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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric sub-3 nm particles at high altitudes" *by* S. Mirme et al.

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We thank the referee for the valuable comments which helped to improve our manuscript.

Comment 1 *The first goal of this work is, according to the introduction bottom of page 19438, to demonstrate the capability of Airborne NAIS to measure neutral sub- 3 nm clusters in the free troposphere. This goal is not evaluated in the conclusions. I would suggest that it is and that the importance of the novelty of these experiments is put forward.*

A paragraph about NAIS was added to the conclusions.

Comment 2 *I have a problem with the statement "indicating a minor overall effect by*

clouds". Assuming that you are referring to an effect on particle formation and small ions I do not think that a comparison between in-cloud and out-of-cloud will be informative in the case the effect takes place in the outflow of clouds. A comparison of outflow vs other out-of-cloud data or outflow vs in-cloud may show a different picture.

Unfortunately we don't have this data available. Even the total in-cloud measurement time was very short, so there are hardly any datapoints for cloud outflow.

Comment 3 *The figures are well chosen and prepared and all very relevant. The figure captions are, however, in some cases very short. I would recommend that you carefully consider if more information is needed. Please see also some detailed comments below.*

We improved the figure captions.

Comment 4 *I have two comments concerning the vocabulary and choice of expressions. The first one is the use of the word aerosol. In many cases in this paper the expression "aerosol formation" is used referring to formation of particles in an existing aerosol and changing its properties. An aerosol is defined as airborne particles AND the carrying gas. It is my opinion that we should be careful not to let the meaning of the word aerosol drift to represent only the aerosol particles. I would therefore suggest that the authors use e.g. the expression "particle formation" and carefully check the use of the word aerosol in the manuscript.*

We changed most instances of "aerosol formation" to "aerosol particle formation".

Comment 5 *In many cases, the expression "size range" is used in combination with numbers in nm. It is, however, not stated if the size refers to radius or diameter. From*

equation 4, expressing mobility as a function of radius, one would assume the size refers to radius but diameter is also mentioned in some cases. I would recommend that you change "size" to "radius/diameter" at least the first time that size ranges are presented in a paragraph and in all figures.

We added the word “diameter” to many places where it was unclear what size was discussed.

Comment 6 *Abstract line 10: What does the word "actively" refer to?*

The word was removed.

Comment 7 *Introduction, line 16: "particle" should be "particles".*

The error was corrected.

Comment 8 *Page 19436 last 3 lines: Do these observations refer to ground based observations or are there also airborne measurements?*

The wording was clarified to “takes place in a wide variety of locations both on ground and in the free troposphere”.

Comment 9 *Page 19437 line 13: "Secondy" should read "Secondly".*

The error was corrected.

Comment 10 *Page 19439 line 19 and page 19442 line 11: In one of these places the flows are given as l/s and the other one as cm3/s. I would recommend that you are consistent.*

We changed both units to l/s.

Comment 11 *Page 18439 line 22: I do not understand the meaning of the expression " which removes analysed particles from the air sample".*

The expression was changed to “removes any particles from the air sample that could be detected by the analyzer”.

Comment 12 *Page 19443 line 15: "3 m" should probably read "3 μm".*

The error was corrected.

Comment 13 *Page 19446 lines 18-23: It is stated that the 2.5-3 nm particles varied little with altitude and that the 4-10 nm particles showed distinct vertical profiles. This indicates a very pronounced different in the vertical distribution of these two sizes, but when looking at figure 8 they look rather similar. This is especially the case for the total particle number. Please comment on this.*

We changed the wording to “In the size range 4–10 nm, both charged and total particles showed a maximum below 2 m and a minimum between about 4 and 6 km”.

Comment 14 *Page 19447 line 2 and figure 8 c,d and 10: Is it possible to give numbers on the fraction of charged particles at charge equilibrium or to indicate the level in figure 8 and 10? To illustrate the statement on the top of page 19447. Also, would you not expect undercharge for growing particles as they may remember their charging state when they were smaller?*

We added some estimates for the equilibrium charging state: “In charge equilibrium and within the boundary layer, the ratio between total and charged particle was calculated to be around 90 for the size range 2.5 – 3 nm and about 30 for the size range 4 – 10 nm.”

If originally undercharged, the growing nuclei tend to remain undercharged until they reach a certain size dictated by their growth rate and cluster ion concentration. The theory behind this “charging dynamics” is explained in detail in given references.

Comment 15 *Page 19447 line 14-16: What is the reason for the loss of negatively charged clusters? Do the inlet losses increase at height? Or does it have something to do with the size range of the instrument at different pressures?*

We changed the last sentence to “Above 4 km the instrument starts to lose part of the negative cluster ions due to the shift of the measurement size range.”

Comment 16 *In the first part of the conclusion section, a new discussion is brought up. Would it be possible to have this discussion earlier?*

The purpose of this finding is to tie up the earlier understanding on FT nucleation and the new findings obtained in the current study. Therefore, we think the appropriate place of this text in conclusions. The background and motivation for FT nucleation studies have been given in Introduction.

Comment 17 *Page 19448, line 25: does continuous refer to continuous with height or continuous in time?*

It refers to continuous in height. We changed the text to “... frequent nucleation taking place throughout the tropospheric column.”

Comment 18 *Table 3: Why are there no data for positive charging and 4-42 nm?*

The table row had accidentally got lost. It was re-added.

Comment 19 *Figure 3: What does the color code refer to?*

Different colors show different flights. It is now mentioned in the figure caption.

Comment 20 *Figure 10: Since different colors are used you can also give the color code.*

We added the color code to the caption.

Comment 21 *Figure 13: Would you not assume a bimodal histogram for nanometer sized particles: either there is a particle formation event (high concentrations) or there is no particle formation (low concentrations) and medium concentrations would be less frequent?*

The aircraft passed different air masses very quickly, so we could not measure a formation event from start to finish but only may have caught glimpses of events in different stages of development. These few data points would not give a distinguishable separate mode especially since the distributions are wide due to high variability of the data.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 19435, 2009.