Forty years of atmospheric radiocarbon variation studies in Slovakia

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ABSTRACT: Radiocarbon variations in the atmosphere have been studied for almost 40 years in two localities of Slovakia, in Bratislava and Zlkovce. Zlkovce is situated about 60 km NE from Bratislava in a flat agricultural area, only a few kilometers from the Jaslovske Bohunice Nuclear Power Station. The accomplished analysis of ^{14}C variations in the atmosphere has proved the existence of annual variations with attenuating amplitude and a decreasing mean value. The observed ^{14}C variations have typical maxima in summer and minima in winter. Winter minima are affected by the heat supply in winter season which is connected directly with the fossil CO_2 emissions and more intensive Suess effect. Summer maxima are affected by a combination of a lower CO_2 emission rate and a higher turbulent transport of the stratospheric ^{14}C to the troposphere. The annual radiocarbon variations were studied using the Fourier harmonic analysis. The obtained results show that the radiocarbon variations have a high degree of symmetry. Furthermore, the obtained basic frequency proves that the cyclic processes with the period of 12 month have a major influence on the ^{14}C amount in the troposphere. The presence of higher-order harmonics is also visible, but a physical interpretation is not clear. The long-time average of the ^{14}C record in Zlkovce during the last 30 years is more variable than that in Bratislava, as it has been clearly affected by operation of the Jaslovske Bohunice Nuclear Power Station.

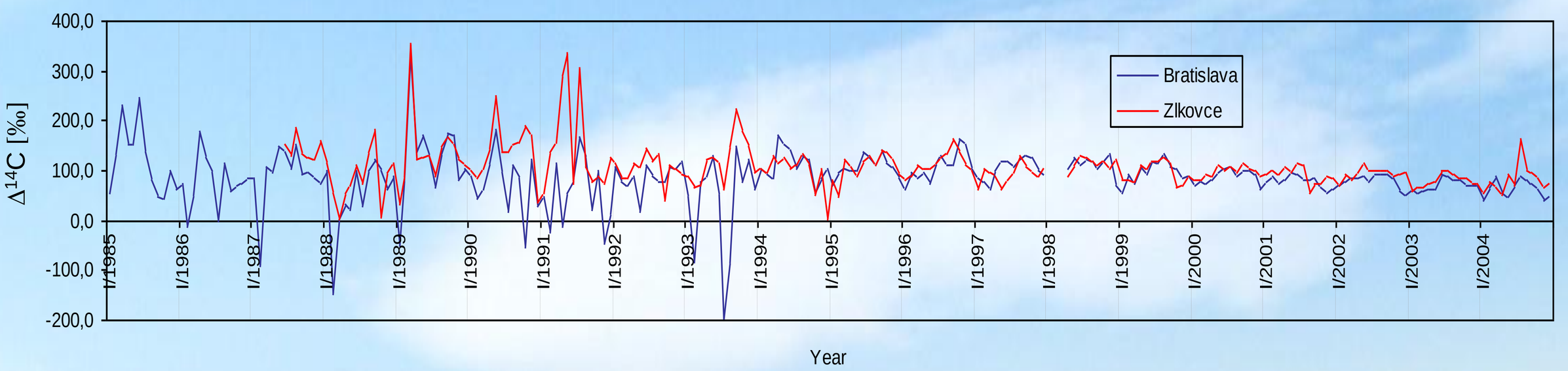

Fig. 1. *Comparison of $\Delta^{14}C$ data series and calculated values of $\Delta^{14}C$ in Bratislava and Zlkovce air.*

^{14}C levels in Bratislava atmospheric CO_2 have been measured since 1966 [1,2], and at Zlkovce since 1987, Fig. 1. Both data sets show the obvious variations with the attenuating amplitude and with the decreasing mean value. The decrease of the mean value is caused by an exchange of ^{14}C between the atmosphere and surrounding carbon reservoirs. Seasonal variations with different periods superposed on the long-term trend, and accidental deviations are seen too. The variations of the ^{14}C activity depend on the meteorological and climatic conditions and on the periodical changes of the carbon reservoirs transition characteristics. The data since 1995 have a typical periodical character and the accomplished analyses show that the annual means and variation amplitudes have been decreasing exponentially. In average, the annual maximum has been shifted to the late July and minimum to the late January.

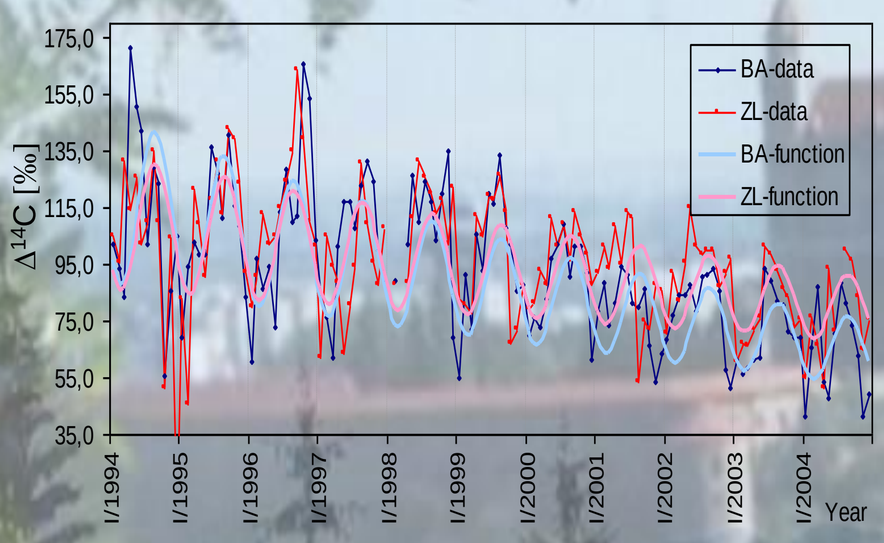

***Fig. 2.** Comparison of $\Delta^{14}C$ data series and calculated values of $\Delta^{14}C$ according to relation (1) in Bratislava and Zlkovce air.*

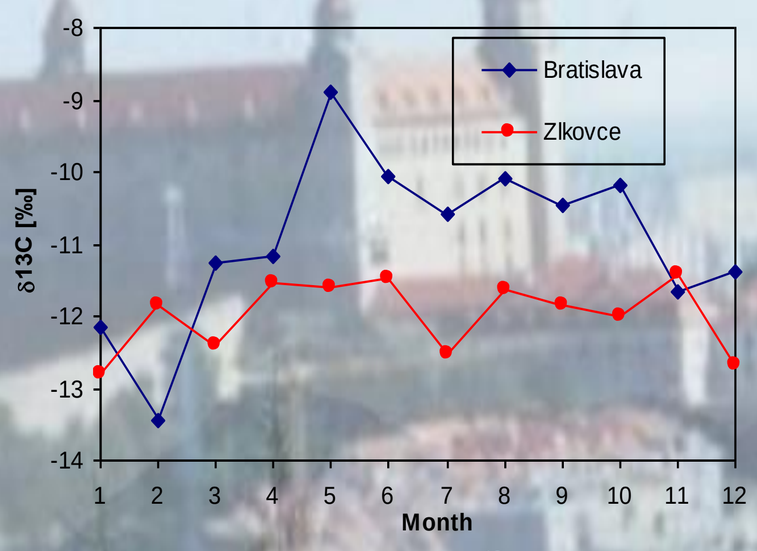

***Fig. 3.** Comparison of $\delta^{13}C$ courses at Bratislava and Zlkovce.*

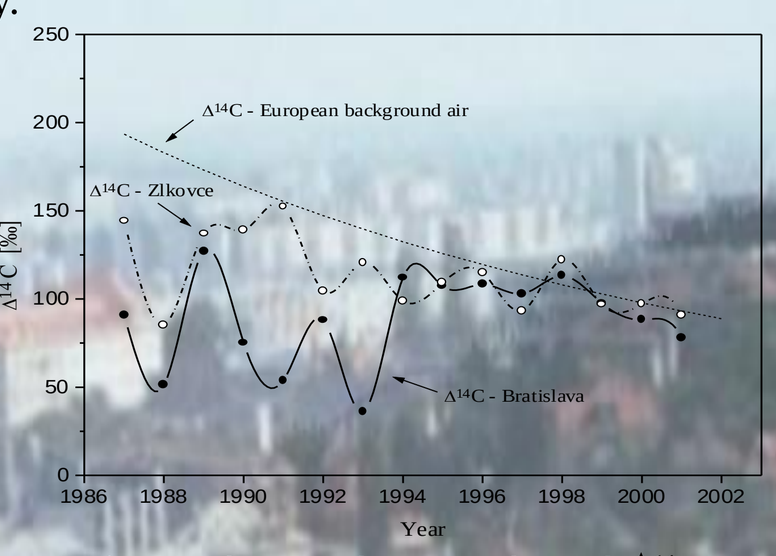

Fig.4. *The annual mean values of $\Delta^{14}C$ in atmospheric CO_2 in Bratislava and Zlkovce. The dashed line represents the long-term of the annual mean $\Delta^{14}C$ in the background air over Europe.*

As a consequence of the mentioned facts we used the following function:

$$y = A\,e^{-at} + B\,e^{-bt}\,\cos(\omega_1 t + \langle\varphi\rangle) \qquad (1)$$

to describe the $\Delta^{14}C$ data courses, where $\omega_1 = 2\pi/T_1$ is the basic frequency, $T_1 = 12$ [month] is the period and $\langle\varphi\rangle$ is the phase shift of the variation. By means of the non-linear regression method, the coefficients A, a, B, b and $\langle\varphi\rangle$ could be obtained.

The courses of $\delta^{13}C$ and $\Delta^{14}C$ levels measured in Bratislava and at Zlkovce are shown in Fig. 3 and 4, respectively. Since 1987 to 1993 the $\Delta^{14}C$ values for both stations are significantly below background air values. However, in the last years for both sites the measured $\Delta^{14}C$ levels are closer to the European background $\Delta^{14}C$ trend line. The relative high increases of $\Delta^{14}C$ in spring and early summer months were caused rather by the depletion of the atmospheric ^{14}C in winter months than by the injection of the stratospheric air into the troposphere (Fig. 3). According to the expectation, the Suess minima of $\Delta^{14}C$ were not as distinct at Zlkovce as they were in Bratislava, although they were also identified. But in some months high $\Delta^{14}C$ values (~ 300 ‰) were measured (in March 1989 in both localities and in April, May and July 1991 only at Zlkovce). The $\Delta^{14}C$ excesses could be of the technogenic origin. Since 1994 there were no longer such marked differences in annual mean $\Delta^{14}C$ values between the two stations. This could be explained by decreasing of fossil fuel CO_2 emissions in Slovakia after 1994. The average annual courses of $\Delta^{14}C$ at both localities for the period 1995 – 2001 are presented in Fig. 5. The courses are similar, reaching maxima in summer months (August) and minima in winter (from December to February), with seasonal variations from 76 to 113 ‰ for Bratislava, and from 85 to 118 ‰ for Zlkovce.

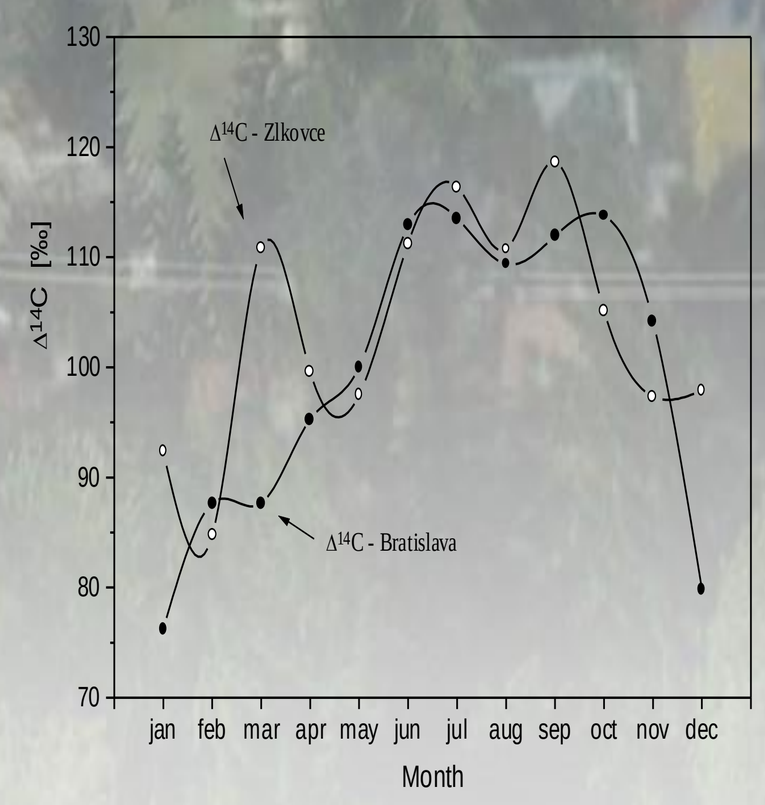

Fig. 5. *The average annual courses of the $\Delta^{14}C$ in the atmospheric CO_2 in Bratislava and Zlkovce.*

Conclusions

A high variability of the annual mean $\Delta^{14}C$ levels in atmospheric CO_2 was observed at two not very distant stations until 1993. In this period the annual mean values of the $\Delta^{14}C$ in heavily polluted atmosphere of Bratislava were about 50 ‰ lower and at Zlkovce about 20 ‰ lower compared to $\Delta^{14}C$ in European background air. After 1993 the annual mean $\Delta^{14}C$ was in both sites close each other and in 2001 reached the value approximately 8 ‰ above the $\Delta^{14}C$ natural level. The amplitudes of the annual courses are decreasing with time and in 2001 they reached the value of 14 ‰. The observed $\Delta^{14}C$ behavior in the atmosphere provides an unique evidence of the decrease of fossil fuel CO_2 emissions into the atmosphere.

References

[1] POVINEC, P. et al. The rapid method of carbon-14 counting in atmospheric carbon dioxide. Int. J. Appl. Rad. Isotopes 19 (1968) 877-881.

[2] POVINEC, P. et al. Seasonal variations of anthropogenic radiocarbon in the atmosphere. Nucl. Instr. and Methods 317 (1986) 556-559.