Response to Referees’ comments

Dear Referees,

Below we attempt to reply thoroughly your comments and we also state the main revisions of the manuscript.

Referees’ comments (in blue) and author’s response (in red)

Referee #1

In general, this was a well-designed study and the results are of sufficient importance and novelty to merit publication in Atmospheric Chemistry and Physics.

We thank the reviewer for the encouraging words.

Although the conclusions are not particularly precise or substantial, the approach to the problem is commendable and the constraints on the dissociation and dehydration equilibrium constants will likely be useful to the modelling community.

We agree. Thus, we reform the summary and conclusions section. We also discuss the importance to include in models accurate descriptions of the sulphate aerosol evaporation process in the stratosphere, and to consider the volatility of sulphate particles in new particle formation studies.

It is clear that the existing literature has been well reviewed and that the current work contributes to this body of literature.

We would like to thank the reviewer and to mention that we put our efforts to connect parts of the atmospheric climate puzzle through a precise experimental and modelling study.

A major issue with the manuscript, however, is that it is too long and thus a bit cumbersome and inaccessible. For example, the introductory material is 4 pages while the methods section is 9 pages. Overall, there are 19 pages of text, a very large table and 8 multi-paneled figures. It is the reviewer’s opinion that the manuscript would benefit greatly from a reduction in length and more succinct presentation of the work.

We consider carefully this remark about the manuscripts’ length. We reduce an extensive amount of the introductory material and also the length of the methods section where it is appropriate. In regard to the description of the modelling work, included in this section, we consider the information valuable and it is difficult to proceed with major changes because it will limit the comprehension and reproducibility of the study. Additionally, we reduce the number of figures from the manuscript and attach them to the supplement. We also attach the very large table to the supplement.

Referee #2

This is a nice, if lengthy, manuscript on sulfuric acid at low RH that will be of interest to the ACP community. It is well written and appears to be soundly executed work.

We would like to thank the reviewer for his supporting comments. We take care of the manuscripts’ length and reduce it where we consider it appropriate (see response comment to Referee #1).

Couple of comments for improvement in regards to equations 2-5: The reason for the change in the basis of the activity coefficients in equations 2,5 vs equation 4 is not clear. A reason should be given. For clarity, the symbols for the activity coefficients should be different if a different basis is used. Please correct for eq 4 and 12.

The simple explanation why to use the mole fraction based form of Eq. 4 in the model due to how we thought it was easiest to implement Eq. 4 in ADCHAM, and how we did it. The eNRTL model that we use provide mole fraction based activity coefficients. Thus, instead of converting them to molality based activity coefficients, we expressed Eq. 4 in the mole fraction based form. Moreover, for a hypothetical very low water activity (a_w) a non-negligible fraction of the solvents will be H_2SO_4 and SO_3 and then we believe that it is questionable to express the species concentrations using molalities, where water is assumed to be the only solvent. On page 11, line 6 we suggest that we add the following clarifying text:

"The Eq. 4 is given in a mole fraction based form for the following reasons: a) the eNRTL provides mole fraction based activity coefficients, and b) if Eq. 4 would be applied for a_w that are even lower than considered in this work, the assumption of using molalities, i.e. where water is considered to be the only solvent, will not be acceptable."

We now correct the notation of the mole fraction based activity coefficients in Eq. 4 and 12. Explicitly, we now use f_{H_2O}, $f_{H_2SO_4}$ and f_{SO_3} instead of γ_{H_2O}, $\gamma_{H_2SO_4}$ and γ_{SO_3}. We change this notation on the manuscript in page 9, Fig. 1, Eq. 4, Eq. 12, and in the supplement Fig. S3, Eq. S1 and Eq. S2.

There are also a few minor typos, including: (4) Abstract (. . . and then measuring evaporation. . .) should be (. . . and then measured evaporation. . .). (5) Throughout, pick either sulfuric or sulphuric. (6) 'gases', not 'gasses'; 'nucleus', not nucleous; 'model' instead of 'module'?

We would like to thank the reviewer for his comment pointing on minor typos. We change accordingly the text.

In general, the main changes in the revised manuscript are:

- Reduction in the number of plots. Figures and corresponding text which are removed from the manuscript are now attached to the supplement.
- Remove the very large table and attach it to the supplement.
- Reformulation of the abstract and sections so as to increase the clarity and/or reduce the length.
- Explanation for change in the basis of activity coefficients and replace symbol γ with symbol f (representing mole fraction based activity coefficients) where is necessary (ex. in text, Fig. 1, Eq. 4, Eq. 12, Fig. S3, Eq. S1 and Eq. S2).
- Additional comments in section 4.5 and 5 relevant to the atmospheric importance of our experimental and modelling work (e.x. on the volatility of sulphate particles, on the accurate description of the thermodynamic properties of sulphate aerosol in stratosphere).
- Add a number to the first equation, the kappa equation. Thus, all the following equation numbers now shift + 1 unit.
- Typos.