

Modelling of aerosol dynamics and particle chemistry using a new nucleation theory

Pontus Roldin¹, Guy Schurgers², Birgitta Svenningsson¹, Erik Swietlicki¹, Axel Eriksson¹, Joakim Pagels³, Erik Nilsson¹, Matthias Ketzel⁴ and Susanna Gustafson⁵

¹Div. of Nuclear Physics, Lund University, SE-221 00, Lund, Sweden, ²Dept. of Physical Geography and Ecosystem Analysis, Lund University, ³Div. of Ergonomics and Aerosol Technology, Lund University, ⁴NERI, Dept. of Atmospheric Environment, Aarhus University, ⁵Environmental Dept., City of Malmö.

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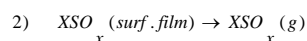
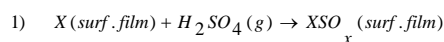
For this work a coupled column trajectory model for aerosol dynamics, gas phase chemistry and radiative transfer calculations (ADCHEM¹) has been applied. The model is an updated version of the model described by Roldin et al., (2008). The main updates since the previous version are: 1) more detailed organic and inorganic particle chemistry with sulphate, nitrate, ammonium, sodium, chloride, EC, POA, anthropogenic and natural SOA, 2) in cloud processing of the aerosol particles including SO₂ and H₂O₂ condensation.

The aim of this work is to study the particle chemistry in the sub-micrometers size range and test a new nucleation theory developed in this work. The modelled results have been compared with measurements from DMPS, AIS, AMS and HTDMA at the Vavihill rural background station in Southern Sweden (13°09'E, 56°01'N).

The model study was focused on an 11 day period between 29th of September and 9th of October 2008. Air mass trajectories were retrieved from the internet based HYSPLIT model. Anthropogenic gas and PM_{2.5} emissions from EMEP were included along the trajectories. For Denmark and Southern Sweden regional emission inventories from the Danish National Environmental Research Institute and Malmö Environmental Department were used instead of the EMEP data. Isoprene and monoterpene emissions over Europe were derived with the vegetation model LPJ-GUESS (Schurgers et al., 2009).

The new nucleation mechanism included in the model assumes that stable organosulphate nucleation clusters (XSO_x), forms from reactions between gas phase sulphuric acid and an organic surfactant (X) at the particle surface water film (eq. 1). In the model the organic surfactants are formed exclusively by condensation of oxidation products from monoterpenes, reacting with OH. After the organosulphate molecules have been formed they can evaporate out into the gas phase (eq. 2), and immediately form stable nucleation clusters with one organosulfate molecule as core (eq. 3). Decreasing RH promotes nucleation since the particle water in the surface film get more concentrated on organosulphates, which then are

forced out to the gas phase. The number of clusters formed, also depends on the particle surface area concentration (A), in each diameter size bin i (eq. 3). The proposed nucleation theory has been compared with simulations using the kinetic nucleation theory. Examples from the model and measurements can be seen in figure 1.



$$3) \quad J_{1nm} = k[H_2SO_4(g)] \sum_{i=1}^N (A_i [X(aq, bulk)]_i)$$

$$k = k_1 k_2 \exp\left(-\frac{\Delta G_{surf}^O - bulk}{RT}\right)$$

$$4) \quad [X(\text{surf. film})] = [X(aq, bulk)] \exp\left(-\frac{\Delta G_{surf}^O - bulk}{RT}\right)$$

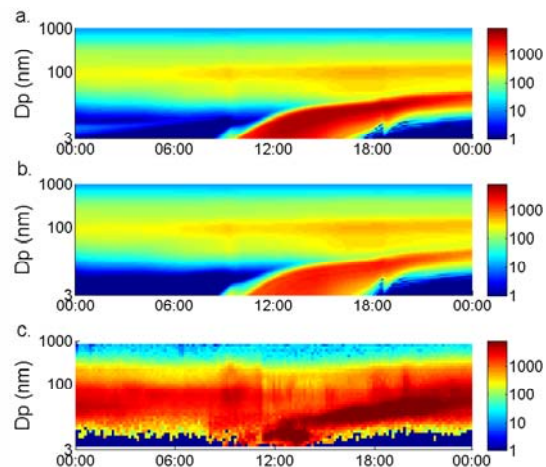


Figure 1: Particle number size distributions ($dN/d\log Dp \text{ cm}^{-3}$) for a. kinetic nucleation theory, b. nucleation theory proposed in this work, and c. measured with DMPS at Vavihill, 6th of Oct., 2008.

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