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Fossil and contemporary sources of organic and elemental carbon at a rural and an urban site in the Netherlands

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Measurement of the radioactive carbon isotope 14C in aerosols can provide a direct estimate of the contribution of fossil fuel sources to aerosol carbon. In aerosol science, measurements of 14C/12C ratios are usually reported as fraction modern (fm), relative to an oxalic acid standard that, by definition, has fm=1. The radiocarbon signature gives a clear distinction between 'modern' carbon sources (fm around 1.1-1.2 for biomass burning and around 1.05 for biogenic secondary organic aerosol) and 'fossil' carbon sources (fm =0 for primary and secondary formation from fossil fuel combustion).

High volume filter samples have been collected since February 2011 at Cabauw, a rural location in the Netherlands, and additionally in May and June at two suburban locations around Rotterdam. We report measurements of fm for total carbon (TC), organic carbon (OC), water insoluble OC (WIOC) and thermally refractory carbon (RC) as a proxy for elemental carbon. The carbon fractions are isolated by combusting TC at 650 °C, OC and WIOC at 360 °C. Refractory carbon is defined as the carbon remaining on the filter after water extraction, combustion at 360 °C for 15 min and at 450 °C for 2 minutes. The method has been tested with test substances and real aerosol filters and shows little charring for water-extracted filters.

First results of 7 filter samples taken from February – Mai 2011 show fm(OC) generally larger than 0.86 at the rural site, except for one case, when a strongly polluted air mass originating in Eastern Europe reached the site. This indicates a strong contribution of natural sources to OC, even in the Netherlands, a very densely populated country with one of the highest levels of aerosol pollution in Western Europe. In particular, WSOC in the rural springtime aerosol seems to originate almost entirely from contemporary sources. Refractory carbon also showed relatively high fm, generally between 0.3-0.5, except in two cases, when marine air masses reached the site from the West and fm(RC) dropped to around 0.1. This could mean that biomass combustion plays a significant role around the Netherlands even in springtime. It is also possible that there exists a biogenic, organic component of the aerosol that evolves at such high temperatures that it is virtually inseparable from elemental carbon. Both hypotheses are consistent with low fM(RC) in marine aerosol. One urban sample was taken concurrently with one of the samples analyzed so far from the rural site. For both OC and RC, fM values were roughly 10-15% lower in the urban area than at the rural sites, which shows only a moderate influence of the urban fossil emissions on fm.

A more detailed comparison between the rural and urban location as well as fm values in other seasons will be presented, as more samples from the rural site become available.