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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

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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 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.

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.

2) The principal differences between the strong on 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.

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.

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

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?

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

The data plotted in Fig 10 should be labeled or filtered by altitude or potential temperature.

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Interactive Comment

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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?

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

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

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

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