JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 113, D11104, doi:10.1029/2007JD009160, 2008

Beryllium-10 and beryllium-7 in precipitation in Dübendorf (440 m) and at Jungfraujoch (3580 m), Switzerland (1998–2005)

U. Heikkilä,[1] J. Beer,[1] and V. Alfimov[2]

Received 12 July 2007; revised 4 January 2008; accepted 26 February 2008; published 5 June 2008.

[1] Beryllium-10 and beryllium-7 have been measured in monthly precipitation samples at a high (Jungfraujoch, 46.32°N 7.59°E, 3580 m) and a low (Dübendorf, 47.25°N 8.27°E, 440 m) altitude station in Switzerland from 1998 till today. The average deposition fluxes of ^{7}Be are 380 ± 30 atoms m^{-2} s^{-1} in Dübendorf and 320 ± 20 atoms m^{-2} s^{-1} at Jungfraujoch, and the fluxes of ^{10}Be are 810 ± 70 atoms m^{-2} s^{-1} in Dübendorf and 810 ± 60 atoms m^{-2} s^{-1} at Jungfraujoch. The ^{10}Be/^{7}Be ratio measured at Jungfraujoch (2.4 ± 0.1) is higher than in Dübendorf (2.1 ± 0.1), which is probably caused by a greater share of stratospheric air at Jungfraujoch in which the ^{7}Be content is reduced because of radioactive decay. The ratios correspond to a mean atmospheric residence time of ~110–120 d. The deposition fluxes show a seasonal change with a summer maximum and a winter minimum. The ^{10}Be/^{7}Be ratio exhibits a maximum in the first half of the year at Jungfraujoch in accordance with seasonal stratosphere-troposphere exchange (STE) variations. In Dübendorf the maximum occurs 1 month later. An analysis with a two-box model shows that the calculated variability due to seasonally varying STE explains only one third of the observed variability in the ^{10}Be/^{7}Be ratio. The rest of the variability is caused by local effects, such as the precipitation rate and the larger than average STE variability at the midlatitudes. According to the model calculations, the global mean ^{10}Be/^{7}Be ratio only varies between 1.1 and 1.5 with a seasonally varying STE. The larger measured values in the midlatitudes point to a latitudinal dependence of the ^{10}Be/^{7}Be ratio.

Citation: Heikkilä, U., J. Beer, and V. Alfimov (2008), Beryllium-10 and beryllium-7 in precipitation in Dübendorf (440 m) and at Jungfraujoch (3580 m), Switzerland (1998–2005), *J. Geophys. Res.*, *113*, D11104, doi:10.1029/2007JD009160.

1. Introduction

[2] Beryllium-10 and beryllium-7 have been repeatedly measured in precipitation samples since 1998 at two different stations in Switzerland, Dübendorf (47.25°N, 8.27°E, elevation 440 m above sea level (asl)) and Jungfraujoch (46.32°N, 7.59°E, elevation 3580 m asl). The Dübendorf station is located in the planetary boundary layer, whereas most of the time the Jungfraujoch station is in the free troposphere. In this work the results from the beginning of the measurement program (year 1998) till the delayed present (year 2005) are presented (~8 years) with a monthly resolution.

[3] Beryllium-10 (half-life ~1.5 × 10^{6} years, see *Nishiizumi et al.* [2007] for reassessment of the half-life) and ^{7}Be (half-life 53.2 d) are natural radionuclides which are produced in the atmosphere through the interaction of cosmic rays with the atmospheric N and O atoms. The dependence of the production rate on the altitude and latitude is similar for both radionuclides, but on a global average the ^{10}Be/^{7}Be ratio is 0.5 (see Figure 1) [*Masarik and Beer*, 1999]. Because of the very different half-lives of the two radionuclides, comparison of the two provides an interesting tool for the study of the atmospheric transport processes, especially stratosphere-troposphere exchange (STE).

[4] The maximum production rate of ^{7}Be and ^{10}Be occurs in the lower stratosphere at an altitude of ~200 hPa (10–13 km). The production gradient is also strong at this altitude. Therefore, small changes in the tropopause height or in STE may cause variations in the deposition fluxes measured at Jungfraujoch. The Dübendorf station is assumed to show these changes less directly because of the further distance from the stratosphere.

[5] When modeling the atmospheric transport of these radionuclides with general circulation models (GCMs) [*Brost et al.*, 1991; *Field et al.*, 2006; *Koch et al.*, 1996; *Land and Feichter*, 2003; *Liu et al.*, 2001], it has turned out that the STE as well as the wet scavenging are uncertain factors in the GCMs and need to be more adequately understood. There is a need for deposition flux observations to improve the wet scavenging routines of the models. A number of ^{7}Be deposition observations exist [e.g., *Baskaran et al.*, 1993; *Bleichrodt*, 1978; *Brown et al.*, 1988; *Crecelius*, 1981; *Dibb*, 1989; *Graham et al.*, 2003; *Hasebe et al.*, 1981; *Huh et al.*, 2006; *Ishikawa et al.*, 1995; *Olsen et al.*, 1985; *Papastefanou et al.*, 1995; *Todd et al.*, 1989; *Turekian et al.*,

[1] Swiss Federal Institute of Aquatic Science and Technology, Dübendorf, Switzerland.
[2] Ion Beam Physics, Paul Scherrer Institute and ETH Zurich, Zurich, Switzerland.

0148-0227/08/2007JD009160$09.00

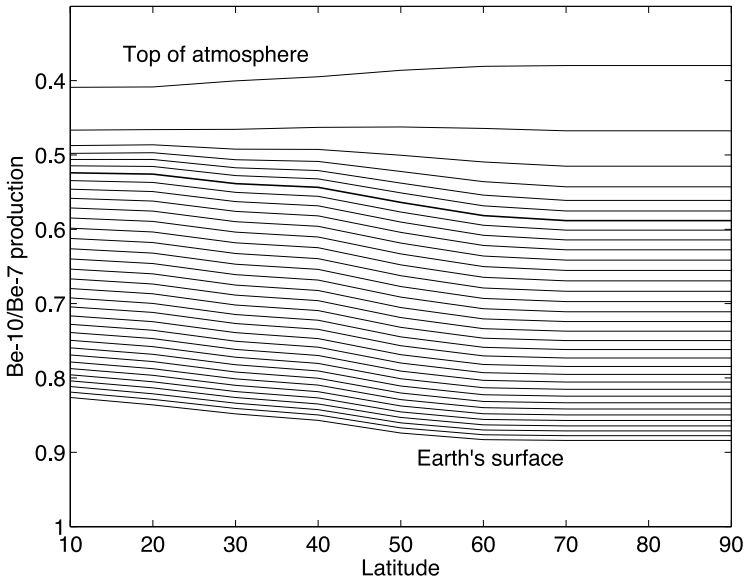

Figure 1. The $^{10}Be/^{7}Be$ production ratio as a function of latitude and altitude calculated by *Masarik and Beer* [1999]. The production ratio is calculated for the present geomagnetic field strength and the long-term mean solar activity of Φ = 550 MeV. The different lines indicate different atmospheric depths from 30 g cm^{-2} (top line, highest altitude) to 1020 g cm^{-2} (bottom line, Earth's surface) with steps of 30 g cm^{-2}. The thicker line shows the production ratio at 210 g cm^{-2}, which is approximately the height of the tropopause. The global average ratio of $^{10}Be/^{7}Be$ production is 0.5 when weighted with the surface area of the Earth, which significantly lowers the contribution of production at 60–90°.

1983; *Uematsu et al.*, 1994; *Young and Silker*, 1980], but many of them cover only a period of less than 5 years, and they are not evenly distributed over the globe. The year-to-year variation of deposition is large due to production changes as well as changes in the local precipitation rate. To smooth out the precipitation fluctuations, observational periods of a few years are preferable. The ^{10}Be deposition fluxes have rarely been observed, especially in combination with ^{7}Be. *Brown et al.* [1988] measured ^{10}Be and ^{7}Be deposition fluxes in Hawaii, Illinois, and New Jersey (1986–1994) and estimated a mean ratio of $^{10}Be/^{7}Be$ of the deposition fluxes of 1.25. Also *Monaghan et al.* [1985/1986], *Baskaran et al.* [1993], and *Knies* [1994] have measured ^{10}Be and ^{7}Be deposition fluxes at three stations in the United States. The ratios measured were 1.7 at College Station, Texas, 1.8 in Indiana, and 2.2 in New Haven, Connecticut. *Graham et al.* [2003] measured ^{10}Be and ^{7}Be at three stations in New Zealand from 1996 to 1998. They found mean $^{10}Be/^{7}Be$ ratios from 1.4 to 1.5 on the northern island and 2.0 on the southern island. They also found a seasonal variation in the ratio measured, with higher values in the winter, reflecting STE.

[6] Moreover, comparing two stations situated close to each other but at different elevations provides information about the STE. That is useful to evaluate the vertical transport processes in models. The ratio of $^{10}Be/^{7}Be$ has been used for this purpose in model studies by *Koch and Rind* [1998] and *Rehfeld and Heimann* [1995].

2. Methods

[7] The precipitation samples were collected on the roof of the Swiss Federal Institute of Aquatic Science and Technology (EAWAG) building in Dübendorf. A cylindric funnel was used with a diameter of 16 cm from 1998 till December 1999, of 20.5 cm till March 2000 and of 24 cm since April 2000.

[8] The precipitation samples from Jungfraujoch were kindly delivered to us by the staff of the observatory. The collector used at Jungfraujoch is cylindric with a diameter

Table 1. Sampling Periods and the Measured Mean Concentrations in the Rainwater and the Deposition Fluxes at Both Stations[a]

	Sampling Period, years		Average Concentrations	
	Dübendorf	Jungfraujoch	Dübendorf	Jungfraujoch
^{7}Be concentration, 10^4 atoms g^{-1}	Mar 1998 to Jun 2005	Jan 1998 to Jul 2005	1.2 ± 0.1	0.8 ± 0.1
^{10}Be concentration, 10^4 atoms g^{-1}	Jan 1998 to Nov 2004	Jan 1998 to Dec 2004	2.8 ± 0.2	2.1 ± 0.1
^{7}Be flux, atoms m^{-2} s^{-1}	Mar 1998 to Jun 2005	Jan 1998 to Jul 2005	380 ± 30	320 ± 20
^{10}Be flux, atoms m^{-2} s^{-1}	Jan 1998 to Nov 2004	Jan 1998 to Dec 2004	810 ± 70	810 ± 60
Precipitation, mm d^{-1}	Jan 1998 to Jul 2005	Jan 1998 to Jul 2005	2.6 ± 0.2	3.5 ± 0.2
$^{10}Be/^{7}Be$	Jan 1998 to Nov 2004	Jan 1998 to Dec 2004	2.1 ± 0.1	2.4[b] ± 0.1

[a]The given uncertainties are 1σ standard errors of the mean.
[b]Calculated without the extreme values of March 2001 and April 2002.

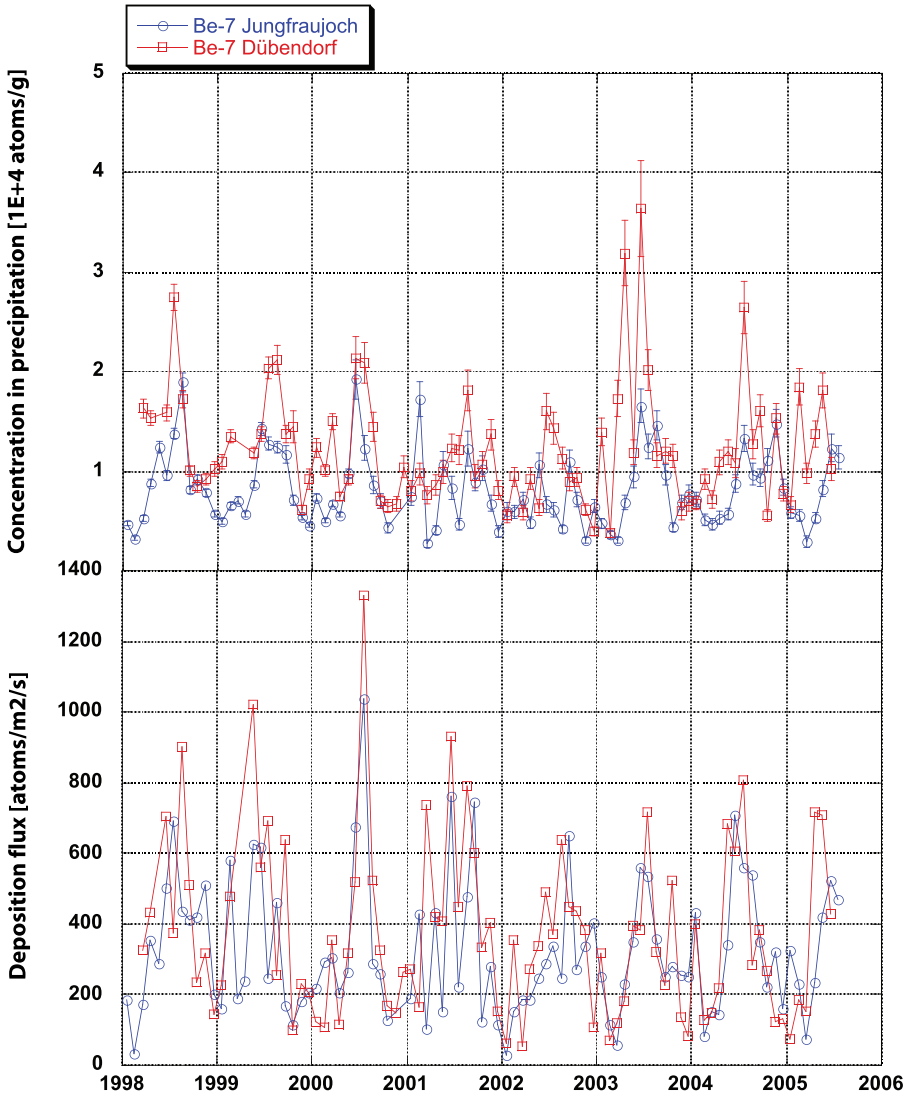

Figure 2. The measured ^{7}Be (top) concentrations in precipitation with the measurement 2σ standard deviations and (bottom) deposition fluxes in Dübendorf (red squares) and at Jungfraujoch (blue circles).

of 23.5 cm. Since precipitation at Jungfraujoch consists mainly of snow it is collected daily and accumulated in an indoor container until the end of the month. 0.3 mg of ^{9}Be carrier was added in the samples. All of the ^{7}Be samples are measured at EAWAG using a Ge-gamma spectrometer, which has been calibrated. The ^{10}Be samples are prepared at EAWAG and measured at the Accelerator Mass Spectrometer (AMS) facility of ETH/PSI. The results were normalized to an in-house standard with a nominal value of ^{10}Be/^{9}Be = 95.5 $\times$ 10^{-12}. It is a secondary standard normalized to the material BEST433 to determine the ^{10}Be half-life of 1.51 $\times$ 10^{6} years [*Hofmann et al.*, 1987].

[9] The monthly sampling period is not exactly the same in Dübendorf and at Jungfraujoch, with deviations of up to ±5 d. The amount of precipitation was between 0.5 and 10 kg month^{-1}. Errors due to evaporation are not taken into account. However, the amount of precipitation measured agrees generally well with precipitation data in Zürich measured by the Swiss Federal Office of Meteorology and Climatology (MeteoSwiss). The ^{7}Be concentrations measured are decay corrected to the last day of the respective month.

[10] The collectors used are open during the whole sampling period. In this way, both wet and dry deposition is collected. This can lead to slightly enhanced ^{10}Be concentrations, caused by recycled dust from the Earth's surface [*Baumgartner et al.*, 1997; *Brown et al.*, 1988; *Monaghan et al.*, 1985/1986]. ^{7}Be can be assumed to be decayed in the dust because of its short half-life and, therefore, it does not contribute to the ^{7}Be concentrations measured. The contribution of recycled ^{10}Be in dust is highly variable depending on the location of the measurement site and the amount of precipitation during the measurement period. During dry months the contribution of recycled ^{10}Be is larger. The samples from Dübendorf and Jungfraujoch were filtered using a coarse filter (>50 micron) to remove the largest impurities. This filtering, however, was not efficient enough to remove the dust. This issue is further discussed in section 3.2.

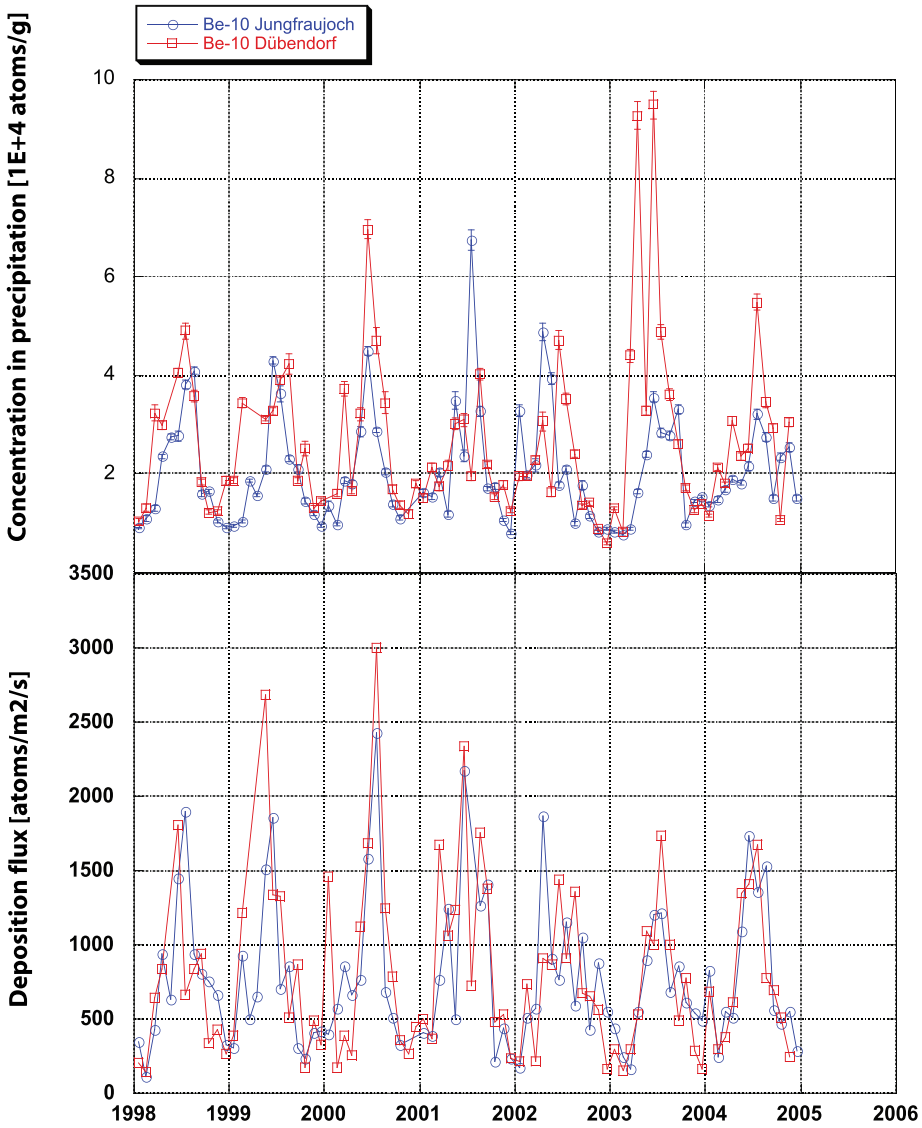

Figure 3. Same as Figure 2 but for ^{10}Be. The error bars of the concentrations indicate the measurement 1σ standard deviations.

[11] A summary of the data used for the analysis is presented in Table 1. The whole data set is available from ftp.eawag.ch/pub/eawag/Beryllium_DD_JJ.

3. Results

3.1. Concentrations and Fluxes

[12] The concentrations measured in rainwater and deposition fluxes of ^{7}Be (Figure 2) and ^{10}Be (Figure 3) in Dübendorf and at Jungfraujoch are shown with the 1σ (^{10}Be) and 2σ (^{7}Be) standard deviations. The deposition fluxes were calculated from the concentrations using the amount of rain measured. The radionuclide concentrations measured are generally higher in Dübendorf than at Jungfraujoch. A possible reason for this is the higher precipitation rate at Jungfraujoch (3.5 ± 0.2 vs. 2.6 ± 0.2 mm d^{-1}), which leads to a dilution of the radionuclide concentrations in rainwater. At Jungfraujoch a variable amount of the precipitation was lost because of nonvertical snowfall caused by strong winds. There are some extremely high values in the rainwater concentrations measured. The fact that the corresponding fluxes do not stand out indicates that these extreme values are due to low precipitation.

[13] The average ^{7}Be fluxes measured are 380 ± 30 atoms m^{-12} s^{-1} in Dübendorf and 320 ± 20 atoms m^{-2} s^{-1} at Jungfraujoch. For ^{10}Be they are 810 ± 70 atoms m^{-2} s^{-1} in Dübendorf and 810 ± 60 atoms m^{-2} s^{-1} at Jungfraujoch. These values (from ~47°N) compare quite well with mean fluxes found on the northern island of New Zealand (~38°S): ~480 atoms m^{-2} s^{-1} (^{7}Be) and ~860 atoms m^{-12} s^{-1} (^{10}Be) and also with the somewhat lower measurements on the southern island of New Zealand (~44°S): ~290 atoms m^{-2} s^{-1} (^{7}Be) and ~600 atoms m^{-2} s^{-1} (^{10}Be) [*Graham et al.*, 2003].

3.2. The ^{10}Be/^{7}Be Ratio

[14] The ^{10}Be/^{7}Be ratio is shown in Figure 4. The horizontal lines show the global mean stratospheric (5) and tropospheric ratios (1.5) determined by a two-box

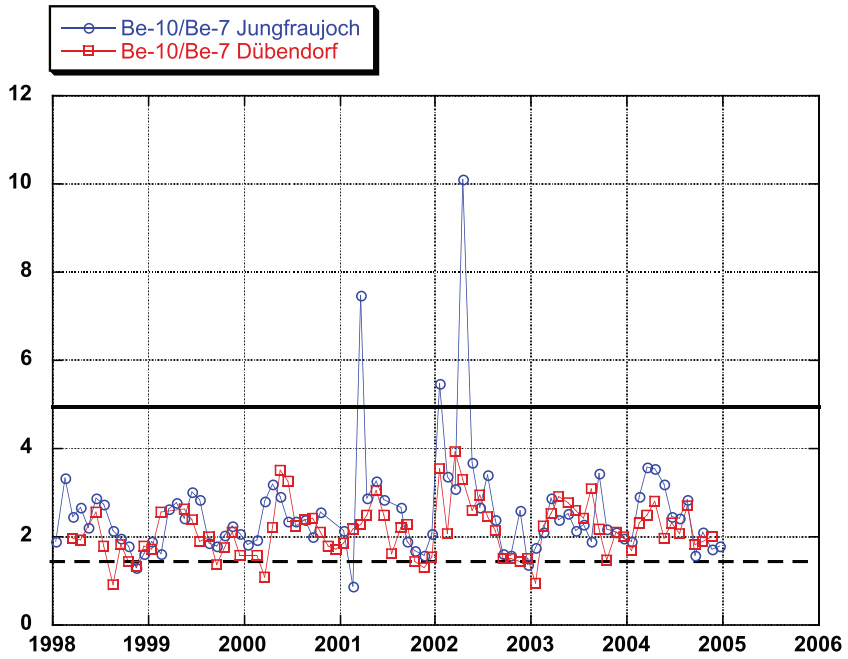

Figure 4. Measured $^{10}Be/^{7}Be$ ratio in Dübendorf (red) and at Jungfraujoch (blue). The horizontal dashed lines indicate the average $^{10}Be/^{7}Be$ ratios in the stratosphere (solid line) and the troposphere (dashed line). The lowest values ($\sim$1) are still above the average production ratio of 0.5.

model (see section 3.5). Even at the low-altitude station, Dübendorf, the $^{10}Be/^{7}Be$ ratios are generally higher than the tropospheric average ratio. The reason for this is that the mean ratio is a global average, whereas the two measurement stations of this study are located in the midlatitudes, where a higher fraction of stratospheric air is expected because of stronger STE [*Holton et al.*, 1995]. The measured $^{10}Be/^{7}Be$ ratios are similar or a bit higher at Jungfraujoch than in Dübendorf. The variability at Jungfraujoch is greater. There are some extreme values even higher than the global mean stratospheric $^{10}Be/^{7}Be$ ratio of 5, which all occur at Jungfraujoch during the winter and spring months. Double checking of the data gave no indication of analytical problems. Extremely high $^{10}Be/^{7}Be$ values could be caused by very old air or dry months, during which the ^{10}Be contamination from dust is higher. However, during March 2001, January 2002, and April 2002, the precipitation measured was not especially low. These extreme values are caused by a combination of rather high ^{10}Be and of lower than normal ^{7}Be. Low ^{7}Be fluxes (<200 atoms m^{-2} s^{-1}), which are observed during winter (see fluxes in Figure 2), point to old air. In the following analysis the effect of these extreme values on the results is evaluated.

[15] The mean values for ^{10}Be and ^{7}Be are also shown in Table 1. The ^{10}Be concentrations and fluxes are approximately 2.1 times higher than those of ^{7}Be in Dübendorf and 2.4 times higher at Jungfraujoch. The higher ratio measured at Jungfraujoch is probably caused by a greater fraction of stratospheric air at the station. It can be assumed that the $^{10}Be/^{7}Be$ ratio reflects the atmospheric ratio when they are averaged over the whole measurement time (8 years). A $^{10}Be/^{7}Be$ ratio of 2.1–2.4 gives a mean residence time of approximately 110–120 d for the radionuclides as an average for the whole atmosphere. *Graham et al.* [2003] calculated an average residence time of 80 to 100 d assuming an average production ratio of 0.53 according to *Masarik and Beer* [1999].

[16] The $^{10}Be/^{7}Be$ ratio of 2.1 in Dübendorf agrees well with other observations (1.3–2.2) [*Baskaran et al.*, 1993; *Brown et al.*, 1988; *Graham et al.*, 2003; *Knies*, 1994; *Monaghan et al.*, 1985/1986]. The $^{10}Be/^{7}Be$ ratio at Jungfraujoch of 2.4 cannot be compared directly with these ratios because Jungfraujoch is the only high-altitude station. Instead, we compared the ratio with ratios measured in air filters at Jungfraujoch and at Zugspitze (2962 m asl), Germany, from March 2000 till February 2001 [*Zanis et al.*, 2003]. At Jungfraujoch, the variability of the ratio measured in the air filters is similar to the one measured in precipitation during the respective months. The ratio varies from 1.0 (0.9) to 3.7 (3.2), with an average of 2.1 (2.3) measured in air filters (precipitation). The ratio measured in surface air is at a maximum from March to June, lasting slightly longer than the maximum measured in precipitation (March, April, and May). At Zugspitze the ratio varies less with an average of 1.8 and exhibits a maximum in March–April and a second one in June–July.

[17] The measured $^{10}Be/^{7}Be$ ratio of 2.1–2.4 is slightly increased because of recycled ^{10}Be from dust. However, it is impossible to quantify this contribution without measuring the dust separately. *Monaghan et al.* [1985/1986] determined the amount of dust in their ^{10}Be measurements in precipitation at seven sites in the United States. The average amount of recycled ^{10}Be varied from 8% to 35% between the sites. *Brown et al.* [1988] also investigated this issue concluding that it is extremely difficult to quantify the amount of recycled ^{10}Be from the data. *Graham et al.* [2003] performed dust capture experiments and found a contribution of recycled ^{10}Be of 10%, 11%, and 8% at the

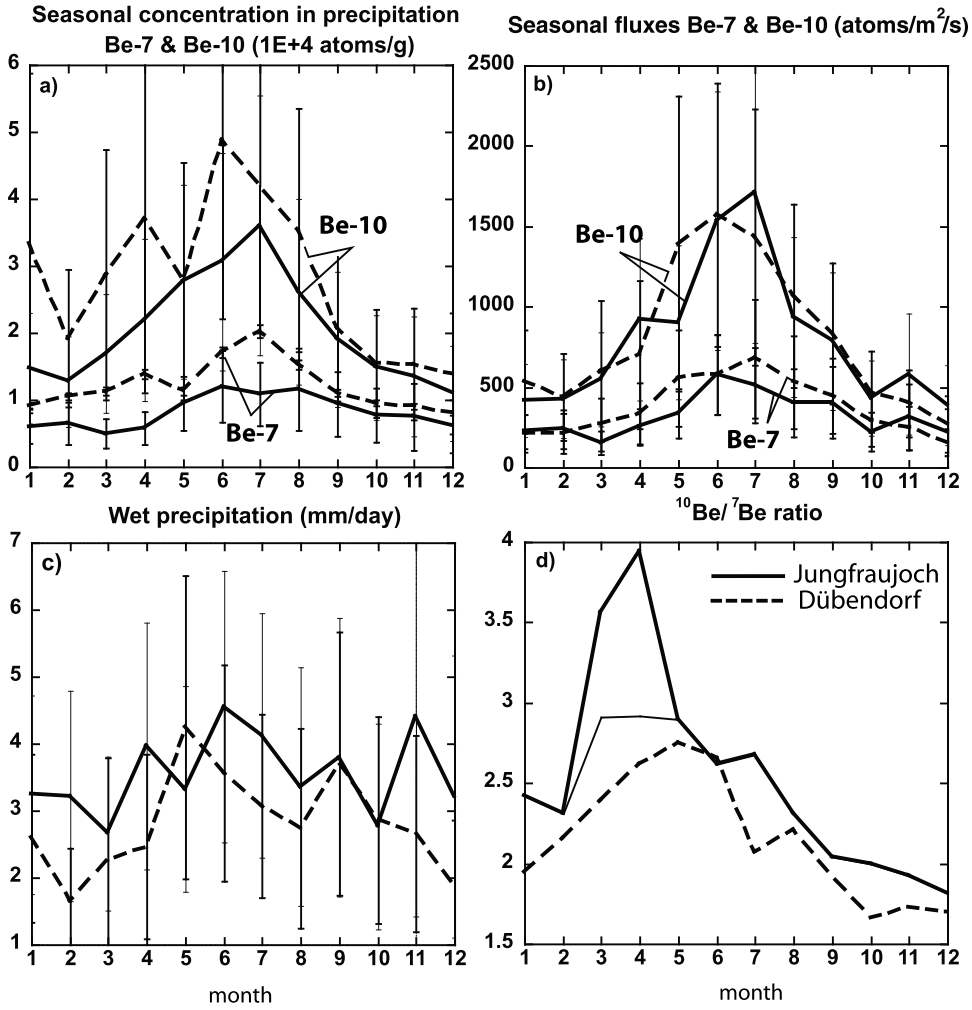

Figure 5. (a) The seasonality of the ^{10}Be (two upper curves) and ^{7}Be (two bottom curves) concentrations measured in precipitation. (b) Deposition fluxes in Dübendorf (dashed line) and at Jungfraujoch (solid line). (c) The measured precipitation. (d) The ^{10}Be/^{7}Be ratio in Dübendorf (dashed line) and at Jungfraujoch (solid line). The thinner solid line shows the ^{10}Be/^{7}Be ratio without the extreme ratios of March 2001 and April 2002.

three New Zealand sites. The ^{10}Be/^{7}Be ratios varied from 1.6 to 2.1, with the correction from 1.4 to 2. *Baumgartner et al.* [1997] estimated the contribution of recycled ^{10}Be in the GRIP ice core during the Holocene at less than 5%. However, the central Greenland area, which is covered by glaciers, is far away from any dust sources. The ^{10}Be/^{7}Be ratio which we measured would be 1.7–1.9 in Dübendorf and 1.9–2.1 at Jungfraujoch, assuming a dust contribution of 10–20%. Since it is not possible to correct our ^{10}Be/^{7}Be ratio, we will continue working with the ratios of 2.1 (Dübendorf) and 2.4 (Jungfraujoch), bearing in mind that this represents an upper limit.

3.3. Correlation With Cosmic Rays

[18] The production rate of ^{7}Be and ^{10}Be is modulated by the solar activity as well as the intensity of the geomagnetic field. Both of these variations are captured by the neutron monitors measuring the cosmic rays entering the atmosphere. To establish whether a correlation between the fluxes measured and cosmic rays exists, we used cosmic ray intensity data measured at the Sodankylä Geophysical Observatory, Finland. These data are available at http://cosmicrays.oulu.fi.

[19] The data are smoothed with a 13-month low-pass Fourier filter to remove the seasonality. The correlation between the neutron monitor data and the ^{7}Be or ^{10}Be fluxes or concentrations is positive, with correlation coefficients ranging from 0.27 to 0.48. The reason for the rather weak correlation is that the radionuclides were measured at only two single stations, and the local effects are much stronger than the production changes. After the production the radionuclide concentrations are influenced by the STE, tropospheric transport and the local precipitation rate at the station. Moreover, the measurement time (8 years) is short compared to an average solar (Schwabe) cycle of 11 years. A stronger correlation (0.72) between ^{7}Be surface air concentration and sunspots was found in Palermo, southern Italy, during the period from 1982 to 2002 [*Cannizzaro et al.*, 2004]. The fact that the correlation between solar activity and ^{7}Be surface air concentrations is higher than

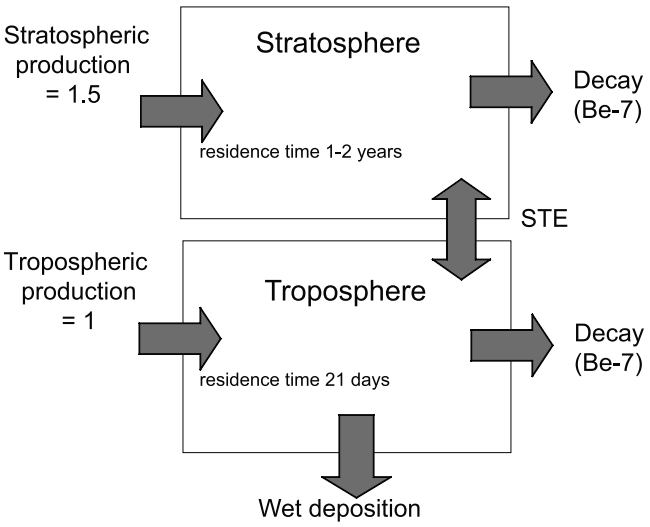

Figure 6. The two-box model used to simulate the exchange processes in the atmosphere. Beryllium-10 and -7 are produced in the stratosphere and in the troposphere. As the averaged $^{10}Be/^{7}Be$ production ratio does not significantly vary between stratosphere (∼0.45) and troposphere (∼0.5–0.6) (see Figure 1 or *Lal and Peters* [1967] and *Nagai et al.* [2000]), it is set to 0.5 in both boxes. The ratio between stratospheric and tropospheric production varies between 1:1 [*Masarik and Beer*, 1999; *Nagai et al.*, 2000] and 2:1 [*Lal and Peters*, 1967]. Therefore, both contributions of 1 (stratospheric) to 1 (tropospheric) and 1.5 (stratospheric) to 1 (tropospheric), which is an average of both estimates, are used. The residence time of the radionuclides in the stratosphere is 1–2 years. To test the sensitivity, both 1 and 2 years are used. The tropospheric residence time used is 21 d without considering the decay of ^{7}Be. *Bleichrodt* [1978] estimated a residence time of 22–35 d for ^{7}Be using atmospheric concentrations and deposition fluxes measured between 30° and 75°. Model studies calculate tropospheric residence times of approximately 21 d, when only deposition is considered [*Koch et al.*, 1996; *Liu et al.*, 2001]. Beryllium-7 decays in both boxes with a half-life of 53.2 d.

the correlation between solar activity and ^{7}Be deposition fluxes again indicates that the local precipitation rate at the measurement site causes strong variability on the radionuclide deposition fluxes.

3.4. Seasonality of the Deposition Fluxes

[20] Seasonal cycles of the concentrations in rainwater, deposition fluxes and wet precipitation are shown in Figure 5. The error bars are the standard errors of the mean and show that the year-to-year variations are large. The precipitation varies strongly at both stations, but has a seasonality with a maximum in the summer months. The amount of precipitation is about 1.5 times higher at Jungfraujoch than in Dübendorf, whereas the concentrations measured in rainwater are approximately 1.5 times higher in Dübendorf than at Jungfraujoch, leading to equal fluxes.

[21] The concentrations peak during the months from June to August. The deposition fluxes of both radionuclides are of the same order of magnitude and have a very similar seasonal variation. They peak during June and July, when also the precipitation is high.

[22] Figure 5 also illustrates the seasonal $^{10}Be/^{7}Be$ ratio in Dübendorf and at Jungfraujoch. The seasonal cycle of $^{10}Be/^{7}Be$ varies between 1.5 and 3–3.5 at Jungfraujoch and 1.5 and 3 in Dübendorf. The difference between Jungfraujoch and Dübendorf is not large, but on the average the ratio at Jungfraujoch is higher during the whole year. The very high ratios in March and April at Jungfraujoch are caused by single extreme values (March 2001 and April 2002). Ignoring these values leads to much lower ratios in March and April (thin solid line in Figure 5d). Still, the asymmetry of the seasonality remains, showing a maximum in the first half of the year and a minimum in the second half of the year. This is in agreement with *Holton et al.* [1995] as the intrusions of stratospheric air into the troposphere are strongest in the late winter and spring in the northern hemisphere. The $^{10}Be/^{7}Be$ ratio in Dübendorf has a maximum between April and June. Considering the high year-to-year variability it is difficult to determine if the $^{10}Be/^{7}Be$ ratio in Dübendorf shows STE variability. At Jungfraujoch the maximum in spring can probably be connected to stratospheric air.

[23] The seasonal variability of the $^{10}Be/^{7}Be$ ratio measured in this study seems to be higher (∼50%) (Figures 4 and 5d) than the variability observed by *Graham et al.* [2003]. This could be related to the fact that stratospheric intrusions are stronger in the northern hemisphere [*Holton et al.*, 1995].

3.5. Comparison of $^{10}Be/^{7}Be$ With Two-Box Model

[24] The ^{10}Be and ^{7}Be deposition fluxes are known to have a strong latitudinal and also a longitudinal dependence [e.g., *Field et al.*, 2006; *Heikkilä et al.*, 2008]. The fluxes can be up to a factor of 4 larger at the midlatitudes than in polar regions, because of the large precipitation rate at the midlatitudes and the maximum in the stratospheric intrusions in the subtropics. Comparing the observed ratios with a global average ratio obtained using a simple two-box atmospheric exchange model gives us a measure of this latitudinal dependence. Figure 6 illustrates the model and the parameters used. Because of the simple character of this box model and the few required parameters we can assume that because of mass conservation, the results are accurate as a global mean. Moreover, this box model also allows us to theoretically investigate how the ratio changes as a function of the 11-year solar cycle or the STE, which has a seasonal variability.

[25] The simulated ratio of the $^{10}Be/^{7}Be$ deposition fluxes varies between 1.1 and 1.4 when varying the stratospheric residence time between 1 and 2 years and the ratio of stratospheric to tropospheric production between 1 and 1.5. The $^{10}Be/^{7}Be$ ratio of 2.1–2.4 measured at midlatitudes is higher than the global average pointing to a strong latitudinal dependence due to strong STE at midlatitudes. This is confirmed by other observed ratios from the subtropics and midlatitudes, which are also higher than those simulated by the two-box model, but slightly lower than ours (1.7–2.2 by *Monaghan et al.* [1985/1986], *Baskaran et al.* [1993], and *Knies* [1994], 1.3 by *Brown et al.* [1988], and 1.4–2.0 by *Graham et al.* [2003]). It also has to be kept in mind that our ratio is not dust corrected.

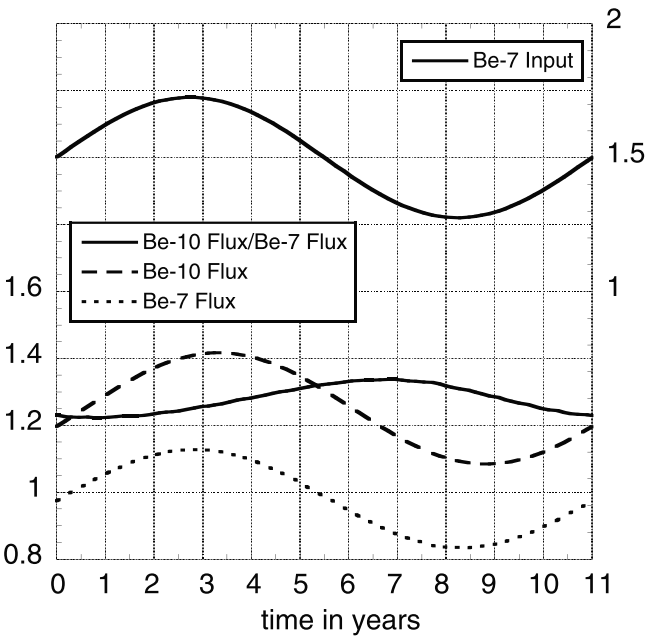

Figure 7. The results of the two-box model when a sinusoidal input function is used (above, in units of production). The ^{7}Be input varies by 15%, but the ^{10}Be/^{7}Be deposition flux varies by only 4–5% (bottom). The ^{10}Be deposition flux lags about 1 year behind the production; the ^{7}Be deposition flux lags only 0.5 year because of decay. As a result of this phase difference, the ^{10}Be/^{7}Be ratio reaches its maximum about 4 years later than production.

[26] To simulate the 11-year Schwabe cycle, a sinusoidal production signal was used. The production varies 30–40% between minimum and maximum during a typical solar cycle [*Masarik and Beer*, 1999]. Therefore, the variation from the average of the sinusoidal production was set to 15% (see Figure 7). Only ^{7}Be input is shown because the production of both radionuclides depends on the solar activity in the same way. The ^{7}Be deposition flux varies as much as the production signal within a solar cycle (15%), whereas for ^{10}Be the variation is slightly less (13%). ^{7}Be reacts more directly to production changes because a large part of it decays and the burden is reduced. The ^{10}Be/^{7}Be ratio of the deposition fluxes varies only 4–5% within a solar cycle. Figure 7 also shows the phases of ^{10}Be and ^{7}Be input and deposition flux variability. Because of the stratospheric residence time of 1 year, the ^{10}Be deposition flux reaches a maximum 1 year later than the production. ^{7}Be deposition flux lags only 0.5 year behind the production because a large part of its stratospheric content decays within the residence time. Therefore, the phases of ^{10}Be and ^{7}Be differ by about 0.5 year. For this reason the ^{10}Be/^{7}Be ratio is almost in antiphase with the production, lagging about half a solar cycle (approximately 4 years) behind the production.

[27] To estimate the seasonal ^{10}Be/^{7}Be variability, the STE (downward fluxes from the stratosphere) was varied sinusoidally so that it has a maximum during the spring (March–April–May) and a minimum in autumn and a 50% deviation from the average [*Appenzeller et al.*, 1996]. Figure 8a shows the downward fluxes from the stratosphere to the troposphere for ^{10}Be and ^{7}Be. The ^{7}Be mass flux varies slightly less (45%) than ^{10}Be mass flux because a part of ^{7}Be has decayed. The ^{10}Be/^{7}Be ratio varies only by 5%. The deposition fluxes shown in Figure 8b vary 27% (^{10}Be), 10% (^{7}Be) and 17% (^{10}Be/^{7}Be ratio). The ^{10}Be/^{7}Be ratio varies between 1.5 (STE maximum in spring) and 1.1 (STE minimum in autumn) on average. The ratios measured from our study as well as from the earlier studies (1.3–2.2) tend to be higher than these values, which seems to point to a latitudinal dependence of the ^{10}Be/^{7}Be ratio. STE is strongest at the midlatitudes, bringing down air with high ^{10}Be/^{7}Be ratios. The global average ratio is probably lowered by low ratios in the tropics, where STE is weak and high precipitation scavenges the particles effectively from the atmosphere. There is a need for more observations, both deposition flux and air filter ratios to understand their global distribution.

[28] We learn from the two-box model that although the solar activity affects the production of ^{10}Be and ^{7}Be in the same way, the decay of ^{7}Be causes a phase lag of almost half a solar cycle in the measured deposition flux ^{10}Be/^{7}Be ratio. However, this variability is strongly damped (only 4–5%) and can hardly be observed. This is in accordance with the measured ratio, as no trend can be observed in Figure 4. The seasonally varying STE is visible in the ^{10}Be/^{7}Be ratio, causing a maximum in the spring, which can also be observed in the measured ratios. Figure 5d shows that the measured seasonal variability of the ^{10}Be/^{7}Be ratio from the average is about 50% at both stations. This measured variability, including the 11-year and the STE variability, is much higher than the modeled variability (17%) obtained with the two-box model. This means that production or STE changes explain only approximately one third of the deposition flux variability. Factors such as the local precipitation rate and atmospheric transport are the reason for the larger observed variability. Furthermore, the STE variability in the midlatitudes is probably larger than the global average variability determined by the two-box model.

4. Summary and Conclusions

[29] Beryllium-7 and beryllium-10 have been measured monthly in precipitation at low- and high-altitude stations in Switzerland, Dübendorf (440 m) and Jungfraujoch (3580 m) from 1998 till 2005. The deposition fluxes of the two radionuclides and their ratio at the two stations were analyzed with respect to stratosphere-troposphere exchange (STE).

[30] The average deposition fluxes measured are 380 ± 30 atoms m^{-12} s^{-1} in Dübendorf and 320 ± 20 atoms m^{-2} s^{-1} at Jungfraujoch (^{7}Be) and 810 ± 70 atoms m^{-2} s^{-1} in Dübendorf and 810 ± 60 atoms m^{-2} s^{-1} at Jungfraujoch (^{10}Be). The absolute precipitation is approximately 1.5 times higher at Jungfraujoch (3.5 ± 0.2 mm d^{-1}) than in Dübendorf (2.1 ± 0.2 mm d^{-1}).

[31] The ^{10}Be/^{7}Be ratio measured is approximately 2.1 ± 0.1 in Dübendorf and 2.4 ± 0.1 at Jungfraujoch, which are comparable with other studies from the midlatitudes. As the ^{10}Be/^{7}Be ratio at the time of production is only marginally latitude dependent and varies between 0.4 and 0.9 from 1000 hPa to 30 hPa, the higher ratios measured provide information about STE and atmospheric residence times of

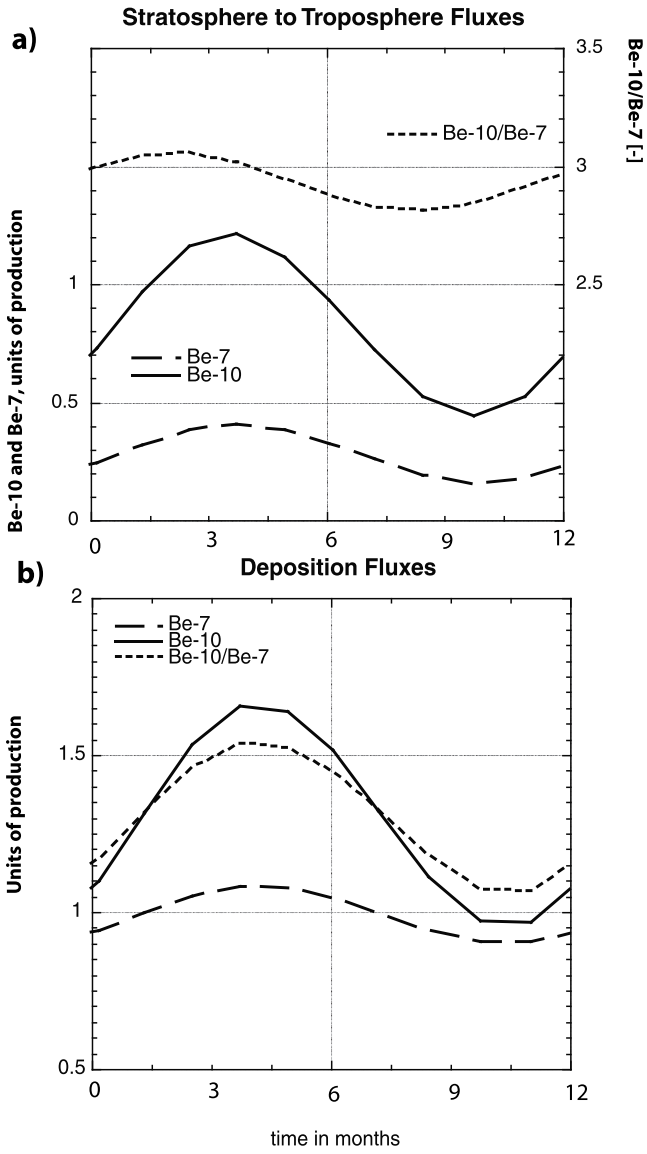

Figure 8. The results of the two-box model for an STE varying sinusoidally by 50% from the average. (a) The downward fluxes from the stratosphere for ^{10}Be (variation 50%), ^{7}Be (45%), and their ratio (5%). (b) The variation of the deposition fluxes (^{10}Be flux 27%, ^{7}Be flux 10%, and their ratio 17%).

the radionuclides. The higher ^{10}Be/^{7}Be ratio measured at Jungfraujoch also probably reflects the higher share of stratospheric air. The ratios include the recycled ^{10}Be from dust, which can contribute up to 10–20% of the concentration. The ratio of 2.1–2.4 corresponds to an average atmospheric residence time of ~110–120 d.

[32] The deposition fluxes exhibit a seasonality, reaching a maximum during the summer months similar to the precipitation rate. The amplitude of the seasonal cycle is in the order of 3–4. The seasonal cycles of ^{10}Be and ^{7}Be are very similar at both stations. The seasonal variation of the ^{10}Be/^{7}Be ratio of the deposition fluxes reaches a maximum during the first half of the year, especially at Jungfraujoch. In Dübendorf the maximum occurs about a month later. The spring maximum at Jungfraujoch provides evidence for STE, which has similar seasonal variability. Still, the year-to-year variations of the deposition fluxes are large.

[33] A simulation with a two-box atmospheric exchange model shows that the production variability during a solar cycle does not cause observable changes in the ^{10}Be/^{7}Be ratio. Indeed, no trend is evident in the measured ratios. Further, the model suggests that the seasonally varying STE causes a larger variability in the ^{10}Be deposition flux than in the ^{7}Be deposition flux, because changes in the ^{7}Be flux are damped by radioactive decay. This is also in accordance with the measured fluxes. The simulated ^{10}Be/^{7}Be ratio reveals an attenuation of the applied STE variation by one third. Moreover, the simulated seasonal variability is one third of the observed seasonal variability. This means that two thirds of the observed variability are caused by local effects, such as the local precipitation rate and tropospheric transport. This, and the fact that the ^{10}Be/^{7}Be ratio is damped suggest that even if the year-to-year variations in the seasonal ^{10}Be/^{7}Be ratios measured are large, the maximum in the spring could be significant and reflect STE.

[34] According to the two-box model, the global average ^{10}Be/^{7}Be ratio varies only little (1.1 to 1.5) with varying production or STE. The measured ratios of ~2.1 (Dübendorf) and ~2.4 (Jungfraujoch) as well as other measurements from the midlatitudes indicate that the ratios have a significant latitudinal dependence. The influence of stronger STE in the midlatitudes is visible in the measured ratios. These findings highlight the interest of using the ^{10}Be/^{7}Be ratio for studying atmospheric processes. More observations from different latitudes and altitudes would improve our understanding of the STE and help improve the global circulation models.

[35] **Acknowledgments.** We are grateful to E. Grieder for measuring ^{7}Be and S. Bollhalder Lück for preparing the ^{10}Be samples for AMS. This work is funded by the Swiss National Science Foundation.

References

Appenzeller, C., J. R. Holton, and K. H. Rosenlof (1996), Seasonal variation of mass transport across the tropopause, *J. Geophys. Res.*, *101*, 15,071–15,078.

Baskaran, M., C. Coleman, and P. Santchi (1993), Atmospheric depositional fluxes of ^{7}Be and ^{210}Pb at Galveston and College Station, Texas, *J. Geophys. Res.*, *98*, 20,555–20,571.

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

Bleichrodt, J. (1978), Mean tropospheric residence time of cosmic-ray-produced beryllium 7 at north temperate latitudes, *J. Geophys. Res.*, *83*, 3058–3062.

Brost, R., J. Feichter, and M. Heimann (1991), Three-dimensional simulation of ^{7}Be in a global climate model, *J. Geophys. Res.*, *96*, 22,423–22,445.

Brown, L., G. J. Stensland, J. Klein, and R. Middleton (1988), Atmospheric deposition of ^{7}Be and ^{10}Be, *Geochim. Cosmochim. Acta*, *53*, 135–142.

Cannizzaro, F., G. Greco, M. Raneli, M. C. Spitale, and E. Tomarchio (2004), Concentration measurements of ^{7}Be at ground level air at Palermo, Italy—Comparison with solar activity over a period of 21 years, *J. Environ. Radioact.*, *72*, 259–271.

Crecelius, E. A. (1981), Prediction of marine atmospheric deposition rates using total ^{7}Be deposition velocities, *Atmos. Environ.*, *15*, 579–582.

Dibb, J. (1989), Atmospheric deposition of beryllium 7 in the Chesapeake Bay region, *J. Geophys. Res.*, *94*, 2261–2265.

Field, C. V., G. A. Schmidt, D. Koch, and C. Salyk (2006), Modeling production and climate-related impacts on ^{10}Be concentration in ice cores, *J. Geophys. Res.*, *111*, D15107, doi:10.1029/2005JD006410.

Graham, I., R. Ditchburn, and B. Barry (2003), Atmospheric deposition of ^{7}Be and ^{10}Be in New Zealand rain (1996–1998), *Geochim. Cosmochim. Acta*, *67*, 361–373.

Hasebe, N., T. Doke, J. Kikuchi, Y. Takeuchi, and T. Sugiyama (1981), Observations of fallout rates of atmospheric ^{7}Be and ^{22}Na produced by cosmic rays: Concerning estimation of the fallout rate of atmospheric ^{26}Al, *J. Geophys. Res.*, *86*, 520–524.

Heikkilä, U., et al. (2008), Modeling cosmogenic radionuclides ^{10}Be and ^{7}Be during the Maunder Minimum using the ECHAM5-HAM general circulation model, *Atmos. Chem. Phys.*, in press.

Hofmann, H. J., J. Beer, G. Bonani, H. R. Von Gunter, M. Raman, M. Suter, R. L. Walker, W. Wölfli, and D. Zimmermann (1987), ^{10}Be: Half-life and AMS-standards, *Nucl. Instrum. Methods Phys. Res., Sect. B*, *29*, 32–36.

Holton, J., P. Haynes, M. McIntyre, A. Douglass, R. Rood, and L. Pfister (1995), Stratosphere-troposphere exchange, *Rev. Geophys.*, *33*, 403–439.

Huh, C.-A., C.-C. Su, and L.-J. Shiau (2006), Factors controlling temporal and spatial variations of atmospheric deposition of ^{7}Be and ^{210}Pb in northern Taiwan, *J. Geophys. Res.*, *111*, D16304, doi:10.1029/2006JD007180.

Ishikawa, Y., H. Murakami, T. Sekine, and K. Yoshihara (1995), Precipitation scavenging studies of radionuclides in air using cosmogenic ^{7}Be, *J. Environ. Radioact.*, *26*, 19–36.

Knies, D. L. (1994), Cosmogenic radionuclides in precipitation, Ph.D. dissertation, Purdue Univ., West Lafayette, Indiana.

Koch, D., and D. Rind (1998), Beryllium-10/beryllium-7 as a tracer of stratospheric transport, *J. Geophys. Res.*, *103*, 3907–3917.

Koch, D., D. Jacob, and W. Graustein (1996), Vertical transport of tropospheric aerosols as indicated by ^{7}Be and ^{210}Pb in a chemical tracer model, *J. Geophys. Res.*, *101*, 18,651–18,666.

Lal, D., and B. Peters (1967), Cosmic ray produced radioactivity on the Earth, in *Handbuch der Physik*, vol. XLVI/2, pp. 551–612, Springer, New York.

Land, C., and J. Feichter (2003), Stratosphere-troposphere exchange in a changing climate simulated with the general circulation model MAECHAM4, *J. Geophys. Res.*, *108*(D12), 8523, doi:10.1029/2002JD002543.

Liu, H., D. Jacob, I. Bey, and R. Yantosca (2001), Constraints from ^{210}Pb and ^{7}Be on wet deposition and transport in a global three-dimensional chemical tracer model driven by assimilated meteorological fields, *J. Geophys. Res.*, *106*, 12,109–12,128.

Masarik, J., and J. Beer (1999), Simulation of particle fluxes and cosmogenic nuclide production in the Earth's atmosphere, *J. Geophys. Res.*, *104*, 12,099–12,111.

Monaghan, M. C., S. Krishnaswami, and K. K. Turekian (1985/1986), The global-average production of ^{10}Be, *Earth Planet. Sci. Lett.*, *76*, 279–287.

Nagai, H., W. Tada, and T. Kobayashi (2000), Production rates of ^{7}Be and ^{10}Be in the atmosphere, *Nucl. Instrum. Methods Phys. Res., Sect. B*, *172*, 796–801.

Nishiizumi, K., M. Imamura, M. W. Caffee, J. R. Southon, R. C. Finkel, and J. McAninch (2007), Absolute calibration of ^{10}Be AMS standards, *Nucl. Instr. Methods Phys. Res., Sect. B*, *258*, 403–413.

Olsen, C., P. Lowry, N. Cutshall, J. Todd, G. Wong, and W. Casey (1985), Atmospheric fluxes and marsh-soil inventories of ^{7}Be and ^{210}Pb, *J. Geophys. Res.*, *90*, 10,487–10,495.

Papastefanou, C., A. Ioannidou, S. Stoulos, and M. Manolopoulou (1995), Atmospheric deposition of cosmogenic ^{7}Be and ^{137}Cs from fallout of the Chernobyl accident, *Sci. Total Environ.*, *170*, 151–156.

Rehfeld, S., and M. Heimann (1995), Three dimensional atmospheric transport simulation of the radioactive tracers ^{210}Pb, ^{7}Be, ^{10}Be and ^{90}Sr, *J. Geophys. Res.*, *100*, 26,141–26,161.

Todd, J., G. Wong, C. Olsen, and I. Larsen (1989), Atmospheric depositional characteristics of beryllium 7 and lead 210 along the southerneast Virginia coast, *J. Geophys. Res.*, *94*, 11,106–11,116.

Turekian, K., L. Benninger, and E. Dion (1983), ^{7}Be and ^{210}Pb total deposition fluxes at New Haven, Connecticut and Bermuda, *J. Geophys. Res.*, *88*, 5411–5415.

Uematsu, M., R. Duce, and J. Prospero (1994), Atmosphere beryllium-7 concentrations over the Pacific Ocean, *Geophys. Res. Lett.*, *21*, 561–564.

Young, J., and W. Silker (1980), Aerosol deposition velocities on the Pacific and Atlantic oceans calculated from ^{7}Be measurements, *Earth Planet. Sci. Lett.*, *50*, 92–104.

Zanis, P., et al. (2003), An estimate of the impact of stratosphere-to-troposphere transport (STT) on the lower free tropospheric ozone over the Alps using ^{10}Be and ^{7}Be measurements, *J. Geophys. Res.*, *108*(D12), 8520, doi:10.1029/2002JD002604.

V. Alfimov, Ion Beam Physics, Paul Scherrer Institute and ETH Zurich, CH-8093 Zurich, Switzerland.

J. Beer and U. Heikkilä, EAWAG, Überlandsstrasse 133, Postfach 611, CH-8600 Dübendorf, Switzerland. (ulla.heikkilae@eawag.ch)