

Fission Products in Antarctic Snow, A Reference Level for Measuring Accumulation

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Abstract. In the area around King Baudouin Station (70°S, 24°E), it is shown that a reference horizon, easy to identify, was formed by the stratospheric fallout of radioactive debris from thermonuclear bomb tests. Gross β activity and Sr^{90} have been measured in snow and firn samples from 1935 to 1960. The samples were dated by stratigraphy and by oxygen isotope-ratio measurements. The contribution of natural radionuclides is negligible, and the gross β activity can be taken as a measure of the fission products concentration. The following average values were found: from 1955 to 1960, 14 dpm/kg of snow; beginning of 1955, 22 dpm/kg ($\text{Sr}^{90} = 4.5$); 1953 and 1954, 2 dpm/kg; and from 1935 to 1952: 0.5 dpm/kg ($\text{Sr}^{90} < 0.1$). The sharp tenfold increase in the β activity at the beginning of 1955 is attributed to the sudden release in the antarctic troposphere of fission products from the Castle thermonuclear tests series (March 1954). The previous and first thermonuclear test (Ivy, November 1952) is less marked. The β activity before 1952 is essentially due to Pb^{210} and K^{40} . There are indications that this radioactive horizon has been formed at the same time over the whole ice cap, but direct checks are needed on more, well-dated firn profiles.

Introduction. A major problem in glaciology of the polar regions and especially of the Antarctic is the estimation of the current ice budget of ice caps. The wastage is very difficult to determine and will not be considered here. The evaluation of the supply in the present budget of the ice caps depends essentially upon knowing the rate of annual accumulation, the average value of which is still not well known despite the great progress made in recent years.

Direct measurements of the accumulation are scarce. In Antarctica the only positive results come from stations established for several years, stations which are scarce in number and mainly located on the eastern coast of the continent.

The value of the annual accumulation away from these stations is generally deduced from stratigraphic observations. These observations are subjected to a great deal of personal interpretation, especially on the polar plateau where the accumulation is very poor (from 5 to 15 cm of water annually) and the seasonal contrasts in the firn are scarcely indicated.

A valuable aid to the interpretation of the stratigraphic characteristics of the firn will be the geochemical methods currently being devel-

oped, notably the isotopic methods based upon the seasonal variations of the stable isotopes of hydrogen or oxygen [Epstein and Sharp, 1959] or upon the decay of radioactive isotopes such as tritium [Kaufman and Libby, 1954; Oeschger et al., 1962], Pb^{210} [Goldberg, 1962], C^{14} [Coachman et al., 1958; Scholander et al., 1962]. These methods are still in their developmental phase. Furthermore they require laboratory equipment not currently available at the polar stations or, a fortiori, in the field.

An ideal method for measuring the mean accumulation of the last several years should consist in finding a reference level which corresponds to a well-defined date which is easily identifiable and which extends over the entire continent.

Because of the rarity of stations and the discontinuity in time of observations, it is practically impossible at this stage to find man-made reference levels.

Several studies are being made of the possibility of using natural reference horizons: layers of volcanic ash [Ragle et al., 1960], frequency of micrometeorites [Thiel and Schmidt, 1961; Langway, 1962], or pollens, but no positive result has yet been obtained.

It has already been suggested that in the polar regions, where the temperature is per-

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manently below 0°C, a reference level could be effected by the radioactive fallout from nuclear bomb tests [Picciotto *et al.*, 1962; Drevinsky *et al.*, cited by Sharp and Epstein, 1962]. In a general paper on radioactive fallout, Martell [1959a] demonstrates implicitly the applicability of this method in Greenland.

In the present study, we demonstrate that, in the area of King Baudouin Base (70°S, 24°E), a horizon, probably valid for all Antarctica, was established in 1955 by the fallout of the artificial radionuclides produced by the first thermonuclear explosions. This horizon is easily identifiable by a simple radioactive measurement.

Radioactive fallout. Since the first works on this subject, a considerable amount of literature has been devoted to the circulation and behavior of fission products released in the atmosphere by nuclear explosions. Here we can only quote a few reviews papers [U. S. Government, 1959; Libby and Palmer, 1960; Martell, 1959a, b; Libby, 1959; Machta, 1961; Friend *et al.*, 1961]. Many important problems still remain controversial; we should recall the following conclusions, which will be especially useful to us further on:

1. The behavior of fission products depends upon the latitude, the altitude, and the season in which they have been released. In the troposphere their residence time is of the order of a month. In the stratosphere the residence time is of the order of several months for fission products injected into the polar stratosphere, and it varies from 1 to 5 years in the equatorial stratosphere. Fission products introduced into the equatorial stratosphere spread rapidly in their latitudinal band of injection but migrate slowly toward the poles.

2. The transport mechanisms, vertical as well as horizontal, of fission products in the stratosphere are not clearly understood. Several models of the general processes of transfer in the stratosphere have been proposed, but none has yet been definitively established. Depending upon the particular author, the transfers are made either by an organized meridional circulation as in the Dobson-Brewer model [Stewart *et al.*, 1958; Goldsmith and Brown, 1961] or that of Goldie [Libby and Palmer, 1960], or by wide-scale turbulent diffusion [Feely and Spar, 1960].

3. Worldwide radioactive fallout is stronger

in the northern hemisphere than in the southern. In both hemispheres, the fallout is subjected to seasonal fluctuations, and it displays a maximum intensity in the spring.

4. Fission nuclear bombs (of the order of several kilotons) yield almost exclusively tropospheric fallout. Thermonuclear bombs (of the order of several megatons), on the other hand, always send a certain amount of fission products into the stratosphere, the amount depending upon the type of device and the conditions of detonation [Libby, 1959].

5. From the chronology of nuclear bomb tests, we ought to retain the following data (Figure 2):

From 1945 to 1952 only fission bombs were detonated, largely in the northern hemisphere. These explosions yielded only tropospheric debris which did not have any appreciable chance of reaching the Antarctic. The total amount of fission products released by the fission bombs is negligible compared with that produced by the later thermonuclear bombs.

The first thermonuclear explosion occurred in November 1952, in the Bikini-Eniwetok area (Ivy test). It was a ground-level explosion whose power was probably several megatons, relatively weak compared with the later explosions. It was followed, in this same area, in March 1954, by the first series of high-yield thermonuclear tests (Castle test). The testing of the thermonuclear bombs by the United States, the Soviet Union, and Great Britain continued until the autumn of 1958. Nuclear tests in the atmosphere were suspended in November 1958 only to be resumed with increased power in September 1961.

If the radioactive debris from nuclear tests is to be used as a marker in the antarctic firn layers, the date of arrival of the fission products on the ground must be well known and must be the same over the entire continent. The more sudden this arrival, the more evident will be the marked level.

After its precipitation, the snow carrying down the fission products must not have undergone melting or mixing sufficient to affect a thickness greater than that of the annual accumulation.

The date and the mode of arrival of fission products from Ivy and Castle tests in Antarctica are difficult to predict theoretically. It is

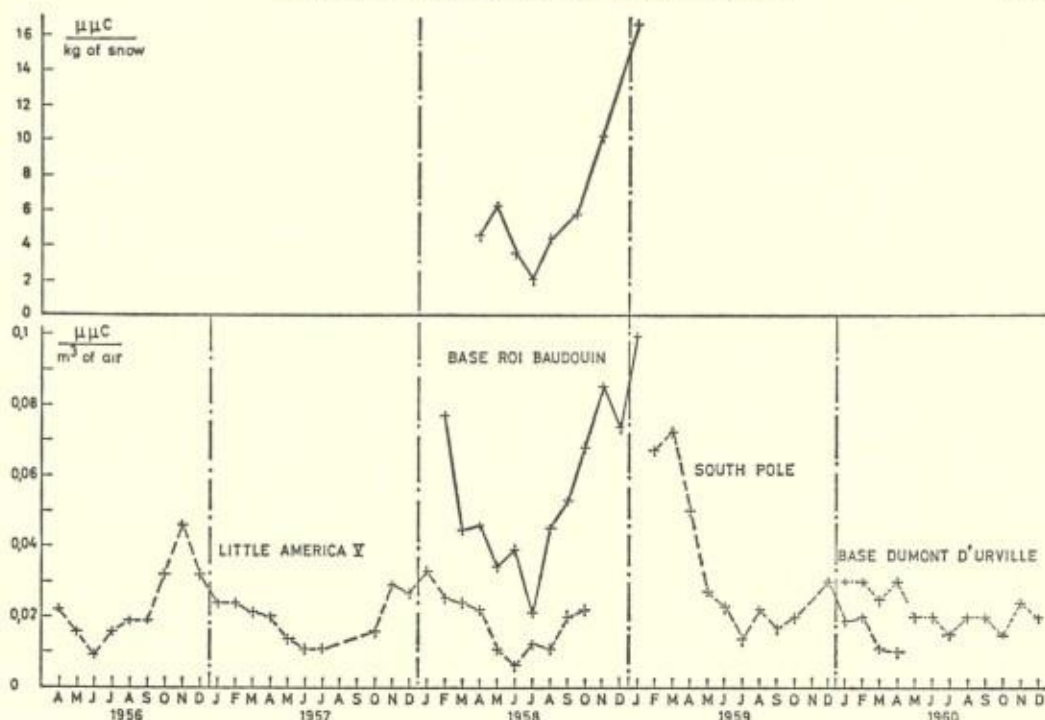


Fig. 1. Concentration of fission products in the air and in the precipitations in Antarctica. Little America 5 and South Pole from Lockhart [1960]. King Baudouin Base from Picciotto *et al.* [1962]. Base Dumont d'Urville, from Ardouin *et al.* [1961].

likely that the first important transfer occurred during the austral spring of 1954. But it is important that this date be established from samples of snow precipitated on known dates or from the measurement of the air radioactivity since 1952 at least.

Results already obtained for the polar regions. In Antarctica, a systematic measuring of air radioactivity at ground level began in April 1956 at station Little America 5 [Lockhart *et al.*, 1958; Lockhart, 1960]. Measurements have been made at King Baudouin Base since February 1958 [Picciotto, 1958; Picciotto *et al.*, 1962]. Similar measurements were also made at Base Dumont d'Urville in 1960 [Ardouin *et al.*, 1961]. The results (Figure 1) show the existence of fission products in the air at the ground level since 1956, with a concentration nearly 50 times lower than that found in the northern hemisphere. A seasonal cycle is clearly indicated with a minimum of radioactivity in the winter. The maximum occurs between November and March, depending upon the year.

The only available data before April 1956 are a series of measurements of Sr^{90} in samples of surface snow collected during January and February 1955 at various spots on the antarctic coast; these probably represent the precipitations of the spring of 1954 [Martell, 1959a]. The mean activity of Sr^{90} found is 3.4 ± 0.5 dpm/kg of snow.

In Greenland, Sr^{90} has been measured [Martell, 1959a] on snow-core samples from site 2 (77°N , 50°W). The dating of the samples was checked by stratigraphy and by oxygen isotope ratio variations. They showed negligible Sr^{90} deposition up to the summer of 1953. The mean activity was found to be approximately 1 dpm/kg in 1953 and 1954; it increased to 3.8 dpm/kg after the summer of 1955.

Sampling. The method proposed here must be checked on samples of snow precipitated at known dates since at least 1952. In the absence of precipitation samples collected at these early dates, we must rely on samples of firn dated by the methods mentioned before.

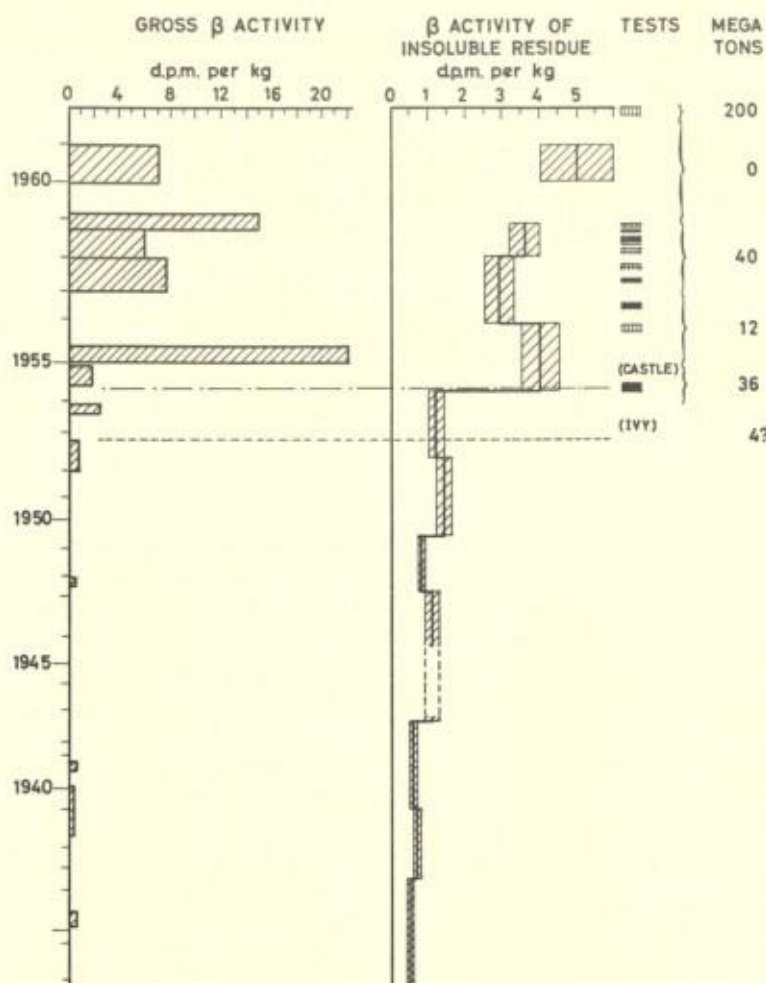


Fig. 2. Radioactivity of the snow, as a function of the date of precipitation at King Baudouin Base. From left to right: Year attributed [Gonfiantini *et al.*, 1963]. Gross β activity. Insoluble β activity. Date of main thermonuclear test series. Total fission yield from nuclear events (partly from Libby [1959]) in megatons (approximately 0.1 Mc of Sr^{90} is produced per megaton [Martell, 1959a]).

We have already emphasized the difficulty of identifying annual layers in firn profiles. However, we think that we have a set of samples meeting the required conditions. It was taken from the 16-m-deep glaciological pit of King Baudouin Base ($70^{\circ}26'S$, $24^{\circ}19'E$) on the ice shelf of the Princess Ragnhild Coast.

The stratigraphy of this pit dug by Y. Van De Can (Belgian Antarctic Expedition 1959) has been carefully studied by W. De Breuck (Belgian Antarctic Expedition 1960).

The identification of the annual layers is based upon the compared interpretation of the

isotopic and stratigraphic profiles, both of which show seasonal variation. A detailed discussion will be found in Gonfiantini *et al.* [1963].

The top of this section corresponds to January 1958. The mean accumulation rate was found to be 38 cm of water.

In Figure 2, we have reported the annual identification of the summer surfaces. These surfaces generally separate the spring or summer precipitations (November to January) from the autumn precipitations (March).

Even though there are uncertainties in the interpretation of parts of the profile, we think

TABLE 1. Long-Lived Natural Radionuclides in Snow, King Baudouin Base

Radionuclide	dpm/kg	Reference
Pb ²¹⁰	≤1	[Picciotto <i>et al.</i> , 1962; G. Crozaz, unpublished]
K ⁴⁰	≤0.7	From the K concentration [Brocas and Delwiche, 1963]
U + Th	<0.5	From the α activity [Picciotto <i>et al.</i> , 1962]
Cosmic-ray- produced Be ¹⁰ and Si ³²	<0.01	Estimated [see for instance Kharkar <i>et al.</i> , 1962]

that the identification of the annual layers leaves no doubt that the first 6 m represent the 1948-1958 interval.

The samples more recent than 1958 are represented by surface snow or precipitations collected in 1958 and 1960.

Natural radioactive nuclides in the snow. The most significant method for expressing and comparing the concentrations of fission products is to measure the concentration of one or several individual nuclides; generally Sr⁹⁰ is selected.

Measuring Sr⁹⁰ involves a series of radiochemical separations which are scarcely compatible with the criteria of simplicity and rapidity that we are looking for.

Fortunately, the Antarctic presents an exceptional situation which allows us to avoid any radiochemical separation and to express the fission-product concentration in terms of the gross β activity. The activity of the long-lived natural radionuclides is negligible indeed (less than 10 per cent) compared with that of the fission products. This is clearly shown in Table 1 and Figure 2.

Experimental procedures. The measurements of total and insoluble β activities, as well as the Sr⁹⁰ activity, are based on standard procedures which will not be discussed here. Experimental details are available in an internal report [Picciotto and Wilgain, 1963].

The total β activity was measured on samples condensed by evaporation. The last fraction was transferred to a stainless steel planchet and dried under an infrared lamp.

The radioactive measurements were made

with low-level proportional counters similar to the one described by Lal and Schink [1960]. The background rate was 0.2 cpm for β and 0.2 cph for α , the sensitive area and the diameter of the window being 3.18 cm. The over-all detection efficiency for the β^- radiation from a Sr⁹⁰-Y⁹⁰ equilibrium mixture was found to be 43 per cent.

Great care must be exercised to avoid radioactive contamination. It is often necessary to wait one or two days between the preparation of the sample and the radioactive assay in order to allow for the decay of Pb²¹⁴ and Pb²¹⁰.

In trying to make the method as simple as possible, we have checked whether a significant and constant fraction of the radioactivity could be retained by filtration on paper. The results of these trials are also given in Figure 2.

We tried several times to estimate the proportion of activity retained by the filter, as well as the fraction possibly adsorbed upon the walls of the containers. The results are variable from one test to another, but they indicate that (1) a significant fraction (10 to 20 per cent) of total β activity is immediately retained on the walls of the polyethylene container and is extracted by washing with concentrated HNO₃, and (2) retention of β activity in the filters may amount to 60 per cent, but it depends upon the thickness and the type of filter used and also upon the total quantity of insoluble matter.

Results and discussion. The results of radioactive measurements are found in Tables 2 and 3 and Figure 2. The striking fact is the sudden increase in β activity at the beginning of 1955.

The β activity values can be divided into three groups: (1) after 1954 (post-Castle), 7 to 21 (average, 13.6) dpm/kg, (2) 1953 and 1954 (post-Ivy), 2 dpm/kg, and (3) before 1952 (pre-Ivy), 0.5 dpm/kg. The pre-Ivy activity is due principally to Pb²¹⁰ and also to K⁴⁰.

The increase by a factor of 10 in β activity at the beginning of 1955 is undoubtedly due to the sudden arrival in the antarctic troposphere of fission products from the Castle test, with a delay of 10 to 11 months.

The small activity increase in 1953 is probably due to fission products from Ivy, but we cannot rule out a possible contribution from the upper layers by percolation of melt water or by mechanical mixing.

To date more accurately the level correspond-

TABLE 2. Fission Products and Gross β Activity in Snow, King Baudouin Base (measured in 1962)

Depth Interval, cm	Attributed Date*	Gross β Activity, dpm/kg†	Sr ⁹⁰ Activity, dpm/kg
Surface	1960	7.0 \pm 0.5	0.48 \pm 0.03
Precipitation	1958 Oct.-Jan.	15 \pm 3	
Surface	1958 Mar.-Oct.	6.0 \pm 0.6	
0-73	1957	7.6 \pm 0.8	
190-224	1955 Feb.-May	22 \pm 2	4.5 \pm 0.3
231-268	1954	1.8 \pm 0.1	
310-330	1953	2.2 \pm 0.2	
387-452	1952	0.82 \pm 0.05	
679-694	1947	0.46 \pm 0.06	
1072-1087	1940	0.56 \pm 0.01	
1119-1224	1938-1940	0.30 \pm 0.05	
1390-1421	1935	0.46 \pm 0.07	<0.12 \pm 0.06

* From *Gonfiantini et al.* [1963] for 1935 to 1957.

† The quoted errors in activities are the standard deviations on the counting rates.

ing to the major increase of activity, the section of firn from the years 1954 and 1955 has been cut into thinner slices. A precipitation date has been attributed to each sample on the basis of its stratigraphic characteristics of its oxygen isotope ratios. A detailed discussion of the criteria used will be found in the paper by *Gonfiantini et al.* [1963]. The results are given in Table 3, where it can be seen that the maximum increase in activity took place between November and March 1955. In our opinion, it was associated with the precipitations of March 1955. This date is in agreement with the pattern, seen in Figure 1, of seasonal variation of artificial radioactivity.

In conclusion, the reference level indicated by the sudden increase in β activity due to fission products corresponds to February 1955, with an uncertainty of ± 2 months. This conclusion is based upon observations made at only one spot; it has to be checked for more localities before making any practical use of it. Yet several arguments indicate that it is valid for the whole continent: (1) the purely stratospheric origin of the fallout, (2) the good agreement of the results and the monthly variations between Little America and Base Roi Baudouin in 1958 (Figure 1) (the systematic difference between the two sets of results can, in our opinion, be ascribed to uncertainties in calibrating the

TABLE 3. β Activity in Snow during 1954 and 1955, King Baudouin Base

Depth Interval, cm	Insoluble β Activity, dpm/kg	Gross β Activity, dpm/kg	Firn Stratigraphy*	Oxygen Isotope ratio,† δ (SMOW)	Attributed Date
190-224	6.6 \pm 0.6	22 \pm 2	Fine-grained unmetamorphosed firn ($d = 0.53$) resting upon the summer surface 1954-1955	-19.5 (summer)	Feb.-May 1955
224-229	3 \pm 1		Coarse-grained metamorphosed ($d = 0.48$)	-24.1 (winter)	Oct.-Nov. 1954
229-231	1.4 \pm 0.1		Fine-grained unmetamorphosed ($d = 0.48$)	-24.7 (winter minimum)	Sept. 1954
231-268	0.6 \pm 0.1	1.8 \pm 0.1	Same as 229-231 ($d = 0.53$)	-23.3 (winter)	May-Sept. 1954

* Firn stratigraphy from De Breuck (unpublished).

† Oxygen isotope ratio from *Gonfiantini et al.* [1963].

counters rather than to a real geographical effect), and (3) the concentration of Sr^{90} (4.5 dpm/kg) attributed to the precipitations from February to April 1955 at Base Roi Baudouin is in excellent agreement with the values published by Martell [1959a] for surface snow collected in February 1955 on the antarctic coast (2.0 to 5.3 dpm/kg).

It is likely that a more recent, accessory reference horizon was marked in 1962, corresponding to the intensive resumption of thermonuclear tests in October 1961, after three years of interruption.

It is possible that the seasonal variations of fallout can be used to identify the annual layers, by a method similar to that of stable isotope ratio.

Practical considerations. With transistorized counting units currently available, the measuring of β activity in the residue of evaporation or of the insoluble fraction would be possible, not only at the polar stations, but also in the field.

With a counter similar to the one used here, the measurements could be performed on 0.5 kg of snow.

In the coast area, where the annual accumulation may reach 1 m of snow, it would be necessary to drill down 12 m and to cut the cores into 50-cm slices (assuming 7.5-cm diameter).

On the polar plateau, drilling down 2 or 3 m would be sufficient, and the cores should be cut into approximately 10-cm slices.

The direct measurement of γ activity from Cs^{137} in the field seems to be at the lower limits of possibility and is worth a theoretical and experimental study [see e.g. Chesselet and Nordemann, 1962].

Acknowledgments. We wish to thank Mr. Y. Van De Can and Dr. W. De Breuck, members of the Belgian Antarctic Expeditions, who collected the samples from the glaciological pit of King Baudouin Base.

We are indebted to Dr. W. De Breuck and to the Laboratory of Nuclear Geology of the University of Pisa (Professor E. Tongiorgi) for allowing us to use the results of stratigraphic observations and those of oxygen isotope-ratio analyses while they were still in the process of being published.

We also thank, for their moral and financial support, the Institut Interuniversitaire des Sciences Nucléaires de Belgique and the Centre National de Recherches Polaires de Belgique.

This work was carried out under contract European Atomic Energy Community (EURATOM)-Université Libre de Bruxelles-Comitato Nazionale Energia Nucleare 013-61-7 AGECE.

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(Manuscript received June 3, 1963;
revised August 7, 1963.)