Modeling of long-term particle formation and growth in the planetary boundary layer

L. Zhou[1,2], M. Boy[1], T. Nieminen[1], D. Mogensen[1], S. Smolander[1], and Markku Kulmala[1]

[1]Department of Physics, University of Helsinki, P. O. Box 48, 00014 Helsinki, Finland
[2]Department of Forest Science, University of Helsinki, P.O. Box 27, 00014 Helsinki, Finland
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Presenting author email: luxi.zhou@helsinki.fi

Natural and anthropogenic aerosols may have a great impact on climate as they can directly interact with solar radiation and indirectly affect the Earth's radiation balance and precipitation by modifying clouds. In order to quantify the direct and indirect effect, we must understand the complex processes that connect an aerosol particle to a cloud droplet. However, while modern measurement techniques are able to detect particle sizes down to nanometre all the way from ground up to the stratosphere, the data does not serve for all of our needs for understanding the processes. Hence we will demonstrate a modelling approach to investigate the complex processes of aerosols in the planetary boundary layer (PBL) over the boreal forest.

SOSAA (model to **S**imulate the concentration of **O**rganic vapours, **S**ulphuric **A**cid, and **A**erosol) was developed by adding an aerosol dynamics model UHMA (Korhonen, 2004) to an exiting chemistry – transport column model SOSA (Boy, 2011). It was constructed to study the emissions, transport, chemistry, as well as aerosols in the PBL. Up to our knowledge, it is the first column model existing in the world with detailed chemistry and aerosol dynamics. Thanks to the parallel computing, the model can be used to study the aerosol processes in the PBL for a few days up to a year with high vertical resolution. As a first application of the model, we present new particle formation study of the year 2010 in a Finnish boreal forest site, Hyytiälä, Finland, where the Station to Measure Ecosystem Atmosphere Relation (SMEAR II) is located (Hari and Kulmala, 2005). Chemistry in the model is validated against the measurements from the HUMPPA-COPEC campaign, which was conducted at SMEAR II during summer 2010 (Williams, 2011).

We tested kinetic nucleation (Weber, 1997) and organic nucleation (Kulmala and Kerminen, 2008) theory. Kinetic nucleation theory states that the nucleation rate is proportional to the square of ambient sulfuric acid concentration. Organic nucleation theory assumes that both sulfuric acid and organics are taking part in the nucleation, although the detailed knowledge about the participating compound is not achieved up today. In the simulations, the nucleating organics are the first stable reaction products from monoterpenes oxidized by hydroxide (OH).

Simulation results show that organic nucleation theory generally gives better prediction in nucleation rates compared to organic nucleation theory. The agreement is relatively well in spring time compared to autumn and winter. One reason could be that emissions were underestimated in autumn and winter. We include the first stable oxidation products from the oxidation of monoterpenes as the organic condensing vapors. Even though the growth of particles in the model suffers from the uncertainties in organic chemistry and the missing knowledge of physical properties of organic compounds. In Figure 1, we see that the model is able to predict the number concentrations of particles between 25 and 100 nm sufficiently good. In general, the model captures 10 out of 11 observed event days in 2010.

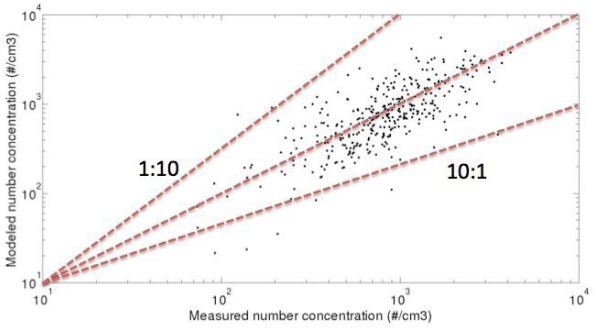

Figure 1.Scatter plot of measured number concentrations of particles between 25 and 100nm against simulated.

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