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Recycling of cosmogenic nuclides after their removal from the atmosphere; special case of appreciable transport of ^{10}Be to polar regions by aeolian dust

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Abstract

In this paper we discuss the general problem of recirculation of atmospheric cosmogenic isotopes after their initial removal from the atmosphere. Practically all nuclides are recycled after their removal from the atmosphere. The noble gas radio-nuclides re-circulate between the atmosphere and the hydrosphere; the fraction of time spent in these reservoirs depends on their solubility and half-lives. Radiocarbon atoms spend most of their life-time in the oceans before being removed to the sediments. All cosmogenic nuclides which are removed from the atmosphere by wet precipitations spend most of their life-time in the hydrosphere or in sediments. A special exception arises in the case of nuclides such as ^{10}Be and ^{26}Al, which are particle-active and adhere to aerosols and top soils in the lithosphere. Strong winds dislodge surface particles and transport the smaller particles to great distances.

We discuss the special case of the transport of particle reactive ^{10}Be via aeolian dust, which can result in appreciable ^{10}Be fluxes in polar regions in cold climates, vis-à-vis its direct deposition from the atmosphere. We compare estimated concentrations of dust transported ^{10}Be with the measured *total* ^{10}Be concentrations in Antarctica in the Vostok and Taylor Dome ice cores, and *atmospheric* ^{10}Be concentrations in Greenland in the GISP and GRIP ice cores. During certain epochs, high aeolian dust concentrations contribute significantly to ^{10}Be fluxes in Greenland, comparable to or greater than atmospheric ^{10}Be fluxes. In the Antarctica, however, even during epochs of high dust fluxes, the aeolian ^{10}Be contributions are $<10\%$ of the atmospheric ^{10}Be fluxes. These results imply that great care should be taken in analyzing ^{10}Be concentrations in Greenland especially!

Keywords: cosmic ray produced isotopes; cosmogenic ^{10}Be in polar ice; re-deposition of particle active nuclides; dust transported ^{10}Be in Greenland and Antarctica

1. Introduction

Cosmic ray interactions with the atmospheric nuclei produce a stable cosmogenic nuclide, ^{3}He and 16 radioactive nuclides of half-lives exceeding 2 weeks, which have been utilized as tracers for studying geophysical and geochemical processes (Lal, 2002). The circulation of cosmogenic nuclides depends on their chemical nature. The cosmogenic nuclides fall in three main categories (Lal, 2002): i) noble gases which do not form compounds; ii) those which form compounds with constituent molecules in the atmosphere and the hydrosphere, and iii) those which

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form compounds but being particle active, also attach to aerosols/particles. As we shall see, it is the circulation of nuclides in the third category that gives rise to appreciable recirculation that is important geophysical and geochemically.

1.1. Noble gas nuclides

Radio-nuclides belonging to the first category, ^{3}He, ^{37}Ar, ^{39}Ar and ^{81}Kr behave as conservative tracers in fresh waters and in oceans. However depending on their half-lives, they re-circulate within the atmosphere and the hydrosphere before they decay. The mean residence times of the noble gases He, Ar and Kr in the atmosphere, as dictated by their solubility, are 1600, 400 and 250 y respectively (Krishnaswami and Lal, 1973). ^{3}He also re-circulates; its removal from the atmosphere is regulated by its much longer time of escape from the atmosphere with the polar wind (Axford, 1968; Banks and Holzer, 1960), and from the exosphere (Macdonald, 1963).

1.2. Chemically active nuclides forming compounds in the atmosphere/hydrosphere

Radio-nuclides belonging to the second category, ^{3}H, ^{7,10}Be, ^{14}C, ^{26}Al, ^{32}Si and ^{32,33}P, form compounds in the atmosphere as well as in hydrosphere. Amongst these nuclides, only ^{14}C re-circulates between the atmosphere, ocean surface waters and the biosphere by molecular exchange. Others, except for the particle active nuclides, ^{7,10}Be and ^{26}Al (discussed below), remain largely confined to the hydrosphere and sediments, after their removal from the atmosphere by wet precipitations.

Except for ^{3}H, the mobilization of these nuclides in authigenic sediments and on to particles on which they (^{7,10}Be and ^{26}Al) are attached, removes them from the hydrosphere. Note however that as a result of strong near surface currents, their fall-out patterns on to sediments usually differ significantly from the pattern in which they are removed from the atmosphere to the hydrosphere. A further modification in their removal pattern to the sediments may occur near the sediment–water interface by turbidity currents. A number of studies based on excess ^{230}Th have shown that extensive sediment focusing occurs near the sediment water interface which increases the apparent deposition rate of properties in the sediments (cf. Marcantonio et al., 1996).

1.3. Nuclides which form compounds and attach to aerosols

Beryllium and aluminum isotopes ^{7,10}Be and ^{26}Al form oxides in the atmosphere. These oxides show high particle activity. The distribution coefficient, defined as the ratio of ^{10}Be amounts in particulate and dissolved phases, have been reported to be in the range of 10^{5}–10^{6} at pH of 6–7, based on laboratory experiments with ^{7}Be (Nyffler et al., 1984; You et al., 1989; Aldahan et al., 1999). These nuclides get attached at first as oxides to aerosols in the atmosphere. Depending on the pH of the water in the hydrosphere, a part of the oxides of these nuclides may dissolve in water at the time of their removal from the atmosphere by wet precipitations.. The concentrations of ^{10}Be are found to be in the range of 10^{8}–10^{10} atoms/g, in aerosols sampled in the troposphere (see Section 2.1), and in near surface soils and in sediments in the open ocean (Lal et al., 1991; Gu et al., 1996; Barg et al., 1997; Gu et al., 1997; Monaghan et al., 1983; Pavich et al., 1984; Bourles et al., 1989). With such concentrations, ^{10}Be can be detected in samples of ~1 mg terrestrial dust using the AMS method.

Recirculation would be expected to be important only for ^{10}Be and ^{26}Al; the half-life of ^{7}Be is too short, 53.3 days for it to recycle between the atmosphere and the land-surface. In view of the foregoing, most of the ^{10}Be inventory on the land is expected to be located in the upper surface of the lithosphere. Surprisingly however, one finds appreciable amounts of ^{10}Be down to depths of few meters in the soil horizons (Barg et al., 1997; Monaghan et al., 1983; Pavich et al., 1984). This owes itself to the fact that the upper lithic horizons are rich in organic matter and therefore acidic, which causes downward transport of ^{10}Be in the lithic horizons (Lal, 2001). The distribution coefficient depends sensitively on the pH. At pH of ~4–5, it is of the order of 10^{3}–10^{4}, two orders of magnitude smaller than at pH of 6–7 (You et al., 1989).

Strong winds dislodge surface particles and transport micron and sub-micron size particles to great distances (Duce et al., 1980; Prospero et al., 2002). In the case of ^{10}Be, this constitutes an important "focusing" mechanism for transporting this nuclide to different geographical locations, as first pointed out by Monaghan et al. (1985/86). As would be expected this contribution may be quite significant in the polar regions where the depositional fluxes of ^{10}Be are relatively small to begin with (Lal and Peters, 1967; Lal, 1987), in spite of the fact that dust fall-out in polar regions is small. This follows from the fact that the concentrations of ^{10}Be are high in surface soils, lying in the range of 10^{8}–10^{10} atoms/g. Experimental data on dust transported ^{10}Be in the GRIP ice core (Baumgartner et al., 1997) support this theory.

Thus we see that geochemical processes involving concentration of cosmogenic nuclides on particles or authigenic sediments can appreciably alter their depositional patterns from the hydrosphere. The re-deposition of particle active nuclides, ^{10}Be and ^{26}Al in the polar regions by continental soils/dust is a special case of their re-deposition after their initial precipitation to the surface of the earth by wet precipitations. In this paper, considering the importance of ^{10}Be as an important atmospheric tracer, and furthermore since it has been extensively studied in polar ice, we make an assessment of the expected dust-transported fluxes of ^{10}Be to the polar regions, and compare the expectations with the measured total amounts of ^{10}Be in ice cores from Greenland and Antarctica. Similar considerations may apply to the cosmogenic ^{26}Al but in view of paucity of data on its distribution in the continental soils/dust, we defer this discussion.

2. Aeolian transport of cosmogenic ^{10}Be after initial deposition to polar regions

Cosmogenic ^{10}Be has been extensively studied in continental soils/dust (Lal et al., 1991; Gu et al., 1996; Barg et al., 1997; Monaghan et al., 1983; Pavich et al., 1984) and in polar ice samples from the Greenland and Antarctica (Beer et al., 1988; Raisbeck et al., 1992; Yiou et al., 1997; Finkel and Nishiizumi, 1997; Steig et al., 2000). In the following, we proceed to determine first the expected concentrations of ^{10}Be in fine aeolian dust and then the expected amounts of dust transported ^{10}Be in the Greenland and Antarctic ice, basing on the measured or estimated dust concentrations in polar ice samples.

2.1. Expected concentrations of ^{10}Be in fine aeolian dust

We are interested in assessing the concentrations of ^{10}Be in aeolian dust in the Northern and Southern Hemispheres. We show here that the directly available data on ^{10}Be concentrations allows us to make a reasonable estimate of this quantity.

In the Northern Hemisphere, direct measurements of ^{10}Be in aeolian dust in the GRIP ice core (European Ice Core Project), near the Summit in Greenland by Baumgartner et al. (1997) yielded values of $\sim 10^{10}$ atoms ^{10}Be/g dust during the Wisconsin, based on studies of GRIP ice core from the Summit, Greenland, by melting the ice and filtering the melt through 0.45 μm size filter.

In the Southern Hemisphere, we have direct estimates of fluxes of aeolian ^{10}Be to the ocean at ~50° S, 6° E (near the present-day position of the Antarctic Polar Front), based on observations of excess ^{10}Be in marine opal. We recently established (Lal et al., 2005) that marine opal concentrates large amounts of several elements (at levels of ~mg/g for Al, Ca, Fe, Mg and Ti, and ~μg/g for Be, Mn, Zn and Sr). Studies of ^{10}Be and ^{9}Be and trace element concentrations in marine opal in the ocean at ~50° S, 6° E in a time bracket encompassing the last glacial and interglacial period (Lal et al., 2005) showed that as the dissolved concentrations of Be, Al, Ti and Fe increased, so did the ^{10}Be concentrations (Lal et al., 2005). The observations of both ^{10}Be concentrations and the ^{10}Be/^{9}Be ratios are completely consistent with the model of a steady flux of ^{10}Be by precipitation and dissolution of aeolian ^{10}Be and ^{9}Be. The results show that $(2.54 \pm 0.63) \times 10^{9}$ atoms ^{10}Be are added per micro-gram aeolian ^{9}Be dissolved in the ocean; this result is based on the observed ranges of concentrations of $(1.8–4.0) \times 10^{9}$ atoms ^{10}Be/g opal and (0.10–0.95) micro-gram ^{9}Be/g opal. The mean concentrations of ^{9}Be in the 62 soil samples from the N. American continent (Lal et al., 1991; Barg et al., 1997) and in Weinan soils from the Chinese loess plateau (Gu et al., 1996; Zhaoyan Gu, priv. comm.) are found to be 2.69 and 2.78 mg ^{9}Be/g soil, corresponding to an average value of 2.73 mg ^{9}Be/g soil. This leads us to the result that the mean concentration of ^{10}Be in the aeolian soil deposited in the Antarctic Polar Front is $(6.9 \pm 1.7) \times 10^{9}$ atoms ^{10}Be/g aeolian dust.

In a composite "inland aeolian dust sample", representing 1900 m^3 air (STP) filtered on the roof of Dr. J. M. Prospero's laboratory in Univ. of Miami (Florida, USA), we measured ^{10}Be amount. The measured ^{10}Be concentration was $(2.12 \pm 0.036) \times 10^{9}$ atoms/g dust. This should be considered as a lower limit of aeolian transported ^{10}Be since finer dust would be transported to the polar ice; smaller dust particles are expected to have higher ^{10}Be concentrations because ^{10}Be is preferentially adsorbed on smaller size particles.

Alternately, we have made estimates of the expected aeolian fluxes of ^{10}Be based on published literature on its concentrations in soils. Soils are expected to be the principal suppliers of aeolian ^{10}Be (Lal et al., 1991; Barg et al., 1997; Monaghan et al., 1983). Considering the processes involved in the adsorption of ^{10}Be in soil grains, the concentrations of ^{10}Be in soils are expected to depend on the local flux of ^{10}Be at a site and the grain size distribution of the soil matrix. Except for changes expected due to temporal changes in the geomagnetic field, the mean global production rate of ^{10}Be is expected to remain invariant with time (within about ±10%). Consequently, the local flux of ^{10}Be would be expected to be proportional to the ratio of local to zonal precipitation rate (Gu et al., 1996). As discussed earlier, cosmogenic ^{10}Be gets attached strongly to particulate

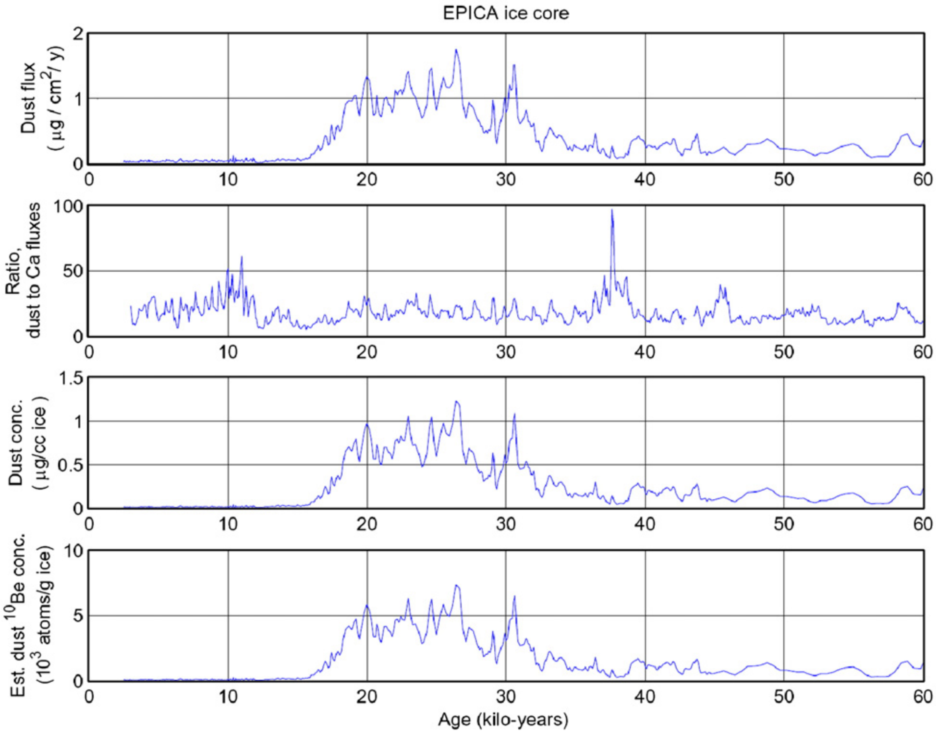

Fig. 1. Measured dust flux, ratio of dust to Ca fluxes and dust concentrations in the EPICA ice core (EPICA community members, 2004; Delmonte et al., 2004) are plotted along with the estimated ^{10}Be concentrations transported by aeolian dust. See text for details.

materials. Based on studies of loess-paleosol sections from several sites in Chinese loess plateau, it was concluded (Gu et al., 1996) that ^{10}Be introduced by wet precipitation is quantitatively retained within a soil layer. The time constant for leakage of ^{10}Be from surface soils due to lowering of soil pH by organic acids (Lal, 2001) is much greater than for losses by surface soil erosion, $\geq 10^4$ years. It should be noted that the bulk soil concentrations of ^{10}Be would grossly underestimate expected aeolian fluxes of ^{10}Be since wind transport favors smaller particles which have higher ^{10}Be concentrations because ^{10}Be is adsorbed preferentially on smaller size particles (cf. Chengde et al., 1992).

A large number of soil samples from different locations in the world, with a greater proportion being from the North American continent (Atlantic Coast and N. American river basins), have been analyzed for their ^{10}Be and ^{9}Be concentrations (Barg et al., 1997; Monaghan et al., 1983; Pavich et al., 1984; Lal, 2001). A detailed mathematical analysis of the ^{10}Be concentrations in soils has been carried out (Lal et al., 1991; Lal, 2001). As stated above, the ^{10}Be concentrations are expected to be a function of local precipitation, surface soil erosion rate, and soil grain size (considerations of surface adsorption lead to $1/r$ (r = grain radius) distribution in the expected ^{10}Be concentrations. Furthermore, the soil grain size distribution depends sensitively on the degree of its pedogenesis. Greater the pedogenesis, larger the fraction of grains of smaller size and higher the ^{10}Be concentration. This expectation is consistent with observations of Chengde et al. (1992). Therefore one would expect that the aeolian transported ^{10}Be would be preferentially contributed by more mature soils. Considering this we take the top quartile mean concentration of ^{10}Be based on 62 soil samples from N. American soils (Lal et al., 1991; Barg et al., 1997), which is found to be 1.35×10^9 atoms/g soil.

The above ^{10}Be concentrations of soils refer to the bulk soil matrix. The ^{10}Be concentrations in the aeolian dust would be much greater than observed in bulk soils, since the aeolian dust is composed of fine particles of ~1 μm size. In air sampled up to 3 km altitude, the differential particle size distributions exhibit a power law spectrum with a slope of (−4) in the 0.1–20 m radii range (Junge, 1963). The differential particle volume spectrum in equal logarithmic intervals of radii of the high altitude dust and particles observed in GISP ice from the Summit, Greenland, shows a broad distribution

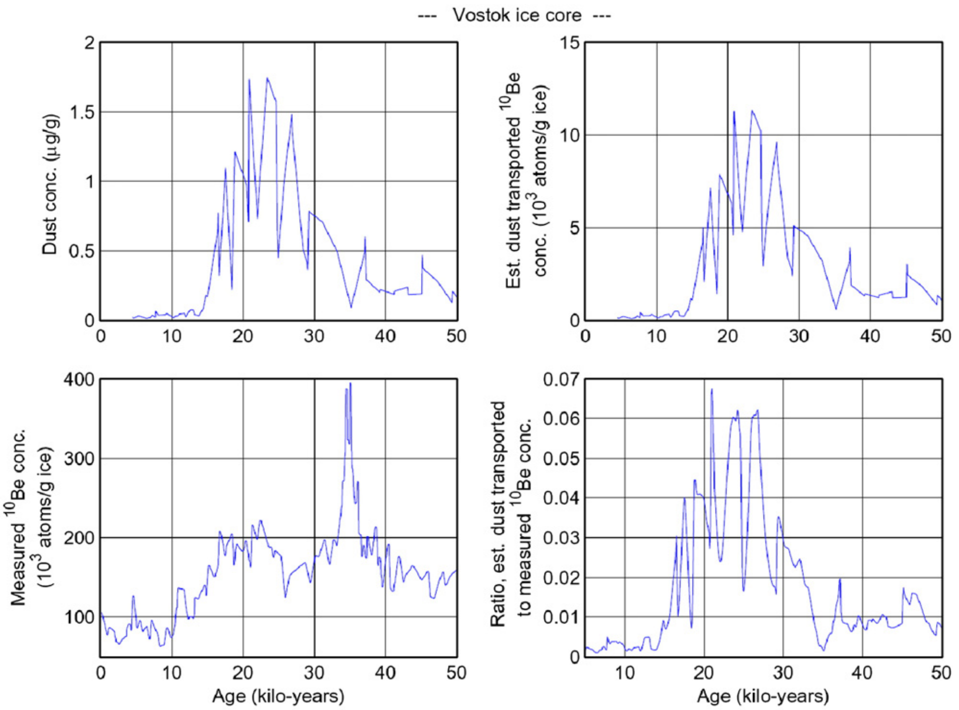

Fig. 2. Measured dust concentrations in the Vostok ice core (Petit et al., 1990; Jouzel et al., 1993), and the estimated dust transported and measured total ^{10}Be concentrations (Raisbeck et al., 1992) are plotted along with the ratios of estimated dust transported and measured total ^{10}Be concentrations. See text for details.

in the 0.3–2 μm range, with a lognormal 1 sigma value of 0.5–1.3 μm (Junge, 1963; Steffensen, 1997).

In order to quantify this we carried out analyses of differential mass and concentrations in two soils from the Chinese loess plateau. For grains of <100 μm size, the differential mass and ^{10}Be distributions follows a power law with mean slopes of (−1.23) and (−1.42), respectively. The mean concentrations of ^{10}Be in the <1 and 1–2 μm size grains were found to be 2.57 and 1.96 times higher, respectively, compared to the concentration in the bulk soil sample. These data are consistent with grain size studies of mass and ^{10}Be concentrations in Luochuan loess from the central area of the loess plateau (Chengde et al., 1992). Therefore, based on the deduced top quartile ^{10}Be concentration in 62 soil samples from N. American soils, we are led to an expectation of $\sim 3\times10^9$ ^{10}Be atoms/g aeolian dust, contributed by soils of grain size <1–2 μm size.

Considering the direct measurements of ^{10}Be concentrations in filtered dust from GRIP ice of $\sim 10^{10}$ atoms/g dust (Baumgartner et al., 1997), the value of $>(2.12\pm0.036)\times10^9$ atoms/g measured in an inland lower troposphere aeolian dust sample, a concentration of $\sim 3\times10^9$ ^{10}Be atoms/g aeolian dust based on soils, and the opal based direct estimate of $(6.9\pm1.7)\times10^9$ atoms ^{10}Be/g aeolian dust in the S. Hemisphere, we consider that a good operational global average value for the aeolian transported ^{10}Be is 6.5×10^9 ^{10}Be atoms/g aeolian dust, with a standard deviation of $\sim 3.5\times10^9$ ^{10}Be atoms/ g aeolian dust.

2.2. Expected concentrations of ^{10}Be in polar ice samples in Greenland and Antarctica

Fairly extensive studies of trace elements, dust and ^{10}Be concentrations in polar ice samples from the Greenland and Antarctica are available to make reasonable estimates of the expected dust-transported ^{10}Be in Greenland and Antarctica samples. In the case of the EPICA core from Dome C, measurements are available for both trace elements and dust (EPICA community members, 2004; Delmonte et al., 2004). In the case of Greenland ice samples from the SUMMIT (GISP and GRIP ice cores), extensive data are available for Ca, which is considered a good proxy archive for dust (Mayewski et al., 1997; De Angelis et al., 1995). We also have some measurements of Ca and dust concentrations in the GRIP core (Steffensen, 1997). Fig. 1 shows the ratio of dust to Ca fluxes and the measured dust concentrations in the EPICA core. The long term averaged dust to Ca ratio is

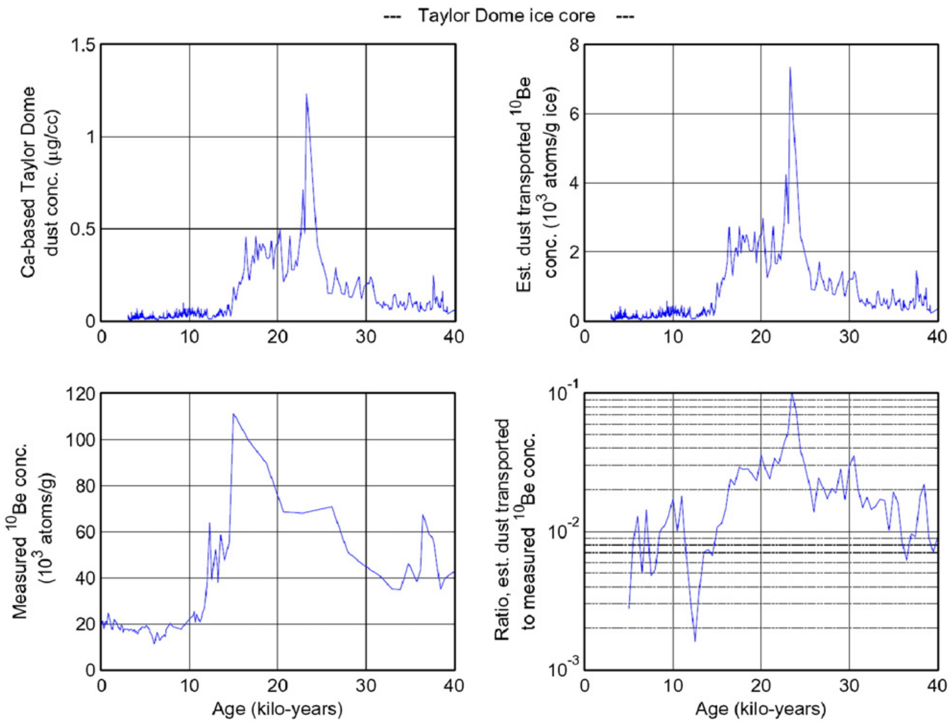

Fig. 3. Estimated dust concentrations in the Taylor Dome ice core based on measured Ca concentrations (Steig et al., 2000), and the estimated dust transported and measured total ^{10}Be concentrations (Steig et al., 2000) are plotted along with the ratios of estimated dust transported and measured total ^{10}Be concentrations. See text for details.

18.2 with a standard deviation of 0.3. The limited data of Steffensen (1997) for the GRIP core are in fair agreement with the EPICA data. During high dust epoch (20–30 kyr B.P.) the average dust to Ca ratios are 12.9 and 9.9 for EPICA and GRIP respectively. We now proceed to discuss the estimated dust transported ^{10}Be concentrations at the Antarctic sites, Vostok and Taylor Dome, and the Greenland sites, GISP and GRIP where dust fluxes are either available or can be estimated from Ca fluxes, and fairly extensive data on ^{10}Be concentrations are available.

2.2.1. EPICA

In Fig. 1, the measured fluxes of dust, dust to Ca flux ratios and the dust concentrations are plotted for the past 60 kyr B.P. (EPICA community members, 2004; Delmonte et al., 2004). The estimated mean dust transported ^{10}Be concentrations, basing on the global average value of 6.5 (±3.5)×10^9 atoms ^{10}Be/g aeolian dust (Section 2.1), are also plotted.

2.2.2. Vostok

The measured dust concentrations in Vostok ice core (Petit et al., 1990; Jouzel et al., 1993), and the measured total ^{10}Be concentrations (Raisbeck et al., 1992) in ice are plotted in Fig. 2, along with the estimated dust transported ^{10}Be concentrations, basing on the global average value of 6.5 (±3.5)×10^9 atoms ^{10}Be/g aeolian dust (Section 2.1). Also plotted are the ratios of estimated dust transported and measured total ^{10}Be concentrations.

2.2.3. Taylor Dome

Direct estimates of dust concentrations in Taylor Dome are not available. We have therefore estimated the dust concentrations based on Ca concentrations (Steig et al., 2000) and the ratio of dust to Ca fluxes for the EPICA core (Fig. 1). The estimated dust and dust transported ^{10}Be concentrations in ice are shown in Fig. 3, along with the measured total ^{10}Be concentrations (Steig et al., 2000). Dust transported ^{10}Be was estimated as in Fig. 1.

2.2.4. GISP

As in the case of Taylor Dome, direct estimates of dust concentrations are not available for GISP. We have therefore estimated the dust concentrations based on Ca concentrations (Mayewski et al., 1997) and the ratio of dust to Ca fluxes for the EPICA core (Fig. 1). The estimated dust and dust transported ^{10}Be concentrations are shown in Fig. 4, along with the measured ^{10}Be concentrations in melt water filtered through 0.45 μm

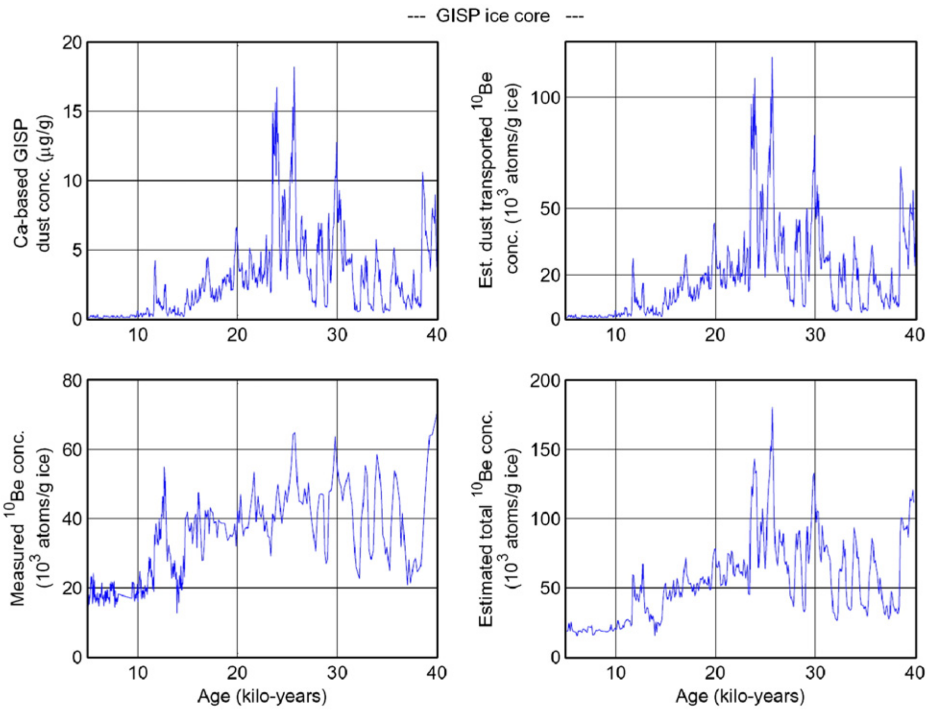

Fig. 4. Estimated dust concentrations in the GISP ice core based on Ca concentrations (Mayewski et al., 1997), and the estimated dust transported and measured ^{10}Be concentrations in melt water filtered through 0.45 µm Millipore filter (Finkel and Nishiizumi, 1997) are plotted along with the estimated total expected ^{10}Be concentrations. See text for details.

Millipore filter (Finkel and Nishiizumi, 1997), and also the estimated *total* expected ^{10}Be concentrations (sum of measured and the estimated dust transported ^{10}Be concentrations). Dust transported ^{10}Be was estimated as in Fig. 1.

2.2.5. GRIP

Direct estimates of dust concentrations are also not available for GRIP. We have therefore estimated the dust concentrations based on Ca concentrations (Jouzel et al., 1993) and the ratio of dust to Ca fluxes for the EPICA core (Fig. 1). The estimated dust and dust transported ^{10}Be concentrations are shown in Fig. 5 along with the measured ^{10}Be concentrations in melt water filtered through 0.40 and 0.45 µm Millipore filters (Yiou et al., 1997, 2002). The estimated *total* expected ^{10}Be concentrations (sum of measured and the estimated dust transported ^{10}Be concentrations) are also plotted in Fig. 5. Dust transported ^{10}Be was estimated as in Fig. 1.

3. Discussion of contributions of dust transported ^{10}Be in Greenland and Antarctica sites

At the outset we state that (i) data on ^{10}Be concentrations are not available at present for EPICA ice core, (ii) in the case of Vostok and Taylor Dome ice, the measured ^{10}Be concentrations refer to the total amount of ^{10}Be in the ice, and (iii) in the case of GISP and GRIP ice, ^{10}Be was measured in melt water filtered through Millipore filters to remove aeolian dust. The measured ^{10}Be concentrations are therefore expected to be largely free of contributions from dust transported ^{10}Be in the GISP and GRIP ice.

The estimated contributions of dust transported ^{10}Be are not appreciable for Vostok and Taylor Dome, as can be seen from Figs. 2 and 3. The ratios of estimated dust transported to measured *total* ^{10}Be concentrations are <10% even during peak dust epochs plotted. The largest dust transported ^{10}Be fractions are about 6% and 10% for Vostok and Taylor Dome respectively, during ~22–25 kyr B.P.

During the Holocene, the estimated dust transported ^{10}Be concentrations in the two Greenland cores, GISP and GRIP cores are <10% of the measured atmospheric concentrations (Figs. 4 and 5). However, the situation is quite different in these cores during 10–40 kyr B.P. The ratios of estimated dust transported ^{10}Be to measured *atmospheric* ^{10}Be concentrations for GISP and GRIP are frequently ≥1 during the period, 20–30 kyr B.P.; the two data sets are fairly similar (Figs. 4 and 5) which is

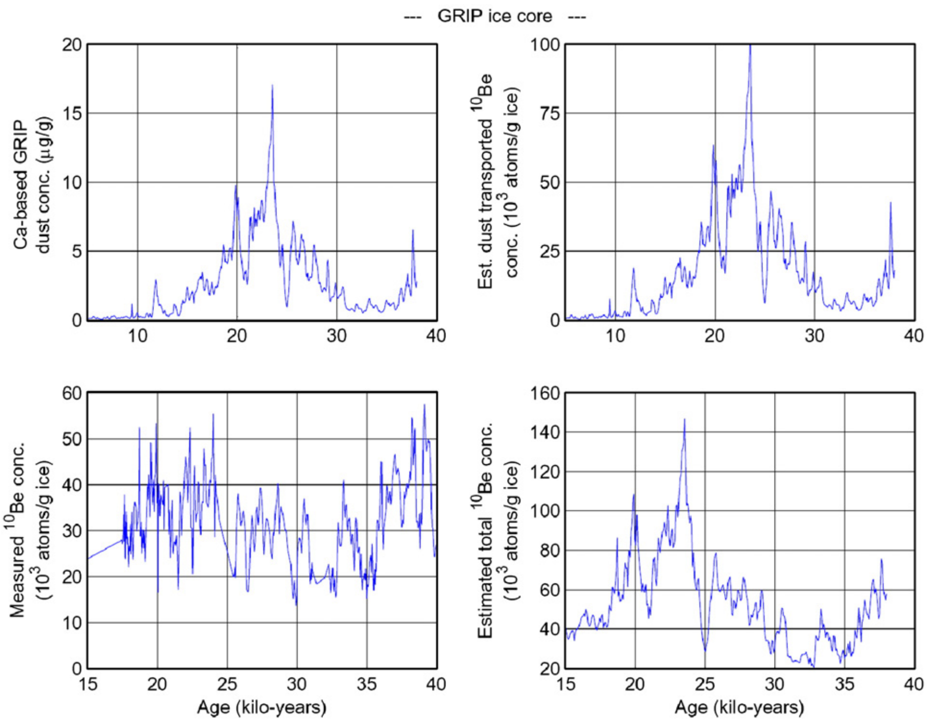

Fig. 5. Estimated dust concentrations in the GRIP ice core based on Ca concentrations (De Angelis et al., 1995), and the estimated dust transported and measured ^{10}Be concentrations in melt water filtered through 0.40 and 0.45 μm Millipore filters (Yiou et al., 1997, 2002) are plotted along with the estimated total expected ^{10}Be concentrations. See text for details.

not surprising since these ice cores are from near the Summit in Greenland. Our model results show that within the uncertainties of the estimates, the dust transported ^{10}Be flux at these sites during 20–30 kyr B.P. was often comparable to or exceeded the atmospheric ^{10}Be flux, by factors of 1.5–2, and that therefore great care should be taken in interpreting the ^{10}Be data. This is in accord with the findings of Baumgartner et al. (1997) who also measured the ^{10}Be in dust particles in several samples from the GRIP ice core, obtained by filtering the melt water through 0.4 and 0.5 μm Millipore filter paper.

As stated earlier, direct measurements of ^{10}Be sticking to dust particles in the GRIP ice core have been made by Baumgartner et al. (1997) by melting the ice and filtering the melt through 0.45 μm size filter. They found that the contribution of dust borne ^{10}Be was <5% during the Holocene, and about 20% (±5%) between 16–100 kyr B.P. Their results are in general agreement with our model calculations for the dust transported ^{10}Be for 10–15 and 30–40 kyr B.P. intervals, but are in disagreement for the time bracket, 15–30 kyr B.P. (Fig. 5), when dust fluxes seem to contribute dominantly to the ^{10}Be present in ice (even considering the 50% uncertainty in the estimate of ^{10}Be concentration of aeolian dust). The disagreement in the 15–30 kyr B.P. time interval may partly be due to the fact that a fraction of the dust transported ^{10}Be may get solubilized during melting of the ice in the experiments of Baumgartner et al. (1997)! The possibility that we are overestimating the dust concentrations in the GRIP core, which are based on the measured Ca concentrations, is ruled out since dust to Ca ratios in the GRIP core have been measured by Steffensen (Steffensen, 1997) for the GRIP core in the time bracket, 20–30 kyr B.P. As mentioned earlier, his data are similar to the dust to Ca ratios measured in EPICA (Section 2.2).

Our model calculations which imply that most of the measured ^{10}Be is dust transported in the time bracket, 15–30 kyr B.P. can be understood as a combined result of the facts that this is also the period of lowest accumulation rates in GISP and GRIP cores, and that the dust concentrations are high with values of ~5–15 μg/gram ice during appreciable part of the period (Figs. 4 and 5).

An interesting conclusion follows from the above discussion. In Fig. 6, we have plotted the ^{10}Be concentrations and the estimated atmospheric ^{10}Be fluxes in the GISP and GRIP ice cores for the past 40 kyr B.P. The latter is estimated by multiplying the

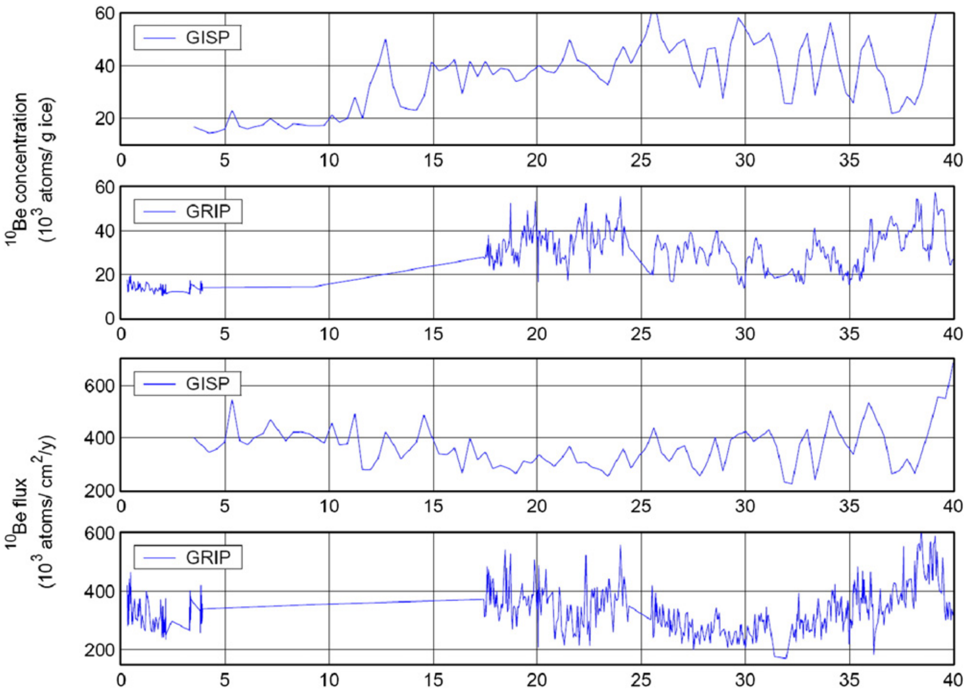

Fig. 6. Measured ^{10}Be concentrations in the Millipore filtered melt water in the GISP (Finkel and Nishiizumi, 1997) and GRIP (Yiou et al., 1997, 2002) ice cores, and the corresponding ^{10}Be fluxes are plotted for the past 40 kyr B.P.

^{10}Be concentrations by the corresponding accumulation rates, as is conventionally done. It is seen that during the time bracket, 20–35 kyr B.P. the atmospheric ^{10}Be fluxes in the GISP and GRIP ice cores are generally lower than that during the Holocene by up to ~50%. This compounded with the fact that during this period Holocene ^{10}Be data are relatively free from dust contributions, and that the data 20–35 kyr B.P. are inferred to be primarily dust contributed, lead to the conclusion that the fall–out fluxes of ^{10}Be at the SUMMIT due to scavenging of air masses by snow are appreciably reduced during 20–35 kyr B.P. Since one can not attribute this anomaly to a decrease in cosmic ray flux, we interpret this as an anomaly caused by an appreciable reorganization of large scale air circulation in the polar atmosphere.

4. Conclusions

Cosmic ray produced isotopes recycle during dynamic exchange processes between the atmosphere, hydrosphere and the lithosphere. We have discussed that amongst different cosmic ray produced isotopes in the atmosphere, the recycling of particle active nuclides, ^{10}Be and ^{26}Al can appreciably alter their initial fall-out patterns on the surface of the Earth. To date, experimental data on ^{10}Be alone allow one to estimate quantitatively the importance of its being transported to the polar regions by aeolian dust deposited in ice sheets.

In an earlier study, Baumgartner et al. (1997) studied dust transported ^{10}Be by melting the ice from the GRIP ice core from the Summit (Greenland) and measuring ^{10}Be concentrations in the dust retained on a 0.45 μm size filter. They found that the contribution of dust borne ^{10}Be to total ^{10}Be was <5% during the Holocene, and it was about 20% (±5%) between 16–100 kyr B.P.. In the present study, we have followed a different approach. We have estimated the ^{10}Be concentrations of aeolian dust, and the expected dust transported ^{10}Be concentrations in ice based on dust concentrations of ice. The latter is based on direct measurements of dust in ice or the concentrations of dust or Ca. In Section 2.1, we show that a good operational global average value for the aeolian transported ^{10}Be is 6.5×10^9 ^{10}Be atoms/ g aeolian dust, with a standard deviation of $\sim 3.5 \times 10^9$ ^{10}Be atoms/g aeolian dust.

The model calculations allow us to reach some important conclusions, besides estimating dust transported ^{10}Be at two sites in Greenland, and at three sites in Antarctica.

Data on ^{10}Be concentrations in EPICA ice core are not yet available. We are however able to compare our estimates of dust transported ^{10}Be with measurements of ^{10}Be in the ice cores, Vostok, Taylor Dome, GISP and

GRIP. Contributions of dust transported ^{10}Be for Vostok and Taylor Dome are estimated to be <3% except during the period 20–30 kyr B.P. when they are at maximum ~6% and ~10%, respectively (Figs. 2 and 3). In the case of GISP and GRIP ice cores, during the Holocene, dust transported ^{10}Be contributions are <10% of the measured atmospheric concentrations (Figs. 4 and 5). During 10–40 kyr B.P., however, dust contributions are often very appreciable in the case of GISP and GRIP. The ratios of estimated dust transported ^{10}Be to measured atmospheric ^{10}Be concentrations for GISP and GRIP are fairly similar, which is not surprising since the two cores are from near the Summit in Greenland. The ratios are greater than 1 during most of 15–30 kyr B.P. (Fig. 6). Even considering the 50% uncertainty in the estimate of ^{10}Be concentration of aeolian dust (Section 2.1), the result implies that during this time bracket, a large fraction of the measured ^{10}Be concentration is dust transported. We have stated above that this result is at divergence with the experimental data of Baumgartner et al. (1997), who melted the GRIP ice samples and measured ^{10}Be amounts in the dust obtained by filtering the melt through 0.45 μm size filter. They found that the dust borne ^{10}Be to total ^{10}Be was <5% during the Holocene, and about 20% (±5%) between 16–100 kyr B.P. Their results are however in general agreement with our model calculations for 10–15 and 30–40 kyr B.P. intervals, The disagreement in the 15–30 kyr B.P. time interval may partly be due to the fact that a fraction of the dust transported ^{10}Be may get solubilized during melting of the ice in the experiments of Baumgartner et al. (1997)! We have discussed that the possibility that we are overestimating the dust concentrations in the GRIP core, which are based on the measured Ca concentrations, is ruled out since dust to Ca ratios in the GRIP core have been measured by Steffensen (Steffensen, 1997) for the GRIP core in the time bracket, 20–30 kyr B.P.

We point out that there must have occurred a major reorganization of the air circulation pattern in the polar region during 20–35 kyr B.P. since we note that the observed ^{10}Be fluxes during this time bracket are appreciably lower than during Holocene (Fig. 6). This implies that that the fall-out fluxes of atmospheric ^{10}Be at the SUMMIT due to scavenging of air masses by snow are lower during 20–35 kyr B.P. Since one can not attribute this anomaly to a decrease in cosmic ray flux, we interpret this as an anomaly caused by an appreciable reorganization of large scale air circulation in the polar atmosphere.

Finally, we would like to note as a caution that since appreciable amounts of ^{10}Be amounts can be transported to the polar regions by dust, even during periods when aeolian dust concentrations are low, e.g. during the Holocene, and most of the time in the Antarctica, significant dust transport by dust can occur during short periods of time when climate cools, causing increased winds and higher aeolian dust fluxes. Short duration pulses of ^{10}Be in such circumstances should be carefully examined for possible transport of ^{10}Be by dust.

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References

Aldahan, A., Haiping, Y., Possnert, G., 1999. Distribution of beryllium between solution and minerals (biotite and albite) under atmospheric conditions and variable pH. Chem. Geol. 156, 209–229.

Axford, W.I., 1968. Polar wind and terrestrial helium budget. J. Geophys. Res. 73, 6855–6859.

Banks, P.M., Holzer, T.E., 1960. High latitude plasma transport: the polar wind. J. Geophys. Res. 74, 6317–6332.

Barg, E., Lal, D., Southon, J.R., Caffee, M.W., Finkel, R.C., Pavich, M.J., 1997. Beryllium geochemistry in soils: evaluation of ^{10}Be/^{9}Be ratios in authigenic minerals as a basis for age models. Chem. Geol. 140, 237–258.

Baumgartner, S., Beer, J., Wagner, G., Kubik, P., Suter, M., Raisbeck, G.M., Yiou, F., 1997. ^{10}Be and dust. Nucl. Instrum. Methods Phys. Res., B Beam Interact. Mater. Atoms 123, 296–301.

Beer, J., Siegentahler, U., Bonani, G., Finkel, R.C., Oeschger, H., Suter, M., Wolfli, W., 1988. Information on past solar activity and geomagnetism from ^{10}Be in the Camp Century ice core. Nature 331, 675–679.

Bourles, D., Raisbeck, G.M., Yiou, F., 1989. ^{10}Be and ^{9}Be in marine sediments and their potential for dating. Geochim. Cosmochim. Acta. 53, 443–452.

Chengde, S., Beer, J., Tungsheng, L., Oeschger, H., Bonani, G., Suter, M., Wolfli, W., 1992. ^{10}Be in Chinese loess. Earth Planet. Sci. Lett. 109, 169–177.

De Angelis, M., Steffensen, J.P., Legrand, M.R., Clausen, H.B., Hammer, C.U., 1995. Primary aerosol (sea salt and soil dust) deposited in Greenland ice during the last climatic cycle: comparison with east Antarctic records. J. Geophys. Res. 102, 26681–26698.

Delmonte, B., et al., 2004. EPICA Dome C Ice Cores Insoluble Dust Data. IGBP PAGES/World Data Center for Paleoclimatology Data Contribution Series # 2004-040. NOAA/NGDC Paleoclimatology Program, Boulder CO, USA.

Duce, R.A., Unni, C.K., Ray, B.J., Prospero, J.M., Merrill, J.T., 1980. Long range transport of soil dust from Asia to the tropical North Pacific; temporal variability. Science 209, 1522–1524.

EPICA community members, 2004. Eight glacial cycles from an Antarctic ice core. Nature 429 (6992), 623–628. doi:10.1038/nature02599 (June 10, 2004).

Finkel, R.C., Nishiizumi, K., 1997. Beryllium 10 concentrations in the Greenland Ice Sheet Project 2 ice core from 3–40 ka. J. Geophys. Res. 102 (#C12), 26,699–26,706.

Gu, Z.Y., Lal, D., Liu, T.S., Southon, J., Caffee, M.W., Guo, Z.T., Chen, M.Y., 1996. Five million year ^{10}Be record in Chinese loess and red-clay: climate and weathering relationships. Earth Planet. Sci. Lett. 144, 273–287.

Gu, Z.Y., Lal, D., Liu, T.S., Guo, Z.T., Southon, J., Caffee, M.W., 1997. Weathering histories of Chinese loess deposits based on uranium and thorium series nuclides and cosmogenic ^{10}Be. Geochim. Cosmochim. Acta. 61, 5221–5231.

Jouzel, J., Barkov, N.I., Barnola, J.M., Bender, M., Chappelaz, J., Genthon, C., Kotlyakov, V.M., Lipenkov, V., Lorius, C., Petit, J.R., Raynaud, D., Raisbeck, G., Ritz, C., Sowers, T., Stievenard, M., Yiou, F., Yiou, P., 1993. Extending the Vostok ice-core record of paleoclimate to the penultimate glacial period. Nature 364, 407–412.

Junge, C.E., 1963. Air chemistry and radioactivity. Academic Press, New York. 382 pp.

Krishnaswami, S., Lal, D., 1973. Exchange of properties/materials at geochemical interfaces. Proc. Symp. on Hydrogeochemistry and Biogeochemistry, Tokyo, September 6–12, 1970. Hydrogeochemistry and Biogeochemistry, vol. I. The Clarke Company, Washington, D.C., pp. 581–600.

Lal, D., 1987. ^{10}Be in polar ice: data reflect changes in cosmic ray flux or polar meteorology? Geophys. Res. Lett. 14, 785–788.

Lal, D., 2001. New nuclear methods for studies of soil dynamics utilizing cosmic ray produced radionuclides. In: Stott, D.E., Mohr, R.H., Steinhardt, G.C. (Eds.), Proc. 10th Int. Soil Conservation Organization Conference, held at Purdue University, W. Lafayette, IN, May 24–29, 1999, pp. 1044–1052.

Lal, D., 2002. Cosmogenic radionuclides. In: Holton, J.R., Pyle, J., Curry, J.A. (Eds.), Encyclopedia of Atmospheric Sciences. Academic Press, pp. 1891–1900.

Lal, D., Peters, B., 1967. In: Flugge, S. (Ed.), Cosmic ray produced radioactivity on the earth. Handbuch der Physik, vol. 46/2. Springer & Verlag, Berlin, pp. 551–612.

Lal, D., Barg, E., Pavich, M., 1991. Development of cosmogenic nuclear methods for the study of soil erosion and formation rates. Curr. Sci. 61, 636–639.

Lal, D., Charles, C., Vacher, L., Goswami, J.N., Jull, A.J.T., McHargue, L., Finkel, R.C., 2005. Paleo-ocean chemistry records in marine opal: implications to aeolian fluxes of suite elements and cosmogenic nuclides, ^{10}Be and ^{26}Al, and to biological productivity. Geochim. Cocmochim. Acta. 70, 3275–3289.

Macdonald, G.J.F., 1963. The escape of helium from the earth's atmosphere. Rev. Geophys. 1, 305–349.

Marcantonio, F., Anderson, R.F., Stute, M., Kumar, N., Schlosser, P., Mix, A., 1996. Extraterrestrial He-3 as a tracer of marine sediment transport and accumulation. Nature 383, 705–707.

Mayewski, P.A., Meeker, L.D., Twickler, M.S., Whitlow, S.I., Yang, Q., Lyons, W.B., Prentice, M., 1997. Major features and forcing of high-latitude northern hemisphere atmospheric circulation using a 110,000-year-long glaciochemical series. J. Geophys. Res. 102, 26345–26366.

Monaghan, M.C., Krishnaswami, S., Thomas, J.H., 1983. ^{10}Be concentrations and the long-term fate of particle-reactive nuclides in five soil profiles from California. Earth Planet. Sci. Lett. 65, 51–60.

Monaghan, M.C., Krishnaswami, S., Turekian, K.K., 1985/86. The global-average production rate of ^{10}Be. Earth Planet. Sci. Lett. 76, 179–187.

Nyffler, U.P., Li, Y.-H., Santschi, P.H., 1984. A kinetic approach to describe trace element distribution between particles and solution in natural aquatic systems. Geochim. Cosmochim. Acta. 48, 1513–1522.

Pavich, M.J., Brown, L., Klein, J., Middleton, R., 1984. ^{10}Be accumulation in a soil chronosequence. Earth Planet. Sci. Lett. 68, 198–204.

Petit, J.R., Mounier, L., Jouzel, J., Korotkevitch, Y., Kotlyakov, V., Lorius, C., 1990. Paleoclimatological implications of the Vostok core dust record. Nature 343, 56–58.

Prospero, J.M., Ginoux, P., Torres, O., Nicholson, S.E., Gill, T.E., 2002. Environmental characterization of atmospheric soil dust identified with the NIMBUS 7 total ozone mapping spectrometer (TOMS) absorbing aerosol product. Rev. Geophys. 40 (1), 1002. doi:10.1029/2000RG000095.

Raisbeck, G.M., Yiou, F., Jouzel, J., Petit, J.R., Barkov, N.I., Bard, E., 1992. ^{10}Be deposition at Vostok, Antarctica, during the last 50,000 years and its relationship to possible cosmogenic production variations during this period. In: Bard, E., Broecker, W.S. (Eds.), The Last deglaciation: Absolute and Radiocarbon Chronologies. NATO ASI Ser., vol. 12, pp. 127–139.

Steffensen, J.P., 1997. The size distribution of microparticles from selected segments of the Greenland Ice Core Project ice core representing different climate periods. J. Geophys. Res. 102 (#C12), 26,755–26,763.

Steig, E.J., Morse, D.L., Waddington, E.D., Stuiver, M., Grootes, P.M., 2000. Wisconsinan and Holocene climate history from an ice core at Taylor Dome, Western Ross Embayment, Antarctica. Geogr. Ann. 82a, 213–235.

Yiou, F., Raisbeck, G.M., Baumgartner, S., Beer, J., Hammer, C., Johnsen, S., Jouzel, J., Kubik, P.W., Lestringuez, J., Stievenard, M., Suter, M., Yiou, P., 1997. Beryllium 10 in the Greenland Ice Core Project ice core at Summit, Greenland. J. Geophys. Res.-Oceans 102 (#C12), 26,783–26,794.

Yiou, F., et al., 2002. GRIP Ice Core Beryllium 10 Data, IGBP PAGES/World Data Center for Paleoclimatology Data Contribution Series #2002-085. NOAA/NGDC Paleoclimatology Program, Boulder CO, USA.

You, C.-F., Lee, T., Li, Y.-H., 1989. The partition of beryllium between soil and water. Geochim. Cocmochim. Acta. 77, 105–118.