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NATURAL TRITIUM DEPOSITION OVER ANTARCTICA AND ESTIMATION OF THE MEAN GLOBAL PRODUCTION RATE

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Abstract. Tritium content was measured in snow deposited in Antarctica before the first arrival of artificial tritium (1954), both at Dome C (74°39'S) and South Pole. The mean tritium values at the time of precipitation are equal to 32 ± 2 and 26 ± 3 TU at South Pole (1939-1942) and Dome C (1951) respectively. A seasonal pattern of natural tritium fallout identical to the artificial pattern (with a winter maximum) is observed in the South Pole profile.

The deposition rate of natural tritium is 0.6 and 0.2 tritium atoms $\text{cm}^{-2} \cdot \text{sec}^{-1}$ at South Pole and Dome C respectively. Its geographical variation over Antarctica appears quite similar to that existing for artificial tritium, clearly showing that the same mechanisms govern the deposition of natural and artificial tritium, in particular tritium injection over polar regions during Antarctic winter. A mean deposition rate of 0.37 tritium atoms $\text{cm}^{-2} \cdot \text{sec}^{-1}$, over Antarctica is derived giving an estimated mean global production rate of 0.2 tritium atoms $\text{cm}^{-2} \cdot \text{sec}^{-1}$.

This strongly suggests that natural tritium results only from cosmic ray production.

Introduction

Analysis of snow which fell before the thermonuclear tests is a useful tool to investigate natural tritium deposition in polar areas, however, few results exist for Antarctica (Taylor, 1966; Ravoire et al., 1970); available data is limited to coastal sites except one sample taken in the South Pole snow mine, which is thought to be contaminated (Taylor, 1966). The present study gives results relative to two stations on the Antarctic Plateau (fig. 1): South Pole and Dome C (74°39'S, 124°10'E). The objective of the study was a fourfold: 1) determination of the natural tritium values; 2) investigation of the seasonal pattern taking advantage of the seasonal snow stratification preservation at South pole; 3) study of the geographical distribution of annual deposition rate and comparison with the present distribution for artificial tritium; 4) estimation of the mean global production rate.

Sampling

Snow samples were taken on the wall of a ten meter deep pit, excavated 3 km from the new South Pole base. This pit covers the 1927-1978 period over which the mean annual accumulation rate was $8.5 \text{ g} \cdot \text{m}^{-2}$ (Jouzel et al., to be published). Sampling was carried out on a basis of ten samples per year. Although tritium content in atmospheric water was low at this time (less than 100 TU; 1 T.U. corresponding to a T/H ratio of 10^{-18}), special care was taken to prevent contamination of the samples both in the field (A slice of 50 cm of snow was removed along the wall before each sampling) and subsequently during conservation of the snow (kept in a plastic bag enclosed in a sealed tin box). For the period before 1955, relatively large amounts of snow (more than 250 g) were sampled.

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The same procedure was followed at Dome C (mean annual accumulation of $3.7 \text{ g} \cdot \text{cm}^{-2}$) in a 2.5 m deep pit covering a shorter period (1950-1978) (Petit et al., 1982). In addition, some «dead» samples (older than 10 000 years) were taken from the 906 m ice core (Lorius et al., 1979) to check the absence of contamination during sampling, conservation and analysis of the samples.

Tritium analysis

Owing to the low values of prebomb tritium in precipitation and radioactive decay (the tritium half-life is equal to 12.43 years; Unterweger et al., 1980), the expected tritium content of snow samples from pre-1950 snowfalls is to-day only a few TU, thereby necessitating high tritium enrichment before counting. The complete system developed in our laboratory was recently described (Jouzel and Merlivat 1981). Tritium enrichment is obtained by electrolysis in Ostlund-type cells with nickel and iron electrodes using a single-step procedure from an initial volume of 250 cm^3 to a final volume of 2.5 cm^3 before gas-counting, giving a 71-fold tritium enrichment on the average. Water is reduced by magnesium. Hydrogen then reacts with «dead» ethylene to form ethane, used as the counting gas in proportional counters with an internal anticoincidence system. Despite the high tritiated environment (highly variable between 500 and 10 000 TU or more), contamination was almost completely eliminated. Nine old ice samples were analysed giving a mean tritium value of 0.25 TU with a dispersion of 0.23 TU (1σ). This dispersion of the blank values is the main source of error and the final accuracy is around 0.3 TU whatever the sample in the tritium range under study (2-5 TU).

Results

A first series of data for the study of natural tritium deposition over the Antarctic plateau is presented below. With the above technique, a continuous sequence of 20 samples (7.40 - 7.80 m) from South Pole and another sequence of 6 samples from Dome C (2.44 - 2.50 m) were analysed.

Annual dating of the ten meter deep South Pole pit was obtained from visual stratigraphic observations and very reliably confirmed from three independent methods based on the examination of the deuterium, tritium (1960-1978) and gross β activity (1955-1978) profiles (Jouzel et al., to be published). Three summer stratigraphic indexes were observed in the 7.40-7.80 m depth increment at 7.71, 7.54 and 7.48 m attributed to the 1940, 1941 and 1942 summers respectively, indicating that the studied samples correspond to snow fallen between 1939 and 1942 (Figure 2). At Dome C the seasonal stratification of snow is not as well preserved, with the absence of some seasonal, stratigraphic and isotopic (D and T) indexes. Reliable dating can thus only be obtained from the well defined 1955 and 1965 β reference levels, due respectively to the arrival in the Antarctic area of radioactive debris from the Castle series and of the 1962 nuclear tests. In the 2.50 m pit, the 1955 level was found at a depth of 2 m. Assuming a constant annual snow accumulation of $3.7 \text{ g} \cdot \text{cm}^{-2}$ (i.e. approximately 10 cm of snow) before 1955, it is deduced that the 2.44-2.50 m

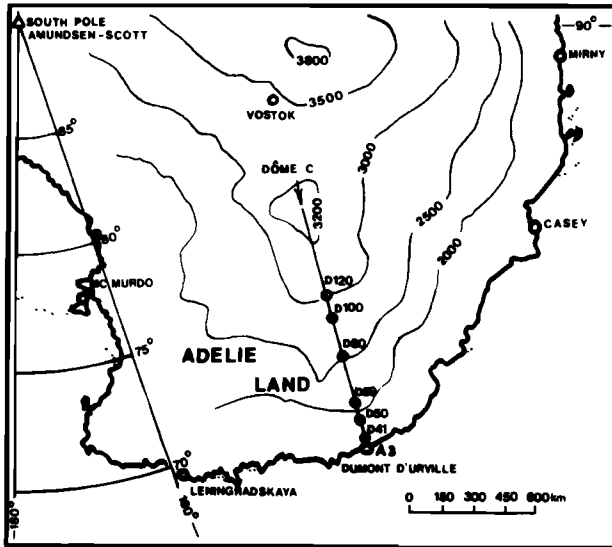


Fig. 1. Partial map of Antarctica.

increment corresponds to snow fallen during 1950. Note however that the accuracy of this dating is probably no better than 1 year.

Tritium results calculated to the precipitation date are presented in figures 2 and 3 as a function of depth, with the accuracy (1σ) indicated for each sample. For Dome C samples, a dating error of 1 year (corresponding to 6% for the radioactive decay) is taken into account.

Natural tritium values

It may be seen that the tritium content of the individual samples lies between 18 ± 2 and 46 ± 3 TU at South Pole (Fig. 2) and between 18 ± 3 and 33 ± 3 TU at Dome C (Fig. 3), illustrating fluctuations far above the analytical errors as discussed below. The mean values are equal to 32 ± 2 TU (South Pole) and 26 ± 3 TU (Dome C). The mean South Pole value of 32 TU corresponding to the 1939-1942 period is quite similar to that derived from the analyses of 6 samples of snow fallen during 1953 at this site, i.e. 34 ± 3 TU (Jouzel et al., 1979). As it was impossible to rule out contamination from the first Ivy explosion (November 1952) this figure was considered to be an upper limit for the mean natural tritium at South Pole. The present results confirm that the first artificial tritium arrived in the Southern hemisphere in 1954, as suggested then (Jouzel et al., 1979).

These figures confirm the dependence of natural tritium concentration on latitude, showing increasing values from the equator towards the pole. Measured values are less than 1 TU in equatorial precipitation and of a few TU in middle latitudes (Craig and Lal, 1961). Although the amount of data is limited, a subsequent increase can be observed over Antarctica with a mean value of 11.3 TU estimated for the 1940-1956 period at the coastal site A3 located at $66^{\circ}40'S$ (Ravoire et al., 1970), 26 TU at Dome C ($74^{\circ}39'S$) and 32 TU at South Pole (Fig. 4b). It should be noted that the tritium increase is probably also enhanced by the drastic increase in altitude towards the pole.

On a global scale, the South Pole and Dome C values compare quite well with those estimated for polar areas of the Northern hemisphere. From the study of ice cores taken near the $71^{\circ}N$ parallel, 20 TU was taken as a good estimate of the lower limit of the natural concentration of tritium in Greenland snow (Aegerter et al., 1969) and a value of 28 TU was recently estimated at Dye 3 located 600 km to the south (Theodorsson, 1977).

Seasonal pattern

An important feature of South Pole data is the appearance of an annual cycle in the natural tritium deposition, with one well marked maximum in both 1940 and 1941 (Fig. 2). As stated by Theodorsson (1977), the tritium content of precipitation was tacitly assumed to have a steady value. However, a strong indication of seasonal fluctuations was observed by that author in the Dye 3 sequence covering the 1944-1948 period (Theodorsson 1977). Although limited to two annual cycles, the South Pole profile proves the existence of seasonal variations. Moreover, it can be added that tritium peaks occur during Antarctic winter as shown by the stratigraphic dating in figure 2 (the stratigraphic levels correspond to summer snow). The seasonal pattern of natural tritium deposition thus appears quite similar to that existing for artificial tritium at the same site (Jouzel et al., 1979), which was also suggested by Theodorsson (1977), owing to the fact that both natural and artificial tritium are stored in the stratosphere after production. The fluctuations observed in the Dome C profile (Fig. 3) are also probably of seasonal origin but the results are less demonstrative since the sequence is too short and the seasonal stratification of the snow not well preserved at this site (Petit et al., 1982).

Deposition rate

The mean deposition rate of natural tritium was calculated from the mean annual accumulation values (8.5 g.cm^{-2} at South Pole between 1927 and 1978 and 3.7 g.cm^{-2} at Dome C between 1870 and 1975). This is justified since the tritium values do not appear correlated with the annual accumulation rate (at South Pole the same means are observed for 1940 and 1941 whereas the accumulation rate is three times higher in the first case). This leads to 0.6 and 0.2 tritium atom g.cm^{-2} (noted hereafter T.A. $\text{cm}^2.\text{sec}^{-1}$). This data and that derived by Ravoire et al. (1970) for the coastal site A3 ($0.5 \text{ T.A. cm}^2.\text{sec}^{-1}$) is given in figure 4a as a function of the latitude.

The geographical distribution of the natural tritium deposition rate can be compared to that of artificial tritium. The artificial tritium deposition rate was first studied along an axis from Dumont d'Urville to Vostok in East Antarctica

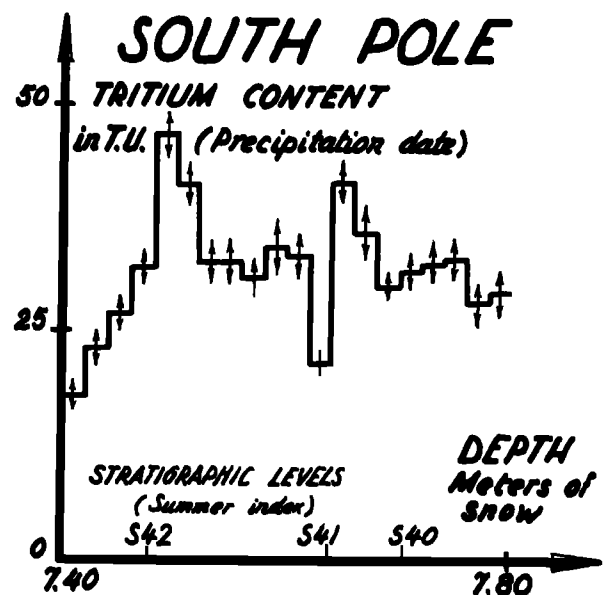


Fig. 2. Natural tritium content at South Pole over the 1939-1942 period as a function of sample depth in meters.

(Fig. 1) and subsequently at Dome C and South Pole (Merlivat et al., 1977) (Jouzel et al., 1979). The latitudinal dependence of the artificial tritium deposition averaged over the August 1956 - January 1972 period is presented in figure 4a, which shows the similarity between natural and artificial tritium fallout. For example, the fallout ratios between South Pole and Dome C are the same (2.8) for natural and artificial tritium. This similarity is obviously also apparent when the tritium contents are considered (Fig. 4b, lower graph).

These seasonal and geographical similarities show that the same mechanisms govern the deposition of artificial and natural tritium. The high tritium values and the existence of a winter maximum registered in each case at South Pole, strongly suggest that tritium is transported from the stratosphere to the troposphere over polar regions during the Antarctic winter. After its production, the tritium formed at high stratospheric altitudes is brought down within the circumpolar vortex, and two mechanisms can be proposed to explain its transfer to the troposphere.

a) No temperature inversion exists at the South Polar tropopause during the Antarctic winter and when these particular conditions exist a continuous rapid exchange of air between the lower polar stratosphere and the troposphere may possibly occur (Taylor, 1968).

b) It was also suggested (Stanford, 1973) that stratospheric clouds can be formed and subsequently precipitated in the higher troposphere during winter. In this process, a significant percentage of tritium-rich water could be removed from the stratosphere (Jouzel et al., 1979). Note however that the first mechanism is probably the more effective one (as the second implicates very low temperatures) except for especially cold years at stratospheric levels.

Our results can be used to calculate an average value of the natural tritium deposition rate over Antarctica. For this, the latitude-deposition rate relationship, sufficiently well known for artificial tritium, was applied for natural tritium. For the area south of 74°S, a linear increase with latitude between Dome C and South Pole was used. This leads to a value of 0.37 T.A. cm⁻².sec⁻¹ south of 66°S, an area covering a large part of Antarctica. However, as the region south of Dome C is poorly documented, a second calculation was carried out assuming that the higher value of 0.6 T.A. cm⁻².sec⁻¹ registered at South Pole applies to

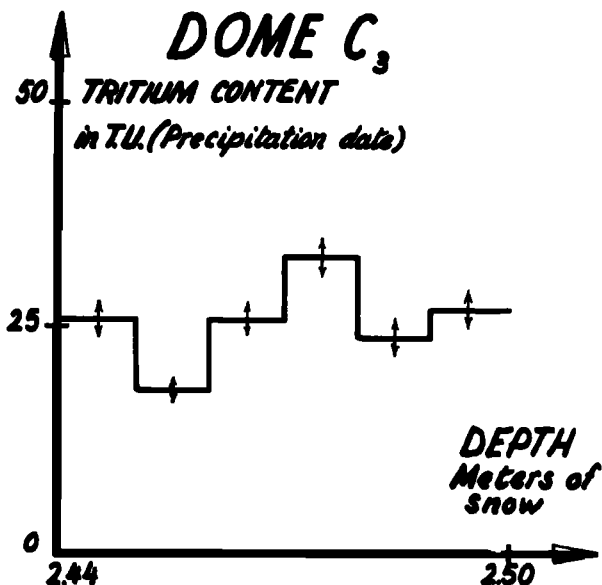


Fig. 3. Natural tritium content at Dome C during 1951 as a function of sample depth in meters.

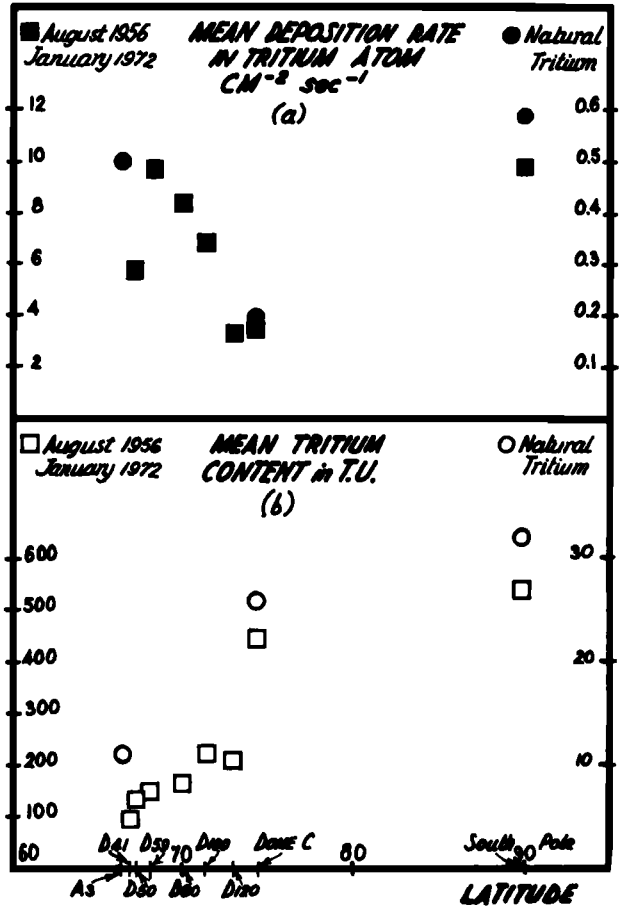


Fig. 4. Mean tritium content in T.U. (4) and tritium fallout in tritium atoms cm⁻².sec⁻¹ (4a) along the axis drawn on figure 1 from A3 to Dome C and at South Pole, as a function of latitude. The squares represent artificial tritium with mean values calculated between August 1956 and January 1972 and the circles represent natural tritium. Note that the scale for natural (right) and bomb (left) values have been selected to show the close relationship between the distributions.

all the region south of Dome C. This leads to a value of 0.48 T.A. cm⁻².sec⁻¹ which can be considered as an upper limit of the mean natural-tritium rate deposition over Antarctica.

Global production rate

Three sources are generally accepted for natural tritium production 1) production by galactic cosmic rays estimated to be about 0.2 T.A. cm⁻².sec⁻¹ (Craig and Lal, 1961 ; Nir et al., 1966 ; Teegarden, 1967) / 2) production by solar flare accelerated particles which, according to Nir et al. (1966) appears to be negligible compared with production by galactic cosmic rays ; 3) accretion of solar tritium with an estimate of 0.4 T.A. cm⁻².sec⁻¹ (Flamm et al., 1962). Since the intensity of galactic cosmic rays is in antiphase with solar activity, there should exist a negative correlation between solar activity and the first production term. On the other hand the two other processes should show a correlation in phase with solar activity.

From their Greenland data, Aegerter et al. (1969) calculated a mean deposition rate of 0.85 T.A. cm⁻².sec⁻¹ and estimated the mean global production rate in the following way. Owing to the strong latitudinal mixing throughout the troposphere and the efficiency of exchange through

the tropopause at high latitudes, they assumed that the natural tritium deposition pattern is similar to the production pattern. A correction of about 5 %, to account for tritium decay in the stratosphere (mean residence time about one year) was then applied leading to a production rate in the Arctic of $0.9 \text{ T.A. cm}^{-2} \cdot \text{sec}^{-1}$. Taking into account the latitudinal dependence of tritium deposition, estimated to give twice the global average at latitudes higher than 70° , a value of $0.45 \text{ T.A. cm}^{-2} \cdot \text{sec}^{-1}$ is obtained for the mean global production rate. Although, the hypothesis of similar deposition and production patterns needs to be thoroughly investigated, we applied the same reasoning to the above value for the Antarctic deposition rate leading to a mean global production rate of $0.2 \text{ T.A. cm}^{-2} \cdot \text{sec}^{-1}$ (which increases to 0.25 if the upper limit of $0.48 \text{ T.A. cm}^{-2} \cdot \text{sec}^{-1}$ over Antarctica is considered).

According to Aegerter et al. (1969), the strong anticorrelation between tritium deposition at station Centrale (Greenland) and solar activity shows that cosmic ray production is dominant. However, it must be noted that a strong increase in tritium deposition is observed during two of the three solar activity maxima, suggesting a contribution from the sun (sources 2 and 3) for which a value of $0.2 \text{ T.A. cm}^{-2} \cdot \text{sec}^{-1}$ can be considered to set an upper limit. On the other hand, Craig and Lal (1961) had concluded as early as 1961, from the good agreement observed between the geochemical tritium inventory and cosmic ray data, that there is no evidence for accretion of tritium from an extra terrestrial source.

Although there are many assumptions in our interpretation, this study strongly supports the last conclusion. The remarkable agreement between the calculated natural tritium production rate from cosmic rays alone and our estimate from Antarctic snow ($0.2 \text{ T.A. cm}^{-2} \cdot \text{sec}^{-1}$ in each case) strongly suggests that the total contribution from the sun is negligible and that natural tritium results only from cosmic ray production.

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