IN-SITU RADIOCARBON PRODUCTION BY NEUTRONS AND MUONS IN AN ANTARCTIC BLUE ICE FIELD AT SCHARFFENBERGBOTNEN: A STATUS REPORT

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ABSTRACT. In the radiocarbon accelerator mass spectrometry (^{14}C AMS) analysis of gases obtained in a dry extraction from a 52-m Antarctic ice core, we observed $^{14}CO_2$ and ^{14}CO concentrations decreasing with depth. The concentrations are explained in terms of in-situ production by neutrons and captured muons in ablating ice. The ratio of the $^{14}CO_2$ concentration to that of ^{14}CO has been found to be constant at 1.9 ± 0.3. The ablation rates obtained of 42 ± 18 cm.yr^{-1} and 40 ± 13 cm.yr^{-1} for the neutron and muon components, respectively, are about three times higher than observed from stake readings. The discrepancy may point to an incomplete extraction of the dry extraction method. Using the constant ratio in $^{14}CO_2$ and ^{14}CO concentrations we correct for the in-situ component in the trapped $^{14}CO_2$ and deduce an age of 10,300 ± 900 BP for the ice core.

INTRODUCTION

Blue ice fields mainly occur in Antarctica at the outer edges of the main ice sheet, where the ice flow runs up against a mountain range and local winds remove the upper ice layer. In many cases blue ice fields form stranding-areas of meteorites, which after a long burial time come to the ice surface (Cassidy et al. 1992). As the ice originates from an accumulation area, where the precipitating snow forms a firn layer and gradually transforms to ice, it brings information to the blue ice field where it crops out. In the firn-to-ice transition at about 70 m (Schwander and Stauffer 1984) air becomes trapped in the ice. The gases are locked in the ice through their extremely small mobility and form a wealthy archive of ancient atmospheres (Neftel et al. 1983), which bears information on climate and greenhouse gases (Jouzel et al. 1987; Raynaud et al. 1993). The special role of the trapped carbon dioxide for radiocarbon dating of ice has been recognized early (Fireman 1982; Andreé 1984).

During the transportation from accumulation to ablation, regional ice is subjected to bombardment of cosmic rays, which produce radionuclides in the ice. In-situ production in the first few meters is dominated by reactions from neutrons, in the next tens of meters by captured muons and deeper by fast muons (Lal and Peters 1967). First reports on in-situ production by muons in rock were made for ^{36}Cl by Kubik et al. (1984) and for ^{10}Be by Brown et al. (1995). Heisinger (1998) determined production rates of various radionuclides in quartz by samples irradiated by muons.

Lal et al. (1987) indicated the ability to extract accumulation and ablation rates of ice sheets from the in-situ production and used ^{14}C in determining the ablation rate of Antarctic ice (Lal et al. 1990). Lal and Jull (1994) also observed in-situ ^{14}C in the firn of accumulation areas and found a variable in-situ fraction retained in the ice. The variability of the retained fraction has been attributed to erosion of the firn by variable wind ventilation in the firn (Lal et al. 2000).

In a previous study, van Roijen et al. (1996) performed ^{14}C accelerator mass spectrometry (AMS) analysis on 3- and 10-m ice cores from an Antarctic blue ice field at Scharffenbergbotnen. Ablation

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rates and ages were deduced. The ratio of the ^{14}CO and $^{14}CO_2$ concentrations showed to be constant in an individual core but different among cores. In addition, the asymptotic value at 10 m, the largest depth, could not be explained by the neutron component alone. For a further investigation of these aspects we returned to the location to take another and, ice core.

METHOD

The 52-m ice core was drilled in the ablating blue ice field at Scharffenbergbotnen, Antarctica (74°34′40″S, 11°02′58″W, elevation 1173 m) during the 1997–1998 SVEA field expedition. Ice samples of about 2 kg were used in a dry extraction technique in which rotating knives scrape ice in a vacuum recipient situated in a freezing box held at –35 °C. For extraction of the carbon monoxide a carrier has been added in advance. In the extraction procedure the carbon dioxide is collected with cold traps, and the remaining carbon monoxide is oxidized to CO_2 and collected subsequently. Yields were obtained from precise pressure measurement. A 95% efficiency is obtained in this on line oxidation procedure using platinum wire at 300 °C. The collected CO_2 fractions are converted into graphite and prepared for ^{14}C measurement with AMS (van der Borg et al. 1987, 1997). The contamination in the total extraction procedure has been determined from blanks and admixtures of CO and CO_2 of known activity and has been found to be 1.5 ± 0.5 pMC.

ANALYSIS AND RESULTS

Table 1 shows details about the ice samples and depth, the gas yields, and the results of the ^{14}C analysis. The mean value of the air yields from the ice samples is 108 mL/kg, which is in agreement with values earlier obtained by van Roijen (1995). The CO_2 concentrations are close to modern values in the surface samples. The average concentration of the other samples is about 300 ppmv, which is higher than Holocene values. The air yields show fluctuations of 20%, which may be due to differences in the ice matrix, e.g. cracks through which gases escape during evacuation of the dry extraction vessel. The ^{14}CO and $^{14}CO_2$ concentrations have about 5% errors, which are mainly due to background and fractionation corrections required in the AMS analysis of the small graphite samples (Alderliesten et al. 1998).

Table 1 Details about the samples of the Sbb52-core, yields, and concentrations

Depth (m)	Mass (kg)	Air yield[a] (mL/kg)	CO_2 conc.[b] (ppmv)	Mass[c] (μg C)	$^{14}CO_2$ conc. (at.g^{-1})	^{14}CO conc. (at.g^{-1})
0.9	2.032	98	321	31	—	134 ± 15
1.7	1.738	100	328	28	587 ± 30	156 ± 17
2.7	2.058	99	319	32	530 ± 27	105 ± 15
4.1	2.376	100	301	35	427 ± 19	62 ± 12
8.4	1.986	119	296	34	453 ± 23	89 ± 14
10.0	2.256	102	310	35	429 ± 19	67 ± 12
16.5	2.246	123	300	40	429 ± 18	60 ± 13
25.5	2.068	104	300	31	378 ± 20	40 ± 13
38.0	2.173	100	299	32	337 ± 18	24 ± 12
45.0	2.292	119	303	41	408 ± 18	36 ± 12

[a]STP, uncertainty estimated at 2%
[b]Uncertainty estimated at 5%
[c]Mass of trapped CO_2

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Figure 1 shows the ^{14}CO and $^{14}CO_2$ concentrations as a function of depth. Both profiles, showing a steep decrease, have close correspondence in shape, even in individual data points, reflecting a constant ratio of the ^{14}CO to $^{14}CO_2$ components formed during ablation. The samples at 8.4, 16.5, and 45.0 m, which have 20% higher air yield, show somewhat higher values in both the ^{14}CO to $^{14}CO_2$ profiles, but less than expected from a full correlation with the air yield. On the other hand, the low values for both the ^{14}CO and $^{14}CO_2$ concentrations of the 4.1-m sample do not show any fluctuation in air yield. Therefore, a correction to the concentrations based upon the air yield fluctuations does not seem to be straightforward.

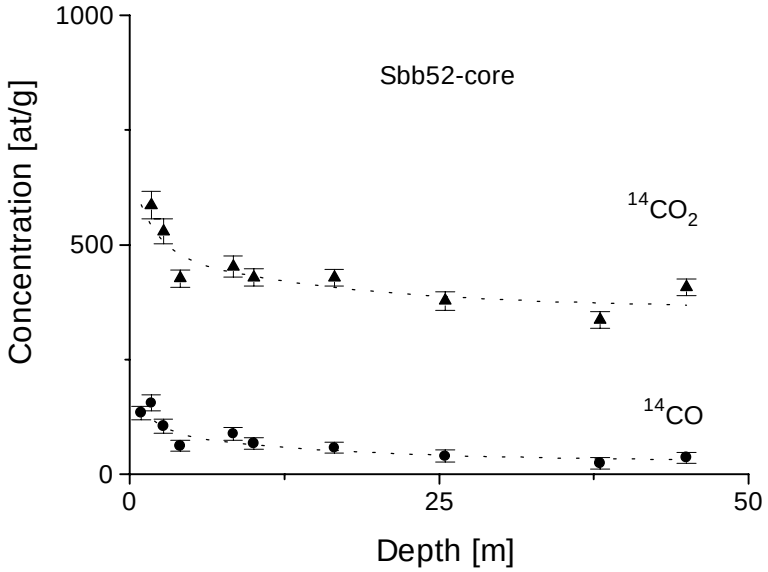

Figure 1 The concentration of ^{14}C as measured in the fractions $^{14}CO_2$ and ^{14}CO in a 52-m ice core taken at an Antarctic blue ice field at Scharffenbergbotnen. The decrease in depth of both profiles shows the in-situ production in an ablation region. The dotted lines are fits to the data.

In-situ production of ^{14}C in ice takes place during accumulation, during transport and during ablation of the ice. The ^{14}C concentration comprises both the $^{14}CO_2$ and ^{14}CO fractions from either trapped component or from in situ production.

The ^{14}C concentration $C(h)$ in ablating ice can be described by a component $C^{abl}(h)$ for in-situ ^{14}C production in the ablation region and by a component C from other origin:

$$C(h) = C^{abl}(h) + C\,. \tag{1.1}$$

The component $C^{abl}(h)$ reflects the depth dependence from the (different) attenuation of the neutrons and muons from the cosmic rays. The concentration C comprises the ^{14}C originating from the accumulation region in the form of trapped CO_2 and in-situ ^{14}C in the firn. The concentration C may have small depth but negligible dependence from the age difference in the core. The surface ice is expected to be the oldest as it has the longest pathway from the accumulation area. However, the time span of a few hundred years in the 52-m core—as estimated from the ablation rate—has a small effect on the ^{14}C concentration of the old ice (see below) with respect to the 5% uncertainty of the concentration.

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The depth dependence of the component $C^{abl}(h)$ is given by the attenuation lengths of the neutron and captured muons (we neglect the contribution from fast muons as this is cannot be discriminated through the attenuation length longer than the length of the core; we assume its contribution contained in C). We use

$$C^{abl}(h) = A_n e^{-\frac{h}{\mu_n}} + A_m e^{-\frac{h}{\mu_m}} \tag{1.2}$$

with

$$A_n = \frac{P_{0n}}{\lambda + \frac{a}{\mu_n}} \quad \text{and} \quad A_m = \frac{P_{0m}}{\lambda + \frac{a}{\mu_m}} \tag{1.3}$$

where A_n and A_m are the ^{14}C surface concentrations produced by neutrons and captured muons, respectively, as related to the ablation rate of a cm.yr^{-1} and the production rates P_n and P_m at.g^{-1}.yr^{-1} (Lal et al. 1990), with λ = ln 2/5730 yr^{-1} the decay constant and μ_n and μ_m the attenuation lengths with μ_n = 1.67 m (Lal et al. 1990) and μ_m = 15.2 m (Bilokon et al. 1989).

We start the analysis with the fit of the ^{14}CO profile, because it is fully attributed to in-situ production and no complication occurs from a trapped component. We use the formulas (1.1) and (1.2) with CO-labeled coefficients. The fit to the ^{14}CO-data, as shown in Figure 1, results in the values: $A^{CO}{}_n = 90 \pm 36$, $A^{CO}{}_m = 68 \pm 19$, and $C^{CO} = 29 \pm 8$ at.g^{-1}.

Subsequently, we fit the $^{14}CO_2$ profile, using the parameters found for the ^{14}CO profile and $A^{CO2}{}_n = r\,A^{CO}{}_n$ and $A^{CO2}{}_m = r\,A^{CO}{}_m$. We find $r = 1.9 \pm 0.3$ and $C^{CO2} = 363 \pm 11$ at.g^{-1}. For A_n, the sum of both components, we find $A_n = (1 + r)\,A^{CO}{}_n = 260 \pm 110$ at.g^{-1} for the neutron component and similarly $A_m = (1 + r)\,A^{CO2}{}_n = 200 \pm 60$ at.g^{-1} for the muon component. Using relation (1.3) for A_n, and the production rate by neutrons $P_n = 67 \pm 4$ at.g^{-1} at altitude 1170 m (as derived from $P_{0n} = 15.7 \pm 0.7$ at.g^{-1}.yr^{-1} for quartz by Lifton et al. (2000), which reproduces the value deduced from the parametrization given by Lal 1991), we obtain an ablation rate $a = 42 \pm 18$ cm.yr^{-1} from the neutron component. Using the value for A_m and the production rate by muons $P_m = 5.3 \pm 0.5$ at g^{-1} yr^{-1}—as derived for ice from $P_{0m} = 3.7 \pm 0.4$ at g^{-1} yr^{-1} in quartz (Heisinger 1998) by taking into account the differences for ^{28}Si and ^{16}O (von Egidy and Hartmann 1982)—we find an ablation rate $a = 40 \pm 13$ cm.yr^{-1} from the muon component.

Assuming in-situ production of $^{14}CO_2$ in the trapped CO_2 is related to the asymptotic value of ^{14}CO by the ratio $r = 1.9 \pm 0.3$ we estimate a total amount of $(1 + r)\,C^{CO} = 85 \pm 27$ at g^{-1}. Correcting with this amount, we find 280 ± 30 at g^{-1} for the trapped $^{14}CO_2$. Compared to 970 at.g^{-1} for modern CO_2 trapped in ice we find an age of 10,300 ± 900 BP. The large uncertainty obtained in the age is due to the sum of the uncertainties from the fitting procedure. The correction for the in situ component in the trapped CO_2 is based on the use of the ratio r obtained from ablating ice. Whether this is allowed for in situ production from other origin, e.g. from the accumulation region, has not been proven. However, for rather small asymptotic ^{14}CO values the influence on this correction by the value of this ratio is rather small in comparison with the total uncertainty in the age.

DISCUSSION

The ablation rates $a = 42 \pm 18$ cm.yr^{-1} and 40 ± 13 cm.yr^{-1} obtained from the in-situ production by neutrons and muons, respectively, are clearly higher that the 12 ± 4 cm.yr^{-1} ablation rate obtained from stake readings (Jonsson 1992). As the uncertainty in the production rates is only 10%, this can-

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not explain the large discrepancy. Although the stake readings only probe the last 10 years, a large change in ablation rate would have been observed in the radionuclide concentrations. A possible explanation of the discrepancy can be an incomplete extraction of the dry extraction method. That would imply an extraction efficiency of only 30% of the in situ ^{14}C components.

The constant ratio for in-situ production of $^{14}CO_2$ and ^{14}CO over the time span of a few hundred years in the 52-m core suggests equilibrium may be reached in the oxidation of ^{14}CO. Such equilibrium is not known and may depend on local ice conditions. Irradiation experiments in which ^{11}C was produced showed a CO_2 fraction increasing with the applied dose (Lambrecht 1997).

The asymptotic value C^{CO} of the ^{14}CO profile may be from in situ production from another origin. A part may originate from in-situ production by fast muons with a large attenuation length. Using P_f = 0.210 at.g $^{-1}$.yr^{-1} for ^{14}C production in ice by fast muons—as derived from P_f = 0.127 at.g $^{-1}$.yr^{-1} in quartz measured by Heisinger (1998)—we estimate about half of the asymptotic value to be ascribed to the production by fast muons. The other half may originate from the accumulation region where it has been produced in the firn by neutrons. As the same decay constant applies to both the in-situ and the trapped fractions retained in the ice, the ratio of both fractions is independent on depth. Lal and Jull (1994) derived for the ratio of the in situ fraction over the trapped fraction $R = 0.176\, P_0/s$, with P_0 the production rate of ^{14}C and s the accumulation rate of ice (Lal and Jull 1994). Using an average accumulation rate s = 6 cm.yr^{-1} and $P_0 = 67 \pm 4$ at.g $^{-1}$.yr^{-1} we find R = 2.0. Assuming 43 ± 13 at.g^{-1}, being the half of the asymptotic in-situ production, originates from the accumulation region we find for the 52-m core R = 0.45, taking into account a 30% extraction efficiency for the in situ components. That would mean a retainment of 23% in-situ ^{14}C of the firn in the ice, which is different from the observation by van der Kemp et al. (2000) who do not observe in situ production in accumulating ice.

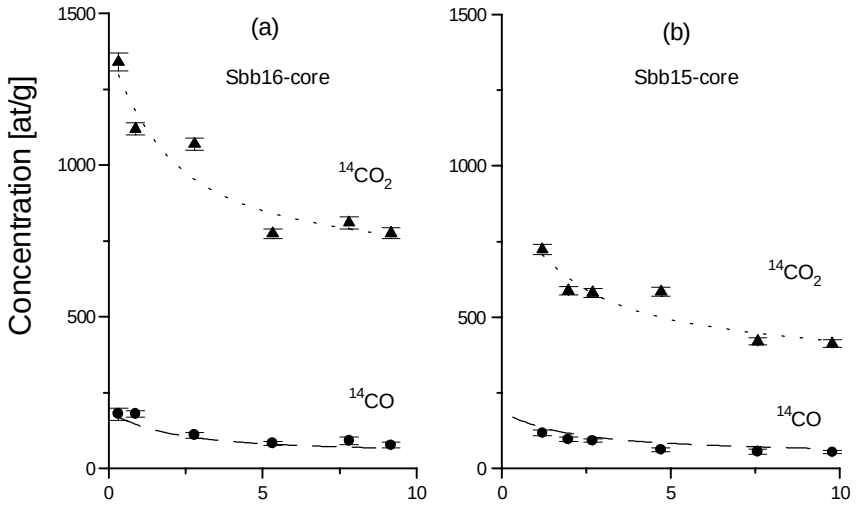

Figure 2 The ^{14}C data obtained by van Roijen et al. (1995) with 10-m ice cores of Scharffenbergbotnen in (a) Sbb-16, and (b) Sbb-15. The dashed lines in the ^{14}CO data have been taken from the 52-m core. The dotted lines are fits to the $^{14}CO_2$ data.

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The ^{14}CO profiles obtained by van Roijen et al. (1995) in shallow ice cores of 3 and 10 m from the same area are almost equal to the one obtained in the 52 m as is shown by the line through the data (Figure 2 above). This similarity indicates an equal extraction efficiency of ^{14}CO for the three cores. Using the same parametrization as used for the 52-m core we find $r = 4.0 \pm 0.2$, 5.1 ± 0.2 and $C^{CO2} = 277 \pm 16$ at.g^{-1}, 575 ± 18 at.g^{-1} for $^{14}CO_2$ profiles of the cores Sbb15 and Sbb16, respectively. Obviously, each core seems to have its own ratio r, which may reflect specific properties of the ice matrix. From the r-values we deduce the ablation rates $a = 25 \pm 10$ and 20 ± 8 cm from the neutron component and $a = 24 \pm 7$ at.g^{-1}, 19 ± 5 at.g^{-1} for the muon component, respectively, which all are about a factor of two higher than the value from stake readings. From the C^{CO2}-values we obtain, similarly as above, the ratio of in-situ to trapped components $R = 1.65$ and 0.66, respectively, which differ from the $R = 0.45$ obtained for the 52-m core, and which may be due to variable wind ventilation in the accumulation area (Lal et al. 2000).

CONCLUSIONS

We observed in-situ ^{14}C over the whole depth range of the 52-m ice core, reflecting the production by neutrons in the first few meters and the production by captured muons at larger depth. We derive an ablation rate $a = 42 \pm 18$ cm.yr^{-1}from the neutron component and 40 ± 13 cm.yr^{-1} from the muon component. Both values are about three times higher than the $a = 12 \pm 4$ cm.yr^{-1} observed from stake readings (Jonsson 1992) and may be explained by 30% efficiency in the dry extraction method for in-situ ^{14}C.

We deduce an age of 10,300 ± 900 BP for the 52-m ice core using a correction for in-situ-produced $^{14}CO_2$ in the trapped CO_2 fraction, which depends on the choice for the ratio of in-situ production of $^{14}CO_2$ and ^{14}CO in ablating ice. Although a constant ratio has been observed in the ablation profiles of individual cores, the large variability among cores, make application of this ratio uncertain for in situ production from other origin, such as e.g. the accumulation region.

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