Authors' response to referee #2

Hang Yin et al.

November 2021

We would like to thank referee #2 for the effort in providing this review. In the following, we repeat each comment by the referee (in black font) followed by our response and, where applicable, related changes to the revised version of the manuscript in blue font. Page and line numbers stated are those from the original manuscript.

General comments:

The manuscript by Yin et al. describes the extension of the AIOMFAC model to iodine and carbonate species. These chemical species are important for sea spray and mineral dust particles in the atmosphere. The extension will play a critical role in future thermodynamic studies on coarse mode particles. Generally, the scientific approach of the manuscript sounds. The output of the developed model agrees well with experimental data when it is available. As the authors mention in the manuscript, experimental data for the chemical species are scarce. I hope that the manuscript will stimulate experimental scientists in the area to conduct high-quality study on the related chemical species, and the authors will update the model parameters in the future when it is needed. The manuscript is well written, and the topic is within the interest of the readers of the journal. I suggest publication of this manuscript after addressing the comments

Specific comments:

1. Table 2: It would be more informative if the authors could add further information about the experimental details of the references (e.g., temperature, range of ionic strength, and experimental method).

   Authors' response: Two extra columns regarding the experiment temperature and ionic strength range have been added to Tables 2 and 6. Conceptual descriptions of various experimental methods are discussed in Section 3.4 and a more detailed description of our own measurements is provided in the supplemental information.

   Changes to manuscript: Two columns T (K) and I ($\mathrm{mol\,kg^{-1}}$) have been added to Tables 2 and 6.

2. I understand that determination of weighting factors for this type of study needs to be arbitrary. It would be ideal to have a little bit more detailed descriptions on how the weighting factors have been decided.

   Authors' response: Additional discussion has been added in the manuscript to address the

rationale behind the (initial) weighting factor assignment; see the following changes.

Changes to manuscript: Line 187: "The initial weightings of the datasets for the fitting process were set to unity unless otherwise specified (see footnote in Table 2)."
Line 209: "The initial weightings of individual datasets were first determined based on an estimated relative uncertainty associated with the experimental method. For example, water activity measurements from bulk solutions were generally assigned with weighting value units higher than those from EDB measurements. After a first round of the fitting process, the relative contributions from different datasets to the total objective fit function value were evaluated. Datasets which indicated potential inconsistencies with other datasets for a certain system, as assessed from their large (or contradicting) contributions to the objective function, were carefully checked for potential errors in input files and associated data. For datasets considered valid but of high objective function contribution, we lowered the initial weighting such that their contribution became more similar to the median objective function value contribution of different datasets. Such manual, iterative inspections of fit progress, associated graphical data comparison, and dataset weighting adjustments, aid in avoiding potential issues of large biases in parameter estimation, which may occur when the fit error is dominated by only a small subset of datasets."

3. L372: 'Instead, we estimate the interaction parameters for carbonate ions and organic compounds based on those for sulfate ions due to the similar ion size and electric charge.' The statement might make sense for the Coulombic force. I wonder if the assumption is valid for middle- and short-range forces.

   Authors' response: The short-range contribution mostly comes from the size/shape parameters of the ions as listed in Table 1. This part is not affected by the substitution method for new interaction parameters, which is intended only for AIOMFAC's middle-range part associated with Eq. 3. We argue that within AIOMFAC's framework, which includes ion–dipole interactions in its middle-range part, such substitutions are reasonable in the absence of sufficient high quality data enabling a direct parameter fit.

4. L438: 'AIOMFAC predictions are substantially lower, by about 0.1 units, than EDB measurements at lower water activity, while they are much better at higher water contents.' Do the authors have any explanation on this observation?

   Authors' response: This discrepancy reflects a possible outcome when adopting the group–contribution approach. Since our model is not attempting to only fit these EDB measurements (nor only this system) well, existing discrepancies, e.g. due to an imperfect water–sorbitol interaction representation in the salt-free case or contradicting influence from iodide interactions with other alcohols/polyols (affecting involved functional group interactions with iodide), affect the observed reduced agreement at lower water contents. Furthermore, the relatively poorer model–measurement agreement in this case compared to other systems containing alcohols with iodide salts, means the interaction between sorbitol and iodide is more sensitive to the parameters, particularly at low water contents. A more detailed related discussion of uncertainties is provided in Section 4.2.3.