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## A new model of cosmogenic production of radiocarbon $^{14}\text{C}$ in the atmosphere

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### ABSTRACT

We present the results of full new calculation of radiocarbon  $^{14}\text{C}$  production in the Earth atmosphere, using a numerical Monte-Carlo model. We provide, for the first time, a tabulated  $^{14}\text{C}$  yield function for the energy of primary cosmic ray particles ranging from 0.1 to 1000 GeV/nucleon. We have calculated the global production rate of  $^{14}\text{C}$ , which is 1.64 and 1.88 atoms/cm<sup>2</sup>/s for the modern time and for the pre-industrial epoch, respectively. This is close to the values obtained from the carbon cycle reservoir inventory. We argue that earlier models overestimated the global  $^{14}\text{C}$  production rate because of outdated spectra of cosmic ray heavier nuclei. The mean contribution of solar energetic particles to the global  $^{14}\text{C}$  is calculated as about 0.25% for the modern epoch. Our model provides a new tool to calculate the  $^{14}\text{C}$  production in the Earth's atmosphere, which can be applied, e.g., to reconstructions of solar activity in the past.

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### 1. Introduction

Radiocarbon  $^{14}\text{C}$  is a long-living (half-life about 5730 yr) radioactive nuclide produced mostly by cosmic rays in the Earth's atmosphere. Soon after production, it gets oxidized to  $^{14}\text{CO}_2$  and in the gaseous form takes part in the complex global carbon cycle (Bolin et al., 1979). Radiocarbon is important not only because it is used for dating in many applications (e.g., Dorman, 2004; Kromer, 2009), but also because it forms a primary method of paleo-reconstructions of solar activity on the millennial time scales (e.g., Stuiver and Quay, 1980; Stuiver and Braziunas, 1989; Bard et al., 1997; Muscheler et al., 2007). An essential part of the solar activity reconstruction from radiocarbon data is computation of  $^{14}\text{C}$  production by cosmic rays in the Earth's atmosphere. First such computations were performed in the 1960–1970s (e.g., Lingenfelter, 1963; Lingenfelter and Ramaty, 1970; Light et al., 1973; O'Brien, 1979) and were based on simplified numerical or semi-empirical methods. Later, full Monte-Carlo simulations of the cosmic-ray induced atmospheric cascade had been performed (Masarik and Beer, 1999, 2009). Most of the earlier models, including O'Brien (1979) and Masarik and Beer (1999) deal with a prescribed functional shape of the galactic cosmic ray spectrum, which makes it impossible to be

applied to other types of cosmic ray spectra, e.g., solar energetic particles, supernova explosions, etc. A flexible approach includes calculation of the yield function (the number of cosmogenic nuclei produced in the atmosphere by the primary cosmic rays of the given type with the fixed energy and unit intensity outside the atmosphere), which can be convoluted with any given energy spectrum of the primary cosmic rays (e.g., Webber and Higbie, 2003; Webber et al., 2007; Usoskin and Kovaltsov, 2008; Kovaltsov and Usoskin, 2010). This approach can be directly applied to, e.g., a problem of the signatures of extreme solar energetic particle events in the cosmogenic nuclide data, which is actively discussed (e.g., Usoskin et al., 2006; Hudson, 2010; LaViolette, 2011). Some earlier models (Lingenfelter, 1963; Castagnoli and Lal, 1980) provide the  $^{14}\text{C}$  yield function however it is limited in energy. Moreover, different models give results, which differ by up to 50% from each other, leading to large uncertainty in the global  $^{14}\text{C}$  production rate. Therefore, the present status is that models providing the yield function are 30–50 yr old and have large uncertainties.

In addition, there is a systematic discrepancy between the results of theoretical models for the  $^{14}\text{C}$  production and the global average  $^{14}\text{C}$  production rate obtained from direct measurements of the specific  $^{14}\text{CO}_2$  activity in the atmosphere and from the carbon cycle reservoir inventory. While earlier production models predict that the global average pre-industrial production rate should be 1.9–2.5 atoms/cm<sup>2</sup>/s, estimates from the carbon cycle inventory give systematically lower values ranging between 1.6 and 1.8 atoms/cm<sup>2</sup>/s (Lingenfelter, 1963; Lal and Suess,

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1968; Damon and Sternberg, 1989; O'Brien et al., 1991; Goslar, 2001; Dorman, 2004). This discrepancy is known since long (Lingenfelter, 1963) but is yet unresolved (Goslar, 2001).

In this work we redo all the detailed Monte-Carlo computations of the cosmic-ray induced atmospheric cascade and the production of  $^{14}\text{C}$  in the atmosphere to resolve the problems mentioned above. In Section 2 we describe the numerical model and calculation of the radiocarbon production. In Section 3 we compare the obtained results with earlier models. In Section 4 we apply the model to calculate the  $^{14}\text{C}$  production by galactic cosmic rays and solar energetic particle events for the last solar cycle. Conclusions are presented in Section 5.

## 2. Calculation of the $^{14}\text{C}$ production

Energetic primary cosmic ray particles, when entering the atmosphere, collide with nuclei of the atmospheric gases initiating a complicated nucleonic cascade (also called shower). Here we are interested primarily in secondary neutrons whose distribution in the atmosphere varies with altitude, latitude, atmospheric state and solar activity. Neutrons are produced in the atmosphere through multiple reactions including high-energy direct reactions, low-energy compound nucleus reactions and evaporation of neutrons from the final equilibrium state. Most of the neutrons with energy below 10 MeV are produced as an evaporation product of excited nuclei, while high-energy neutrons originate as knock-on neutrons in collisions or in charge exchange reactions of high-energy protons. While knock-on neutrons are mainly emitted in the forward direction (viz. downwards), evaporated neutrons of lower energy are nearly isotropic. Radiocarbon  $^{14}\text{C}$  is a by-product of the nucleonic cascade, with the main channel being through capture of secondary neutrons by nitrogen:  $\text{N}14(\text{n,p})\text{C}14$ . Other channels (e.g., via spallation reactions) contribute negligibly, but are also considered here.

We have performed a full Monte-Carlo simulation of the nucleonic component of the cosmic ray induced atmospheric cascade, using the Planetocosmic code (Desorgher et al., 2005) based on GEANT-4 toolkit for the passage of particles through matter (Geant4 Collaboration, 2003) (see details in Appendix). The secondary particles were tracked through the atmosphere until they undergo reactions with an air nucleus, exit the atmosphere or decay. In particular, secondary neutrons were traced down to epi-thermal energy. Simulations are computationally intensive. Simulations of single energies (ranging from 0.1 to 1000 GeV/nucleon) were conducted, to determine the resulting flux of secondary neutrons. Since the calculations require very large computational time to keep the statistical significance of the results for low energies, we applied an analytical approach for atmospheric neutrons with energy below 10 eV (see details in Appendix). Cross-sections have been adopted from the Experimental Nuclear Reaction Database (EXFOR/CSISRS) <http://www.nndc.bnl.gov/exfor/exfor00.htm>. The number of simulated cascades induced by primary CR particles was chosen as  $10^5$ – $10^6$  to keep the statistical stability of the results at a reasonable computational time. Computations were carried out separately for primary protons and  $\alpha$ -particles. Because of the

similar rigidity/energy ratio, nuclei with  $Z > 2$  were considered as effectively  $\alpha$ -particles with the scaled number of nucleons (cf. Usoskin and Kovaltsov, 2008).

As the main result of these detailed computations we calculated the  $^{14}\text{C}$  yield function. The yield functions for primary protons and  $\alpha$ -particles are tabulated in Table 1 and shown in Fig. 1 (the energy range above 100 GeV/nucleon is not shown). Note that the yields (per nucleon with the same energy) are identical for protons and  $\alpha$ -particle, viz. an  $\alpha$ -particle is identical to four protons, at energies above 10 GeV/nucleon. Details of the computations are given in Appendix A. All further calculations are made using these yield functions.

In order to compute the  $^{14}\text{C}$  production  $q$  in the atmosphere at a certain place and conditions/time, one can use the following method:

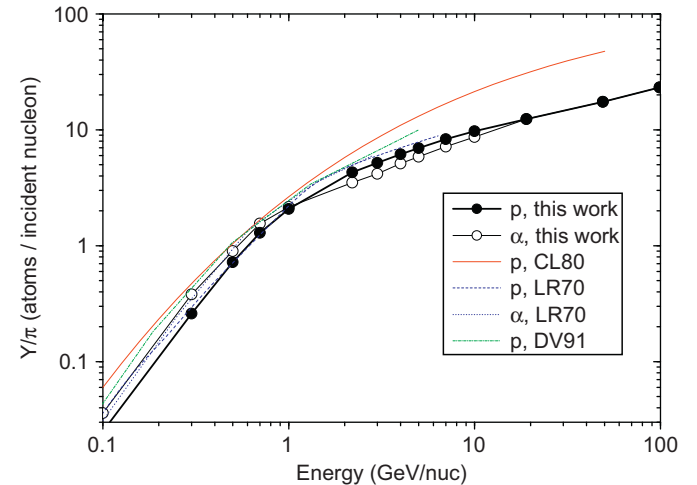
$$q(t) = \sum_i \int_{E_{ic}}^{\infty} Y_i(E) J_i(E, t) dE, \quad (1)$$

where  $E$  is the particle's kinetic energy per nucleon,  $J_i$  is the spectrum of primary particles of type  $i$  on the top of the atmosphere,  $E_{ic}$  in GeV/nucleon is the kinetic energy per nucleon corresponding to the local geomagnetic rigidity cutoff  $P_c$  in GV

$$P_c = \frac{A_i}{Z_i} \sqrt{E_{ic} (E_{ic} + 2E_r)}, \quad (2)$$

where  $E_r = 0.938$  GeV/nucleon is the proton's rest mass. Summation is over different types of the primary cosmic ray nuclei with charge  $Z_i$  and mass  $A_i$  numbers. The local geomagnetic rigidity cutoff is roughly defined via the geomagnetic latitude  $\lambda_G$  of the location as following (Elsasser et al., 1956):

$$P_c [\text{GV}] = 1.9 \cdot M \cdot \cos^4 \lambda_G, \quad (3)$$



**Fig. 1.** Yield function  $Y/\pi$  of  $^{14}\text{C}$  production in the Earth's atmosphere by primary cosmic ray protons and  $\alpha$ -particles (as denoted by "p" and " $\alpha$ " in the legend, respectively) with given energy per nucleon. Different curves correspond to the present work (Table 1) and earlier models Castagnoli and Lal (1980, CL80), Lingenfelter and Ramaty (1970, LR70) and Dergachev and Veksler (1991, DV91), as denoted in the legend.

**Table 1**

Normalized yield functions  $Y_p/\pi$  and  $Y_\alpha/\pi$  of the atmospheric columnar  $^{14}\text{C}$  production (in atoms sr) by a nucleon of primary cosmic protons and  $\alpha$ -particles, respectively, with the energy given in GeV/nucleon. For energy above 20 GeV/nucleon, an  $\alpha$ -particle is considered to be identical to be identical to four protons.

| E (GeV/nucleon) | 0.1   | 0.3  | 0.5  | 0.7  | 1    | 3    | 7    | 10   | 19    | 49    | 99    | 499   | 999   |
|-----------------|-------|------|------|------|------|------|------|------|-------|-------|-------|-------|-------|
| Proton          | 0.025 | 0.26 | 0.72 | 1.29 | 2.07 | 5.19 | 8.32 | 9.72 | 12.40 | 17.45 | 23.24 | 48.30 | 72.73 |
| $\alpha/4$      | 0.036 | 0.38 | 0.89 | 1.55 | 2.16 | 4.18 | 7.17 | 8.67 | 12.40 | 17.45 | 23.24 | 48.30 | 72.73 |

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