

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 H₂SO₄/H₂O” by F. Khosrawi et al.

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We thank reviewer 1 for the constructive, helpful criticism. We followed the suggestions of reviewer 1 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.

Major concerns:

The result of the analysis is that the nucleation depends on the type of vertical upwards motion,

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slow or fast. But such a dependency on the motion of the air mass could be expected, or? What I am missing is that the authors convince me as a reader, why such a study is of relevance to atmospheric science.

The binary homogeneous nucleation is strongly dependent on temperature, relative humidity and relative acidity (or partial pressure of H_2SO_4). Due to the cold temperatures binary homogeneous nucleation is mainly occurring in the upper tropical troposphere (Brock et al., 1995). In other altitude regions and especially at higher latitudes particle formation by binary homogeneous nucleation is relatively small (Raes et al., 1995). Nevertheless, on some occasions particle formation has been observed in these regions under temperature and relative humidity conditions that are otherwise unfavorable for nucleation. In the upper troposphere particle formation is caused by dynamical processes like uplifting of air masses due to convection. In the high latitudes air masses air generally stable stratified and convection is not that frequently occurring. We included a sentence concerning the dependence of the binary homogeneous nucleation on T, RH and sulfuric acid mixing ratio sentence as well as a sentence where we refer to Raes et al. (1995) in the introduction to emphasize that particle formation by binary homogeneous nucleation is not that frequently observed in the polar regions: *The binary homogeneous nucleation of H_2SO_4 and H_2O is mainly dependent on temperature, relative humidity and relative acidity (or partial pressure of H_2SO_4). [...] At other altitude and latitudes regions particle formation by binary homogeneous nucleation is relatively small (Raes et al., 1995).* Further, the following text has been included: *Previous studies investigating particle formation due to vertical motion were performed for the midlatitude and tropical regions. Thus, our study is the first one investigating this relationship in the polar regions.* In the conclusion we following sentence has been added: *Especially, here for the first time it has been shown that vertical motion also are important in the polar regions.*

The model indicates particle formation for all investigated trajectories, which makes me doubt if it is the appropriate tool to investigate why nucleation mode particles were found on

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one day, but not on the other two. Furthermore, differences in air mass history is not visible, because there is no trace gas information available, and the shown trajectories do not include altitude information. The authors consider the three days (within a period of five days) to be comparable concerning chemical composition and atmospheric conditions, only upward motion seems to be different. Again, the authors might be correct, but please convince the reader that not different air mass composition instead of the vertical motion is the main reason for the observed differences in particle number density.

Our box model simulation show indeed for all trajectories particle formation. This states that the condition during that time in that area were particularly favorable for particle formation. We can trust our model concerning this results since we did not get this behavior for other case studies we did, e.g. we applied the model in a study simulating a nucleation event observed in the midlatitudes during the STREAM 1998 campaign (Khosrawi and Konopka, 2003). Performing box model simulations for this case study we did not encounter such favorable conditions. Further, pressure and temperature along the trajectories give information on the upwards motion of the air mass (see discussion in section 4.1.3. To make this more clear we included a figure showing the pressure along the trajectories. Concerning the chemical composition, indeed there are now measurements of trace gases available. However, simulations with FLEXPART were performed for the ASTAR 2004 campaign wich can give a hint on the SO₂ source contribution. The FLEXPART simulations have been included in our discussion (see our answer to comment on p21965, l4.)

Specific comments:

p. 21961, Introduction, first paragraph: the link between the climate issue and particle formation is missing, why is particle formation important, and why in the Arctic?

See our answer above to the major comments. We are sure that the improvements we made on the introduction make now the importance of particle formation in the Arctic more clear.

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p. 21961, Introduction, second paragraph: the authors present the different particle formation processes discussed in the literature, but this presentation does not include the recent papers to this topic, e.g., Laaksonen et al., ACP 8, 7255-7264, 2008. Furthermore, they should argue more clearly, why it is valid to use the binary homogeneous nucleation for modeling particle formation in the free troposphere (cf. e.g., introduction in Laaksonen et al, 2008, Weber et al.,1999).

We included the more recent references of Laaksonen et al. (2008) and Curtius et al. (2006) for the ion-induced nucleation as well as Yu (2006) and Benson et al. (2009) for the ternary nucleation. Further, we argue now more clearly why it is valid to use binary nucleation for modeling particle formation in the free troposphere. We included the following sentence in the introduction: *The observations of particle formation above 4 km can be well explained by binary homogeneous nucleation while at altitudes below 4 km other nucleation processes seem to be involved in the particle formation (Weber et al., 1999).*

p. 21962 l. 1, the Hermann et al., 2003 reference is poorly cited. These authors did not measure the particle size distribution, but only integral particle number concentrations in two size ranges, or? Furthermore, they found that atmospheric dynamics (in this case deep convective clouds) also play an essential role in the tropics, not only at mid-latitudes.

We are sorry that we did not cite the work of Hermann et al. (2003) correctly. We changed the sentence as follows: *Hermann et al. (2003) measured particle number concentrations over three years on a German commercial aircraft in the latitude range from 5° to 50° and found that photochemistry and solar radiation (tropics) as well as atmospheric dynamics (midlatitudes, tropics) were the driving forces for aerosol formation in the tropopause region.*

p. 21962 l. 6, particle formation due to stratosphere-troposphere exchange was also found by Zahn, A. et al., J. Geophys. Res. 105, 1527-1535, 2000, please add this reference.

We would like to thank reviewer 1 for pointing this out and added the reference of Zahn et al. (2000).

p. 21964 l. 3 . . . , please state exactly which instruments you were using, i.e. the manufacturer and the model types.

We have added the information on manufacturer and model types of the instruments that have been used in our analysis.

p. 21964 l. 8, the authors emphasize the uncertainties associated with counting statistics, but what about the uncertainties caused by the particle sampling efficiency or the inlet system, pressure dependent counting efficiencies, etc. Please add this important information.

The performance of the whole inlet-instrumentation package was tested during flyby and intercomparison with measurements carried out at ground based station at Ny Ålesund were performed. There is a very good agreement between these measurements for both DMPS as well as OPC size ranges. The CPC data have not been corrected for a pressure dependence of sampling efficiency. According to our own laboratory measurements this effect is for the type of Butanol-based CPC we use down to a pressure level of 250 hPa small (smaller than 5 %). The aircraft inlet was designed for this type of aircraft (and its typical air speeds) based on common practical and theoretical considerations in inlet design. Particles of a size of up to roughly 1 micron (and smaller) should pass through the inlet with no significant loss. In particular in the altitude regions we focus on, no coarse mode aerosol particles larger 1 μm are present. Thus, inlet effect do not affect the CPC and other measurements.

p. 21964 l. 17, the stated uncertainty of the FSSP-300 refers to what, sizing or concentration? Other investigators give higher numbers, e.g., <http://www.eol.ucar.edu/raf/Bulletins/B24/fssp300.html>.

We agree with reviewer 1 that the uncertainty in the measurement of the FSSP-300 can be higher and can concern both concentration and size. The error in concentration depends on counting statistics which in turn depends on the size and integration time. Further, the error in size depends on calibration and on knowing the refractive index. In this study the FSSP is only used to define the in-cloud sequences during the flights.

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In this case, the concentration and sizing error is not relevant. Further, the probe was calibrated before and after the campaign. The following sentence has been included to clarify this: *The FSSP-300 is used in this study only to define the in-cloud sequences during the flights (using a threshold criterion for the number concentration of particles larger than $\approx 3 \mu\text{m}$).*

p. 21964 l. 21, in terms of the above statement, I don't believe the 5% accuracy of the CPSA to be true, it should be higher.

The CPSA is a battery of CPCs, thus the accuracy of each CPC-channel is the same like for any other CPC. 5% is a very reasonable value for number concentrations determined by CPCs. The error represents uncertainties in sampling efficiency (instrument-to-instrument variation, and pressure dependence), flow calibration and counting statistics. During a campaign we specifically check the agreement between the 4 CPC-channels and adjust, if necessary, an efficiency factor according to comparison with the reference instrument. In conclusion, we support to give a typical statistical error for the CPC measurement of about 5%. However, there might be some systematic underestimation of particle concentrations due to diffusion losses of sub-10 nm particles (see elsewhere).

p. 21965 l. 1, the estimation about the particle losses is based on what? And if 35% of the 4 nm particles get lost in the sampling system (on average maybe 20% for the size range 4-10 nm) this number is large and the data should be corrected for using an average value, even without the knowledge of the exact size distribution.

The estimate on the particle loss is based on aerosol text book (Willeke and Baron) description on diffusional losses for laminar flow through a tube and assumptions on the range of possible particle sizes. How to correct the concentration data is a difficult problem if the size distribution between, in this case, 4 and 10 nm, is not known. It makes a huge difference if the main population of nucleation mode particles would sit closer to 10 nm or closer to 4 nm. We are therefore not in favor of the suggestion of the reviewer of applying some "average" correction factor. We just don't know it.

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Therefore it is more true to the situation to state the possible range of underestimation (according to our estimate) and allow the reader to draw his own conclusion of the magnitude of the effect. In terms of the analysis presented in this paper, we do not believe it is critical to apply a correction or not. However, to make the reader aware of this possible uncertainty in the measurements we included the following sentence into section 4.1.2: *It should be noted here, that there is possible underestimation of 10-35% due to diffusion losses in the measured number densities of nucleation mode particles (see section 2.1).*

p. 21965 l. 4, are there any trace gas measurements available, which might help interpreting the data?

Trace gas measurements would be indeed very helpful to interpret data. However, unfortunately there are no trace gas measurements available. The DO-228 aircraft is only a small aircraft with limited payload capacity and priorities were given to aerosol related instruments. However, simulations with the FLEXPART model were provided by Andreas Stohl. These simulations (20-d backwards) give a hint about the SO₂ source contribution during the three measurement days. On all three days the SO₂ source contribution was low. However, if one compares the three days with each other, on 24 May the contribution was highest. The following sentence has been included in section 4.1.3: *Further, simulations (20-d backwards) that were performed with the FLEXPART model (Stohl et al., 2005) for this campaign show that the SO₂ source contribution on all three days was low though the highest contribution of SO₂ was found on 24 May. Further, the sentence describing the differences in new particle formation on 24 May compared to the other two days has been changed as follows: Besides the fact that FLEXPART shows a higher SO₂ contribution on 24 May the nucleation was caused by a slow updraft.*

p. 21965 l. 11, please add a figure showing the flight tracks.
We added a figure showing the flight tracks.

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p. 21965 l. 11, it might be nitpicking, but personally I feel that the first figure mentioned in the text should also get the figure number “1”, and the order of figures should not be mixed.

We apologize for this mistake and corrected the order of the figures.

p. 21966 l. 2, I am not a specialist in modeling particle nucleation, but aren't there more recent, improved formulations for the homogeneous nucleation theory, e.g., F. Yu, J. Chem. Phys. 127, 054301, 2007?

Though there are more recent publications on binary homogeneous nucleation there are no major differences compared to the nucleation code by Kärcher et al. (1998) which we apply in our model. The Kärcher et al. (1998) nucleation model is based on a revised form of the classical binary homogeneous nucleation and considers the formation of gaseous sulfuric acid hydrates. Further, e.g. the model of Yu (2007) has only been compared to experimental data which were derived by laboratory measurements under boundary layer conditions. Thus there is no evidence that the Yu (2007) model gives better results in the upper/free troposphere than former models based on a revised nucleation theory.

p. 21966 l. 5, condensation of material leads to changes in particles size, which influences the particle coagulation rate. So, if you do not consider condensation, how larger is the uncertainty in your modeling due to this neglect?

Indeed, condensation leads to changes in particle size. However, condensation is a quite slow process so that in case of a nucleation burst as we encounter it in our simulations coagulation will be much faster and therefore the aerosol size distribution will be stronger affected by coagulation than by condensation. To make this more clear we changed the text as follows: *However, this should not affect our results since condensation is a very slow process. The atmospheric concentrations of H_2SO_4 are much lower than the concentrations of H_2O , so that rather H_2O will condense onto the particles than H_2SO_4 (Hamill et al., 1997). Further, since the concentration of H_2SO_4 is decreasing with*

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altitude we can assume that H_2SO_4 condensation is more important for aerosol growth in the lower troposphere than in the upper troposphere. Furthermore, Kerminen and Kulmala (2002) have shown that after a nucleation burst coagulation is the more dominant process affecting the aerosol size distribution than condensation.

p. 21966 l. 7, please provide a reference for the statement in the last sentence of this paragraph.

See our reply to the previous comment. Since there is no reference for the statement we changed the sentences so that it is clear now for the reader how we came to this conclusion.

p. 21966 l. 27, which “three” trajectories are meant? Either there are four (2x 24 May, 1x 22 and 26 May, respectively) or six, if there were trajectories for both altitudes for each of the three days.

There are 12 trajectories for each day of measurement. For each the three measurement days trajectories were calculated at 2 altitudes and 6 coordinates, thus all in all there are $3 \cdot 2 \cdot 6 = 36$ trajectories. The three trajectories we refer to are the three derived for 7000 m at the locations where the flights were performed. We changed the sentence as follows to clarify this: *The path of these three trajectories (22, 24 and 26 May, 7000 m) are shown in Fig. 2.*

p. 21967 l. 4, sorry, but I could not follow your counting, how did you come to 36 trajectories in total? Please explain in a way that the reader understands why you chose this number.

As described in the answer to the previous comment, trajectories are calculated for each of the three measurement days at 2 altitudes and 6 different coordinates, thus all in all there are $3 \cdot 2 \cdot 6 = 36$ trajectories. We changed the sentence as follows to make this more clear: *In total 36 back trajectories (3 measurement days \times 2 altitudes \times 6 coordinates) were calculated with TRAJKS.*

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p. 21967 l. 12, why do you need the stability parameters to know if vertical motion has occurred along the trajectory? What kind of additional information do they provide, which is not provided by the trajectory altitude or pressure?

The trajectory altitude or pressure gives only the information over which altitude or pressure range the air mass has been lifted. The stability parameters gives an information on the general dynamical state of the atmosphere, e.g. if the atmosphere was stable stratified or not. In our study we use dry and wet stability to investigate if vertical motion were involved in the particle formation as it is described in more detail in section 4.1.1. We changed the sentence as follows to make this more clear: *Dry and wet stability ($\gamma_d = \frac{d\Theta}{dp}$ and $\gamma_w = \frac{d\Theta_e}{dp}$) are used here as a proxy to investigate if vertical motion have occurred along the back trajectories and if vertical motion were involved in the formation of new particles.*

p. 21968 l. 5, did I understand it right that the box model calculations were initialized with measurements made six days later? If, yes, please provide some evidence that this approach is valid.

We assume that the background distribution is the same for the entire period considered here. To proof if we are right we only need to check that the distribution used for initialization is the same at the end of simulation, thus at the time of the flight when these measurements were performed. We included the following sentence to clarify this: *Note, the initialization with the measured aerosol distributions is done six days before the measurements were performed. However, for obtaining 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 measurements.*

p. 21968 l. 28, please indicate that the sulfuric acid concentrations are gas phase concentrations. The chosen values are high, according to your references typical for polluted air

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masses. Why did you chose these high values, the trajectories suggest rather clean air masses, or?

We changed the sentence so that it is clear that the used concentrations are gas phase concentration. The chosen values are representative for both clean (1 pptv) and polluted (40 and 80 pptv) air masses. Though the polluted values are rather high we still get the best agreement between measurements and simulations for a gas phase mixing ratio of 40 pptv. We changed the sentence as follows to make clear that we consider both polluted and unpolluted conditions. *The simulations were performed along the 36 trajectories for three different gas phase sulfuric acid mixing ratios $\mu_{H_2SO_4} = 1, 40$ and 80 pptv (1 pptv = 2.67×10^7 molecules cm^{-3} at $0^\circ C$ and 1013 hPa), where a mixing ratio of $\mu_{H_2SO_4} = 1$ pptv is representative for clean air and mixing ratios of $\mu_{H_2SO_4} = 40$ and 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. We would not say that the trajectories suggest rather clean air masses since the air were transported over inhabited areas in Scandinavia. However, to get clear answer to the question if the air masses were polluted or not we would need additional measurements, like e.g. CO which are unfortunately not available.*

p. 21969 l. 9, in line 6 you used the order 40 pptv, 80 pptv, and 1 pptv for the sulfuric acid mixing ratio, hence in line 9 one would assume that the nucleation time steps 17 h, 16 h, and 2 h are for the same order of sulfuric acid mixing ratios, which is not the case, or?

The three time steps are an example for that nucleation occurs at three different time steps. These number refer to the three sulfuric acid mixing ratios used in this analysis. We changed the sentence as follows to avoid misunderstandings: (3) *Nucleation occurred for all sulfuric acid mixing ratios used at different time steps e.g. for $\mu_{H_2SO_4} = 1$ pptv at $t = 17$ h, for $\mu_{H_2SO_4} = 40$ pptv at $t = 16$ h and for $\mu_{H_2SO_4} = 80$ pptv $t = 2$ h (10 occurrences).*

p. 21969 l. 15, I do not fully understand the differentiation between temperature and vertical motion as trigger for new particle formation. Vertical motion (disregarding the

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reduction of particle surface area due to scavenging processes inside clouds and different photochemistry at higher altitudes) is equivalent to temperature change, so why make a difference? Which process, besides altitude change should be responsible for the temperature change? Please explain.

The binary homogeneous nucleation is strongly dependent on temperature, relative humidity and the sulfuric acid concentration. If temperatures are quite low (due to synoptical processes) then binary homogeneous nucleation can occur without any dynamical effects. However, if the synoptic temperatures are too warm, dynamical processes as vertical motion cause can an uplifting and thus a decrease of temperatures that in turn leads to particle formation.

p. 21969 l. 26, as figures 4 to 6 show, the model indicates particle formation along all 36 trajectories. Is this realistic? I would assume that there are trajectories which are not accompanied by particle nucleation. Otherwise, it would mean that in the region under investigation particle formation is always taking place.

Indeed, our simulation show particle formation along all 36 trajectories. This states that the condition during that time in that area were particularly favorable for particle formation. We can trust our model concerning this results since we did not get this behavior for other case studies we did, e. g. we applied the model in a study simulating a nucleation event observed in the midlatitudes during the STREAM 1998 campaign (Khosrawi and Konopka, 2003) where we did not had such favorable conditions.

p. 21969 l. 28, the gamma values are what? Mean values along the trajectory, stability at the time when the maximum particle concentration occurred, maximum stability along the trajectory, ...?

The gamma values were calculated with TRAJKS along the trajectories. Thus, at each time step we have one gamma value. The values used for the figures are the ones which correspond to n (here we took the maximum number concentration of freshly nucleated particles encountered during the simulation). We included a “corresponding”

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before γ_d to make this more clear.

p. 21970 the two paragraphs comparing the three cases in Fig. 4-6: I read the paragraphs two times, but I did not get the message (sorry, maybe I am too tired). What do you want to show with these figures? How is your “uniformity” defined (for the 1 pptv data in Fig. 4 the number densities vary over nearly three orders of magnitude)? And why does the “uniformity agree with the behavior found for case 1”? What “relationship” are you talking about?

In figures 4-6 we consider atmospheric stability together with the maximum number concentration along the trajectory (thus at the time step where the nucleation event occurred) for the three different cases we described earlier to investigate if vertical motion were involved in the particle formation. In these two paragraphs the figures and thus the relationship between n and γ_d and γ_w , respectively, is described. Further, we intend to relate the behavior between n and γ_d and γ_w found here to the behavior we found for the three cases from the model simulations. We have no definition for “uniformity”, we use “uniformity” to describe what we see. It is true that there is a certain spread concerning the particle number, however, we use the term “uniformity” to express that all points of n lie almost on a line independent on which dry or wet stability was prevailing in the atmosphere. Thus, the important thing here is, that obviously the dynamic state of the atmosphere did not contribute to the particle formation. This in turn agrees with our definition of case 1 where we found that the temperature alone was the driving force for particle formation. To make this more clear we included the following two sentences: *Thus, it seems that the stability of the atmosphere did not had an effect on the particle formation. Here, except for one occurrence particle formation occurred during a stable stratification of the atmosphere thus indicating that vertical motion were not involved in the particle formation.* Further, the subsequent sentence has been changed as follows: *This uniformity agrees with the behavior we found for case 1 from the box model simulations showing that nucleation occurs always at the same time step and that temperature only was the driving force.*

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p. 21970 l. 13, is it justified to declare a “slight decrease” in the number density based on only two data points. I don't think so.

We agree, with only two points it is hard to judge a certain “real” relationship between these parameters. We included the following disclaimer: *However, since we only have two data points for each sulfuric acid mixing ratio it is difficult to say if this relation is arbitrarily.*

p. 21970 l. 26, if you discuss the relation between the number density and the updraft velocity displayed in Fig. 7 in only one sentence, is it necessary to show this figure?

This figure is necessary for our study since it shows that the same relation which we found between number density and stability is also found between number density and vertical velocity. Thus, this figure corroborates our previous findings, that for case 1 the the temperature only was the driving force for particle formation while for case 2 and 3 vertical motion were involved in the particle formation. However, we agree that our discussion of this figure is with only one sentence somewhat short and included the following text: *Case 1 is uniformly distributed with regard to updraft velocity while for case 2 a uniform distribution is only found at 6000 m for a sulfuric acid mixing ratio of 1 pptv. For sulfuric acid mixing ratios of 40 and 80 pptv an increase with a subsequent decrease is found for n . Case 2 shows at 7000 m a decrease for 1 pptv and increase of n with w for 40 and 80 pptv. However, since we only have two data points for each sulfuric acid ratio it is difficult to say if this behavior is only arbitrarily. Case 3 is at both altitudes extending over a larger range of vertical velocities (from -2 to $+6$ m/s) than the other two cases showing that for this case the influence of vertical motion was strongest.*

p. 21971 l. 16, the 24 May is not “dominated” by case 1, because case 2 has nearly the same number of occurrence. Please change this statement.

Indeed, the 24 May is only dominated by case 1 at 7000 m, but by case 2 at 6000 m. We changed the sentence as follows: *Considering each day, the 26 May was dominated by case 1 in both altitudes while the 22 May was dominated to the same amount by case 3 at*

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6000 m and by case 1 and case 3 at 7000 m. The 24 May was dominated by case 2 at 6000 m and by case 1 at 7000 m.

p. 21972 l. 5, which were the criteria by which the trajectories were chosen for Fig. 8, i.e., why there are some trajectories included and others not?

Figure 8 displays all trajectories while Figure 9 only displays the ones which were started near the location where the flight was performed. The purpose of Figure 9 is to get a better comparison between measurements and simulations. This is stated in the figure captions as well as in the manuscript where the figures are described (p. 21972 at l. 5 and p. 21972 at l. 12, respectively).

p. 21972 l. 5, what do you mean by "... some trajectories ... lie also within this area"? Do you refer to a geographical area?

We refer to the gray shaded area mentioned two sentences earlier. We changed the sentence as follows: *The 24 May trajectory (7000 m) as well as some trajectories of the other days (22 May and 26 May) and other starting points in the grid around the measurements lie also within the gray shaded area and thus in the range of nucleation mode particles measured on 24 May.*

p. 21973 l. 1, if on 26 May 1 pptv sulfuric acid leads to a too low nucleation mode particle number density and 40 pptv as well as 80 pptv lead to too strong nucleation events (and thereby again to a low nucleation mode particle number density at the measurement time), a sulfuric acid mixing ratio "somewhat lower than 40 ppt" should lead to a nucleation mode particle number density which should have been observable, or?

We agree that a sulfuric acid mixing ratio "somewhat lower than 40 pptv would possible lead to a observable nucleation mode particle number. However, a sulfuric acid mixing ratio higher than 40 pptv can be excluded since with these high values we would not receive the background distribution at the end of the simulation which we used for initialization and (thus the measurements). Thus, the sulfuric acid mixing ratio

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must have been definitely below 40 pptv. We skipped “somewhat” and changed the sentence as follows: *Since on that day no nucleation mode particles were observed we conclude that the H₂SO₄ mixing ratio on that day must have been definitely lower than 40 pptv.*

Technical Comments:

again, it seems to me that the manuscript was written hastily, because there are several simple language errors, e.g., p. 21960, l. 6, “vertical motion are the reason”, p. 21963 l. 17, “aircrafts”, p. 21967, l. 1, “coordinates which represents”. Please check the whole document again.

We checked the entire document again and removed all language errors.

p. 21961 l. 10, I never heard the term “nucleation . . . conversion”, and I believe it to be wrong. It should be either “particle nucleation”, “nucleation process”, or “gas-to-particle conversion”, but not a mixture of these two terms (which are not the same, by the way).

Yes, this is true. The closing bracket was at the wrong place. The sentence correctly reads: *The nucleation (gas-to-particle conversion) is a source for new particles in the atmosphere.*

p. 21962 l. 8, please insert “particle” before “properties”.

We included “of aerosol particle” after “properties”.

p. 21964 l. 7, “CPC” must be “OPC”.

This has been corrected.

p. 21967 l. 1, please use a consistent nomenclature for the geographical coordinates, i.e., either always separated by a slash or never.

Coordinates are given with a slash. This is now done consistently in the entire manuscript.

p. 21969 l. 13, the trajectory start should be at 19.0°E and 78.7°N, or is it a new tra-

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jectory? Please see also Fig. 3.

This is correct, the decimals were missing. They have been added now.

p. 21969 l. 18, please remove “previously” because 1999 (and there are other references as well) is ten years ago.

“Previously” should not be mixed up with “recently”. Recently we cannot apply for studies done several years ago, however, previously means only “before”, thus we can apply “previously” also for studies which have been done ten years ago.

p. 21969 l. 20, the reference should be to Table 2, not Table 1.

This has been corrected.

p. 21971 l. 3, please remove the “and” before “to case 3”.

The “and” has been removed.

Tab 1: “uK/Pa” should be “ μ K/Pa”, or? Same in Tab. 3.

We changed uK into μ K

Tab 3: in the table “Altitude” without “d”.

This has been corrected.

Fig. 1: the trajectories do not start at 10° or 11°E, they all start at 22.5°E! The coordinates given in the Figure caption are correct (see also chapter 3.1).

Fig. 1: please include trajectory altitude information in the figure, e.g., by using different shades of color.

We included an additional figure showing the pressure along the trajectory. The pressure range at which the air masses were lifted and the resulting temperature decrease is discussed in section 4.1.3

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Fig. 2: the chosen representation of data wastes 20% of the figure, because they are not used, why do you not expand the x-axis scale, which would make the structures more visible?
The figure has been changed as suggested.

Fig. 2: there are gaps in the N4 time series, in particular for the flight altitude 7000 m, please explain and add this information to the text. What are the implications for your simulation?

The gaps are there because when the aircraft was flying curves the data was routinely excluded. However, this does not mean that data is not useful, it is just for precaution. Looking on the raw data, no essential data with nucleation mode particles present were missed. Thus, there are no implications due to this gaps on our analysis. We included the following sentence in the figure caption: *Note: The gaps in the figure are due to elimination of data when the aircraft was turning (high bank angle).*

Fig. 2, legend: “begin” should be “beginning”, or?
This has been corrected.

Fig. 2, legend: the reference to the simulation is a little bit confusing, because one might think that the data were simulated.
This was a mistake. We replaced “simulation” by “measurement”.

Fig. 4: symbols are hard to see, please use solid symbols. Same for Fig. 5, 6, and 7.

Unfortunately, we cannot use solid symbols since then the symbols which lie over each other will not be as good visible as they are now. However, to make the symbols more visible we increased the symbol size.

Fig. 4, x-axis: “uK” should be “μK”, or? Same for Fig. 5 and 6.

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This has been corrected.

Fig. 5, upper, right graph: there seems to be one green and one blue symbol missing, or do they fall together with two other data points? If yes, please notify this.

No, these points were solely beyond the plot range. We extended the x-axis to include these points in the figure.

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

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