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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.

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This work poses an interesting and important question. Identifying the conditions where nucleation is absent should help to improve the understanding of the complex unresolved question of the mechanisms of new particle formation in the earths atmosphere. This work presents some interesting observations of size resolved nanometer sized particles in the free troposphere. Unfortunately the particle observations are only weakly constrained by observations of other relevant parameters and the analysis and discussion are qualitative and inconclusive. This work may be publishable in ACP following extensive reanalysis and revision. Specific comments follow.

The title doesn't seem appropriate for this work, for two reasons. (1) Based on the criteria presented here for the classification of "new particle events" (N4-9>1 cm-3,

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N4-9>0.15N4-2000, N4-6>N6-9), a "non-event" does not necessarily correspond to the absence of nucleation. (2) The non-events are not the focus of this manuscript. There are no plots of non-event data, and most of the discussion concerns weak and strong events.

The present work and others show that nucleation is very common in the UT. What fraction of the observations has 4-9 nm particle concentrations above the detection limit? In the end, it seems that very few observations will show absolutely no evidence of nucleation. This is an intriguing possibility considering that the sub 4 nm particles are not even detected. The value of the nucleation event criteria is not clear.

The nucleation criteria used in this work require that the 4-9 nm particles are at least 15% of all (4-2000nm) particles for the observation to be classified as a nucleation event. Does this discriminate against data that show nucleation at high surface areas (a few small new particles in the presence of many large particles), and bias the conclusions regarding the influence of surface area on nucleation?

p. 14210 Line 13: "the events were closely associated with convection" is too strong a statement considering the sparse data and qualitative analysis. I am not convinced that this work has established a direct link between convection and nucleation. See additional discussion below.

p. 14210 Line 14: There is not a statistically significant difference between the average surface areas of the events and non-events. This data should be analyzed graphically as e.g. N4-9 versus surface area, to examine possible correlations. See additional comments below.

p. 14210 Line 17: "where precursor concentrations are relatively low" implies that we know the identity of the aerosol precursors and their concentrations, which is not the case. Better wording would be "expected precursors" and state the identity of the expected precursors. Same comment for text in the conclusions.

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p. 14210 Line 20 abstract: What is the basis of the comment "nucleation is thermodynamically favorable"? What is the significance of 250 K?

p. 14211 Line 3: How are nucleation events classified by other investigators? Do they use similar criteria as in the present work? This is important for comparison of the occurrence statistics.

p. 14211 Line 8: km-3=cm-3

p. 14211 line 20: Lee et al (2003) = Lee et al. (2004)?

p. 14212 line 17: "nighttime studies in this area are rare". Are there other studies, and what was observed?

p. 14214 Line 2 and table one: The average values of the surface area are different for non-events and events, but the standard deviations encompass both averages. This is not "a clear indication that low surface area is necessary for new particle formation" (line 4 p. 14214). The authors should plot number of 4-9 nm particles (or some other measure of recent nucleation) as a function of the surface area to examine possible correlations. Evaluate the statistical significance of the slopes and intercepts of these plots. These figures should be included in the manuscript. Similar analyses should be used to explore possible correlations between nucleation and temperature, RH, convection, rainfall, etc. Use all of the available data in these plots.

p. 14214 line 4: Are the authors suggesting that the observations are consistent with ion-induced nucleation, or simply that the apparent anticorrelation with surface area is consistent with nucleation models? If the latter, then there are many more appropriate references than Lovejoy et al. Same issue p. 14218 Line 18 of the conclusions. If the authors are proposing that the observations are consistent with ion induced nucleation, then there needs to be more discussion.

p.14214 line 8: The criteria used to classify convection and non-convection seem arbitrary and should be justified. It would be better to quantify some important charac-

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teristics of the convection process, e.g. by rate of uplift, and plot N4-9nm versus these convection parameters to examine possible correlations.

p. 14214 line 15: The authors state that they "analyzed several strong new particle formation events and non-events", with reference to the data presented in figures 1 through 8. The figure captions refer to these events as strong and weak (not non-events). This is an important difference.

In this work, observations of 4-9 nm particles are referred to as "new particle events", whereas by definition the new particles are formed when they grow past the critical cluster, which is very likely much smaller than 4 nm. Hence the particles are "formed" hours before they are observed. Therefore statements like "showing a strong new particle formation event that occurred at night" (p. 14214 line 27) may be more accurate as "showing high concentrations of 4-9 nm particles observed at night". Note that it will take $>$4 (40) hrs to grow to $>$4 nm with 10**7(6) H2SO4 cm-3, assuming that H2SO4 is the main growth agent. Note also that [H2SO4] should drop when the actinic flux drops at night. It would be a very important observation if it could be shown that nucleation occurred in the dark. However one cannot rule out nucleation during the previous day as the source of the nighttime ultrafines, without knowing the particle growth history. Does the growth history information exist?

P 14215 last paragraph: The possibility that convection enhances aerosol precursor concentrations in the UT is intriguing. However, without measurements of aerosol precursors and even knowledge of the identity of aerosol precursors, these arguments are highly speculative. Also, one can not rule out the importance of earlier trajectory history due to possible long lifetimes of aerosol precursors. For example, the atmospheric lifetime of SO2 with respect to OH reaction is roughly two weeks. Therefore it may be very difficult to draw any conclusions regarding the role of convection based on 5 day trajectories without knowledge of precursor concentrations.

p. 14216 line15: Please clarify if "polluted" and "clean" are supported by trace gas

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measurements, or just assumed. These are not good assumptions solely based on the trajectories presented in figure 6.

p. 14216 line 19: The authors state that rainfall reduces surface area. However the weak nucleation event (fig. 5) has a surface area comparable to the strong event (fig. 4) despite more intense recent rainfall. Do the observations presented in this work support the postulate that rainfall reduces surface area? Is there enough data to plot surface area versus a measure of recent rainfall to test this postulate?

The authors should indicate the number of observations that are the basis for the data presented in table 1.

p. 14218 line 2 conclusions: The statement that new particle formation is active in the UT due to low temperatures needs some justification and references.

p. 14218 conclusions: The conclusions are overstated. See comments above.

Figure 2: It is difficult to discern the number concentrations from the plot. It would help to make the number concentration scale 0-1000 cm-3. Panels (a) and (b) are not self consistent (although this may be an optical illusion due to the scaling). The size distribution has a significant fraction of large particles, but panel (a) suggests that all the particles are small. In all of the data figures the time period that the "average size distribution" represents should be indicated.

Figure 3 caption "...where the event occurred" is really "where the elevated ultrafine concentrations were measured".

Figure 1,2,4,5,7, and 8 captions: x,000 km = x km?

Figure 10: It would be very interesting to see the altitude and/or temperature of the observations on the plot.

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