

Past and recent tritium levels in Arctic and Antarctic polar caps

Elise Fourné^a, Philippe Jean-Baptiste^{a,*}, Arnaud Dapoigny^a,
Dominique Baumier^a, Jean-Robert Petit^b, Jean Jouzel^a

^a *IPSL/LSCE, UMR CEA-CNRS 1572, CEA-Saclay, 91191 Gif/Yvette, France*

^b *CNRS-LGGE, 38402 Saint Martin d'Hères, France*

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Abstract

Tritium concentration was measured in snow deposited at the GRIP site (central Greenland) and at the Vostok station (east Antarctica) from snow pits covering the period 1980–1990. The objective of the study was to investigate tritium concentrations in polar regions several decades after the bomb peak of the sixties and to put them in the context of available data for environmental tritium in the Arctic and the Antarctic over the last five decades. The tritium content of the samples was measured by mass spectrometry using the helium-3 regrowth method. In Antarctica, the tritium concentrations are in the range 70–110 TU. The comparison of the bomb tritium history at different locations show that tritium levels increase moving inland, where vapour pressure becomes extremely low and therefore more sensitive to the intrusion of stratospheric air masses highly enriched in tritium. Although most tritium fallout occurred in the Northern hemisphere, the tritium levels in central Greenland in the 80's, in the range 10–40 TU, are significantly lower than at Vostok. Unlike Antarctica, no such continental effect is observed in Greenland, due to the higher water vapour content of the air masses, as evidenced by the much higher snow accumulation rate. Whereas tritium fallout in Antarctica appears to occur as a result of direct injections of stratospheric tritium during winter, Arctic fallout are the result of the dominant spring injection of stratospheric air at mid-latitude, in line with the deposition of other stratospheric tracers.

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1. Introduction

Natural tritium (³H) is produced mainly by the interaction of cosmic radiations with the upper atmosphere, at a rate of ~ 0.25 atoms/cm²/s [1]. This tritium enters the hydrological cycle in the form of tritiated water molecules (HTO). ³H has a radioactive half-life of 4500 ± 8 days. By equating the production and decay rates, a steady-state global inventory of tritium of ~ 3.6 kg can be inferred.

Since 1954, vast amounts of thermonuclear tritium have been injected into the upper troposphere and the stratosphere. Estimates are that atmospheric detonations of thermonuclear devices produced ~ 525 kg of tritium in the form of HTO [2]. Approximately 13 kg of tritium gas (HT) was also released by military tritium facilities and leaks from underground testing of nuclear weapons [3]. Over the same period (i.e., 1950–2000), the contribution of the civil nuclear industry to the tritium budget was comparatively quite small with modest airborne releases of 0.35 kg of tritium [2]. Since the Nuclear Test Ban Treaty came into force in 1963, tritium

* Corresponding author.

E-mail address: Philippe.Jean-Baptiste@cea.fr (P. Jean-Baptiste).

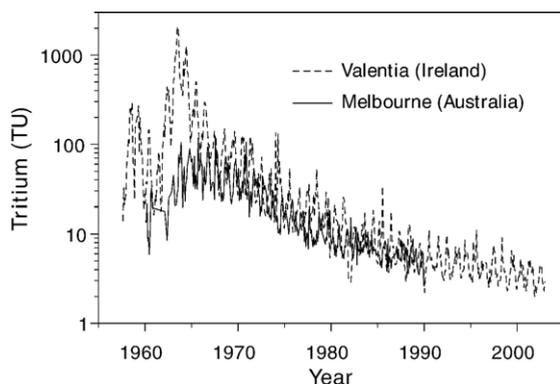


Fig. 1. Tritium in precipitation at Valentia, Ireland and Melbourne, Australia [11].

levels in precipitation have been steadily decreasing (Fig. 1) due to radioactive decay and dilution within the vast oceanic reservoir.

Natural tritium levels are very low. In pre-nuclear times (before 1950), the limited accuracy of measurement methods explains the sparseness and poor reliability of the data for tritium in natural waters. Ice cores provide an alternative and offer the opportunity to reconstruct with a good accuracy the recent history of tritium including pre-nuclear levels. Here we present tritium data measured in snow deposited in central Greenland at Summit (GRIP site, 72°N–37°W) and at the Vostok station, Antarctica (78°S–106°E). The objective of the study was to investigate tritium concentrations in polar regions several decades after the bomb peak of the early sixties and to put them in the context of available data for environmental tritium in the Arctic and the Antarctic over the last five decades. This allows us to draw a global picture of the history of the bomb tritium deposition in polar regions and to study its main characteristics.

2. Materials and methods

Eighty snow samples were taken at the GRIP site (Fig. 2), on the wall of a 5 m deep pit covering the period between 1984 and 1992. In the same manner, sixteen samples were also collected at the Vostok station (Antarctica) from a 2.5 m deep pit (Fig. 2). The tritium content of the ice was measured by mass spectrometry using the helium-3 regrowth method [4]. The principle of the method is to remove the ^3He initially dissolved in the sample by degassing the water sample under high vacuum, then to store it in a sealed glass bulb (Corning 1724 glass) to allow for the accumulation of tritiogenic ^3He . Unlike Pyrex glass which is permeable to helium,

the diffusivity of helium in Corning 1724 glass is low [5], thus preventing ^3He from diffusing through the glass wall. The tritium content of the sample is subsequently deduced from the mass spectrometric determination of the amount of ^3He produced during the storage time. $^3\text{He}^+$ ions are detected on an electron multiplier operated in the pulse mode. The main advantage of this technique is its higher sensitivity compared to conventional tritium counting, with a detection limit of the order of 0.01 TU, corresponding to ~ 1 mBq/kg of ice (1 Tritium Unit means a T/H ratio of 10^{-18}).

3. Results

The results from the GRIP site are shown in Fig. 3. Tritium concentrations, between 10 and 40 TU, are modulated by a clear seasonal cycle similar to the seasonal pattern already observed in precipitation (see Fig. 1). The mean accumulation at the GRIP site deduced from these annual oscillations for the period 1984–1992 is 57.75 cm of snow per yr (Fig. 4), in excellent agreement with that given by the GRIP group based on annual layers counting [6].

The Vostok data include 15 samples covering the period 1980–1990 and one pre-nuclear sample corresponding to the year 1953. The results are shown in Fig. 5 along with historical data. The poor temporal resolution due to the very low accumulation rate at Vostok (~ 2 – 3 cm/yr) does not allow us to observe any seasonal cycle. The northern hemisphere sharp tritium peak of the year 1963/64, which expressed itself in the southern hemisphere record as a broader maximum centered around the year 1966, is less pronounced in the southern hemisphere owing to the fact that most nuclear devices were detonated in the northern hemisphere (Fig. 1). However, the tritium levels recorded at Vostok between 1980 and 1990 (in the range of 70–110 TU) as well as in our 1953 sample (63 TU) are significantly higher than in central Greenland. We interpret this fact as basically reflecting the stronger influence of stratospheric tritium inputs at Vostok due to the very low atmospheric water vapor content and the quasi-absence of precipitation at the site (see discussion below).

4. Discussion

4.1. Stratospheric tritium input

Fig. 6 shows the time-evolution of the average tritium content in the stratosphere [7]. Tritium levels in the stratosphere are higher by several orders of magnitude compared to tropospheric levels, due to the large injection

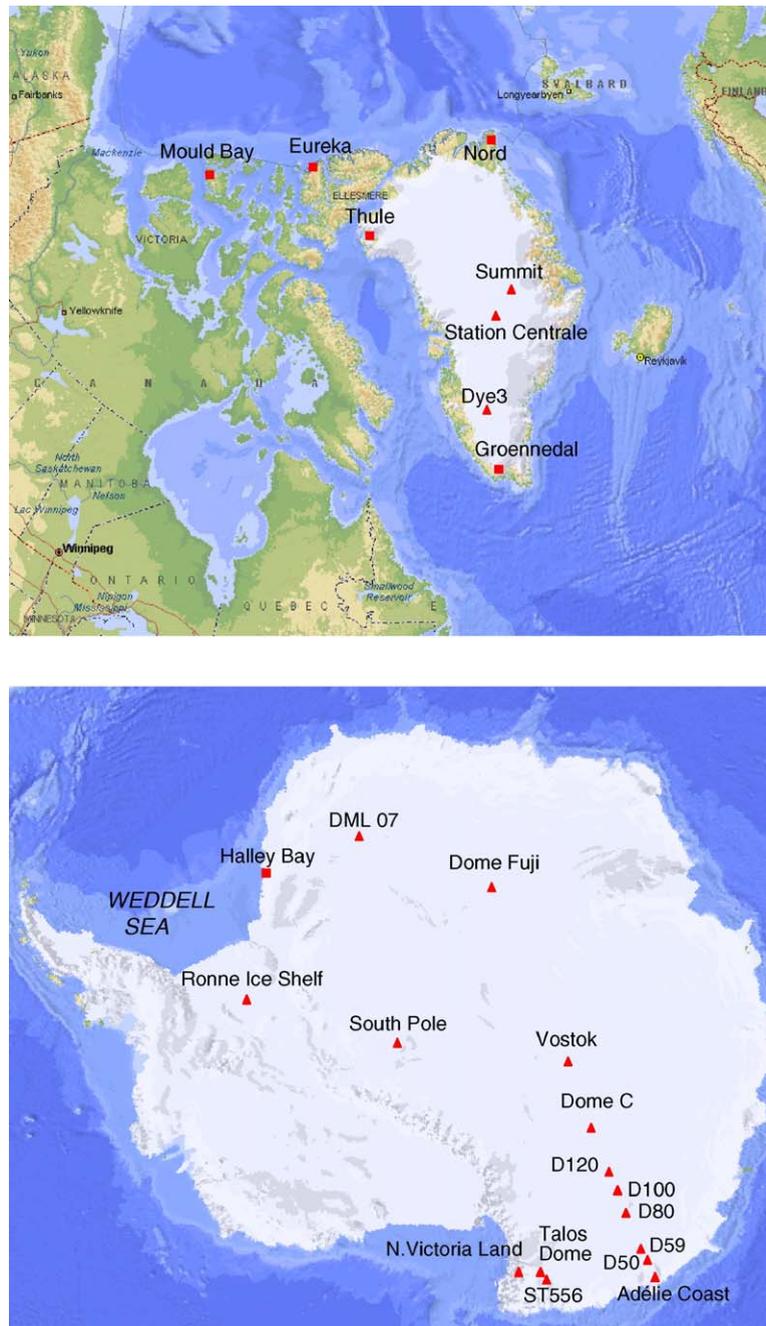


Fig. 2. Location map for Greenland and Antarctica showing the stations with tritium data.

of the bomb tritium above the tropopause during the atmospheric nuclear tests.

In Antarctica, an increasing trend between the tritium content of the snow and the distance from coast is observed on two transects between the Adélie coast and Dome C [8] and between Terra Nova Bay and Dome C [9]. This positive correlation is also clearly noticeable in

Fig. 5: coastal stations, characterized by somewhat milder temperatures and higher accumulation rates, show lower tritium levels than those situated more inland where the temperatures and the atmospheric water vapour content are extremely low. At Vostok for instance, located at a high altitude (3480 m) on a dry and frigid polar plateau (mean temperature = $-55\text{ }^{\circ}\text{C}$),

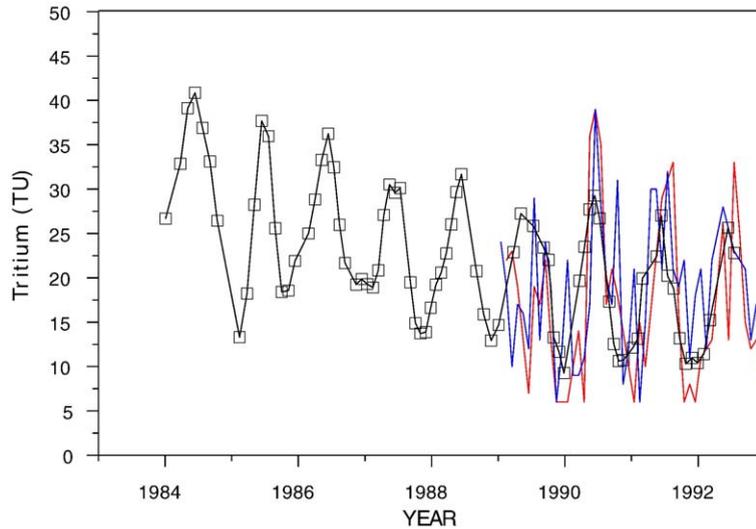


Fig. 3. Tritium concentration at the time of deposition measured in snow at Summit, central Greenland (in black), and comparison with tritium in precipitation at two canadian arctic coastal stations, Mould Bay, 76°N/119°W (in blue) and Eureka, 80°N/85°W (in red) [11]. The chronology is deduced from the counting of the seasonal cycles.

the annual snow accumulation is at its lowest (~ 2–3 cm of water equivalent). In such extreme meteorological conditions, stratospheric intrusions of air masses with a high tritium content have a maximum effect on the local air tritium concentration, especially during the austral winter, when the air is particularly dry and the Antarctic interior is isolated from marine air masses by the polar vortex. This is consistent with the fact that the seasonal maximum in bomb tritium fallout observed in the ice deposited at south Pole since 1954 [8] takes place during the austral winter (April to September).

The influence of the addition of a given fraction α of stratospheric air enriched in tritium (tritium concentration T_{strato}) to a local air mass of marine origin (initial tritium content T_{tropo}) can be described by the following Eq. (1):

$$T(\text{in TU}) \sim \alpha T_{\text{strato}} [\text{H}_2\text{O}]_{\text{strato}} P_{\text{atm}} / h P_{\text{sat}} + (1 - \alpha) T_{\text{tropo}} \quad (1)$$

where P_{sat} is the vapor pressure of water at the local temperature, P_{atm} is the local atmospheric pressure, h is the relative humidity (typically 60%) and $[\text{H}_2\text{O}]_{\text{strato}}$ is the mean water content of the stratosphere (~5 ppm [10]).

The tritium content of the stratosphere in 1966, T_{strato} , can be extrapolated from the tritium stratospheric data [7] at 80×10^6 TU (Fig. 6). T_{tropo} in 1966 is of the order of 75 TU (Fig. 1). Fig. 7 shows the evolution of the atmospheric tritium content T calculated from Eq. (1) as

a function of the local temperature for two typical values of the atmospheric pressure, respectively 600 hPa (in red) and 900 hPa (in black) and for various amounts of stratospheric air (α between 0.001% and 0.05%). The comparison of this very simple model with the amplitude of the 1966 tritium peak at various stations (Table 1 and Fig. 7) shows that the incursion of minute amounts of stratospheric air can explain the continental tritium enrichment observed in the data.

At Vostok, if we take the 1953 value of 63 TU as a measure of the natural tritium concentration, the 1980–

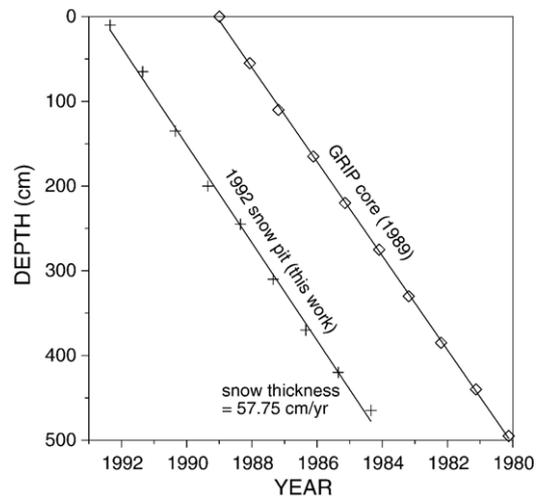


Fig. 4. Plot of the depth of the annual spring tritium maximum at GRIP snow pit versus the year of deposition and comparison with the annual layer counting in the top five meters of the GRIP ice core [6].

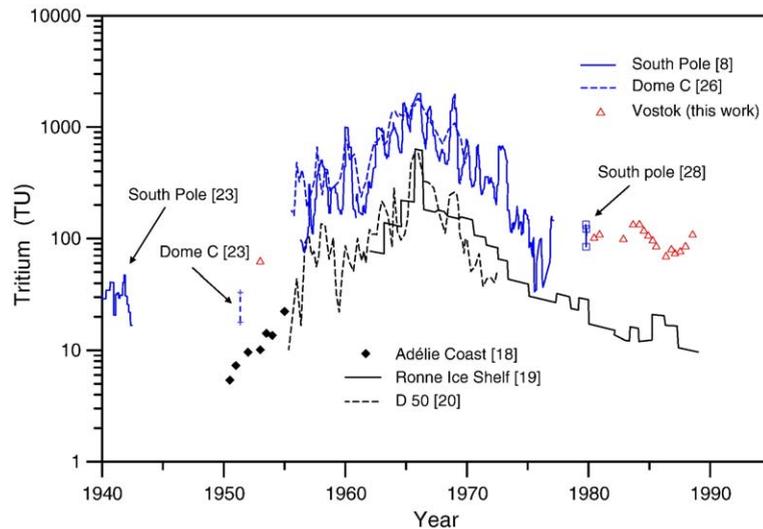


Fig. 5. Composite profile for Antarctic tritium, at the time of deposition. (N.B. the chronology of the Vostok samples was deduced from the Grenoble's accumulation model constrained by the beta activity's 1966 maximum). (Data from [8,18–20,23,26,28] and this work.)

1990 concentrations appears still to be above the pre-nuclear background. On the other hand, the facts that there is no sign of tritium decline in the Vostok dataset and that stratospheric tritium levels are approaching their natural background (see Fig. 6) suggest that tritium concentrations at Vostok may have reached a level where the interannual variability is greater than the residual decreasing trend. This interannual variability is likely linked to the varying strength of the stratospheric subsidence, also recorded in the size distribution of the dust deposited at Vostok (B. Delmonte, pers. comm.).

In Greenland, the effect of direct stratospheric air incursions is expected to be less pronounced owing to the larger moisture content of the air masses, as

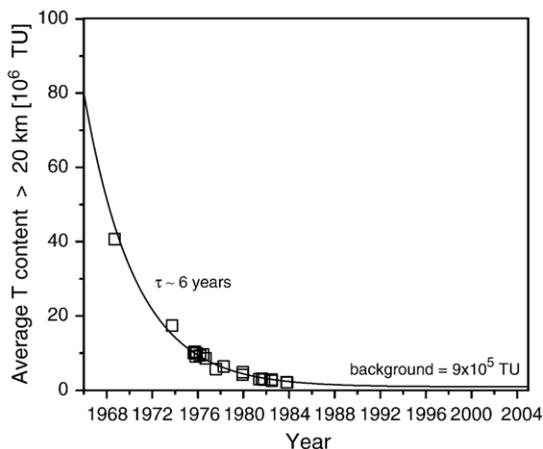


Fig. 6. Tritium levels in the stratosphere (after [7]). The solid line is the best fit of the data using a one-box model of the stratosphere (see text).

evidenced by the high accumulation rate in central Greenland (20 to 25 cm of water equivalent/yr at Summit) compared to central Antarctica (2 to 8 cm of water equiv./yr). Our tritium data for central Greenland in the early 90's are indeed surprisingly similar to those measured in precipitation at two canadian arctic coastal stations, Mould Bay, Melville island, 76°N/119°W and Eureka, 80°N/85°W [11] (Fig. 3). From tritium

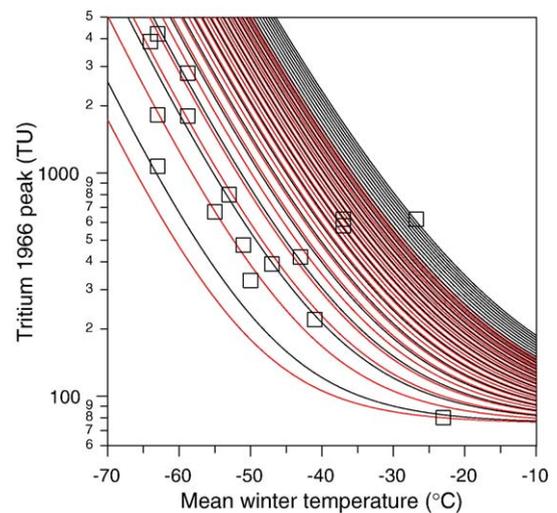


Fig. 7. 1966 tritium peak recorded at various Antarctic sites (open squares) plotted as a function of the mean local winter temperature (see Table 1). The solid lines represent the calculated 1966 tritium concentrations (after Eq. (1)) which result from the addition of various fractions of stratospheric air (from 0.001% to 0.05%) to the ambient atmosphere for two typical atmospheric pressures, respectively 600 hPa (in red) and 900 hPa (in black).

Table 1
Summary of available tritium data in Antarctica

Site	Mean annual temperature (°C)	Mean winter temperature (°C)	Elevation (m)	Mean atmospheric pressure (hPa)	Tritium peak (1966) (TU)	Natural tritium (TU)	Tritium ref.
Halley Bay	-18 ⁺	-26.8 ⁺	30	991*	620	–	[17]
Adélie Coast (site A3)	-15 ⁺	-23*	220	966*	80	5–10	[18]
Ronne Ice Shelf (site 5)	-28.6 ⁺	-37*	1000	872*	620	–	[19]
D50	-28.2 ⁺	-37*	1700	793*	580	–	[20]
D59	-34.3 ⁺	-43*	2220	739*	420	–	[20]
ST556	-38.1 ⁺	-47*	2246	737*	390	–	[21]
Talos dome	-41 ⁺	-50*	2316	730*	330	–	[21]
D80	-42.1 ⁺	-51*	2430	718*	475	–	[20]
Dronning Maud Land /DML07	-44.3 ⁺	-53*	2680	694*	800	5–20	[22]
D100	-46.5 ⁺	-55*	2810	682*	670	–	[20]
South pole (1978 pit)	-49.4 ⁺	-58.8 ⁺	2835	681 ⁺	2800	–	[8]
South pole (1975 pit)	-49.4 ⁺	-58.8 ⁺	2835	681 ⁺	1800	17–47	[8,23]
Northern Victoria Land	-33 ⁺	-41*	2960	669*	220	–	[24]
D120	-53.5 ⁺	-63*	3010	664*	1070	–	[20]
Dome C	-53.5 ⁺	-63 ⁺	3240	645 ⁺	4200	–	[25]
Dome C (1975 pit)	-53.5 ⁺	-63 ⁺	3240	645 ⁺	1820	18–32	[23,26]
Vostok	-55.1 ⁺	-65 ⁺	3480	624 ⁺	–	63	this work
Dome Fuji	-54.5 ⁺	-64 ⁺	3810	595*	3880	–	[27]

⁺Measured.

*Estimated.

measurements made on firn samples along an East–West transect from coast to coast across central Greenland, Merlivat and et al. [12] also concluded that the amount of tritium deposited at each station depends primarily on the precipitation rate, which is to say that the tritium content of the snow is basically independent of the geographical position of the station. This indicates that, unlike Antarctica where increasing

tritium levels are observed moving inland, there is no such marked continental effect in Greenland. The homogeneity of the tritium distribution over Greenland is also apparent in Fig. 8 where the various stations have recorded the same tritium history irrespective of their location and distance to coast.

The seasonal timing of the tritium maximum in Arctic precipitation is also markedly different from that observed

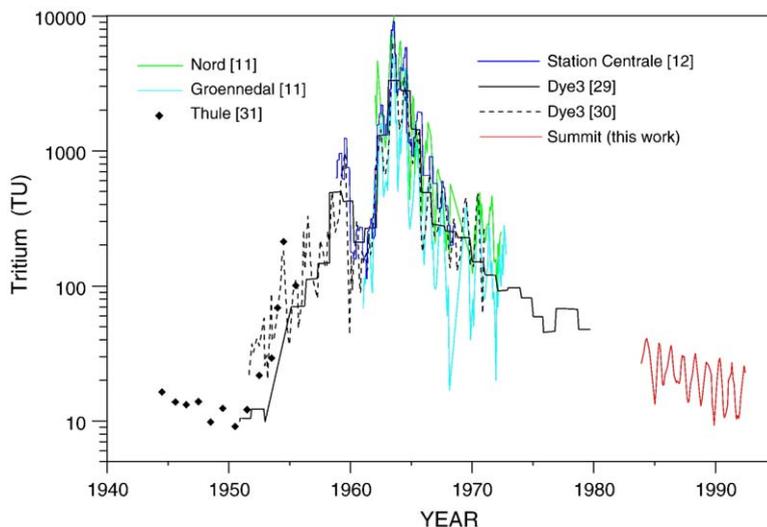


Fig. 8. Composite profile for Greenland tritium (at the time of deposition). (Data from [11,12,29–31] and this work.)

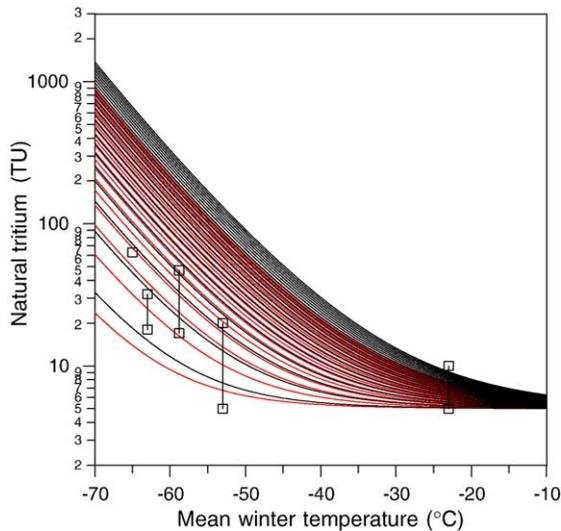


Fig. 9. Same as Fig. 7 for natural tritium in Antarctica.

in Antarctica : the annual tritium peak seen in our GRIP data, as well as in Arctic precipitations [11], occurs in spring instead of winter, thus following the same seasonal pattern as that generally observed at mid-latitude over the northern hemisphere [13,14]. Hence, Arctic tritium fallout do not seem to occur as a result of a direct injection from the stratosphere during winter as observed in Antarctica, but as a result of the dominant spring injection of stratospheric air at mid-latitude, also observed for the deposition of other stratospheric tracers such as ^7Be and ^{10}Be [15].

4.2. Natural tritium

The decay in tritium content of the stratosphere which followed the end of nuclear atmospheric tests is due to the radioactive decay of HTO (radioactive constant $\lambda=0.0563 \text{ yr}^{-1}$) and to the transfer of tritiated water vapour to the troposphere. On the other hand, tritium naturally produced by cosmic radiations is a source of HTO. Only half of the production rate of $\sim 0.25 \text{ atoms/cm}^2/\text{s}$ [1] actually takes place in the stratosphere [16], corresponding to an annual source of stratospheric tritium $\text{Prod}=33.67 \text{ mol}$. Thus, the temporal evolution of the tritium content of the stratosphere (T in TU) since $t_0=1966$ can be computed by time steps Δt from the following Eq. (2):

$$T(t + \Delta t) = T(t) - \Delta t \lambda T(t) - \Delta t T(t) / \tau + \Delta t \text{Prod} / N_{\text{H}_2\text{O}} \quad (2)$$

where $N_{\text{H}_2\text{O}}$ is the total amount (in mol) of water in the stratosphere ($N_{\text{H}_2\text{O}}=m_{\text{H}_2\text{O}}/18$ with $m_{\text{H}_2\text{O}} \sim 3 \times 10^{15} \text{ g}$) and

τ is the residence time of water vapor in the stratospheric reservoir (equal to $N_{\text{H}_2\text{O}}$ divided by the annual water vapour flux exchanged through the tropopause). We find (Fig. 6) that $\tau=6 \text{ yr}$ is the value that fits the best the stratospheric tritium data. The value of the stratospheric tritium extrapolated back to 1966 (i.e., the year of the Antarctic peak) is $\sim 80 \times 10^6 \text{ TU}$. This simple calculation also suggests that the stratosphere has now returned to its natural background concentration value of $\sim 9 \times 10^5 \text{ TU}$. This steady-state value remains high compared to natural tritium level in the troposphere.

In the same manner as in Fig. 7, Fig. 9 shows the natural tritium level in Antarctica (calculated from Eq. (1)) as a function of the local temperature for two values of the atmospheric pressure, respectively 600 hPa (in red) and 900 hPa (in black) and for various amounts of stratospheric air (α between 0.001% and 0.05%). The high natural tritium level in the stratosphere coupled with the very low atmospheric water vapor content in the Antarctic interior explains why Antarctic background tritium values are significantly higher than anywhere else. The calculated stratospheric air contribution to the 1966 peak at each individual station in East Antarctica is between 0.002% and 0.007%. Applied to Vostok, this range leads to a natural tritium level in the range 25–70 TU, consistent with the measured pre-nuclear 1953 value of 63 TU.

In Greenland, the natural background is lower, between 10 and 20 TU (Fig. 8). Although the tritium concentrations measured at GRIP between 1984 and 1992 still show a slight decreasing trend, the comparison with pre-nuclear values suggests that the legacy of the nuclear atmospheric tests is vanishing and that present-day levels may have reached the natural background.

5. Conclusion

In Greenland, the extrapolation of the decreasing trend observed on the samples from Summit covering the period 1984–1992 shows that present-day levels are close to the natural background measured at Thule [31] before the nuclear tests, between 10 and 20 TU. In Antarctica, the comparison between our measurements on samples from Vostok covering the 80's and pre-bomb levels is more difficult since only one pre-bomb sample from Vostok was available for this study and data from other sites published in the literature are of no direct use; as a matter of fact, and unlike the situation in Greenland, tritium contents are highly variable depending on the location with higher values in the drier and colder inland than on the coastal regions of Antarctica due to a maximal effect of

stratospheric air intrusions. Even though it would be interesting to further constrain pre-bomb tritium contents all over Antarctica, we can say from our measurements that in Antarctica as well, tritium contents in the 80's (70–110 TU) are not far from pre-bomb ones. However no decreasing trend can clearly be seen as it is the case in the Greenland record: the interannual variability which is very likely due to the strength of stratospheric subsidence is too important. From the extrapolation of tritium measurements in the stratosphere [7] and the data of the 1966 tritium peak recorded in various stations (Table 1), we used a simple mixing model to estimate the amount of stratospheric air required to explain the high tritium values observed in Antarctica: between 0.002% and 0.007%. Along with other stratospheric tracers, tritium in polar ice could be a useful tool to reconstruct and better understand stratospheric air inputs in the atmosphere.

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