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Atmospheric Chemistry and Physics Discussions

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7, S8935–S8941, 2008

Interactive Comment

Interactive comment on “When does new particle formation not occur in the upper troposphere?” *by* D. R. Benson et al.

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We would like to thank the reviewer for their helpful comments that were incorporated into our revised manuscript. Any references to figures or tables are refering to the revised manuscript.

ACPD-2007-0383 "The Effects of Convection on New Particle Formation in the Free Troposphere: Case Studies" By Benson et al.

Response to the Anonymous Referee 2’s Comments by S.-H. Lee

Ref2: *This manuscript describes tropospheric and stratospheric ultrafine aerosol data measured aboard the GV aircraft platform during the Progressive Science Missions in December 2005. The authors contend that new particle formation (NPF) events, or lack*

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thereof, are strongly related to air mass history, specifically recent convection and the level of preexisting aerosol surface area. Both topics have been addressed in previous publications that are cited in the manuscript, and the effect of particle surface area in particular has been heavily discussed from a theoretical perspective. The current work is a systematic study using a suite of field measurements. The field data is unfortunately hampered by lack of measurements of trace gases, aerosol precursors, and oxidants. The authors rely solely on NOAA HYSPLIT back trajectories to provide information about air mass history.

In general the arguments in the manuscript do not strongly support the conclusions. To properly investigate surface area, convection or ascent, and origin of air on NPF potential, the case studies and mission averages must be selected with greater care. The manuscript does not provide compelling new evidence of NPF trends, and it does not warrant publication unless the authors can provide more definitive examples from their dataset as described in the points below.

Re: We thank the reviewer for the constructive and helpful comments that improved our manuscript significantly. Based on these comments, we made in-depth reanalysis and reconstructed the manuscript, by carefully re-selecting case study events from similar altitude regions and by including an absolute non-NPF event (Figures 2-7). We have also included more detailed analysis on surface area (Table 1 and Figure 1a). In this study, we show distinct examples of strong-, weak-, and non- new particle formation events from the measured aerosol size distributions to see how air mass history affects NPF. Even with the lack of chemical information, these atmospheric observations will provide an important dataset to the nucleation modelers to test and improve their nucleation theories.

Our reanalysis shows that for weak- or non-event cases, the air masses did not experience convection; for "strong" events, air masses often had convection but there were also event cases where convection did not occur. As for surface areas, the surface area ranges are in fact the same for the event and non-event cases (Figure 1a) and there is

ACPD

7, S8935–S8941, 2008

Interactive Comment

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no strong anti-correlation, although the median values are higher for the non-NPF than the NPF cases (Table 1). This is because we sampled at a wide range of altitudes (up to 14 km) and latitudes (18 °N to 62 °N) and 95% of the samples are NPF cases and also because surface areas are very low in this region in general.

Ref2: *1) In the abstract (lines 4-5) the authors state that case studies show the effect of surface area on NPF. However, all of the cases presented involve low surface area conditions, and all are described as NPF events, either strong or weak. No contrasting cases are given, and within the presented cases surface area does not anti-correlate with ultrafine concentrations. The only data that support the conclusions relating surface area and NPF are given in Table 1. Although the average surface areas support the overall conclusion, they have very large standard deviations (500%) that encompass zero. Clearly, NPF was observed for some high surface area conditions, and conversely, some instances of very clean air showed no NPF. If the authors were to further divide the NPF and non-NPF formation cases, there may be clearer trends in altitude (eg, stratosphere vs troposphere), origin of air, or other parameters. Furthermore, the authors should consider a case example clearly illustrating the effect of surface area. In its current form, the surface area conclusion is not strongly supported by the data presented. Additionally, Table 1 shows that the non-NPF cases are observed at an average of 20K higher temperature, which has an enormous impact on the potential for nucleation to occur. When viewed as a mission average, it is not surprising that temperature is the dominant parameter determining NPF.*

Re: We agree and the extensive revision is made on selecting case study events (Figures 2-7) and on the discussion on surface area.

(1) For the surface area discussion, please see the above summary response. Surface area is discussed in detail in Section 3.1 (lines 111-129) and Section 4 (lines 245-255).

(2) We have also included here new Figure 1 for N4-9 vs. surface area, temperature and RHI. And this is discussed in detail in Section 3.1 (lines 111-129).

(3) Table 1 shows that there is indeed some altitude dependence of N4-9. Lines 125-129: "The higher median surface area for non-NPF is probably related to the fact that most of the non-NPF events were measured in the lower altitudes (Figure 1b and 1c and Table 1). For example, for the non-event samples, the median temperature was $\sim$ 244 K, higher than that for NPF cases ($\sim$ 228 K) (Table 1)". Surface area is usually associated with altitudes (Young et al., 2007) Also, as mentioned in lines 290-295, "For these specific case studies, however, the altitude rather seems to be a dominating factor in determining the strength of the NPF event. Both strong events had a median altitude of less than 2.5 km during the previous five days and had minimum altitudes very close to the ground level (< 1 km), whereas for the weak events the median altitudes were both above 6 km and the air never fell below 2 km (Table 2). And, this may again point to the significance of convective events in determining the extent of NPF."

Ref2: *2) The principal differences between the strong and weak nucleation events for Cases I and II appear to be the sampling altitude. Observations of both weak NPF appear to be within the stratosphere or very near the tropopause, whereas the strong NPF regions were clearly in the free troposphere. Back trajectories for the weak cases appear to be stratospheric. This presentation is somewhat convoluted. Lower stratospheric air may very well have lower precursor concentrations than the free troposphere (although Young et al show that stratospheric/tropospheric mixing may still lead to strong NPF). There is also likely an overall altitude trend in NPF within the authors' dataset. However, the effect of convection cannot be unambiguously determined by comparing cases from the stratosphere and troposphere. A better comparison would be cases at similar altitudes with lofted air and air that has resided exclusively in the free troposphere for several days without strong ascent. Is there always NPF? Does HYSPLIT always show convection at those altitudes? When this NPF-convection relationship is violated, what is the cause? The conclusions that are drawn here, that ascent of precursor gases strongly influences NPF, is not clearly demonstrated by Cases I II.*

ACPD

7, S8935–S8941, 2008

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Interactive Discussion

Discussion Paper

Re: We agree that one cannot compare the stratosphere and troposphere cases to each other. Based on the reviewer's comments and to support our non-/weak-NPF and non-convection conclusion, we have re-selected case studies of non-, weak-, and strong events all at similar altitudes (the upper troposphere region) (Figures 2-7). 95% of samples satisfied NPF criteria (Table 1), likely because of low temperatures and low surface areas in this region. For weak- or non-event cases, the air masses did not experience convection; for "strong" events, air masses often had convection but there were also event cases where convection did not occur.

Ref2: *3) Case III seems to contradict the hypothesis that ascent of air brings enhanced precursor concentrations that lead to NPF. Air originated from a similar geographical location in the lower troposphere for IIIa and IIIb, and surface area was low for both cases. Only IIIa showed significant NPF. The only observed difference is that air from the strong event ascends quickly, apparently in a convective system. This may provide a case for fast convective lofting versus slow ascent, but both cases are defined as convective in the manuscript. The authors do not offer a convincing explanation for Case III given the available data, and overall this Case does not support the manuscript's conclusions regarding convection.*

Re: We defined convection from the rate of uplifting (Section 3.1 lines 130 - 139): "In the present study, convection is defined based on the NOAA HYSPLIT backward trajectory outputs (e.g., air mass altitude dependence with time) (Draxler and Rolph, 2003). Convection is referred to as the cases in which the air mass was uplifted from a lower altitude, usually less than 2 km above ground level, to higher altitudes at an uplift rate greater than 3 km per day and the air mass was exposed to these low altitude source regions for at least 2 days before the vertical motion. On the other hand, if the rate of uplift was less than 3 km per day or if the air mass spent less than two days at an altitude of 2 km or less, we considered such a case as a non-convection event." We have reselected case studies (Figures 2-7) to see the convection effects on NPF.

Ref2: *4) The question posed in the manuscript title is never answered.*

ACPD

7, S8935–S8941, 2008

Re: We have changed the title to "The effects of convection of NPF in the free troposphere: Case studies". Our reanalysis shows that usually non-/weak-NPF events usually did not experience convection.

Ref2: *5) Minor points: The authors should consider photochemical lifetimes to produce precursor gases upon ascent. How does this affect their analysis using the HYSPLIT trajectories?*

Re: We agree - SO2 has a lifetime of approximately two weeks. But as shown in atmospheric observations, SO2 shows clear altitude dependence in the free troposphere (Thornton et al., 1999) and in this case, convection and air mass history can be very important (Lines 264-272). Since aerosol precursor measurements are very rare for aircraft studies, trajectory calculations can be particularly useful and with such help, one still can provide useful information on air mass history and the associated aerosol precursor source information indirectly.

Ref2: *Describe how surface areas were determined. Were fine mode aerosol included?*

Re: Surface areas are calculated from FCAS measurements only (> 90 nm) and fine mode aerosols are not included. Our previous studies have shown that the surface area calculated from FCAS alone is similar to that calculated from NMASS (4-100 nm) and FCAS measurements together (Lee et al., 2003).

Ref2: *The data plotted in Fig 10 should be labeled or filtered by altitude or potential temperature. The aircraft apparently changed altitudes during both the Dec19 events, changing ambient temperatures by 10°. This represents a significant change in nucleation potential, and the fine particle concentration has a range of x10. Are Fig 8b and 9b average size distributions over the entire time period?*

Re: A new Figure 8 is now provided and filtered by temperature (thus altitude). We have also revised all figures (Figures 2-7) to show the periods corresponding to the

ACPD

7, S8935–S8941, 2008

case study region.

Ref2: *Clarify statements about air mixing: pp 14215, lines 25-29.*

Re: On air mixing (line 188-192): "This also implies that air mixing can take place during abrupt uplifting. It has been shown that the nucleation rate can be increased one order of magnitude with a temperature decrease of 2-3 K (Nilsson et al., 2000), and therefore it is also possible that such air mixing also contributed to strong NPF events." Furthermore, "Abrupt air mixing can also take place during strong convection. As shown in previous theoretical predictions (Nilsson et al., 2000), because nucleation is a non-linear process, when two air masses mix with each other with different RHI, temperatures, and aerosol precursors, nucleation rates can be much higher than without mixing" (lines 272-276).

Ref2: *In Figures 2c and 5c the HYSPLIT 0% RH line does not appear correct since HYSPLIT indicated rainfall.*

Re: New case study events are provided.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 14209, 2007.

ACPD

7, S8935–S8941, 2008

Interactive Comment