

# Natural and anthropogenic levels of tritium in a Canadian Arctic ice core, Agassiz Ice Cap, Ellesmere Island, and comparison with other radionuclides

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**ABSTRACT.** Numerous studies of the ice caps in Greenland and Antarctica have observed accumulations of transuranic radionuclides and fission products from nuclear weapons testing, particularly during the period 1945–75. Recently, the concentrations of radionuclides in the annually deposited surface layers of Agassiz Ice Cap, Ellesmere Island, Canadian Arctic, from 1945 to the present have been measured and have demonstrated a continuous record of deposition of <sup>137</sup>Cs and <sup>239,240</sup>Pu in ice and snow. In this study, <sup>3</sup>He-ingrowth mass spectrometry has been used to measure the low levels of tritium (<sup>3</sup>H) in some of these samples. Pre-nuclear-bomb tritium levels in ice-core samples were approximately 12 TU in high-latitude meteoric waters and 3–9 TU in mid-latitude meteoric waters. Comparisons of <sup>3</sup>H levels and <sup>3</sup>H/<sup>137</sup>Cs + <sup>239,240</sup>Pu ratios, which were quite low during the earliest fission-bomb detonations (1946–51) and substantially higher during thermonuclear hydrogen-fusion bomb testing (1952–64), provide a clear indication of the type of nuclear device detonated. This finding accords with the results from other ice-core studies of the distribution of anthropogenic radionuclides from bomb fallout.

## INTRODUCTION

Permanent ice sheets can be used as a proxy to provide a detailed record of fallout materials from the atmosphere to the Earth's surface at any latitude (e.g. Thompson and others, 1989; Cecil and Vogt, 1997). A long period of research on glacier ice cores has indicated that, with noted exceptions (Koerner and Taniguchi, 1976; Goto-Azuma and others, 1997), summer melting and the effects of ice scouring may be negligible to non-existent; hence, the continuous yearly accumulations will be largely undisturbed, with the snow or ice and dissolved and particulate matter remaining largely intact. Numerous studies of annual ice layers in ice caps in Greenland and Antarctica have observed the fallout deposition of transuranic radionuclides and associated fission products from nuclear weapons testing (Koide and Herron, 1977; Jouzel and others, 1979; Koide and others, 1979, 1982; Elmore and others, 1982; Cecil and Vogt, 1997; Levchenko and others, 1997), particularly during the period 1945–75. Similar studies have been carried out to a limited extent on lower-latitude, higher-altitude glaciers, such as those by Cecil and Vogt (1997) and Naftz and others (1993) who measured <sup>36</sup>Cl and tritium (<sup>3</sup>H) on the Wind River alpine glaciers in Wyoming, Colorado, U.S.A.

Concentrations of radionuclides (<sup>137</sup>Cs and <sup>239,240</sup>Pu) have been measured on the annual ice layers of Agassiz Ice Cap, Ellesmere Island, Canadian Arctic, and indicate a continuous record of deposition of anthropogenic nuclides between 1945 and the present. Kudo and others have docu-

mented the record of fallout from the earliest fission weapons test and nuclear detonations in Japan (Hiroshima and Nagasaki) and elsewhere starting in 1945, and also from subsequent worldwide fusion-bomb testing beginning in the early 1950s (Carter and Moghissi, 1977). In this study we have used <sup>3</sup>He-ingrowth mass spectrometry to measure the tritium concentrations in 1935–64 ice horizons in spatially associated core samples, and comment on: (1) the tritium concentrations in high-latitude, pre-1945 precipitation and in precipitation from the earliest nuclear detonations, and (2) correlations between transuranic radionuclides and tritium in the high-latitude snowpack.

## MATERIALS AND METHODS

Ice samples supplied for tritium analysis were collected from a site located at the dome of Agassiz Ice Cap (~81° N; 1740 m a.s.l.). Details of the ice-core drilling and sampling can be found in Zheng (1996), Zheng and others (1998) and Kudo and others (1998). Samples equivalent to 6 months' snow accumulation were taken from depths of 13.7–7.0 m, representing the time interval 1935–64, and were predominantly firn with some interlayers of ice (Kudo and others, 1998). Frozen ice-core samples (~180–530 g) were sealed in Nalgene containers, were allowed to melt at Canadian National Research Council laboratories in Ottawa, and were then transported to the Low Background Laboratories at Atomic Energy of Canada Ltd, Chalk River, Ontario, for tritium measurements.

Determination of  $^3\text{H}$  in the ice-core samples was primarily carried out using  $^3\text{He}$ -ingrowth mass spectrometry (Clarke and others, 1976) utilizing a noble-gas mass spectrometer (MAPL 215-50 Mass Spectrometer; Lynch and Kay, 1981) and a high, vacuum ( $<10^{-7}$  torr) stainless-steel gas-inlet and helium-cryogenic gas-separation system (Lott and Jenkins, 1984; Bayer and others, 1989; Kotzer and others, 1994; Mahara, 1995). For  $^3\text{He}$ -ingrowth mass spectrometry, the prepared water standards and melted ice waters were transferred using  $\text{N}_2$  gas and capillary transfer, evacuated to remove all atmospheric gas components and stored at low temperatures ( $-20^\circ\text{C}$ ) for up to 9 months in specialized vessels constructed of Al-Si glassware (Dow Corning 1720/1724; Suckow and others, 1990). Tritium concentrations in some of the ice-core samples were also determined by direct liquid-scintillation counting methods using a Wallac 1220 scintillation counter and equal volumes ( $\sim 10$  mL) of Packard Picofluor LLT scintillator and sample waters in Packard low-diffusion polyethylene vials. Tritium concentrations are given in tritium units (TU) where  $1 \text{ TU} = 1 \text{ } ^3\text{H} \text{ per } 10^{18} \text{ H atoms}$ .

Duplicate and triplicate measurements using  $^3\text{He}$ -ingrowth mass spectrometry on selected ice samples and previously measured International Atomic Energy Agency (IAEA) and Chalk River Laboratories (CRL) intercom-

parison waters were combined with the results from liquid-scintillation counting on several of the ice samples (Nos. 1–4 and 14–19; Table 1). Good agreement was obtained for the measurements, with maximum errors of  $\pm 25\%$  for the lowest-level samples ( $<1 \text{ TU}$ ) and  $\pm 2\text{--}10\%$  for higher levels ( $>1 \text{ TU}$ ).

## RESULTS AND DISCUSSION

### Tritium results

The range of tritium concentrations in ice-core samples at depths of 7.5–13.92 m below the Agassiz Ice Cap surface is 0.35–474 TU (Table 1; Fig. 1). These depths represent deposition of snow between 1935 and 1964 and are discussed in the sequence oldest to youngest. The ice cores measured for tritium were from the same location as several other ice cores which were analyzed for other transuranic nuclides as reported by Kudo and others (1998). Where applicable, the samples have been decay-corrected using a half-life for tritium of 12.39 years.

#### (a) Pre-bomb ice samples

Triplicate analyses on the deepest ice samples (13.77 and

Table 1. Sampling depths, descriptions and tritium concentrations for prepared standard waters and ice-core samples from Agassiz Ice Cap

No.	Depth m	Year <sup>1</sup>	Length cm	Weight g	Description	Measured tritium		Corrected <sup>2</sup> $^3\text{H}/\text{Cs} + \text{Pu}^3$ tritium	
						$^3\text{He}$ mass spectrometry TU	Liquid-scintillation counting TU	TU	Bq $\text{cm}^{-2}$
1	7.5–7.6	1964	9.5	238.9	Firn with 4 mm full ice layer at one end	474	470 $\pm$ 14	2976	2933
2	8.5–8.6	1958	9.5	244.0	Firn with a half-circle of ice about 1.5 cm thick about 8 mm from one end	58	60 $\pm$ 7	538	311
3	8.9–9.0	1957	9.5	229.0	Homogeneous, clear firn	65.3	67 $\pm$ 7	605	
4	9.5–9.6	1955	9.5	218.9	Almost completely homogeneous firn	10.2	10 $\pm$ 6	105	78
5	9.8–9.9	1952–53	9.8	245	Almost completely homogeneous firn sample with 5 mm thick ice layer at one end	4.7	n/a	54.3	63
6	10.1–10.2	1951	8.8	188	Homogeneous firn with no visible ice	2.2	n/a	27.4	
7	10.2–10.3	1950	9.8	233	Firn with discontinuous ice layer approx. 8 mm wide. Ice lens is approximately 1/3 of sample	1.8	n/a	23.6	
8	10.5–10.6	$\sim$ 1949	10.0	270	Firn with very faint ice layer which is 2 cm long, 4 mm wide and 2 cm from one end of sample	4.6	n/a	61.5	
9	10.9–11.0	$\sim$ 1946	9.5	261	Firn with small ice layer (6 mm wide) near one end	5.1	n/a	82.2	
10	11.5–11.6	1944–45	10.0	272	Firn	0.63	n/a	11.4 <sup>6</sup>	87
11	11.6–11.7	1944–45	9.5	278	Firn containing several faint, small ( $<5$ mm thick) ice layers	0.86	n/a	14.7 <sup>6</sup>	
12	13.77–13.92	$\sim$ 1935	15.5	528.1	Firn with numerous ice layers ( $\sim 25\%$ ice by volume)	0.53, 0.42	n/a	14.9, 11.8 <sup>6</sup>	
13	13.77–13.92	$\sim$ 1935	14.5	419.8	Firn with two small ice layers ( $<1.4$ cm wide)	0.35	n/a	10 <sup>6</sup>	
<i>Low-level water analyses</i>									
14	IAEA A <sup>4</sup> (N = 1)					0.29	0.139 $\pm$ 0.018		
15	IAEA B <sup>4</sup> (N = 3)					1.13 $\pm$ 0.039	1.074 $\pm$ 0.026		
16	IAEA C <sup>4</sup> (N = 2)					4.8–5.1	4.926 $\pm$ 0.029		
17	IAEA D <sup>4</sup> (N = 4)					19.52 $\pm$ 1.8	19.551 $\pm$ 0.085		
18	CRL 30 <sup>5</sup> (N = 2)					35–37	38 $\pm$ 9		
19	CRL 70 <sup>5</sup> (N = 2)					69–72	75 $\pm$ 9		

<sup>1</sup> Based on layer counting and associated radio-isotope analyses.

<sup>2</sup> Decay-corrected to 1996.

<sup>3</sup> Ratio of  $^3\text{H}/^{137}\text{Cs} + ^{239,240}\text{Pu}$ .

<sup>4</sup> Waters supplied from 1994 IAEA tritium intercomparison; decay-corrected to 1994 (Østlund and others, 1995).

<sup>5</sup> Samples prepared at CRL by dilution of natural waters.

<sup>6</sup> Pre-anthropogenic high-latitude meteoric precipitation is approximately 12.5 TU (mean + std dev. = 12.54  $\pm$  1.95).

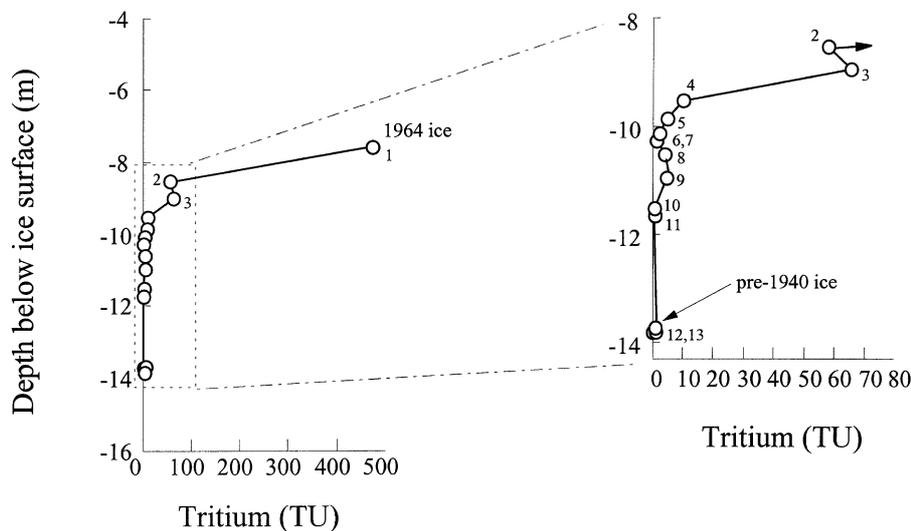


Fig. 1. Measured tritium–depth profile for ice-core samples from Agassiz Ice Cap, 1930–64. (Numbers beside data points correspond to sample numbers from Table 1; tritium values not decay-corrected.)

13.92 m;  $\sim$ 1935) yield tritium concentrations of 0.35–0.53 TU, which, when decay-corrected, result in an average tritium concentration of 12.2 TU (Table 1; Fig. 1). Duplicate analyses on ice samples from 11.5–11.7 m depth ( $\sim$ 1944–45) have concentrations of 0.63–0.86 TU (Table 1) and decay-corrected tritium values of 11.4–14.7 TU. Combined, the ice samples for the time interval 1935–44 indicate a decay-corrected tritium value of  $12.5 \pm 1.9$  TU (samples 10–13; Table 1).

#### (b) 1945–64 ice samples

Tritium levels were measured in two ice-core samples at 10.5–11.0 m depth, which represents snow accumulation between approximately 1946 and 1949. During this period, initial detonation of U-fission bombs occurred (Carter and Moghissi, 1977). Measured tritium values for these depths are 4.55 and 5.08 TU, with decay-corrected tritium values of 61.5 and 82.2 TU (Table 1; Fig. 1).

Ice-core samples from 7.5–10.2 m depth have measured tritium concentrations in the range 2.24–474 TU, with decay-corrected tritium values of 27.4–2976 TU (Table 1; Fig. 1). These depths represent snow accumulations during the period 1950–64, when substantial testing of thermonuclear hydrogen-fusion bombs occurred (Carter and Moghissi, 1977).

## Discussion

Koide and others (1982) and Koerner and Taniguchi (1976) have reported on tritium concentrations in ice samples from the Greenland ice cap (Dye-3) and from Devon Island, Canadian Arctic, although no tritium determinations were carried out on samples of ice older than 1950. As some of the ice-core samples in this study are from snow-accumulation periods on Agassiz Ice Cap beginning as early as 1935, the measured tritium levels have been used to estimate the natural, cosmogenic levels and production rates of tritium in high-latitude precipitation over North America before detonation of nuclear devices started in 1945.

Tritium levels for snow deposited between 1935 and 1945 (pre-bomb) on Agassiz Ice Cap are approximately 12.5 TU, which is higher than the tritium levels for mid- to northern-latitude North American and European meteoric waters and wines during the period 1927–45 (Fig. 2). The measured tritium values for high-latitude, Agassiz precipitation agree well with pre-nuclear values of  $<20$  TU in snow sampled in

1944 and 1945 from Thule, Greenland (Begemann, 1958). Koerner (1970) found that melting on Devon Ice Cap diminished during the 1940s, so it is not considered that tritium concentrations in the Agassiz ice over this same time interval (samples 10–13; Table 1) have been affected to any great extent by processes associated with ice ablation. Higher tritium levels are expected in pre-bomb, meteoric precipitation for the Canadian Arctic and Greenland than in precipitation at lower latitudes, as the natural production of tritium varies as a function of geomagnetic latitude, with greater cosmogenic tritium production and maximum stratospheric–tropospheric water-vapor transfer occurring at higher latitudes.

Using the tritium contents along with the annual accumulation rates of precipitation for pre-bomb Agassiz samples (Nos. 10–13; Table 1), a value for the natural production rate of tritium has been estimated. Here, an average tritium concentration of 12.5 TU, and an upper limit for the accumulation rate of  $18 \text{ g w.e. cm}^{-2} \text{ a}^{-1}$ , results in a calculated natural production rate of  $0.54 \pm 0.05 \text{ tritons cm}^{-2} \text{ s}^{-1}$ . This value is of the same order of magnitude as empirical observations for Greenland ( $1.05 \pm 0.15 \text{ tritons cm}^{-2} \text{ s}^{-1}$ ; Begemann, 1958) and calculated worldwide values ( $0.16\text{--}0.20 \pm 0.09 \text{ tritons cm}^{-2} \text{ s}^{-1}$ ; Nir and others, 1966).

During the period 1946–49, tritium concentrations which have been measured in the Agassiz ice-core samples are elevated and, although still quite low, are up to five times greater than the decay-corrected, pre-bomb tritium levels in deeper ice-core samples (Nos. 8 and 9; Table 1; Figs 1 and 2). There are several possible explanations for this. One is that isolated periods of intense summer ablation and melting on the snowpack have resulted in percolation of snowmelt waters into the underlying layers (Koerner, 1970). Such processes have been documented for mid-latitude alpine glaciers (Naftz and others, 1993; Cecil and Vogt, 1997) and during select time intervals for Canadian Arctic glaciers (Koerner and Taniguchi, 1976; Fisher and Koerner, 1994). Although melting would be expected to be minimal at the high elevations and latitudes of some Arctic glaciers, several periods of melting have been documented on Agassiz Ice Cap, the early 1950s being amongst the most significant (cf. Fisher and Koerner, 1994, fig. 10). This observation may suggest that the 1946–49 snow layers, which are within 0.5–1 m of the early 1950 depositional intervals (samples 5–9; Table

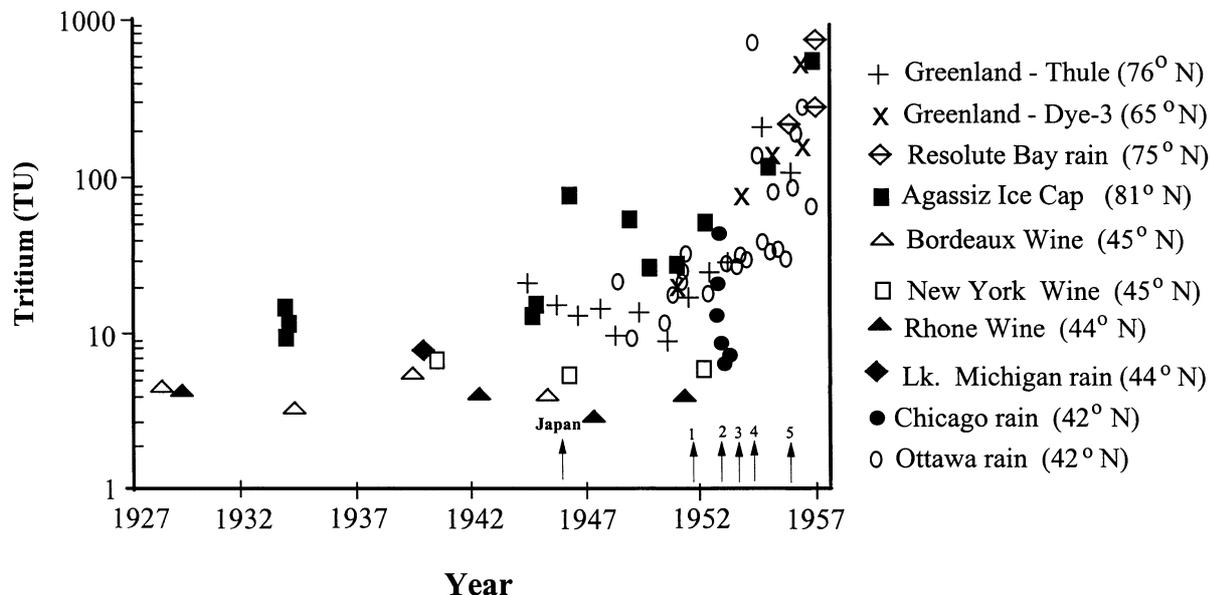


Fig. 2. Comparison between (i) tritium levels measured in precipitation and wine samples from mid-latitude meteoric waters, and (ii) tritium levels measured in precipitation and ice-core samples from higher-latitude precipitation. Mid-latitude tritium data adapted from Clark and Fritz (1997). Other symbols used: ■, tritium measurements from this study; ◇, tritium data for Resolute Bay (Canadian Arctic) rain (IAEA Global Network of Isotopes in Precipitation database); ×, tritium data on Greenland ice core (Dye-3) from Koide and others (1982); +, tritium data for snow from Thule, Greenland (Begemann, 1958). Small arrows on bottom of diagram refer to Japan: Nagasaki atom bomb and earliest atmospheric tests of thermonuclear devices, namely, 1. George (75 kilotons, U.S.A.); 2. Ivy-Mike (10 megatons, U.S.A.); 3. RDS-6s (400 kilotons, U.S.S.R.); 4. Castle series, (total 47 megatons, U.S.A.); 5. second Soviet test (2 megatons) (from Clark and Fritz, 1997).

1), may have been affected by percolation of snowmelt waters from the 1950s, which would have contained more tritium from initial testing of hydrogen-fusion bombs (Brown, 1970; Carter and Moghissi, 1977). Concentrations of  $\text{NO}_3$ ,  $\text{SO}_4$  and  $\text{Cl}^-$  in annual layers from Agassiz Ice Cap suggest limited post-depositional changes to the ice layers as a result of melting and percolation in the early 1950s (Goto-Azuma and others, 1997), although this may not have been the case for tritium in the ice core, which would be more mobile in the vapor phase (as tritiated water vapor), and may also serve to explain the immobile behavior of some of the other radionuclides ( $^{239+240}\text{Pu}$ ,  $^{137}\text{Cs}$ ) during these time periods, as they, like the anion complexes, would exist as particulate phases (Kudo and others, 1998).

A second possibility is that the elevated tritium levels measured in the 1945–49 Agassiz ice-core samples (Nos. 8 and 9; Table 1) represent a record of the deposition of anthropogenic tritium fallout from the earliest fission-bomb detonations between late 1945 and 1950 (Hiroshima, Nagasaki, Bikini, Eniwetok; Carter and Moghissi, 1977), as the small increases in tritium levels coincide with increasing amounts of  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$  in the ice horizons, which have been interpreted as fallout transuranics from the earliest fission-bomb detonations (Fig. 3; Koide and others, 1982; Kudo and others, 1998). The absence of an adequate mechanism to produce substantial concentrations of tritium in the earliest fission-bomb detonations, and the lack of data reflecting increased tritium levels in Greenland snow samples and in lower-latitude meteoric precipitation during the earliest period of nuclear fission-bomb testing (Brown and Grummitt, 1956; Begemann, 1958), does not support this interpretation (Fig. 2).

The proliferation of hydrogen-fusion bomb tests between 1952 and 1962 produced large inventories of tritium which were injected into the stratosphere and significantly increased the tritium levels observed in mid- to higher-lati-

tude, Northern Hemispheric meteoric precipitation (Fig. 2; Brown and Grummitt, 1956; Brown, 1970; Carter and Moghissi, 1977). As has been observed in ice cores from Dye-3 and elsewhere (Koide and others, 1979, 1982), detonation of the earliest fission bombs, followed by testing of hydrogen-fusion bombs, resulted in increases in the deposition of tritium relative to other radionuclides. These changes were particularly evident for the years 1958 and 1961–64, during which significant thermonuclear testing took place. On Agassiz Ice Cap, annual ice layers deposited over the same time periods record increases in the levels of tritium (Fig. 2) which are comparable to those in synchronous ice-core samples from Greenland (Dye-3; Koide and others, 1982). A limitation here is that, because of the discontinuous nature of the 1952–64 samples available for tritium measurements, the data do not define two distinct peaks for 1958 and 1963, as observed for ice cores at the Dye-3 site (Koide and others, 1982). Rather, for these data, there is an overall trend of increasing tritium in the early 1950s (samples 3–6; Figs 1 and 2), with a slight decrease around 1958–59 (sample 2; Fig. 1) and a subsequent substantial increase in tritium in the 1964 sample (No. 1; Fig. 1). Although less precisely defined than in other studies, these tritium concentrations reflect increases in Northern Hemispheric, atmospheric tritium as a result of the earliest Soviet and U.S. (Ivy–Castle–Redwing series) hydrogen-fusion bomb tests and the subsequent large infusion of atmospheric tritium following increased hydrogen-fusion bomb tests in the early 1960s (Carter and Moghissi, 1977). Despite the lack of a detailed tritium profile for the Agassiz core, the decay-corrected tritium values are consistent with those in temporally equivalent snow layers for the Dye-3 core and for snow samples collected near Thule (Fig. 2), suggesting equivalent transport mechanisms for both of these high-latitude areas.

The data from Agassiz Ice Cap indicate increasing  $^3\text{H}$ ,  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$  concentrations and  $^3\text{H}/^{137}\text{Cs} + ^{239,240}\text{Pu}$

activity ratios, trending from low activity ratios and tritium concentrations between 1935 and 1952, and substantial increases between 1958 and 1964 (Table 1; Fig. 3). Thus, similar to what has been recorded for deposition of tritium and other radionuclides in other ice-core sites worldwide, the changes in radionuclide concentrations and their deposition ratios in the Agassiz ice-core samples are a reflection of the type of nuclear device detonated, providing an isotopic “fingerprint” to characterize these depositions in the natural environment and to aid in establishing the ice-layer chronology.

## CONCLUSIONS

Measurement of tritium in ice cores from Agassiz Ice Cap using  $^3\text{He}$ -ingrowth mass spectrometry has yielded accurate data which are comparable to the chronologic distribution of anthropogenic tritium on other polar ice caps and in meteoric precipitation, even after several decades of radioactive decay. The following conclusions have been drawn from these measurements of tritium and from comparison of these data with previously measured radionuclides in several ice-core samples taken from the same area (Kudo and others, 1998).

- (1) With the exception of two ice-core samples (10.9–11.0 and 10.5–10.6 m; Table 1), the record of tritium deposition for annual layers on Agassiz Ice Cap indicates that the site is a suitable archive for delineating the fallout patterns of cosmogenic and anthropogenic radionuclides in high-latitude meteoric waters.
- (2) Pre-nuclear-bomb tritium levels in high-latitude meteoric waters were approximately 12.5 TU, compared to values of 3–9 TU in mid-latitude meteoric waters (Fig. 2).
- (3) The record of the timing of tritium fallout on Agassiz Ice Cap is similar to that observed by measuring fission-product  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$  (Fig. 3). As elsewhere, the nature of the nuclear detonation can be estimated from the ratio of  $^3\text{H}/^{137}\text{Cs} + ^{239,240}\text{Pu}$  which was quite low during the earliest

fission-bomb detonations and substantially higher during subsequent thermonuclear hydrogen-fusion bomb testing.

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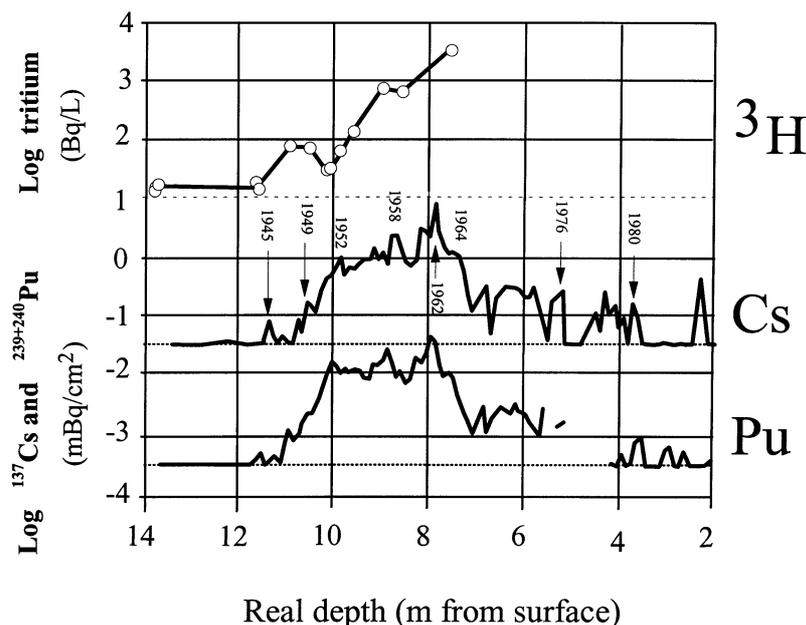


Fig. 3. Comparison between tritium levels (this study) and the concentrations of  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$  previously measured by Kudo and others (in press) on annually deposited ice samples from several other ice cores taken at the same site on Agassiz Ice Cap. Note units used for tritium levels ( $\text{Bq L}^{-1}$ ) relative to those for transuranic radionuclides ( $\text{mBq cm}^{-2}$ ). For tritium 1 TU =  $0.1184 \text{ Bq L}^{-1}$ .

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