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Carbonaceous Aerosol in Europe out of the woods and into the blue?

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Abstract

Particulate matter (PM) in the atmosphere influences weather and climate and may have important health impacts. Regional scale chemical transport modelling aims to describe the composition of particulate matter, to track different sources, estimate their relative importance, and to give realistic predictions of responses to changes in emissions and atmospheric conditions. The focus of this thesis is the modelling of an important constituent of PM — carbon containing PM.

The EMEP MSC-W chemical transport model is used for European policy making regarding air pollution, to provide scientific support to the convention on long-range transboundary air pollution (CLRTAP). The organic aerosol (OA) treatment in the EMEP model has been extended to include more realistic primary OA emissions, and new schemes for the formation of secondary OA, based on the volatility basis set method.

Long-term model simulations of OA and elemental carbon (EC) over Europe have been performed for the period 2002-2010. The model results were compared to observations, including source-apportionment data. Total organic carbon concentrations matched measured concentrations for summer periods, but problems were found during winter, with poor agreement between modelled and measured organic carbon, and tracers of wood-burning. To tackle these problems a new inventory for emissions of OA and EC from residential wood combustion (RWC) was developed. Total European OA emissions from RWC are almost 3 times larger in the new inventory than in the old one. According to the new inventory, about 60% of the primary OA emissions in Europe are due to RWC. EC emissions are to a larger extent due to fossil fuel combustion; RWC emissions contribute about 1/5 of the total anthropogenic fine particle EC-emissions in Europe.

The model results indicate that many sources contribute to OA in Europe. During summer, fossil fuel combustion, biomass burning and biogenic secondary OA all contribute considerably. RWC is the dominant OA source during winter, contributing more than 50% to the model OA. According to the model results, non-fossil sources contribute more to regional scale OA than fossil fuel, except in the Po Valley during summer. EC comes mainly from fossil fuel during the warm seasons, but EC from RWC is important during winter.

Modelling is useful to investigate potential impacts of newly discovered sources of organic aerosol. Biotic stress-induced emissions (SIE) were investigated in this thesis. The fractions of stressed trees in European forests were estimated, based on observed tree damage. Emission estimates for sesquiterpenes, methyl salicylate and unsaturated C₁₇-compounds, and the SOA yield from the oxidation of these SIE, were based on plant chamber experiments. The model results suggest that SIE may contribute substantially to SOA in Europe. During some periods, SIE may contribute more to OA than the non-stressed biogenic emissions of volatile organic compounds. Thus, further research on SIE is warranted.

Keywords: organic aerosol, elemental carbon, chemical transport modelling, residential wood combustion, biotic stress induced emissions, source apportionment, EMEP MSC-W model