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Key Points:

- Radiocarbon production by thunderstorm neutrons in regions with severe thunderstorm activity can be compatible to that by cosmic rays
- The observed enhancements of atmospheric neutron flux caused by thunderstorms may have important consequences for the radiocarbon dating

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Radiocarbon Production by Thunderstorms

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Abstract In view of the neutron flux enhancements observed in thunderstorms, a contribution of thunderstorm neutrons to atmospheric radiocarbon (isotope ${}^{14}_{6}C$) production is analyzed in connection with the archaeometry. Herein, estimates of neutron fluence per lightning electromagnetic pulse in regions with severe thunderstorm activity, at which a local rate of the ${}^{14}_{6}C$ production is comparable to the observed rates, are shown to be consistent with the measured magnitudes of thunderstorm neutron fluence. At present, available observations of atmospheric neutron and parent gamma ray flashes correlated with thunderstorms do not allow making final conclusions about thunderstorm contributions to ${}^{14}_{6}C$ production. For this, numerous studies of high-energy phenomena in thunderstorms are required, especially in the tropical belt where the thunderstorm activity is especially severe and where the ${}^{14}_{6}C$ production by galactic cosmic rays is almost independent of the solar activity disturbing the Earth's magnetic field shielding the Earth from cosmic rays.

Plain Language Summary Decay of radiocarbon accumulated in fossil biota is widely used in archaeometry. A capture by atmospheric nitrogen nuclei of neutrons, produced by cosmic rays, is causal to the radiocarbon production. In view of that thunderstorms produce neutrons their contribution in the radiocarbon production is analyzed. In regions with severe thunderstorm activity, evaluated neutron numbers per unit area (fluence) per lightning required for a rate of the radiocarbon production by thunderstorms to be comparable to the measured rate magnitudes are consistent with the measured magnitudes of thunderstorm neutron fluence. Available data on thunderstorm neutrons and parent to them gamma ray flashes do not allow making final conclusion about the thunderstorm deposition in the radiocarbon production. For this, numerous observations are required, especially in tropics where the thunderstorm activity is extremely strong and where the radiocarbon production is almost independent of the solar activity disturbing the Earth's magnetic field shielding the Earth from cosmic rays.

1. Introduction

Neutrons from natural sources are of invaluable importance "… because they provide quantitative information about the processes that create them and because the transmutations they produce can provide tools for understanding other physical, chemical, or chronological processes" (Fleisher, Plumer, & Crouch, 1974). As thunderstorm neutrons are injected directly into the troposphere, their significance would be enhanced on a small geographic scale provided that they constitute an appreciable fraction of atmospheric neutrons.

An accuracy of the radiocarbon (isotope ${}^{14}_{6}C$) dating (Bronk Ramsey, 2008; Libby, Anderson, & Arnold, 1949) is affected by variations in the atmospheric ${}^{14}_{6}C$ concentration. The variations are connected with variations in the galactic cosmic ray flux producing the radiocarbon and the Earth's magnetic field that shields the Earth from cosmic rays. On the radiocarbon dating time scales the cosmic ray flux is rather constant, but the screening ability of the Earth's magnetic field is affected by the solar activity; hence, variations in the latter lead to variations in the radiocarbon production rate (RPR) (Bronk Ramsey, 2008; Dorman, 2004, section 17; Gosse & Klein, 2015; Lingenfelter, 1963; Rakowski et al., 2015; Usoskin et al., 2013). On the other hand, Pavlov et al. (2013) connected the impulsive increase in the cosmogenic radiocarbon concentration in tree rings (of 12%) since with A.D. 750 with violent galactic gamma ray burst. The fluctuations of any origin limit "… the precision of radiocarbon calibration of single samples to at best a century… or in many cases much worse, quite regardless of the measurement precision" (Bronk Ramsey, 2008). The thunderstorm neutrons may have important consequences for the radiocarbon dating; in particular, they may add to the above uncertainties. A possible contribution of thunderstorm neutrons to the ${}^{14}_{6}C$ production is analyzed in this paper.

2. History of the Problem

The idea of C. Wilson (1924) about electron acceleration ("runaway" (Eddington, 1926)) in electric fields of thunderclouds is widely known. Less known is his idea of nuclear reaction occurrence in thunderstorms. The state of nuclear physics of those days, however, did allow him merely mentioning the nucleus disintegration or synthesis. Now it is known that neutrons are among possible daughter products of nuclear reactions, but in 1924 the neutron was not even discovered. Half a century later, Libby and Lukens (1973) revived the Wilson's hypothesis. Their interest to a possibility of the thunderstorm activity to generate neutrons was motivated by an attempt to account for the short-term variations of the ${}_{6}^{14}C$ concentration in tree rings. Suess (1965) found that the ${}_{6}^{14}C$ variations in tree rings correlate with sunspot numbers; therefore, he related the secular variations with fluctuations in the solar cosmic ray intensity. The latter is too low to affect directly the RPR; however, the Sun activity influences the atmosphere electric activity such that the frequency of thunderstorms depends on it. Libby and Lukens believed that plausible modulation of the ${}_{6}^{14}C$ production by lightning discharges and accumulation of this isotope in biota would explain the correlation noted by Suess.

The first, with null result, attempt (Fleisher, 1975) to detect thunderstorm-related neutrons was followed by communications claiming neutron flux enhancements in thunderstorms (Bratolyubova-Tsulukidze et al., 2004; Chilingarian et al., 2010, 2012, 2013, 2016, 2017; Gurevich et al., 2012; Ishtiaq et al., 2016; Kuroda et al., 2016; Kuzhewskiĭ, 2004; Martin & Alves, 2010; Shah et al., 1985; Shyam & Kaushik, 1999; Starodubtsev et al., 2012). Possibly, after Libby and Lukens the neutron generation in thunderstorms was related to $d(d,n)$ ${}_{2}^{3}He$ reactions in lightning channels (Fleisher, 1975; Kuzhewskiĭ, 2004; Shah et al., 1985; Shyam & Kaushik, 1999). However, the energy of deuterons in lighting channels is too low for these reactions to occur (Babich, 2006, 2007; Babich, 2014; Babich et al., 2014; Babich & Roussel-Dupré, 2007). As bright terrestrial γ ray flashes (TGFs) with duration Δt_{γ} on the order of 1 ms, detected in near space (Briggs et al., 2010; Connaughton et al., 2011; Fishman et al., 1994; Marisaldi, Fuschino et al., 2010; Marisaldi, Tavani et al., 2010; Smith et al., 2005; Tavani et al., 2011), aboard aircraft (Smith et al., 2011) and at the ground (Tran et al., 2015), and prolonged γ bursts (Δt_{γ} up to 10 min) detected on the ground (Chilingarian et al., 2010, 2012, 2013, 2016, 2017; Khaerdinov, Lidvansky, & Petkov, 2005; Kuroda et al., 2016; Torii et al., 2009; Tsuchiya et al., 2007, 2009, 2011, 2012) are correlated with thunderstorms and their spectra extend up to 100 MeV (Tavani et al., 2011), i.e., high above the thresholds $\varepsilon_{th,N}$= 10.55 MeV and $\varepsilon_{th,O}$= 15.7 MeV of photonuclear ${}_{7}^{14}N+\gamma\to{}_{7}^{13}N+n$, ${}_{8}^{16}O+\gamma\to{}_{8}^{15}O\ +n$ and electro-disintegration ${}_{7}^{14}N\ +e^{-}+\varepsilon_{e}\ \to{}_{7}^{13}N\ +\ n+e^{-}$, ${}_{8}^{16}O\ +e^{-}+\varepsilon_{e}\ \to{}_{8}^{15}O\ +\ n+e^{-}$ reactions; these reactions are the most obvious processes accounting for the thunderstorm neutrons (Babich, 2006, 2007, 2014; Babich et al., 2014; Babich & Roussel-Dupré, 2007; Carlson, Lehtinen, & Inan, 2010; Chilingarian et al., 2010, 2012). As the thunderstorm γ rays are the bremsstrahlung of relativistic runaway electron avalanches, capable of developing in thundercloud electric fields (Gurevich, Milikh, & Roussel-Dupre, 1992), a significant fact is that the average electron energy in the avalanches, of 6–7 MeV (Dwyer, 2008; Dwyer, Smith, & Cummer, 2012; Kutsyk et al., 2012a, 2012b), in electric field with the intensity below the self-breakdown limit in the atmosphere is not much below than $\varepsilon_{th,N}$.

In view of the first observations (Kuzhewskiĭ, 2004; Shah et al., 1985; Shyam & Kaushik, 1999) of neutron flux enhancements correlated with lightning electromagnetic pulse (EMP), Babich and Roussel-Dupré (2007) analyzed the ${}_{6}^{14}C$ production by thunderstorms. Allowing for only one competing reaction ${}_{7}^{14}N(n,\gamma\ ){}_{7}^{15}N$, they evaluated the volumetric (m^{-3}s^{-1}) RPR by thermal neutrons:

$$\dot{N}_{\text{thund,vol}}\left({}_{6}^{14}C\right)=N_{n1}\times R_{\text{flash,glob}}\times\delta/V_{\text{trop}},\tag{1}$$

where N_{n1} is the number of neutrons per lightning stroke, $R_{\text{flash, glob}}$ is the global rate of lightning flashes, δ is a portion of lightning EMPs correlated with the neutron production, $V_{\text{trop}}\ \approx\ 4\pi(R_{\text{Earth}})^{2}\times l_{\text{trop}}$ is the troposphere volume, $R_{\text{Earth}}\approx 6370$ km is the Earth's radius, and l_{trop} is the troposphere height. Magnitudes of all quantities in equation (1), especially δ and N_{n1}, strongly depend on measurements and calculations. With $R_{\text{flash, glob}}\approx 100$/s, $l_{\text{trop}}\approx 10$ km, $\delta=0.01$ (according to Shah et al. (1985)), and a number of thunderstorm neutrons $N_{n1}=10^{15}$ per flash, as Babich and Roussel-Dupré predicted, they obtained magnitude $\dot{N}_{\text{thund,vol}}\left({}_{6}^{14}C\right)\approx 2\times10^{-4}$ m^{-3}s^{-1} that is 2 orders of the magnitude lower than the rate 1.85×10^{-2} m^{-3}s^{-1} due to the cosmic irradiation of the atmosphere, which the authors estimated using the ${}_{6}^{14}C$ half-time and rather uncertain magnitude of the ${}_{6}^{14}C$ concentration in the atmosphere available in Prokhorov (1988). Besides, photonuclear neutrons, initially distributed in the MeV range (Babich, Bochkov, Kutsyk et al., 2010), experience a variety of interactions with the air nuclei during moderation. Babich and Roussel-Dupré (2007) concluded that thunderstorm neutrons nowadays deposit weakly into the $^{14}_{6}C$ concentration on average across the globe, but locally, especially in tropics, the $^{14}_{6}C$ production by thunderstorms, possibly, compete with that by cosmic irradiation.

In view of the shortcomings in Babich and Roussel-Dupré (2007) and given new experimental (Briggs et al., 2010; Chilingarian et al., 2010, 2012, 2013, 2016, 2017; Connaughton et al., 2011; Cummer et al., 2005, 2011; Gurevich et al., 2012; Ishtiaq et al., 2016; Kelley et al., 2015; Kuroda et al., 2016; Marisaldi, Fuschino et al., 2010; Marisaldi, Tavani et al., 2010; Martin & Alves, 2010; Starodubtsev et al., 2012; Tavani et al., 2011; Tsuchiya et al., 2007, 2009, 2011, 2012) and computational (Babich, 2014; Babich et al., 2007, 2008; Babich, Bochkov, Kutsyk et al., 2010; Babich, Bochkov, Donskoĭ et al., 2010; Babich, Bochkov, Kutsyk et al., 2013; Babich, Bochkov, Dwyer et al., 2013; Babich et al., 2014; Carlson et al., 2010; Celestin, Xu, & Pasko, 2012; Dwyer, 2008; Dwyer, Grefenstette, & Smith, 2008; Kelley et al., 2015; Tsuchiya et al., 2012; Xu, Celestin, & Pasko, 2012) data on thunderstorm γ rays and neutrons, reanalyzing of the problem of the $^{14}_{6}C$ production by thunderstorms is expedient. The new analysis is based not on a limited number of detections of thunderstorm neutrons as in Babich and Roussel-Dupré (2007) but on a vast amount of new data obtained from detecting of thunderstorm ground enhancements, i.e., enhanced fluxes of high-energy electrons, hard γ rays, and neutrons on the Earth's surface, which have been copiously collected on Mount Aragats, Armenia (3250 m above sea level) (Chilingarian et al., 2010, 2012, 2013, 2016, 2017) and at other sites worldwide: Japan (sea level and 2770 m above sea level) (Tsuchiya et al., 2007, 2009, 2011); Tibet, China (4300 m above sea level) (Tsuchiya et al., 2012); Himalayas, India (2743 m above sea level) (Ishtiaq et al., 2016; Shah et al., 1985); and Tien Shan, Kazakhstan (3340 m above sea level) (Gurevich et al., 2012).

It is pertinent to note that, as ranges of photons with energies $\varepsilon_\gamma \geq \varepsilon_{th,N}$ greatly exceed transverse sizes of lightning channels, the neutron-producing reactions occur outside the channels (Babich, 2014; Babich, Bochkov, Kutsyk et al., 2013; Babich, Bochkov, Dwyer et al., 2013; Babich et al., 2014). The neutron pulses may only correlate with lightning EMPs; rather frequently, they occur in advance of them or even do not correlate with them. Thus, the prolonged γ flashes (up to 1 min), parent to neutrons, were observed in advance of the EMPs (Tsuchiya et al., 2007, 2009, 2011) and, hence, were not produced by lightning discharges. Even TGFs, usually less than 1 ms in duration, occur within $-3/+1$ ms of the EMPs (Cummer et al., 2005, 2011). Therefore, the wording "correlation with lightning EMPs" is to be understood as a correlation with thunderstorms or even with thundercloud formations, because there is no other way to measure the global thunderstorm rate as via detecting of the lightning EMPs.

3. Numbers of Thunderstorm Neutrons Required for the Radiocarbon Production by Thunderstorms to be Comparable to the Observed Production

The neutron number N_{n1} produced in correlation with one lightning EMP may be calculated using the following balance equation:

$$\frac{dN_{n1}}{dt} = I - \frac{N_{n1}}{\tau_a}, \quad I = \begin{cases} S_{\text{source}}, & 0 \leq t \leq \Delta t; \\ 0, & t > \Delta t. \end{cases} \tag{2}$$

Here S_{source} is the neutron source with a duration equal to the parent γ pulse duration $\Delta t \approx \Delta t_\gamma$, $\tau_{abs} = 1/2N_{air}\upsilon_n\sigma_{abs}$, N_{air} is the air number density, υ_n is the neutron velocity, $\sigma_{abs} = \sum_i \sigma_i$ is the neutron absorption cross section, and σ_i is the neutron cross section for the *i*-type interaction with the air nuclei. A solution of this equation with the initial condition $N_{n1}(0)$ is as follows:

$$N_{n1}(t) = S_{\text{source}} \times \tau_a \times \begin{cases} 1 + \left(\dfrac{N_{n1}(0)}{S_{\text{source}} \times \tau_a} - 1\right) \times \exp(-t/\tau_a), & 0 \leq t \leq \Delta t; \\ \left(1 + \left(\dfrac{N_{n1}(0)}{S_{\text{source}} \times \tau_a} - 1\right) \times \exp(-\Delta t/\tau_a)\right) \times \exp\left(\dfrac{\Delta t - t}{\tau_a}\right), & t > \Delta t. \end{cases} \tag{3}$$

The $^{14}_{6}C$ production per lighting EMP can be evaluated using the balance equation:

$$\frac{dN_{\text{str}}\left(^{14}_{6}C\right)}{dt} = \frac{N_{n1}}{\tau_{14}}, \tag{4}$$

where $\tau_{14} = 1/2N_{air}\upsilon_n\sigma_{14}$ and σ_{14} is the ${}^{14}_{7}N(n,p){}^{14}_{6}C$ cross section. A decay of the ${}^{14}_{6}C$ nuclei is neglected because of too large half-time (5730 years) in comparison with τ_{14}.

Integrating equation (4), combined with equation (3), in the range from $t = 0$ to ∞ (actually, to $\Delta t + (2-3)\cdot\tau_{abs}$) with obvious conditions $N_{n1}(0) = 0$ and $N_{str}\left({}^{14}_{6}C\right) = 0$ at $t = 0$, gives

$$N_{str}\left({}^{14}_{6}C\right) = S_{source}\Delta t \times \frac{\tau_{abs}}{\tau_{14}} = N_{n1} \times \frac{\sigma_{14}}{\sigma_{abs}}. \tag{5}$$

This formula can be obtained by simply inserting equation (2) in equation (4) and integrating the result in the same range with the same conditions.

The global RPR is evaluated as follows:

$$\dot{N}_{thund}\left({}^{14}_{6}C\right) = N_{str}\left({}^{14}_{6}C\right) \times R_{flash} \times n_{str} \times \delta, \tag{6}$$

where R_{flash} is the global rate of lightning flashes per unit area (m^{-2} s^{-1}) and n_{str} is a number of strokes comprising one flash.

Babich and Roussel-Dupré (2007) evaluated the RPR using the neutron number $N_{n1} = 10^{15}$ which they predicted. But there are only a few measurements of the thunderstorm neutron numbers such that the N_{n1} magnitude is very uncertain; therefore, it is reasonable to calculate N_{n1} required to fit the global RPR (m^{-2} s^{-1}), which should include the rate due to the cosmic irradiation of the atmosphere $\dot{N}_{cosm}\left({}^{14}_{6}C\right)$ and thunderstorm rate $\dot{N}_{thund}\left({}^{14}_{6}C\right)$. A number of thunderstorm neutrons N_{n1}, required for the rate $\dot{N}_{thund}\left({}^{14}_{6}C\right)$ to constitute a portion k of the $\dot{N}_{cosm}\left({}^{14}_{6}C\right)$, are then obtained combining equations (5) and (6):

$$N_{n1} = \frac{\sigma_{abs}}{\sigma_{14}} \times \frac{k \cdot \dot{N}_{cosm}\left({}^{14}_{6}C\right)}{n_{str} \cdot R_{flash} \cdot \delta}. \tag{7}$$

This relation includes the RPR per unit area $\dot{N}_{cosm}\left({}^{14}_{6}C\right)$ that directly fits a dimension ($m^{-2}s^{-1}$) of the measured and computed rate magnitudes, whereas using the global flash rate $R_{flash,\,glob}(s^{-1})$ in relation (1) forced Babich and Roussel-Dupré (2007) to include division by $V_{trop} = 4\pi(R_{Earth})^2 l_{trop}$ with arbitrarily set $l_{trop} = 10$ km for comparing the result with rather uncertain magnitude of the volumetric RPR, which they themselves roughly evaluated.

For further evaluations, numerical magnitudes of the quantities in relation (7) are required. It is very convenient that N_{n1} is independent of the duration Δt_γ and altitude of the parent thunderstorm γ ray flashes because Δt_γ varies from hundreds of milliseconds up to tens of minutes (e.g., Chilingarian et al., 2010, 2012; Dwyer et al., 2012; Tsuchiya et al., 2012, and references therein). Besides, neutrons are produced at different altitudes and, consequently, at different air density because ranges of γ photons with energies above the photonuclear threshold $\varepsilon_{th,N}$ are extremely long.

Depending on the longitude, latitude, and season, the rate R_{flash} of lightning flashes (actually, the rate of EMPs) varies in a very wide range. In areas with high thunderstorm activity (the highest is on the land in the tropical belt (Christian & Latham, 1998)), the R_{flash} magnitude varies from 1 to 80 per square kilometer per year (Smith et al., 2005; Global Hydrology Resource Center data, http://ghrc.msfc.nasa.gov), i.e., from 3.3×10^{-14} to 2.7×10^{-12} m^{-2} s^{-1}. The global lightning frequency measured by Christian et al. (2003) and Ushio (2003) is of 45 s^{-1} (see also citation in Sato et al. (2008)). Dividing it by the area of the Earth's surface $S_{Earth} \approx 4 \times 10^{14}$ m^2 gives a magnitude of the global flash rate of 10^{-13} m^{-2} s^{-1}, which is within the above range of R_{flash}. The number of strokes is of $n_{str} = 3$ (Bazelyan & Rizer, 2000; Rakov & Uman, 2003). For the prolonged events (Chilingarian et al., 2010; Khaerdinov et al., 2005; Tsuchiya et al., 2007, 2009, 2011, 2012) with the γ bursts preceding the lightning EMPs, obviously, we should let $n_{str} = 1$.

The computed magnitudes of the rate $\dot{N}_{cosm}\left({}^{14}_{6}C\right)$ vary from 1.76×10^4 m^{-2} s^{-1} to $(2.5 \pm 0.5) \times 10^4$ depending on the longitude, latitude, and elevation above sea level (Bronk Ramsey, 2008; Damon, Lerman, & Long, 1978; Lingenfelter, 1963; Masarik & Reedy, 1995; O'Brien et al., 1978; Poluianov et al., 2016; Suess, 1965). Bronk Ramsey (2008) noted that "… attempts to directly measure the production rate either at ground level (Mak, Brenninkmeijer, & Southon, 1999) or at altitude (Bronk Ramsey et al., 2007) do not agree well with these estimates." On the other side, the magnitude $\dot{N}_{cosm}\left({}^{14}_{6}C\right) \approx 1.96 \times 10^4$ $m^{-2}s^{-1}$ computed by Poluianov et al.

(2016) agrees with the magnitude $(1.83 \pm 0.05) \times 10^4\ \text{m}^{-2}\text{s}^{-1}$ measured by Kanu et al. (2016) in stratosphere. So, for further estimations, it is reasonable to set $\dot{N}_{\text{cosm}}\left({}^{14}_{6}\text{C}\right) = 2 \times 10^4\ \text{m}^{-2}\text{s}^{-1}$.

Initially, the photonuclear neutrons, produced by γ rays with the bremsstrahlung spectrum of relativistic runaway electron avalanche (Babich et al., 2004), are distributed in the energy range from 10 to 16 MeV with a maximum at ≈13 MeV (Babich, Bochkov, Kutsyk et al., 2010) or below 10 MeV with a maximum at ≈1–2 MeV (Carlson et al., 2010). These neutrons interact with nuclei of the main constituents of the atmosphere (nitrogen and oxygen) and protons of the water hydrogen inside thunderclouds. The corresponding cross sections, available, for instance, in the database (https://www-nds.iaea.org/exfor/endf.htm), should be weighted by the abundance of the target species; however, to evaluate the ratio $\sigma_{\text{abs}}/\sigma_{14}$ within the accuracy of the present consideration, it is sufficient to allow for the reactions with nitrogen:

$$ {}^{14}_{7}\text{N}(n,\ \alpha){}^{11}_{5}\text{B},\ {}^{14}_{7}\text{N}(n,\ t){}^{12}_{6}\text{C},\ {}^{14}_{7}\text{N}(n,\ d){}^{13}_{6}\text{C},\ {}^{14}_{7}\text{N}(n,\ \gamma){}^{15}_{7}\text{N},\ {}^{14}_{7}\text{N}(n,\ p){}^{14}_{6}\text{C}. \tag{8} $$

The reasons are as follows:

First, consider the neutron capture by protons ${}^{1}_{1}\text{H}(n,\ \gamma){}^{2}_{1}\text{H}$. Inside cumulonimbus clouds, a magnitude of the water density $\rho_{\text{water}} \approx (0.33\text{–}1.7)\ \text{g m}^{-3}$ was measured at the altitudes h = 1–2.5 km (Shishkin, 1964). In a model "gas" of droplets with a radius $r_{\text{dr}} = 1\ \mu\text{m}$ suspended with a number density $n_{\text{dr}} = 10^9\ \text{m}^{-3}$ (McCarthy & Parks, 1992), the water density is somewhat higher: $\rho_{\text{water}}\ {}^{1}_{1}\text{H}(n,\ \gamma){}^{2}_{1}\text{H}$. Both ρ_{water} values are orders of the magnitude less than the dry air density $\approx\ 1.29\ \text{kg m}^{-3} \times \exp(-h/7.1)$ at the altitudes h of interest (troposphere and lower stratosphere). The ${}^{1}_{1}\text{H}(n,\ \gamma){}^{2}_{1}\text{H}$ cross section is much less than the ${}^{14}_{7}\text{N}(n,\ p){}^{14}_{6}\text{C}$ cross section: in the range of neutron energies from 10^{-5} eV to approximately 100 keV, a ratio of the ${}^{14}_{7}\text{N}(n,\ p){}^{14}_{6}\text{C}$ cross section to that of ${}^{1}_{1}\text{H}(n,\ \gamma){}^{2}_{1}\text{H}$ is approximately 5–10, and at the higher energies, it is on the order of hundreds (https://www-nds.iaea.org/exfor/endf.htm). Hence, the neutron capture by water protons is insignificant.

A deposition of other carbon-producing reactions is many orders of the magnitude less than that of ${}^{14}_{7}\text{N}(n,\ p){}^{14}_{6}\text{C}$, which gives more than 99% of the total RPR in the Earth's atmosphere (Dorman, 2004, p. 673). Interactions with oxygen, the second main constituent of the atmosphere, can also be neglected because of too low concentration (≈3.25 times less than the nitrogen concentration) along with that that the cross sections of reactions with oxygen, similar to equation (8), are rather close to or less than cross sections of reaction (8). As a result, for the initial neutron spectrum (10–16 MeV) (Babich, Bochkov, Kutsyk et al., 2010), the ratio $\sigma_{\text{abs}}/\sigma_{14}$ is about ≈5. Neutrons moderate in atmosphere; accordingly, $\sigma_{\text{abs}}/\sigma_{14}$ varies between 5 and 10 in the range from 10 MeV down to 1 MeV; below 1 MeV, where the reaction ${}^{14}_{7}\text{N}(n,\ p){}^{14}_{6}\text{C}$ dominates, $\sigma_{\text{abs}}/\sigma_{14} \approx 1$.

The δ magnitudes are the most uncertain. Now it is not clear if neutrons are produced by each thunderstorm. Obviously, the emitted neutron numbers per one thunderstorm EMP may vary in a wide range. Possibilities to detect thunderstorm neutrons and numbers of detected neutrons strongly depend on the distance to the neutron source, relief, air humidity, precipitations, etc. Thus, Shah et al. (1985) selected 124 events with the number of detected neutrons three or more above the background of 11,200 lightning EMPs, which gives $\delta \approx 0.011$. An inclusion of two-neutron events threefold increases the δ magnitude. On the other hand, during the observational period from year 2006 to 2009, Ishtiaq et al. (2016) recorded events with more than two neutrons correlated with each of sensored 150 EMPs; 19 and 13 of these events contained more than five and more than 10 neutrons, respectively. During May and June of the year 2006, when the thunderstorms occurred in the vicinity of the neutron monitor, 60 EMPs were sensored, with which 50 recorded events were correlated with ≥4 neutrons per event. In five of them more than 20 neutrons per event were recorded (Ishtiaq et al., 2016). Hence, according to these observations δ may vary from about 0.01 to 1 depending on the number of detected neutrons. According to the vast experimental data collected since 2009 at Mount Aragats station each thunderstorm is accompanied with thunderstorm ground enhancements of high-energy electron, γ ray, and neutron fluxes (Chilingarian et al., 2010, 2013, 2016, 2017). The γ photon energies exceed 20 MeV $> \varepsilon_{\text{th, N}}$= 10.55 MeV (Chilingarian et al., 2013). And what is more important, each lightning flash was preceded by the thunderstorm ground enhancements (Chilingarian et al., 2017). Hence, according to Mount Aragats observations $\delta = 1$. Therefore, in view of the δ uncertainty, the further evaluations will be carried out with $\delta = 0.01$, as in Babich and Roussel-Dupré (2007), and $\delta = 1$.

Table 1
Neutron Numbers N_{n1} Per Thunderstorm EMP Required for the Thunderstorm RPR $\dot{N}_{\mathrm{thund}}\left({}^{14}_{6}\mathrm{C}\right)$ to Constitute a Portion k of the Observed Rate $\dot{N}_{\mathrm{cosm}}\left({}^{14}_{6}\mathrm{C}\right) = 2\times10^{4}\ \mathrm{m}^{-2}\mathrm{s}^{-1}$ for $R_{flash} \approx 3.3\times10^{-14} - 2.7\times10^{-12}\ \mathrm{m}^{-2}\ \mathrm{s}^{-1}$ and n_{str} = 1–3

k	0.1				0.5			
n_{str}	1		3		1		3	
$\sigma_{\mathrm{abs}}/\sigma_{14}$	1	5	1	5	1	5	1	5
$N_{n1}/10^{15}$, $\delta = 1$; $N_{n1}/10^{17}$, $\delta = 0.01$	61–0.7	305–3.5	20–0.3	102–1.2	305–3.5	1525–18	100–1.5	510–6

Thereby, with the above magnitudes of R_{flash}, n_{str}, $\dot{N}_{\mathrm{cosm}}\left({}^{14}_{6}\mathrm{C}\right)$, δ, and $\sigma_{\mathrm{abs}}/\sigma_{14}$ for the initial and moderated neutron spectra, numbers of neutrons per lightning stroke N_{n1} required to account for 10% (k = 0.1), and a half (k = 0.5) of the observed RPR, at different areas of the globe varies as given in Table 1. Note that the fraction k = 0.1 corresponds to the observed fluctuations in the ${}^{14}_{6}\mathrm{C}$ production (Dorman, 2004, section 17; Lingenfelter, 1963; Pavlov et al., 2013; Rakowski et al., 2015; Suess, 1965; Usoskin et al., 2013).

The observational data on parent γ ray flashes do not allow computing initial neutron numbers because any experiment only provides gamma count rates and energies at the detector location, not the γ photon numbers and spectra at the γ source altitudes, but precisely, the latter are required for computing neutron numbers and energies to be compared with the data in Table 1. Conventionally, to obtain characteristics of photoneutron pulses, numerical simulations are used to compute rates and spectra of parent γ photons in the source placed at variable altitude; the source altitude is selected to fit the simulation results to the γ detector data. To avoid the inevitable uncertainties, inherent to this approach, it is reasonable to evaluate neutron fluence, required for the thunderstorm RPR to constitute a significant portion of the cosmic RPR, and compare the result directly with the fluence given by the measured neutron count rates.

4. Neutron Fluence Required for the Radiocarbon Production by Thunderstorms to be Comparable to the Observed Radiocarbon Production

The on-ground local neutron fluence per EMP Φ_{n1}, with which the thunderstorm RPR $\dot{N}_{\mathrm{thund}}\left({}^{14}_{6}\mathrm{C}\right)$ constitutes the portion k of the rate $\dot{N}_{\mathrm{cosm}}\left({}^{14}_{6}\mathrm{C}\right)$, is evaluated dividing equation (7) by a portion of the Earth's surface area with high thunderstorm activity $S \approx 0.3 \times S_{\mathrm{Earth}}$:

$$\Phi_{n1} = \frac{N_{n1}}{S} = \frac{\sigma_{\mathrm{abs}}}{\sigma_{14}} \times \frac{k\cdot\dot{N}_{\mathrm{cosm}}\left({}^{14}_{6}\mathrm{C}\right)}{n_{\mathrm{str}}\cdot R_{\mathrm{flash}}\cdot S\cdot\delta}. \tag{9}$$

Table 2 presents Φ_{n1} magnitudes satisfying relation (9) with k = 0.1 and the same $\dot{N}_{\mathrm{cosm}}\left({}^{14}_{6}\mathrm{C}\right)$, $\sigma_{\mathrm{abs}}/\sigma_{14}$, R_{flash}, n_{str}, and δ magnitudes as in Table 1. With k = 0.5 the Φ_{n1} data are to be correspondingly multiplied.

In Table 3, magnitudes of the neutron fluence Φ_n are presented either detected at various altitudes h_{det} or obtained by numerical simulations in the framework of the photonuclear origin of the neutrons for different h_{det} and altitudes of the parent γ ray source h_{γ}. It is seen that with $\delta = 1$ the Φ_n magnitudes are rather close or even exceed the evaluations in Table 2. With $\delta = 0.01$ the Φ_n magnitudes are consistent with the evaluations in Table 2 for regions with severe thunderstorm activity. Hence, there is a chance that thunderstorm RPR locally constitutes a significant portion (at least 0.1) of the cosmic RPR.

Note that observations are only capable of providing local magnitudes of the neutron fluence; therefore, it is impossible to identify the Earth's surface area they relate to, because neither coordinates or dimensions of the γ source are known. But observations of thunderstorm neutrons are not numerous; therefore, the data in Table 3 cannot be considered as close to the average neutron fluence per thunderstorm EMP throughout the globe.

Table 2
Neutron Fluence Per Thunderstorm EMP Required for the Thunderstorm RPR to be Compatible to the Observed Rate, k = 0.1

n_{str}	1		3	
$\sigma_{\mathrm{abs}}/\sigma_{14}$	1	5	1	5
Φ_{n1} (m^{-2}), $\delta = 1$; $\Phi_{n1}/100$ (m^{-2}), $\delta = 0.01$	556–6	2780–30	185–2	925–10

Table 3
On-Ground Neutron Fluence Φ_n at Different Detection Altitudes h_{det}

		Neutron fluence (m^{-2})							
		Experiment					Computed		
h_γ (km)	h_{det} (km)	(a)	(b)	(c)	(d)	(e)	(f)	(g)	(h)
?	2.74	30–670		56–700					
?	3.25		5×10^4						
15–5	0						$0.03–7 \times 10^2$		
12–8	3						$(0.35–4) \times 10^2$		
?	3.34					$(2–3) \times 10^4$			
4–2	0							$2\ (10^3–10^5)$	
5–3.5	3							$(0.9–2) \times 10^7$	
5	0								3×10^2
2.5	0								10^4
5.2	4.3				1.4×10^4				

Note. (a) Shah et al. (1985); (b) Chilingarian et al. (2010, 2012); (c) Ishtiaq et al. (2016); (d) Tsuchiya et al. (2012); (e) Gurevich et al. (2012); (f) Babich, Bochkov, Kutsyk et al. (2010); (g) Babich, Bochkov, Donskoĭ et al. (2010); (h) Carlson et al. (2010).

5. Conclusions

Computed magnitudes of the neutron fluence per thunderstorm EMP (an indicator of the global thunderstorm activity), required for the ${}^{14}_{6}C$ production by thunderstorms in regions with severe thunderstorm activity (flash rate R_{flash} of 10^{-12} $m^{-2}s^{-1}$, δ of unity) to be comparable to that due to cosmic irradiation, are compatible with the available measured and computed magnitudes of the thunderstorm neutron fluence. Thus, nowadays the thunderstorm-produced neutrons may locally contribute to the ${}^{14}_{6}C$ concentration. Babich and Roussel-Dupré (2007) concluded that "if … some sizable fraction of the ${}^{14}_{6}C$ is deposited locally (e.g., absorbed by the local biota) on a time short compared to the redistribution by circulation, and account for the fact that lightning is concentrated over land and that the lightning rate varies by orders of magnitude over various regions then it is possible for lightning produced ${}^{14}C$ to compete with the cosmic irradiation in these regions." The enhancements of atmospheric neutron flux caused by thunderstorms may have important consequences for the radiocarbon dating. In particular, thunderstorm-produced neutrons may relate to anomalies in the radiocarbon dating. The ages of various materials may be underestimated unless the historical occurrence rate and geographical distribution of thunderstorms are taken into account.

However, only few papers, communicating observations of thunderstorm neutrons, are available by now. Most of them only communicate observations of neutron flux enhancements roughly correlated with lightning EMPs without presenting numerical data. Now it is not even clear if neutrons are produced by each thunderstorm. Thus, Shah et al. (1985) at High Altitude Research Laboratory, Gulmarg, Kashmir, India, only selected 124 events with the number of detected neutrons three or more above the background of 11,200 lightning EMPs. On the other hand, Ishtiaq et al. (2016) detected neutrons correlated almost with each registered EMP at the same setup. According to vast data from observations at Aragats Space Environmental Center of the Yerevan Physics Institute, Armenia, conducted since 2009, neutrons with energies above the photonuclear threshold $\varepsilon_{th,N}$= 10.55 MeV are produced by each thunderstorm (Chilingarian et al., 2013, 2017). Moreover, the high-energy phenomenon in thunderstorms is a new and, therefore, weakly elaborated field of the atmospheric electricity. The number of observations of the X-ray, γ ray, and neutron pulses correlated with thunderstorms is rather limited.

Besides, as Bronk Ramsey (2008) noted, the global RPR is "… surprisingly poorly known." The difference between computed and measured magnitudes is significant; thus, Mak et al. (1999), who measured the RPR at different attitudes and latitudes, noted that the calculated rate (Lingenfelter, 1963) "… at low altitudes overestimates the actual production rate by a factor of 2."

In view of the problem significance, more accurate and numerous experimental studies of the thunderstorm X-rays, γ rays, and neutrons are required at different longitudes, latitudes, and altitudes. It would be very interesting to measure the ${}^{14}_{6}C$ production in tropics, where the thunderstorm activity is especially severe. The thunderstorm activity is strongly affected by the solar activity; but the ${}^{14}_{6}C$ production by galactic cosmic rays at low latitudes is almost independent of the solar activity (Dorman, 2004, p. 676; Lingenfelter, 1963). Hence, if the ${}^{14}_{6}C$ production in tropics appears dependent on the solar activity, these additional ${}^{14}_{6}C$ nuclei are possibly produced by thunderstorms.

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