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TEMPORAL ¹⁰ BE VARIATIONS IN ICE

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INTRODUCTION

¹⁰ Be $(T_{1/2} = 1.5 \cdot 10^6 y)$ is mainly produced in the atmosphere by cosmic ray spallation reactions on nitrogen and oxygen. About 70 % of the production takes place in the stratosphere. ¹⁰Be becomes attached to aerosols within a very short time. If ¹⁰Be is produced in the stratosphere some latitudinal mixing occurs. Most of the ¹⁰Be is transferred to the troposphere during spring and early summer when, mainly at median latitudes, large stratospheric air masses enter the troposphere. Tropospheric ¹⁰Be is deposited rapidly on the earth's surface by precipitation. The mean residence time of ¹⁰Be in the atmosphere is call to 2 years. ¹⁰Be removed from the atmosphere by precipitation is either preserved in snow and ice layers, in the topsoil and the biosphere, or it enters the hydrosphere (oceans and lakes), where it is transported to the sediments.

Many precise 14 C measurements on tree rings covering the last 8000 years (Suess, 1980; Stuiver and Quay, 1980) clearly show that atmospheric 14 C concentration has not been constant. Possible causes of these variations with amplitudes of 1 to 2 % are 1) changes of the production rate due to changes of the galactic cosmic ray intensity. Incoming

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cosmic ray flux is modulated by the magnetic properties of the solar wind plasma and the intensity of the geomagnetic field; 2) changes of the global carbon cycle (Siegenthaler, Heimann, and Oeschger, 1980). Reservoir sizes and exchange fluxes can be influenced by changes of environmental conditions.

Fluctuations of the 14 C production rate are strongly dampened due to the large size of the atmospheric CO₂ reservoir and the exchange processes with the ocean. Model calculations show that, eg, the production variation induced by the ll-year solar cycle is attenuated by a factor of 100. Because of its rapid transfer from the atmosphere to the geosphere 10 Be responds with a much greater amplitude to changes of the production rate. The ll-year production variation is attenuated only by ca 20 %.

Until 1977 only a few ¹⁰Be measurements were made mainly on ocean sediments (Somayajulu, 1977; Finkel, Krishnaswami, and Clarck, 1977). Because of its \sim 100 times smaller global production rate compared to ^{14}C and its \sim 300 times larger half-life, the detection of 10_{Be} by conventional low-level counting techniques is very difficult. For the first ¹⁰Be measurement on polar ice which represents the best record of precipitation, 1.2.106kg of meltwater were processed (McCorkell, Fireman, and Langway, 1967). Since the development of accelerator mass spectrometry in 1977, samples of ca lkg containing $\sim 10^7 \ ^{10}$ Be atoms are sufficient for a measurement. The first sets of ¹⁰Be measurements in ice cores from Antarctica using the accelerator technique show very promising results (Raisbeck et al, 1981). To study ¹⁰Be variations in ice cores for at least the last 10^5 years, we started with two sets of samples drilled at Dye 3, Greenland. The main goal of set A for 1900 - 1976 is to study short-term fluctuations caused by changes of solar activity, ie, the ll-year solar cycle. Set B contains 14 samples distributed over the depth range 1300 to 1950m corresponding to a period from 3600 BP to \sim 30,000 BP. These samples should provide some information on long-term fluctuations and on the changing conditions during the transition from glacial to postglacial times (10,000 -13,000 BP).

SAMPLE PREPARATION

Samples were prepared from two ice cores drilled at Dye 3, Greenland (65° ll' N, 43° 50'W) for GISP (Greenland Ice Sheet Project) an American-Danish-Swiss collaboration. Samples of set A were prepared from a 70m shallow firn core. The $\delta^{18}{\rm O}$

profile (ca 8 samples per year) measured by the Danish group was used to cut the core into pieces containing the precipitation of one year. Each sample was mechanically cleaned and melted. Aliquots for tritium and chemical analysis were taken. Before adding Be- (1.18 mg Be) and Cl-carrier (2 mg Cl) the precipitation of two years was combined to one sample to increase the 10_{Be} concentration. First, the water volume (2 to 6kg, cf table 1) was reduced by evaporation to ca 30ml. Then the chlorine was separated by precipitation of AqCl and purified as described elsewhere (Nishiizumi et al, 1979). After an additional volume reduction to 2ml, the samples were analyzed by gamma spectroscopy. Beryllium acetylacetonate in the presence of EDTA was extracted into CHCl3 and evaporated after adding HCl. The organic material was oxidized with aqua-regia. Be(OH)) was precipitated with NH_AOH and converted to BeO by ignition at 950°C in a quartz crucible. The 14 samples of set B were prepared from ice of the 2037m long deep core, reaching bedrock. Depending on the depth, one sample represents the precipitation of ca 10 to 250 years. The samples were processed in the same way as the samples of set A.

MEASUREMENTS

The 10 Be concentrations were measured using the EN-tandem accelerator facility of the ETH Zürich. This system was designed in 1978 to detect, in a first step 14 C and 10 Be, and in a second step, 36 Cl and 26 Al in natural samples. The Cs sputter ion source produced BeO currents of up to 1 μ A leading to count rates of up to 10^3 cph for a typical 10 Be/ 9 Be ratio of $5 \cdot 10^{-13}$. The background is of the order of 10^{-14} depending on the boron content of the sample. The accelerator mass spectrometer is described in more detail by Woelfli et al (1983). The gamma activity of the samples was measured with a 64cc Ge(Li) detector. The tritium content was determined by a commercial liquid scintillation counter. Both counters were operated in a well-shielded underground laboratory (Oeschger et al, 1981).

RESULTS

The shape and the maximum of the nuclear bomb pulses of ${}^{3}\text{H}$ and ${}^{137}\text{Cs}$ were used to confirm the dating of the ice core based on $\delta^{18}\text{O}$ variations. The ${}^{10}\text{Be}$ concentrations and the weights of set A and B are given in tables 1 and 2. All samples (except 2) were measured twice, at different times. The measuring time (20-30 min) was divided into intervals of

Year	Sample weight	¹⁰ Be concentration 104 atoms/g
1900-1901	3853	1.11 ± 0.05
1902-1903	3192	0.91 ± 0.05
1904-1905	3354	1.09 ± 0.06
1906-1907	2295	1.27 ± 0.07
1908-1909	4236	0.79 = 0.05
1910-1911	2970	0.81 ± 0.06
1912-1913	4623	0.87 ± 0.05
1914-1915	3558	1.10 ± 0.10
1916-1917	3460	0.85 ± 0.05
1918-1919	3584	0.77 ± 0.09
1920-1921	3255	0.98 ± 0.14
1922-1923	3892	0.74 ± 0.09
1924-1925	3028	1.01 ± 0.20
1926-1927	3853	0.85 ± 0.14
1928-1929	4604	0.57 ± 0.07
1930-1931	2836	0.98 ± 0.13
1932-1933	3230	0.97 ± 0.09
1934-1935	3392	0.86 ± 0.10
1936-1937	2941	0.97 ± 0.13
1938-1939	6559	0.72 ± 0.05
1940-1941	4493	
1942-1943	3401	0.74 ± 0.11
1944-1945	2821	0.80 ± 0.12
1946-1947	4274	0.90 ± 0.08
1948-1949	2387	0.93 ± 0.11
1950-1951	2673	0.98 ± 0.08
1952-1953	2538	1.40 ± 0.12
1954-1955	3652	0.81 ± 0.12
1956-1957	2660	0.83 ± 0.11
1958-1959	3560	0.62 ± 0.06
1960-1961	3573	
1962-1963	2256	0.57 ± 0.15
1964-1965	3199	1.41 ± 0.12
1966-1967	2140	1.17 ± 0.17
1968-1969	2843	1.10 ± 0.10
1970-1971	3105	0.93 ± 0.08
1972-1973	3629	0.78 ± 0.07
1974-1975	3118	0.78 ± 0.07
1976-1977	3026	1.47 ± 0.10

TABLE 1.¹⁰Be concentrations in the Dye 3 firm core

TABLE 2. 10 Be concentrations in the Dye 3 deep core

Depth (m)	Sample weight (g)	<pre>10Be concentration (10⁴ atoms/g)</pre>
	1056	0 50 + 0 10
1314-1315	1826	0.59 - 0.10
1397-1398	1701	1.16 ± 0.14
1517-1517.5/1537-1537.5	1932	1.20 ± 0.15
1636-1636.5/1657-1657.5	1842	0.72 ± 0.13
1714.5-1715.5	2175	1.05 ± 0.12
1775.5-1776.5	1837	1.11 ± 0.12
1800.5-1801.5	1687	1.60 ± 0.15
1810.5-1811.5	1781	0.86 ± 0.09
1832.5-1833.5	1200	2.45 ± 0.13
1852.5-1853.5	1445	2.87 ± 0.14
1873.5-1874.5	1804	2.43 ± 0.16
1895.5-1896.5	1272	1.43 ± 0.11
1913.5-1914.5	1689	1.93 ± 0.12
1930-1931	2004	1.52 ± 0.15

 $50~{\rm sec.}$ The final result was obtained by calculating the weighted mean value of the two measurements. For absolute calibration the measurements were periodically compared to a $^{10}{\rm Be}$ standard with a known $^{10}{\rm Be}/^{9}{\rm Be}$ ratio.

The data of set A are plotted together with the sunspot numbers and the Δ^{14} C data of the period 1915-1940 (Stuiver and Quay, 1981) in figure 1. A comparison of the 10 Be spline function fit with the sunspot curve shows general agreement. The maximum of the spectral density function is at 13 \pm 3 years. The cross-correlation between the Be data and the sunspot numbers yields a phase lag of the 10 Be variations of 1.5 years which is consistent with the mean atmospheric residence time of 10 Be. Model calculations of the variation of the 10 Be production rate induced by the 11-year solar cycle predict changes of ca 60 % (Oeschger et al, 1970) in agreement with the variations shown in figure 1.



Fig 1. Comparison of $^{10}\textsc{Be}$ concentrations (two-year means) in the Dye 3 firn core with sunspot numbers $\,$ and $\Delta^{14}\textsc{C}$ variations

Between 1900 and 1960 the 10 Be concentration shows a slight decreasing trend which could be explained by the observed slow rise of the solar activity. After 1960 the 10 Be concentration rises again with greater amplitudes. There is no clear indication of a bomb peak due to nuclear weapon tests as observed for 137 Cs, T and 36 Cl (Elmore et al, 1982). The mean value between 1900 and 1976 is (0.93 \pm 0.22) 10⁴ atoms per gram of ice. The 14 C values shown in figure 1 were obtained by subtracting the Suess effect using linear regression. Data before 1915 were not used, since according to Stuiver, contamination cannot be excluded. The cross-correlation with the 10 Be curve reaches a maximum of 0.4 when 14 C lags 4 \pm 2.5 years behind 10 Be while CO₂ model calculations predict a lag of 3 years between the solar activity and the atmospheric 14 C concentration.

Table 2 and figure 2 give the results of the samples from the deep core. Because there is no general agreement about the age below 1780 m (corresponding to 10,000 BP) the data are listed as a function of depth. The time scale added in figure 2 is based on an ice flow model (Hammer <u>et al</u>, 1978). The $10_{\rm Be}$ concentration changes dramatically between 1810 and 1830m corresponding to the end of the last ice age.



Fig 2. 10_{Be} concentration in the deep ice core from Dye 3

The mean value of the first 6 points above 1780m is $(0.97 \pm 0.25) 10^4$ atoms/g concurring with the value found for the 20th century $(0.93 \pm 0.22) 10^4$ atoms/g.

DISCUSSION

The 10 Be concentration of the time period 1900 to 1976 reflects, to some extent, the ll-year solar cycle. This conclusion is supported by the size of the amplitude, the spectral density function, and the phase lag. Since measurements of 10 Be concentration in monthly rainwater in France (Raisbeck, 1979) show variations of a factor of 3, it is not surprising to find some "meteorological noise" in the two-year mean vaules. Absence of a clear nuclear bomb peak is expected because the environmental ⁹Be concentration and the n-activation cross-section are small.

Using the fallout pattern of Lal and Peters (1967) and the average rainfall for the latitude of Dye 3 (Moller, 1951) the mean 10 Be concentration of (0.93 \pm 0.22) 10⁴ atoms/g corresponds to a global deposition rate of 0.016 cm⁻²sec⁻¹, which agrees well with the value 0.018 cm⁻²sec⁻¹ derived by Amin, Lal, and Somayajulu (1975). In spite of a general similarity between the 10 Be and the 14 C curve, the number of data points is not sufficient for a clear correlation. More measurements covering longer periods are needed.



Fig 3. Comparison of the ^{10}Be concentrations around the transition from glacial to postglacial time (\sim 10,000 BP) with $\delta^{18}\text{O}$ values and CO $_2$ concentrations

There are three possible explanations for the significant change of ¹⁰Be concentration at the transition from Wisconsin to Holocene. The increase by a factor of 2.5 to 3 can be caused by an enhanced production rate, by changes of atmospheric circulation and mixing processes, or by changes of precipitation rates. Other parameters like δ^{18} O, CO₂ content, anion and dust concentration which have been measured on the same ice core also show significant variations at this depth. In figure 3 the 10Be values around the transition are plotted together with Danish δ^{18} O data and the CO $_2$ concentration (Stauffer et al, 1982). The correlation between these three data sets is surprisingly good. Raisbeck et al (1981) found the same features in an ice core from Dome C, Antarctica; an increase by a factor of 2 to 3 between 10,000 and 15,000 BP and a good correlation with the δ^{18} O curve. Considering that the increase of 10_{Be} concentration coincides with a strong climatic change, it is very probable that this effect can be attributed to serious changes of atmospheric circulations and precipitation rates. Herron and Langway (1982) find that, based on sulfate measurements, precipitation during the last ice age at Dye 3 was lower by a factor of 2 to 3. However, there are indications that the deposition rate in Antarctica was rather constant during this time. With the present information it is difficult to decide if the production rate of ¹⁰Be was higher during Wisconsin. Figure 1 and results deduced from ice samples of the Maunder minimum (Raisbeck et al, 1981) show that the production rate during periods of very low solar activity is increased by < 100 %. Thus, it seems improbable that changing solar magnetic properties alone could account for the observed threefold higher ¹⁰Be concentrations.

CONCLUSIONS

Despite some meteorologic disturbances, the $^{10}{\rm Be}$ data seem to reflect solar activity as well as climatic changes. If confirmed by more measurements from other sites, this observation has important implications. Records of solar activity provide basic information for understanding the solar cycle mechanism. Comparisons of $^{10}{\rm Be}$ and $^{14}{\rm C}$ data sequences enable us to distinguish variations due to fluctuations of the global carbon cycle. The correlation of the $^{10}{\rm Be}$ concentration with $\delta^{18}{\rm O}$ values and other climatic parameters could be very helpful in studying not only the climate of the last 10^5 to 10^6 years but also the extent to which climatic changes are influenced by the sun.

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REFERENCES

- Amin, BS, Lal, D, and Somayajulu, BLK, 1975, Chronology of marine sediments using the ¹⁰Be method: intercomparison with other methods: Geochim et Cosmochim Acta, v 39, p 1187-1192.
- Elmore, D, Tubbs, LE, Newman, D, Ma, XZ, Finkel, R, Nishiizumi, K, Beer, J, Oeschger, H, and Andrée, M, in press, The ³⁶Cl bomb pulse measured in a 100 m ice core from Dye 3, Greenland: submitted to Nature.
- Finkel, R, Krishnaswami, S, and Clarck, DL, 1977, ¹⁰Be in arctic ocean sediments: Earth Planetary Sci Letters, v 35, p 199-204.
- Hammer, CU, Clausen, HB, Dansgaard, W, Gundestrub, N, Johnsen, SJ, and Reeh, N, 1978, Dating of Greenland ice cores by flow models, isotopes, volcanic debris and continental dust: Jour Glaciol, v 20, p 3-26.
- Herron, MM, and Langway CC, 1982, Chloride, nitrate and sulfate in the Dye 3 and Camp Century, Greenland, ice cores: Am Geophys Union mtg, Philadelphia, May 31-June 14, 1982.
- Lal, D, and Peters, B, 1967, Cosmic ray produced radioactivity on the earth, in Flügge, S, ed, Handbuch Physik: Berlin, Springer, v 462, p 551-612.
- McCorkell, R, Fireman, EL, and Langway, CC, 1967, Aluminum-26 and Beryllium-10 in Greenland ice: Science, v 158, p 1690-1692.
- Moller, F, 1951, Vierteljahreskarten des Niederschlages für die ganze Erde: Petermanns Geog Mitt, v 95, p 1-7.
- Nishiizumi, K, Arnold, JR, Elmore, D, Ferraro, RD, Gove, HE, Finkel, RC, Beukens, RO, Chang, KH, and Kilius, LR, 1979, Measurements of ³⁶Cl in antarctic meteorites and antarctic ice using a van de Graaff accelerator, in Earth Planetary

Sci Letters, v 45, p 285-292.

- Oeschger, H, Beer, J, Loosli, HH, and Schotterer, U, 1981, Low level counting systems in deep underground laboratories, in Methods of low-level-counting and spectrometry: IAEA symposium, Berlin 1981, Proc: p 459-474.
- Oeschger, H, Houtermans, J, Loosli, HH, and Wahlen, M, 1970, The constancy of cosmic radiation from isotope studies in meteorites and on the Earth, in Olsson, IU, ed, Radiocarbon variations and absolute chronology: New York, John Wiley + Sons, p 471-498.
- Raisbeck, GM, Yiou, F, Fruneau, M, Loiseaux, JM, Lieuvin, M, and Ravel, JC, 1979, Cosmogenic ¹⁰Be; mean deposition rate and seasonal concentration variations in precipitation: Nature, v 282, p 279-280.
- Raisbeck, GM, Yiou, F, Lieuvin, M, Ravel, JC, Fruneau, M, and Loiseaux, JM, 1981, ¹⁰Be in the environment: some recent results and their applications, <u>in</u> Kutschera, W, ed, Symposium on accelerator mass spectrometry, Proc: Argonne, Illinois, p 228-243.
- Siegenthaler, U, Heimann, M, and Oeschger, H, 1980, ¹⁴C variations caused by changes in the global carbon cycle: <u>in</u> Stuiver, Minze and Kra, Renee, eds, Internatl radiocarbon Conf, 10th, Proc: Radiocarbon, v 22, no. 2, p 177-191.
- Somayajulu, BLK, 1977, Analysis of causes for the beryllium-10 variations in deep sea sediments: Geochim et Cosmochim Acta, v 41, p 909-913.
- Stauffer, B, Neftel, A, Oeschger, H, and Schwander, J, 1982, CO₂ concentration in air extracted from Greenland ice samples: Am Geophys Union mtg, Philadelphia, May 31 -June 14, 1982.
- Stuiver, Minze, and Quay, PD, 1980, Changes in atmospheric carbon-14 attributed to a variable sun: Science, v 207, p 11-19.
- Stuiver, Minze, and Quay, PD, 1981, Atmospheric ¹⁴C changes resulting from fossil fuel CO₂ release and cosmic ray flux variability: Earth Planetary Sci Letters, v 53, p 349-362.
- Suess, HE, 1980, The radiocarbon record in tree rings of the last 8000 years, <u>in</u> Stuiver, Minze and Kra, Renee, eds, Internatl radiocarbon Conf, 10th, Proc: v 22, no. 2, p 200-209.
- Woelfli, W, Bonani, G, Suter, M, Balzer, R, Nessi, M, and Stoller, Ch, 1982, Radioisotope dating with the ETHZ-ENtandem accelerator, in Stuiver, Minze and Kra, Renee, eds, Internatl radiocarbon Conf, 11th, Proc: Radiocarbon, v 25.