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Spatial distribution of the atmospheric radionuclide production by galactic cosmic rays and its imprint in natural archives

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ABSTRACT

We used the GEANT4 toolkit to simulate the altitude and latitude profiles of the production rate of ^{14}C, ^{10}Be and ^{36}Cl radionuclides by the galactic cosmic ray (GCR) interactions in the terrestrial atmosphere at a varying geomagnetic field. We found that applying two intranuclear cascade models incorporated in GEANT4 (Binary Intranuclear Cascade, BIC, and Bertini Intranuclear Cascade, BERT) result in significantly different production rate values. We present the conclusions about the certain model relevance to the abundance of these isotopes in the surface fallout, ice-core records and lunar soil depth profile. Comparison of our simulations with the recent publication of Poluianov et al. (2016) shows a good agreement for ^{14}C (BIC) and ^{10}Be (BERT) and a definite by the factor 2–3 difference in the ^{36}Cl (BIC) atmospheric yield functions. Also, the mean level and amplitude of the ^{10}Be variations in polar ice from central regions of Antarctica and Greenland could be accounted for its tropospheric production by GCRs. The fallout rate of ^{36}Cl there can be explained assuming its additional input from the stratosphere. Significant additional variations of radionuclide sedimentation rate in polar regions may arise due to tropopause height changes even at a constant atmospheric production rate of the certain isotope.

1. Introduction

Stationary production of long-lived radionuclides (^{14}C, ^{10}Be, ^{36}Cl etc.) in the atmospheric nuclear reactions induced by galactic cosmic rays (GCRs) has attracted a strong interest since its discovery (Libby, 1946) up to now (Beer et al., 2012). GCRs are the dominant source of cosmogenic radionuclides in the environment compared to the sporadic pulses of atmospheric production from the solar energetic particles (SEPs) and the mobile fraction of the surface *in-situ* production. The measurable manifestation of these processes is the fallout of the nuclides on the surface and their following fixation in the so-called natural archives, which are natural objects with known history and stable chemical composition. The interpreting of measured concentrations of cosmogenic radionuclides in terms of their connection to cosmic and geophysical processes was thoroughly discussed in (Lal and Peters, 1967) and later reexamined and complemented by many authors. However, a model of nuclide's atmospheric production forms the foundation of all the applications of their concentration measurements to particular physical problem, atmospheric physics included. The informative review of cosmogenic production models can be found in Beer et al. (2012), which covers the history of the question and the variety of its modern applications. The applicability and reliability of the production calculation for ^{14}C may be directly checked by the independent estimate of its global abundance balance. Its altitude-latitude production variations are globally equalized by the fast air transport of atmospheric ^{14}CO$_2$. The same confirmation of calculation results is hardly possible for ^{10}Be and ^{36}Cl because of the strong dependence of their local fallout on the atmospheric processes. Therefore, we propose as additional tests for these radionuclides to compare calculations of their production by GCRs in lunar soil with their measured concentration depth profiles. Also, the ^{10}Be local concentration in the ice cores from central regions of Antarctica and Greenland is used as a limiting factor for its calculated tropospheric production because of its fast local fallout.

In the present paper we carry out the modeling of the atmospheric production of long-lived ^{14}C, ^{10}Be and ^{36}Cl by GCRs using GEANT4 toolkit. GEANT4 utilizes Monte Carlo method both for atmospheric internuclear and intranuclear cascade modeling within the same procedure in contrast to previous approaches of Masarik and Beer (2009); Kovaltsov and Usoskin (2010); Kovaltsov et al. (2012); Poluianov et al. (2016). In these papers, the Monte Carlo approach was used to calculate secondary particle fluxes only, which then were multiplied by the corresponding radionuclide nuclear cross sections. Such an approach may

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lead to incorrect production rate estimation (Section 4) because this flux should be also formed by the isotope production reactions theirselves. Secondary particles may also contribute to the additional nonphysical generation of isotopes when they move in the direction close to horizontal one of the atmospheric layer (for more details see Section 4). Our calculation scheme avoids these inaccuracies. Also, we present a comprehensive study of the influence of the optional models of intranuclear cascades built in GEANT4 on radionuclide production rates which has not been done before (see next Section).

The aim of the present paper is to give a detailed analysis of the individual cosmogenic nuclide atmospheric yield functions by GCRs. Then, we compare our production results with the records of ^{14}C in tree-rings, ^{10}Be and ^{36}Cl in polar ice-cores and all of three isotopes in lunar rocks to determine the most relevant intranuclear cascade model and, hence, the most reliable yield function for each of the studied radionuclide. Such standardized yield functions would become a foundation stone for the following research as was previously pointed out by Webber and Higbie (2003) and Poluianov et al. (2016).

The paper is organized as follows. The results of GCR's production modeling are given in the first part along with their critical comparison with recently published results of other authors. The second part gives the connection of the atmospheric production with the measured surface concentration and considers the role of the atmospheric transport of the cosmogenic radioactivity, especially the stratosphere to the troposphere air exchange, in the regulation of the surface fallout. In the last part we discuss the origin of the temporal variations in the fallout of ^{10}Be and ^{36}Cl revealed from their concentrations in dated polar ice and draw a number of final conclusions.

2. Modeling of the cosmogenic nuclides' atmospheric production

Historically, the first calculations of cosmogenic nuclide production were based on the semiempirical models describing the depth development of the atmospheric nuclear cascade. The altitude profiles of the differential energy spectra of secondary neutrons provided by these calculations resulted in estimates of the mean atmospheric production rate of ^{14}C (Lingenfelter, 1963; Mendell et al., 1973). Calculations of spallation reaction products, such as ^{10}Be and ^{36}Cl, were carried out with more sophisticated models of nucleonic interactions (Lal and Peters, 1967). The concept of a yield function of the given nuclide in the atmosphere was firstly introduced by O'Brien (1979) as a mean relative production of the nuclide per a “nuclear star” – an equivalent of the strong inelastic nuclear interaction. Given the experimental data on the altitude-latitude distribution of stars in nuclear emulsions exposed in the atmosphere, calculations of the production rate of a nuclide became straightforward. Later on, much interest was given to the estimates of the variations of the nuclides' production rate caused by the extra-atmospheric GCR temporal variations (Castagnoli and Lal, 1980; Blinov, 1988).

Application of innovative accelerator mass-spectrometry method, which allowed measurements of isotopic ratios as low as 10^{-16} in a sample, to studies of cosmogenic nuclides in natural archives revolutionary increased the sensitivity of the measurements and demanded an adequate progress in the accuracy of the production calculations. At that time several codes were already available for modeling the high-energy nuclear cascade in matter with applications to experimental physics and extensive air showers. They allowed proceeding from the calculations of the mean global production rate to latitude-altitude resolved isotope production models predicting its atmospheric 3D distribution. It gave a necessary clue for interpreting the nuclide records measured at certain locations and attributed to certain production conditions. The FLUKA code was used by Webber and Higbie (2003) for studies of solar modulation influence on ^{10}Be and ^{36}Cl atmospheric production, while the GEANT4 toolkit has become more common for atmospheric calculation since then (Masarik and Beer, 2009; Kovaltsov and Usoskin, 2010; Kovaltsov et al., 2012; Poluianov et al., 2016). Recently the comprehensive study has been published (Poluianov et al., 2016) with recommendations of the standardized calculation algorithm, which we are going to discuss in more details in the following.

Our production rate simulations of ^{10}Be, ^{14}C and ^{36}Cl in the terrestrial atmosphere by protons and alpha-particles were also carried out with the GEANT4.10 software package. The choice of these nuclides is explained, firstly, by their long lifetime, allowing studies of the astrophysical phenomena on the millennium time scale and, secondly, by the comprehensive experimental data available on their concentration in natural archives. The professional software toolkit GEANT4 (GEANT4 collaboration, 2013; Allison et al., 2016) was initially developed by CERN coworkers for high-energy experimental applications. It is a Monte-Carlo code that includes the internuclear and intranuclear cascades, which computation parts are complemented by the relational databases of nuclear parameters like reaction cross-sections etc. The GEANT4.10 version was successfully used in our previous papers (Pavlov et al., 2013, 2014b) for calculations of the production of the same nuclides by solar energetic particles from superflares and by gamma-radiation from the Galactic gamma-ray bursts. Also, we used similar method for simulation of these isotopes production in Martian rocks (Pavlov et al., 2014a).

The approach applied in the present paper is based on the formalism of the yield function. It is defined in the same way as in the papers of Webber and Higbie (2003) and Poluianov et al. (2016). Namely, it is a number of radionuclides produced in an atmospheric column by an incident incoming particle of a given energy E. However, we normalize our simulations to one incident particle “isotropically distributed” at the top of the atmosphere while Poluianov et al. (2016) made similar normalization to a unit particle flux. It results in the systematic factor of π difference of the values.

For the most effective for nuclide production energy interval, that encompasses the bulk of incident GCRs, GEANT4 offers optional nuclear reaction models, and namely the Binary Intranuclear Cascade (BIC) and the Bertini Intranuclear Cascade (BERT), because there is no universal theory of nuclear reactions in a wide interaction energy range. Two abovementioned models differ in describing hadron-nucleon interactions (different account of resonances) and in the model developing of the intranuclear cascading process based on binary collisions in a nuclear medium (different nuclear field potentials). They also distinguish the description of nuclear density and nucleon momentum distributions, precompound models of residuals, de-excitation modeling etc. As a result, these models provide different fluxes of cascading secondary particles in the terrestrial atmosphere (Kang et al., 2013). Similar difference in a secondary neutron flux (up to 70%) was found for GEANT4 simulations with laboratory experimental results (Lo Meo et al., 2015). Independently on the secondary particle fluxes there is also difference in radionuclide yields during the intranuclear process itself. So, the yield function approach should be compared with radionuclide measurements to select relevant intranuclear cascade model. The difference between the results of modeling using BIC and BERT is significant. The results of our simulations are presented in Fig. 1a and b for protons and alpha-particles in comparison with those of Poluianov et al. (2016). We see good agreement of both approaches excluding significant (on the average by 2 times) difference of ^{36}Cl production. Below we discuss the possible origin of this discrepancy.

To obtain the local production rate of the isotopes dependent on altitude, latitude and longitude one should convolute the obtained yield functions with the GCR spectra of incident particles, which are solar activity dependent.

Apart from the most abundant particles in GCR – protons, the Earth is also bombarded by alpha-particles (~10% by number), which were also included in our calculations. The simulations showed that the contribution of alpha-particles to the isotopic production rate was about ~40% in the BIC model. Fig. 1 a,b demonstrate the yield functions for ^{14}C, ^{10}Be and ^{36}Cl produced by one primary proton and alpha-particle of energy E, MeV/nucleon, for BIC and BERT models of intranuclear cascades. As the BERT model did not allow us to consider alpha-particle as a projectile we