Estimating pre-existing aerosol effects on tropospheric aerosol production

M. Dal Maso[1], L. Liao[2], H. Vehkamäki[2], H.Korhonen[3], and K.E.J. Lehtinen[3]

[1]Tampere University of Technology, Dept. of Physics, PO Box 692, 33101 Tampere, Finland
[2]University of Helsinki, Department of Physics, PO Box 48, 00014, Helsinki, Finland
[3]Finnish Meteorological Institute, Kuopio Unit, P.O.Box 1627, 70211 Kuopio
Keywords: atmospheric aerosol, coagulation, new particle formation, modelling
Presenting author email: miikka.dalmaso@tut.fi

Understanding the source strength of tropospheric aerosol production is key to successfully predicting aerosol concentrations and their effect on human health and climate (Kulmala et al., 2004). Aerosol sources generally add aerosol particles or gas-phase aerosol precursors into atmospheric volumes already containing a significant number of pre-existing aerosol particles. This pre-existing aerosol acts as a sink for fresh aerosol particles as well as gaseous aerosol precursors.

Aerosol source processes are generally described as a source rate of aerosol particles into a given size range. Source rates from aerosol production processes such as combustion or secondary gas-to-particle conversion are often obtained in idealized conditions with no pre-existing aerosol present. Alternatively, the production rate is reported in context of realistic ambient concentrations but without characteristic information of the pre-existing aerosol.

The sink processes, caused by condensation and coagulation during the early steps of aerosol lifetime, can significantly alter the aerosol size distribution on timescales that are shorter than those implemented in conventional models predicting tropospheric aerosol concentrations. Parameterizations to take these effects already exist (eg. Lehtinen et al.,2007; Anttila et al, 2010), but they do not take into account effects caused by the size dependence of the particle growth (Vuollekoski et al., 2012) and the changes to the time dependence of the size distribution evolution caused by this.

In this study, we explore the different mechanisms that cause pre-existing aerosol to affect new aerosol sources. We use two models to study the effect of pre-extising aerosol on the source strength of atmospheric aerosol production processes. The molecular resolution model MOREAD, developed from the model by Lehtinen and Kulmala (2003), is used to quantify the effect of nanoparticle dynamics on the source rate of secondary nanoparticle production, and specially how these dynamics affect parameterizations of the formation rate of such particles. We also applied the 0d box model CHAMMO to study the carry-on effect of the pre-existing particles on the survival rate of particles to climatically significant sizes.

In addition to pure aerosol dynamics, pre-existing aerosol affects the particle formation process and the perceived dependencies on precursor vapors in the case of multiple chemical compound participating in the particle production. We studied the formation of secondary particles from sulphuric acid and VOC oxidation, and present results the describe particle formation dependencies in presence of sinks for nucleating and condensing vapours as well as additional sinks, such as dilution.

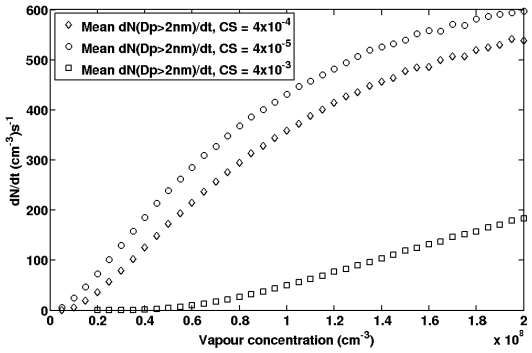

Figure 1: The perceived particle formation rate in a model aerosol with a prescribed formation rate of 1000 $cm^{-3}s^{-1}$ under varying background aerosol concentrations

Our model showed a strong influence of background aerosol on perceived particle formation rate even if the formation rate is technically derived very close to the actual formation size (see Fig. 1). The background aerosol is also key to the survival of fresh aerosol to CCN size; additionally, the relative chemical and condensation sink determine the dependency of particle formation rate on VOC precursor and oxidant concentration. We will illustrate these effects with field data, which shows clearly reduced particle formation activity with increasing background aerosol.

Anttila, T., et al: *J. Aerosol Sci.*, 41, 621–636, doi:10.1016/j.jaerosci.2010.04.008, 2010. 18783, 18785

Kulmala, M., et al: Formation and growth rates of ultrafine atmospheric particles: A review of observations *Aerosol Science* 35, 143-176, 2004

Lehtinen, K E J. and Kulmala, M. (2003): A model for particle formation and growth in the atmosphere with molecular resolution in size, *Atmos. Chem. Phys*., 3, 251-258

Lehtinen, K.E.J. et al: Estimating nucleation rates from apparent particle formation rates and vice versa: Revised formulation of the Kerminen–Kulmala equation *J. Aerosol. Sci* 38 (9), 988-994, 2007

Vuollekoski, H., et al: A numerical comparison of different methods for determining the particle formation rate *Atmos Chem Phys*, 12, 2289–2295, 2012