

Antarctic Snow Chronology with Pb²¹⁰

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Abstract. After a short discussion on the Pb²¹⁰ dating method, the results of Pb²¹⁰ measurements on snow and firn samples from four Antarctic stations are reported as well as the experimental procedure used. Pb²¹⁰ was also measured in the air at ground level, at Base Roi Baudouin, the average Pb²¹⁰ activity of aerosols in 1958 being 1.3×10^{-2} dpm/kg of air. In samples of surface snow accumulated during the last two to five years, the average Pb²¹⁰ activity is of the order of 100 dph/kg of snow. This activity seems to be independent of the local rate of accumulation. The variations of Pb²¹⁰ activity as a function of depth were studied on two vertical firn profiles spanning an interval of more than 200 years, at Base Roi Baudouin (70°S, 24°E) and at Amundsen Scott Station (South Pole). Within experimental errors, the Pb²¹⁰ activity was found to decrease exponentially with depth at both stations. The most plausible interpretation is that the rate of water accumulation and the initial Pb²¹⁰ concentration in the snow have remained constant at both places over the last 100 years. The annual rates of water accumulation calculated from the decay curves are 6 ± 1 cm at the South Pole and 45 ± 3 cm at Base Roi Baudouin, in good agreement (to better than 10 per cent) with accumulation rates so far obtained by other methods.

INTRODUCTION

The importance of and need for reliable methods of measuring the accumulation of snow on the Antarctic ice cap have frequently been stressed [see Sharp and Epstein, 1962; Picciotto and Wilgain, 1963]. Current measurements are principally based on the interpretation of the stratigraphic features of firn sections or, less frequently, of variations in the stable oxygen or hydrogen isotope ratios [Epstein and Sharp, 1959; Lorius, 1961; Gonfiantini et al., 1963]. A comparison among these methods may be found in Sharp and Epstein [1962] and in Gonfiantini et al. [1963].

Whatever their merits, methods based on identification of annual layers, whether stratigraphic or isotopic, involve a certain amount of personal interpretation. Their application to the polar plateau is likely to lead to an underestimate of the number of years and an overestimate of the accumulation rate.

To avoid these difficulties, Picciotto and Wilgain [1963] recently proposed a method based on the measurements of radioactive fallout from nuclear bomb tests.

Goldberg [1963] has developed a radiometric method of glacier chronology based on the decay of the naturally occurring Pb²¹⁰. He has applied it to a 14-meter-deep core from Greenland which represents a period of about 20 years.

In this paper we report the results of Pb²¹⁰ measurements on Antarctic snow samples. We have studied the variation of Pb²¹⁰ activity with depth at two sites which differ greatly in their annual rate of water accumulation: the South Pole and the Base Roi Baudouin. Both sections spanned an interval of more than 200 years.

PRINCIPLE OF THE METHOD

Air contains a certain amount of atmospheric aerosol-bound Pb²¹⁰ originating in the decay of radon. Information on the Pb²¹⁰ cycle in the atmosphere as well as the related literature will be found in Goldberg [1963] and in Junge [1963].

Like all other aerosol-bound elements, the Pb²¹⁰ is brought to the ground principally by precipitation. The average concentration of Pb²¹⁰ in rainfall in the intermediate latitudes

of the northern hemisphere is of the order of 5 dpm/l. The average Pb^{210} fallout, assuming that 1 meter of precipitation falls annually, is estimated at 15 atoms/min cm^2 of earth's surface. The tropospheric residence time of Pb^{210} , deduced from parent- and daughter-element ratios ranges from 5 to 20 days.

The principles involved in the chronology of firn layers by the Pb^{210} method are simple. If, at the time of precipitation, the snow contains an amount of Pb^{210} corresponding to a specific activity A_0 , this activity will decrease exponentially, with a half-life of 21.4 years [Eckelmann *et al.*, 1960] as a function of the time elapsed since the fall.

At first sight, this method is applicable only in polar regions where the snowfall undergoes little or no fusion and where it is preserved in the original order of deposition.

An age determination requires a knowledge of the activity A_0 of the snow at the time of its fall. This value is not directly known and we are obliged to rely on hypotheses which can, however, be indirectly verified.

Let us consider two extreme cases, as was done by Goldberg [1963], but with a somewhat different formulation.

The initial Pb^{210} concentration in the precipitation at a given place remains constant. In this case, the age t of a sample lying at a level h will be given in years by

$$A_h = A_0 e^{-\lambda t} \quad t = \frac{1}{\lambda} \ln (A_0 / A_h) \quad (1)$$

where $A_0 = \text{Pb}^{210}$ activity at level 0, $A_h = \text{Pb}^{210}$ activity at level h , and $\lambda =$ radioactive constant for $\text{Pb}^{210} = (0.032 \pm 0.001) \text{ yr}^{-1}$. This relation remains valid even in the case of variable annual rates of water accumulation or of variable rates of Pb^{210} fallout. When the annual rate of water accumulation remains constant, we have as a special case

$$t = h/a \quad \ln A_h = \ln A_0 - \lambda/a \cdot h \quad (2)$$

where $h =$ depth of the sample level, expressed in water equivalent, and $a =$ annual rate of water accumulation. A plot of $\ln A_h$ as a function of h will give a straight line with a slope inversely proportional to a .

The rate of Pb^{210} deposition remains constant, but the initial Pb^{210} concentration in the snow and the rate of water accumulation are variable.

In such a case, a sample from a given level cannot be dated independently. The total Pb^{210} activity integrated on a column of unit cross-sectional area and of depth h , corresponding to an interval of time t , is given by

$$S_h = F \int_0^t e^{-\lambda t} dt = \frac{F}{\lambda} (1 - e^{-\lambda t}) \quad (3)$$

where F is the rate of Pb^{210} deposition in number of atoms per year and per unit area. The age t of the base of a column whose total activity is S_h can be calculated from (3). The annual rate of Pb^{210} deposition, F , is given by

$$F = S_\infty \cdot \lambda$$

where S_∞ is the Pb^{210} activity integrated over a column of infinite height (or within 1 per cent for a column whose height is equivalent to 150 years).

ORIGIN OF THE SAMPLES

The firn samples analyzed here were collected at four Antarctic localities, in the vicinity of stations Roi Baudouin, Amundsen-Scott, McMurdo, and New Byrd.

Table 1 gives the main information on the origin of the samples. More details can be found in the articles quoted.

EXPERIMENTAL PROCEDURE

All samples were brought back and stored frozen until the time of analysis. This procedure seems indispensable to prevent adsorption of Pb^{210} and/or contamination by the containers.

The samples were collected with the utmost care, the surfaces being trimmed just before use. The containers used were mostly of polyethylene, previously washed with HNO_3 and triple-distilled water.

Approximately 5 mg of Pb in concentrated HNO_3 solution is added, as carrier, to the firn sample (between 2 and 20 kg) at the moment of melting. This small amount of added Pb does not appreciably contaminate the sample and makes it possible to measure the final activity from thin sources. The Pb (kindly supplied by Professor Tongiorgi) was more than 500 years old, with a gross β -particle activity lower than 1 dpm/g. The lead-free analytical-grade nitric acid was distilled in a quartz still, its gross β -particle activity being of the order of 0.01

TABLE 1. Origin of the Samples

Station	Sample	Origin of Sample	Date of Collection	Operation
Base Roi Baudouin	W ₂	Base glaciological pit, 22 m deep	Winter 1960	1960 Belgian Antarctic Expedition [De Breuck, 1961]
Ice shelf of the Princess Ragnhild Coast (70°26'S, 24°19'E) [Picciotto, 1961; Loodts, 1960]	BRB and S ₂ serie	120-m-deep drilling, 2 km SE of the base	Jan. 1961	CNEN-EURATOM drilling contract No. 002/60/12/GEO-C [Tongiorgi et al., 1962]
Amundsen-Scott, South Pole [Giovinetto, 1960]	P ₁	2.50-m-deep pit, 500 m NW of the aurora dome	Dec. 1962	NSF contract AA-422 [Picciotto and De Breuck, 1963]
	HH	2.50-m-deep trench, 300 m N of the station	Dec. 1962	NSF contract AA-422 [Picciotto and De Breuck, 1963]
	SM serie	Base glaciological pit 'Snow Mine,' 26 m deep	Dec. 1962	NSF contract AA-422 [Picciotto and De Breuck, 1963]
McMurdo (77°21'S, 166°41'E)	RIS	2.50-m-deep pit on the Ross Ice Shelf, 2 km S of Pram Point	Dec. 1962	NSF contract AA-422 [Picciotto and De Breuck, 1963]
New Byrd (80°00'S, 120°00'W)	B serie	CRREL glaciological trench	Dec. 1962	NSF contract AA-422 [Picciotto and De Breuck, 1963] (Collected by the CREEL 1962-1963 summer team)

dpm/ml. This low activity nevertheless introduced an appreciable correction for the samples of lowest activity. After fusion and equilibration with the Pb carrier, the sample is transferred, in a dust-free atmosphere, to a quartz distillation apparatus in which it was concentrated to about 10 ml.

The original polyethylene container was rinsed with concentrated HNO₃, the rinsings being added to the residue of the distillation. This liquid was transferred to a small quartz beaker and evaporated in a pyrex recipient under an atmosphere of flowing filtered nitrogen. The residue was redissolved in 1.5 M HCl, the solution was centrifuged, and the insoluble fraction (principally silicate and polyethylene debris) was discarded. The solution was then passed through an anion exchange column (Dowex 1 × 8, 100/200 mesh in 1.5 M HCl medium). Two column volumes (±5 ml) of 1.5 M HCl were passed through, followed by 10 ml H₂O which eluted the Pb; the Bi²¹⁰ re-

mained on the column. The Pb recovery yield was determined by polarography on an aliquot.

The solution of Pb was evaporated in a quartz beaker, the residue being taken up in 1 ml HNO₃ which was then transferred and evaporated by small fractions in a stainless-steel planchet. The Pb²¹⁰ activity was measured by counting the regrowing Bi²¹⁰ activity. The Pb²¹⁰ radiochemical purity was checked from the Bi²¹⁰ build-up curve (*T* = 5 days). At the start of the measurements a short-lived activity due to Pb²¹⁴ and Pb²¹³ is usually detected.

In samples younger than 1955, the main source of contamination should be Y⁹⁰ building up from its parent Sr⁹⁰, with a half-life of 64 hours. The absence of Sr⁹⁰ in the Pb fraction was ascertained by adding a known amount of stable Sr to the samples; the Pb fraction was found to contain less than 1 per mil of the added Sr.

The radioactivity was measured with two low-level proportional flow counters with anti-coincidence guard, of the type described by Lal

and Schink [1960]. Their characteristics are as follows:

	Counter 1	Counter 2
Window diameter and sensitive area	32 mm	13 mm
Window thickness	800 $\mu\text{g}/\text{cm}^2$	800 $\mu\text{g}/\text{cm}^2$
Background	0.2 c/m	0.04 c/m
Over-all detection efficiency for Pb^{210} + Bi^{210} at equilibrium	38%	13%

The counters were calibrated with a standard Pb^{210} solution prepared by allowing a known amount of radon sealed in a glass tube to decay [see *Begemann and Houtermans*, 1952]. The radon was measured, with a precision of 1 per mil, by comparing its γ -ray activity with that of a radium standard. Samples of snow more than 200 years old were tested for background level. The limit of significant detection under our conditions is 2 dpm of Pb^{210} per samples (2 to 20 kg).

This limit could easily be lowered by a factor of 10 by counting the α -particle activity of the Po^{210} ($T = 140$ days) building up from the Pb^{210} , but it would be necessary to wait several months before making the final measurements. The direct measurement of the Po contained in the snow is an uncertain process because of the absence of an available Po -isotopic carrier.

Pb^{210} IN THE ANTARCTIC ATMOSPHERE AND PRECIPITATIONS

To establish the reliability of the Pb^{210} dating method, precise knowledge of the Pb^{210} cycle in the atmosphere in general, and in the Antarctic atmosphere in particular, is necessary. In view of the particular situation of the Antarctic continent (its ice cover preventing the escape of radon, its distance from any important continental masses) we can expect some particularities in the behavior of the Pb^{210} found in these regions.

The Po^{210} concentration in Antarctic air at ground level was first estimated in 1958 at the Base Roi Baudouin [Picciotto, 1958]. This estimation was based on a measurement of the total α -particle activity of the aerosols. It led to a value of the order of 10^{-10} curie/ m^3 or 0.17×10^{-8} dpm/kg of air, which must be considered as a lower limit for the Pb^{210} activity value.

We have checked this value by radiochemically analyzing the air filters for 1958 which had served for the α -particle activity measurements. The paper filters, grouped by month, were completely dissolved in a mixture of nitric and perchloric acid, and approximately 1 mg of inactive Pb was added. The solution was evaporated, the residue being redissolved in 1.5 M HCl and all subsequent operations being the same as for the snow samples. Monthly averages thus obtained are shown in Table 2. The average Pb^{210} concentration for the year 1958 is 7×10^{-10} curie/ m^3 or 1.3×10^{-8} dpm/kg of air, thus being about 1/20 of the average activity observed in the northern hemisphere.

Monthly variations are small, and no clear seasonal effect was noted. More recent results have been obtained by the Belgian Antarctic Expedition in the last two years, but they will be discussed elsewhere.

To study the variations of Pb^{210} in precipitation, we have measured the Pb^{210} activity on recent snows (1962 to 1955), collected at the surface or at shallow depths from four sites displaying annual precipitation accumulation which varied from 7 to 40 cm.

The results are given in Table 3. It is seen that in spite of a great variability from sample to sample the results favor the hypothesis that the initial concentration of Pb^{210} in the snow

TABLE 2. Pb^{210} Concentration in the Air at Ten Meters above Ground Level during the Year 1958, Base Roi Baudouin, Monthly Averages

Month	10^{-8} dpm/kg of air*
February	1.9
March	0.79
April	0.82
May	1.2
June	1.4
July	0.87
August	1.5
September	0.86
October	1.2
November	1.8
December	1.6
Mean value	1.3 ± 0.1

* The experimental error, including the uncertainties of air-flow measurement, chemical analysis, and radioactivity counting, is estimated to be approximately ± 10 per cent.

TABLE 3. Pb²¹⁰ Concentration in Recent Firn from Different Regions

Sample	Region	Thickness of the Firn Sample, cm	Estimated Year	Pb ²¹⁰ Concentration, dph/kg	Annual Precipitation Accumulation, cm
P ₁ 1962	Amundsen-Scott Station	30	1962-1961	108 ± 5	7
HH 1960		10	1960	65 ± 3	
RIS	McMurdo Sound	100	1962-1960	145 ± 6	16*
B ₁	New Byrd station	10	1962	27	18†
B ₂		10	1961	56	
B ₂ - B ₁₁		100	1960-1959	113 95†	
B ₁₂ - B ₂₁		100	1958-1956	101	
B ₂₂ + B ₂₄		18	1955	31	
BRB 1960	Base Roi Baudouin	100	1960	74 ± 3	40

* *Stuart and Bull* [1963].

† Weighted mean from thickness.

‡ *Giovinetto* [1960].

averaged over 1 or 2 years is constant and independent of the accumulation rate. Before this hypothesis is accepted it should be checked by analyzing many measurements.

The sample RIS 1 which had the highest specific activity was collected in the vicinity of Ross Island, where the active Mount Erebus volcano is located. Its high Pb²¹⁰ content may be explained by the proximity of an important continental mass producing radon and also by some contribution of Pb²¹⁰ in equilibrium with uranium contained in the continental dust.

VARIATIONS IN THE Pb²¹⁰ CONCENTRATION AS A FUNCTION OF DEPTH IN THE FIRN

The variations in the Pb²¹⁰ concentration in the firn as a function of the depth at Base Roi Baudouin and at the South Pole are shown in Tables 4 and 5 and in Figure 1.

In cases where the sample thickness is appreciable, the mean activity measured has been taken to correspond to the middle of the depth interval covered by the specimen. If the thickness corresponds, as it does, to a time interval

TABLE 4. Pb²¹⁰ Concentration as a Function of Firn Depth, Base Roi Baudouin (70°26'S, 24°19'E)

Sample	Depth of Snow, cm*		Depth of Water, cm†		Estimated Time Interval‡	Sample Weight, kg	Chemical Yield, %	Pb ²¹⁰ Concentration, dph/kg§
	Top	Bottom	Top	Bottom				
BRB 1960	0	100	0	40	1960	22.54	54	74 ± 3
S ₂ 8 to 20	805	1360	410	670	1950-1942	5.73	69	45 ± 4
W ₂	1105	1810	593	1041	1944-1933	8.53	67	44 ± 2
S ₂ 35 to 51	1955	2570	1170	1622	1932-1921	8.71	68	29 ± 2
S ₂ 67 to 94	3230	4325	2111	2987	1910-1887	17.95	73	12 ± 1
S ₂ 119 to 123	9630	10790	7602	8612	1772-1747	2.90	76	6 ± 4

* The depths are measured from the zero level, January 1961.

† After measured densities.

‡ This estimate is based on the interpretation of the stratigraphy and of the variations of the oxygen isotope ratio.

§ The error in the specific activity takes into account the standard deviation on the radioactive measurement and an uncertainty of ±3 per cent in the chemical yield.

TABLE 5. Pb^{210} Concentration as a Function of Firn Depth, South Pole

Sample	Depth of Snow, cm*		Depth of Water, cm†		Estimated Time Interval‡	Sample Weight, kg	Chemical Yield, %	Pb^{210} Concentration, dph/kg§
	Top	Bottom	Top	Bottom				
P ₁ 1962	0	22	0	9	1962-1961	8.2	71	108 ± 5
HH 1960	22	38	9	14	1960	6.8	68	65 ± 3
SM 11-12	406	436	159	172	1940-1939	1.5	75	37 ± 4
SM 24-25	802	833	336	350	1918-1917	9.4	47	15 ± 2
SM 35-36	1138	1168	498	512	1890-1894	14.6	41	6 ± 2
SM 55-56	1748	1778	812	822	1851-1850	7.9	73	2 ± 1
SM 79	2510	2632	1236	1297	1778-1770	3.3	75	0 ± 3
SM 80-83								

* The depths are measured from the zero level, December 1962.

† Data of Giovinetto (except for the two first samples of which we have measured the density).

‡ After Giovinetto [1960] for the samples collected before 1960.

§ The error in the specific activity takes into account the standard deviation on the radioactive measurement and an uncertainty of ± 3 per cent in the chemical yield.

shorter than a half-life, this approximation introduces an error of less than 2 per cent, which has not been taken into account.

Within the limits of error, the experimental points for both stations fall on a straight line.

On the basis of the preceding discussion, the most likely interpretation is that the initial Pb^{210} concentration in the snow and in the annual rate of water accumulation remained constant over the last century. Thus the rate of accumulation is deducible from equation 2.

For the two sets of experimental points the line of best fit satisfying the equation

$$\log A_h = \log A_0 - \frac{1}{2.303} \frac{\lambda}{a} h$$

has been drawn.

The annual rates of water accumulation a are found to be: Base Roi Baudouin, $a = (45 \pm 3)$ cm; South Pole, $a = (6 \pm 1)$ cm.

COMPARISON WITH OTHER MEASUREMENTS OF ACCUMULATION

Base Roi Baudouin. In Table 6 the results obtained up to the present by other methods are collected: direct measurements, stratigraphy, variations in the stable oxygen isotope ratios.

It is to be remembered that 1958 was a year of exceptionally low accumulation over the whole of East Antarctica [Kotlyakov, 1961].

On the basis of these results we estimate the

most likely mean rate of annual accumulation of water over the last hundred years to lie between 37 and 41 cm.

Unpublished oxygen-isotope data relating to the interval 1880-1870, kindly communicated by Giontini, confirm that the rate of accumulation, a hundred years ago, did not appreciably differ from that of the present.

The value of the annual accumulation calculated from the Pb^{210} data is slightly higher; a comparison of the results may nevertheless be considered as a fair confirmation of the validity of the assumptions used.

South Pole. The results obtained at the

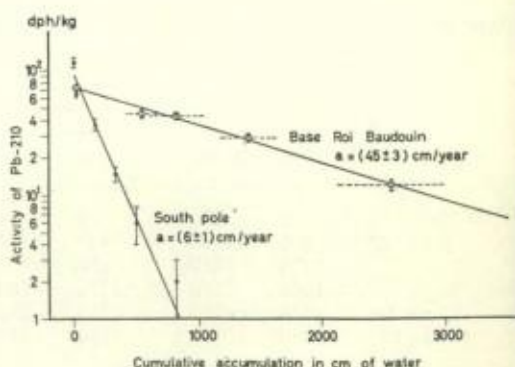


Fig. 1. South Pole station and Base Roi Baudouin. Variation of the Pb^{210} activity in the firn as a function of equivalent water depth. The vertical lines indicate experimental errors in the Pb^{210} measurements. The horizontal dotted lines indicated the thickness of the specimens.

TABLE 6. Estimations of the Annual Accumulation at the Base Roi Baudouin

Date (estimated)	Method of Measurement	Water Accumulation, cm	Reference
1960	Direct	45.5	<i>De Breuck</i> [1961]
1958	Direct	20	<i>Picciotto</i> [1961]
(1961-1953)	Stratigraphy and O^{18}/O^{16}	39.5	<i>Tongiorgi et al.</i> [1962] <i>Gonfiantini</i> (private communication)
(1957-1933)	Stratigraphy and O^{18}/O^{16}	37	<i>Gonfiantini et al.</i> [1963]
(1880-1870)	O^{18}/O^{16}	41	<i>Gonfiantini</i> (private communication)
(1960-1887)	Pb ²¹⁰	45 $\pm$ 3	Present work

Amundsen-Scott South Pole station are given in Table 7. The accumulations measured directly were very kindly communicated to us by Dr. Luis Aldaz; we have estimated the water accumulation by assuming a density of 0.35 for the surface snow.

The value for the period 1963-1951 was obtained from our own stratigraphic observations on a pit dug 500 m from the station (Figure 2). The value for 1963-1955 is deduced from the distribution of the gross β -particle activity attributed to the fission products in the same pit (Figure 2) [*Picciotto and Wilgain*, 1963], and these results will be discussed in greater detail in another paper.

Sharp and Epstein [1962], on the basis of their measurements of the O^{18}/O^{16} ratio, note a systematic disagreement between their values of the annual accumulation and those either measured directly or deduced from firn stratigraphy in a number of locations in West Antarctica. They suggest the possibility that the rate of accumulation, for the whole of West Antarctica, may have been underestimated by

a factor of 2. They propose 15 cm of water as the accumulation at the Pole. This value was obtained by analyzing a section 80 cm thick showing only three isotopic alternations.

The value deduced from Pb²¹⁰ measurements is 6 $\pm$ 1 cm, in good agreement with all estimations except that of *Sharp and Epstein*, which does not appear to be acceptable. Possible causes of such a disagreement have been discussed by *Gonfiantini et al.* [1963].

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TABLE 7. Estimations of the Annual Accumulation at the South Pole

Annual Interval (estimated)	Method of Measurement	Water Accumulation, cm	Reference
February to November 1962	Direct (New field, 140 stakes)	7.0	Aldaz (private communication)
February 1958 to November 1962	Direct (Old field, 36 stakes)	6.6	Aldaz (private communication)
(1947-1850)	Stratigraphy	7.4	<i>Giovinetto</i> [1960]
(1958-1954)	O^{18}/O^{16}	15	<i>Sharp and Epstein</i> [1962]
(1963-1951)	Stratigraphy	7.5	<i>Picciotto and De Breuck</i> [1963]
(1963-1955)	Fission products	6.5 $\pm$ 0.5	Unpublished
(1963-1850)	Pb ²¹⁰	6 $\pm$ 1	Present work

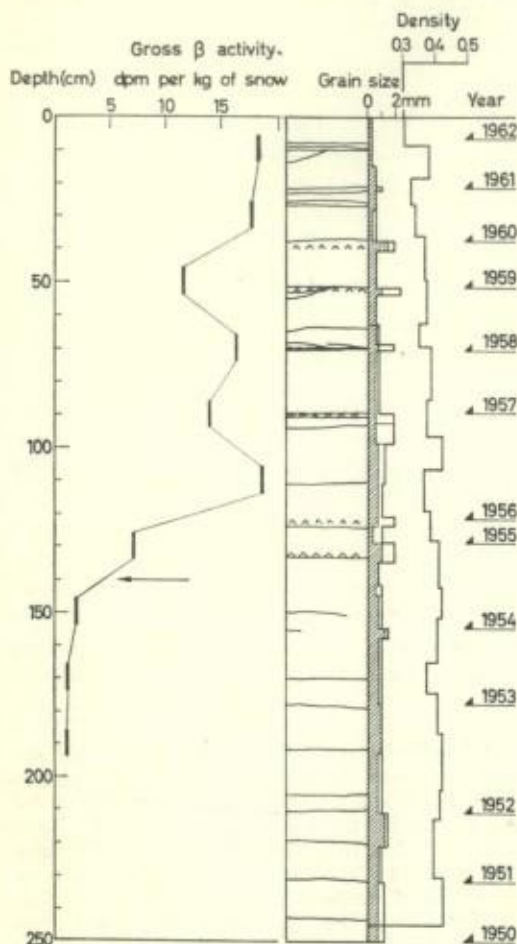


Fig. 2. South Pole station, Pit 1. Stratigraphic profile and gross β -particle activity. The interpretation of the stratigraphic profile in terms of annual layers is given on the right side of the figure.

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