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ON THE INFLUENCE OF CLIMATIC FACTORS ON THE RATIO BETWEEN THE COSMOGENIC ISOTOPE ^{14}C AND TOTAL CARBON IN THE ATMOSPHERE IN THE PAST

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Abstract: Radiocarbon ^{14}C is a cosmogenic isotope, which is most extensively used by scientists from a wide variety of fields. Its rate of generation in the atmosphere depends on solar modulation and thus, studying ^{14}C concentration in natural archives, one can reconstruct solar activity level in the past. The paper shows results of box-model calculations of generation of the ^{14}C isotope in the atmosphere and its relative abundance during the time interval 1389–1800 AD, taking into account influence of changing climate. This interval includes the deep minimum of solar activity and period of significant change in atmospheric concentration of CO_2 and global temperature. The performed analysis showed that concentration of ^{14}C in the atmosphere reflects not only variations of the galactic cosmic rays intensity but as well changes of temperature and atmospheric CO_2 concentration. It is shown that the decrease in CO_2 concentration in the atmosphere during 1550–1600 can be connected with absorption of CO_2 by the ocean surface layer. Thus, taking into account the climatic changes is an important condition for the reconstruction of solar activity in the past using data based on cosmogenic isotopes.

Keywords: solar activity, climate change, cosmogenic isotopes ^{14}C and ^{10}Be.

1. INTRODUCTION

It is well known that the cosmogenic isotopes ^{10}Be and ^{14}C are generated in the Earth's atmosphere due to the influx of galactic cosmic rays (GCR) (Beer *et al.*, 1990; Dergachev and Veksler, 1991). The major part of ^{14}C is produced by secondary thermal neutrons in the reaction $^{14}N(n,p)^{14}C$. Neutrons are present in the atmosphere as a product of the cosmic-ray-induced cascade originated by cosmic ray particles with energy more than 1 GeV/nucleon. After formation ^{14}C is oxidized rapidly to ^{14}CO, and after 1.0–1.5 months to $^{14}CO_2$ (Lowe and Allan, 2002). $^{14}CO_2$, in turn, is mixed and homogenized in the atmospheric $^{12}CO_2$ pool and involved in a chain of geophysical and geochemical processes forming the global carbon cycle. Cosmogenic beryllium is generated in the atmosphere in nuclear spallation reactions $^{14}N(Ha,X)^{10}Be$, $^{16}O(Ha,X)^{10}Be$, where Ha are hadrons of

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the cosmic-ray-induced atmospheric cascade, X are the other reaction products.

The intensity of cosmic rays is modulated by both variations of interplanetary magnetic and changes in the geomagnetic dipole field, which shields terrestrial atmosphere from cosmic particles at low and middle latitudes. The interplanetary magnetic field extends from the Sun with the solar wind towards the interplanetary space and changes with the corresponding change of solar activity. That is why radiocarbon concentration in tree rings, varves and peat sediments as well as ^{10}Be in polar ice are sensitive to variations in both Sun's activity (SA) and the geomagnetic field.

The ^{10}Be generated in the atmosphere is deposited on the Earth's surface and accumulates in the terrestrial reservoirs (polar ice, oceanic and lake sediments *etc.*) while the generated ^{14}C takes part in the natural carbon cycle through exchange between different carbon reservoirs. It is taken up by plants as $^{14}CO_2$ and fixed for instance in annual tree rings (Libby, 1955). Thus both ice core layers and tree rings contain information about past changes in production of cosmogenic isotopes. In that case, records of the abundance of these isotopes in well dated samples help us to investigate changes of SA in the past, solving direct (e.g. Ogurtsov, 2004) and inverse (e.g., Nagovitsyn, 2007) problems of a ^{14}C balance between natural reservoirs. Climatic changes leading to variation of CO_2 concentration in the atmosphere influence on the atmospheric concentration of ^{14}C and also on ^{14}C concentration in terrestrial archives. Climatic effect in ^{14}C concentration can be manifested more visible during the sharp climatic changes, for example, during the abrupt cold event — the Younger Dryas, which took place in the Northern Hemisphere during 12900–11500 years BP. Simulations, performed by Meissner (2007) showed that both the rise in atmospheric CO_2 concentrations derived from ice core and sharp rapid changes in the atmospheric ^{14}C concentration (Δ^{14}C), comparable to the peak recorded in the paleoceanographic records of the Cariaco Basin, during the Younger Dryas can be a result of a complete shutdown and recovery of the Atlantic Meridional Overturning Circulation. Singarayer *et al.* (2008) analyzed the atmospheric Δ^{14}C increase at Younger Dryas and arrived at the conclusion that this increase was likely caused by ocean reorganization rather than by increase of ^{14}C production rate. Butzin *et al.* (2012) showed that processes of ocean ventilation influenced the concentration of ^{14}C in the atmosphere during the last deglaciation (17.5–14.5 cal ka BP).

When investigating and reconstructing solar activity in the past, one should take into account that the abundance of ^{14}C measured in laboratories is determined relative to the stable carbon isotope, *i.e.* laboratory measurement reconstructs the ratio of abundance of ^{14}C to the total abundance of carbon in the atmosphere in the year of the annual tree ring formation. Thus, reconstructed values for Δ^{14}C contain information on ratio of concentrations of ^{14}C to ^{12}C concentration in the atmosphere. It is therefore important to distinguish between changes of Δ^{14}C, caused by GCR variations, and variations of Δ^{14}C caused by climatic changes, because terrestrial records of Δ^{14}C are influenced not only by atmospheric production but also by changes in climate, which influence on the exchange of carbon between different reservoirs and therefore on the ^{14}C/^{12}C ratio. Climatic changes can influence the radiocarbon cycle as a result of different processes: the above mentioned oceanic circulation, changes in biosphere, variations in rate of exchange between the surface and deep layers of the ocean and between ocean and atmosphere due to changes in global temperature. Changes in sea-ice cover could also influence the exchange between ocean and atmosphere. An increase in the sea ice cover can diminish the intensity of ocean-atmosphere exchange.

Dergachev and Ostriakov (1978) emphasized that temperature variations influencing the concentration of ^{14}C in the atmosphere should be taken into account. It is known that warming of water results in a decrease of the concentration of dissolved carbon dioxide, *i.e.* degassing appears. That is why it is necessary to take into account temperature dependence of the rate of exchange of carbon between different reservoirs during periods of variations in global temperature (e.g., during the Little Ice Age). Variations in the surface ocean temperature near Antarctica over the last 12 000 years, studied by Shevenell *et al.* (2011), show that amplitudes of the variations reached up to a few degrees and were similar to global temperature variations. Moreover, in the middle of the second millennium AD, a sharp cooling of the ocean surface water, coinciding with the Little Ice Age, was observed. The decrease of the water temperature reached 2–3 degree (Shevenell *et al.*, 2011). After this a warming appeared. Such variations of the surface ocean layer temperature should result in a redistribution of carbon between ocean and atmosphere and the effect should consequently be taken into account when investigating solar activity in the past using data on ^{14}C abundance in annual tree ring records.

2. CALCULATION OF THE ABUNDANCE OF ^{14}C IN THE ATMOSPHERE

In order to study the abundance of ^{14}C in the atmosphere we perform a model simulation based on a five-reservoir carbon exchange model as shown in **Fig. 1** (see, e.g., Dorman, 1978). In that case abundance of ^{14}C in the atmosphere, biosphere, humus, upper and deep ocean layers is described by the following system of differential equations:

$$\frac{dN_a}{dt} = Q(t) - (\lambda + \lambda_{ab} + \lambda_{amO})N_a + \lambda_{ba}N_b + \lambda_{ha}N_h + \lambda_{mOa}N_{mO} \quad (2.1)$$

$$\frac{dN_b}{dt} = \lambda_{ab}N_a - (\lambda + \lambda_{ba} + \lambda_{bh})N_b \quad (2.2)$$

$$\frac{dN_h}{dt} = \lambda_{bh} N_b - (\lambda + \lambda_{ha}) N_h \quad (2.3)$$

$$\frac{dN_{mO}}{dt} = \lambda_{amO} N_a - (\lambda + \lambda_{mOa} + \lambda_{mOdO}) N_{mO} + \lambda_{dOmO} N_{dO} \quad (2.4)$$

$$\frac{dN_{dO}}{dt} = \lambda_{mOdO} N_{mO} - (\lambda + \lambda_{dOmO}) N_{dO} \quad (2.5)$$

N_a, N_b, N_h, N_{mO}, N_{dO} are values of the ^{14}C isotope content in the atmosphere, biosphere, humus, and upper (mixed) and deep layers of the ocean. $Q(t)$ is the ^{14}C production rate in the atmosphere.

$\lambda = 1.21 \times 10^{-4}$ year^{-1} — the rate of radiocarbon decay; λ_{ab}, λ_{amO}, λ_{ba}, λ_{ha}, λ_{mOa}, λ_{bh}, λ_{mOdO}, λ_{dOmO} — are the rates of ^{14}C exchange between atmosphere and biosphere, atmosphere and upper ocean layer, biosphere and atmosphere, humus and atmosphere, upper ocean and atmosphere, biosphere and humus, upper ocean and deep ocean, deep ocean and upper ocean correspondingly. In our calculations we used the rates of ^{14}C exchange after Dorman (1978) — see **Table 1**. We calculated the rate of ^{14}C production ($Q(t)$) in the atmosphere, according to Beer *et al.* (1994), as proportional to the production rate of ^{10}Be which, in turn, is proportional to the concentration of ^{10}Be in polar ice.

It should be noted, however, that a link between the production rate of ^{10}Be and beryllium concentration in polar ice might be more complicated. Since the rate of ice generation is determined by precipitation and, therefore, concentration of ^{10}Be in polar ice is dependent on local meteorological conditions. The flux of ^{10}Be is influenced by variations in tropospheric inter-latitude mixing and exchange between stratosphere and troposphere. That is why fluctuations of local and regional meteorological parameters could affect both short-term (periods less than 10 year) (Beer *et al.*, 1990) and longer-term (multidecadal and longer periods) (Lal, 1987; Bard *et al.*, 2000) variations of beryllium concentration in ice. Recently, researchers began to use ^{10}Be flux instead of concentration (Berggren *et al.*, 2009). Beryllium flux tells us the rate at which beryllium atoms were deposited per unit area and time (Beer *et al.*, 2012). Some researchers believe that the flux is less influenced by meteorological fluctuations. However, Beer *et al.* (2012) noted that in fact neither concentration nor flux reflect the production rate correctly and that is one of the limiting factors of the cosmogenic radionuclide method. More correct account for the complexity of meteorological factors, influencing ^{10}Be transport is needed to solve the problem.

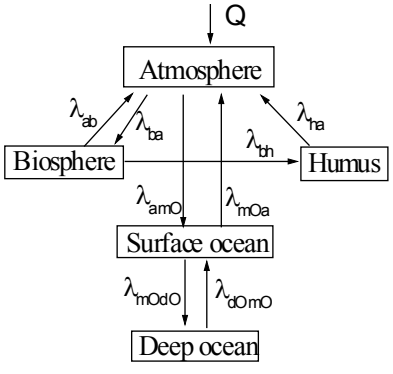

Fig. 1. *The five-reservoir model of the carbon exchange system used in the calculations.*

Thus, the abundance of cosmogenic beryllium (as well as radiocarbon) in natural archives should reflect time variations of: (a) a flux or the form of GCR spectrum in the vicinity of the solar system; (b) solar magnetic activity; (c) intensity of a dipole geomagnetic field of the Earth; (d) global and regional climate.

In general, all the rates of carbon exchange between reservoirs, described above, depend on climatic change but in this work we consider the influence of temperature only on the rate of CO_2 transition between the upper ocean layer and atmosphere, as this is the most important process in the carbon cycle, at least during the reference time interval. Possible influence of variations of other exchange rates (particularly between surface and deep ocean layers) will be considered in further works.

To take this into account we replace λ_{mOa} with a temperature dependent parameter $\lambda_{mOa} = (1+k\cdot\Delta T)\,\lambda^0_{mOa}$, where ΔT is the variation of global temperature and k a numerical coefficient. We consider that the temperature of the upper ocean layer changes synchronously with the change of the global atmosphere air temperature, and we describe the surface air temperature using the available proxy reconstructions.

When we selected the value of k, we took into account the following circumstances. The net air-sea flux (F) of CO_2 is a sum of the flux coming to the surface ocean layer from the atmosphere and the opposite flux from the surface ocean layer can be described by the following equation (see for example, Siegenthaler and Sarmiento, 1993)

$$F = k_g (pCO_{2,a} - pCO_{2,s}) \quad (2.6)$$

where k_g is a gas exchange coefficient, $pCO_{2,s}$ is the equilibrium partial pressure of CO_2 in surface water layer, $pCO_{2,a}$ is partial pressure of CO_2 in the air.

Eq. 2.6 shows that if the partial pressure of CO_2 in the surface ocean layer is less than in air the total flux will be positive, *i.e.* the ocean will take up carbon dioxide from the atmosphere. If $pCO_{2,a} < pCO_{2,s}$ then flux from the ocean to the atmosphere will exceed flux from the atmosphere to the ocean and F becomes negative.

Table 1. *Parameters of the five-reservoir model.*

λ_{ab} = 1/30 year^{-1}	λ_{amO} = 1/6.84year^{-1}	λ_{ba} = 1/30.6 year^{-1}	λ_{ha} = 1/101.2 year^{-1}	λ^0_{mOa} = 1/8.5 year^{-1}
λ_{bh} = 1/30.6 year^{-1}	λ_{mOdO} = 1/3 year^{-1}	λ_{dOmO} = 1/193year^{-1}		
$N_a(t_0)$ = 7.52×10^9 cm^{-2}	$N_b(t_0)$ = 3.88×10^9 cm^{-2}	$N_h(t_0)$ = 1.25×10^{10} cm^{-2}	$N_{mO}(t_0)$ = 8.77×10^9 cm^{-2}	$N_{dO}(t_0)$ = 5.51×10^{11} cm^{-2}

Change of the surface layer temperature influences appreciably the value of partial pressure of CO_2 in surface water and hence the CO_2 flux from the ocean to the atmosphere. According to (Takahashi *et al.*, 1993), an increase (or decrease) of temperature of the surface water layer by 1 degree results in an increase (or decrease) of the partial pressure of CO_2 dissolved in water by ca 4% for a temperature range (2–28)°C and for a salinity range of (34–36) per mil. As it has been noted above during the Little Ice Age the temperature of the surface water layer can change by a few (2–3) degrees (Shevenell *et al.*, 2011). In that case, the flux of carbon dioxide (and, hence, λ_{mOa}) from the surface layer to the atmosphere could have changed by up to ≈10%, and that we take into account in our calculations. As we have noted above, in our calculations we use the data on change of the temperature of the surface layer of air (not the surface layer of ocean). We take into account that variations of this temperature are less than 1°C and, hence, if k = 0.1°C^{-1} variations of λ_{mOa} are less than 10%.

Using the temperature reconstruction from Esper *et al.* (2002) (**Fig. 2A**) and the ^{10}Be concentration data from the NGRIP ice core (Berggren *et al.*, 2009) (see **Fig. 2B**) we obtain the results shown in **Fig. 2C**. Results of calculations showed with lines 1-3 in **Fig. 2C** correspond to the initial conditions, listed in **Table 1**. These concentrations of radiocarbon in the work of Dorman (1978), cited above, are considered as the equilibrium. The initial time used in our calculations is t_0 = 1389 AD.

According to Dorman (1978), the mean value of Q in the middle of the 20th century was 7.06·10^7 cm^{-2}·year^{-1}. In our calculations we use the value $Q(t_0)$ = (6–8)·10^7 cm^{-2}·year^{-1}. The results of calculations performed for Q(1389) = 7·10^7 cm^{-2}·year^{-1} are shown in **Fig. 2**. Results, obtained for other Q values, are similar.

Curves 1, 2 and 3 correspond to k = 0, 0.05 and 0.10 respectively that corresponds to the change of CO_2 transition from the upper ocean layer to atmosphere by 0, 5 and 10% when the global air temperature varies by 1 degree.

The results of calculations of the abundance of ^{14}C in the atmosphere (curves 1–3) for the time interval 1389–1800 AD and data on Δ^{14}C from Stuiver and Braziunas (1993) (curve 5) are shown in **Fig. 2C**. The data are given as deviations (in %) from the level of 1822 AD, because Δ^{14}C = 0 in this year according to the record of Stuiver and Braziunas (1993). One can see from the figure that the curves have apparent common features: minima of solar activity, which correspond to maxima of ^{14}C abundance, are clearly manifested. However some differences between the calculated curves 1–3 and curve 5 are obvious.

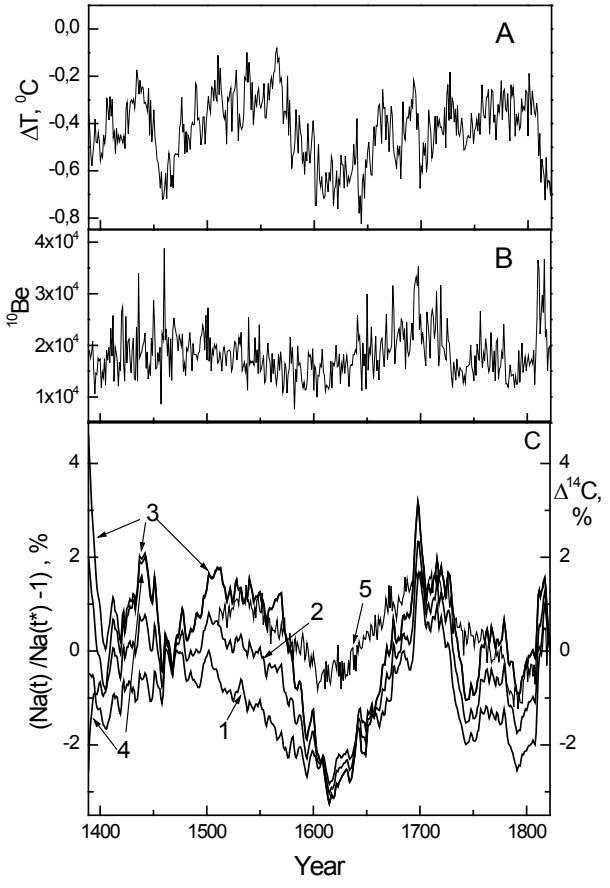

Fig. 2. *A — Variation of global surface temperature according to Esper* et al. *(2002); B — Concentration of ^{10}Be in Greenland ice layers according to Berggren* et al. *(2009); C — Calculated values of variations of ^{14}C concentration in the atmosphere; $(N_a(t)–N_a(t^*))/N_a(t^*)$ for* k = *0, 0.05 and 0.10 — curves 1–3 respectively, t^* = 1822. Curve 4 corresponds to the same parameters as curve 3 but with other abundance of ^{14}C in the atmosphere ($N_a(t_0)$ = 7×10^{9}cm^{-2}). Curve 5 corresponds to experimental Δ^{14}C according to Stuiver and Braziunas (1993).*

Here we should note that the sharp decrease of the calculated value of ^{14}C abundance in the atmosphere (curve 2 and 3) during 1389–1405 is connected with the tendency of the radiocarbon concentration in the atmosphere to the equilibrium state. As at the starting point (t_0 = 1389 AD) temperature anomaly $\Delta T \neq 0$ for every value of the coefficient k values λ_{mOa} will be different (because $\lambda_{mOa} = (1+k\cdot\Delta T)\,\lambda^0_{mOa}$) and, thus, equilibrium values of ^{14}C concentration will be different as well. Moreover, as at t_o = 1389 AD $\Delta T < 0$, the value λ_{mOa} will decrease when coefficient k increases. A smaller equilib-

rium value of radiocarbon concentration in the atmosphere will correspond to a smaller value of $\lambda_{mOa,}$ because the flux of radiocarbon from ocean to atmosphere will decrease. Let us consider changes of the results of calculation due to a choice of the initial value $N_a(t_0)$. Curve 4 describes results of calculations performed using parameter values of the analogous curve 3, but with $N_a(t_0) = 7\times10^9$ cm^{-2}. As one can see from the figure, the value N_a will increase at the initial time moment and that testifies that the chosen value $N_a(t_0)$ exceeds a bit the equilibrium value for the considered λ_{mOa}. The differences between curves 3 and 4 (**Fig. 2**) disappear after 1450 and have no influence during the time interval 1500–1800 considered in the article.

It should be noted that direct comparison of the variations, calculated above, and variations of $\Delta^{14}C$ is fully substantiated only for periods over which concentration of CO_2 in the atmosphere is constant. The cause of this could be that the value $\Delta^{14}C$ delineates variations of ratio of the isotope ^{14}C to the concentration of the stable isotope ^{12}C and concentration of CO_2 in the atmosphere (and isotope ^{12}C) change with time.

3. VARIATIONS OF CO_2 CONCENTRATIONS IN THE ATMOSPHERE FROM 16th TO 18th CENTURIES

Let us calculate change of CO_2 concentration in the atmosphere from the beginning of 16th up to the end of 18th century by comparing the values of the calculated ^{14}C abundance in the atmosphere with the measured $\Delta^{14}C$ values. We assume the following:

1) Variations of the absolute abundance of ^{14}C in the atmosphere correspond to that, calculated above;
2) Variations of $^{14}C_a/^{12}C_a$ ratio (ratio of concentration ^{14}C and ^{12}C in the atmosphere) are equal to the values, obtained by Stuiver and Braziunas (1993).

I.e. the following condition is satisfied:

$$\frac{^{14}C_a(t)/^{12}C_a(t) - {}^{14}C_a(t^*)/^{12}C_a(t^*)}{^{14}C_a(t^*)/^{12}C_a(t^*)} = \Delta^{14}C(t)/100 \quad (3.1)$$

where $\Delta^{14}C$ it is expressed in percent.

Then, from **Eq. 3.1**, we obtain the variations;

$$\frac{^{12}C_a(t)}{^{12}C_a(t^*)} = \frac{^{14}C_a(t)}{^{14}C_a(t^*)} \quad \frac{1}{1+\Delta^{14}C(t)/100} \quad (3.2).$$

Because

$$\frac{^{14}C_a(t)}{^{14}C_a(t^*)} = \frac{N_a(t)}{N_a(t^*)} \quad (3.3)$$

we obtain from **Eq. 3.2**:

$$\frac{^{12}C_a(t)}{^{12}C_a(t^*)} = \frac{N_a(t)}{N_a(t^*)} \quad \frac{1}{1+\Delta^{14}C(t)/100} \quad (3.4),$$

where $N_a(t)$ is calculated from **Eqs. 2.1–2.5**.

Thus, when changes in $^{12}C_a$ abundance (and CO_2 concentration in the atmosphere) have such a form, the calculated variations $^{14}C/^{12}C$, will coincide with the values measured by Stuiver and Braziunas (1993). The results of the calculations for different values of the coefficient k mentioned above are shown below in **Fig. 3**.

One can see from **Fig. 3** that the calculated concentration of ^{12}C (and, hence, CO_2) in the atmosphere should be practically stable over the period 1550–1650 AD, if the rate of carbon transport from the upper ocean layer to atmosphere (**Fig. 3**, curve 1) is constant. However, the measurements show that a decrease of the CO_2 concentration takes place over this time interval (**Fig. 4**), which decreases simultaneously with the global temperature (**Fig. 2A**). If we take into account temperature dependence of the rate of exchange of carbon between the upper

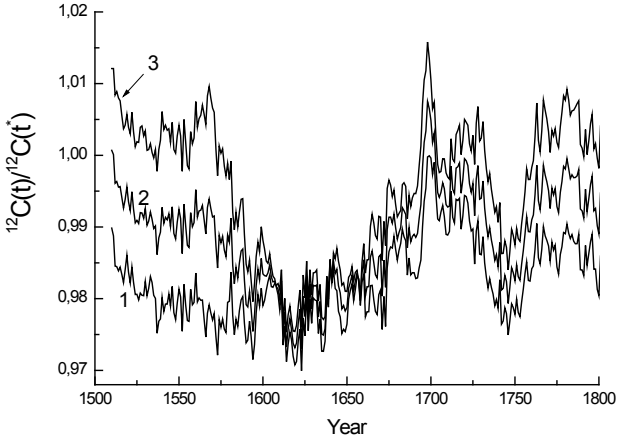

Fig. 3. *Variation of ^{12}C (and CO_2) concentrations (curves 1, 2, 3) calculated using ^{10}Be records by Berggren* et al. *(2009), reconstruction of temperature after Esper* et al. *(2002) and the five-reservoirs model for k = 0, 0.05 and 0.10 consequently.*

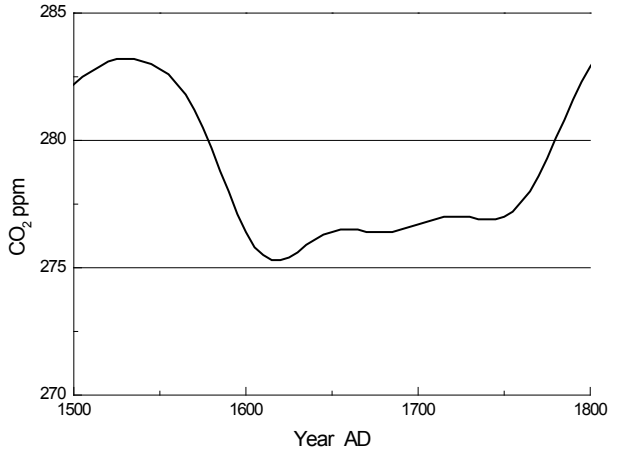

Fig. 4. *Variation of CO_2 concentration in the atmosphere from Etheridge* et al. *(1998). http://cdiac.ornl.gov/ftp/trends/co2/lawdome.smoothed.yr75*

ocean layer and the atmosphere, the calculated values exhibit decrease in the concentration of carbon in the atmosphere during the reference time interval (**Fig. 2**, curves 2 and 3). This result confirms that a decrease of CO_2 concentration in the atmosphere derived from Antarctic ice cores (Etheridge *et al.*, 1998) (see also **Fig. 4**), which takes place through 1550–1650 AD is a result of capture of carbon dioxide from the atmosphere by the upper ocean layer due to decrease of the temperature over this time interval. Increase of the CO_2 concentration in the atmosphere starts since the middle of the 18th century. At the end of the 18th century the concentration reaches values as those of the first part of the 16th century (**Fig. 4**). Curves 2, 3 (**Fig. 3**) demonstrate the same picture. Results of calculation performed with $N_a(t_0) = 7\times10^9$ cm^{-2} are not shown in **Fig. 3** because they entirely coincide with the curve 3 (see, also, curve 3 and 4 in **Fig. 2**).

At the time interval 1650–1750 AD one can see differences between **Fig. 3** and **Fig. 4**. These differences could be connected, for example, with changes in vegetation due to temperature variation that could result in a corresponding change of CO_2 abundance in the atmosphere. A well-documented example from Boreal forests is the variation in tree ring width with changing temperature. Also, the changes in sea-ice cover could influence exchange between ocean and atmosphere. Likely the effect is not a global but has a local character (e.g., Zhilina, 2010). It has been manifested in the North Atlantic as earlier river freezing and an increase of the sea-ice cover of the seas of European section of Arctic. It is unknown how large were variations of the sea-ice cover in a global scale at that time. The answer to this question requires additional investigation.

In addition, if the deep ocean temperature starts to change it would also result in change of concentration of carbon dioxide in the atmosphere. This factor is of particular importance when dealing with the recent global warming. Possible influence of the factors described above will be considered in further works. Besides this the mentioned differences can be a result of influence of the local climatic conditions on ^{10}Be concentration in Greenland ice.

4. CONCLUSION

A possible connection between radiocarbon concentration in the atmosphere and global temperature was noted by Dergachev and Veksler (1991). Peristykh and Damon (1998) reported that the observed increase of Δ^{14}C during the Maunder Minimum (1645–1715 AD) is explained by both, an increase in GCR intensity and cooling of the ocean. These statements were however only qualitative estimations limited by the absence of more or less precise and reliable long-term temperature data. Millennium-scale climatic proxies, obtained over the last 10–15 years, make it possible to test this linkage in more detail.

A comparison of the calculated values of the variations of carbon dioxide concentrations in the atmosphere based on the box model described with the results of measurements, for the period 1500–1800 AD, shows that a) data on Δ^{14}C (Stuiver and Braziunas, 1993) reflect not only variations of the GCR intensity but also changes of temperature and atmospheric CO_2 concentration; b) the decrease of the concentration of CO_2 in the second part of 16th and beginning of 17th can be result of cooling of the ocean surface; c) dependence of the rates of carbon exchange between the different natural reservoirs should be taken into account when studying solar activity and climatic changes in the past.

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