Chemical and stable carbon isotope composition, sources and chemical processing of fine aerosol at urban, coastal and forest background sites

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The chemical and isotopic composition of organic aerosol collected on PM_1 filters samples was determined as a function of desorption temperature from the filter to investigate the main sources of organic carbon and the effects of photochemical processing on atmospheric aerosol. The filter samples were collected at an urban, a coastal and a forest site in Lithuania (Europe) in March 2013, which due to very low temperatures throughout the whole month, can be interpreted as winter-time samples.

The detailed chemical composition of organic compounds was analysed by a thermal desorption PTR-MS. The mass concentration of organic aerosol at the forest site was roughly a factor of 30 lower than at the urban and coastal site, indicating that in this cold month biogenic SOA formation was still very low. Moreover, the organic aerosol collected at the forest site was more refractory and contained a larger fraction of heavy molecules with m/z > 200.

The isotopic composition of the aerosol was used to gain information on the main sources of organic carbon. This is possible because the main winter-time sources of organic aerosol in Lithuania are biomass burning (Garbaras et al., 2015) and fossil fuel combustion and organic aerosol from biomass burning is enriched in ¹³C compared to organic carbon from fossil fuel emissions (Masalaite et al., 2015). δ^{13} C values of the organic aerosol samples showed a positive correlation with the mass fraction of several individual organic compounds (tentatively identified as biomass burning tracers). Most of these organic compounds contained nitrogen, indicating that nitrogen compounds may serve as tracers for biomass burning (Fig. 1). Other compounds that showed negative correlations with $\delta^{13}C_{OC}$ were identified as possible fossil fuel tracers. These compounds include heavy hydrocarbons and were on average less oxidized than the bulk organic carbon.



Figure 1. PTR–MS analysed organic mass fraction via $\delta^{13}C_{OC}$ values for CHN ions

The correlation of the $\delta^{13}C_{OC}$ via O/C ratio was positive at low but negative at high desorption temperatures at the forest site. We propose that this might be due to photochemical processing in the atmosphere, which could lead to accumulation of isotopically depleted carbon to the more refractory and more oxidized organic fraction. Detailed laboratory experiments are necessary to further investigate the photochemical processing of aerosol particles, before firm conclusion can be drawn.

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