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Comment on acp-2022-238

Anonymous Referee #2

Referee comment on "Mass spectrometric measurements of ambient ions and estimation of gaseous sulfuric acid in the free troposphere and lowermost stratosphere during the CAFE-EU/BLUESKY campaign" by Marcel Zauner-Wieczorek et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-238-RC2>, 2022

The manuscript entitled "Mass spectrometric measurements of ambient ions and estimation of gaseous sulfuric acid in the free troposphere and lowermost stratosphere during the CAFE-EU/BLUESKY campaign" by Zauner-Wieczorek and co-authors presents ion measurements in the UTLS region. To this end, an API-ToF-MS was operated onboard the HALO aircraft and sampled air masses primarily over Western Europe. The negative ion mode was found to be dominated by NO₃⁻ and HSO₄⁻ as well as clusters thereof. Based on the measured ion concentrations the number concentration of sulfuric acid was derived. The positive ion mode was studied in less detail but protonated pyridine was identified as a major ion. Based on the data presented an increase of nitrate ions with altitude was found while hydrogen sulfate ions as well as sulfuric acid showed a more evenly distributed trend. My overall assessment of this manuscript is quite positive and it clearly fits the scope of ACP. Especially the introduction is well written and gives a nice overview of previous work, however, a few things need clarification and improvement before final approval.

Let me start with section 3.1.3 which I feel least comfortable with. While the in-cloud measurement shows some interesting features, the interpretation seems speculative and immature to me. In a way the section sounds vague and does not quite fit the rest of the manuscript. Apart from the fact that this was a one-time signal over 30 seconds only, there are a couple of questions that need clarification. As I understand this measurement took place at an altitude of >5km. I actually can't believe that outside temperature at this height and latitude will be positive. What does the temperature reading refer to? What were temperatures during other flights/altitudes? On the other hand, reported values of RH exceeding 130% sound completely unfamiliar to me. Basic literature (e.g. Seinfeld & Pandis) reports supersaturations in convective clouds not exceeding 2%, so I'd expect RH to be clearly below 110%. Even if the reported numbers are correct they must be put into context otherwise readers will get confused. In addition, I'd be surprised that during such a number of flights there was only one period of 30 s in-cloud flight. What makes this cloud different from the others? Unless this section is improved considerably I'd recommend putting this topic into supplemental material or keep it for another publication

when data are clearer. Along these lines the introduction of C-TOF-AMS and OPC in the instruments section appears quite unexpectedly as they do not relate to the ion (distribution) measurements. These should better be mentioned together with the in-cloud measurements.

A few minor issues:

Page 8, line 182: "...data were averaged to 30 s". What distance does this period relate to at cruise speed? Again, put numbers into context.

Page 9, line 239: "... the value of q applied here MUST be 90%..." This is quite a strong formulation that should be relaxed, maybe by giving a range.

Page 11, Table 2: the exact mass is only given for one ion. Why not show all exact masses for reference? Or do all measured masses agree exactly with the nominal masses?

Regarding section 3.2 "Positive ions": For me and probably for many other readers it would be interesting to see an averaged mass spec of the positive ions. It is shown for negative ions (Figure 2) but not for positive ones. I would very much appreciate it if such a plot could be added.