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Combined measurements of organic aerosol isotopic and chemical composition to investigate day-night differences in carbonaceous aerosol

Ulrike Dusek (1,2), Rupert Holzinger (1), Harro A.J. Meijer (2), and Thomas Röckmann (1)

(1) Utrecht University, Institute for Marine and Atmospheric research Utrecht (IMAU), Utrecht, Netherlands (u.dusek@uu.nl), (2) Centre for Isotope Research, Groningen University, Groningen, the Netherlands

PM2.5 filter samples have been collected during the Pegasos (Mai, 2012) and Actris (June/July 2012) campaigns at the CESAR site near Cabauw, the Netherlands. This site lies in a rural location surrounded by major urban centers and highways and is a good location for measuring the regional aerosol contamination in the Netherlands. High volume filter samples were taken over several days, but the aerosol was collected on separate filters during day and night time periods.
We analyzed these filters for carbon isotopes (14C and 13C) and detailed chemical composition of the organic fraction, which can be a powerful tool, for investigating sources and processing of the organic aerosol. Measurement of the radioactive carbon isotope 14C in aerosols can provide a direct estimate of the contribution of fossil fuel sources to aerosol carbon. The stable carbon isotopes 12C and 13C can be used to get information about sources and processing of organic aerosol. We use a method to measure d13C values of OC desorbed from the filter samples in He at different temperature steps. The chemical composition of the organic fraction at the same temperature steps can be determined using a Proton Transfer Reaction Time-of-Flight Mass Spectrometer (PTR-ToF-MS). The PTR-MS method is applied to the filter samples as well to aerosol collected in situ by a impaction using a Collection-Thermal-Desorption Cell.
First results show that the mass concentration of the carbonaceous aerosol is higher during night time than during day time, dominated by a strong increase of biogenic organic aerosol. This is at least partially caused by a shallow night time boundary layer combined with decreased traffic sources and increased condensation of semi-volatile biogenic gases during night-time. Evidence for the role of semi-volatile compounds in enhancing organic carbon (OC) night time concentrations comes from several observations: (1) semi-volatile OC with desorption temperatures lower than 250 °C increases more strongly during night time than less volatile OC. (2) PTRMS measurements show that concentrations of small molecules with m/z <100 amu increase more strongly during night-time than concentrations of larger molecules. (3) d13C values of OC only show day-night differences for semi-volatile carbon desorbed at 150 °C. This indicates that isotopic properties of semi-volatile compounds are more indicative of regional sources and processes and of low volatility compounds by large-scale processes.
The day-night differences of individual chemical compounds and compound classes will be further analyzed and discussed.