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Assessing diurnal changes of carbon budget in the urban atmosphere: combined use of CO_2, $^{13}CO_2/^{12}CO_2$, $^{12}C^{18}O^{16}O/^{12}C^{16}O_2$ and $^{14}CO_2/^{12}CO_2$ mixing ratios

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Emissions of carbon dioxide related to burning of fossil fuels constitute an important component of the carbon budget, both on global and regional scales. For heavily industrialized and populated areas such as western and central Europe, a large proportion of the total CO_2 flux entering the atmosphere is attributed to this source. Global and regional models of carbon cycle rely mainly on emission statistics to quantify the magnitude and variability of the fossil CO_2 flux into the atmosphere.

Krakow (50^o04'N, 19^o55'E, 220 m a.s.l.) is a large urban agglomeration located in the southern Poland, with about 1 million inhabitants, rapidly growing car traffic and significant industrial activities. Consumption of coal, gas and oil for communal and transport purposes generates major fluxes of anthropogenic carbon dioxide and carbon monoxide within the region. In addition, due to prevailing westerly air circulation, the Krakow region is under substantial influence of a large coal mining and industrial centre (Upper Silesia) located approximately 60 km west of the city.

The $^{14}CO_2/^{12}CO_2$ ratios measured in bi-weekly composite samples collected in Krakow since 1983 testify major changes in economy of the region which have occurred over the past two decades. The ^{14}C signature of atmospheric CO_2 reflects significant changes of anthropogenic CO_2 fluxes released into the local atmosphere. The contribution of fossil-fuel derived CO_2 in the total CO_2 load of the lower atmosphere in Krakow decreased from approximately 21 ppm in 1989 to around 10-12 ppm in the last few years. This change is linked with major reduction in coal consumption in Poland, from ca. 160 Mt in 1985 to 84 Mt in 2004.

The regional models of carbon cycle run with high temporal resolution require detailed information on structure and variability of CO_2 fluxes entering the atmosphere. Bottom-up approaches to quantify anthropogenic fluxes of CO_2 based on emission statistics require independent verification. Combined, high-resolution measurements of CO_2 concentration and its isotopic composition ($\Delta^{14}C$, $\delta^{13}C$, $\delta^{18}O$) may serve as an independent tool to assess variability of CO_2 fluxes (anthropogenic and biogenic) on local scale. The methodology adopted here is based on mass and isotope balance of local CO_2 load.

Several short-term pollution events which have occurred in the urban atmosphere of Krakow during 2007 have been analysed. The following measurements were performed during each pollution event: (i) quasi-continuous measurements of $CO_2$and CO mixing ratios, supplemented by continuous measurements of PAH (polycyclic aromatic hydrocarbons), (ii) CO_2 and CO concentration as well as $\Delta^{14}C$, $\delta^{13}C$ and $\delta^{18}O$ of CO_2 in several samples of air collected in chosen intervals during the given event, (iii) meteorological parameters (air temperature, pressure, wind speed and wind direction). In addition, backward-trajectory modelling was used to identify the origin of air masses passing the sampling point during the event. With the mass and isotope balance of CO_2 solved for each sampling interval it was possible to assess short-term changes of anthropogenic and biogenic CO_2 flux during the analysed pollution events. Significant diurnal changes of both fluxes were observed.

In addition, it was possible to assess diurnal changes of the $\Delta CO/\Delta CO_2$(fossil) ratio. This ratio depends on the quality of the combustion process. During the analysed pollution events the $\Delta CO/\Delta CO_2$(fossil) ratio varied between ca. 20 and 70 ppb CO per ppm of fossil CO_2. These are significantly higher ratios than those reported for major industrial sources in the Krakow region for the period 1998-2005 (between ca. 7 and 11 ppb CO per ppm of fossil CO_2). However, those estimates are based on emission statistics of major industrial enterprises and do not comprise emissions related to car traffic and domestic heating systems (low emission) which is a dominating source of fossil fuel CO_2 in urban environment of Krakow and its suburbs.

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