The Si^{32} fallout in Scandinavia

A new method for ice dating

By W. DANSGAARD, H. B. CLAUSEN, *Physical Laboratory II, University of Copenhagen, Denmark,* and A. AARKROG, *Danish A. E. C. Research Establishment, Risø, Denmark.*

(Manuscript received December 2, 1965)

ABSTRACT

The specific Si^{32} activity of air in Denmark showed a seasonal variation by a factor of 5 in 1963. The total Si^{32} fallout was $5.5 \cdot 10^{-5}$ dpm cm^{-2} $year^{-1}$ in the period Sept. 64–Sept. 65. Measurements on accumulated snow and old glacier ice in Norway show Si^{32} as an usable element for dating of up to 2000–3000 years old ice.

Introduction

Cosmic ray produced Si^{32} was first detected by LAL *et al.* (1960). Estimates on its half life time vary from 500 years (HONDA & LAL, 1964) to 650 years (GEITHOFF, 1962).

The preparation of samples for Si^{32} determination is essentially the concentration procedure for silica (scavenging by $Fe(OH)_3$) followed by a milking of the daughter P^{32} (LAL *et al.*, 1960). Even samples heavily contaminated by fission products can be sufficiently rinsed. Reduction of the total β-activity by a factor of higher than 10^8 has been obtained with a nearly 100% recovery of the silica (CLAUSEN, 1965). In spite of the collection and preparation of the samples being relatively complicated, Si^{32} would seem to become an important tool in geochemical research, because (*a*) it is constantly being produced in the atmosphere by spallation of argon nuclei, (*b*) a half life of approx. 600 years is well suited for many geochemical problems (e.g. in glaciological work it fills up the gap between 19 years for Pb^{210} and 5700 years for C^{14}, cp. section 3) and (*c*) unlike tritium and C^{14}, the deposition of Si^{32} from the atmosphere is a one way process (which has not yet been studied in details).

Production of Si^{32} in the nuclear weapons tests would, of course, disturb the application of Si^{32} in many respects, at least for several years.

LAL & PETERS (1962) calculated the cosmic ray production of Si^{32} to $1.6 \cdot 10^{-4}$ atoms $cm^{-2} \cdot sec^{-1}$, of which 30% is assumed to be produced in the troposphere and to be deposited practically independent on the latitude (curve I, Fig. 1). Most of the 70% produced in the stratosphere is assumed to be transferred to the troposphere at mid latitudes in early summer and to be deposited with the same latitudinal distribution as observed for Sr^{90}, i.e. with a bulk at mid latitudes. The expected pattern of the total annual fall out, A_p, is given by curve II (LAL, 1963) in Fig. 1.

If we consider the latitudinal distribution of precipitation (MØLLER, 1951, curve III, Fig. 1) and ignore the minor difference between geomagnetic and geographic latitude, which is hardly important in this respect, a simple division gives the average specific Si^{32} activity of precipitation, $\bar{a}_p$, as a function of the latitude (curve IV, Fig. 1). Assuming a relative uncertainty of no less than 15%, $\bar{a}_p$ is expectedly independent on λ for $\lambda > 30°$.

In our mind, the available data are too sparse to give clear experimental evidence for the global production and for the latitude effect on the $\bar{a}_p$.

The data presented in this paper is considered only as a modest contribution to the solution of these problems. Our data show (1) the seasonal variation of the Si^{32} fallout in Denmark in 1963 and 1964–65, (2) the total fallout of Si^{32} in Denmark in 1964–65, and they also show the possibility of (3) ice dating by Si^{32}.

1. The seasonal effect

The half life of Si^{32} ($\sim$600 years) is long compared to its residence times in the stratosphere and in the troposphere. These are 1–10 years

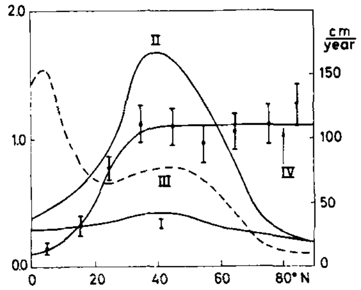

FIG. 1. *Curve I:* The production in the troposphere of an isotope, the total production of which averages 1 atom cm^{-2} sec^{-1} (after LAL, 1963). Scale to the left. *Curve II:* The expected pattern of the total fallout of the isotope scavenged from the atmosphere by precipitation (after LAL, 1963). *Curve III:* Latitudinal distribution of precipitation on the Northern hemisphere (after Møller, 1951). Scale to the right. *Curve IV:* The expected pattern of the specific activity of precipitation.

and 1 month, respectively, assuming Si^{32} to be scavenged from the atmosphere by the same processes as other radioactive aerosols by precipitation according to the GREENFIELD theory (1957). Since the residence time in the troposphere (1 month) is short compared with the period of injection from the stratosphere (1 year), one must expect a seasonal variation of the specific Si^{32} activity, a_a, of tropospheric air and, consequently, of the specific Si^{32} activity, a_p, of precipitation. However, the stratospheric contribution (curve II minus curve I, Fig. 1) to the total Si^{32} fallout at a given location varies from approx. 80 % at mid latitudes till very little at high latitudes. Therefore, the "peak to valley ratio" of a_a and a_p is probably strongly dependant on λ.

(a) *Air samples*

CAMBRAY *et al.* (1964) showed that the Cs^{137} concentrations in air at 4000 ft altitude and at ground level vary in a parallel fashion. Assuming a similar parallelism to hold for Si^{32} one should be able to measure the seasonal variation of a_p for Si^{32} by investigating air filters. This procedure would even make us independent on possible accidental periods of more or less local drought.

G. F. A. Wattman glass fibre filters are more than 99.9 % effective in filtering radioactive fallout (LOCHHART *et al.*, 1964). Six series of filters were passed by $5 \cdot 10^5$ to 10^6 m^3 of air, each series covering a 2-month period. Altogether they give a record of the specific Si^{32} activity (a_a dpm/kg air) of near ground level air at Risø within the year March 63 to Feb. 64 (Fig. 2 and Table 1).

The highest activity is $7.7 \cdot 10^{-3}$ dpm ton^{-1} in May–June, the lowest $1.4_4 \cdot 10^{-3}$ dpm ton^{-1} in Nov.–Dec., the average of all 6 samples is $3.7 \cdot 10^{-3}$ dpm ton^{-1}. The seasonal variation, i.e. the summer to winter value ratio in 1963, is 5.3.

If one subtracts $1.0 \cdot 10^{-3}$ dpm ton^{-1} from the 3 values on the last half of 1963, the 3 corrected values fall practically on a straight line with a slope corresponding to the observed residence time of Sr^{90} in the troposphere (approx. 1 month). The contribution $1.0 \cdot 10^{-3}$ dpm ton^{-1} represents, firstly, the continuous Si^{32} production in the troposphere and, secondly, the amount of Si^{32}, which is constantly brought down through the tropopause by diffusion, turbulence etc. The line reflects the exponential decrease due to washout of the remains of the stratospheric input in early summer.

(b) *Precipitation samples*

Systematic collection of precipitation was initiated Sept. 1964 from a roof at Glostrup, 12 km West of Copenhagen. Approx. 50 m^2 was covered by plastic foil, which had no detectable Si^{32} activity after the collection of rain. Thus, we are sure of having collected the total Si^{32} fallout. All dust was prepared together with the rain.

Fig. 3 and Table 1 show the results on samples No. 7 to 12 covering almost a full year. The mean value, weighted with the amounts of precipitation, is 0.95 dpm ton^{-1}. As in the case

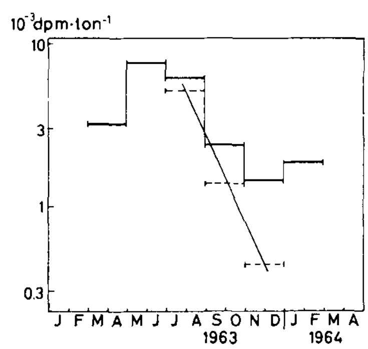

FIG. 2. Specific Si^{32} activity of air 3 meters above the ground at Risø, Denmark. The values indicated by dashed lines are corrected for estimated "tropospheric contribution", cp. text.

TABLE 1

No.	Sample	Location	Period represented	Amount (tons)	Counting rate (cpm)	Specific activity (dpm ton $^{-1}$)
1	Air	Risø	Mar.–Apr., 63	1190	0.82	$3.2 \cdot 10^{-3}$
2			May–June, 63	1260	1.46	$7.7 \cdot 10^{-3}$
3			July–Aug., 63	1240	2.00	$6.2 \cdot 10^{-3}$
4			Sept.–Oct., 63	1230	0.67	$2.4 \cdot 10^{-3}$
5			Nov.–Dec., 63	1270	0.47	$1.4_4 \cdot 10^{-3}$
6			Jan.–Feb., 64	730	0.24	$1.8_4 \cdot 10^{-3}$
7	Rain	Glostrup	Sept. 9–Nov. 5, 64	1.8	0.40	0.85
8			Nov. 6–Dec. 16, 64	2.4	0.38	0.63
9			Dec. 17, 64–Jan. 17, 65	1.8	0.29	0.64
10			Mar. 16–May 17, 65	3.9	0.56	0.69
11			May 18–June 23, 65	1.8	1.05	2.7
12			June 24–Sept. 3, 65	5.4	1.02	1.07
13	Ground water	Copenhagen	?	5	0.00	0.00
14	Snow	Leirvassbu	Early (?) winter 61–62	14.5	1.75	0.23[a]
15	Melt water	Store Breen	As above	10	0.40	0.19
16	*Glacier ice*		(1250 ± 120) A.D.? according to C^{14} dating	20	0.24	0.11

The estimated accuracy is ±0.04 dpm ton^{-1} on sample No. 13, 20 % on No. 14 and less than ±10 % on the rest. [a] See added note on p. 191.

of air, the extremes occurred in May–June (2.7 dpm ton^{-1}) and in Nov.–Dec. (0.63 dpm ton^{-1}) with a ratio of 4.3.

2. The total Si^{32} fallout in Denmark 1964–65

The total precipitation at the site of collection was 58 cm in the period Sept. 64 to Sept. 65. With $\bar{a}_p = 0.95$ dpm ton^{-1} one finds $A_p = 5.5 \cdot 10^{-5}$ dpm cm^{-2} year^{-1} $= 8.0 \cdot 10^{-4}$ atoms cm^{-2} sec^{-1}, assuming a half life of 600 years.

3. Dating of glacier ice by Si^{32}

(a) *Introduction*

One of the most important parameters in glaciological research is the age of the considered snow and ice, which leads to estimates on the net accumulation or on the velocity of moving glacier ice. In the past decade a number of methods have been developed on the basis of the measurements of stable and radioactive isotopes as well as chemical compounds.

The first attempt to use the radio-carbon dating method on old ice (Store Breen in Jotunheimen, Norway) was made by COACHMAN *et al.* (1958). The C^{14} content corresponded to an age of 710 ± 120 years in excellent agreement with estimates made by the Norwegian Polar Institute. The same technique was later applied to 11 West Greenland icebergs (SCHOLANDER *et al.* 1962). The same authors considered various sources of error and found most of them negligible. Nevertheless, the addition of recent CO_2 or organic compounds at the later stages of the movement towards the coastal glaciers stands out as a possibility, which is difficult to exclude: Whereas meltwater in the ablation zone one may just run off an ice mass with a temperature of 0°C in the summer time, it may be partly trapped in the cold, deeper strata of a high polar glacier. Consequently, it would be of great interest to date icebergs by

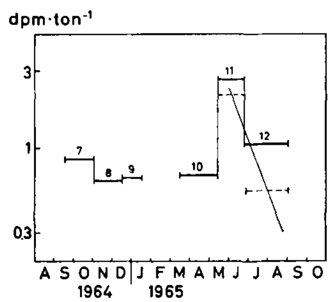

FIG. 3. Specific Si^{32} activity of precipitation at Glostrup, Denmark. The values indicated by dashed lines are corrected for estimated "tropospheric contribution", cp. text.

means of C^{14} and, simultaneously, by another, independent method.

The use of the fallout of cosmic ray produced Si^{32} for ice dating was proposed by LAL *et al.* (1960). Comparison between the C^{14} and Si^{32} "ages" of several icebergs might lead to indications on the constancy of the cosmic ray flux (LAL, 1964); the C^{14} concentration in the atmosphere would, namely, be much less sensible to changes (due to the buffer effect of the oceans) than the Si^{32} fallout.

(b) *Results*

Our techniques for snow and ice melting and for silica extraction (by precipitation of Fe $(OH)_3$) in the field were tested on Store Breen, Jotunheimen, Norway, in the summer 1962. The Si^{32} content of the old glacier ice (sample 16) had to be compared with that of recent net-accumulated snow. The latter sample was replaced, firstly, by the remains of winter snow (sample 14) found at an accessible nereby location and, secondly, by melt water from winter snow (sample 15).

(c) *Discussion*

If the specific activities of samples 14 and 15 are significantly different, the reason might be adhesion to or exchange with inactive silica from the silicates in the sand and gravel, over which the melt water flowed on its way down to the site of collection.

Accepting a half life of 600 years for Si^{32}, a comparison between samples No. 14 and No. 16 gives an apparent age of 660 ± 160 years for the old ice in agreement with the C^{14} age 710 ± 120 years.

The agreement between the C^{14} and Si^{32} ages of the Store Breen ice indicates that the C^{14} dating method is reliable when applied on temperate glaciers, in spite of the intense melting. However, the reason could be that all the melt water runs off, and the conclusion should, therefore, not be generalized to concern high Polar glaciers. A Si^{32} dating requires samples of the order of 5 tons, but a much simpler field equipment than a C^{14} dating. The technique to be used in future experiments of this kind will involve, firstly, melting the ice for extraction of CO_2 and, secondly, extraction of Si^{32} from the same melt water.

(d) *The limit of the method*

As to the limit of dating by Si^{32} we consider a sample of M tons of water with the initial specific activity a_0 dpm. ton^{-1}. After the time t years the total activity will be

$$a_0 M \exp\left(-\frac{0.69}{T}t\right) \text{ dpm},$$

T being the half life time of Si^{32}. In a counter with $E\%$ efficiency this activity will appear with a counting rate of

$$C = \frac{Ea_0 M}{100} \exp\left(-\frac{0.69}{T}t\right) \text{ cpm}.$$

The lowest activity to be measured with an accuracy of $p\%$ is given by

$$c = \frac{100}{p}\sqrt{\frac{2b}{\tau}},$$

b being the counter background and τ the counting time. In practise all samples were counted several days spread over at least 3 weeks for identifying the P^{32} by its half life time and for measuring possible impurities. However, by the assumption of no radioactive contamination counting during 5 days interrupted by 5 days background counting would be a reasonable procedure. This would give standard deviations on both c and b corresponding to $\tau =$ 120 hours.

The highest age, t_{max}, to be measured with a standard deviation of $p\%$ is then given by putting c equal to C, i.e.

$$t_{max} = -\frac{T}{0.16} \ln\left(\frac{10^4}{Ea_0 Mp} \cdot \sqrt{\frac{2b}{\tau}}\right) \text{ years.}$$

Considering the best counter used in this work, choosing a reasonable size of the sample with the same initial activity as the Norwegian snow, and requiring a fairly high accuracy, we insert

$b = 0.07$ cpm, $\tau = 120 \cdot 60$ min., $E = 50\%$, $M = 5$ tons, $a_0 = 0.2$ dpm ton^{-1}, $p = 20\%$, $T =$ 600 years,

and get $t_{max} = 2600$ years.

The limit of detection of Si^{32} corresponds to an age of the ice, which is found by inserting $p =$ 100%:

$$t_{max} = 4200 \text{ years.}$$

Of course, only one half life time (600 years) would be added to these ages by preparing the double amount of ice.

This means that if a_0 on the Greenland ice cap is of the same order as in Norway, and if the radiocarbon ages were correct, at least ten out of the eleven hitherto C^{14} dated icebergs could easily have been dated by Si^{32}.

Note: Since this was written we have found weighty indication for contribution of bomb produced Si^{32} to the fallout, especially in 1963 and 1964 (Dansgaard et al., 1966). The specific Si^{32} activity of sample No. 14 has been corrected for the contribution of bomb produced Si^{32}, which was estimated from comparative measurements on air filters from early winter 1961–62, and from the rest of the period 1960–65. The Si^{32} fallout is expected to be close to normal by the end of the 1960's.

Acknowledgements

The authors are greatly indebted to Miss E. M. Olsen, F. Carlsen, M.Sc., Mr. J. H. Jensen and J. D. Bunch, M.Sc., for excellent and enthusiastic assistance during the Norway expedition. Our sincere thanks are due to the Danish Defence Research Board and to Mr. Å. Elveseter for valuable help in collecting the glacier sample, to Mr. F. Foldager Petersen for excellent assistance in the difficult collection of precipitation samples, to Cand. Polyt. K. Mossin and Cand. Polyt. Frederiksen, technical director of NOVO Industry Ltd., for lending to us a separator, to Cand. Polyt. F. B. K. Karlsborg and Cand. Polyt. E. J. Maglekilde-Petersen, the Copenhagen Water Works, for obliging help in connection with the ground water sampling, and last, but not least, to Professors J. Koch and D. Lal for their continuous support and interest throughout the work.

Financial support was granted by the Danish State Research Foundation.

REFERENCES

Cambray, R. S., Miss Fisher, E. M. R., Spicer, G. S., Wallace, C. G., and Webber, T. J., 1964, Radioactive fall-out in air and rain: Results to the middle of 1964. *AERE–R*4687, Harwell.

Clausen, H. B., 1965, Extraction of P^{32} from heavily contaminated air filters. Manuscript in preparation.

Coachman, L. K., Hemmingsen, E., Scholander, P. F., Enns, T., and de Vries, H., 1958, Gases in glaciers. *Science*, 127, p. 1288.

Dansgaard, W., Clausen, H. B., and Aarkrog, A., 1966, Evidence for bomb produced Si^{32}, *Journ. of Geophys. Res.* (in press).

Geithoff, D., 1962, The production of Si^{32} by means of $a(t,p)$ process. *Radiochim. Acta*, 1, p. 3.

Greenfield, S. M., 1957, Rain scavenging of radioactive particulate matter from the atmosphere. *Journ. of Met.*, 14, p. 115.

Honda, M., and Lal, D., 1964, Spallation cross-sections for long lived radio-nucleides. *Nuclear Physics*, **51**, p. 363.

Lal, D., 1963, On the investigations of geophysical processes using cosmic ray produced radioactivity. In *Earth Science and Meteoritics*, compiled by J. Geiss and E. D. Goldberg, North-Holland Publishing Co., Amsterdam, 1963.

Lal, D., 1964, Personal communication.

Lal, D., Goldberg, E. D., and Koide, M., 1960, Cosmic ray produced Si^{32} in Nature. *Science* **131**, p. 332.

Lal, D., and Peters, B., 1962, Cosmic ray produced isotopes and their applications to problems in geophysics. *Progress in Cosmic Ray and Elementary Particle Physics*, Vol. **6**, North-Holland Publishing Co., Amsterdam.

Lockhart, L. B., jr., Patterson, R. L., jr., and Anderson, W. L., 1964, Characteristics of air filter media used for monitoring airborne radioactivity. *N.R.L. Rep.* **6054**.

Møller, F., 1951, Vierteljahrskarten des Niederschlages für die gane Erde. *Petermanns Geogr. Mitt.*

Scholander, P. F., Dansgaard, W., Nutt, D. C., de Vries, H., Coachman, L. K., and Hemmingsen, E., 1962, Radio-carbon dating and oxygen-18 analysis of Greenland icebergs. *Medd. om Grønland* **165**, No. 1.

ВЫПАДЕНИЕ Si^{32} В СКАНДИНАВИИ, И НОВЫЙ МЕТОД ДЛЯ ОПРЕДЕЛЕНИЯ ВОЗРАСТА ЛЬДА

В 1963 г. в Дании удельная радиоактивность Si^{32} воздуха изменялась в 5 раз. Полное выпадение Si^{32} равнялась $5{,}5 \cdot 10^{-5}$ dpm cm^{-2} $год^{-1}$ за период с сентября 64-сентябрь 65 г. Измерения, выполненные для старого ледникового льда и скопившегося снега в Норвегии, показали возможность использования Si^{32} для датировки льда возростом до 2000–3000 лет.