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Nickel Content of Antarctic Snow: Implications of the Influx Rate of Extraterrestrial Dust

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The concentration of Na, Mg, K, Ca, Cl, and Ni has been measured in firn samples collected in the vicinity of King Baudouin Base (70°S, 24°E) and Amundsen-Scott Station (90°S). The Ni content is of the order of a few ppb at both stations. About 35% of the Ni was probably present in the firn as soluble salts. Arguments are presented in support of an extraterrestrial origin for nearly all the Ni found in the south pole samples. The rate of Ni deposition at the south pole is of the order of 10^{-8} g/cm²/yr. Under the assumption of a Ni abundance of 1.3% (chondrite average), the influx rate of extraterrestrial matter over the entire earth's surface should lie between 3 and 10 million tons per year depending on the assumption made in the extrapolation.

INTRODUCTION

It is well established that meteorites represent but a negligible fraction of the influx of extraterrestrial matter to the earth. The major part of this matter is accreted by earth as particles with dimensions of the order of 10^{-4} to 10^{-2} cm. Some of these particles are formed by ablation or fusion of meteorites or meteors during atmospheric entry, but most of them are already present in interplanetary space, forming a cloud of dust around the sun. The existence and the properties of these dust particles are inferred from a wide variety of observations: satellites and rocket measurements, zodiacal light, solar corona, meteors, etc. (see bibliography in *Hodge et al.* [1961], *Schmidt* [1963], *McCracken and Alexander* [1963], *Whipple* [1964]).

Until now, very little has been known about the origin, the physical properties, and the chemical and isotopic composition of extraterrestrial dust particles. Estimates of their accretion rate show a spread of 3 to 4 orders of magnitude.

Observations made by rockets and satellites are rapidly increasing our knowledge of this matter, but observations on earth are still of great interest.

Studies on earth are generally carried out on deep-sea sediments and on firn and ice from Antarctica and Greenland, where the contribution from terrestrial sources is at a minimum value. A major problem in these studies is the identification of the extraterrestrial components in the collected material. For practical reasons, most observations have dealt with opaque magnetic spherules which are easy to concentrate from large volumes of ice or sediment and which are easily identified under the microscope. The origin of these objects is, however, still a matter of controversy [*Fredriksson and Martin*, 1963; *Hodge and Wright*, 1964; *Giovinetto and Schmidt*, 1965; *Schmidt*, 1966]. Even if their extraterrestrial origin were ascertained, they could only represent an undetermined fraction of the total mass accreted. As pointed out by *Tilles* [1966], spherule estimates do not take into account the contribution of transparent or nonspherical particles or the water soluble compounds and the material volatilized into the atmosphere.

The most convincing criterion for an extraterrestrial origin is the presence of stable or radioactive nuclides produced by interaction with the cosmic radiation. (An excellent review of this question may be found in *Wasson* [1963].) Unfortunately, the application of this criterion requires very large samples, of the order of several kilograms of pelagic clay or several tens of tons of polar ice. Interesting attempts have been made in this direction [*Merrihue*, 1964; *Schaeffer et al.*, 1964; *Tilles*,

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1965; *Fireman and Langway*, 1965; *Lal and Venkatavaradan*, 1966], but they have led either to negative results or to results difficult to interpret univocally in terms of accretion rates.

The antarctic ice sheet, specially the central part of eastern Antarctica, is one of the most favorable sites for collecting extraterrestrial matter for the following reasons:

1. It is particularly remote from sources of industrial or geological contamination.

2. Because the temperature is permanently below 0°C, the chemical composition and even the size and shape of the deposited particles should remain unaltered in the course of time. This makes Antarctica particularly well suited for searching micrometeorites (in the definition of *Whipple* [1949], 'extraterrestrial bodies that are sufficiently small to enter the earth's atmosphere without being damaged by encounter with the atmosphere').

3. The low rates of snow accumulation can be measured with fair accuracy at the actual sampling site, thus enabling an accurate estimate of deposition rates. The classical methods based on firn stratigraphy [*Giovinetto*, 1960; *Kotliakov*, 1961] can be usefully supplemented by isotopic methods based on O^{18}/O^{16} ratio [*Epstein et al.*, 1965], on Pb 210 radioactive decay [*Crozaz et al.*, 1964], or on artificial radioactivity reference levels [*Picciotto and Wilgain*, 1963].

4. Drilling into the ice sheet would provide a continuous section spanning a time interval of several thousand years, thus offering a unique opportunity to study the influx variations over this time interval.

Investigations on extraterrestrial particles in the snow of polar regions have been reported as early as 1874 [*Nordenskjold*, 1874]. As a result of the International Geophysical Year, a number of modern investigations were made both on Greenland and on antarctic snow samples [*Thiel and Schmidt*, 1961; *Mrkos*, 1962; *Parkin and Hunter*, 1962; *Schmidt*, 1963*b*; *Hodge et al.*, 1964; *Langway*, 1965]. Although they have provided very valuable information, most of these examinations dealt with black metallic spherules and gave no definite data on the total mass accretion of extraterrestrial matter.

In the present work, an attempt has been made to estimate the amount of extraterrestrial matter in antarctic snow samples on the basis of their chemical composition. The number of samples available was too small to allow a search for cosmogenic nuclides. The content of magnesium, sodium, potassium, calcium, chlorine, and nickel were measured. The choice of these elements was somewhat arbitrary, based partly on the availability of analytical methods of sufficient sensitivity and partly on a consideration of the abundance ratios most characteristic of an extraterrestrial origin. The main drawback of such a method is obviously the complete lack of knowledge on the composition of extraterrestrial dust. In the absence of direct information, we have assumed that the relative abundance of these elements in extraterrestrial dust is the same as in chondritic meteorites, these objects being generally considered as the best available samples of undifferentiated solar matter, at least for the nonvolatile elements [*Suess and Urey*, 1956].

In this respect, particular attention was paid to the content of nickel, the abundance of which is much higher in chondritic matter than in terrestrial crustal rocks.

Origin of Snow Samples

Firn samples were collected with utmost care in the vicinity of two antarctic stations: the King Baudouin Base (KBB) on the Princess Ragnhild Coast and the Amundsen-Scott Station (ASS) at the south pole.

Details on the sampling procedures will be found in *Picciotto and De Breuck* [1963, 1964]. Table 1 summarizes the main information on both stations.

King Baudouin Base. The samples for nickel measurement were collected in February 1964 in a 3-meter pit dug 2 km south (upwind) of the base, over the depth interval 150–200 cm. This layer was unambiguously dated by stratigraphy and by oxygen isotope variations at 1962, a year during which the base was not in operation. The samples for Ni measurements were stored in the frozen state until the time of analysis.

The samples assayed for Cl, Na, K, and Ca content were collected in October 1960 by *De Breuck* [1961]. The data reported here are averages over 7 cores about 1 meter long; each core represents the snow accumulated since approximately January 1960 and collected at 7

TABLE 1. Main Characteristics of King Baudouin Base (KBB) and Amundsen-Scott Station (ASS)

	KBB	ASS
Position	70°26′S–24°19′E	90°S
Elevation, meters	40	2800
Distance to nearest coast, km	10	1200
Distance to nearest mountain range	200 km upwind from the station	300 km downwind from the station
Mean annual temperature	−15°C	−51°C
Maximum temperature	+4°C	−15°C
Annual rate of snow accumulation, g/cm² yr	40	7

sites, 30 to 50 km south of the station. These samples were stored in liquid form in polythene containers. Numerous tests have attested to the absence of adsorption or of contamination from the containers for the measured elements at their actual level of concentration (0.1 to 1 ppm).

Amundsen-Scott Station. All the samples were collected in December 1962 in a pit dug 2 km upwind from the station, over the depth interval 190–240 cm, attributed to the years 1950–1952, that is at least 5 years before the establishment of the station. The firn samples were transported and stored in the frozen state. The concentration of Mg, Na, Cl, K, and Ca were measured on aliquots of a 5-kg melted sample. Ni measurements were made on fifteen 500-gram ice samples.

Analytical Procedure

A detailed description of the analytical procedures can be found in *Brocas* [1966]. Chlorine was measured by neutron activation and by colorimetric method based on the exchange reaction between Cl^- and $CrO_4^=$. Sodium has been determined by flame photometry; and magnesium, potassium, and calcium, by stable isotope dilution.

The main problem encountered in measuring Ni at the ppb level was laboratory contamination. It was overcome by using neutron activation techniques on the frozen samples.

The 500-gram samples were cored with a plastic tube in the center of large blocks of firn. They were then irradiated for 2 to 3 hours in a neutron flux of the order of $10^{12} n/cm^2$ sec, in the large channel of the reactor BR1 of the Center of Nuclear Study (C.E.N.) at Mol, Belgium. The samples were kept in the frozen state during the irradiation by wrapping them in a thick layer of polystyrene foam. The whole package was cooled at liquid air temperature before irradiation. After the irradiation, the ice was melted in the presence of a stable nickel carrier.

In most cases, we have tried to separate soluble and insoluble Ni fractions by filtering the melt water through Millipore filters of 0.05-μ porosity and by measuring the Ni activity on both fractions separately.

The nickel content was measured by the β activity of Ni 65 ($T_{1/2} = 2.6$ hours). Chemical separations and yield estimates were based on the classical method of precipitation and extraction of the Ni salt of dimethylglyoxime. Recovery yields ranged from 60 to 90%.

The radiochemical purity of Ni 65 was checked by the shape of the decay curve. Decontamination factor measurements enabled us to rule out possible interferences from manganese 56, strontium 87, or silicon 31 whose half-lives are in the region of 2.6 hours. The probability of forming Ni 65 by side reactions such as Cu 65 (n, p) or Zn 68 (n, α) was also found to be negligible.

Blank tests were performed by applying the identical procedure to frozen water, thrice distilled in a quartz still. The activity found in the nickel fraction was of the order of 1% of the average Ni content found in the samples. Table 2 gives the detailed data for a typical sample.

TABLE 2. Nickel Analysis Data Sheet for Sample KBB1

Weight of sample	325 g
Neutron flux	$\sim 10^{12} n/cm^2$ sec
Irradiation time	2 h 30 m
Activity of Ni fraction 4.7 hr after irradiation	312 cpm
Activity of Ni fraction 26 hr after irradiation	13 cpm
Ni^{65} activity extrapolated at end of irradiation	900 cpm
Chemical yield	60%
Counter yield	~30%
Over-all experimental error	±10%
Ni content	2.8×10^{-9} *g Ni/g ice*

Discussion of Results

General. Average concentrations of the measured elements are given in Table 3. The detailed results can be found in *Brocas* [1966]. We report in Table 4 the detailed results of the nickel measurements.

A complete discussion of these results would be beyond the scope of this article. We shall only point out the following observations:

1. The concentration values found in the snow from the south pole are the lowest ever recorded in precipitations (see Table 5). The ionic concentrations reported by *Matveev* [1961, 1962] on snow samples collected along the Mirnyy-Vostok profile are not consistent with our results. Our values for Na, K, and Cl are in general agreement with the results reported by *Wilson and House* [1965].

2. On the average, 35% of the total nickel was found in the fraction passing through 0.05-μ Millipore filters. It seems reasonable to assume that this nickel was present in the snow as soluble salts. We cannot, however, exclude completely the possibility that very small particles were solubilized during the melting of the ice, even though no acid was added to the water before filtration.

3. Table 4 shows that the total nickel content varies considerably from sample to sample but that the largest deviations (samples KBB 4, KBB 15, ASS 3) are likely to be caused by the presence of insoluble particles. The spread of the soluble nickel values is less pronounced.

The soluble nickel fraction could be partly ascribed to the matter vaporized during the entry of extraterrestrial objects into the atmosphere. A fairly large fraction of the total mass accreted is expected to undergo such a vaporization [Öpik, 1961, quoted in *Wasson*, 1963; *Whipple*, 1951].

As pointed out by *Wasson* [1963], this fraction will behave in a way similar to radioactive tracers, such as Rh 102 [*Kalkstein*, 1962] and Cd [*Salter*, 1964] injected into the high stratosphere. It will become attached to the atmospheric aerosols and will be carried down to the ground along with the precipitations; its deposi-

TABLE 3. Abundance of Chemical Constituents in Antarctic Snows (10^{-9} g/g)

Sample	Cl	Na	K	Ca	Mg	Ni
KBB (total)	630	740	190	67	...	4.3
KBB (excess)*		325	175	53	...	4.3
ASS	<10	3	8	8	6	1.5
Blank	<10	<1	0.03	0.5	2	0.05

* Tentative estimate of the nonoceanic contribution. It has been assumed that all Cl is of oceanic origin and the other elements are present in proportion to their presence in ocean water.

TABLE 4. Nickel Content of Antarctic Snows (10^{-9} g/g Snow)

Sample No.	Total Ni	Soluble Ni	Soluble Fraction %
	King Baudouin Base Area		
KBB 1	2.80	...	...
2	0.30	...	...
3	2.78	...	...
4	19.13	0.44	2.3
5	3.45	1.00	30
6	1.27	1.07	84
7	2.12	0.85	40
8	1.20	1.08	91
9	3.64	2.69	74
10	6.75	5.33	79
11	3.07	2.45	80
12	3.29	3.22	98
13	3.50	0.73	21
14	2.70	1.11	41
15	8.41	1.26	15
Average	4.3	1.8	36
Range	0.3–19	0.4–5.3	2.3–91
	South Pole		
ASS 1	1.10	...	...
2	0.90	0.80	89
3	13.46	3.23	24
4	1.03	0.36	35
5	0.19	...	...
6	0.43	0.29	67
7	0.26	...	...
8	0.39	...	...
9	0.51	0.41	80
10	2.05	...	...
11	0.17	...	...
12	0.25	0.14	55
13	0.62	0.57	92
14	0.41	...	...
15	1.08	...	...
Average	1.5	0.8	34
Range	0.2–13.5	0.1–3.2	24–92
Blank	0.05		

Experimental error on individual run: ±10%.

TABLE 5. Abundance of Chemical Constituents in Snow and Ice from Various Locations (10^{-9} g/g)

	Temperate (1)	Antarctic Coast (2)	Greenland Ice Sheet (3)	Greenland Ice Sheet (4)	Vostok, Antarctica (5)	South Pole, Antarctica (6)
Na	460	1390	77	29	180	3
K	310	470	60	11		8
Mg	160	...	24	...	160	6
Ca	390	220	38	35	490	8
Cl	500	1830	27	37	350	<10

(1) Sierra Nevada snow [*Feth et al.* 1964].
(2) Princess Ragnhild Coast, firn, partly from *Brocas and Delwiche* [1963].
(3) Weighted average of samples of snow, firn, and ice from four locations [*Langway*, 1965].
(4) Firn-ice from site 2, average values for the time period 1915–1957 [*Junge*, 1960].
(5) Vostok, surface snow [*Matveev*, 1961, 1962].
(6) Present work.

tion will be maximum in the temperate latitude belts of both hemispheres and minimum at the pole and at the equator.

To find some clue to the origin of the nickel, we may compare the chemical ratios found in the antarctic snow samples with the same ratios in three possible sources (Table 6):

The earth's crust, represented by the average igneous rocks.

The basalts that are the richest in nickel among the common rocks.

The chondrites.

We must admit that such factors as the heterogeneity of the samples, the poor sampling statistics, and the fact that the nickel and the other elements were not measured on the same samples render the drawing of any definite conclusion difficult. Nevertheless, the ratios in the south pole samples are strikingly lower than the ratios expected from any conceivable terrestrial source. To attribute a terrestrial origin to the nickel found in these samples, one must assume a process that would have enriched the nickel by a factor of 100.

Assuming a terrestrial contribution of basaltic composition, the nickel fraction from extraterrestrial origin would amount to more than 95% in the south pole samples. At King Baudouin Base, this Ni fraction would be of the order of 50%. As can be seen from Table 3, the Na/Ni ratio at this location should be divided by a factor of 2 as a result of the contribution of Na of marine origin. Thus the Na/Ni and K/Ni ratios in snow are half the value of the same ratios found in basalts.

Rate of accretion of extraterrestrial matter. By taking into account the rates of snow accumulation (Table 1) and by assuming a Ni content of 1.35% in weight (average of the chondrites according to *Wasson* [1963]), we find the annual deposition rates reported in Table 7.

The difference between the two stations could be partly ascribed to the poor sampling statistics and to the relatively high terrestrial contribution at King Baudouin Base which may have been underestimated, but it is of the order

TABLE 6. Chemical Ratios in the Antarctic Snow Samples and in Three Possible Sources (Weight Ratios)

Source	Na/Ni	K/Ni	Ca/Ni	Mg/Ni
Snow KBB (1961–1963)	172	44	15	...
Snow ASS (1950–1953)	≤2	5.4	5.4	≤4
Chondrites	0.41*	0.06*	0.82*	8.4*
	0.63†	0.08†	1.22†	13.6†
Basalts‡	185	93	500	290
Average igneous rocks§	360	320	455	260

* After *Mason* [1962]; Ni = 1.7%.
† After *Suess and Urey* [1956], composition of L chondrites group of Urey-Craig (1953); Ni = 1.06%.
‡ After Turekian (1956) and Daly (1933), compiled by *Goldberg and Arrhenius* [1958].
§ *Rankama and Sahama* [1950].

TABLE 7. Nickel Deposition Rate in Antarctica (in 10^{-9} g/cm²/yr)

Origin of Samples	Total Ni	'Soluble' Ni	'Insoluble' Ni	Presumed Extraterrestrial Ni	Total Extraterrestrial Matter (Ni = 1.35%)
King Baudouin Base Area	170	61	109	85	6300
Amundsen-Scott Station	10	3.4	6.6	10	740

TABLE 8. Recent Estimates of the Rate of Accretion of Black Spherules Over the Earth

Reference	Source of Spherules	Years of Fall	Deposit, tons/yr	Minimum Diameter, μ
Schmidt [1963b]	Antarctic Peninsula firn	1952(?)–1962	1.2×10^5	10
Langway [1965]	Greenland ice (site 2)	1960(?)	2.1×10^5	5
	Greenland firn (Camp Century)	1955(?)	6.6×10^5	5
Crozier [1966]	New Mexico air	1956–1965	10^6	5
Picciotto (unpublished)	Antarctic firn (KBB)	1960	1.2×10^6	3

to be expected from the difference of snow accumulation at both stations.

In view of estimating the total mass accretion, we shall consider only the results from Amundsen-Scott Station, where the terrestrial contamination is the lowest. In trying to extrapolate these data to the entire earth, we shall consider two extreme cases:

1. The mass accretion is uniform and independent of geographical factors. This assumption should lead to a lower limit, since we extrapolate to the entire earth surface the accretion rate found at a site of expected minimum deposition. By multiplying 0.7 γ by the earth area we obtain a total accretion of 3.7×10^6 tons per year.

2. The mass accretion is a function of latitude. In the absence of better information, we shall assume that the totality of extraterrestrial matter behaves in a way similar to the stratospheric strontium 90 in the southern hemisphere in 1964 and 1965, when its distribution with latitude was approximately uniform [*Feely et al.*, 1966]. In 1965 the average Sr 90 deposition over the whole southern hemisphere was 1.23 mCi/km² [*Volchok*, 1966], whereas its value at the south pole was 0.47 mCi/km² (by extrapolating the results of *Wilgain et al.* [1965]). This factor of 2.5 leads to a total accretion close to 10×10^6 tons per year. This value is considered as an upper limit, since a fraction of the extraterrestrial dust reaches the ground under the action of gravity independently of any atmospheric process.

Conclusions

The main aim of this work was to appraise the suitability of the eastern Antarctic Plateau for collecting and studying the extraterrestrial matter falling to earth. The extreme chemical purity of the snow samples collected at the south pole strongly supports the assertion that this area (and even more so, the area of the so-called Pole of Inaccessibility) is the most favorable environment for making such studies.

A tentative conclusion of the present work is that the total extraterrestrial mass accreted by earth is comprised between 3 and 10 million tons per year. These limits themselves are subject to considerable uncertainties arising mainly from the following restrictions:

1. The sampling statistics, both in time and over area, are very poor. The conclusions are based on analyses of snows fallen in the south pole region during the year 1952 or 1953 over an area of less than 0.1 m².

2. The extraterrestrial origin of the nickel in our samples is not definitely ascertained, although we have no better explanation for the chemical ratios observed.

3. The Ni concentration of 1.35% assumed for extraterrestrial dust is based on an analogy with meteoritic matter but has no experimental basis.

We do not intend to make a detailed comparison with other estimates here, but a few remarks may be worth underlining.

Estimates based on spherule observations are subject to errors both by excess (some of the spherules possibly being of terrestrial origin) and by defect (spherules representing only a fraction of the total extraterrestrial mass accretion). The most recent estimates (Table 8) are in general agreement around a value of 10^5 to 5×10^8 tons of spherules per year and are thus not inconsistent either with our lower limit for the total mass accreted or with an extraterrestrial origin of the spherules.

The value of 2.4×10^9 tons per year proposed by *Grjebine* [1964] for the total mass accretion is totally inconsistent with our upper limit.

Our estimates agree with recent data obtained by completely independent methods: satellite measurements [*Alexander et al.*, 1963] lead to accretion rate estimates of 4×10^8 tons per year; the Cl 36 activity found in marine sediments by Schaeffer et al. (reported by *Tilles* Z1966]) enables them to estimate a minimum influx of extraterrestrial dust of about 10^8 tons per year.

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