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

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Carbonaceous material constitutes a significant fraction of the atmospheric aerosol. We used the LOTOS-EUROS model to calculate elemental carbon (EC) and primary organic carbon (OC) as well as the contribution of fossil sources for the year 2011 using different emission inventories. The model results are compared to a year-long source apportionment record of fossil versus contemporary carbon sources at a regional background station in the Netherlands. This record was derived from measurements of the radioactive carbon isotope 14C on carbonaceous aerosol and gives a unique opportunity for detailed model evaluation, since a comparison of model results to measured OC and EC concentrations alone is often inconclusive.

First results using the EUCARII EC-OC emission inventory show that at a regional background site in the Netherlands the modeled fossil fraction for EC is on average 0.9, with higher values around 0.95 in the summer and lower values in the winter due to increased biomass combustion. Overall, 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, whereas the EC from biomass combustion was only slightly overestimated. Consequently the LOTOS-EUROS model gave a somewhat higher fossil fraction for EC than the measurements. Overall the temporal trend in EC concentrations was well reproduced in the model, with exception in air masses arriving from Eastern Europe, where concentrations could be severely underestimated.

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 fossil 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 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 model results of total OC concentrations including SOA formation, which closely resemble the measured OC concentration in the Netherlands.