Response to the Editor

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

I would like to thank the author for their response and revision, which improved the manuscript. Before the manuscript can be published in ACP, I would like to reiterate on two points brought up by the reviewers.

1 - Comparison with AIOMFAC (Reviewer 1, Comment 1). The supplied graphic R1 is very useful for the reader to gauge the overall uncertainty/variability between different methods. I suggest to include such a graphic into the electronic supplement along with a qualifying sentence in the main manuscript.

Additional question: What determines the aw range for calculations (ends of the lines in R1), is this x_salt = 1? It would be helpful if this could be indicated.

Author response: I have added the figure to the Supplement with a description of the comparison in the main text. The water activities in Fig. R1 (now Fig. S1) were calculated for salt mole fractions between 0 and 0.2, corresponding to all other calculations in the paper. This was added to the caption of the figure.

Changes in manuscript (line 33): COSMO-RS and AIOMFAC predict similar water activities in $(NH_4)_2SO_4$, NH_4HSO_4, NH_4NO_3 and NH_4IO_3, within a factor of 1.08 and 1.30 in aqueous solutions with 0.1 and 0.2 mole fraction of the salt, respectively (see Fig. S1 of the Supplement).

Changes in Supplement:

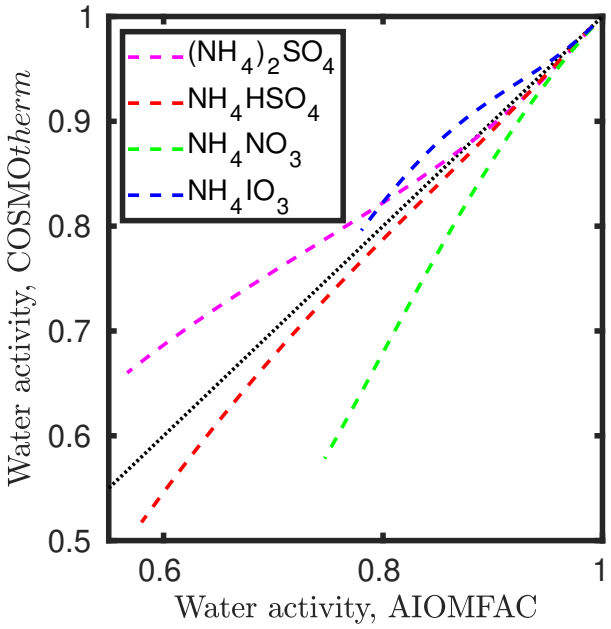

Figure S1: Comparison between COSMO*therm*- and AIOMFAC (AIOMFAC-web, 2023)-derived water activities in ammonium salts at 298.15 K. Water activities were calculated for solutions with salt mole fractions from 0 to 0.2. The black dotted line shows the 1:1 line.

2 - Both reviewers asked for an estimate of model uncertainty (Reviewer 1, Comment 3; Reviewer 2, Comment 1). While a good point was made by the author that there are no measurements for comparison, the reader would still massively benefit from a general estimation. For example, it would be interesting to give such a comparison for species for which data are available (e.g. including data into a figure complementing Fig. R1), together with a statement on whether similar uncertainties could be expected for the aminium species in question.

I am aware that this is not the main focus of this manuscript, which are aminium salts, so also a reproduction of or simple reference to existing literature could be helpful here.

Author response: I have added calculated error estimates based on the existing ammonium and aminium salt experiments of Figs S1–S3. This should provide the most relevant error estimate for the aminium salts.

Changes in manuscript (line 79: Using the selected parametrizations, COSMO*therm* agrees with the experimental water activities of aminium sulfate, bisulfate and nitrate solutions (x_{salt} <0.2) within factors of 1.39, 1.18 and 1.23, respectively. Similar uncertainties can be expected for other aminium salts. Additionally, as can be seen from Figs S2–S4 of the Supplement, the disagreement between calculations and experiments increases with the increasing salt mole fraction in the solution.

Furthermore, I have small remark:

3 - l. 73 - "The highest level of theory parametrization in COSMOtherm (BP_TZVPD_FINE_21; abbreviated FINE; using BP/TZVPD//BP/TZVP level cosmo files) works poorly with the strong charges of small ions, such as atomic ions and strong

small semi-spherical ions (such as SO42-, NH4+)." - Can you clarify the literature reference on this statement and other statements in this paragraph?

Author response: Thank you for this clarification, I have added the reference BIOVIA COSMO*therm* (2021) to all of the sentences instead of the end of the paragraph.

After these minor comments are addressed, I would be happy to accept the manuscript for publication.

Thank you very much and best regards, Thomas Berkemeier

References

AIOMFAC-web: version 3.05, http://www.aiomfac.caltech.edu, last access: 28 August 2023, 2023.

BIOVIA COSMO*therm*: Reference Manual, 2021.