0	0.06	0.215	n.a.
1975	9–10 Jun	15.7	0.03	0.232	138.03
1976	9 Jun	7.4	b.d.	b.d.	137.98
1977	21 Jun	11.4	0.05	0.262	138.34
1978	19–20 Jun	14.1	n.a.	n.a.	138.30
1979	19–20 Jun	9.0	0.02	b.d.	137.98
1980	4 Jun	6.1	b.d.	b.d.	n.a.
1981	9–10 Jun	13.3	b.d.	b.d.	137.91
1983	15 Jun	5.7	b.d.	b.d.	138.23
1985	1 Jul	4.8	b.d.	b.d.	137.88
1986	12 Jun	37.6	b.d.	b.d.	138.03
1987	29 Jun	9.6	b.d.	b.d.	137.85
1988	14–15 Jun	7.9	b.d.	b.d.	n.a.
1990	2 Jul	3.4	b.d.	b.d.	n.a.

The profiles of $^{240}Pu/^{239}Pu$ for the ice core and particularly the Rothamsted grass hold a good record of nuclear testing and also permit identification of some tests. One of these sources is the Nevada Desert Test site where 84 atmospheric nuclear test were carried out between 1951 and 1962 [21,22] (Table 5). A 1953 air sampling study of NTS-derived radioactive fallout showed north-

Table 7
Radionuclide data for the Mont Blanc ice core

Plutonium				Uranium	
Depth (m)	$^{239,240}Pu$ (mBq/kg)	$^{240}Pu/^{239}Pu$	Ice weight (kg)	Depth (m)	$^{238}U/^{235}U$
−42.65	0.017	n.a.	99.73	−51.532	137.61
−54.14	0.004	b.d.	38.25	−54.14	138.22
−64.21	0.018	b.d.	46.51	−55.2756	138.49
−68.42	0.019	b.d.	22.70	−56.4111	137.36
−73.37	0.019	b.d.	19.41	−59.25	139.03
−77.82	0.060	0.186	22.17	−64.21	138.20
−85.22	0.061	0.205	24.90	−68.42	138.70
−86.43	0.166	0.202	4.66	−73.37	135.74
−88.12	0.140	0.214	4.62	−77.82	137.11
−88.81	0.383	0.213	1.72	−85.22	138.17
−89.7	0.802	0.206	2.20	−85.82	133.05
−90.28	0.907	0.232	1.92	−86.43	132.26
−91.16	2.165	0.247	2.05	−87.32	134.08
−92.02	1.141	0.217	2.53	−88.12	132.17
−92.8	0.794	0.221	2.65	−88.81	131.14
−93.66	0.578	0.165	2.64	−90.28	131.95
−95.55	0.108	0.191	5.16	−91.16	132.49
−96.36	0.240	0.181	2.78	−92.02	128.77
−97.19	0.271	0.177	2.82	−92.8	137.53
−97.96	0.540	0.159	2.11	−93.66	131.45
−98.77	0.797	0.161	2.54	−94.6	133.68
−99.69	0.183	0.160	2.17	−95.55	134.43
−100.52	0.275	0.184	2.41	−96.36	133.51
−101.36	0.627	0.255	2.35	−97.19	133.83
−102.14	0.460	0.255	6.01	−97.96	132.14
−103.63	0.453	0.261	2.42	−98.77	131.13
−104.51	0.159	b.d.	2.25	−99.69	128.29
−105.19	0.080	b.d.	2.55	−101.36	132.67
−105.94	0.042	b.d.	3.06	−102.89	135.83
−106.4	0.038	b.d.	2.72	−103.63	131.72
−107.06	0.089	b.d.	2.49	−104.51	131.99
−107.61	b.d.	b.d.	2.68	−105.94	134.13
−108.46	b.d.	b.d.	1.67	−107.06	134.70
−109.14	b.d.	b.d.	1.35	−109.14	134.84
−109.96	b.d.	b.d.	1.76	−109.96	134.58
−110.83	b.d.	b.d.	1.33	−111.66	133.58
−111.66	b.d.	b.d.	1.86	−112.35	135.42
−112.35	b.d.	b.d.	1.47	−113.05	134.61
−113.05	b.d.	b.d.	1.28	−113.85	134.05
−113.85	b.d.	b.d.	1.48	−114.65	135.33
−114.65	b.d.	b.d.	1.42		
−115.16	b.d.	b.d.	1.88		

b.d., < detection limit (see detection limits); n.r., not recorded; n.a., not analysed (either due to selection or to analytical problems).
2 S.D. uncertainties: $^{239,240}Pu$ activity: < 1%rel, $^{240}Pu/^{239}Pu$: < 1% rel, ^{137}Cs: 5–20% rel.

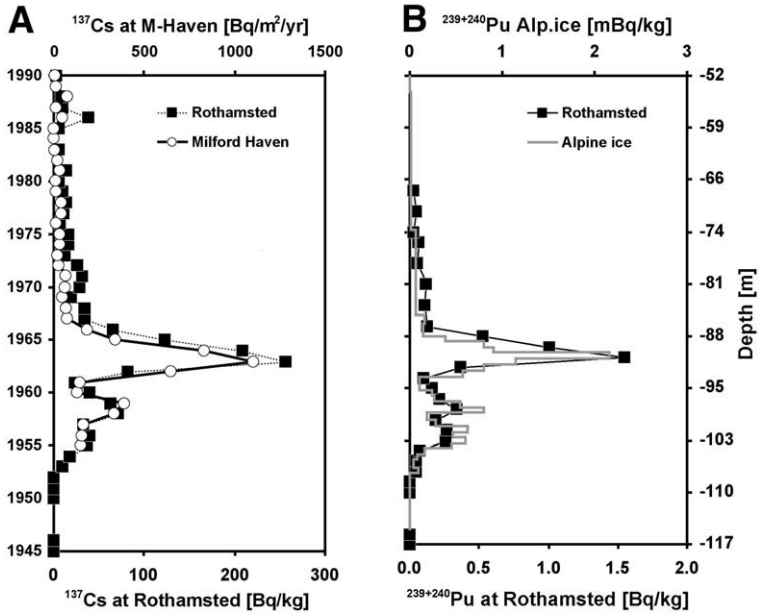

Fig. 3. (A) ^{137}Cs specific activities in the Rothamsted grass (Bq/kg) and deposition flux at Milford Haven (Bq/m^2/yr) (from AEA monitoring from 1957 to 1991) [24]. Both ^{137}Cs profiles are decay corrected to the indicated dates. (B) ^{239,240}Pu specific activities in the Rothamsted grass and in Alpine ice.

easterly dispersal of the test plume toward western Europe [9] (Fig. 1). Hitherto the only record of this fallout in the UK was from measurements of β/γ activity in air carried out by AEA (Atomic Energy Authority) [25]. The current work confirms that plutonium was also deposited since the 1952 Rothamsted grass (June–July harvest) has a ^{240}Pu/^{239}Pu of 0.06. This ratio in the grass cannot be related to the first nuclear test with a stratospheric input, the 10.4 Mt 'Mike' shot at Eniwetok Atoll which occurred on 31 October 1952. Since there were no USSR tests in 1952 this plutonium can only be fallout from one or more of the eight tests at the NTS between April and June 1952. The time-lag between a Nevada test and subsequent deposition in the UK could have been as short as 5 days if suitable weather conditions existed [26]. The sample from the 1952 second harvest (Sep–Nov) shows no measurable plutonium, which is consistent with the records of no summer testing at the NTS. Previously reported low isotope ratios in North Atlantic, Gulf of Mexico and Mississippi Delta sediments [3–6] have been attributed to NTS testing but they did not have the good time control of the grass samples. Additional evidence for tropospheric fallout is also seen for later samples. For example, ^{240}Pu/^{239}Pu in the 1953 grass is 0.154 for the first cut and 0.135 for the second cut. Since fallout from the Oct 1952 'Mike'-Shot had a ^{240}Pu/^{239}Pu greater than 0.30, as inferred from ice cores [23], the lower 1953 ratios must represent mixtures of tropospheric fallout and stratospheric fallout. Between the first and second 1953 Rothamsted cuts there were no tests conducted at NTS and no plutonium isotopic ratio differences would be expected for purely stratospheric fallout. Any differences in ^{240}Pu/^{239}Pu between the first and second cuts would mostly reflect tropospheric fallout. The slightly lower ^{240}Pu/^{239}Pu of 0.135 in the second 1953 cut could include material derived from the first Soviet thermonuclear test (Kazakhstan site) in August 1953 (440 kt).

The main stratospheric events that dominate the ratios during 1954–1961, are the US and UK tests on Bikini, Eniwetok and Christmas Island in 1954, 1956 and 1958 and Soviet tests in the Arctic in 1958, 1961 and 1962. In November

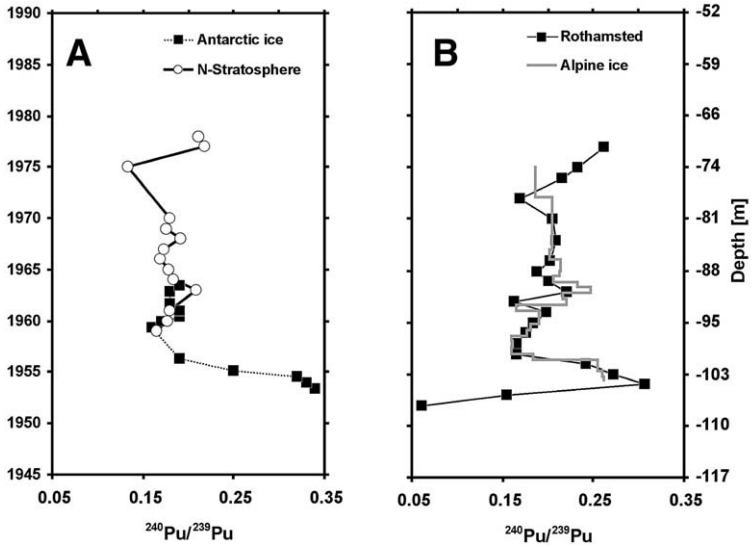

Fig. 4. (A) $^{240}Pu/^{239}Pu$ atom ratio in an ice core from the J-9 Ross Ice Shelf, Antarctica [23] and in the northern stratosphere [24]. (B) $^{240}Pu/^{239}Pu$ atom ratio in Rothamsted grass and Alpine ice. Full details of the mass spectrometer and the analytical procedures developed for uranium and plutonium are published elsewhere [8,20].

1958 the Partial Test Ban Treaty inhibited nuclear testing until the resumption of the Soviet test series in September 1961. The USA and the USSR finally ceased atmospheric testing in October 1962 and December 1962 following the ratification of the International Test Ban Treaty. Data from the stratospheric sampling in the Northern Hemisphere from 1959 onwards [24] are useful for correlation purposes. The ratios derived from stratospheric and Antarctic ice core [23,24] sampling show the same trends as the Rothamsted grass except where tropospheric inputs have occurred

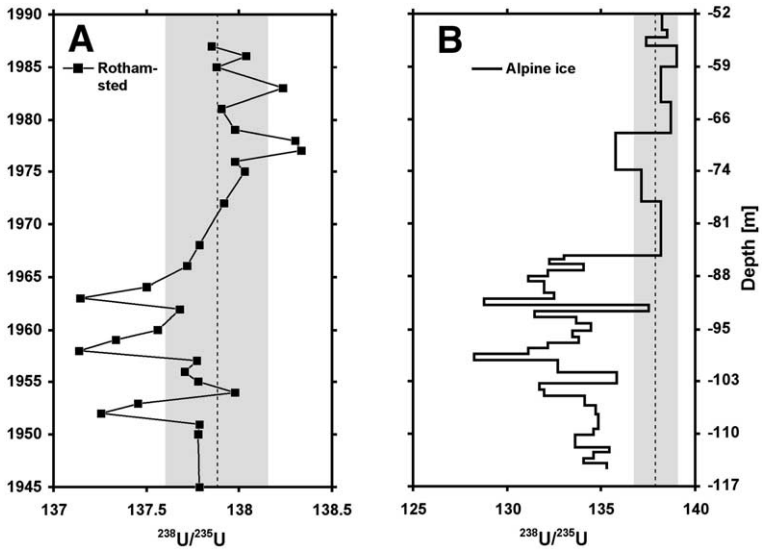

Fig. 5. (A) $^{238}U/^{235}U$ atom ratios in Rothamsted grass samples. (B) $^{238}U/^{235}U$ atom ratios in Alpine ice from Dome du Gouter (4300 m), Mont Blanc, France. The shaded area represents the 3-sigma analytical limit.

and the grass record therefore provides an important extension of the record.

Chinese and French atmospheric thermonuclear tests were carried out between 1964 and 1980 and 1966 and 1974 respectively [21] and can also be related to the ratios seen in the grass. The small increase in the ratio during the late 1970s seen in N-stratosphere and Rothamsted data must be due to Chinese tests. There are no ice core $^{240}Pu/^{239}Pu$ data for this period although stratospheric aerosol measurements exist for 1975, 1977 and 1979. The Rothamsted isotope ratio data provide a useful alternative for this hiatus in the record.

Uranium isotope ratio data for atmospheric fallout from weapon testing have not been previously reported, perhaps because the small amounts of isotopically altered uranium contamination are masked by the presence of natural uranium. The variation of the $^{238}U/^{235}U$ in the Rothamsted grass shows three distinct negative inflections (enriched U ratio) at 1952, 1958 and 1963 and two small but significant positive inflections (depleted U ratio) at 1977/78 and 1983 (Fig. 5A). The two negative inflections in the 1958 and 1963 grass data are also present in the ice record but no positive inflections are seen later in the ice core. The 1952 deviation in the Rothamsted grass, where Pu was also first detected, is likely to have been caused by tropospheric fallout from NTS. The 1953 deviation is likely to result from a mixture of tropospheric and stratospheric fallout, as also inferred from the Pu data, while the 1954 data can be attributed mostly to stratospheric fallout. The strongest deviations in $^{238}U/^{235}U$ in the grass record occur in 1958 and 1963 which coincide with the years of greatest fallout from weapon testing. The ratios in both cases are about 0.55% lower than the natural ratio but the magnitudes of the deviations for 1958 and 1963 are greater in the Mont Blanc ice (Fig. 5B). Enriched uranium isotopic ratios in the ice persist for the whole period of high yield atmospheric testing. It is likely that the reason for the differences between ice and grass is a higher content of natural uranium in the grass compared with the ice. The discovery of slightly enriched uranium in weapon fallout and its small magnitude can be used to argue against a weapon fallout origin for the anomalous uranium isotopic composition that was reported for samples around Greenham Common [2]. Those data showed a $^{238}U/^{235}U$ of less than 100 which is considerably smaller than is seen in the 1958 Rothamsted grass or the Alpine ice data.

Explanations for the small inflections seen for depleted uranium ratios in the Rothamsted grass and not the Alpine ice core might indicate that the depleted $^{238}U/^{235}U$ in the grass in 1977, 1978 and 1983 is due to a local, but presently unidentified source. A possible source for these very small perturbations could be from industrial uses of depleted uranium and/or the testing or fabrication of uranium-based weaponry similar to that reported by other workers [27,28].

4. Conclusions

Ground level records for $^{240}Pu/^{239}Pu$ and $^{238}U/^{235}U$ atom ratios for northern temperate latitudes have been obtained from an annual UK herbage archive and a core from a French Alpine glacier using a recently developed mass spectrometric technique. Evidence is provided, for the first time, that plutonium contamination originating from Nevada Desert atmospheric weapon tests in 1952 and 1953 extended eastwards as far as northwestern Europe. Hitherto unseen fluctuations in $^{238}U/^{235}U$ atom ratios in the atmospheric fallout record are also reported, some of which are directly related to nuclear testing and some of which may indicate an origin from military or industrial sources. Some later fluctuations in uranium isotopes may result from uses associated with uranium-based weaponry. The results presented show that there is now a ready capability to detect and precisely identify sources of plutonium in the environment which has implications for the development of atmospheric transport models, recent geochronology and environmental studies.

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