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Isotope source apportionment of carbonaceous aerosol as a function of particle size and thermal refractiveness

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The stable carbon isotopes can be used to get information about sources and processing of carbonaceous aerosol. We will present results from source apportionment of carbonaceous aerosol as a function of particle size thermal refractiveness. Separate source apportionment for particles smaller than 200 nm and for different carbon volatility classes are rarely reported and give new insights into aerosol sources in the urban environment.

Stable carbon isotope ratios were measured for the organic carbon (OC) fraction and total carbon (TC) of MOUDI impactor samples that were collected on a coastal site (Lithuania) during the winter 2012 and in the city of Vilnius (Lithuania) during the winter of 2009. The 11 impactor stages spanned a size range from 0.056 to 18 μ m, but only the 6 stages in the submicron range were analysed.

The δ^{13} C values of bulk total carbon (δ^{13} C $_{TC}$) were determined with an elemental analyser (Flash EA 1112) coupled with an isotope ratio mass spectrometer (Thermo Finnigan Delta Plus Advantage) (EA – IRMS). Meanwhile δ^{13} C $_{OC}$ was measured using thermal–desorption isotope ratio mass spectrometry (IRMS) system. This allows a rough separation of the more volatile OC fraction (desorbed in the oven of IRMS up to 250 0 C) from the more refractory fraction (desorbed up to 400 0 C).

In this study we investigated the composition of organic aerosol desorbed from filter samples at different temperatures using the thermal–desorption proton–transfer–reaction mass spectrometry (TD–PTR–MS) technique.

During winter-time in Lithuania we expect photochemistry and biogenic emissions to be of minor importance. The main sources of aerosol carbon should be fossil fuel and biomass combustion. In both sites, the coastal and the urban site, δ^{13} C measurements give a clear indication that the source contributions differ for small and large particles. Small particles < 200 nm are depleted in 13 C with respect to larger particles by 1-2 %. This shows that OC in small particle arises mainly from fossil fuel sources, whereas OC in larger particles from 200 nm to 1 μ m has higher contribution from biomass burning/other sources.

Moreover, there is a clear distinction in source contribution between the more volatile OC fraction and the more refractory fraction. The more refractory fraction is enriched in 13 C by 1 to 2 % for both small and large particles. These results show that the fossil fuel combustion is associated to a larger degree with more volatile carbon, whereas biomass burning is the main source of the more refractory particles.

According to our source apportionment, the more volatile carbon fraction in the smallest particles is almost completely from fossil fuels, whereas the more refractory carbon fraction in the large size range is almost complete from biomass burning. The more refractory small particles and the less refractory large particles are roughly an even mix of these two sources.

The detailed chemical speciation of the carbonaceous aerosol will be presented as well.

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