On theory, modelling and data analysis of atmospheric aerosol formation and growth

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New atmospheric aerosol particle formation and subsequent growth have been observed frequently at various locations all over the world. Kulmala et al. (2012) presented a protocol of methods how the atmospheric particle formation rate, nucleation rate and growth rate can be determined from measurements of particle size distribution evolution.

In this plenary presentation, I will revisit this 'protocol' as well as some more recent developments on the topic (e.g. Vuollekoski et al., 2012; Leppä et al., 2013; Korhonen et al., 2014; Olenius et al., 2015; Kontkanen et al., 2015). I will also discuss some limitations in applying the first nucleation theorem (Kupiainen-Määttä et al, 2014; Malila et al., 2015) and differences in the implications when using continuous or discrete size distribution dynamics (Olenius et al., 2015).

Many of the studies have focused on atmospheric nucleation events, where particle formation starts typically before noon and competition between growth and scavenging onto background particles determines if the particles grow to sizes at which they are climatically of significance. Then the dynamics is essentially the formation of a nucleation mode and its growth by condensation. Very different type of size distribution dynamics can be observed in chamber experiments where nucleation, growth, scavenging and wall deposition start in either an empty chamber or with seeds, sometimes producing a nearly steady-state size distribution (Lehtipalo et al., 2014; Olenius et al., 2014).

Our method of analysis has been to use synthetic experiments, i.e. generate either field-experiment or chamber experiment type data by using aerosol dynamic simulations, and, analyse the data by the same methods as have been used for the real experiments. As the 'answers' are now known, we can directly evaluate how well our methods work. This approach has already resulted in several improvements in the methodology how new particle formation data is analysed.

Not only measurement techniques (e.g. Zhao et al., 2011; Jokinen et al., 2012) and data analysis methods but also detailed modelling techniques (McGrath et al., 2012) now extend to the smallest clusters. Look-up tables of particle formation rates calculated by a detailed multicomponent cluster dynamics model (ACDC), with evaporation rates from quantum chemistry computations, have now been implemented also to large-scale atmospheric models with promising results (Baranizadeh et al., 2016). Without any artificial fudge factors we were able to obtain correct order of magnitude vertical profile number concentrations in the atmosphere above Europe. This is a huge improvement compared to several previous studies in which only empirical nucleation rates or classical theories with correction factors of the order 10^6 have had to be used in order to reach similar performance.

With respect to continuous vs. discrete size distribution representation, our main focus is the condensation term in the GDE. The continuous GDE is typically applied in a form where the condensation 'diffusion term' is missing (Seinfeld and Pandis, 2006). This may result in inaccuracies when relating e.g. growth rate with apparent formation rate (particle flux in size space; Olenius et al, 2015).

Baranizadeh et al. (2016) *Geosci. Model Dev. Discuss*., in review.
Jokinen et al. (2012) *Atmos. Chem. Phys.* 12, 4117.
Kontkanen et al. (2015) *Atmos. Chem. Phys. Discuss.*, in review.
Korhonen et al. (2014) *J. Aerosol Sci.* **69**, 13.
Kulmala et al. (2012) *Nature Protocols* **9**, 1651.
Kupiainen-Määttä et al. (2014) *J. Aerosol Sci.* **77**, 127.
Lehtipalo et al. (2014) Boreal Env. Res. **19**, 215.
Leppä et al. (2013) *Atmos. Chem. Phys.* **13**, 463.
Malila et al. (2015) *J. Chem. Phys.* **142**, 011102.
McGrath et al. (2012) *Atmos. Chem. Phys.* 12, 2345.
Olenius et al. (2014) *J. Aerosol Sci*., **78**, 55
Olenius et al. (2015) *J. Aerosol Sci.* **90**, 1.
Seinfeld and Pandis (2006) Atmospheric Chemisty and Physics. 2nd edition, Wiley.
Vuollekoski et al. (2012) *Atmos. Chem. Phys.* **12**, 2289.
Zhao et al. (2011) *Atmos. Chem. Phys.* 11, 10820.