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Atmospheric Chemistry and Physics Discussions

Interactive comment on "In situ detection of electrified aerosols in the upper troposphere and in the stratosphere" *by* J.-B. Renard et al.

Anonymous Referee #2

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General comments:

The authors present balloonborne measurement of charged aerosols using an improved version of their instrument, the STAC aerosol counter. These data were obtained during a flight in March 2011 above Kiruna, up to 24 km. The authors compare their measurement with a stratospheric ion-aerosol model. The authors conclude to a possible impact of charged aerosols on the electrical field in the middle stratosphere and above, and further on sprite formation. We regret the quite superficial analysis of the measurements, the too fast conclusion on "good agreement" and the absence of error calculations, while a correct uncertainty is crucial to conclude on the presence of the suspected layer of charged aerosols. Further, the use of the English language should be revised very carefully.

Specific comments:

Here are my detailed remarks:

- L. 13, p.7063: The first sentence should be precised. Any molecule has electric charges !

- L. 23-24, p.7063: This fact concerning the mesosphere seems not relevant in this context where aerosols are studied up to 24 km.

- L. 3-4, p.7064: What do the authors mean by "one by one" ?

- L. 3-4, p.7064: Why do the authors say that the intensity of scattered light is proportional to aerosol size? This is for sure not a result from the scattering theory!

- L. 3-4, p.7064: It would be interesting that the authors provide the detection limit of their instrument and the uncertainty for measurements of this order of magnitude.

- L. 5-7, p.7064: This sentence is not well formulated. The 13 size classes are not a property of particles counting technique, but well a feature of the instrument. The 13 classes could be given, at least by a reference to figure 1.

- L.8-9, p.7064: Why is the light scattered by solid particles smaller than light scattered by liquid particles?

- L. 52, p.7064-l. 1, p.7065: What about the uncertainty obtained on the measurements? Values of 85%, 25% and 8% are found for concentration of 0.001, 0.1 and 1 cm-3 [Deshler et al., JGR, 2003] and the uncertainty is higher for thick particles than for thin particles. In the present case, errors on both measurements are cumulative. In view of the orders of magnitude obtained for the concentration of charged and uncharged particles (See Fig. 5), there is no real evidence that the structures the authors detect are real. The authors should provide a detailed discussion of their error calculation. The only mention about error is the error bars in Fig. 2, that are all of equal size over the whole figure, which means that the relative error would be the same for both charged and uncharged particles, independently of the pressure/altitude, the particle concentration (from about 10-5 to 1 cm-3) and particle size (from 350 nm to more than 3 μm)! This seems to be a quite unrealistic estimate of the uncertainty. This impression is corroborated by the occurrence, in Fig. 2, of concentration measurements that are appreciably larger for the uncharged particle fraction than for the total particle concentration (without overlap of the error bars).

- L. 3-10, p.7065: The absence of bias in the measurement of charged particles is really not straightforward. It can be expected that the electrical properties of the particles may differ from one species to another (for instance, between solid particles of different types), making their response to an electrical field different. The authors don't give any explanation about the laboratory tests supposed to verify this assumption. They should bring much more convincing arguments, again with a reliable estimate of the uncertainty of their laboratory tests.

- L. 6, p.7065: The authors should precise on what no bias is introduced.

- L. 19-20, p.7065: The use of the expression "typical conditions of aerosol content" looks strange.

- L. 23, p.7065: What do the authors mean by "change of the concentration envelope"?

- L. 1-10, p.7066: How did the authors choose the limiting values of the particle class? Stratospheric aerosol particles, of which the typical size varies between 50 nm to 500 nm, are expected to be all in the first class. It is besides confirmed by Figure 2 that the concentrations in the 2 last classes are only marginal. The percentage of charged looks impressive, but the corresponding concentrations of uncharged particles are really small and, compared to Figure 1, seem to be at the limit of what the instrument is able to detect. Therefore, again, the authors should provide the detection limit of the instrument, and a proper error estimate to assess the relevancy of these estimates.

- L. 19-21, p.7066: The authors should add some short explanation about the reason

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why the ionization is the highest at 14-17 km, and which processes make that SO4- and NH4+ are the most encountered ions (is it what the authors mean by “major”?).

- L. 25, p.7066: What do the authors mean by the words “the rate of change ... concentrations”; this sentence is incomprehensible, and it “ve” is supposed to be some quantity/parameter, it should be correctly defined, as well as n+ and n-.

- L. 9-11, p.7067: The authors could give a minimum details about the Hoppel and Frick model to give a better insight about what they do. At least, it would give a sense to the information give about the ionic mobility and mean free path.

- L. 10, p.7068: What does the expression “simulated probability of charge distribution” mean ?

- L. 10-19, p.7068: These numbers and orders of magnitude give the impression that the phenomenon considered by the authors is really marginal. As already mentioned above (See rem. On ll. 1-10, p.7066), the concentration in particle classes 1-3.3 μm. And >3 μm can be considered as negligible. The class 0.33 -1 μm already excludes a great part of the aerosol population, and we can infer moreover that the particles that effectively contribute positively to this “probability” are the particles with size close to 1 μm, i.e. particles found in very small quantities. The authors should recompute their estimate by using realistic particle classes reflecting the real aerosol granulometry.

- L. 20-24, p.7068: I don’t see how the authors can conclude that the concentration of neutral particles shows a good agreement between the model and observation below 10 km and above 20 km. There is a factor 1 to 4 between the measurement and modeled concentration of uncharged particle for the class 0.35-1 μm, whereas the model foresees (L. 11-12, p.7068) that about 20% of the particles remain neutral. It seems that, either the uncertainty on the measurements is much too high to be useful for the detection of such a small change in the amount of charged particles, or there are other effects dominating the process and making the model insufficient to describe the problem. Again, a proper error assessment is crucial and really missing.

- L. 24, p. 7068-l.2, p.7069: This study seems definitely not mature enough to speculate on further possible interpretations.

- L. 12, p.7069: The presence of sporadic transient layers of charged aerosols, in view of what was said before, seems very uncertain.

- L. 24, p.7069-l.3, p.7070: How can the authors extrapolate what happens up the mesosphere from measurements reaching at highest 24 km altitude? This kind of speculation should be supported by convincing arguments, which is not the case at all. Overall, the significance of the presence of charged aerosols in the stratosphere is still to be supported by strong experimental measurement and a rigorous estimation of the uncertainty.

Technical corrections :

- L. 3, p.7063: Neely et al. is referred to as from 2011 in the introduction and from 2001 in the bibliography.

- L. 13-16, p.7063: This sentence seems incorrect.

- L. 21, p.7063: Reference by Thomas et al. is not given in the bibliography.

- L. 4, p.7068: a ")" is missing.

- Eq. (4) and L.6, p.7068: Why do the authors introduce the quantities n-, n+? What is the link with the notations used in Eqs (1-3)? This new notation seems redundant and is confusing.

- L. 7-9, p.7068: This sentence repeats a previous sentence.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 7061, 2013.