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Bulk carbon isotopic ratio of size-resolved aerosol at background and urban sites in Lithuania

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Stable carbon isotopes have been successfully used in source apportionment studies of atmospheric aerosols where two or more sources have distinct isotopic composition. It has been shown that areal and temporal isotopic variabilities of aerosol samples are related to the diversity of the sources (Cahier, 1989), but there is a lack of studies combining the MOUDI impactor and carbon isotopic ratio measurements (Norman et al. 1999).

The samples collections were performed at the Air pollution research station in Preila during 17 - 22 September, 2008 and in Vilnius during 02 – 08 December 2008. The Preila station (55˚ 55′N and 21˚00′E) is located in western Lithuania on the coast of the Baltic Sea, on the Curonian Spit. Sampling in Vilnius (54˚ 39′N and 25˚ 14′E) was performed on the roof of the Institute of Physics, which is located in a forested area in the southwestern part of Vilnius.

The samples were collected with a Micro-orifice Uniform Deposit Impactor (MOUDI) model 110. The flow rate was 30 l/min, and the 50% aerodynamic cutoff diameters of the 10 stages were 18, 10, 5.6, 3.2, 1.8, 1.0, 0.56, 0.32, 0.18, 0.10 and 0.056 μm. Aluminum foil was used as the impaction surface. Total carbon isotopic ratios ($\delta^{13}C/^{12}C$) were measured at each stage using the elemental analyzer (*FlashEA 1112*) connected to the stable isotope ratio mass spectrometer (*ThermoFinnigan Delta Plus Advantage*). Simultaneously the carbon amount at each stage was measured using the elemental analyzer with a thermal conductivity detector (Garbaras et al. 2008). The $1/6^{th}$ part of the foil was placed into the tin capsule and combusted in the oxidation furnace at the temperature of 1020º C and with the oxygen excess. Carbon of the sample oxidized into CO_2 gas, which passed to the mass spectrometer ionization cell through the gas distribution device *ConFlow III*.

The MOUDI cascades were analyzed for total carbon by the elemental analyzer (Flash EA1112). At the Preila station the total carbon mass concentration was highest in the range of 0.32 – 0.56 μm and 3.2 – 5.6 μm (0.79 μg/m^3 and 0.32 μg/m^3). In Vilnius the total carbon mass concentration was highest in the range of 0.56 – 1 μm (51.48 μg/m^3). The total carbon mass in Vilnius samples was about 28 times higher than in samples in cascades 1.8 – 5.6 μm, and about 74 times higher in cascades 0.056 – 1 μm and $>$10 μm. This demonstrates that coarse mode in Preila is more pronounced than in Vilnius. The absence of apparent coarse mode in Vilnius is typical of a rural site (Chow et al., 2008), probably with one dominant aerosol source.

The carbon isotopic ratio variation between accumulation and coarse modes in Vilnius was from -26.5 ‰ to -23.8 ‰The total carbon isotopic value ($\delta\ ^{13}C/^{12}C \sim$ -20.5 ‰) of coarse mode at Preila site implies that carbonaceous particles in this mode were of marine origin emitted from the Baltic Sea, while accumulation mode aerosols had some other source ($\delta\ ^{13}C/^{12}C \sim$ -29 ‰). Air mass transport during the Preila experiment was from the continent (Latvia, Estonia). In such transport scenario the main source of carbonaceous aerosols in accumulation mode could be vegetation and transport emissions (Kelly et al., 2005; Huang et al., 2006). During the experiment in Vilnius air mass transport was from Belarus, Poland and Ukraine. In these areas coal burning is used for domestic heating and industry. Relatively small differences in the carbon isotopic ratio in accumulation and coarse modes suggest that in wintertime aerosol sources of similar origin are dominant. It can be coal burning and transport.

The precision of isotopic measurements was better than 0.3 ‰ except a few cases at the first and last stage due a small amount of carbon.

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