Response to Referee #2

Thank you for the insightful comments. Below are the referee comments in black, my responses to each point in red and changes made to the manuscript in blue.

Hyttinen used the quantum chemistry based conductor-like screening model for real solvents (COSMO-RS) for estimating water activities atmospheric-relevant aminium salts as well as their mixtures with organic proxies. To the best of my knowledge, the author is the first to consistently apply density functional theory to determine the thermodynamic properties of atmospherically relevant aerosol-water systems. The data obtained are important for understanding the mechanism of aerosol particle formation and their hygroscopic and CCN properties. I recommend this manuscript to be published in ACP after the following issues to be addressed and modified.

Major comments:

1. Throughout the manuscript, I did not find error estimates of the modeled values. Is it possible to provide an estimate of the uncertainty of the computational results?

Author response: Thank you for this important question. See the response to comment 3 from referee 1.

2. To assess the degree of non-ideality, I recommend that Figures S2-S5 show the water activity for an ideal solution as it was done in Fig.1.

Thank you for this suggestion. The ideality lines have been added to Figures S1-S5 in the Supplement.

3. It is difficult to analyze the calculation results presented in Fig.1. I recommend to split this data into several graphs.

Author response: Thank you for the suggestion. I have made a new figure that better highlights the differences between both anions and cations. See the response to comment 4 from referee 1.

4. As a potential user, I am concerned that different parameterization methods are used for different systems. Does this mean that additional parameterizations are required for systems with a different chemical composition? If so, it may be worthwhile to consider this issue, particularly to evaluate the prospect of using the DFT method to determine the thermodynamic properties of atmospherically relevant species as compared to other models that are easier to use.

Author response: I fully agree with this. Ideally, one could use the FINE parametrization for all calculations in the future, if the electrolyte model is extended to the BP/TZVPD level of theory. The ELYTE parametrization was used here for ammonium sulfate only because the clear disagreement between experiments and the FINE parametrization. The parametrizations of COSMO-RS programs are generally based on the level of theory used to obtain the quantum chemistry input files, so that only one parametrization exists for one level of theory and the same parametrization should be used for all compounds. One exception are systems containing small ions. For this reason, the developers of COSMO*therm* have added an electrolyte parametrization for one of the levels of theory. The choice between levels of theory is usually based on time constrains, since the BP/TZVPD calculations are significantly more time consuming than the BP/TZVP calculations. For larger molecules, e.g.

proteins, using the BP/TZVPD level of theory may not be feasible. Otherwise, the COSMO*therm* developers recommend using the BP_TZVPD_FINE_21 parametrization (BIOVIA COSMO*therm*, 2021). See also my response to comment 1 from referee 1.

Minor comments:

Line 35. Sulfate and bisulfate ions are not acids. Please correct.

Author response: Thank you for noticing this. The sentence has been changed in the manuscript.

Changes in manuscript (line 35): Sulfuric acid (H_2SO_4) is derived from anthropogenic emissions of SO_2, nitric acid (HNO_3) is produced from both anthropogenic and natural processes, and iodic acid (HIO_3) and methylsulfonic acid (CH_3SO_3H) are more abundant in marine environments. In aqueous solutions, these strong acids are deprotonated to form sulfate (SO_4^{2-}) and bisulfate (HSO_4^-), nitrate (NO_3^-), iodate (IO_3^-) and methylsulfonate ($CH_3SO_3^-$), respectively.

Line 51 ... "**x** is the mixing state in mole fraction". Why not a ... mixing ratio.. or mole fraction of compound i ?

Author response: **x** contains the mole fractions of all compounds in the system, not just the mole fraction of compound i. To avoid confusion, the wording was changed.

Changes in manuscript (line 51): where $\mathbf{x}$ is the composition of the mixture, ...

References

BIOVIA COSMO*therm*: Reference Manual, 2021.