Review of Metzger et al. for Atmospheric Chemistry and Physics

General Comments

In "Aerosol water parameterization: a single parameter framework", Metzger et al. present a substantial extension of the thermodynamic parameterization developed in Metzger et al. (2012). Now, the framework is shown to treat mixtures of semi-volatile and non-volatile inorganic compounds in addition to single solutes. Extensive descriptions of the algorithm are provided. Evaluation of the aerosol water content predicted by the parameterization against other aerosol thermodynamic models, field campaign observations, and aerosol optical depth (AOD) is provided. Detailed computational cost information is missing though the formulation of the algorithm almost necessitates tremendous increases in efficiency over other approaches.

This manuscript describes a meaningful advancement over the previous work of the authors, which should be as carefully described and documented as the authors have made an effort to accomplish here. Nevertheless, some clarifications and additional information are necessary to increase the utility of the work to the reader. Thus, I would support publication in Atmospheric Chemistry and Physics after minor changes and responses to the comments included below.

1. Derivation of parameterization

The authors explain that parameters were empirically derived by comparison with ISORROPIA results in a number of important equations (Eq. 3, 17, 21, A4). However, the authors do not describe the process by which these parameters were determined nor do they provide a sense of the representativeness of the selected parameter for the characteristics that were being evaluated. Given that the primary advancement of this work is the parameterization of the ISORROPIA II results, this manuscript needs to be augmented by a careful description of that process. With this type of information provided, it may become apparent that the parameterization does not reduce uncertainty as claimed (p. 33526, ll. 14) but instead increases it.

2. Range of applicability

In terms of model input, temperature shapes thermodynamics significantly, and very little attention is given to the range of temperatures across which this parameterization holds. Figure 1 uses 10˚C as the lower bound; nevertheless, in the AOD comparison, the parameterization must be applied to much cooler temperatures.

As output, aerosol water is the primary outcome evaluated, and the conclusions indicate the value of the parameterization for predicting aerosol water mass. Nevertheless, others could use the model to obtain different information (i.e., aerosol speciation). Bounding the range of input conditions and the evaluated output for which the parameterization is useful is necessary to guide the reader.

3. Efflorescence v deliquescence relative humidity

The authors show comparisons only to ISORROPIA II results that include the solid and liquid partitioning. Certainly, this result is valuable to replicate, but in the majority of chemical transport models, ISORROPIA II is executed in metastable mode, which does not allow formation of solid salts. This selection is generally made in order to account for the lack of knowledge about the history of the aerosol. Specifically, if an aerosol had previously deliquesced, the efflorescence relative humidity would be the relative humidity at which salts form rather than the deliquescence relative humidity. Could this parameterization currently be used in a mode similar to the metastable mode of ISORROPIA II? If not, how will the authors account for the lack of knowledge about the history of the aerosol that could lead to an underprediction of aerosol water?

Specific Comments

A. Text

Page | Lines	***Comment***
33500 | 5-6	Please consider rewording as “subsequent” does not make sense in the context of it being considered first.
33504 | 21	(NRO, max) is explained later in Sect. 2.5. Please consider reordering the explanation for the sake of clarity.
33507 | 9	“Despite the large differences in both approaches” This phrase seems to be an overstatement of the differences between ISORROPIA II and this parameterization.
33510 | 19	Please include this equation in the text or point the reader to the Appendix where it is included.
33512 | 1-4	Please consider making this series of statements into a flow chart. This would allow a reader to more carefully understand the algorithm introduced in this manuscript.
33512 | 14	“(partial)” is confusing. On line 21, “partial aerosol water” is used, so consider using that here.
33513 | 21	Since the formulation of the parameterization has not been explained, it is hard to understand which portions of the following analysis are evaluation and which might be the material used in the formulation of the parameterization, particularly with respect to ISORROPIA. Please differentiate.
33515 | 16-17	The evaluation here is primarily against SP2006 since EQSAM4clim is designed to follow ISORROPIA; therefore, the SP2006 results should be digitized and included in Figure 5.
33516 | 23	“gas-liquid-solid-partitioning” to “gas-liquid-solid partitioning”

33517 | 7	"to this article" - Please specify the article with a reference.
33517 | 14-16	Please provide summary statistic data to quantify the agreement rather than stating qualitatively that the agreement is close.
33517 | 18	"extent" to "extend"
33526 | 23-24	"sneak preview" to "sample"

B. Tables and Figures

Table 1. Please define the terms in the left-most column.

Figure 5. Why are ISORROPIA and EQSAM4clim predicting the greatest amount of aerosol water when no ions are present?

Figure 6. Please clarify. Are the sulfate ratios fixed or various? "various sulfate molar ratios, fixed"

Figure 7. Please simplify the title "20 Cases Comparison - Case 16".