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Variation of atmospheric $^{14}CO_2$ and its spatial distribution

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ABSTRACT

The atmospheric $^{14}CO_2$ is usually presented in the $\Delta^{14}C$ notation, which cannot reflect its absolute quantity change. This article presents the atmospheric radiocarbon activity concentrations (a_{acn}, reported in mBq/m^3) in recent years at nine observation stations. The a_{acn} at Schauinsland decrease from 1977 to 1993 but between 1993 and 2003 keeps at a relative steady state. Atmospheric a_{acn}s were higher in the northern hemisphere than that in the southern hemisphere. The a_{acn}s in the northern hemisphere show clear seasonal cycle with higher value in winter and lower value in summer, while this seasonality is not obvious in the southern hemisphere. Vegetation plays as a role of sink in summer and a role of source in winter, and atmosphere-biosphere radiocarbon exchange might be the main driver of the a_{acn}s seasonality. The annual mean a_{acn}s in both hemispheres show slightly increasing trends since 2002, which may be mainly caused by decreasing air-sea ^{14}C flux as the air-sea ^{14}C gradient decline.

1. Introduction

Radiocarbon is an important cosmogenic radioactive nuclide which is widely used in geological dating and environmental tracing. The content of radiocarbon is usually expressed in the $\Delta^{14}C$ notation (Stuiver and Polach, 1977). In period of preindustrial $\Delta^{14}C$ was in a relatively steady state, regulated by the ^{14}C production. Over the last century, the natural level of $^{14}CO_2$ in the atmosphere has been disturbed by human activities, including the ongoing input of fossil fuel CO_2 into the atmosphere (Suess, 1955), ^{14}C productions mainly from nuclear bomb tests and some contributions from the nuclear energy facilities. Atmospheric nuclear bomb tests primarily in the 1950s and 1960s nearly doubled the atmospheric ^{14}C (Levin et al., 1985; Manning et al., 1990; Nydal and Lövseth, 1983). Atmospheric $\Delta^{14}C$ reached its maximum in mid-1960s and subsequently declined exponentially (Hua and Barbetti, 2004; Levin and Kromer, 2004). Observations suggest that the $\Delta^{14}C$ decline in the 1960s and 1970s was mainly caused by the atmospheric ^{14}C exchange with the ocean and terrestrial biosphere (Broecker and Peng, 1974; Broecker et al., 1985; Nydal et al., 1984). But at present, the atmospheric $\Delta^{14}C$ decline is mainly caused by fossil fuel CO_2 emissions (Levin et al., 2010; Turnbull et al., 2009). However, there is lack uniform understanding on the contribution of surface carbon reservoirs to the atmospheric $\Delta^{14}C$ change (Graven et al., 2012b; Levin et al., 2010), because the $\Delta^{14}C$ can be affected by so many factors, such as fossil fuel CO_2 dilution, stratosphere-troposphere exchange, atmosphere-ocean exchange and so on. What's more, though the use of $\Delta^{14}C$ notation has the advantage in accounting for isotopic fractionations in the carbon exchanging process, it masks the fact how the absolute quantity of atmospheric $^{14}CO_2$ change.

Here the atmospheric radiocarbon specific concentration (10^{-3} becquerel per cubic meter of air, mBq/m^3) (Světlík et al., 2010) was adopted to explore how the atmospheric $^{14}CO_2$ changes in the past decade. The radiocarbon specific concentration was expressed in a_{acn}. The advantage of the a_{acn} is that it's not affected by the fossil fuel CO_2 emissions (because fossil fuel is zero ^{14}C content) and can directly reflect the atmospheric $^{14}CO_2$ molecules quantity change. In this article I present the a_{acn}s variations with time at nine $^{14}CO_2$ observation stations and try to analyze the reasons for these changes.

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2. Methods

2.1. Calculation of a_{acn}

Because the $\Delta^{14}C$ has been isotopic fractionation corrected by $\delta^{13}C$, it should be first converted to its natural fractionation using $\delta^{13}C$. The $\Delta^{14}C$ is transferred to $\delta^{14}C$ by deform the equation given by Stuiver and Polach (1977) to an alternation form,

$$\delta^{14}C = \frac{1}{0.95 + 0.002 \times \delta^{13}C}[\Delta^{14}C + 2(\delta^{13}C + 25)] \quad (1)$$

The $\delta^{14}C$ and $\Delta^{14}C$ are both given relative to radiocarbon standard (95% of the NBS oxalic acid I). Then the a_{acn} was calculated by the equation given by Světlík et al. (2010) as below,

$$a_{acn} = a_{stdd}(1 + 0.001 \times \delta^{14}C)\frac{C_{CO2} \times M_C}{V_{Mair}} \quad (2)$$

Here a_{stdd} is the activity of radiocarbon standard, 0.226 Bq/g, C_{CO2} represents the atmospheric CO_2 concentration (parts per million, M_C and V_{Mair} represent the mole atomic mass of ^{12}C and unit volume of air in standard condition (0 °C, 1.013 × 10^5 pa), equal to 12.01 g/mol and 22.468 × 10^{-3} m^3/mol respectively.

2.2. Data descriptions

To derive the accurate a_{acn}, the following conditions must be satisfied: (1) $^{14}C/^{12}C$, $^{13}C/^{12}C$ and the CO_2 should be measured from the same sample; (2) For time-integrated ^{14}C samples, there should be no isotopic fractionation occur and the CO_2 concentration can be derived from the continual measurement of CO_2 during the ^{14}C sample collecting period at the same site.

Though there are plenty of atmospheric $^{14}CO_2$ monitoring stations around the world in the past decades (Graven et al., 2012c; Hesshaimer et al., 1994; Levin and Kromer, 2004; Levin et al., 2013; Turnbull et al., 2007), most of them collected atmospheric $^{14}CO_2$ samples by time-integrated method, that is absorbing atmospheric CO_2 by basic solutions (e.g. NaOH solution), using a pump or not, called bubble method (Levin et al., 1980) or static absorption method (Currie et al., 2011), respectively. For both

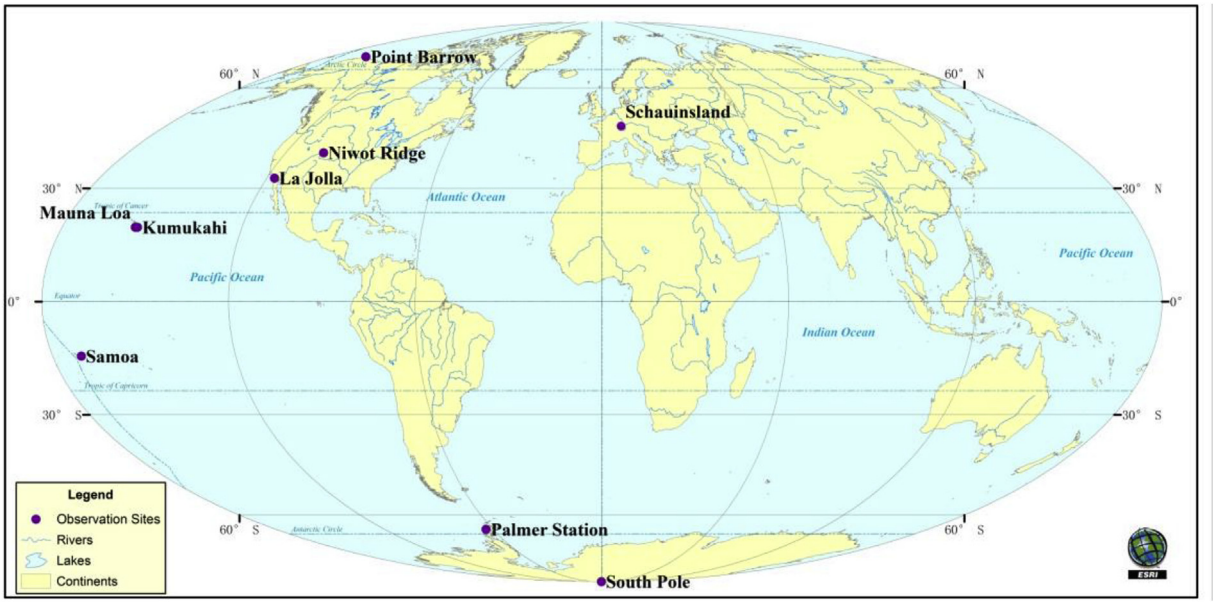

Fig. 1. Observation stations of data used in this article.

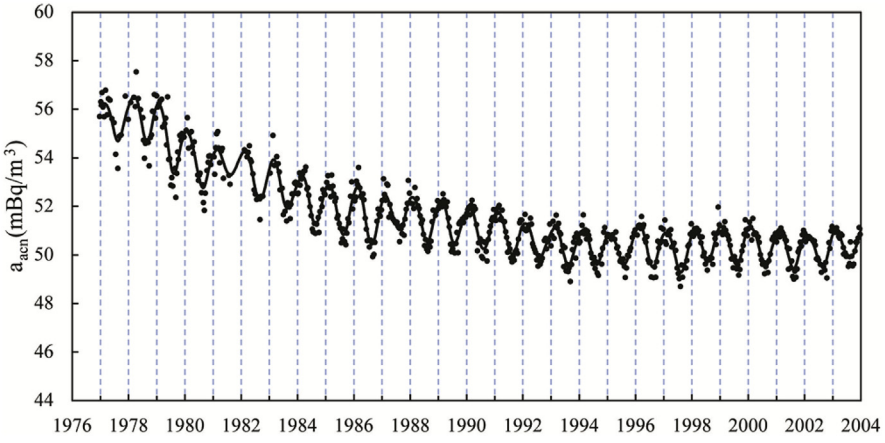

Fig. 2. Atmospheric a_{acn} at Schauinsland.