We thank the anonymous referee for her/his time to review this work, the valuable and constructive comments on the derivation of the parameterization, the range of applicability of the vi-framework, the efflorescence vs deliquescence relative humidity and the specific comments. We hope to have addressed all points satisfactorily by the answer given below and by our answer to referee 2. Please also note the additional results which are shown in our reply to referee 2.

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.

We agree that the manuscript does not include a careful description of the process by which the vi-parameterization were determined. We will modify the sentence to:

“Besides significant computational speed-up, another advantage is that our framework minimizes the number of thermodynamic data that are typically required, while it enables greater flexibility with respect to the extension to other compounds, not considered in this evaluation.”

To improve the final manuscript version, we will add the following paragraph after p.33497,ll. 4:

“The mixed solution parameterization framework has been determined by a multi-functional fitting, limited to one parameter per compound, to match the results of ISORROPIA II. Subsequently, the multi-phase and multi-component thermodynamic system has an analytical solution by using our consistent set of analytical equations together with the vi-coefficient of M2012. Therefore, our mixed solution parameterization framework has similar applicability as ISORROPIA II.”
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.

We agree that temperature significantly influences the thermodynamics calculations. The current vi-parameterization can be applied to the same range of ambient conditions as ISORROPIA II. To be consistent with ISORROPIA II, the compound specific vi-coefficients have been derived from water activity data at room temperature. This can introduce uncertainty for certain atmospheric applications, but this limitation applies to all modeling studies that rely on available water activity data due to lack of T-dependent measurements. Within our parameterization framework we could use a T-dependent vi-parameter, provided that a set of data pairs (e.g., of solute molality and water activity at saturation) is available for the whole range of temperatures. Unfortunately, such a consistent data set is not available for the compounds considered in this work. A sensitivity study will be subject of a follow-up study.

Finally note that the same limitations as for ISORROPIA II apply to the range of input conditions for the vi-parameterization framework (EQSAM4clim), but both can be applied for typical tropospheric conditions. The model output of EQSAM4clim has been evaluated in another study (that will be presented separately) for various aerosol concentrations and different aerosol modes (aikten, accumulation coarse). Considering single solute and mixed solutions, solute compounds, their anions, cations and precursor gases, the total (lumped) particulate matter and associated aerosol water uptake are all valid output values. But the vi-parameterization has been designed, and is particularly most useful, to obtain the bulk aerosol water mass.

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?

We agree with referee 1 that for the majority of chemical transport models, ISORROPIA II is executed in metastable mode in case of decreasing RH (efflorescence path). Thus, a mutual deliquescence RH range (or RHD) is not required. Since this case is “trivial” compared to the deliquescence case, it is not considered here (“trivial” since mostly constant RH-thresholds such as 40% are used in 3-D applications due to the lack of aerosol composition dependent efflorescence measurements). Please also see our answer to referee 2 on this issue and note the additional results that show long-term applications of EQSAM4clim and ISORROPIA II by considering the full hysteresis loop (i.e., with solids and metastable mode).

We will add the following note to the MS (below p.33513,ll. 20):

“Note that Eq.(22) can be equally used for so-called metastable aerosols, for which the formation of solid salts is generally not considered.”

Specific Comments

33500 | 5-6 Please consider rewording as “subsequent” does not make sense in the context of it being considered first

Will be changed to: “Our implicit assumption is that all sulfates are neutralized first through the reactions with cations (Sect. 2.3).”

33504 | 21 (NRO, max) is explained later in Sect. 2.5. Please consider reordering the explanation for the sake of clarity.

This will be changed: (33505| ll 1) from “… concentrations constant, which we directly obtain from the NRO (see Sect. 2.5)” to “… concentrations constant. Since we do not use at this computation step any dissociation constant, we obtain the maximum concentrations (NRO, max) directly obtain from the NRO.”

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.

Will be changed to: “Despite fundamental differences in both approaches”

33510 | 19 Please include this equation in the text or point the reader to the Appendix where it is included.

Will be changed to: “Eq. (5b) of M2012 (or Eq. A6 of the Appendix).”

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.

Will be changed to: “See Fig. S2.2 for a flowchart of the Mixed solution RHD calculation.”
Figure S2.2: Mixed solution RHD calculation (B9 of the EQSAM4clim flowchart, Fig. S2.1).

33512 | 14 “(partial)” is confusing. On line 21, “partial aerosol water” is used, so consider using that here.

Will be changed to: “partial aerosol water”.

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.

Will be changed to: “We apply our parameterization using EQSAM4clim.”

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 Figure5.

Here we disagree. The SP2006 results are more or less nicely reproduced by EQSAM4clim and ISORROPIA II. The SP2006 results are too coarse to be digitized and to included in Figure 5.”


Will be changed to “gas-liquid-solid partitioning”.

33517 | 7 “to this article” - Please specify the article with a reference.

Will be changed to: “we refer to Metzger et al. (2006)”.

33517 | 14-16 Please provide summary statistic data to quantify the agreement rather than stating qualitatively that the agreement is close.
Will be changed to: “(see Table 5 for the statistics and the Appendix for the evaluation metrics).”

The summary statistic data show in Table 5 and Appendix will be added to the final version.

Table 5. MINOS aerosol statistics (Fig.8-9): (top) fine mode, (bottom) coarse mode.
EQSAM4c (EQ4c) and ISORROPIA II (ISO2) versus MINOS observations (Aug 2001).

<table>
<thead>
<tr>
<th></th>
<th>HNO3</th>
<th>ISO2</th>
<th>HNO3</th>
<th>ISO2</th>
<th>HCl</th>
<th>ISO2</th>
<th>PM</th>
<th>ISO2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean</td>
<td>EQ4c</td>
<td>SO44</td>
<td>EQ4c</td>
<td>SO44</td>
<td>EQ4c</td>
<td>SO44</td>
<td>EQ4c</td>
<td>SO44</td>
</tr>
</tbody>
</table>
| Mean   | 19.56±13.17 | 19.56±13.17
| r_m   | 16.81±0.24 | 16.81±0.24
| r_s   | 16.15±0.26 | 16.15±0.26
| RMSE  | 2.56 | 2.02 | 33.13 | 21.44 | 0.10 | 0.05 | 0.08 | 0.08 |
| R     | 0.99 | 0.99 | 0.73 | 0.67 | 1.00 | 1.00 | 0.99 | 0.99 |
| MBE   | 0.30 | 0.61 | 0.30 | 0.61 | 0.30 | 0.61 | 0.30 | 0.61 |
| GFE   | 0.04 | 0.05 | 0.04 | 0.05 | 0.04 | 0.05 | 0.04 | 0.05 |
| SS1   | 0.99 | 0.99 | 0.99 | 0.99 | 0.99 | 0.99 | 0.99 | 0.99 |
| PF3   | 0.00 | 1.00 | 0.00 | 1.00 | 0.00 | 1.00 | 0.00 | 1.00 |
| PF10  | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 |
| NPOINTS | 124 | 124 | 124 | 124 | 124 | 124 | 124 | 124 |

Will be changed.

33517 | 18 “extent” to “extend”

Will be changed.

33526 | 23-24 “sneak preview” to “sample”

Will be changed.

B. Tables and Figures

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

Will be defined.

Figure 5. Why are ISORROPIA and EQSAM4c predicting the greatest amount of aerosol water when no ions are present?

Because of H2SO4 (which is shown in the supplement only). Will be noted.

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

The sulfate ratios are fixed. Will be changed to: “different sulfate molar ratios, fixed…”

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

Reply to Referee 1   Page 5 of 6   S. Metzger
Appendix D: Evaluation metrics

- **RMSE** – Root Mean Square Error between the model \((m)\) and the observations \((o)\):
  \[
  RMSE = \sqrt{\frac{1}{N} \sum (X_m - X_o)^2}
  \]  
  (D.1)

- **\(\sigma\)** – Standard deviation of the model \((\sigma_m)\) and the observed \((\sigma_o)\) value:
  \[
  \sigma = \sqrt{\frac{1}{N} \sum_{i=1}^{N} (X_i - \bar{X})^2}, \quad \text{where} \quad \bar{X} = \frac{1}{N} \sum_{i=1}^{N} X_i
  \]  
  (D.2)

- **R** – Correlation coefficient between the model \((m)\) and the observations \((o)\):
  \[
  R = \frac{\sum_{i=1}^{N} (X_m^i - \bar{X}_m)(X_o^i - \bar{X}_o)}{\sqrt{\sum_{i=1}^{N} (X_m^i - \bar{X}_m)^2 \sum_{i=1}^{N} (X_o^i - \bar{X}_o)^2}}
  \]  
  (D.3)

- **MBE** – Mean Bias Error between the model \((m)\) and the observations \((o)\):
  \[
  MBE = \frac{1}{N} \sum (X_m - X_o)
  \]  
  (D.4)

- **\(r\)** – Geometric mean of the model \((r_m)\) and the observations \((r_o)\):
  \[
  r = \sqrt[N]{\prod_{i=1}^{N} X}
  \]  
  (D.5)

- **GFE** – Growth Factorial Error:
  \[
  GFE = \frac{1}{N} \sum \frac{|(X_m - X_o)|}{X_m + X_o}
  \]  
  (D.6)

- **SS1** – Skill score between the model \((m)\) and the observations \((o)\):
  \[
  SS1 = \frac{4(1 + R)}{(\sigma_f + 1/\sigma_f)^2(1 + R_0)}, \quad \text{where} \quad \sigma_f = \frac{\sigma_o}{\sigma_m}, \quad R_0 = 0.9
  \]  
  (D.7)

PF2 is fraction of the number of points within a factor of two of the observations, PF10 is fraction of the number of points within a factor of ten of the observations, and NPoints is the number of points used.