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TEMPORAL <sup>10</sup> BE VARIATIONS IN ICE

JUERG BEER, MICHAEL ANDREE, HANS OESCHGER AND BERNHARD STAUFFER

Physics Institute, University of Bern Sidlerstrasse 5, 3012 Bern, Switzerland

and

RICHARD BALZER, GEORGES BONANI, CHRISTIAN STOLLER, MARTIN SUTER AND WILLY WOELFLI

Laboratorium für Kernphysik Swiss Federal Institute of Technology ETHZ 8093 Zürich, Switzerland

and

ROBERT C FINKEL

Mt Soledad Laboratory Scripps Institution of Oceanography S - 002 La Jolla, California 92093, U S A

### INTRODUCTION

<sup>10</sup> 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. <sup>10</sup>Be becomes attached to aerosols within a very short time. If <sup>10</sup>Be is produced in the stratosphere some latitudinal mixing occurs. Most of the <sup>10</sup>Be is transferred to the troposphere during spring and early summer when, mainly at median latitudes, large stratospheric air masses enter the troposphere. Tropospheric <sup>10</sup>Be is deposited rapidly on the earth's surface by precipitation. The mean residence time of <sup>10</sup>Be in the atmosphere is call to 2 years. <sup>10</sup>Be 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 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 CO<sub>2</sub> 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 <sup>10</sup>Be 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 <sup>10</sup>Be 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 <sup>10</sup>Be measurements in ice cores from Antarctica using the accelerator technique show very promising results (Raisbeck et al, 1981). To study <sup>10</sup>Be 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° 11' 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  $10_{\text{Be}}$  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  $\mu$ 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

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| Year      | Sample weight<br>(g) | <sup>10</sup> Be concentration<br>10 <sup>4</sup> atoms/g |
|-----------|----------------------|---|
|           |                      | 1.11 ± 0.05   |
| 1900-1901 | 3853                 | $0.91 \pm 0.05$   |
| 1902-1903 | 3192                 | $1.09 \pm 0.06$   |
| 1904-1905 | 3354                 | $1.09 \pm 0.08$<br>$1.27 \pm 0.07$                        |
| 1906-1907 | 2295                 |   |
| 1908-1909 | 4236                 | 0.79 = 0.05   |
| 1910-1911 | 2970                 | $0.81 \pm 0.06$<br>$0.87 \pm 0.05$                        |
| 1912-1913 | 4623                 |   |
| 1914-1915 | 3558                 | $1.10 \pm 0.10$   |
| 1916-1917 | 3460                 | 0.85 ± 0.05   |
| 1918-1919 | 3584                 | $0.77 \pm 0.09$   |
| 1920-1921 | 3255                 | $0.98 \pm 0.14$   |
| 1922-1923 | 3892                 | $0.74 \pm 0.09$   |
| 1924-1925 | 3028                 | $1.01 \pm 0.20$   |
| 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                 | $1.41 \pm 0.12$   |
| 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.<sup>10</sup>Be concentrations in the Dye 3 firm core

TABLE 2.  $^{10}$  Be concentrations in the Dye 3 deep core

| Depth<br>(m)            | Sample weight<br>(g) | <sup>10</sup> Be concentration<br>(10 <sup>4</sup> atoms/g) |
|-------------------------|----------------------|---|
|                         |                      |   |
| 1314-1315               | 1856                 | 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  $\Delta^{14}$ 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.

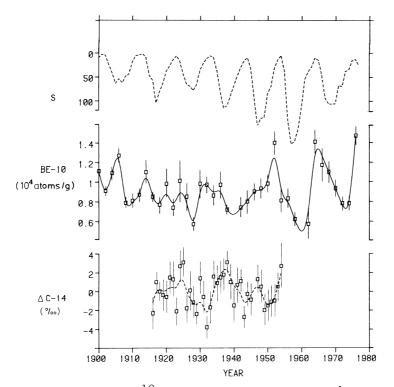


Fig 1. Comparison of  $^{10}\textsc{Be}$  concentrations (two-year means) in the Dye 3 firn core with sunspot numbers  $\,$  and  $\Delta^{14}\textsc{C}$  variations

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) 10<sup>4</sup> 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 CO<sub>2</sub> 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.

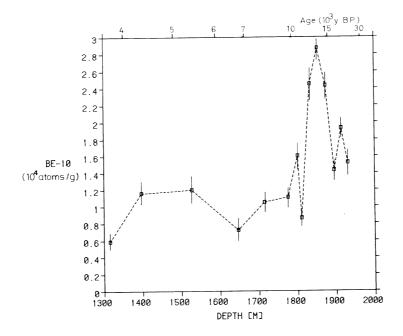


Fig 2.  $10_{\text{Be}}$  concentration in the deep ice core from Dye 3

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 <sup>9</sup>Be 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) 10<sup>4</sup> atoms/g corresponds to a global deposition rate of 0.016 cm<sup>-2</sup>sec<sup>-1</sup>, which agrees well with the value 0.018 cm<sup>-2</sup>sec<sup>-1</sup> 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.

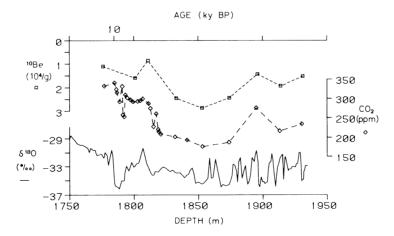


Fig 3. Comparison of the  $^{10}\text{Be}$  concentrations around the transition from glacial to postglacial time (  $\sim$  10,000 BP) with  $\delta^{18}\text{O}$  values and CO $_2$  concentrations

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There are three possible explanations for the significant change of <sup>10</sup>Be 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  $\delta^{18}$ O, CO<sub>2</sub> 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  $\delta^{18}$ 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  $\delta^{18}$ O curve. Considering that the increase of  $10_{\text{Be}}$  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 <sup>10</sup>Be 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 <sup>10</sup>Be 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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