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

S. Mirme et al.
sander.mirme@ut.ee

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

Comment 1 This manuscript deals with sub-3 nm particles at high altitudes. The authors analyze data from several CPC’s and NAIS instrument onboard DLR Falcon 20 aircraft. They present data that has not been measured or published earlier. In this respect it is unique paper. As a result they conclude that new particle formation takes place actively throughout the tropospheric column and these particles are formed via neutral pathways in boundary layer and there were no signal of increasing role by ion-induced nucleation towards the upper troposphere. However it should be kept in mind that these measurements do not present the whole atmosphere, rather one experiment in May 2008 in western Europe. Authors do say that they results “indicate”. So this is by no means any closure, rather just one observation, and I would state this more clearly.

We added a sentence to the end of the conclusions sections to reaffirm that.

Comment 2 Move text page 19438, line 8-19 to materials and methods

We moved most of the paragraph to “Materials and methods” first paragraph, but left the lines about what NAIS measures and why it was flying.

Comment 3 Page 29439, line 25; ...is able to measure at varying altitudes from inside the aircraft. I know what authors mean I get impression that the sample is taken from inside the aircraft. Also, is the instrument in environmental pressure or in the pressure inside cabin.

The wording was clarified: “We developed a new version of NAIS, which is able to measure ambient air at varying altitudes while being situated inside a pressurized aircraft.”

Comment 4 Page 19441, equation 1, please define Epsilon_0

We added “(\(\varepsilon_0\) is the vacuum permittivity)” to preceding paragraph.

Comment 5 Section 2.6, the calibration of the CPC are done close to sea level pressure, the cabin is at lower pressure. The CPC’s are tuned up to measure particles larger than 10 and 4 nm. Especially the 4 nm limit is very sensitive to small changes in supersaturation. This is strong function of diffusion coefficient which is inversely proportional to pressure. Does author have idea of the magnitude of this effect?
The reviewer is correct in pointing out that the CPC detection efficiency (being a function of size) may in principle change with altitude (pressure). It is however the outside pressure which is relevant, not the cabin pressure, because the sample lines and exhaust lines for the CPCs have no connection to the cabin. We do ensure in the CPCs used in this study that the temperature difference between the saturator and condenser parts of the instruments (controlling inside butanol supersaturation) is monitored and kept constant during the flight. It is possible that the lower cut-off size of the CPC slightly changes with pressure and the effect is not quantified in detail for the CPCs used in this study. We refer however to the paper by Hermann and Wiedensohler, 2001, in Journal of Aerosol Science 32, which showed for similar CPCs that the effect of change of the minimal particle detection limit with pressure is fairly small.

Given that there are also uncertainties in ultrafine particle number concentrations caused by diffusion losses and which are difficult to quantify as briefly mentioned in the manuscript, we do not believe it is in any way critical for the conclusions of this paper should the actual cut-off size of the CPCs change slightly change with pressure. There is clear observational evidence that particle formation events can be detected with two CPCs set to different temperature settings can be observed throughout the altitude range covered by the Falcon.

In the text we added one sentence referring to the above mentioned paper.

**Comment 6** Page 19445, lines 16-23, does author have idea of the magnitude of overcharging?

The overcharging estimates are given in section 2.4. It should be no larger than 10 – 20%.

**Comment 7** Page 19447, lines 17-19, to me it seems that in figure 11 the histograms are shifted to higher particle/ion ratios as the altitude increases.

There is indeed a small shift towards higher particle/ion ratios. Is is visible in figure 8c where medians are presented, but the histograms on figure 11 show a significant shift only at the very highest altitude range. We changed the wording to “distribution of the ratios remains nearly constant regardless of altitude” from “virtually consistent”.

**Comment 8** Section 3.3: please define “in-cloud” and “out-of-cloud”

We added “The presence of cloud around the aircraft was decided based on the concentration of 3 – 20 µm particles, which was measured by an on-board FSSP-300.” to the second paragraph of section 3.3.

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