Day-night differences in the sources of carbonaceous aerosol determined by ^{14}C measurements at a regional background site

U. Dusek[1], E. Broekema[1], R. Holzinger[2], T. Röckmann[2], A. Hensen[3], and H. A. J. Meijer[1]

[1]Centre for Isotope Research, Groningen University, Groningen, the Netherlands
[2]IMAU, Utrecht University, Utrecht, the Netherlands
[3]ECN, Petten, the Netherlands

Keywords: carbonaceous aerosol, ^{14}C source apportionment, radiocarbon, PM2.5
Presenting author email: u.dusek@rug.nl

The origin of carbonaceous aerosol differs during day- and night-time, because emissions from major sources such as traffic, biomass combustion, and secondary organic aerosol formation show a distinct diurnal pattern. Source apportionment of aerosol carbon can give interesting insights into these varying source contributions. Moreover, assessing day-night differences can give an indication of the importance of local/regional carbon sources, since day-night differences should be averaged out during long-range transport. If local sources dominate, one could expect a strong diurnal variation in the source profile, but if long-range transport dominates the diurnal variation would be much weaker.

In this work we use radiocarbon (^{14}C) measurements on PM2.5 samples to estimate three main aerosol sources of organic and elemental carbon (OC and EC): Fossil fuel combustion (ff), biomass combustion (bb), and biogenic sources (bio). High volume filter samples of PM2.5 were collected at a regional background site in a polluted region of the Netherlands over several days. During this period, the aerosol was collected on separate filters, either during day (07:00 – 19:00) and night time periods (19:00-07:00), or during four 6-hour time periods: morning (04:00 – 10:00), midday (10:00 – 16:00), evening (16:00 – 22:00), and night (22:00 – 04:00). Day-night samples were collected for 3-4 days and the samples with higher time resolution for at least 6 days. The samples were only collected during time periods with similar air mass origin for the whole sampling period. Two sets of samples were analyzed for each season during 2012/13.

First results show that the contribution of fossil fuel combustion to EC and OC is higher during day-time then during night-time. This is valid for all seasons. During night-time biomass combustion plays a bigger role as a source of carbonaceous aerosol. Even in the summer, when biomass combustion is a small source, its contribution increases between 19:00 and 07:00. Figure 1 shows an example of source contributions during winter midday (10:00 – 16:00) compared to night-time (22:00 – 04:00). A smaller contribution of carbon from fossil sources during the night can be explained by the decrease in traffic, which is the major source of fossil carbon in the Netherlands. It is however not entirely clear, if the relative increase in biomass burning carbon during the night is due to the absence of fossil emissions, or if biomass combustion sources increase significantly during night-time. This question will be addressed with the help of radon data, which can serve as a proxy for change in boundary layer height and will allow to compare aerosol burdens during day and night.

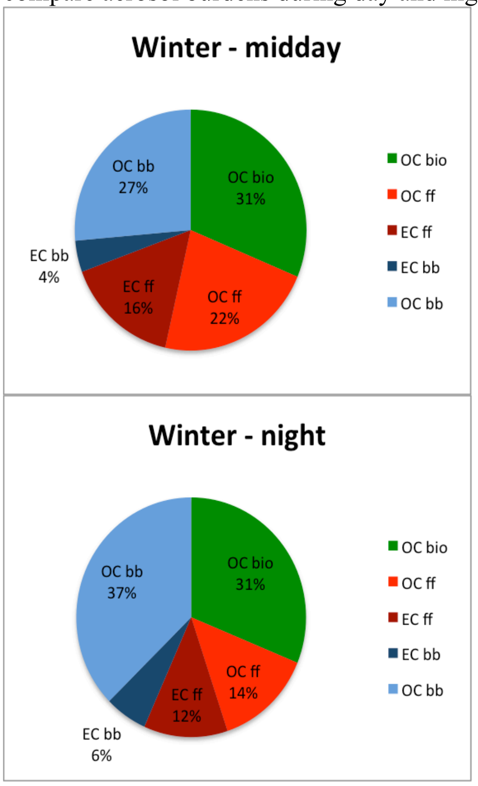

Figure 1. Comparison of source contribution to the carbonaceous aerosol during day- and night-time.

For a few sets of samples we conducted detailed case studies, where we measured the chemical composition of the organic carbon at different desorption temperatures. In all samples the concentration of less refractory organic carbon (desorption temperature up to 200 °C) increases at night-time much more strongly than the concentration of the more refractory carbon. Similarly, the concentration of small organic compounds with m/z < 100 also increases more strongly than heavier compounds. This indicates partitioning of semi-volatile OC as a possible additional night-time source. Overall, day night variations are clearly present, but not very strong, highlighting the importance of long-range transport of carbonaceous aerosol.

This study was funded by the Dutch Science Foundation (NWO grant Nr. 820.01.001).