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Development of radiocarbon-based methods to investigate atmospheric fossil carbon pollution

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Gaseous and solid state carbon containing compounds significantly affect global climate change based on current atmospheric research results. Major part of the anthropogenic changes of the atmospheric carbon dioxide can be attributed to the combustion of fossil fuels and 95% of their emission is realised in the industrially active areas of the northern hemisphere. Anthropogenic carbonaceous aerosol particles play also a key role in the atmosphere modifying indirectly climate change and the quality of the environment and affecting directly human health.
Since September 2008, the CO_2 concentration of the air and its specific radiocarbon content (14C) has been monitored in the city of Debrecen (Hungary) and in a rural background site, Hegyhátsál (Hungary). To obtain a more representative view regarding anthropogenic contribution of the atmospheric carbon species, our measurement programme was enhanced by including the investigation of atmospheric aerosols in 2010. An aerosol cascade sampler for continuous monitoring was installed close to the atmospheric CO_2 sampling station in the inner city of Debrecen. For 14C measurements, special sample preparation system and method was developed for the tiny total carbon content of the aerosol samples collected synchronously with the carbon dioxide observations. The radiocarbon measurement of the aerosol samples was performed by a high-sensitivity accelerator mass spectrometer (AMS) dedicated to environmental samples (EnvironMICADAS) developed together with ETH Zürich. The δ13C values of the samples were measured by the Dual Inlet system of a Delta PLUS XP Isotope Ratio Mass Spectrometer from the tiny CO_2 amount aimed to reserve.
The atmospheric fossil CO_2 and fossil PM2.5 concentration variations show high similarity in the air of Debrecen city. During the winter heating period, due to the meteorological conditions (frequent thermal inversion, decreasing rate of mixing and upwelling), significantly higher total PM2.5 concentrations can be observed close to the surface, which is also reflected in the quantity of the total carbon. The trend of the fossil CO_2 excess is very similar to the fossil carbon content of the total PM2.5. On the other hand during the winter time heating periods the extreme high observed aerosol concentrations in the city air seems to be caused by biomass combustion. It is well-visible from the aerosol C-14 measurements that the relative 14C content of the aerosol increases in the heating period in the PM2.5, even compared to the summer period. The trend of the measured stable isotope ratio also shows high similarity with the fossil carbon content of the PM2.5.
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