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Cosmogenic ^{7}Be and ^{22}Na in Finland: Production, observed periodicities and the connection to climatic phenomena

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ABSTRACT

This study presents theoretical production calculations and time series analyses of two cosmogenic isotopes, ^{7}Be and ^{22}Na, from the four high-latitude stations of Kotka, Kajaani, Rovaniemi and Ivalo located in Finland. We used published results for ^{7}Be but performed full simulations of the ^{22}Na production in atmosphere. For the first time, lookup tables of ^{22}Na production by cosmic rays in the atmosphere are presented. In conjunction with calculations using the new model of ^{22}Na production, the ^{7}Be/^{22}Na ratios in the atmosphere were also calculated. The wavelet transform of the ^{7}Be and ^{22}Na time series revealed sets of periodicities in the 2.5–8 year range. The wavelet coherence method was used to study coherences between ^{7}Be and ^{22}Na and ^{7}Be/^{22}Na data and AO, NAO, AMO, QBO and SO teleconnection indices representing different climatic variations in Northern Europe. In the wavelet coherence analyses, the ^{7}Be activities were found to be mainly modulated by NAO and AMO at an interannual scale, while ^{22}Na activity was found to be less effected by climatic phenomena. The ^{7}Be coherence with other indices was intermittent where the coherence with SO was limited to Ivalo data and in the case of QBO, to Kotka data. The ^{22}Na data was not found to be in coherence with any of the studied indices. In the ^{7}Be/^{22}Na ratio a clear seasonal pattern was observed where low ^{7}Be/^{22}Na ratios were observed during summer and high ratios during winter. This was speculated to be caused by the height of atmospheric vertical mixing. During 2006–2011, the ^{7}Be/^{22}Na ratios from Kotka, Kajaani and Rovaniemi showed variance at synoptic time scales but were nearly identical at the seasonal scale. The ^{7}Be/^{22}Na ratio was proposed to be a radiochronometer to estimate residence times of aerosols carrying ^{7}Be and ^{22}Na.

1. Introduction

Cosmogenic nuclides have been used in many applications from palaeoclimatology to solar activity reconstructions. In particular, the ratio of the cosmogenic isotopes produced in spallation reactions, such as ^{7}Be and ^{22}Na, has been found to be a useful tool for studying atmospheric processes (e.g., Bhandari et al., 1966; Tokuyama and Igarashi, 1998; Jasiulionis and Wershofen, 2005). One of the most studied cosmogenic isotopes is ^{7}Be, while ^{22}Na is significantly less known. The ^{7}Be isotope is formed in the galactic cosmic-ray-induced spallation of atmospheric nitrogen and oxygen while ^{22}Na is produced in the spallation of atmospheric argon. These cosmogenic isotopes are radioactive having different half-lives, ^{7}Be $T_{1/2}=53.22$ d and ^{22}Na $T_{1/2}=2.603$ years. ^{7}Be is abundant in ambient air where typical activity concentration varies from few hundred to few thousand μBq/m^3. Compared to ^{7}Be, the activity concentrations of ^{22}Na are roughly four orders of magnitude lower, typically below 1 μBq/m^3 (Grabowska et al., 2003; Jasiulionis and Wershofen, 2005; Rulík et al., 2009). This is mainly due to low concentrations and higher spallation threshold energy of atmospheric argon ($E_T \approx$ 200 MeV) compared to oxygen ($E_T \approx$ 30 MeV).

The concentrations of cosmogenic nuclides in ground level air are, despite production changes, dependent on four factors: (1) wet scavenging, (2) stratosphere-to-troposphere exchange, (3) vertical transfer in troposphere and (4) horizontal transfer between different latitudes (Feely et al., 1989). About 75% of ^{7}Be is produced in the stratosphere and 25% in the upper troposphere (Johnson and Viezee, 1981; Usoskin and Kovaltsov, 2008). The chemical properties of ^{7}Be and ^{22}Na differ from each other. However, these differences should not affect their behavior in the upper troposphere or stratosphere. After formation and adsorption onto local

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aerosols ^{7}Be and ^{22}Na are transported to ground level via atmospheric vertical mixing (Lal and Peters, 1962; Vecchi and Valli, 1997). ^{7}Be attaches predominantly to aerosols of the (sub)micron size (El-Hussein and Ahmed, 1994; Papastefanou and Ioannidou, 1995). Thus their behavior, excluding radioactive decay, is governed by the dynamics, transport and removal processes of the aerosol particles carrying them. ^{7}Be was already proposed as a natural atmospheric tracer in the late 1950s (Lal et al., 1958). However, the use of ^{7}Be as an atmospheric tracer is difficult due to the complexity of atmospheric processes affecting ground-level concentrations. Recently, a cosmic ray model was combined with an atmospheric model enabling novel possibilities to study ^{7}Be as an atmospheric tracer in more detail (Usoskin and Kovaltsov, 2008; Usoskin et al., 2009).

The ambient air ^{7}Be and ^{22}Na activity can be largely affected by seasonal variability. The effect of particularly spring and/or autumn stratospheric–tropospheric mixing on ambient air ^{7}Be activity concentrations has been studied widely, see e.g. Feely et al. (1989), Gerasopoulos et al. (2003), and Aldahan et al. (2008). In addition, the concentrations of ^{7}Be are known to be modulated by large-scale atmospheric phenomena (NAO and ENSO) and by the 11-year solar cycle (see e.g. Koch and Mann, 1996; Talpos et al., 2005; Kikuchi et al., 2009; Leppänen et al., 2010). The seasonal cycle in ^{22}Na activities has been studied from aerosol samples and monthly radioactivity measurements of rainfall (Tokuyama and Igarashi, 1998; Leppänen and Grinsted, 2008; Rulík et al., 2009). Finland is located in the transition zone between continental Eurasia and the North Atlantic. Westerly winds that transport maritime air from the Atlantic region are common. These winds generally carry low concentrations of ^{7}Be partly due to significant wet and dry depositions in the North Atlantic along the path of the Gulf Stream (Field et al., 2006). In Northern Finland, the source areas of ^{7}Be-rich air are in Central Russia and, in the springtime, in the southwest of Finland (Paatero and Hatakka, 2000). Thus depending on the origin of air masses, different ^{7}Be and ^{22}Na activity concentrations are observed.

2. Production of ^{7}Be and ^{22}Na in the atmosphere

2.1. Modeling of the isotope production in the atmosphere

Several models have been published earlier (Lal and Peters, 1962; O'Brien, 1979; Masarik and Beer, 1999; Luyanas, 2004; Webber et al., 2007) to calculate the production rates for these isotopes in the atmosphere. These earlier models do not provide a 3D picture of the isotope production over the wide range of solar activity variations, which is necessary to study the details of the production and transport. Moreover, many of the earlier models are based on simplified assumptions thus reducing their accuracy, especially in the lower atmosphere. Nowadays, the production of cosmogenic isotopes can be modeled by a specially developed code, CRAC (Cosmic Ray induced Atmospheric Cascade), originally built for the beryllium isotopes ^{7}Be and ^{10}Be (Usoskin and Kovaltsov, 2008; Kovaltsov and Usoskin, 2010). Since ^{22}Na is produced in a similar way to beryllium isotopes, i.e., in the spallation of atmospheric argon nuclei, we can also apply the same approach, called CRAC:22Na. Here we only briefly describe some details for the sodium isotope, while full details of the model are given elsewhere (Usoskin and Kovaltsov, 2008; Kovaltsov and Usoskin, 2010).

The cosmogenic isotope ^{22}Na is produced in the atmosphere mainly as a result of the spallation of nuclei of atmospheric argon by energetic protons, neutrons and α-particles. These energetic particles can be either primary cosmic rays in the upper atmosphere or secondary nucleonic components of the cascade initiated by interactions of cosmic rays in the atmosphere. We have modeled the development of the atmospheric cascade by means of a Monte–Carlo simulation tool CORSIKA (Cosmic Ray Simulations for Kascade, version 6.617, August 2007) (Heck et al., 1998) linked to the FLUKA tool (version 2006.3b, March 2007) (Fassò et al., 2001) to simulate interactions between low energy (below 80 GeV of total energy) hadrons, as described by Kovaltsov and Usoskin (2010). The chemical composition of the atmosphere was taken as N_2, O_2 and Ar in the volume fractions of 78.1%, 21% and 0.9%, respectively.

The efficiency of the ^{22}Na isotope production in air by a particle (p, n or α) with the kinetic energy per nucleon E

$$S_x(E) = \kappa_{\mathrm{Ar}} \cdot \sigma_{x\mathrm{Ar}}(E), \tag{1}$$

where x is the type of impinging particle (proton p, neutron n or α-particle), $\sigma_{x\mathrm{Ar}}$ is the cross section of ^{22}Na production by particle of type x on argon, $\kappa_{\mathrm{Ar}} = 1.94 \times 10^{20}$ g^{-1} is the numbers of argon atoms per gram of air.

A cross-section for protons $\sigma_{p\mathrm{Ar}}$ has been adopted from Reyss et al. (1981). We found no information on the cross-section for neutrons and assumed $\sigma_{n\mathrm{Ar}} \approx \sigma_{p\mathrm{Ar}}$, which is in accordance with the high spallation threshold. Following Silberberg and Tsao (1973) and Tatischeff et al. (2006), we consider that $\sigma_{\alpha\mathrm{Ar}}(E) = 2\sigma_{p\mathrm{Ar}}(4E)$. The corresponding efficiencies of ^{7}Be and ^{22}Na are shown in Fig. 1.

As the next step we computed the yield function, which is defined as the production of the isotope by primary particles of type X with the unit intensity J (i.e., one primary particle with

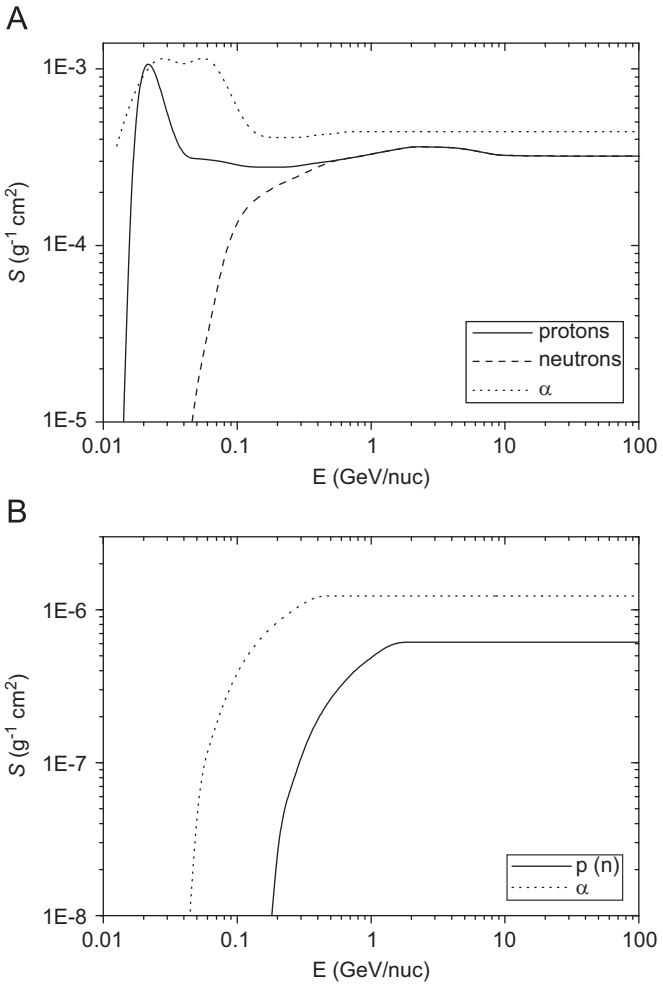

Fig. 1. Efficiency of production of cosmogenic isotopes ^{7}Be (panel A—cf. Fig. 1 in Usoskin and Kovaltsov, 2008) and ^{22}Na (panel B) in air.