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Photonuclear production of cosmogenic beryllium-7 in the terrestrial atmosphere

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Average N(γ,X)^{7}Be, O(γ,X)^{7}Be, and C(γ,X)^{7}Be cross sections of the natural composition of target isotopes in an energy range from threshold to 90 MeV are measured. By using cross sections of the photoproduction of ^{7}Be and a simulation of the nuclear-electromagnetic cascade in the atmosphere, the contribution from the photonuclear mechanism into the production of a cosmogenic ^{7}Be radionuclide in the Earth's atmosphere is investigated. It is shown that the contribution from the photonuclear channel into the production of cosmogenic ^{7}Be is to be taken into account in processes of the accumulation and transfer of ^{7}Be in the ground layer.

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I. INTRODUCTION

Cosmogenic isotopes with a relatively short lifetime have been recognized as useful tools to study atmospheric transport of air masses [1]. Particularly suitable for this purpose is the cosmogenic isotope ^{7}Be (the half-life time of 53.6 days), which is produced through interactions of atmospheric nuclei and the nucleonic component of the atmospheric cascade induced by galactic cosmic rays. To date, the monitoring of radionuclides in the atmospheric boundary layer suggests that a substantial contribution into the radioactivity of surface air stems from a short-lived ^{7}Be isotope of the cosmogenic origin. Variations in the contents of ^{7}Be in air are associated with the solar activity and have the characteristic seasonal variation and the latitude dependence. Shortly after formation, ^{7}Be atoms become attached to atmospheric aerosols, and thus, their fate is related to the aerosol transport. Therefore, ^{7}Be appears to be an excellent tracer for the atmospheric circulation and often is used to constrain atmospheric circulation models [2]. Due to the rapid decay, ^{7}Be isotope activity in plants varies depending on the synoptic conditions. Therefore, ^{7}Be is of interest not only in terms of radiation exposure to biological systems, but also as an indicator of rates of an exchange in plants and, as a consequence, an indicator of an accumulation of pollutants by natural environments in the atmosphere. This is what makes ^{7}Be the convenient indicator for the rapid assessment of a potential air pollution and air exchange in the environment. Therefore, the study of occurrence processes, transportation, and migration of the ^{7}Be radionuclide in the environment is of great interest. For this purpose, one needs to precisely know the features of its production in the atmosphere. A number of models has been developed to compute the ^{7}Be production in the atmosphere as presented in Ref. [3].

It is believed that the main reactions, which lead to the formation of beryllium isotopes in Earth's atmosphere, occur in the interaction of cosmic rays with nuclei of nitrogen and oxygen [3,4], which are the main air components. There are the so-called "spallation" reactions: ^{14}N(p,X)^{7}Be, ^{16}O(p,X)^{7}Be, ^{14}N(n,X)^{7}Be, and ^{16}O(n,X)^{7}Be. Another possible mechanism of the formation of a ^{7}Be isotope in the upper atmosphere can be the photonuclear reactions that have not been investigated yet.

To calculate the production rate of the ^{7}Be isotope in the Earth's atmosphere due to the aforementioned mechanism, it is necessary to know the cross sections of the photonuclear reactions. Unfortunately, there are very limited data on the reactions of the many-body photodisintegration of nuclei to the registration of all the final products. For example, the cross section of the ^{12}C(γ, $n\alpha$)^{7}Be reaction was studied in detail in Ref. [5]. The reaction to a natural mixture of oxygen isotopes ^{0}O(γ,X)^{7}Be in the energy range from 300 to 1000 MeV was investigated in Ref. [6]. The average value of the cross section of the photonuclear reaction ^{0}O(γ,X)^{7}Be in this energy range was about 0.3 mb (the details of the cross section can be found in the database "CDFE" [7]). The data on the ^{14}N(γ,X)^{7}Be reaction are absent. Since nitrogen and oxygen are the main air components, the reaction to these nuclei is of particular interest to analyze the mechanism of the photonuclear production of ^{7}Be in the terrestrial atmosphere.

II. PHOTONUCLEAR CROSS SECTIONS

The cross section of the ^{7}Be photoproduction in the ^{12}C(γ,X)^{7}Be, ^{14}N(γ,X)^{7}Be, and ^{16}O(γ,X)^{7}Be reactions was measured in an experiment on irradiation of the targets, which included oxygen, nitrogen, and carbon by the bremsstrahlung from the electron linear accelerator at the National Science Center "Kharkov Institute of Physics and Technology," Ukraine. The energy of the accelerated electrons was 90 MeV,

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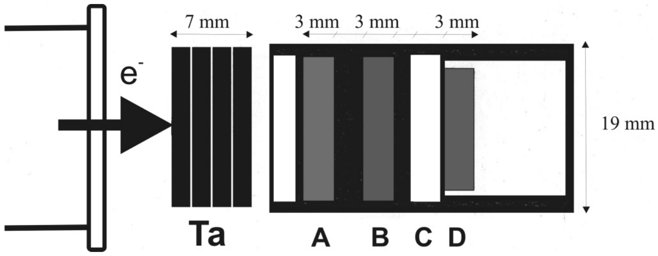

FIG. 1. Layout of the target irradiation on the accelerator. "Ta" denotes the four tantalum plates converter; A and B denote the Al containers with powdered nitrides AlN and BN, respectively; C denotes the corundum target (Al_2O_3), and D denotes the carbon target.

and the current was 3.26 μA. The chemical substances, which contained the given elements, were as follows: boron nitride (BN), aluminum nitride (AlN), corundum (Al_2O_3), and pyrographite (C). The bremsstrahlung converter was designed as four tantalum plates, each 1-mm thick separated by a 1-mm interval. The composite target device is shown in Fig. 1.

The irradiation of the targets was carried out simultaneously for 1 h. A rapid and nondestructive analysis of ^{7}Be is possible by the direct measurement of 477.6-keV γ rays. Since the photonuclear channel of^{7}Be production on aluminum nuclei does not exist (the reaction threshold is greater than 90 MeV), the activity of the targets AlN and Al_2O_3 is only due to the ^{4}N(γ,X)^{7}Be and ^{16}O(γ,X)^{7}Be reactions.

At the same γ-ray flux on nitrogen in the BN and AlN targets, it is possible to estimate the activity of the N channel in the BN target in the proportion of mass ratios of nitrogen in the BN and AlN targets. Thus, we can find the activity due to the boron and nitrogen nucleus in the BN target. The measurement of the ^{7}Be activity of each target after irradiation was performed on the spectrometer complex "CANBERRA InSpector-2000" with a sufficient energy resolution of not less than 1.74 keV in the region of 1332 keV.

To calculate the photoproduction cross section of the ^{7}Be isotope, one needs the knowledge of the bremsstrahlung photons' flux density in the target's location. For a computer simulation of the primary electrons passage through the target, we used the library of the GEANT4 classes [8] and the low-energy electromagnetic processes models (EPDL97 and EEDL97 [9]). The GEANT4 classes handle the following processes: Rayleigh scattering of γ's, Compton scattering of γ's, photoelectric effects, (e^+,e^-) pair production by γ's, multiple scattering for electrons and positrons, ionization and energy loss by electrons and positrons, bremsstrahlung by electrons and positrons, and the positrons' annihilation. The diameter of the electron beam was 5 mm, and the geometry of the electron beam was set by the class G4UniformRand. Target parameters are described by using the methods of the class G4DetectorConstruction (defined components of the target, geometric parameters and materials, rendering options, etc.).

Figure 2 represents the results of our numerical calculation of the bremsstrahlung photons' flux in the target's location. The γ-quanta flux density was defined for each target by taking the thresholds of photonuclear reactions into account. It should be noted that, since the thresholds of the photonuclear reactions are different for different targets, the integrated γ-ray fluxes

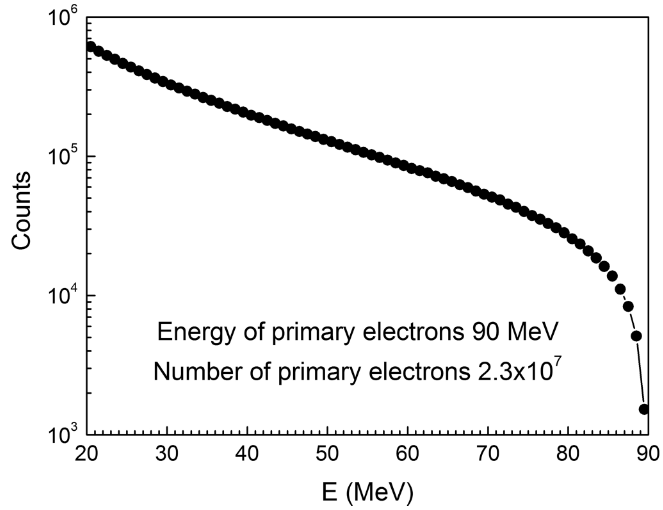

FIG. 2. Calculated bremsstrahlung photons' flux in the target's location.

computed in the range from the threshold to the maximum energy are different too.

The average cross section σ from various targets was computed using the following formula:

$$\sigma = \frac{A_0 A_m}{\Phi_0 m N_A (1 - e^{-\lambda t}) \times 10^{-24}}, \tag{1}$$

where σ is the cross section (barn), Φ_0 is the γ-quanta flux density (cm^{-2} s^{-1}), A_0 is the activity of a target (Bq), A_m is the atomic mass of a target isotope, N_A is Avogadro's number, m is the mass of a target isotope (g), λ is the decay constant of ^{7}Be, and t is the irradiation time of the target. The results of the calculation of the integrated fluxes of γ rays (from threshold to 90 MeV) and the average cross sections of ^{7}Be photoproduction on various targets are shown in Table I. The relative error in determining the cross section by taking the error of the activity measurement and the fluctuations of the beam current into account does not exceed 12%.

The average of the ^{7}Be cross section in this experiment on carbon coincides with the results from Ref. [5] according to which, the maximum cross section is equal to 0.3 mb. It should be noted that the cross section strongly depends on the correct choice of the threshold of the photonuclear reaction, which is determined not only by the number of nucleons knocked out of the nucleus, but also by the type of emitted particles, for instance, n, d, α, etc. In this experiment, it was impossible to fix the type of the emitted particles from the target nucleus. The thresholds of photonuclear reactions on nitrogen and oxygen were calculated by assuming that the ejected particles are nucleons with energies of 56 and 72 MeV, respectively. For a cross section of the ^{12}C(γ, $n\alpha$)^{7}Be reaction, the value of the threshold was assumed to be equal to 25 MeV, according to the data [5].

Although boron is not a component of the atmosphere, Table I also shows the data on measurements and the calculated cross section of the ^{11}B(γ,X)^{7}Be reaction. The cross section of this reaction is an order of magnitude smaller than the cross section of the ^{14}N(γ,X)^{7}Be reaction.

TABLE I. Average cross sections of photonuclear reactions with the ^{7}Be production in the energy range of γ rays from the threshold up to 90 MeV.

Target	Atomic mass (arb. units)	Target mass (g)	Integrated γ-ray flux per 1-μA electron beam (cm^{-2} s^{-1})	Activity (Bq)	Average cross section (barn)
C	12	0.910	8.77×10^{11}	1.83×10^{4}	$(2.58 \pm 0.30) \times 10^{-4}$
AlN		0.850			
N (from AlN)	14	0.300	1.00×10^{11}	1.32×10^{3}	$(5.77 \pm 0.70) \times 10^{-4}$
Al_2O_3		1.123			
O	16	0.480	5.60×10^{10}	6.78×10^{2}	$(3.79 \pm 0.45) \times 10^{-4}$
BN		0.561		3.14×10^{3}	
B (from BN)	11	0.247	8.77×10^{11}	1.76×10^{3}	$(8.42 \pm 1.01) \times 10^{-5}$
N (from BN)	14	0.314	1.00×10^{11}	1.38×10^{3}	$(5.77 \pm 0.70) \times 10^{-4}$

III. COMPUTER SIMULATION OF γ RAYS IN THE ATMOSPHERE

To perform numerical calculations with the γ rays in the energy range from ten to several hundred MeV, we used the software of the libraries GEANT4. The model is based on detailed Monte Carlo simulations, which use "PLANETOCOSMIC" numerical packages [10]. The module PLANETOCOSMIC enables one to simulate the nuclear-electromagnetic cascades due to the cosmic rays in the terrestrial atmosphere. The numerical package includes a wide range of functions to set the system geometry, particles, and interaction processes. The list of the interaction processes is rather extensive and includes electromagnetic and hadronic processes as well as decay and evolution processes of short-living particles in the energy range from 250 eV to several TeV. The simulation takes into account the terrestrial magnetic field as well as the influence of the Earth's surface (soil type) and enables one to calculate the resulting flux of different particles at a given altitude. All the details of the model can be found in Ref. [11].

We believe that the nucleonic-muon-electromagnetic cascade is initiated by primary galactic protons in the Earth's atmosphere. The energy distribution of primary galactic protons is given by formula [12],

$$J(E_p,\Phi) = C_p \frac{E_p(E_p + 2m_pc^2)(E_p + x + \Phi)}{(E_p + \Phi)(E_p + 2m_pc^2 + \Phi)}, \qquad (2)$$

where E_p is the proton kinetic energy (MeV), m_pc^2 is the proton rest energy (938 MeV), and Φ is the so-called modulating multiplier associated with the solar activity, which changes in the range from 400 to 1000 MeV at minimum and maximum solar activities, respectively. The empirical constants C_p and x take the following values: $C_p = 1.244 \times 10^{6}$ cm^{-2} s^{-1} MeV^{-1}, $x = 780\ \exp(-0.000\ 25E_p)$ (the numerical coefficient in the argument of the exponent has the dimension of inverse energy 1/MeV).

Some results of the computer simulation are shown in Figs. 3 and 4. The calculated equatorial γ-ray's flux at the altitude of 10 km in the Earth's atmosphere is shown in Fig. 3. To validate the results, we have compared the calculations with the available experimental data. The results of the calculation of the full γ-ray flux at moderate solar activity at the altitude of 5 km in the terrestrial atmosphere at temperate

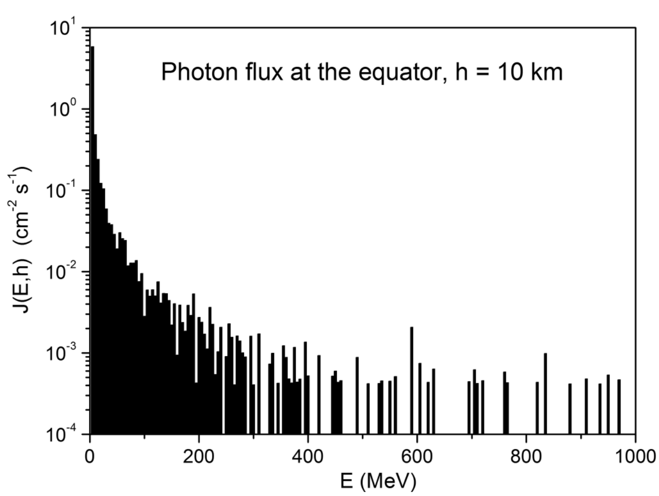

FIG. 3. Calculated equatorial γ-ray flux at the altitude of 10 km in the Earth's atmosphere at moderate solar activity.

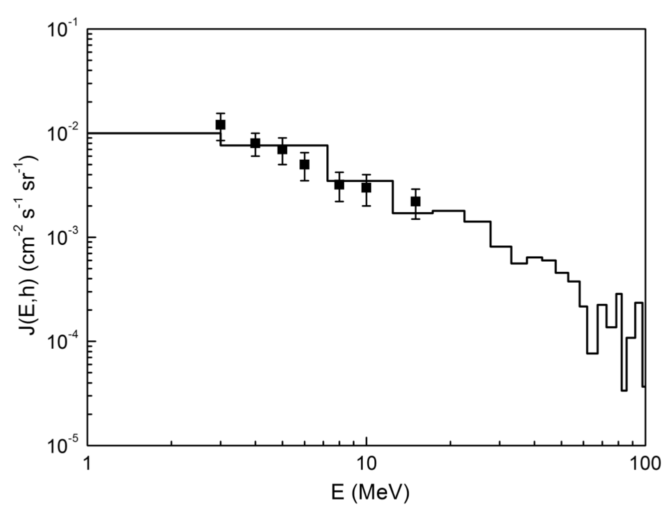

FIG. 4. Results of the calculation of the γ-ray flux at moderate solar activity at the altitude of 5 km in the terrestrial atmosphere (solid line) at temperate latitudes. Squares are the experimental data from Ref. [13].

latitudes show a satisfactory agreement (see Fig. 4) with the experimental data [13].[1]

IV. PHOTONUCLEAR ^{7}Be PRODUCTION IN THE ATMOSPHERE

The calculation of the production rate $P_i(h)$ of the ^{7}Be isotope in the ith photonuclear channel at the given altitude of the atmosphere h can be carried out by using the following formula:

$$P_i(h) = N_i(h) \int_{E_1}^{E_2} dE\, \sigma_i(E) J(E,h) \approx N_i(h)\langle\sigma_i\rangle \int_{E_1}^{E_2} dE\, J(E,h), \quad (3)$$

where $N_i(h)$ is the number of target atoms per 1 g of air at the altitude h, $\sigma_i(E)$ is the cross section of the corresponding photonuclear reaction, $\langle\sigma_i\rangle$ is the average cross section in a range from E_1 to E_2, and $J(E,h)$ is the photon flux. By setting the photon flux as $[J(E,h)] = \text{cm}^{-2}\,\text{s}^{-1}\,\text{MeV}^{-1}$, Eq. (3) gives the number of the ^{7}Be isotopes produced in 1 g of air at the given altitude for 1 s, i.e., $[P_i(h)] = \text{g}^{-1}\,\text{s}^{-1}$. Here, $E_1 = E_i^{\text{min}}$ is the photoformation threshold in the corresponding channel of the reaction, and E_2 is a maximum value. For the oxygen channel, according to Ref. [6], the integration interval can be extended up to $E_2 = 1$GeV. The lack of similar data for the nitrogen channel does not allow one to extend the integration to the high-energy region. However, analogous to the ^{16}O(γ,X)^{7}Be reaction, we can make an upper estimate by extrapolating the measured average cross section ^{14}N(γ,X)^{7}Be reaction (see Table I) up to energies of 1 GeV. Obviously, this leads to a significant increase in the ^{7}Be yield because nitrogen constitutes almost 80% of the atmosphere's composition.

Figure 5 shows the results of the numerical calculation of the photoproduction rate of the ^{7}Be isotope as a function of the altitude on the Earth's equator. The maximum production occurs at an altitude of 15–17 km. Figure 5 also shows the results of the calculation [3] of the ^{7}Be production due to protons and neutrons. The dotted line shows the predictive estimate by extrapolating the data in the nitrogen channel (Table I) for the energy of γ rays of 1 GeV. As can be seen, a contribution from the photonuclear mechanism to the ^{7}Be production in the equatorial region of the Earth's atmosphere is not small and is comparable to the contribution from protons and neutrons.

[1]Unfortunately we have to rely on findings from a conference poster because data [12] have not yet been published in a peer-reviewed piece of work.

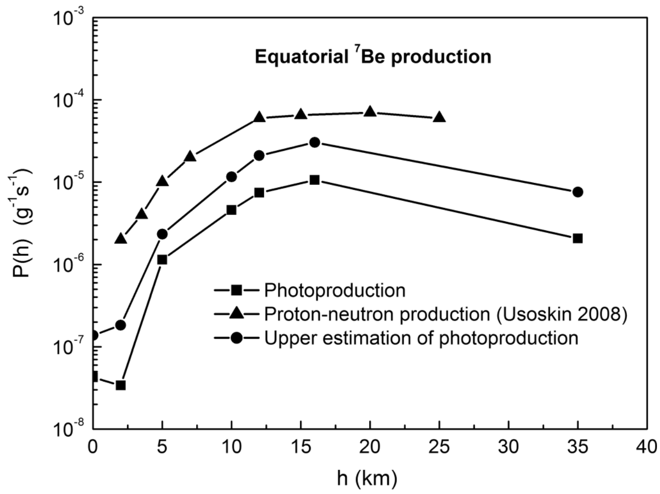

FIG. 5. Results of the numerical calculation of the rate of the photoproduction of ^{7}Be as a function of the altitude in the equatorial region. The contribution of the proton-neutron mechanism is shown for comparison (Ref. [3]).

V. CONCLUSION

The contribution from the photonuclear mechanism into the production of the cosmogenic ^{7}Be radionuclide in the Earth's atmosphere has been studied on the basis of the measured cross sections of the photoproduction of ^{7}Be. It has been shown that the contribution of the photonuclear mechanism is comparable to the contribution from the proton and neutron channels of the ^{7}Be formation in the atmosphere. The contribution of the photonuclear reactions to the total ^{7}Be production in the atmosphere is not less than 10%. The results obtained in this paper should be regarded as a lower limit for the photoproduction of ^{7}Be in the atmosphere. For a more detailed evaluation, one needs to know the data for the nitrogen photonuclear channel in the high-energy γ-ray region. Thus, our paper shows that the contribution from the photonuclear mechanism in the production of cosmogenic ^{7}Be should be taken into account in processes of accumulation and transfer of ^{7}Be in the terrestrial atmosphere.

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