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# Record of cosmogenic in situ produced $^{14}\text{C}$ in Vostok and Taylor Dome ice samples: Implications for strong role of wind ventilation processes

D. Lal,<sup>1</sup> A. J. T. Jull,<sup>2</sup> D. J. Donahue,<sup>2</sup> G. S. Burr,<sup>2</sup> B. Deck,<sup>1</sup> J. Jouzel,<sup>3</sup> and E. Steig<sup>4</sup>

**Abstract.** We present results of studies of in situ cosmogenic  $^{14}\text{C}$  in several ice samples from the Vostok and Taylor Dome cores, spanning the time intervals of 20 and 11 k.y. B.P., respectively. The results are in variance with our findings for the Greenland Ice Sheet Project 2 (GISP 2) samples, where near quantitative  $^{14}\text{C}$  retention was observed. The partitioning of  $^{14}\text{C}$  in the CO and CO<sub>2</sub> phases is, however, quite similar in the GISP and two Antarctic ice samples. Noting that most of the in situ  $^{14}\text{C}$  is produced in the ice during its accumulation to thicknesses of up to ~10 m, we interpret the observed  $^{14}\text{C}$  deficiencies in Antarctic ice samples as due to grain metamorphism (recrystallization and sublimation or evaporation caused by wind ventilation). Simplified models for wind ventilation exhibit an interesting feature of the firnification processes; they differently affect the concentrations of in situ  $^{14}\text{C}$  and of the cosmogenic nuclides  $^{10}\text{Be}$  and  $^{36}\text{Cl}$  scavenged from the atmosphere in the accumulating firn. By studying the concentrations of in situ cosmogenic  $^{14}\text{C}$  and the atmospheric cosmogenic nuclides in the same ice samples, one can hope to obtain fairly realistic models of the chemical impacts of firnification processes: specifically, relationships between precipitation and accumulation and the main processes contributing to modifications in the nuclide concentrations.

## 1. Introduction

It is now well known that nuclear interactions of energetic cosmic ray neutrons with oxygen in ice produce measurable quantities of in situ  $^{14}\text{C}$  in firn as it accumulates to depths of a few meters and in ablating ice as it outcrops [Lal *et al.*, 1990, 1997]. Furthermore, in the case of Greenland Ice Sheet Project 2 (GISP 2) samples, of ages up to 17 k.y. B.P. and accumulation rates of 10–30 cm yr<sup>-1</sup>, the results of Lal *et al.* [1997, 2000] have established that the in situ  $^{14}\text{C}$  produced in the firn during accumulation is retained even through the bubble-closing depth, which can vary between 50 and 100 m. It should be noted here that besides the in situ cosmogenic  $^{14}\text{C}$ , ice samples may also have significant amounts of trapped atmospheric  $^{14}\text{C}$ . The concentration of the latter can be estimated by measuring the amount of air present in individual samples, which is the procedure used in the previous studies cited above. Furthermore, the in situ  $^{14}\text{C}$  is partitioned between two phases: CO and CO<sub>2</sub>. Initially, when in situ  $^{14}\text{C}$  is produced, it is expected that most of it is in the CO phase [Lal *et al.*, 1987]. For previous work on  $^{14}\text{C}$  in ice, reference is made to Lal *et al.* [1990, 1997], Lal and Jull [1990, 1992], Wilson and Donahue [1992], Jull *et al.* [1994a], and van

Roijen *et al.* [1994]. The paper by Lal and Jull [1992] has an overview on cosmogenic nuclides in ice sheets.

The concentration of in situ cosmogenic  $^{14}\text{C}$  in ice is expected to be inversely proportional to the accumulation rates and directly proportional to the cosmogenic production rate of  $^{14}\text{C}$  at the surface of the ice field (as discussed in section 4). In the case of GISP 2 samples we found in situ  $^{14}\text{C}$  concentrations in the range of 1500–5000  $^{14}\text{C}$  atoms g<sup>-1</sup> ice. In the case of Vostok one would expect concentrations to be greater by a factor of ~15–20, considering the higher elevation and the lower accumulation rate, i.e., in situ  $^{14}\text{C}$  concentrations in excess of 20,000 atoms g<sup>-1</sup> ice. Considering the high-resolution temporal climatic record found in the Vostok ice core [Lorius *et al.*, 1993; Jouzel *et al.*, 1993; Raynaud *et al.*, 1993], the availability of the chronology and accumulation history of the Vostok ice [Jouzel *et al.*, 1993], and the expected high concentrations of in situ  $^{14}\text{C}$ , we decided to study the  $^{14}\text{C}$  record in Vostok. In this paper we present the results of our studies of several Vostok samples covering the period from 20 k.y. B.P. to the present. For completeness, we also studied several Holocene ice samples from Taylor Dome [Grootes and Steig, 1992; Steig *et al.*, 1998]. Our data show that the in situ  $^{14}\text{C}$  concentrations in both Vostok and Taylor Dome samples are much lower than was expected, in contrast to the observed  $^{14}\text{C}$  concentrations in GISP 2 samples, where theoretically expected amounts are observed. The concentrations in Taylor Dome are much lower than those in Vostok, as would be expected because of higher ice accumulation rates at this site. We consider the effects of wind ventilation on the accumulation of  $^{14}\text{C}$  in the firn layers in terms of simple models, along with its effect on the concentrations of the cosmogenic atmospheric isotopes,  $^{10}\text{Be}$  and  $^{36}\text{Cl}$ . By “wind ventilation,” we imply the movement of air through firn causing sublimation, transport and

<sup>1</sup>Scripps Institution of Oceanography, La Jolla, California.

<sup>2</sup>Arizona Accelerator Mass Spectrometer Facility, National Science Foundation, University of Arizona, Tucson, Arizona.

<sup>3</sup>Laboratoire des Sciences du Climat et de l'Environnement, Commissariat à l'Énergie Atomique, Saclay, Gif-sur-Yvette, France.

<sup>4</sup>Department of Earth and Environmental Science, University of Pennsylvania, Philadelphia, Pennsylvania.

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fragmentation of snow grains, etc. The stronger the "wind ventilation," the deeper its effects. These models allow us to make some estimates of how ventilation can change the concentrations of different isotopes. They make certain predictions on losses of in situ  $^{14}\text{C}$ , which can be verified by studies of cosmogenic atmospheric isotopes,  $^{10}\text{Be}$  and  $^{36}\text{Cl}$ .

## 2. Samples and Methods

Ice core samples analyzed were obtained from the Antarctic stations, Vostok (78° 28' S, 106° 48' E; elevation 3490 m) and Taylor Dome (77° 50' S, 158° 40' E; elevation 2400 m). Vostok ice samples were obtained from the BH5 and BH7 shallow cores drilled (dry) in January 1992 and 1996, respectively, and from the long core drilled during the 35th Soviet Antarctic Expedition (core sections numbered 35CA3 and 35CA7). The BH5 and BH7 core sections were removed from the site soon after drilling, but the 35CA3 and 35CA7 core sections were left at the site for quite some time before being removed to the storage site. For assignment of chronology and accumulation rates in the Taylor Dome ice core, based on  $^{18}\text{O}$  and  $\text{CO}_2$  records, reference is made to *Steig et al.* [1998] and *Indermuhle et al.* [2000].

The experimental procedures for the extraction of  $^{14}\text{C}$  activity from ice, separately in the two phases, CO and  $\text{CO}_2$ , has been described in detail earlier [*Lal et al.*, 1997]. Briefly, the outer layers of the ice sample (in the range 1.5-3 kg) are removed by scraping with a serrated stainless steel saw in a cold room; the sample is broken up into rectangular pieces of ~1-2 cm and placed in a large cooled flask (5-20 L volume, depending on the size of the ice sample) to which ~1-2 cm<sup>3</sup>  $\text{H}_3\text{PO}_4$  had been added and frozen (to ensure that after ice melted the resulting pH will be < 3). The flask is evacuated and  $^{14}\text{C}$ -free carrier gases of CO and  $\text{CO}_2$ , ~1 cm<sup>3</sup> STP each, are added. The flask is then warmed to allow the ice to melt, and the melt water equilibrated CO and  $\text{CO}_2$  carrier gases (using a magnetic stirrer) are then recovered sequentially through a vacuum line with liquid-nitrogen ( $\text{N}_2(\text{l})$ ) traps to remove  $\text{CO}_2$  and  $\text{N}_2(\text{l})$ -

cooled molecular sieve, Linde 5A, to trap air and CO. The air volumes are measured in each sample before converting CO to  $\text{CO}_2$  by passing the gas over hot CuO. (We note that the extremely small amounts of  $\text{CH}_4$ , expected as trapped in ice with air, would be included in the CO fraction. This would constitute < 0.3% of the  $^{14}\text{C}$  signal.)

In each case we performed a second extraction using the procedure for the first extraction. On the basis of our experience with nominally ~80 polar ice samples, we determined that except in the first 10 samples, our extraction efficiencies are generally better than 80% in the first extraction and that the average line blanks are 40 and 115 atoms  $^{14}\text{C}$  kg<sup>-1</sup> ice for CO and  $\text{CO}_2$  phases, respectively.

Carbonate dust is expected to be  $^{14}\text{C}$  free. *Smith et al.* [1997] estimated that the concentration of  $\text{CO}_2$  from this source might cause an apparent excess of  $\text{CO}_2$  of up to 90 ppm  $\text{CO}_2$  in the air trapped in ice. This does not significantly affect the amount of  $\text{CO}_2$  carrier.

## 3. Results

The results of our measurements are presented in Tables 1 and 2 for Vostok and Taylor Dome samples, respectively. The net  $^{14}\text{C}$  concentrations in ice are determined after removing the contributions from atmospheric  $^{14}\text{C}$  trapped with air, estimated on the basis of the amount of air in the individual ice samples (Table 2), atmospheric  $p\text{CO}_2$  values [*Smith et al.*, 1997], and the past  $^{14}\text{C}/^{12}\text{C}$  ratios in the atmosphere. The net in situ  $^{14}\text{C}$  concentrations, after correcting for the atmospheric  $\text{CO}_2$  contributed  $^{14}\text{C}$ , and  $^{14}\text{C}$  decay, are also given in Tables 1 and 2. The partitioning of in situ  $^{14}\text{C}$  in the CO and  $\text{CO}_2$  phases as given by the ratio of in situ  $^{14}\text{C}$  concentrations in the CO phase to the total in situ  $^{14}\text{C}$  concentrations is provided in the last columns in Tables 1 and 2.

In Tables 3 and 4 we compare the observed in situ  $^{14}\text{C}$  concentrations in Vostok and Taylor Dome ice samples, respectively, with those expected on the basis of estimated ice

**Table 1.** Measured  $^{14}\text{C}$  Concentrations in Vostok Ice Samples<sup>a</sup>

Lab. Code	Core Section	Dept h, m	Weight of Ice, kg	Age, years	Measured $^{14}\text{C}$ Conc., atoms		Age and Air-Corrected $^{14}\text{C}$		Total in situ $^{14}\text{C}$ Conc., <sup>b</sup> atoms g <sup>-1</sup>	$^{14}\text{CO}/(^{14}\text{CO}+^{14}\text{CO}_2)^c$
					CO	$\text{CO}_2$	CO	$\text{CO}_2$		
I001	BH5	7	1.53	5	>20±50	690±85	>25±50	685±85	>710±100	—
D3	BH5	10	2.50	190	825±50	4,040±100	845±150	2,310±90	3,160±100	0.37±0.03
I006	BH5	25	2.85	550	35±25	20±25	50±50	0±50	50±70	—
O11	BH5	56	4.40	1,450	80±10	90±35	100±15	80±60	180±60	0.56±0.21
I003	BH7	60	3.55	1,710	>20±50	560±30	25±50	590±35	615±60	0.04±0.01
I005	BH5	70	3.52	2,100	55±50	505±35	70±60	510±50	580±80	0.12±0.02
V112	BH5	112	2.50	3,820	2,785±80	1,710±60	4,420±130	1,910±95	6,330±160	0.70±0.04
L1	BH5	116	2.50	3,990	1,110±35	2,450±70	1,800±60	3,110±120	4,410±130	0.37±0.02
L8	35CA7	211	2.47	8,270	255±25	880±35	690±65	1,900±95	2,590±115	0.27±0.03
O16	35CA7	266	5.38	10,78	405±35	135±35	1,490±125	495±130	1,985±180	0.75±0.09
I002	35CA3	267	5.03	10,90	465±35	960±20	1,735±130	3,085±70	4,820±150	0.36±0.03
I004	35CA3	297	4.37	14,00	375±35	1,230±30	2,040±190	4,420±145	6,460±240	0.32±0.03
L7	35CA3	333	2.00	14,56	160±25	550±45	945±150	2,780±310	3,720±310	0.25±0.05
I007	35CA3	396	8.50	19,50	480±30	20±30	5,070±105	0±130	5,070±170	1.0±0.05

<sup>a</sup>Lab., laboratory; Conc., concentration.

<sup>b</sup>Corrected for trapped  $^{14}\text{C}$  and decay of  $^{14}\text{C}$ .

<sup>c</sup>Refers to age-corrected in situ  $^{14}\text{C}$  concentrations.

**Table 2.** Measured <sup>14</sup>C Concentrations in Taylor Dome Ice Samples

Lab. Code	Depth, m	Weight of Ice, kg	Age, years	Measured <sup>14</sup> C Conc., atoms g <sup>-1</sup>		In Situ <sup>14</sup> C Conc., <sup>a</sup> atoms g <sup>-1</sup>		Total in Situ <sup>14</sup> C Conc., <sup>a</sup> atoms g <sup>-1</sup>	<sup>14</sup> CO/ ( <sup>14</sup> CO+ <sup>14</sup> CO <sub>2</sub> )
				CO	CO <sub>2</sub>	CO	CO <sub>2</sub>		
O7	1.5–1.89	5.95	10	100±10	320±25	100±15	300±35	400±40	0.25±0.05
O10	4.0–4.3	5.80	80	65±10	45±25	65±20	40±35	105±40	0.74±0.43
L6	30–31	3.20	520	2,300±50	15±45	2,445±50	10±50	2,455±70	1.0±0.04
L4	54–55	3.65	970	90±25	10±55	120±30	90±65	210±70	0.58±0.23
L3	84–85	3.00	1,650	240±10	795±45	295±10	260±55	555±55	0.53±0.06
O19	251–252	3.95	5,530	105±25	630±55	205±50	525±100	730±115	0.28±0.08
O20	200–201	2.41	4,220	5±60	1,130±35	0±90	1,180±55	1,180±110	<0.15 (2 s.d)
O22	327–328	3.51	8,330	75±30	600±60	205±80	975±160	1,180±180	0.17±0.07
O24	355–356	2.68	10,688	135±25	585±50	495±60	1,410±155	1,905±165	0.26±0.04

<sup>a</sup>Corrected for trapped <sup>14</sup>C and decay of <sup>14</sup>C.

<sup>b</sup>Refers to age-corrected in situ <sup>14</sup>C concentrations.

accumulation rates for Vostok [Jouzel *et al.*, 1993] and Taylor Dome [Steig *et al.*, 1998]. These are based on the estimated values for the in situ <sup>14</sup>C production rates at the surface at Vostok and Taylor Dome, of 234 and 120.3 atoms <sup>14</sup>C g<sup>-1</sup> ice, respectively [see Lal *et al.*, 1997] (see section 4). It becomes apparent from Tables 3 and 4 that the measured in situ <sup>14</sup>C concentrations are lower than expectations by factors ranging from 1.5 to 55. Vostok concentrations are generally higher than that in Taylor Dome samples, as would be expected also because of the higher accumulation rates at Taylor Dome. However, we also note that the deficiency factors, defined as the ratio of expected to observed in situ <sup>14</sup>C concentrations, are generally higher for Taylor Dome than they are for Vostok. It seems likely that the higher deficiency factors at Taylor Dome are related to higher temperatures at Taylor Dome.

It should be noted here that the in situ <sup>14</sup>C concentrations reported in Tables 1 and 2 include some contributions from secondary in situ produced <sup>14</sup>C after the ice core was extracted. Reference is made to Lal *et al.* [1997] for the procedure used for correcting for the secondary production. These contributions

are very small in the case of the Vostok BH5 core section and for all of the Taylor Dome samples. For the Vostok 35CA3 core section, secondary contributions could be significant, in the range of ~50%, but cannot be estimated accurately, because of the present lack of information on how the samples were stored at the extraction site. The results on the deficiency factors for the 35CA3 core section samples should therefore be considered as lower limits.

In section 4 we discuss some simple models for quantifying processes for losses of implanted in situ produced <sup>14</sup>C in ice during the accumulation process due to wind ventilation. As stated earlier, by “wind ventilation”, we imply movement of air through firn causing sublimation, transport and fragmentation of snow grains, etc.

#### 4. Interpretation of Data

The oversimplified basic equations governing the in situ production of <sup>14</sup>C, without considering firnification effects, are [Lal *et al.*, 1987]

**Table 3.** Observed <sup>14</sup>C Concentration Deficiencies in Vostok Ice Samples

Lab. Code	Depth, m	Age, years	Total in Situ <sup>14</sup> C Conc., <sup>a</sup> atoms g <sup>-1</sup>	Accumulation Rate, cm yr <sup>-1</sup>	Expected <sup>14</sup> C Conc., <sup>b</sup> atoms g <sup>-1</sup>	Deficiency Factor
I001	7	5	710±100	2.15	9,760	14±2
D3	10	190	3,160±100	2.14	9,810	3.1±0.10
I006	25	550	50±70	2.13	9,855	—
O11	56	1,450	180±60	2.12	9,900	55±20
I003	60	1,710	615±60	2.12	9,900	16±2
I005	70	2,100	580±80	2.12	9,900	17±2
V112	112	3,820	6,330±160	2.22	9,455	1.5±0.04
L1	116	3,990	4,410±130	2.20	9,540	2.2±0.06
L8	211	8,270	2,590±115	2.26	9,290	3.6±0.16
O16	266	10,785	1,985±180	1.80	11,660	5.9±0.54
I002	267	10,900	4,820±150	1.80	11,660	2.4±0.08
I004	297	14,000	6,460±240	1.80	11,660	1.8±0.07
L7	333	14,568	3,720±310	1.57	13,370	3.6±0.30
I007	396	19,500	5,070±170	1.30	16,150	3.2±0.11

<sup>a</sup>Age-corrected in situ <sup>14</sup>C concentration.

<sup>b</sup>Based on <sup>14</sup>C production rate at surface of 234 atoms <sup>14</sup>C g<sup>-1</sup> ice yr<sup>-1</sup>.

**Table 4.** Observed  $^{14}\text{C}$  Concentration Deficiencies in Taylor Dome Ice Samples

Lab. Code	Depth, m	Age, years	Total in Situ $^{14}\text{C}$ Conc., <sup>a</sup> atoms $\text{g}^{-1}$	Accumulation Rate, $\text{cm yr}^{-1}$	Expected $^{14}\text{C}$ Conc., <sup>b</sup> atoms $\text{g}^{-1}$	Deficiency Factor
O7	1.5–1.89	10	400±40	5.35	3,925	9.8±1.0
O10	4.0–4.3	80	105±40	5.70	3,680	35±13
L6	30–31	520	2,455±70	5.80	3,620	1.5±0.04
L4	54–55	970	210±70	4.93	4,260	20±7
L3	84–85	1,650	555±55	5.55	3,780	6.8±0.7
O19	251–252	5,530	730±115	5.00	4,200	5.8±1.0
O20	200–201	4,220	1,180±110	4.80	4,375	3.7±0.35
O22	327–328	8,330	1,180±180	1.48	14,180	12±1.8
O24	355–356	10,688	1,905±165	0.835	25,140	13±1.0

<sup>a</sup>Age-corrected in situ  $^{14}\text{C}$  concentration.<sup>b</sup>Based on  $^{14}\text{C}$  production rate at surface of 120.3 atoms  $^{14}\text{C}$   $\text{g}^{-1}$  ice  $\text{yr}^{-1}$ .

$$\frac{dC}{dt} = -C\lambda + \varepsilon Q_0 e^{-\mu\rho z}, \quad (1)$$

$$\frac{dz}{dt} = s(t), \quad (2)$$

where  $C(z,t)$  is the  $^{14}\text{C}$  concentration in accumulating ice of age  $t$  at depth  $z$  (ice density equivalent depth),  $t=0$  and  $z=0$  at the surface,  $\rho$  is the mean ice density,  $\lambda$  is the disintegration constant of the radionuclide  $^{14}\text{C}$  ( $1.2094 \times 10^{-4} \text{ yr}^{-1}$ ), whose production rate is  $Q_0$  (atoms  $\text{g}^{-1} \text{ sec}^{-1}$ ) at the surface, and  $s(t)$  is the ice density equivalent accumulation rate. The fraction of in situ  $^{14}\text{C}$  retained in the firm,  $\varepsilon$ , is assumed to be constant with depth in (1). The parameter  $\mu$  is the inverse of the absorption mean free path for the nucleonic component of the cosmic radiation in the ice,  $\sim 160 \text{ g cm}^{-2}$ . For the case of constant accumulation rate, i.e.,  $s(t) = \text{const}$ , and  $\varepsilon = 1$ , a transition occurs in the in situ  $^{14}\text{C}$  concentrations at a depth of  $\sim 1.5\text{--}2 \text{ m}$ . At greater depths below the maximum the in situ  $^{14}\text{C}$  concentration rapidly reaches a value,  $C(t)$  in an ice layer of age  $t$  [Lal and Jull, 1990; Lal et al., 1997]:

$$C(z,t) = \frac{\varepsilon Q_0}{s(t)\mu\rho} e^{-\lambda t}, \quad (3)$$

where  $s(t)$  is the average ice accumulation rate between  $t$  and  $t - \Delta t$ ;  $\Delta t$  is the time required to accumulate a thickness of  $\sim 5 \text{ m}$  ice equivalent, by which time essentially all the in situ  $^{14}\text{C}$  is produced in an ice layer as it descends to depths owing to fresh accumulation at the surface.

The cosmic ray production rate of  $^{14}\text{C}$  due to spallation of oxygen nuclei and due to neutron capture by nitrogen present in ice has been estimated [Jull et al., 1994b; Lal and Jull, 1994]. The principal contribution is due to the former, which can be determined since in situ  $^{14}\text{C}$  production rate in quartz has been measured [Lal and Jull, 1994; Jull et al., 1994b]. At the present altitude of Vostok and Taylor Dome the  $^{14}\text{C}$  production rates in ice exposed at the surface are estimated to be  $234 \pm 25$  and  $120 \pm 15$  atoms  $^{14}\text{C}$   $\text{g}^{-1}$  ice  $\text{yr}^{-1}$ , respectively.

An important note here is that in situ  $^{14}\text{C}$  concentrations are a sensitive function of the rate of accumulation of ice above the layer under consideration (see equation (1)). For the simplest case of a layer-by-layer accumulation (i.e., no mixing in the upper layers before consolidation and no mass loss by ventilation or erosion), the  $^{14}\text{C}$  buildup in a test layer deposited at  $t=0$  is controlled by the rate at which the firm

accumulates over this layer; see the results of the simulation by Lal et al. [2000]. The “accumulation control” on  $^{14}\text{C}$  buildup decreases as the thickness of the overlying layer increases in absolute magnitude: exponentially, as  $\exp[-\mu\rho z(t)]$ , where  $z(t)$  is the accumulated equivalent ice thickness at time  $t$ .

#### 4.1. Simplified Firnification Models

We interpret the observed  $^{14}\text{C}$  deficiencies in Antarctic ice samples as being due to grain metamorphism due to recrystallization and sublimation/evaporation caused by wind ventilation in the early stages of firn accumulation [cf. Waddington et al., 1993, 1996; Waddington, 1996]. Most of the in situ  $^{14}\text{C}$  is, in fact, produced in the firn during its accumulations to thickness of up to  $\sim 10 \text{ m}$ . The firnification processes are expected to become more significant in low accumulation rate situations. In the case of the Vostok and Taylor Dome samples we are dealing with accumulation rates in the range of 1–2 and 1–5  $\text{cm yr}^{-1}$ , respectively, in contrast to an order of magnitude higher accumulation rates of 10–30  $\text{cm yr}^{-1}$  in the case of the GISP 2 samples studied. For purposes of illustration we first discuss below two interesting and probably realistic continuous accumulation models for cosmogenic  $^{14}\text{C}$  and  $^{10}\text{Be}$  (note that the former is implanted in the ice as it accumulates and that the latter is present in the accumulating firn, owing to scavenging of atmospheric  $^{10}\text{Be}$  during condensation of vapor in the atmosphere): (1) a mass loss occurs from accumulating firn layers, owing to sublimation and/or erosion by wind ventilation, and (2) no net mass loss occurs during recrystallization and also during sublimation/evaporation losses, owing to the redeposition of vapor within the layer.

Under these conditions the expected behaviors of  $^{14}\text{C}$  and  $^{10}\text{Be}$  are displayed in Table 5. In situ  $^{14}\text{C}$  is, of course, irreversibly lost during mass loss; in the above we assumed that  $^{10}\text{Be}$  remains adhering to the residual ice grain during mass loss processes, since it is particle-active.

The differential equations governing the growth of ice sheet and nuclide concentration for cases 1 and 2 are given below. They differ from the oversimplified equations (1) and (2), where firnification effects were not considered. Since firn density  $\rho$  is depth dependent, we express depth as  $(\rho z)$ , in units of mass  $\text{cm}^{-2}$ , which is an explicit function of time  $t$ . In the following,  $s$  is the accumulation rate of firn of density  $\rho$ .

**Table 5.** Expected Behaviors of Cosmogenic in Situ <sup>14</sup>C and Atmospheric <sup>10</sup>Be in Hypothetical Accumulation Processes

	In situ <sup>14</sup> C		Atmospheric <sup>10</sup> Be	
	Concentration, atoms g <sup>-1</sup>	Total Number of Atoms in a Layer	Concentration, atoms g <sup>-1</sup>	Total Number of Atoms in a Layer
Sublimation or evaporation with mass loss from the layer	no change	decreases	increases <sup>a</sup>	no change
Recrystallization or evaporation and redeposition of vapor within the layer, with no mass loss	decreases	decreases	no change	no change

<sup>a</sup>In the case of grain erosion/fragmentation by ventilation, the <sup>10</sup>Be concentration would remain unchanged if the fine eroded material is blown away from the accumulation site.

Case 1: Mass loss

$$\frac{dC_{14}}{dt} = -\lambda_{14} C_{14} + Q_0 e^{-\mu \rho z}, \quad (4)$$

$$\frac{d(\rho z)}{dt} = \rho s - \int_0^{\rho z} f(z) d(\rho z), \quad (5)$$

$$\frac{dC_{10}}{dt} = -\lambda_{10} C_{10} + C_{10} f(z). \quad (6)$$

Case 2: No mass loss

$$\frac{dC_{14}}{dt} = -\lambda_{14} C_{14} - C_{14} f(z) + Q_0 e^{-\mu \rho z}, \quad (7)$$

$$\frac{d(\rho z)}{dt} = \rho s, \quad (8)$$

$$\frac{dC_{10}}{dt} = -\lambda_{10} C_{10}. \quad (9)$$

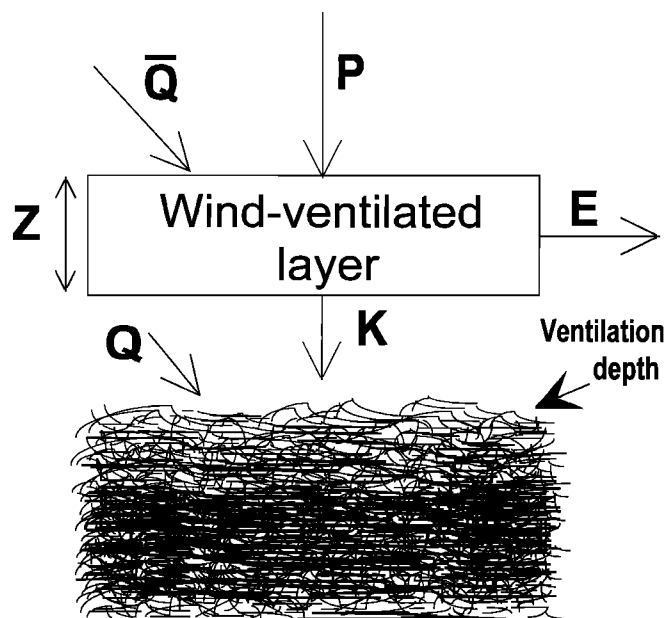
The subscripts 10 and 14 on  $C$  (nuclide concentration) and  $\lambda$  (nuclide disintegration constant) refer to <sup>10</sup>Be and <sup>14</sup>C, respectively. Other symbols,  $Q_0$ ,  $\mu$ , and  $s$ , are as defined earlier. The fractional mass loss rate in case 1 is assumed to be given by the function  $f(z)$ . In case 2, mass loss and mass gained are both of the form  $f(z)$ , i.e., no net mass loss from a layer. Consequently, this function does not appear in the differential equation for  $z$ .

We summarize here the results of a few calculations based on (4)–(9), purely as illustration. We assumed the functional form of  $f(z)$  to be  $K \exp(-\alpha z)$ . For  $s = 10 \text{ cm yr}^{-1}$ ,  $K = 0.1$ , and  $\alpha = 1/50 \text{ cm}$ , the surface accumulation rates level to  $\sim 5 \text{ cm yr}^{-1}$  in the case of mass loss from the layers (case 1), within 2 m depth. In this depth interval the <sup>10</sup>Be concentrations rise to 2 times the value in precipitation. In the case of in situ <sup>14</sup>C, for case 2 (no mass loss), the <sup>14</sup>C concentrations decrease by more than 10% compared to the value for  $s = 10 \text{ cm yr}^{-1}$ , with no ventilation processes. In the case of mass loss (case 1) the in situ <sup>14</sup>C concentrations in fact increase by 68% over the no-effect case, owing to increased cosmic ray implantation (since the effective  $s$  value is reduced).

The model calculations show large effects on <sup>14</sup>C and <sup>10</sup>Be concentrations. The interesting feature is that in some cases the firnification models produce opposite effects in the <sup>14</sup>C and <sup>10</sup>Be concentrations, as seen from Table 5. Therefore simultaneous data on <sup>14</sup>C and <sup>10</sup>Be concentrations in the same ice samples would lead us to realistic firnification models, we hope.

## 4.2. Wind-Ventilated Surface Layer Model

For the present, i.e., with data available only for in situ <sup>14</sup>C concentrations in individual ice samples, we proceed to obtain an idea of the magnitude of firnification processes by invoking a constant thickness surface layer, of effective thickness  $Z$ , in which wind ventilation processes are effective (Figure 1). Note that this approach differs radically from that discussed above in cases 1 and 2, where we tacitly assumed a progressive depth-dependent firnification. In the “wind-ventilated surface layer” model (Figure 1), as accumulation continues, a discrete layer is buried below the surface layer. This constitutes the “undisturbed layer” (Figure 1), below a fixed ventilation depth, where continued accumulation and in situ <sup>14</sup>C production occur, without any grain metamorphism. In the model equations we assume that the fraction of in situ <sup>14</sup>C surviving in the ventilated layer is  $f$ ; the rest is lost during grain metamorphism.



**Figure 1.** A schematic of a simplified model of snow accumulation. Precipitation,  $P$ , continuously adds fresh ice to the “wind-ventilated surface layer,” which is considered well mixed to a depth  $Z$  cm (water equivalent). In situ cosmogenic production of <sup>14</sup>C occurs in this layer, as well as in the lower “undisturbed layer.” Mass losses from the turbulent layer are represented by the first-order rate constant  $E$ , while  $K$  similarly represents ice accumulation in the undisturbed layer.

For a constant precipitation rate,  $P$  ( $\text{g cm}^{-2} \text{yr}^{-1}$ ), thickness of turbulent layer  $Z$  (centimeters of water equivalent thickness), and first-order rate constants  $K$  and  $E$ , which contribute to ice accumulation  $s$  ( $\text{g cm}^{-2} \text{yr}^{-1}$ ) and attrition/erosion  $\varepsilon$  ( $\text{g cm}^{-2} \text{yr}^{-1}$ ), respectively, we obtain the following equations at steady state for the in situ <sup>14</sup>C concentrations  $C$ . The value of  $C$  in the undisturbed layer refers to modern ice at depths of, say, 10 m below the surface where in situ production of <sup>14</sup>C and also decay of accumulated <sup>14</sup>C are not significant. For the ventilated layer,

$$C = \frac{f \bar{Q}}{\lambda + P(\bar{\rho}z)^{-1}} \quad (10)$$

For the undisturbed layer,

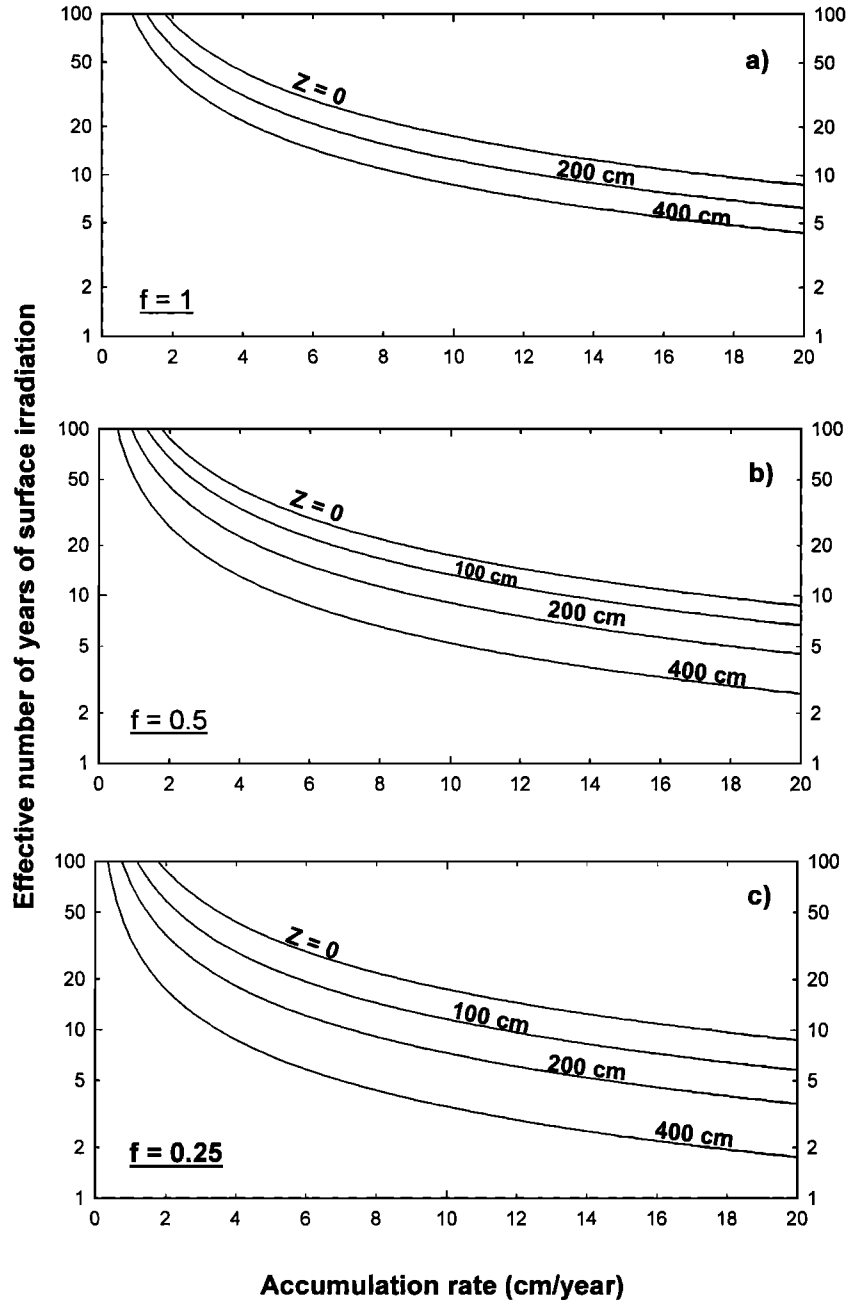
$$C = \frac{f \bar{Q}}{\lambda + P(\bar{\rho}z)^{-1}} + \frac{Q_0 e^{-\bar{\rho}\mu z}}{\bar{\rho}\mu s - \lambda} \quad (11)$$

with

$$\bar{Q} = \frac{Q_0(1 - e^{-\bar{\rho}\mu z})}{\bar{\rho}\mu z} \quad (12)$$

$$P = s + \varepsilon = \bar{\rho}z(K + E) \quad (13)$$

where  $\bar{\rho}$  is the average density of firm in the ventilated layer and  $\rho$  is the density of ice in the undisturbed layer. The concentration of in situ <sup>14</sup>C in the “ventilated layer” is only a



**Figure 2.** Results of the calculations for the effective number of years of implantation of cosmogenic <sup>14</sup>C in the upper layers of the “undisturbed layer” for the accumulation model depicted in Figure 1, based on equations (10) - (13) for the case  $P = s$ : (a)  $f = 1$ , (b)  $f = 0.5$ , and (c)  $f = 0.25$  (see text for the model and the parameters).

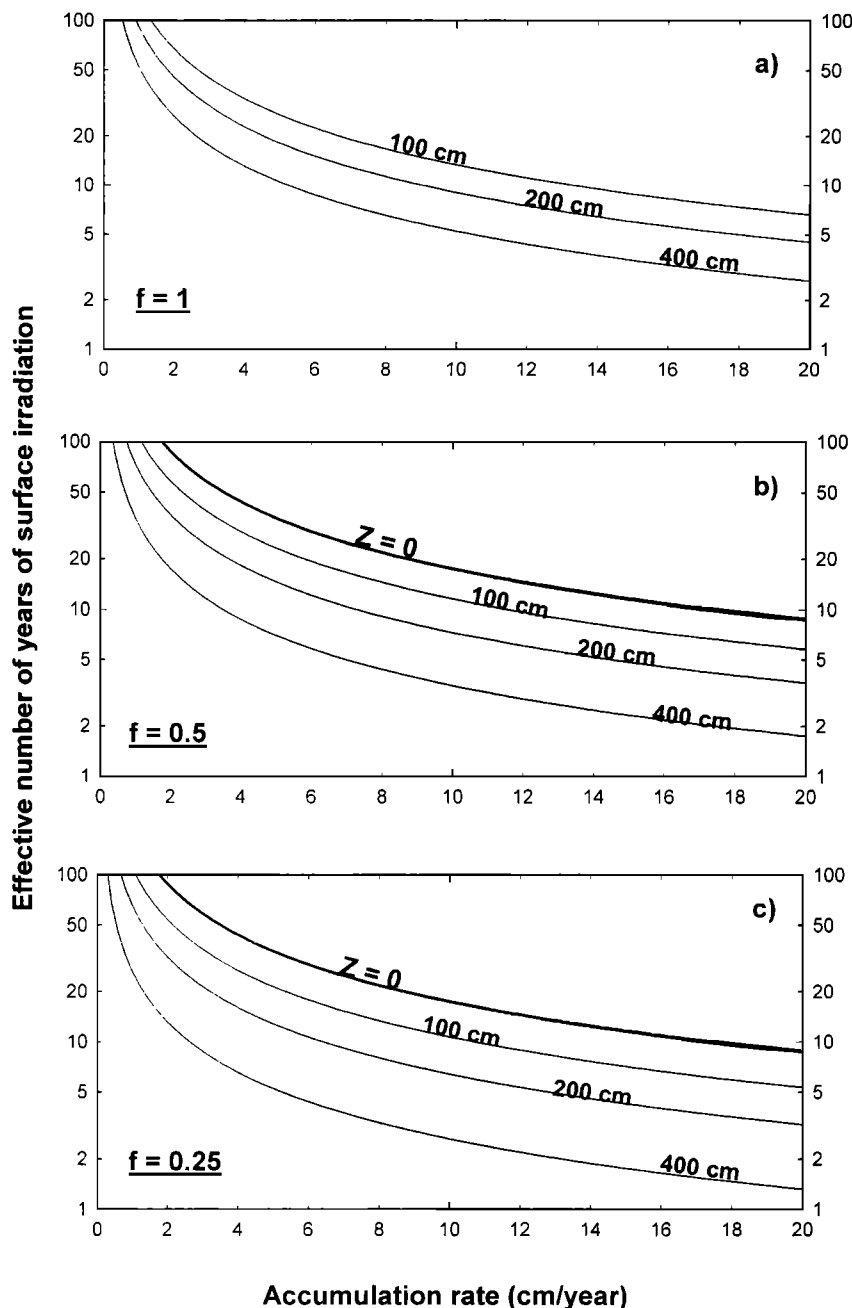


Figure 3. (a-c) Same as in Figure 2, but for the case  $P = 2s$ .

function of  $P$ , which is partitioned between  $s$  and  $\epsilon$ , according to (13). The in situ  $^{14}\text{C}$  concentration of the “undisturbed layer” is a sum of the steady state concentration in the turbulent layer and the depth-dependent accumulation of in situ  $^{14}\text{C}$  as the ice layer accumulates. The latter is obviously a function of the thickness of the turbulent layer  $Z$  and the accumulation rate of the undisturbed layer.

In Figures 2 and 3 we have plotted the effective number of years of surface irradiation in recent ice in the undisturbed layer for  $Z = 100, 200,$  and  $400$  cm (water equivalent depth) and  $f$  values of 1, 0.5, and 0.25; Figure 2 is for the case  $P = s$ , while Figure 3 is for the case  $P = 2s$ . These are based on calculated values of  $^{14}\text{C}$  concentrations,  $C$  (11) with  $Q_0 = 1$   $^{14}\text{C}$  atom  $\text{g}^{-1}$  ice  $\text{yr}^{-1}$ . (Note that the  $Z = 0$  curves in Figures 2 and 3

correspond to the case of absence of the ventilated layer.) It can be seen from Figures 2 and 3 that compared to the case of continued implantation of in situ  $^{14}\text{C}$ , layer by layer, with no losses due to firnification processes (as shown by the dark lines labeled  $Z = 0$  in Figures 2 and 3), the in situ  $^{14}\text{C}$  concentrations are appreciably reduced for the cases considered. In situ  $^{14}\text{C}$  concentration deficiencies are higher for smaller  $f$  values and for the  $P = 2s$  case, compared to the case of  $P = s$ , i.e., no losses from the layer due to erosion or sublimation.

## 5. Discussion and Conclusions

In Vostok and Taylor Dome samples we observe a wide range of deficiency factors for in situ produced  $^{14}\text{C}$ . *Wagon et*



*al.* [1999] observed similar effects in their analyses of volatile acid species in upper firm layers ice samples from Vostok. It is, however, not possible to make a detailed comparison between the observations of *Wagnon et al.* [1999] and our in situ  $^{14}\text{C}$  concentration data since in their case, one would have to also consider the impact of some chemical reactions due to high sulfuric acid levels of volcanic origin.

Some of the Vostok ice samples show a marked lowering of in situ  $^{14}\text{C}$  concentrations with deficiency factors of 1.5-4 (Table 2), which account for 60% of the samples. Model calculations (as summarized in Figures 2 and 3 for some chosen parameters) essentially point to a thickness of the wind-ventilated layer of  $\sim 2$  m and in situ  $^{14}\text{C}$  concentration losses by firnification processes of  $\sim 50\%$ , i.e.,  $f = 0.5$ . Higher losses of in situ  $^{14}\text{C}$  concentrations can be explained only by larger effective  $Z$  values and a higher degree of grain metamorphism ( $f \ll 0.5$ ). Such parametric values would be necessary to explain the higher deficiency factor ( $> 4$ ) samples in Tables 2 and 3. In particular, we note that deficiency factors are higher for the Taylor Dome samples and indicate a higher degree of grain metamorphism compared to Vostok samples.

If losses of in situ  $^{14}\text{C}$  concentrations resulted from sublimation and mass losses from the turbulent layer, one would expect higher  $^{10}\text{Be}$  concentrations in ice accumulating in the undisturbed layer, as was discussed earlier (see section 4.1 and Table 5). In the absence of any  $^{10}\text{Be}$  concentration results in our samples, we may, however, attempt to see if any information can be derived from its reported concentrations in polar ice samples.  $^{10}\text{Be}$  concentrations have been reported for Vostok [*Yiou et al.*, 1985; *Raisbeck et al.*, 1992]. The estimated  $^{10}\text{Be}$  fluxes during the past 20,000 years are in the range of  $1.5\text{--}2.5 \times 10^5$  atoms  $\text{cm}^{-2} \text{yr}^{-1}$ . The corresponding flux estimate at the Taylor Dome drill site is  $1.5 \times 10^5$  atoms  $\text{cm}^{-2} \text{yr}^{-1}$ , which is a good representative average for all the samples studied at the Taylor Dome site [*Steig and Suiver*, 1995; *Steig et al.*, 1996]. In contrast, the  $^{10}\text{Be}$  fluxes for the past 20,000 years at the Summit (Greenland) are close to  $4 \times 10^5$  atoms  $\text{cm}^{-2} \text{yr}^{-1}$  [*Yiou et al.*, 1997], greater than Vostok and Taylor Dome values by a factor of  $\sim 2$ . The only higher value reported [*Aldahan et al.*, 1998] for an Antarctic station is  $4.2 \pm 0.3 \times 10^5$  atoms  $\text{cm}^{-2} \text{yr}^{-1}$  for Dronning Maud Land ( $73^\circ 36' \text{ S}$ ,  $12^\circ 26' \text{ W}$ ), which is comparable to the Summit (Greenland)  $^{10}\text{Be}$  flux. The mean annual accumulation rate at this site is 32 cm water equivalent. These data do suggest the importance of both accumulation rate and the latitude in regulating the  $^{10}\text{Be}$  concentrations and fluxes.

On the basis of studies of  $^{14}\text{C}$  in the Summit samples, we know that firnification effects are minimal in these samples back to 17 k.y. B.P. since the observed agree well with the expected values [*Lal et al.*, 1997, 2000]. On the other hand, we note large in situ  $^{14}\text{C}$  concentration deficiencies in the Vostok and Taylor Dome samples. If these resulted from mass losses from ice accumulates during grain metamorphism,  $^{10}\text{Be}$  concentrations in ice would be enhanced. On the other hand, the rate of ice accumulation would be reduced correspondingly. Therefore it seems plausible that in the process of treating accumulation as a black box, one may still obtain the approximately correct value of the  $^{10}\text{Be}$  flux. It should be noted here that *Steig et al.* [1996] observed a large amplitude in the variation of  $^{10}\text{Be}$  concentrations at Taylor Dome on 11-year solar cycle timescales. These variations presumably owe themselves to firnification processes. However, it is difficult at present to estimate the magnitude of the effect in individual

samples. The measured 2 times higher  $^{10}\text{Be}$  flux at the Summit compared to Vostok and Taylor Dome may imply that besides sublimation, grain attrition with loss of  $^{10}\text{Be}$  sticking to ice grains may also be important at Vostok and Taylor Dome.

We see that we get into interesting situations if we assume that (1) the  $^{10}\text{Be}$  flux,  $F_{10}$ , is given by the commonly used relationship,  $F_{10} = C_{10} s$  and (2) that the  $^{10}\text{Be}$  flux is the same in the Arctic and Antarctic sites. It is not easy to resolve the questions arising from these assumptions at this time, because we do not have a better recourse to estimate  $^{10}\text{Be}$  fluxes. We do not yet know the stratospheric contributions of  $^{10}\text{Be}$  production in the polar sites. Also, we do not know if all the  $^{10}\text{Be}$  produced in the polar troposphere is precipitated within the polar regions. For a detailed discussion of related problems, based on cosmic ray production systematics and a numerical simulation using the observed data on the geographical distribution of fallout of nuclear weapon-released  $^{90}\text{Sr}$ , reference is made to *Lal* [1987], who has discussed the possibility that the fallout of  $^{10}\text{Be}$  in the polar regions specially may be sensitive also to meteorological factors. It is interesting to note that the estimated  $^{10}\text{Be}$  flux at Vostok and Taylor Dome is smaller than even its tropospheric production by  $\sim 50\%$ .

More definitive conclusions can be reached when simultaneous measurements of cosmogenic in situ  $^{14}\text{C}$  and atmospheric  $^{10}\text{Be}$  and  $^{36}\text{Cl}$  are available in the same ice samples. From such studies one should be able to improve the algorithms for estimating cosmogenic nuclide fluxes in the polar regions, which are presumably uncertain today by factors of 2 or more. It would be a significant result of such studies if one can explicitly determine the  $P$ - $s$  relationships, which we have seen do play an important role in determining the fluxes of properties scavenged by ice.

In conclusion, we note that every tracer occupies a special niche in the geochemical domain, making it uniquely suitable for the study of certain specific processes. In this paper the complementary roles of the in situ cosmogenic  $^{14}\text{C}$  and the cosmogenic atmospheric  $^{10}\text{Be}$  are quite manifest. The other cosmogenic atmospheric nuclide,  $^{36}\text{Cl}$ , should also provide useful information considering its distinctly different chemical behavior.

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- G. S. Burr, D. J. Donahue, and A. J. T. Jull, Arizona Accelerator Mass Spectrometer Facility, National Science Foundation, University of Arizona, Tucson, AZ 85721.
- B. Deck and D. Lal, Geosciences Research Division, Scripps Institution of Oceanography, La Jolla, CA 92093-0244. (dlal@ucsd.edu)
- J. Jouzel, Laboratoire des Sciences du Climat et de l'Environnement, CEA Saclay, 91191 Gif-sur-Yvette Cedex, France.
- E. Steig, Department of Earth and Environmental Science, University of Pennsylvania, 240 South 33rd St., Philadelphia, PA 19104-6316.

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