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In situ formation of cosmogenic ¹⁴C by cosmic ray nucleons in polar ice

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ABSTRACT

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1. Introduction

Interactions of cosmic rays with the Earth's atmosphere produce cascades of secondary particles and a variety of cosmogenic nuclides. Some of the particles created in these cascades can reach the surface of the Earth and induce nuclear reactions with the appearance of some cosmogenic nuclides. The records of radionuclides produced in the atmosphere and deposited in natural archives have been used as a proxy of changes in the primary cosmic ray flux in the past [1]. However, the cosmic ray signal in these records is in some cases obscured by natural processes on the Earth.

The radiocarbon deposition in polar ice is a complex process depending on many factors. Radiocarbon is incorporated in ice by trapping atmospheric gases during transformation of firn¹ into glacier ice. In addition, nuclear interactions of energetic neutrons and muons of cosmic rays create ¹⁴C in firn and ice, as these are accumulated and in ablating ice as it outcrops. This additional in situ produced ¹⁴C appears to be oxidized to ¹⁴CO and ¹⁴CO₂ [2]. In situ ¹⁴C has been used to determine the ablation rate of outcropping ice in Antarctica [3–5], and the presence of an in situ signal in an accumulating ice has been confirmed in experiments [6–11].

The factors which control deposition of in situ ¹⁴C in polar ice are discussed in Refs. [2,9,12]. The production rate of in situ ¹⁴C in ice depends on the intensity of cosmic ray flux at the polar site.

We study interactions of cosmic ray particles with the Earth's atmosphere and polar ice focusing on in situ formation of radiocarbon in polar ice. We calculate the production rate of the nuclide for sea level high geomagnetic latitudes using various sets of cross section data and compare our results with experimental data. The effective attenuation length of cosmic ray spallation reactions in ice is found to be 130 g/cm² for high geomagnetic latitudes. Accurate determination of this parameter is important for radiocarbon concentration calculations for ice samples from ablating areas of ice sheet. The recalculation of the radiocarbon production rates for different glacier elevations is discussed.

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BEAM INTERACTIONS WITH MATERIALS AND ATOMS

The main parameter controlling the intensity of cosmic rays at polar latitudes is the level of solar activity. Changes in the geomagnetic field do not affect the cosmic ray flux at high geomagnetic latitudes. Hence, the in situ ¹⁴C record in polar ice can provide a direct measure of changes in solar activity in the past [9]. An important problem related to in situ ¹⁴C is the efficiency of retention in firn grains during ice formation. Some amount of ¹⁴C, in situ produced in firn grains, can be lost via the gas diffusion between firn grains and firn air [2,12]. Moreover, sublimation and condensation cycles during firn grain metamorphism have the potential to release species dissolved in ice [10,12,13]. The accurate estimation of the in situ ¹⁴C production rates in ice is necessary for accurate determination of deficiencies in experimental concentrations and, hence, to better understand processes affecting the efficiency of in situ ¹⁴C retention in firn grains.

There is no consensus in the literature about radiocarbon production rates in ice and published values differ in two times (for example, see Refs. [6,11]). In the present work, we study interactions of cosmic rays with the Earth's atmosphere and ice exposed at the Earth's surface. The emphasis of this work is on radiocarbon in situ formation by cosmic ray nucleons in polar ice. The differences between our results and previous works are discussed.

2. Calculational model

2.1. Galactic cosmic rays

Cosmic rays at energies above several hundred MeV per nucleon are mostly of galactic origin, about 90% of particles being protons and 10% helium nuclei. The particle fraction of heavier nuclei does not exceed 1%. The flux of galactic cosmic rays is highly isotropic.

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¹ Firn is an intermediate stage in the transformation of snow to glacier ice.

At energies below few GeV per nucleon the flux of galactic cosmic rays in space at the Earth's orbit depends on the level of solar activity. The differential energy spectrum of cosmic ray nuclei of type i in the force field approximation is given by [14]:

$$J_i(E) = J_{i,\text{LIS}}(E + \Phi_i) \frac{E(E + 2m_p c^2)}{(E + \Phi_i)(E + \Phi_i + 2m_p c^2)},$$
(1)

where $J_{i,LIS}(E)$ gives the local interstellar spectrum of nuclei *i*, *E* is kinetic energy of nucleus per nucleon, m_pc^2 is proton's rest-mass energy, $\Phi_i = (eZ_i/A_i)\varphi$, Z_i and A_i are nucleus charge and mass numbers, respectively, *e* is the elementary charge, and φ is the modulation potential of cosmic rays in the heliosphere. We take the parameterization of the local interstellar spectrum from Ref. [15]:

$$J_{i, \text{ LIS}}(E) = C_i \frac{p(E)^{-2.78}}{1 + 0.487 p(E)^{-2.51}},$$
(2)

where *E* is expressed in GeV per nucleon, $p(E) = \sqrt{E(E + 2m_pc^2)}$, *C_i* is the normalization factor; $C_p = 1.9 \times 10^4 \text{ (m}^2 \text{ s r GeV})^{-1}$ for protons, and $C_{\text{He}} = 9.5 \times 10^2 \text{ (m}^2 \text{ s r GeV/nucleon})^{-1}$ for helium nuclei.

The modulation potential φ , reconstructed for the period of 1951–2004 years using the data from the worldwide neutron monitor network, ranges from 0.3 to 1.3 GV, the mean value of φ is 0.69 GV [15]. We adopt this mean value of the modulation potential in our simulations. The present-day mean solar modulation appears to be similar to the long-term mean, see discussion and references in Ref. [16]. Note, that for solar modulation parameters within 25% of the modern mean, cosmogenic neutron fluxes in the atmosphere vary about ±5% at high latitudes [17].

The Z/A ratio determines the shape of the differential energy spectrum at low energies. This ratio is close to 1/2 for nuclei with charge numbers $Z \ge 2$. The contribution of all $Z \ge 2$ nuclei to particle cascade in matter is determined by applying a scaling factor k to the results, obtained for α -particles, where k is the ratio of nucleon number densities of all $Z \ge 2$ nuclei to α -particles in the galactic cosmic rays. The data on energy spectrum of cosmic-ray nuclei from Ref. [18] give k = 1.44. All Z = 2 particles are treated as ⁴He nuclei, i.e., the abundance of ³He in the helium flux is neglected.

The vertical cutoff rigidities for geomagnetic latitudes $\lambda > 60^{\circ}$ generally do not exceed 1 GV [19]. This cutoff value corresponds to proton kinetic energy about 430 MeV and α -particle kinetic energy about 125 MeV/nucleon. Particles at these energies provide a minor contribution to the cascade processes in the atmosphere. Hence, cosmic ray fluxes in the atmosphere are unaffected by changes in the geomagnetic field at high geomagnetic latitudes. We disregard the effect of geomagnetic cutoff on energy spectra of cosmic rays because we consider the radiocarbon production in ice at high geomagnetic latitudes.

2.2. Model of the Earth's atmosphere and surface

The Earth is modeled as a sphere of a radius of 6371 km. The Earth's surface is assumed to contain H_2O ice with the density of 0.917 g/cm³.

The atmosphere is considered as a spherical shell of 100 km thickness. The chemical composition of the air (by mass) is nitrogen 75.5%, oxygen 23.2% and argon 1.3%. At altitudes from the surface to about 80 km the chemical composition is nearly constant due to atmospheric mixing [20]. The total thickness of the atmosphere is taken to be equal to the atmospheric depth of 1034 g/cm² at sea level. The atmosphere is divided into concentric subshells with a thickness of 15 g/cm² and constant air density within one subshell. Additional division is done near air–ice interface. The dependence of air density on altitude is taken from the COSPAR reference atmosphere data for high geographic latitudes $\lambda > 60^{\circ}N$ [21].

The cosmic ray particle flux at a given altitude is controlled by the mass of atmosphere (atmospheric depth) traversed by the particles. To apply simulation results to a given location on the Earth one needs to convert site altitude to atmospheric depth using pressure data or appropriate air density dependence on altitude [22–24].

2.3. Physics input

Particle fluxes in matter are calculated using simulation toolkit GEANT4 9.4 [25]. The processes included are those of production, propagation and interaction of baryons (nucleons, short-lived baryons and their antiparticles), mesons (pions and kaons), light nuclei, leptons (electrons, positrons, muons) and gamma rays. Standard electromagnetic processes, photonuclear and electronuclear processes are taken into account [26]. Particles of electronphoton component with energies less than 10 MeV are excluded from calculations. Low-energy and high-energy parameterized models are used to describe inelastic scattering of hadrons and nuclei. The Bertini intranuclear cascade model is employed for describing nucleon-nucleus and meson-nucleus inelastic scattering at hadron energies up to 6 GeV. The binary intranuclear cascade model is adopted for inelastic scattering of light nuclei. Processes of negative meson capture, neutron capture and neutron fission are included. High precision neutron models are used for simulating neutron-nucleus interactions. These models are based on the G4NDL data library (version 3.14) that comes largely from the ENDF-B VI and JENDL libraries for neutron energies below 20 MeV. For neutron energies in the range from 20 MeV to about 3 GeV the JENDL/HE cross section data are employed.

2.4. Calculation of particle fluxes

Let us define the angle-integrated differential flux of particles of type *i*:

$$I_i^{\rm diff}(E) = \int_{4\pi} d\Omega J_i (E)$$

where J_i (E) is a directed differential flux or differential energy spectrum of particles *i*. Let us introduce also the angle- and energy-integrated flux of particles *i* with energies above *E*:

$$I_i^{\rm int}(E) = \int_E^\infty dE' \, I_i^{\rm diff}(E') = \int_E^\infty dE' \int_{4\pi} d\Omega J_i \, (E').$$

The 4π angle-integrated integral flux of galactic cosmic rays is found to be 2.66 cm⁻² s⁻¹ for protons, and 0.27 cm⁻² s⁻¹ for helium nuclei at φ = 0.69 GV and *E* = 100 MeV/nucleon employing the particle differential energy spectra (1) and (2).

The flux of galactic cosmic rays passing through unit area of the upper atmosphere boundary is:

$$F_{0i} = \int_{2\pi} \mathrm{d}\Omega\,\cos\theta\,\int_{E}^{\infty}\mathrm{d}E'J_{0i}\,(E') = \pi\int_{E}^{\infty}\mathrm{d}E'J_{0i}\,(E'),$$

where $J_{0i}(E)$ is directed differential flux of galactic cosmic ray nuclei of type *i* at the Earth orbit, θ is the angle between particle momentum and nadir point. The distribution over θ of the primary cosmic rays penetrating the atmosphere satisfies the relation $dF_{0i}/d\cos\theta \sim \cos\theta$.

Angle-integrated differential flux of particles i with energy E in matter at a depth z is calculated according to:

$$I_{i}^{\text{diff}}(E,z) = \frac{F_{0p}}{N_{0p}\Delta E} \sum_{l} \frac{1}{|\cos\theta_{li}|} + k \frac{F_{0\alpha}}{N_{0\alpha}\Delta E} \sum_{l} \frac{1}{|\cos\theta_{li}|},$$

where N_0 is the number of primary particles (protons or α -particles) for which the cascade simulations are performed, the inner summation is done over all particles *i* crossing a fixed level in matter at

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