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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Particle formation in the Arctic free troposphere during the ASTAR 2004 campaign: a case study on the influence of vertical motion on the binary homogeneous nucleation of H_2SO_4/H_2O" *by* F. Khosrawi et al.

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We thank Shan-Hu Lee for the constructive, helpful criticism. We followed the suggestions of Shan-Hu Lee and revised the manuscript. The grammar and spelling of the text has been checked thoroughly and to avoid any confusion we would like to mention here that the paper has been written in American English.

Main scientific comments include the range of H_2SO_4 concentrations used in this simulations (0, 40, 80 pptv) are way too high – I understand that H_2SO_4 levels in free troposphere

are less than e^7 *cm*$^{-3}$ *range (pptv) so this simulation should be made more reasonably at atmospherically relevant conditions; such as* e^5, e^6, e^7, *and* e^8 H_2SO_4 *cm*$^{-3}$ *range.*
The sulfuric acid mixing ratios of 40 and 80 pptv are indeed on the upper limit of what has been measured up to know while 1 pptv is a typical concentration for the considered altitude region. Due to the fact that earlier measurements have been always made in clean air higher values than 1 pptv seem to be unreasonable. However, more recent measurements of H_2SO_4 in polluted air show indeed that higher sulfuric acid concentrations occur. Further, one should consider that 40 pptv or 80 pptv have not the same concentration in cm^{-3} at each altitudes. At the altitudes considered here the sulfuric acid mixing ratios are correspondent to a concentration of 4.5 to 6 $\times$ 10^{-8} cm^{-3} which are roughly in the range of values measured in polluted air (see page 21986, line 24pp). We changed the the first two sentences beginning at line 27 on page 21968 as follows: *The simulations were performed along the 36 trajectories for three different sulfuric acid mixing ratios* $\mu_{H_2SO_4} = 1$, *40 and 80 pptv (1 pptv=2.67×10*^{7} *molecules cm*$^{-3}$ *at 0° C and 1013 hPa), where a mixing ratio of* $\mu_{H_2SO_4} = 1$ *pptv is representative for clean air and* $\mu_{H_2SO_4} = 40$ *and mixing ratios of 80 pptv are representative for polluted air. It should be noted here that the mixing ratios we used for being representative for polluted air are on the higher range of previously measured concentrations.* Further, concerning the suggestion to include some intermediate steps in the sulfuric acid concentration like 10, 20, 40, 60, 80 pptv. This indeed would be interesting, however this is beyond the scope of this study. More importantly, this even would not change the results since we found the best agreement between model and measurements using a mixing ratio of 40 pptv.

While the authors focused on why NPF took place on May 24 and not on other days, it would be more important to also address when NPF does not occur. It is well recognized now that NPF takes place in almost all parts of the world, except some specific regions such as Amazon forest boundary layer; however, what is unclear is when NPF does not occur and why. This is a far more important question. The authors claimed that during the entire ASTAR 2004 *campaign, there was only one NPF event.*

One important thing one has to consider here is that we focus on NPF of H_2SO_4/H_2O aerosols and not on NPF in general. The binary homogeneous nucleation of H_2SO_4 and H_2O is strongly dependent on temperature, relative humidity and relative acidity (or partial pressure of H_2SO_4). Thus, new particle formation is not occurring unless favorable conditions for particle formation are reached. A region which is generally favorable for particle formation is the tropical upper troposphere. However, in other regions the particle formation due to binary homogeneous nucleation of H_2SO_4 and H_2O is generally quite low. Dynamical effects like mixing of air masses, convection or vertical updrafts can help to reach the conditions that are favorable for particle formation even in that regions. We improved the introduction to make this more clear (see also reply to the comments made by reviewer 1). Further, our statement that particle formation was only observed once during the ASTAR 2004 campaign, namely on the 24 May, refers to the upper troposphere only. Particle formation has been quite frequently observed below 4 km as discussed on page 29165, l4-6. However, in general, very little is known about particle formation in the polar (upper) troposphere. Therefore, it is quite important to investigate why particle formation occurred - and not to investigate why there was no particle formation during the other situations.

It would be useful to mention how binary nucleation schemes Karcher (1998), Jaecker-Voirol et al., 1987) and Laaksonen Kulmala (1991), used in the authors' modeling simulations, stand as opposed to current nucleation parameterizations.

The theory behind the parameterization of Vehkamäki et al. (2002) and the nucleation code by Kärcher (1998) is the same. However, a parameterization is a approximation of the model equations as used e.g. in our box model. The advantage of a parameterization compared to solving the model equations numerically is that a lot of computer time can be saved. This is especially important if it is intended to simulate nucleation processes in the frame of a large-scale model simulation. Especially with the Vehkamäki et al. (2002) parameterization computer time can be saved by a factor

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of 1/500. However, a disadvantage is that a parameterization is only valid in a certain temperature range. Especially, the Vehkamäki et al. (2002) parameterization is valid only down to temperatures of 230 K which makes it difficult to apply it to stratospheric conditions where also temperatures below 230 K are frequently reached. Our model was originally developed to simulate the particle formation especially in the upper troposphere/lower stratosphere.

Also, it is unclear to me why HNO_3 *is included in this nucleation simulations, as* HNO_3 *vapor pressures are too high to contribute condensation for nucleation of small particles considered in this work (as opposed to micron size PSCs).*

We are sorry for this misunderstanding. HNO_3 is not included in the nucleation simulation. The model set up is for simulating H_2SO_4/H_2O aerosols in the upper troposphere and lower stratosphere. Thus, in that region HNO_3 uptake (like e.g. for PSC type Ib formation) is also of interest. However, HNO_3 is only used in the model if condensation (uptake of HNO_3 and H_2O) is considered. In the present model simulations we only consider nucleation and coagulation, thus HNO_3 is not used in the simulation, but given in the initialization of the model since the initialization is independent from which processes are considered in the box model simulation. To make this more clear in the model description we included the following sentence (at the end of the section 'Microphysical box model'): *In the present study box model simulations were performed considering nucleation and coagulation.*

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 21959, 2009.

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