The contribution of fossil sources to carbonaceous aerosol derived from the LOTOS-EUROS model and radiocarbon measurements

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A year-long record of the radioactive carbon isotope ^{14}C in carbonaceous aerosol was used to estimate fossil, biomass burning and biogenic sources at a regional background station in the Netherlands. The sampling was optimized to separate air mass conditions with continental and marine origin. Since marine carbon sources are small, samples collected in these air masses are indicative of regional sources in and around the Netherlands. Additionally, we used the LOTOS-EUROS model to calculate the contribution of fossil sources to elemental carbon (EC) and primary organic carbon (OC) using different emission inventories. Combining the ^{14}C source apportionment record with model results gives a unique opportunity for detailed model evaluation, which is difficult to achieve using only OC and EC concentrations. The model source apportionment of the primary OC will also be used to better estimate the sources of secondary OC.

Carbon from fossil sources accounts on average for around 80% of EC and 30% of OC and does not show a strong seasonal variation. Carbon from biomass combustion sources is much lower in the summer than in the winter, whereas biogenic carbon peaks in the spring, which was exceptionally warm in the year 2011. Concentrations of all carbon fractions are on average much higher in continental air masses, which indicates the importance of long-range transport of carbonaceous aerosol to the Netherlands. Interestingly, very little biomass burning carbon is detected in air masses with marine origin even in the winter, indicating that biomass combustion sources in the Benelux area are small.

First results from the LOTOS-EUROS model calculations show that the temporal trend in EC concentrations was well reproduced in the model, with exception of air masses arriving from Eastern Europe, where concentrations could be severely underestimated. Using the EUCAARI EC-OC emission inventory EC is overestimated by a factor of 1.8 on average by the LOTOS-EUROS model. Radiocarbon data show that this is mainly due to an overestimate of the fossil carbon. EC from biomass combustion was slightly overestimated and showed an excellent agreement with the measurement record (Fig.1). Consequently the modeled fossil fraction for EC is somewhat higher than in the measured record, on average 0.9.

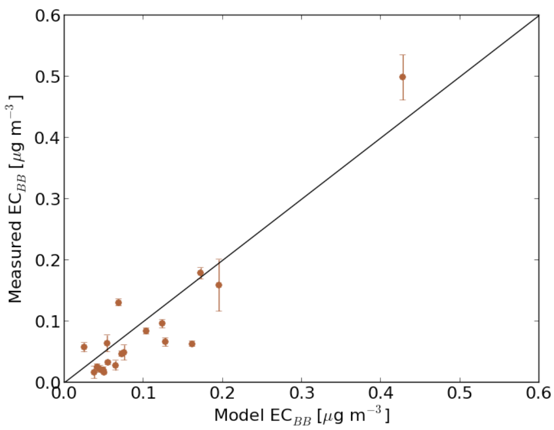

Fig.1: Comparison of modeled and measured biomass combustion EC (EC_{BB}) for the year 2011 at a regional background site in the Netherlands. The solid line indicates a 1:1 relationship

The primary OC calculated by LOTOS-EUROS was on average 1/3 of the measured OC, which implies that the majority of the OC is secondary in origin. The modeled fraction of primary OC was around 0.7 on average, indicating a dominance of fossil sources for primary OC. The measured fossil fraction of the total OC is significantly lower, around 0.3, which indicates that most secondary organic aerosol (SOA) should be from contemporary sources. Even water insoluble OC, which is considered a better proxy for primary OC shows lower fossil fractions around 0.5. This might indicate that OC from biomass burning is underestimated, however we will present strong evidence that primary fossil OC is overestimated by a similar factors as EC.

These results show that secondary organic aerosol formation needs to be included to adequately model the organic aerosol. We will present first LOTOS-EUROS model results of total OC concentrations including secondary OC formation, using a newly implemented volatility basis set (VBS) module. The modeled total OC closely resembles the measured OC concentration in the Netherlands.