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Estimation of $^{14}CO_2$ amount in the atmosphere

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Abstract Carbon from fossil CO_2 emissions, without a significant presence of ^{14}C, causes dilution of ^{14}C in the carbon isotopic mixture (Suess effect). Reported ^{14}C activities are usually connected to radiocarbon amount in the carbon isotopic mixture. Our paper is aimed on estimation of $^{14}C/^{14}CO_2$ amount in the atmosphere (and its trend), utilizing calculation of a ^{14}C activity concentration. A parameter connected only with a ^{14}C quantity in the volume or mass unit of air is not influenced by a fossil carbon amount. Such a "robust" parameter can be influenced only by processes connected with ^{14}C emissions/depositions.

Keywords $^{14}CO_2$ · Global Suess effect · Anthropogenic CO_2 · Delay of atmospheric CO_2

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Introduction

Radiocarbon is a radionuclide with a half-life of 5,730 years, global occurrence, and partly anthropogenic origin. In nature, radiocarbon is produced by nuclear reactions, generated by cosmic rays in the atmosphere. Radiocarbon production rate is equilibrated with ^{14}C removal into other environmental compartments (biota, oceanic water, and sediments) and the role of direct radioactive decay in the atmosphere is only negligible. In the last century, nuclear weapon tests were important sources of anthropogenic ^{14}C. Consequently, activity of atmospheric ^{14}C was found twice higher on the north hemisphere during 1963 and 1964 than activity level given by natural production [1]. During the following years, the ^{14}C activity decreased exponentially. In the 1980s the decrease has decelerated and since the beginning of 1990s it can be represented by a linear relation, see Fig. 1.

On the planetary scale, the total radiocarbon inventory is predominantly given by natural production [2]. Both the atmosphere and biosphere contain only a small part of the total environmental ^{14}C inventory. Radiocarbon is transferred relatively promptly from the atmosphere and biota into other environmental compartments (long-term carbon sinks, namely oceans). The carbon isotopic mixture of fossil fuels (oil, coal) does not contain a significant quantity of radiocarbon. Fossil carbon (CO_2), entering the atmosphere from the process of fuel combustion, dilutes a radiocarbon amount in the carbon isotopic mixture and decreases the resulting radiocarbon activity (activity of ^{14}C per gram of carbon isotopic mixture according to Stuiver–Polach convention) [3]. This is called the Suess effect [4]. As seen in Fig. 2, the anthropogenic emissions of fossil carbon (fossil CO_2) into the atmosphere increases annually [5]. A similar trend is visible in the case of atmospheric CO_2

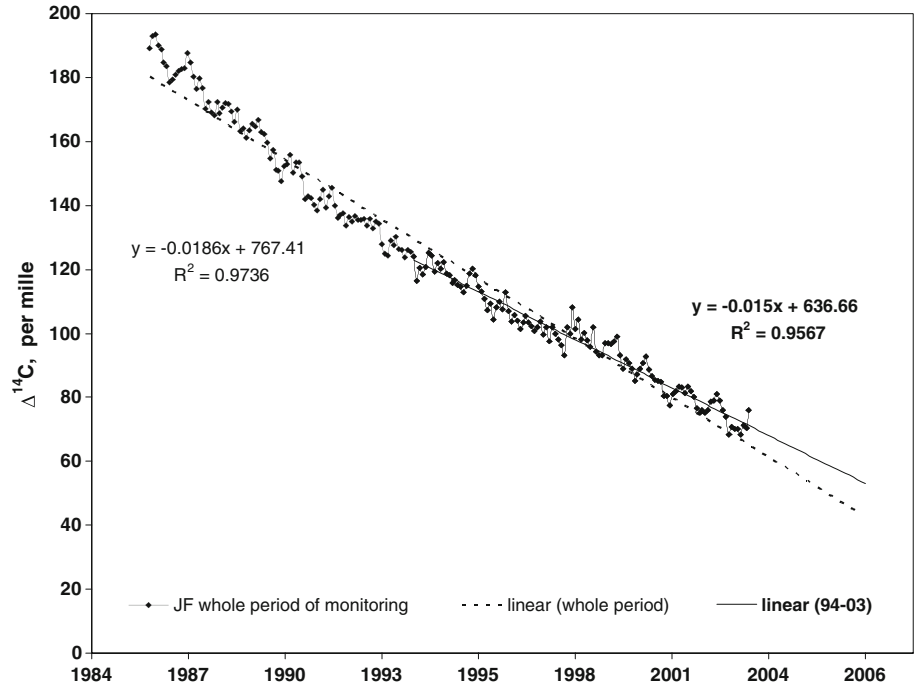

Fig. 1 Time behavior of atmospheric $^{14}CO_2$ activity, since June 1986. Reported data are from high-mountain monitoring station Jungfraujoch (JF) in Switzerland, 3,450 m above sea level [7]

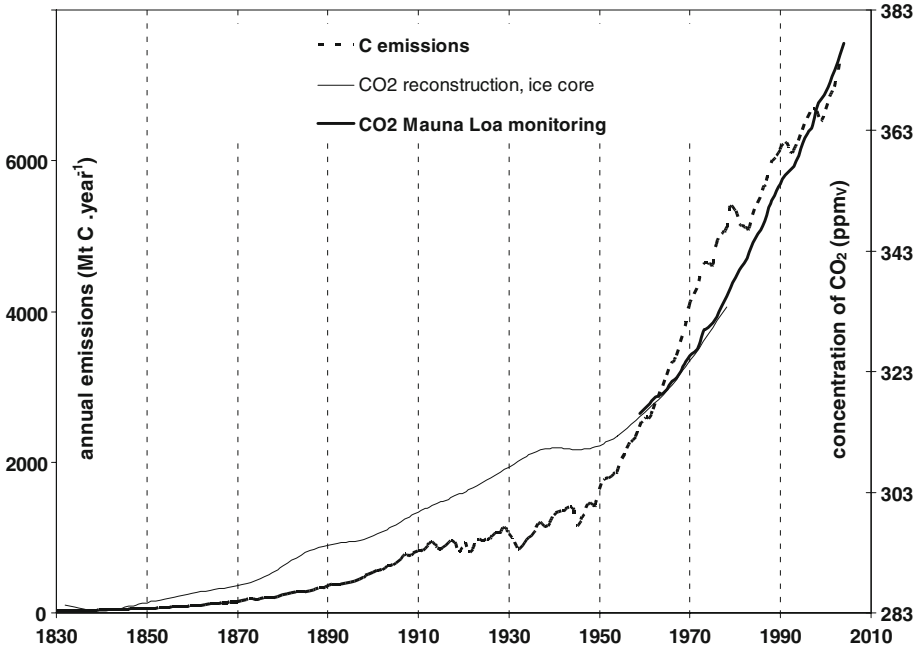

Fig. 2 Annual fossil carbon emissions (*left vertical axis*, in Mt of fossil carbon per year) and its comparison with concentration of atmospheric CO_2 (*right axis*, in ppm). $^{14}CO_2$ is prevailing chemical form of radiocarbon in the atmosphere. The curve of CO_2 concentration (*fine line*) is assembled from the from Antarctic ice core data (Law Dome, No. DE08). Monitoring of atmospheric CO_2 concentration is performed at station Mauna Loa on Hawaiian Islands, USA since the beginning of 1950s (*bold line*) [5]

concentrations. Increasing concentration of this greenhouse gas is probably caused by anthropogenic releases [6].

Experimental

To compare activities of atmospheric $^{14}CO_2$, data from several European monitoring stations were utilized. These stations were Jungfraujoch, Switzerland (until July 2003, a high mountain station, probably with a minimal influence of local and regional Suess effect in the Europe); Schauinsland, Germany (until December 2003, a station located in area with a relative small Suess effect influence); B-24 and A-6 monitoring localities in Hungary, close to the cities of Dunaföldvár and Paks, respectively (the average of observed values); Košetice, Czech Republic (since 2004, monitoring station in the Czech-Moravian highlands, located in the area with relatively weak local sources of

CO_2 from fossil fuel combustion); Prague-Bulovka, Czech Republic (since 2001, monitoring locality situated at the border part of Prague, where relatively enhanced local load from fossil fuel combustion can be expected) [7–9]. Since August 2001, the $^{14}CO_2$ activity concentration is measured in the locality Prague-Bulovka. This parameter is calculated from the observed values of atmospheric CO_2 concentration and $^{14}CO_2$ activities [9]. Analytical routines for determination of ^{14}C activity in Czech and Hungarian laboratories were described elsewhere [8, 9].

Results and discussion

The observed time behavior of $^{14}CO_2$ activities shows seasonal variations, see Fig. 3. A relatively slight activity reduction in winter period is typical for the high mountain stations at Jungfraujoch and Schauinsland, related to the minimal local and regional influence from fossil CO_2 emissions. At the Košetice station and in the Hungarian regions no major local sources of fossil CO_2 occur, so that the observed influence may particularly be given by regional sources. The most distinct winter minima are visible at the monitoring locality Prague-Bulovka, as was expected, although spring/summer values observed at this locality approximates activities from the stations Jungfraujoch and Schauinsland, which are almost free from Suess effect.

On the local scale, the $^{14}CO_2$ activity concentration can be influenced only by ^{14}C releases, such as effluents from nuclear energy facilities or $^{14}CO_2$ emissions from saprophytic processes of organic substance decomposition in soils (such carbon based substances also contain ^{14}C). On the other hand, the $^{14}CO_2$ activity concentration can also be affected by $^{14}CO_2$ intake by photosynthesis in biota.

No significant trends of the atmospheric $^{14}CO_2$ activity concentrations were observed during the monitoring period in the locality Prague-Bulovka. In other words, based on data obtained for the period (2002–2007), no significant long-term change of the $^{14}CO_2$ amount in the atmosphere was found. Supposing that the number of ^{14}C atoms or $^{14}CO_2$ molecules in the atmosphere did not change, a possible explanation of the systematic interannual decrease of $^{14}CO_2$ activity (Fig. 1) seems to be related to the global Suess-effect from the interannual increase of emissions of fossil CO_2. Unfortunately, the values of atmospheric $^{14}CO_2$ activity concentrations observed in locality Prague-Bulovka are charged by relatively larger uncertainties due to the method used for CO_2 concentration determination (volumetry).

To validate our results, the existing data from the German station in Schauinsland were employed. Time behavior of activity concentration of atmospheric $^{14}CO_2$ was calculated from concentration of CO_2 and $^{14}CO_2$ activity for the period from January 1977 to December 2003 [5, 7, 10]. The activity concentration curve (Fig. 4) shows seasonal variations. As seen from comparisons with the CO_2 concentration curve, CO_2 concentration is the dominant parameter controlling seasonal changes of $^{14}CO_2$ activity concentration.

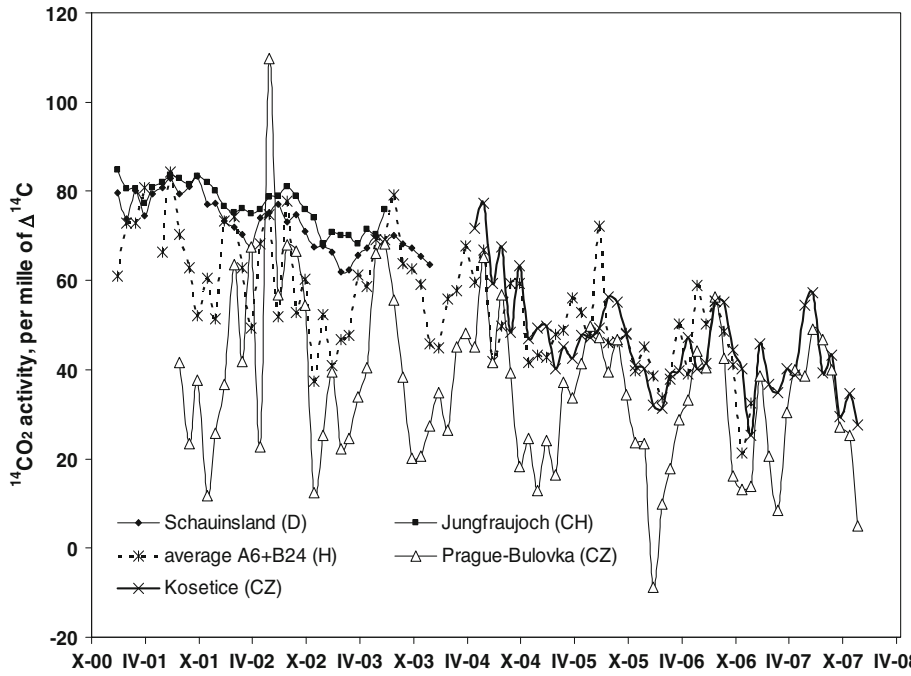

Fig. 3 Comparison of seasonal changes of atmospheric $^{14}CO_2$ activities in several European monitoring localities [7]. Activities are reported in per mile of $\Delta^{14}C$ (i.e., related to ^{14}C amount in the carbon isotopic mixture) [3]

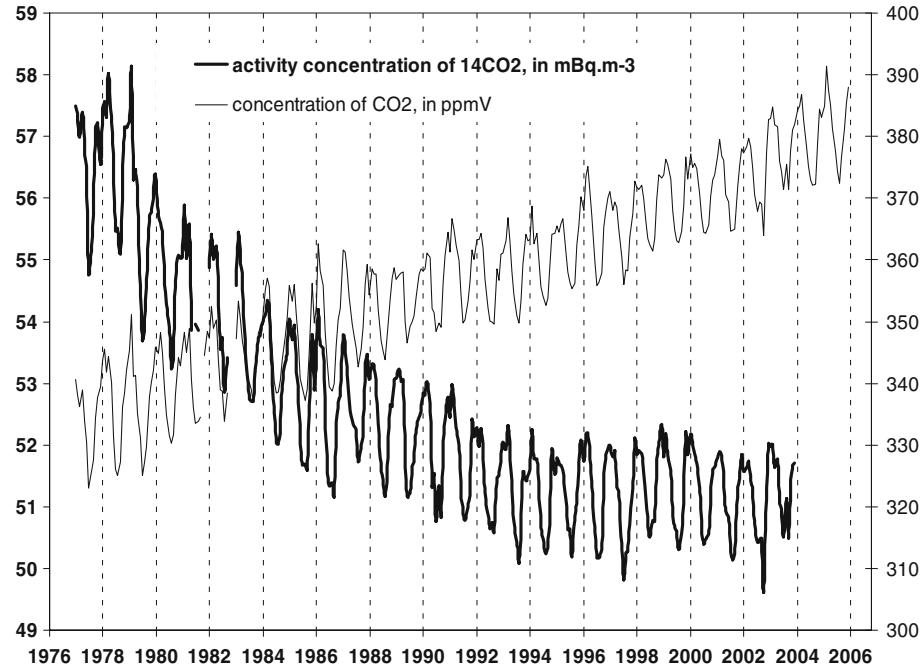

Fig. 4 Activity concentration of the atmospheric $^{14}CO_2$ (in mBq m^{-3}, *bold line*), calculated from the published data from monitoring station Schauinsland, Germany. Activity concentration was calculated from the activity of the atmospheric $^{14}CO_2$ and concentration of CO_2 [5, 7, 10]

The mean annual activity concentrations of atmospheric $^{14}CO_2$, calculated for the data from the localities Schauinsland (Germany) and Prague-Bulovka (Czech Republic) do not show any systematic trend after 1994. Assuming that the atmospheric air volume remains stable, the results obtained prove that the ^{14}C content in the atmosphere (the number of ^{14}C atoms or $^{14}CO_2$ molecules) does not show any significant systematic change since 1994.

The mean activity concentration, calculated from the data published from the Schauinsland station amounts to 51.2 ± 0.7 mBq m^{-3} (1994–2003 period) and for the locality Prague-Bulovka (period 2002–2007) equals to 53.4 ± 1.6 mBq m^{-3}. The observed difference of the mean values of both regions (with different monitoring periods) is close to the level of statistical significance. This difference may be partially due by the systematic error of calibration and volumetric determination of CO_2 concentrations in Prague-Bulovka. Another reason for this difference may be the lack of additional data (air temperature, humidity, atmospheric pressure, etc.) in the calculation of activity concentration for the Schauinsland station.

An amount of radiocarbon in the atmosphere depends on its production rate in the atmosphere (cosmogenic production, current releases from nuclear energy facilities, saprophytic processes) and its transfer into other compartments of the environment (biota, oceans, sediments). The high ^{14}C quantities, appearing in the air as a result of nuclear bomb testing in the past has probably reached a balanced distribution in the atmosphere and associated compartments of the environment. Supposing a stable quantity of ^{14}C in the atmosphere (i.e. stable activity concentration), a linear decrease of ^{14}C activity since the beginning of 1990s (see Fig. 1) can be explained by an increase of global Suess effect.

Conclusions

Values of ^{14}C activity concentrations were calculated utilizing the data from the Prague-Bulovka locality and other monitoring sites and new information has been obtained on radiocarbon quantity in the atmosphere. Although the annual decrease of atmospheric $^{14}CO_2$ activity is relatively uniform during the last decade, the mean amount of ^{14}C ($^{14}CO_2$) in the atmosphere seems to be stable since 1994 (if we suppose stable amount of air in the atmosphere). The decrease of activity of atmospheric $^{14}CO_2$ seems to be caused by the increase of global Suess effect originating from boosting of fossil fuel combustion. Stable amount of ^{14}C ($^{14}CO_2$) in the atmosphere, as a tracer of atmospheric CO_2, indicates a stable delay time of CO_2 in the atmosphere (without any cumulation effect), which seem to be independent of the actual CO_2 concentration.

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References

1. Meijer, H.A.J., van der Plicht, J., Gislefoss, J.S., Nydal, R.: Comparing long-term atmospheric ^{14}C and ^{3}H records near

Groningen, the Netherlands with Fruholmen, Norway and Izaca, Canary Islands^{14}C stations. Radiocarbon **37**, 39–50 (1995)
2. UNSCEAR: Report of the United Nations scientific committee on the effects of atomic radiation to the general assemble: Report 1; (2000)
3. Stuiver, M., Polach, H.A.: Reporting of ^{14}C data. Radiocarbon **19**, 355–363 (1977)
4. Suess, H.E.: Radiocarbon concentration in modern wood. Science **122**, 415–417 (1955)
5. CDIAC: http://cdiac.ornl.gov/trends/trends.htm. Accessed 8 Jan 2009
6. IPCC: Climate change 2007—the physical science basis. Contribution of working group I to the fourth assessment. Report of the IPCC; (2007)
7. Levin, I., Kromer, B.: The tropospheric $^{14}CO_2$ level in mid-latitudes of the northern hemisphere (1959–2003). Radiocarbon **46**, 1261–1272 (2004)
8. Molnar, M., Bujtas, T., Svingor, E., Futo, I., Svetlik, I.: Monitoring of atmospheric excess ^{14}C around Paks Nuclear Power Plant, Hungary. Radiocarbon **49**, 1031–1043 (2007)
9. Svetlik, I., Molnar, M., Svingor, E., Futo, I., Pinter, T., Rulik, P., Michalek, V.: Monitoring of atmospheric $^{14}CO_2$ in central European countries. Czechoslov. J. Phys. **56D**, 291–297 (2006)
10. WDCGG: http://gaw.kishou.go.jp. Accessed 8 Jan 2009