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Interactive Comment

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 reviewer 3 for the constructive, helpful criticism. We followed the suggestions of reviewer 3 and revised the manuscript. Especially, we improved the introduction and added some more recent publications. Further, we state now more clearly in the manuscript why this study is of importance.

Major concerns:
In general, the introduction is somewhat poorly cited and perhaps a few more recent references

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are notably missing. Hegg and Baker (2009) provide a good overview.
The following more recent references have been included in the discussion on the different nucleation processes: Laaksonen et al. (2008) and Curtius et al. (2006) for ion-mediated nucleation and Yu (2006) and Benson et al. (2009) for ternary homogeneous nucleation involving ammonia. Further, we included the following sentence where we refer to the overview paper from Hegg and Baker (2009): *A good overview over all these different nucleation processes and some further references can be found in Hegg and Baker (2009)*.

Minor comments:
I believe the use of the expression "none nucleation" should be replaced with "non-nucleation"
We have replaced the expression "none nucleation" with "non-nucleation"'.

In section 2, the POLAR 2 aircraft is not described, while the POLAR 4 is.
We included the description of the Polar 2 aircraft. The text reads now: *The Polar 2 and Polar 4 are research aircrafts of the Dornier 228-101 and 228-200 type, respectively, owned by the Alfred Wegener Institute for Polar and Marine Research (AWI) in Bremerhaven and operated by the DLR flight facility department. Both are two-engine turboprop aircraft well capable of operating under the harsh conditions of the polar environment.*

p21963, l10: change "extend" to "extent"
This has been corrected.

p21964, l3: change "were a Condensation...", to "was a Condensation..."
This has been corrected.

p21964, l12: remove respectively
This has been corrected.

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p21966, l6: provide references for the statement: "However, this should not affect our results..."

Unfortunately, there exists no reference for this statement. This is a conclusion we made from what has been published earlier about condensation of H_2SO_4. To make this more clear we changed the sentence as follows: *However, this should not affect our results since the H_2SO_4 condensation is a very slow process. Further, in the atmosphere H_2SO_4 concentrations are much lower than H_2O concentration, so that rather H_2O will condense onto the particles than H_2SO_4 (Hamill et al., 1997). Since the H_2SO_4 concentration is decreasing with altitude the H_2SO_4 condensation will be most probably be even less important for aerosol growth in the upper troposphere than in the lower troposphere. Furthermore, Kerminen and Kulmala (2002) have shown that for a nucleation burst coagulation is the more dominant process affecting the aerosol size distribution than condensation.*

p21968, l4: The use of the measurement data to initialize the model is not clear to me. Perhaps this could be clarified.

We use the measurements to initialize the model since we need information on the background aerosol distribution. Since there are now measurements available at other times and locations than the flights we use these measurements to initialize the model. However, to get an agreement between the measurements and the model simulation the requirement is that the background distribution found at the end of the simulation is the same as it was at the begin of the simulation (thus, in the simulation at the time of the flight the size distribution should be the same as the one measured). We included the following text in the manuscript to make this more clear: *Note, the initialization with the measured aerosol size distributions is done six days before the measurements actually were performed. However, for getting an agreement between model simulations and measurements an additional requirement besides the agreement of the modeled and measured nucleation mode particles is that at the end of the simulation also the background aerosol distribution is in agreement with the measured aerosol size distribution.*

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p21973, section 4.1.3: Overall one of my largest concerns comes from the use of simple backward trajectories - which, to my knowledge do not adequately characterize the convective processes of the air parcel. The statement is made on p21973, l15: "Further, the paths of the air mass trajectories... have the same origin". I'm not convinced about this as no information is provided regarding the vertical motion of the trajectories.

Here, we refer to the trajectory path to make clear that the nucleation event cannot be simply explained by the origin of the air mass. The uplifting processes are characterized by the parameters that are calculated along the trajectory. For example, we use the pressure to estimate over which altitude range the air has been lifted. This is described in the paragraph beginning on line 26 on page 21973. Thus, using the pressure and the corresponding altitude the airmass was lifted we can conclude that there are differences in the uplifting of the air masses. In the revised version we additionally included a figure showing the pressure along the trajectories to make this more clear. The information of pressure along the trajectory for characterizing uplifting of air masses and relating this uplifting to aerosol formation has also been used in several other studies (e.g. de Reus et al. 1999, Benson et al., 2008). Further, several other parameters than pressure and temperature were available along the trajectories used in this study that can be used to characterize convective processes, as e.g. dry and wet stability, cloud fraction, cooling rate and updraft velocity. Especially, cooling rate, dry and stability and vertical velocity were applied in our analysis (see section 3.1 and 4.1.1).

Summary: Overall the paper presents an analysis of in flight data using a box model to investigate the process of vertical lifting on particle formation. It is not written in a compelling manner, nor in such a way that makes clear the goal (or conclusion) of the study. For instance, the conclusion has a sentence: "Due to the fact that the nucleation event occurred later than on the other days (thus closer to the time of the measurements) not all newly formed particles were removed due to coagulation until the measurements were preformed.", which essentially – as I read it – states that the two other days may have had similar results if the measurements

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were made at a different time. So, it thus hard to take anything away regarding the different 'cases' employed in the analysis.

The goal of our study is to investigate the influence of vertical motion on particle formation. Our study is performed in the polar region where particle formation is rarely observed. Thus, a event of particle formation in this region is definitely of interest. The formation of H_2SO_4/H_2O particles need either sufficiently high precursor gas concentrations (e.g. H_2SO_4), sufficiently low temperatures or sufficiently high relative humidities. To get these conditions dynamical processes like uplifting of air masses are necessary. For the mid and lower latitudes this will happen due to convection or synoptic scale uplifting quite frequently. However, the polar regions are quite stable stratified and uplifting or convection occurs with a much lower frequency than in other latitude regions.

Our main results are that we can divide our simulations into three cases that reflect different nucleation behavior due to the conditions under which nucleation occurred. For case 1 the driving force was the temperature alone while for case 2 or case 3 vertical motion were involved in the particle formation. Our simulations show that particle formation occurs along all trajectories, however, only on 24 May nucleation mode particles were observed. We can explain why the nucleation mode particles were observed on 24 May but not on the other days by the conditions under which nucleation occurred. Here, we found that on one day the nucleation was caused by a fast updraft and on the other day due to a slow updraft. This difference in the strength of the updraft caused that on one day a higher nucleation rate was reached than on the other day which resulted in a fast removal of all newly formed particles by coagulation by the time of the measurements. On the other day the updraft caused a lower nucleation rate and the particles were not removed that fast by coagulation. The connection between particle formation and the strength of the updraft is a quite new result. Except in our study such a behavior has up to know only been described by Benson et al. (2008). Further, our study emphasizes in connection with other studies

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the importance of vertical motion on particle formation and our study is the first one showing this for the polar regions.

Concerning our statement in the conclusion, if one would have measured at a different time and location they would have probably measured during both days nucleation mode particles. However, nucleation is occurring at different places at different times with different strength. However, in all cases the newly formed particles grow quite fast to larger sizes by coagulation. Thus, to catch a nucleation event, especially in the upper free troposphere, while measuring is quite difficult. In the entire campaign period only one nucleation event was measured. We hope that the improvements we have made in the introduction and in other parts of the paper make the importance of particle formation in the polar regions more clear.

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