General comments:

I would like to thank the editor for inviting me to review this manuscript.

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

Minor comments:

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

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