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Atmospheric impact on beryllium isotopes as solar activity proxy

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[1] Reconstructing solar activity variability beyond the time scale of actual measurements provides invaluable data for modeling of past and future climate change. The ^{10}Be isotope has been a primary proxy archive of past solar activity and cosmic ray intensity, particularly for the last millennium. There is, however, a lack of direct high-resolution atmospheric time series on ^{10}Be that enable estimating atmospheric modulation on the production signal. Here we report quasi-weekly data on ^{10}Be and ^{7}Be isotopes covering the periods 1983–2000 and 1975–2006 respectively, that show, for the first time, coherent variations reflecting both atmospheric and production effects. Our data indicate intrusion of stratosphere/upper troposphere air masses that can modulate the isotopes production signal, and may induce relative peaks in the natural ^{10}Be archives (i.e., ice and sediment). The atmospheric impact on the Be-isotopes can disturb the production signals and consequently the estimate of past solar activity magnitude.

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1. Introduction

[2] The coupled effect of enhanced solar irradiation and atmospheric CO$_2$ may add further loading on global climate warming, particularly with present day high atmospheric CO$_2$ level and expected increase during the near future. An accurate estimate of past solar modulation is thus needed for evaluating changes in the solar total radiation (particularly in the ultraviolet waves) that may affect stratospheric ozone and ultimately the climate [*Foukal et al.*, 2006; *Rottman*, 2006; *Ammann et al.*, 2007; *Schöll et al.*, 2007]. The cosmogenic isotope ^{10}Be (half-life 1.3–1.5 Myr) [*Fink and Smith*, 2007] plays a critical role in retrieving information about past solar activity variations and changes in cosmic rays and geomagnetic field intensity [*Beer et al.*, 1990, 2006; *Aldahan et al.*, 1999, 2001; *Webber and Higbie*, 2003; *Caballero-Lopez et al.*, 2004; *Usoskin et al.*, 2003, 2006; *Muscheler et al.*, 2007]. ^{7}Be (half-life 53.4 days) and the ratio of ^{10}Be/ ^{7}Be have been used to link atmospheric production of Be isotopes to short-term atmospheric processes, such as source and ages of air masses [*Jordan et al.*, 2003].

[3] About 99.9% of ^{10}Be and ^{7}Be natural production occur in the atmosphere, with a residence time of up to a few months in the troposphere and up to 2 years in the stratosphere [*Raisbeck et al.*, 1981; *Lal*, 1992, *Aldahan et al.*, 2001]. Presently little is known about post-production incorporation modes of Be-isotopes and we expect that a large portion adsorb to aerosol particles, due to its small ionic radii, valence charge (Be^{+2}) and strong chemical adsorption affinity. Despite the complex atmospheric effect of mixing and circulation, solar modulation influence (11-year solar cycle) on production of Be-isotopes has been traced in aerosol ^{7}Be [*Aldahan et al.*, 2001] and ice core ^{10}Be [*Beer et al.*, 1999, 2006; *Usoskin et al.*, 2003]. However, scarcity of time series data on atmospheric ^{10}Be has led to the assumption of a constant fallout rate on reconstruction of solar activity from ice core ^{10}Be [*Muscheler et al.*, 2007]. Accordingly, any high-resolution archive for atmospheric ^{10}Be will add new significant information as shown by the presented weekly record of aerosols Be isotopes at high latitudes (southern and northern Sweden; 56.08°N, 13.23°E, 43 m asl and 67.84°N, 20.34°E, 408 m asl; hereafter referred to as sites 56°N and 68°N) covering the period 1975–2006 (sampling, analytical techniques and raw data are given in the auxiliary material[1]).

2. Results and Discussion

[4] The datasets presented here show that the average ^{10}Be concentration is about double that of ^{7}Be in both latitudes. There is also a 20% and 30% higher concentration at latitude 56°N compared with latitude 68°N in the two isotope records respectively (Figures 1a and 1b). In both records there is a clear enhancement during spring-summer months, when concentrations are up to 70% higher than during winter months. These enhancements may also be partly reflected in the ratio of ^{10}Be/^{7}Be (Figure 1c). The seasonal pattern is removed when calculating annual averages (mean value of the six weeks for ^{10}Be; Figure 1d), which is particularly important for estimates of past solar activity parameters from the ^{10}Be data, as further discussed in the text. Sporadic events having concentrations 2–3 times the average are found in both the ^{10}Be and ^{7}Be records (Figure S1 and Data Set S1 of the auxiliary material). These events are better observed in the ^{7}Be dataset because of the higher spatial and temporal resolution compared with ^{10}Be. Despite the latitudinal, seasonal and sporadic events, the datasets show a production modulation signal corresponding to the solar activity 11-year cycle (Figure 2). The difference between solar maxima and minima is reflected by $\leq$ 50% difference in the beryllium isotope concentrations (Figure 2). However, the translation of a combined solar activity (modulating production) and cosmic rays (production source) effects into cosmogenic isotope deposition is non-

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[1] Auxiliary material data sets are available at ftp://ftp.agu.org/apend/gl/2008gl035189. Other auxiliary material files are in the HTML.

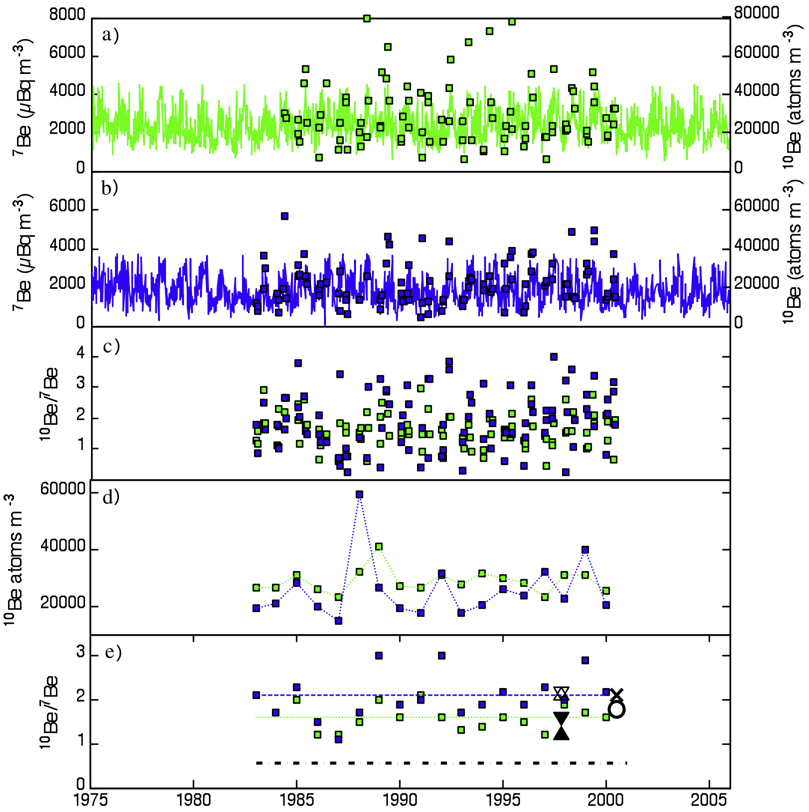

Figure 1. ^{7}Be (solid lines) and ^{10}Be (squares with error bars included) at (a) 56°N and (b) 68°N. ^{7}Be are continuous weekly samples while ^{10}Be are based on selected weeks (2, 4, 6, 19, 21 and 24). Stratospheric intrusions (sample-[mean + 2σ]) have been subtracted from the raw data. (c) ^{10}Be/^{7}Be (atom/atom) between 1983 and 2000 at 56°N (green squares) and 68°N (blue squares) and 7 extreme ratios (reaching 16, Data Set S1) were removed. (d) Annual mean ^{10}Be for 56°N (green squares) and 68°N (blue squares). (e) Annual ^{10}Be/^{7}Be (atom/atom) data of the studied sites plotted together with data from Jungfraujoch, 46°N (**X**) and Zugspitze, 47°N (**O**) (20) and the SONEX flights 6 and 8 flown from latitude 44°N (∇) to 53°N (Δ) and 54°N (▼) to 68°N (▲), respectively (13). The theoretical production ratio (0.5) is shown as a black dashed line.

linear and can be affected by both atmospheric and production factors [*Field et al.*, 2006; *Usoskin et al.*, 2006]. The valuable data on ^{7}Be and ^{10}Be in precipitation by *Heikkilä et al.* [2007] cover a period of 8 years and could not expose the effect of an 11-year solar cycle. The contemporaneous reflection of seasonal (Figure 3) and solar activity (Figure 2) cycles in the ^{10}Be and ^{7}Be presented here represents the first, to our knowledge, published direct atmospheric observation.

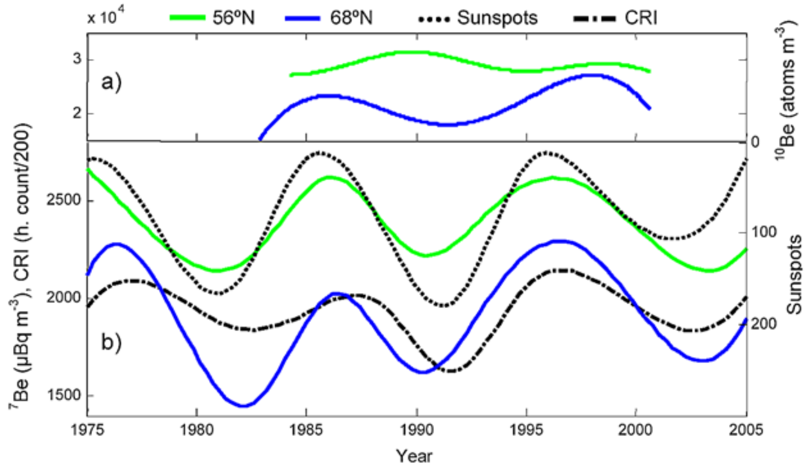

Figure 2. Smoothed curves (using a spline algorithm) of annual mean (a) ^{10}Be and (b) ^{7}Be at 56°N and 68°N (green and blue lines respectively) are plotted together with smoothed Colorado Climax data of cosmic ray intensity, CRI, (black dash-dotted line) and sunspot number (black dotted line). The cosmic ray data and sunspot numbers are available at http://www.ngdc.noaa.gov/stp/SOLAR/COSMIC_RAYS/cosmic.html.

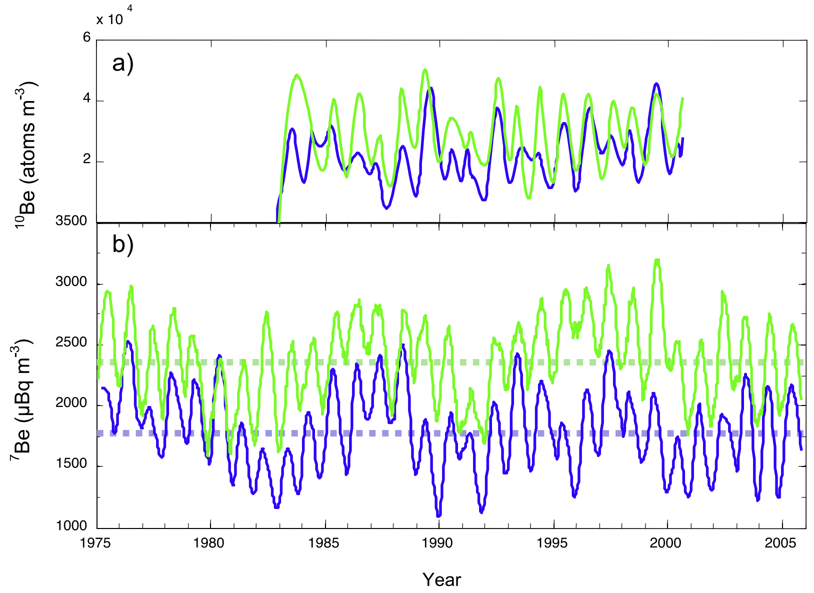

Figure 3. Seasonal variations of (a) ^{10}Be and (b) ^{7}Be at 56°N and 68°N (green and blue lines respectively). The average values are indicated with straight dashed lines for both time series in Figure 3b. In addition to the seasonal fluctuations, the ~10 year solar cycle is clearly visible at both locations in the ^{7}Be data.

Such an empirical finding provides a new perspective for the understanding of pathways, sources and atmospheric effects on the spatial and temporal variability of ^{10}Be in the atmosphere. The correlation between solar activity, cosmic rays and beryllium isotopes is better elucidated for ^{7}Be than for ^{10}Be due to the much higher resolution and longer time span of the ^{7}Be data. Furthermore, the timing between low and high Be values and solar activity extremes seems to be more in phase for ^{7}Be than for ^{10}Be. A possible explanation for this feature can be a lag in the response to mixing rates between stratospheric and tropospheric air masses at the two investigated sites, since the Be spring signals are up to 3 months earlier at site 56°N compared with site 68°N.

[5] Causes of the distinct seasonal differences in the beryllium concentration (Figure 3) apparently reflect the influence of both tropopause folding, most effective during spring-summer [*Feely et al.*, 1989], and temperature, enhancing aerosol abundance during spring-summer season [*Tunved et al.*, 2003]. Evidently both effects are seasonally delayed going northward from middle to higher latitudes. Although there is a coherent seasonal variability in the isotopes signal, a perfect correlation between the weekly data is lacking. This feature can be related to differences in regional air mass trajectories (even on a daily scale) that provide variable input of ^{7}Be and ^{10}Be to a sampling site (example is given in the auxiliary material, Figure S2). Local re-suspension may induce some discrepancy in the ^{10}Be due to its long half-life. However, the weekly differences seem to be damped when averaging the data on seasonal and annual scales. The sporadic peaks found in both the ^{7}Be and ^{10}Be records likely reflect intrusions of Be-rich air mass that may have a stratospheric source, because 50–70% of the Be-isotope production occurs in the stratosphere. Be-rich air masses may also occur within the upper troposphere [*Jordan et al.*, 2003], but both studied sites (latitude 68°N and 56°N) show relatively frequent occurrences of the peaks suggesting a mixed stratosphere/troposphere effect. If tropospheric air masses are the only source, then more peaks are expected at latitude 56°N due to higher reoccurrence of turbulent low pressure systems at the lower latitudes. Irrespective of the source, we used information from flight observations [*Jordan et al.*, 2003] that suggest about > 200 times higher loading of Be-isotopes in stratosphere/upper troposphere air mass, compared to the lower troposphere. Accordingly, an empirical formulation using intrusion = Rs- (mean+2σ), where Rs is a sample value and ó is standard deviation, was used to remove intrusions, which represent about 2–4% and 4–5% of ^{10}Be and ^{7}Be records respectively. This filtration (although of small magnitude) clearly enhances the solar modulation cycle picked up by the Be-records and provides, for the first time, a physical explanation for one of the possible reasons behind relative peaks in the natural ^{10}Be archives (i.e., ice and sediment).

[6] Our results also show that if the intrusions are not removed from the dataset, they may cause an amplitude difference (by up to 15%) in the smoothed Be isotope curves that can induce misleading information about the annual averages. The effect is that a year with relatively frequent and large input of beryllium intrusions will have higher isotopes average values than other years and thus also a higher noise level in the whole dataset. This effect may not be catastrophic when fitting the production (approximation of solar activity) to a long-term beryllium isotope time series (Figure 2). However, if several years have frequent intrusions (elevated concentrations), then the reconstruction of the solar activity cycle may be affected and filtration of the raw data will be necessary. Absence of intrusions (peaks), best exemplified by the high-resolution ^{7}Be record, occurs in some consequent years (e.g., 1988–1993, 2000–2002; Figure S1 of the auxiliary material). The reason behind this phenomenon is not clear, but it may relate to the effect of the North Atlantic Oscillation/Arctic Oscillation [*Hurrell*, 1995; *Thompson and Wallace*, 1998].

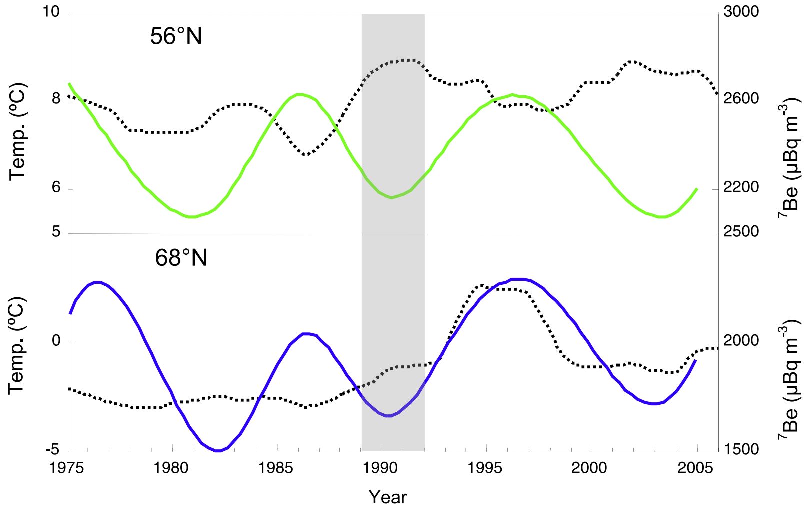

Figure 4. Temperature data (black dotted line) versus ^{7}Be at 56°N (green line) and 68°N (blue line). A relative increase in average temperature in the period 1989–1992 (gray band) is associated with an absence of intrusions in the ^{7}Be record and a positive NAO (Figure S1).

Penetration of stratosphere/upper troposphere air masses are markedly reduced during NAO/AO strong pressure gradients (positive index) at high latitudes [*Baldwin and Dunkerton*, 2001].

[7] Presently we have no direct information about the strength of the intrusions, but further indication about the source can be gained from ^{10}Be/^{7}Be values (Figure 1). Our weekly results show that >90% of the ^{10}Be/^{7}Be values lie between 1 and 3, and most of the ratios outside this range occur at latitude 68°N (Figure 1c). The dominating part of the ^{10}Be/^{7}Be values, which occurs within the range of 1–3, is expected to represent various degrees of mixing within the troposphere and between the tropospheric and stratospheric air masses, due to the very different decay rates of ^{10}Be and ^{7}Be. The source of high ratios (>3) likely relates to relatively aged air masses enriched in ^{10}Be which can be attributed to the stratosphere, as suggested by the direct measurements of Be-isotopes [*Jordan et al.*, 2003] and stratospheric observations [*Baldwin and Dunkerton*, 2001]. The source of low ratios (<1) is here related to relatively freshly produced Be-isotopes that are of tropospheric origin and represent impact of the polar front. This explanation relies on a theoretical atmospheric ^{10}Be/^{7}Be production of 0.5, which may be best preserved in the polar region, due to less troposheric modifications compared to middle latitudes, as also supported by the more frequent occurrence at latitude 68°N compared to 56°N. The mean ^{10}Be/^{7}Be values, in a few days atmospheric measurement [*Jordan et al.*, 2003] and a one-year high altitude daily surface measurement [*Priller et al.*, 2004] (Figure 1e), lie within the 1–3 range shown by our data. This range of ratios is above the theoretical estimation (0.5) and suggests that the empirical values are strongly modified due to the fact that ^{7}Be decays much faster (a few months) compared to ^{10}Be, which may lead to the commonly observed enhancement in the ^{10}Be/^{7}Be measured values.

[8] Although not constant, the average difference in past solar total irradiance between low and high activity in the 11-year cycle is about 0.1% based on ^{10}Be data, which can be translated to an average global forcing difference of about 0.25 Wm^{2} [*Fröhlich*, 2006; *Beer et al.*, 2006]. Solar variability modification on beryllium isotopes production is expected to be stronger at high latitudes (>50°N), where the production rate is high [*Masarik and Beer*, 1999] and atmospheric mixing less effective The intrusions we have observed add a further 10–20% variation to reconstructions of past solar irradiance. The direct effect on past global surface temperature by minor irradiation variations may be insignificant, but amplifications by albedo effects on insolation due to changes in ice and cloud cover and stratospheric ozone are still not well-quantified parameters. We did find some indications of a connection between intrusion frequency and surface air temperature at the studied latitudes. Intrusion-free periods apparently show elevated average temperatures compared to periods with frequent intrusions (Figures 4 and S1). This discernable signal may offer further opportunities to model minute effects of stratospheric/tropospheric air intrusions on surface air temperature.

[9] Translating the link between aerosol and ice-core ^{10}Be is certainly not straightforward. A variety of mathematical techniques were used in order to remove secondary noise in raw data of ^{10}Be from ice cores and to provide a visible fit between ^{10}Be, sunspot number and cosmic ray intensity [*Beer et al.*, 1990; *Muscheler et al.*, 2007]. It was also difficult to link the source and estimate partial effects of the "noise" in the ^{10}Be ice-core records. To obtain some feeling about the aerosols-precipitation connection, we have analyzed 6 samples of precipitation (rain and snow) collected at different times through the year in Sweden. The results show ^{10}Be concentrations of $(0.8–2.6) \times 10^{4}$ atoms g^{-1}, which lie within the ranges obtained in ice cores from Greenland. Furthermore, a theoretically derived estimate of global ^{10}Be flux [*Field et al.*, 2006] suggests approximately comparable fallout for both Greenland and our investigated sites, particularly for the 68°N. Accordingly, with the results obtained here, stratospheric/upper tropospheric intrusions can be a major source of noise in ice core

^{10}Be-data. Additionally, the relatively higher altitudes in Greenland may intensify the atmospheric effect on the ^{10}Be production signals, through enhanced interaction between the stratosphere polar vortex and troposphere [*Baldwin and Dunkerton*, 2001]. These effects, which alter the production signal, should be quantified or eliminated before accurate estimates of past solar irradiance variations can be made. The ultra-sensitive Be isotopes provide an excellent tool for detection of minute changes in the atmosphere, that are otherwise not exposed by conventional atmospheric indicators (e.g., O_3, N_2O, ClO, HCN). Such a tool can further improve our understanding of stratosphere-troposphere perturbations and their effect on climate, which may not have an explicit signature in the Earth's surface temperature records.

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