Simulation of the Size-Composition Distribution of Atmospheric Nanoparticles over Europe

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New particles are introduced in the atmosphere by direct emission from a variety of sources and nucleation (in-situ formation). These ultrafine particles can grow to larger sizes and become cloud condensation nuclei (CCN) affecting the formation and properties of clouds. In this way aerosols can indirectly affect the energy balance of the planet. Sulfuric acid plays a dominant role in the formation of new particles, but sulfuric acid condensation can only explain a small fraction of their growth. In most environments organics appear to be responsible for a significant fraction of the fresh particle growth to larger sizes (Riipinen et al., 2010). However, significant uncertainties remain about both the processes of new particle formation and growth.

In this work, we continue the development of a three-dimensional chemical transport model, PMCAMx-UF, focusing on the simulation of the ultrafine particle size distribution and composition. The Volatility Basis Set (VBS) approach is used for the simulation of organic aerosol. PMCAMx-UF is applied to Europe to quantify the contribution of organic vapors to total number concentrations and fresh particles' growth. PMCAMx-UF uses the new version of the Dynamic Model for Aerosol Nucleation and the Two-Moment Aerosol Sectional algorithm to track both aerosol number and mass concentration using a sectional approach. We evaluate the model predictions for the European domain against field observations collected in the PEGASOS campaigns during 5 June to 8 July 2012 and 24 April to 18 June 2013. The measurements include both ground stations across Europe and airborne measurements from a Zeppelin.

PMCAMx-UF reproduces reasonably well the N_{10} (number concentration of particles larger than 10 nm) hourly observations both aloft (over the Po Valley in Italy) as well as at the ground level. The ground level concentrations of N_{100} are well predicted (normalized mean error of 60%, normalized mean bias -25%) while there is tendency to overestimate N_{10} by approximately 50%. The condensation of organics increases the N_{100} concentration mainly in central and northern Europe by 50-200% and the highest predicted N_{100} concentration is in East Mediterranean Sea (Fig.1) while, on the other hand, it decreases by 10-40% the N_{10} concentration in central and eastern Europe. This counterintuitive result can be explained by an increase of the aerosol surface area leading to an increase of the rate of coagulation and then to a decrease of the ultrafine particle concentration. Sensitivity tests highlight the importance of chemical aging reactions of secondary organic aerosol for the ultrafine particle growth and total particle number concentration.

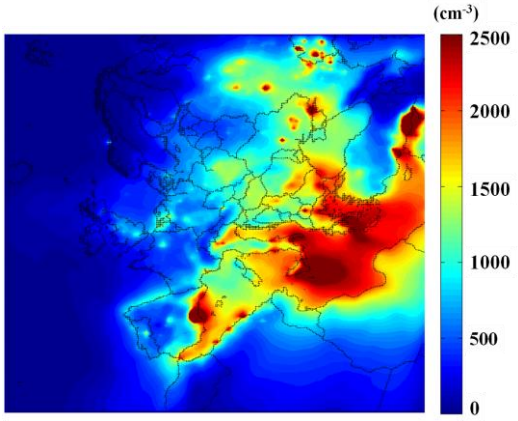

Figure 1. Predicted aerosol number concentration for particles larger than 100 nm with condensation of organics vapors at the ground level during June-July 2012.

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Adams, P. J., and J. H. Seinfeld (2002): Predicting global aerosol size distributions in general circulation models, *J. Geophys. Res.*, 107, 4370.

Fountoukis, C., et al. (2012): Simulating ultrafine particle formation in Europe using a regional CTM: contribution of primary emissions versus secondary formation to aerosol number concentrations, *Atmos. Chem. Phys.*, 12, 8663–8677.

Patoulias, D., Fountoukis, C., Riipinen, I., and Pandis, S. N. (2014): The role of organic condensation on ultrafine particle growth during nucleation events, *Atmos. Chem. Phys. Discuss.*, 14, 30761-30798.

Riipinen, I., et al. (2011): Organic condensation: a vital link connecting aerosol formation to cloud condensation nuclei (CCN) concentrations, *Atmos. Chem. Phys.*, 11, 3865–3878.