Fifth. Indian Expedition to Antarctica, Scientific Report, 1988
Department of Ocean Development, Technical Publication No. 5, pp. 181-188

Polar Fallout of Radioisotopes ^{32}Si, ^{7}Be, ^{210}Pb, ^{137}Cs and ^{239}Pu at Dakshin Gangotri, Antarctica

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Abstract

The concentrations of radioisotopes ^{32}Si, ^{7}Be, ^{210}Pb, ^{137}Cs and ^{239}Pu have been measured in the snow samples in the shelf ice near Dakshin Gangotri Station, Antarctica. The annual fallout of Cosmic Ray produced ^{32}Si — the first measurement of its kind in Antarctica—has been estimated to be 3×10^{-5} dpm/cm^2. yr which corresponds to the global production rate of ^{32}Si to be 0.8×10^{-4} atoms/cm^2 sec, using half-life of ^{32}Si to be 140 yrs. This estimate is lower than that calculated by Lal and Peters (1967) which suggests that the half life of ^{32}Si is closer to 270 yrs as observed by geophysical methods. The fallout of the other natural radioisotope ^{7}Be and ^{210}Pb are consistent with the earlier work. As expected, the fallout of artificial radioisotopes ^{137}Cs and ^{239}Pu are an order of magnitudes lower than the peak production during the nuclear explosions tests conducted during last 3 decades.

Introduction

During the past three decades, the fallout of natural and artificial radioisotopes has been extensively studied for the understanding of atmospheric circulation and washout processes (Bhandari and Rama, 1963; Koide et *al*, 1982; Lal and Peters, 1967; Lal et *al*, 1966, 1974; Turekian, 1977). Polar ice sheets maintain a detailed history of fallout materials deposited from the atmosphere to the earth's surface through the latitude belt of 60°-90° in both the hemispheres. Measurements of the concentrations of various radioisotopes in dry precipitation (fresh snow) occurring in Polar region helps in dating the ice and in estimation of accumulation rates in the remote icy regions where direct measurements pose several logistics problems. For the purpose of dating ice samples, the average fallout values of different radioisotopes, selected on the basis of their half lives, covering the time span of processes under investigation, is to be known. The fallout of different radioisotopes has been studied quite extensively in the Arctic Polar region compared to that in Antarctica during past few decades due to remoteness of the region and logistics problems.

Fallout of different natural and artificial radioisotopes like Cosmic ray produced isotopes ^{32}Si (tl/2 = 140 yrs; Somayajulu er *al.*, 1987), ^{7}Be (tl/2 = 53 days), natural isotopes ^{210}Pb (tl/2 = 22.3 yr), and artificial radioisotopes ^{137}Cs (tl/2 = 30 yrs), and ^{239}Pu (tl/2 = 2.44 X 10^4 yr) (produced as a result of testing of nuclear devices during the past four decades) in the snow samples collected near the Indian Station Dakshin Gangotri (D.G.) (70°S, 12°E), were studied during the Fifth Indian Scientific Expedition to Antarctica. Some of these natural radioisotopes like ^{7}Be, ^{32}Si are also produced during the testing of nuclear explosions and upset the natural balance. However, these natural levels have been upset only for brief periods (Lal et *al*, 1979). For example, ^{32}Si produced during the nuclear explosions is insignificant (< 1% of its natural level) to invalidate its applications to glaciology (Nijampurkar *et al.*, 1982, 1985). Silicon-32 and Lead-210 have been used in past to date glacier ice from Himalayas; Alps and Greenland (Nijampurkar *et al.*, 1982; Gaggler *et al*, 1983; Clausen, 1973).

Whereas the Polar fallout of some of these radioisotopes like ^{210}Pb, ^{137}Cs, ^{239}Pu, have been measured in past at different locations in Antarctica (Picciotto et *al*, 1968; Koide et *al.*, 1982), the estimation of ^{32}Si fallout is perhaps the first measurement of its kind in Antarctica and Southern hemisphere. Measurement of ^{32}Si in Antarctica would not only help in understanding the Polar fallout of ^{32}Si but also improve our understanding on the global production rate of ^{32}Si and its half life (t 1/2 = 100—300 yrs) yet not known accurately.

Average annual fallout values of ^{7}Be and ^{210}Pb in Antarctica are useful in dating fresh snow and old ice (upto 100 yrs).

Activity levels of artificial isotopes ^{137}Cs and ^{239}Pu in the Polar regions of Antarctica would provide data for better understanding of the nature and behaviour of nuclear debris as well as their travel through the atmosphere and diffusion of these radionuclides in these glacial columns. The present day fallout values of these radioisotopes as compared to that observed in the past few decades, based on ice samples obtained from shallow ice cores is expected to improve our knowledge about the atmospheric circulation and washout processes gained by previous researchers.

Materials and Method

(a) Sample collection

Snow samples ranging from 4-300 kg were collected from the shelf ice at Dakshin Gangotri Station near the Indian Bay in Antarctica. The top few inches of the surface snow was removed (to avoid any contamination) and the samples collected upto 50 cms so as to represent the annual snow deposition. The snow samples were melted in clean plastic drums and acidified to pH-3 with- 1:1 HNO_3. Stable carriers were added for different isotopes. After homogenisation the activities were scavanged by iron hydroxide in an ammonical medium following the procedures described earlier (Nijampurkar er *al.*, 1982; Koide et *al.*, 1982).

(b) Chemical Procedures and Counting Methods

The chemical procedures for radiochemical separation and purification of radionuclides have been discussed elsewhere (Nijampurkar *et* al., 1982; Koide et al., 1982).

The counting procedures required that the beta activity of ^{32}P from ^{32}Si and ^{210}Bi from ^{210}Pb was followed for several weeks on a gas flow GM counter with NaI (Tl) anticoincidence having a background of about 1.5 CPh and counting efficiency of 35%. A typical decay curve for ^{32}P and growth curve for ^{210}Bi is shown in Figs. 1 and 2.

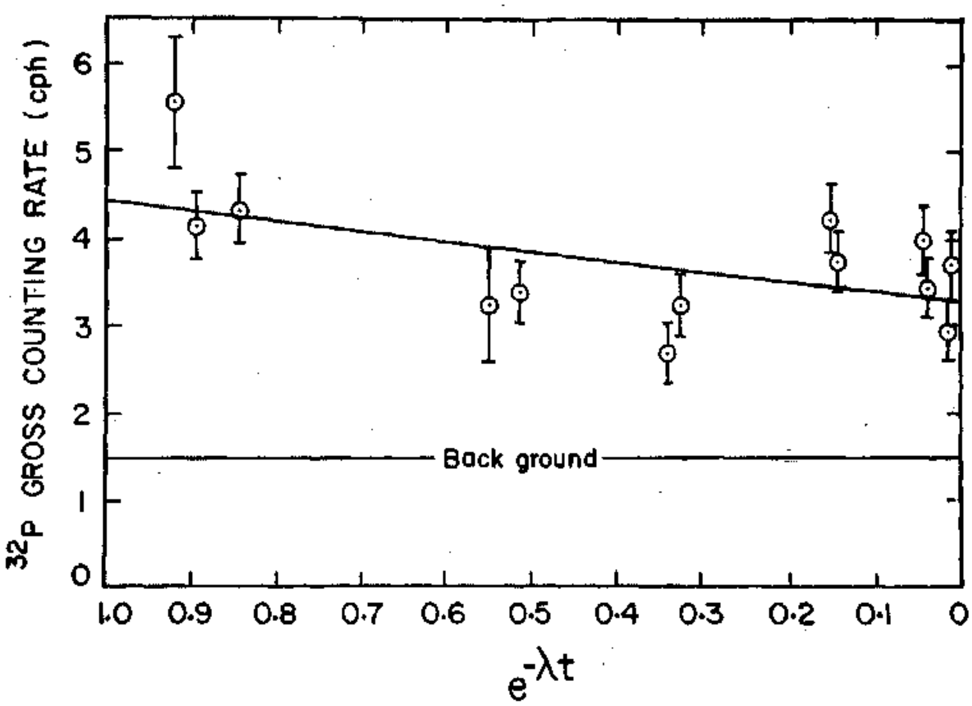

Fig. 1. Variation of gross ^{32}P beta activity milked from surface ice melt water as a function of $e^{-\lambda t}$ The sample has been counted for over 2 months to observe the decay of ^{32}P activity.

The γ activity of ^{7}Be and ^{137}Cs by non-destructive method of analysis was counted on a HPGe detector system located in a 10 cm lead shield. Background in ^{7}Be was 0.14+0.006 cpm whereas for ^{137}Cs was 0.11±0.01 cpm. The counting efficiency of the system is 3.8%. The typical spectra of the γ activity of ^{7}Be (E_{max} = 0.477 Mev), is shown in Fig. 3.

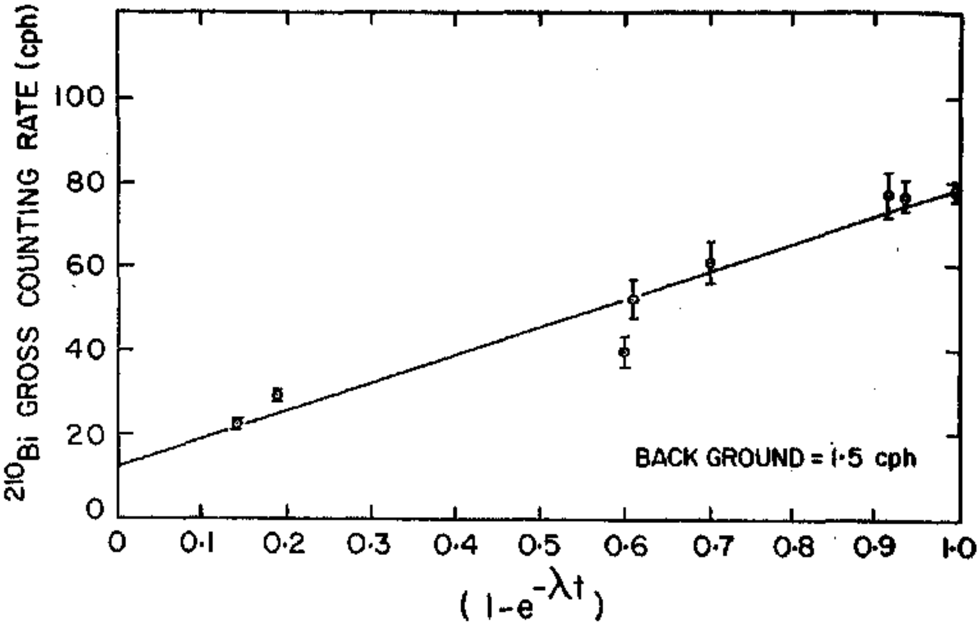

Fig. J. *Growth curve of ^{210}Bi extracted from ^{210}Pb from the windborne snow collected near DG. Station. The growth has been observed over a period of about one month.*

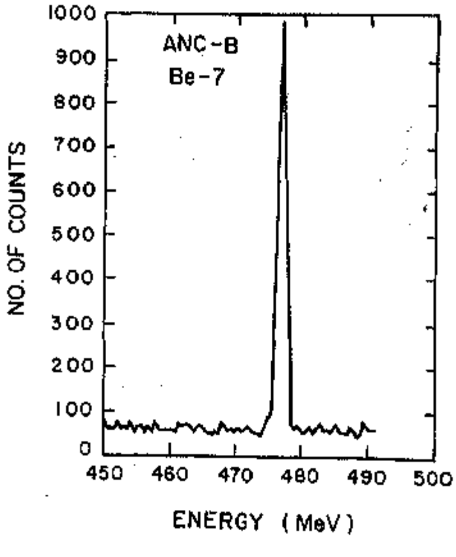

Fig. 3. A typical spectra of ^7Be activity extracted from a windbome snow sample near D.G. Station. A sharp peak of gamma activity has been observed at the energy 0.477 Mev as expected.

The ^{239}Pu activity was scavanged by $Fe(OH)_3$ matrix from 300 L of snow melt water at the site. The radiochemical separation of ^{239}Pu from the matrix and its assay was carried out at BARC, Bombay using $\propto$ spectrometry by the procedures discussed earlier (Pillai, 1987—Personal communication).

Results and Discussion

Concentrations (dpm/L) and fallout (dpm/cm^2 yr) of the radioisotopes ^{32}Si,. ^{7}Be, ^{210}Pb, ^{137}Cs and ^{230}Pu in snow precipitation are given in Table I.

Table I. Concentration and fallout of Radioisotopes ^{7}Be, ^{210}Pb, ^{32}Si, ^{137}Cs and ^{239}Pu at Dakshin Gangotri Station, Antarctica (Samples collected on 4-1-1986)

Radio-isotope	Half-life	Mode of decay and energy (mev)	Volume of water processed (L)	Concentration (dpm/L)	Annual fallout (dpm, cm^2. yr)
^{7}Be	53 days	E_γ = 0.477	25	(8.8+0.2)	3.0
210pb	22.3 yrs	Emaxβp- = 0.061	25	(0.62+0.10)	0.12
^{32}Si	140 yrs	$E_{„ulx}$fi- = 0.21	300	(0.78+0.4) x1O"^{3}	3.0 XlO^{-5}
^{137}Cs	30 yrs	E-y = 0.661	4.5	(0.2+0.2)	8.0
^{239}Pu	2.44x10^{4}yrs	E_v = 5.15	300	(0.32+0.20) X10"^{5}	1.2X10^{-9}

(i) Polar fallout of ^{32}Si

Concentration of ^{32}Si in the snow accumulated over the shelf-ice (0-30 cms) at Dakshin Gangotri station, which represents annual precipitation has been observed to be (0.78 ± 0.4) X 10^{-3} dpm/L. On the basis of this available single measurement at 70°S and the average precipitation of ~ 40 cms [mean of estimated precipitation, 30 cms, (Nijampurkar er *al.,* 1988, this volume) and the average precipitation of 50 cm's in the coastal Antarctica regions] in this region the fallout of ^{32}Si has been estimated to be 3 X 10^{-5} dpm/cm^2. yr. This can be taken as average fallout of ^{32}Si in the latitude belt, of 30—90° in the absence of data at other latitudes in this belt, and the fallout of ^{32}Si being independent of the latitude > 30° (Lal and Peters, 1967; Clausen, 1974). It has been found from the cosmic ray source function that approximately 40 and 60 percent of ^{32}Si activities will be deposited in the 0-30° and 30-90° cells respectively. Hence the fallout of ^{32}Si should be lower in 0-30°S cell by a factor of 1.5 i.e. 2 X 10^{-5} dpm/cm^2. yr than that observed in 30-90°S cell.

The global mean fallout of ^{32}Si is estimated to be 2.5 X 10^{-5} dpm/cm^2. yr based on the measured value of 3 X 10~^{5} dpm/cm^2 yr for the 30-90°S cell and the calculated value of 2 X 10^{-5} dpm/cm^2 . yr for 0-30° tropical cell (the surface areas are same for the two cells). In fact the average fallout in the 0-30°S and 30-90°S should be measured by collecting samples at every 10°latitude in Southern hemisphere, which will give correct picture of the mean global fallout of ^{32}Si in the Southern hemisphere.

The mean global fallout of 2.5 X 10^{-5} dpm/cm. yr of ^{32}Si corresponds to a mean global production rate of 0.8 X 10^{-4} atoms/cm$^{2.}$ sec. (calculated using 140 yr as the half life of ^{32}Si). This estimate is lower than the calculated value of 1.6 X 10^{-4} atoms/cm^2, sec. (Lal and Peters, 1962). This discrepancy could be due to the uncertainty either in the cross sections used for calculating the ^{32}Si production rate or in the half life of ^{32}Si. If it is due to the latter, to obtain agreement with the measured production rate, the half life of ^{32}Si may be closer to 270 yr, similar to the values reported by Clausen (1973) and Demaster, (1980). More direct measurements in this region might help to settle the discrepancy.

Additionally, the higher values of ^{32}Si observed in the circumpolar waters of the Atlantic waters (Somayajulu *et* al., 1987) can be explained by the high values of ^{32}Si observed in the present work compared to that of 0.5 X 10^{-3} dpm/L in the same latitude belt in northern hemisphere and the subsequent contribution of meltwaters of the Antarctic ice sheet to coastal Antarctic waters and their mixing with the mid latitude ocean waters in the Southern hemisphere.

(ii) Fallout of ^7Be and ^{210}Pb

The observed concentration of 8.8 ± 0.2 dpm/L for ^7Be is consistent with the earlier estimates (Bhandari *et* a/., 1984) and comparable to that observed at tropical latitudes in the northern hemisphere from 15° to 31°N (Lal *et al,* 1979). This confirms that the snow samples collected for analysis were about 6-8 months old and would have been deposited in the shelf ice from June to September, 1985 (Table I).

The ^{210}Pb activity in the fresh snow sample is observed to be 0.62±0.1 dpm/L which is consistent with the earlier findings (Nijampurkar *et* al., 1984) and estimates a fallout value of 0.15 dpm/cm^2 yr. The shallow ice cores from Antarctica near the South Pole far away from the coastal areas have been dated by ^{210}Pb using the surface ice value of 1.8 dpm/L (Picciotto *et al.,* 1968) The fallout values of ^{210}Pb in the northern hemisphere in Greenland, Alps and Himalayas lie in the range of 3 to 9 dpm/L (Crozaz and Langway, 1966; Nijampurkar et *al.,* 1982; Gaggler, 1983). The ^{210}Pb values observed at Dakshin Gangotrj at shelf ice near the coast are slightly lower than those observed near the pole but are consistent within the statistical errors. This can be explained by partial contributions from the coastal seawater moisture which do not have any ^{210}Pb activity condensing over the shelf ice in summer, thus lowering the ^{210}Pb concentration in the snow deposited over the near coastal areas. However, assuming the total fallout of ^{210}Pb remains constant over a period of few hundred years, the deep seated ice can be safely dated using the ^{210}Pb value from the average surface ice values in Antarctica.

(iii) Fallout of ^{137}Cs and ^{239}Pu

The artificial radioactivities of ^{137}Cs and ^{239}Pu in surface snow melt waters at Dakshin Gangotri are found to be 0.2 and 0.32 X 10^{-5} dpm/L (Table I) respectively, which are very close to background and order of magnitudes lower than the peak fallout observed during last 4 decades. The peak activities of ^{137}Cs (15 dpm/L) and $^{239+240}Pu$ (0,2 dpm/L), produced as a result of testing of nuclear devices in early 60's at the Dye-3 station of Greenland are reported by Koide et a/. (1982).

This work suggests that there has been no testing of nuclear devices in the atmosphere during the late 80's and that the activity produced in late 70's by French tests in the Southern Hemisphere has been completely washed out during the last decade.

Even though no activity of ^{137}Cs and ^{239}Pu has been observed in surface snow samples, the systematic nuclear records preserved in the Antarctic ice sheet can be investigated if a shallow ice core of about 20 metres is raised from this region. Both these radioisotopes can be used as time markers to estimate past accumulation rates of ice in Antarctica.

Suggestions

This preliminary investigation suggests that (i) systematic efforts are to be made to raise shallow ice cores upto 100 m to understand past nuclear and climatic records, accumulation rates of ice, and deposition of windborne as well as extra-terrestrial matter; and (ii) for better understanding of the knowledge about the global production rate of ^{32}Si and its half life, more ^{32}Si fallout measurements need to be made in Antarctica at different locations.

Acknowledgement

The authors are extremely thankful to Prof. S.P. Pandya, Director P.R.L. for giving us financial support and to Profs. B.L.K. Somayajulu and S. Krishnaswami for useful scientific discussions. Help in providing logistics support for collecting samples in adverse circumstances rendered by Scientists of the Geological Survey of India and other members of the expedition is gratefully acknowledged.

Thanks are due to Prof. H. Pillai of Health Physics Division of BARC, Bombay for measuring ^{239}Pu fallout in snow samples, and for useful discussions.

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