Atmos. Chem. Phys. Discuss., 13, C5323–C5326, 2013
www.atmos-chem-phys-discuss.net/13/C5323/2013/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Influence of aerosol lifetime on the interpretation of nucleation experiments with respect to the first nucleation theorem" *by* S. Ehrhart and J. Curtius

S. Ehrhart and J. Curtius

ehrhart@iau.uni-frankfurt.de

Received and published: 29 July 2013

We thank Referee #1 for the comments and questions.

RC: In the manuscript "Influence of aerosol lifetime on the interpretation of nucleation experiments with respect to first nucleation theorem" the authors present the usage of SAWNUC model with three schemata, the neutral, ion-induced and neutral barrier free (kinetically limited) particle formation at one nucleation temperature (248K) and one lifetime (500 s). Their overall conclusions are reasonable, but it is bit disappointing that authors have not even try to compare their simulations to any experimental data

available. Since the authors refer through the manuscript to "... different laboratory conditions ...", one would expect to find such a comparison, e.g. in studies of Berndt et al. (2010) and also Sipila et al. (2010), but there is many more other studies with wide variability in experimental conditions, the comparison of particle counters with different cut-off efficiencies is reported in wide range of experimental conditions (RH, residence time), also in Brus et al. (2011) and Kirkby et al. (2011) three different nucleation temperatures are reported. Moreover similar comparison of PARNUC model to experimental data was done in Kirkby et al. (2011), so it should not be a big problem.

AC: The main reason for not including these studies was that the effect of walls is most pronounced at low GR. As can be seen in Figure 1. At [H2SO4] $\sim$ 1e8 cm-3 the characteristic time for particles to nucleate and grow to detectable size has only a small influence on the determined n*. Most studies on binary nucleation however were conducted at high GR and high nucleation conditions. Due to the Kirkby et al 2011 results we must also conclude that some experiments may be influenced by contaminants and are therefore in fact measurements of ternary nucleation. It should also be noted that each reactor has an individual loss behaviour. A chamber has losses that are determined mainly by it's geometry and turbulence created by mixing fans. An important loss term for flow reactors is the flow out of the reactor which ends any process occurring regardless of the particle size. In addition the flow reactors have wall loss terms that depend on the individual flow conditions. We think that our findings are quite general and should not just be restricted to a comparison with one experiment, e.g. the CLOUD experiment, as results from other experiments are influenced in a similar way whenever GR are small and losses cannot be neglected. A detailed comparison of CLOUD nucleation rates with SAWNUC results over a wide temperature range is beyond the scope of this paper and is subject of another paper with more authors involved (Ehrhart et al., in preparation, 2013).

RC: Some reasoning or justification why authors have chosen particularly nucleation temperature of 248 K and lifetime 500 s is needed.

AC: main reason to use 248 K was that at this temperature neutral binary nucleation gives a significant nucleation rate. And there is a region where neutral nucleation exceeds ion-induced nucleation with the classical SAWNUC thermodynamics. The lifetime of 500 s was chosen as it represents the CLOUD chamber lifetime and a typical lifetime due to condensational sinks in the troposphere, e.g. Kulmala et al. 2005. A variation of the lifetime is given in Figure 4 for kinetically limited nucleation.

RC: What RH was used in simulations?

AC: An RH of 40% was used. The information is added to the revised version of the paper.

RC: Wider discussion how will develop n_d vs. n* as a function of nucleation temperature, lifetime, RH, and discussion and recommendations for nucleation lab experiments would be appreciated. Where one would expect greater errors?

AC: The development of n_d and n* as function of temperature is covered by the 2nd nucleation theorem. In fact, most nucleation experiments were done at a single temperature or without temperature control. The RH dependency of nucleation rate is out of the scope of this paper. The authors think that the most appropriate way is to minimize losses to walls and aerosols such that the growth time is smaller than the wall loss life time (see answers to Reviewer #2 and new discussion and eq (6) in revised manuscript). Furthermore it is best to analyse nucleation experiments is to use a micro physical model to simulate the general dynamics equation starting from the monomers.

RC: Otherwise the manuscript reads well and especially appreciated is its length.

AC: Thank you.

References: Kulmala, M.; Petäjä, T.; Mönkkönen, P.; Koponen, I. K.; Dal Maso, M.; Aalto, P. P.; Lehtinen, K. E. J. & Kerminen, V.-M. On the growth of nucleation mode particles: source rates of condensable vapor in polluted and clean environments Atmospheric Chemistry and Physics, 2005, 5, 409-416

Berndt, T. et al.: Laboratory study on new particle formation from the reaction OH + SO2: influence of experimental conditions, H2O vapour, NH3 and the amine tert-butylamine on the overall process, Atmos. Chem. Phys., 10, 7101-7116, doi:10.5194/acp-10-7101-2010, 2010. Brus, D. et al.: Homogenous nucleation of sulfuric acid and water at close to atmospherically relevant conditions, Atmos. Chem. Phys., 11, 5277–5287, doi:10.5194/acp- 11-5277-2011, 2011. Kirkby, J et al.: Role of sulphuric acid, ammonia and galactic cosmic rays in atmospheric aerosol nucleation, Nature, 476, 429–433, 2011. Sipila, M. et al.: The role of sulphuric acid n atmospheric nucleation, Science, 327, 5970, 1243–1246, doi:10.1126/science.1180315, 2010.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 9733, 2013.