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13C measurements on organic aerosol – ambient samples versus source studies

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The stable carbon isotopes 12C and 13C can be used to get information about sources and processing of organic aerosol (OA). We developed and tested a method to measure δ13C values of OA collected on filter samples in different volatility classes. These filter samples are introduced into an oven, where organic compounds are thermally desorbed in He at different temperatures. The compounds released at each temperature step are oxidized to CO_2 using a platinum catalyst at 550 °C. The CO_2 is then passed on to an isotope ratio mass spectrometer (IRMS) to measure δ13C ratios. With a similar setup the chemical composition at each temperature step can be determined using a Proton Transfer Reaction Time-of-Flight Mass Spectrometer (PTR-ToF-MS).

System evaluation with controlled test compounds showed that organic compounds usually start evaporating from the filter when their melting point is reached. Isotopic fractionation occurs only, if one temperature step is within a few degrees of the melting point of the substance, so that the substance only partially evaporates. However, this effect should be limited in an ambient sample containing thousands of individual chemical compounds.

We analysed aerosol samples collected in a tunnel in Brazil (vehicular emissions), laboratory generated secondary organic aerosol (SOA) from alpha-pinene ozonolysis, and ambient filter samples from a regional site in the Netherlands and an urban site in Belgium. First results indicate that SOA is more volatile than organic aerosol from ambient or tunnel filters. The δ13C ratios of SOA and vehicular emissions do not change strongly with oven temperature, i.e. the more refractory organic compounds have similar isotopic composition as the more volatile compounds. This is in contrast to ambient organic aerosol where the more volatile compounds evaporating below 200°C are depleted with respect to the refractory compounds. Possible reasons for this difference (mixture of sources vs the role of heterogeneous oxidation in the ambient filter samples) will be discussed and related to the chemical composition of the aerosol.