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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.

D. R. Benson et al.

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

Ref2: The following is a review of the manuscript, "The effects of convection on new particle formation in the free troposphere: Case studies" by Benson et al. This review pertains to a revised version of the manuscript. The case studies in this revision are improved and provide a clearer representation of the relationship between air history and new particle formation. However, the discussion of this relationship and implications have not become more clear in this version. A number of statements regarding air mixing, convection, and surface area, and model output are not explained, not clear, or not supported by the current presentation. This manuscript may yet be publishable in ACP provided that the four major and several minor points below are addressed by the authors.

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RE: We greatly appreciate the reviewer's comprehensive and very helpful comments and revised the manuscript accordingly. Revisions were made to clarify the implication of the relationship between airmass history and NPF. Major revisions include: we replaced the term of "convection" with "uplifting", explained air mixing more in detail, and added references for surface areas.

Ref2: 1. From the relative humidities, it appears that all cases presented were taken from cloud free sections of the flight. Please state this explicitly, and also how in-cloud data was either filtered out of the overall analysis in Table 1 or how in-cloud data might affect those values.

RE: We added lines 113-119 now: "There were only less than 2% of the measurement data from this mission showed RH values greater than 100%. All case studies presented here were taken from cloud free sections of the flight (e.g., RH < 60%) so new particles were not affected by clouds. Our previous studies also have shown that shattering of clouds in the inlet of the NMASS and FCAS instruments has little effects on the measured aerosol number concentrations (while there are some effects on mass concentrations) (Lee et al., 2004), so the measured new particles were unlikely affected by cloud processing."

Ref2: 2. NPF events and non-events are defined as in Young et al. The definitions of strong and weak NPF events are defined relative to a 'background' N_{4-9} value from Table 1 (lines 95-97). Also, concentrations for non-NPF events are compared to 'background' values (lines 104-107). It is unclear what the 'background' value represents since the value in Table 1 is heavily weighted (95%) with NPF cases. The authors should state how the strong/weak criterion is determined and what is meant by 'background'. The distribution of N_{4-9} may not be easily split into strong and weak categories. In the absence of an appropriate physical criterion, a statistical method could be applied, eg, strong and weak defined as N_{4-9} at >1 standard deviation.

RE: Yes - it is correct that strong and weak events are determined by comparing with

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"background" concentrations and we agree that it is important to define "background". Lines 95-102: "Each NPF event is further classified as a strong or week event by comparing with "background" concentrations, which are referred to as the median concentration values from all events shown in Table 1. Strong events are for the cases when N_{4-9} > 500 cm^{-3} [which is approximately the same value as one median absolute deviation value higher than the median N_{4-9} for all events, (275 + 198) cm^{-3} in this case; similarly to Young et al., (2007)]; weak events are defined when N_{4-9} < 100 cm^{-3} (one median absolute deviation value lower than the median N_{4-9} for all events, which is (275 - 198) cm^{-3}, and we took 100 cm^{-3} here for simplicity)."

Ref2: 3. In lines 119-121 the presentation of N_{4-9} for NPF and non-NPF cases seems a little circular since N_{4-9} was used to define NPF and non-NPF. Reword these statements. The important point here is that a small fraction of measurements, the non-NPF cases, showed an obvious and large deviation from the N_{4-9} median.

RE: We agree. Lines 131-135: "On the other hand, non-NPF events had a median N_{4-9} of 4.93 $\pm$ 4.88 cm^{-3} and a median N_{4-2000} of 60.8 $\pm$ 42.3 cm^{-3}, both much lower than the overall N_{4-9} of 275 $\pm$ 198 cm^{-3} and N_{4-2000} of 457 $\pm$ 273 cm^{-3}; the important feature here is, however, that a small fraction of measurements (5%), the non-NPF cases, showed an obvious and large deviation from the N_{4-9} median."

Ref2: 4. The definition of convection (lines 134-139) is not conventional. Convection is usually defined as a small scale process kilometers or less in size. The NOAA HYSPLIT model using GDAS/FNL output has a grid resolution of 1 degree and is unable to resolve convective systems. The air motion referred to in the manuscript is usually described as large scale ascent or uplift. Labeling this as convection is somewhat misleading, and language throughout the manuscript should make this clear, including the title.

RE: We agree. We have replaced "convection" with "uplift" throughout the manuscript and the title is also changed now to "The Effects of Airmass History on New Parti-

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cle Formation in the Free Troposphere". We also clarified the definition of "uplifting" compared to "convection", lines 148-159: "In the present study, uplifting of the airmass is defined based on the NOAA HYSPLIT backward trajectory outputs (e.g., airmass altitude dependence with time) (Draxler and Rolph, 2003). Uplifting is referred to as the cases when the airmass 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 airmass was exposed to these low altitude source regions for at least 2 days before the vertical motion. On the other hand, if this rate was less than 3 km per day or if the airmass spent less than two days at an altitude of 2 km or less, we considered such a case as a non-uplifting event. It is noted that this "uplifting" process is slightly different from the conventional "convection", which is usually defined as a small scale process on the order of kilometers or less in size (the model output from NOAA HYSPLIT calculations only has a grid resolution of 1 degree and cannot truly resolve convective systems)."

Minor Points

Ref 2: Lines 124-125. Add values and a reference(s) for previous surface areas measurements to put these low values in perspective.

RE: Revised. Lines 138-142: "Our low surface areas are consistent with other studies [4 – 6 $\mu m^2 cm^{-3}$ on average (Young et al., 2007), 3.4 $\pm$ 1.7 $\mu m^2 cm^{-3}$ (Lee et al., 2003) and less than 10 $\mu m^2 cm^{-3}$ (Twohy et al., 2003; Carslaw and Karcher, 2006)] and these low surface areas in general also explain the high frequency of NPF observed in this region."

Ref 2: Lines 174-177 and 212-214. The statements about HYSPLIT precipitation and solar flux should be moved to the air mass history discussion in the next paragraphs.

RE: Corrected.

Ref 2: Line 178. Should read Fig. 4.

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RE: Corrected

Ref 2: Line 187. What are 'expected' aerosol precursors?

RE: Lines 216-218: "„ the expected aerosol precursors (e.g., H_2SO_4, NH_3, organic compounds and water vapor, as well as OH and sulfur compounds that can be oxidized to form H_2SO_4, including SO_2) „."

Ref 2: Line 188-192. What evidence is there for entrainment and mixing in the free troposphere? Please explain these statements.

RE: We clarified this statement. Lines 219-223: "It is also possible that air mixing might occur when the humid and warm air was rapidly uplifted to higher altitudes and mixed with the cold and dry air at the higher altitudes and this case, a steep gradient of temperature and RH took place to enhance nucleation rates, because nucleation is a non-linier process as discussed in Nilsson and Kulmala (1998)."

Ref 2: Lines 193-195 and 256-260. Lack of convection for non-NPF cases is an important result. At a number of points throughout the text the authors state that non-NPF events lacked convection. Please explicitly state how convection or uplift was determined for cases not presented in Table 2. Were HYSPLIT trajectories systematically calculated for all NMASS data points? Please provide specific results including numbers of NPF and non-NPF cases that did and did not experience recent convection.

RE: We agree. We added, lines 160-168: "HYSPLIT trajectories were run for a large number of cases other than those presented in two case studies in Sections 3.2 and 3.3. However, because of the tremendous amount of data points we did not calculate for each individual data point. Also, when calculating HYSPLIT trajectories, one can only input the UTC time in hours for the starting time and our measurements were in 1 second and the data presented here were averaged in 30 seconds. Regardless, for NPF events (Table 1), in general it seemed that at least the majority of the time (> 50 %) the events displayed some degree of uplift. On the other hand, all non-NPF events

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found in the free troposphere region did not experience uplifting of airmasses (Section 3.2).” Also, in lines 329-330: “However, there were also some NPF cases where vertical motion clearly did not occur (< 50% of the NPF cases)„”

Ref 2: Line 216. The statement about ’typical differences’ is vague and not necessary.

RE: Removed.

Ref 2: Line 227. Should read Fig 8.

RE: Corrected

Ref 2: Lines 232-234 “This is because ...” It is unclear to what this statement refers. Please reword and clarify.

RE: Lines 261-265: “These results show that at altitudes from 9 to 14 km, particle concentrations are higher in the subtropics and midlatitudes than in the tropics, consistent with the Hermann et al. (2003) trend; both the present study and Hermann et al. (2003) were mostly conducted near the tropopause region in the midlatitudes at similar latitude ranges.”

Ref 2: Line 235-236 Add references for this statement.

RE: Pan et al. (2007) is cited (Lines 267).

Ref 2: The statements about Lee et al are convoluted: Lower concentrations at low latitudes is due to data being mostly in the subtropics and poles? Please clarify. It might be instructive to state data from Lee et al 2003 for comparison.

RE: Lines 270-275: “On the other hand, this trend is different from the previous report by Lee et al. (2003) which showed higher concentrations of ultrafine particles in the lower latitudes. Because a majority of the data in Lee et al. (2003) were taken in the subtropics and polar regions, rather than in the midlatitutes, while the present study was made mostly in the midlaltitude region, a direct comparison between Lee et al. (2003) and this study is difficult.”

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Ref 2: Lines 252-255. This statement needs to be supported by giving surface area values for other regions.

RE: Lines 288-290: "„ such low surface areas measured in this region in general [4 – 6 μm^{2}cm^{-3} on average (Young et al., 2007); 3.4 $\pm$ 1.7 μm^{2}cm^{-3} (Lee et al., 2003); < 10 μm^{2}cm^{-3} (Twohy et al., 2003; Carslaw and Karcher, 2006); 1.58 $\pm$ 0.87 μm^{2}cm^{-3} from the present study] „"

Ref 2: Fig 2. The temperatures listed in the caption for a) and c) don't match the plot.

RE: Corrected

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