Authors' Response to Anonymous Referee's #2 Comments:

Summary:

This EMAC study investigates differences in aerosol modeling results using ISORROPIA II v1, ISORROPIA II v2.3, and ISORROPIA-lite. Notably, disparities in major aerosol components between ISORROPIA II v2.3 and ISORROPIA-lite are consistently less than 10%. Moreover, the application of ISORROPIA-lite results in a notable 5% acceleration in EMAC's computational performance. Despite ISORROPIA-lite's limitation to supersaturated aqueous (metastable) solutions, the authors endorse it as a dependable replacement for the previous thermodynamic module in EMAC. The paper's content is sufficiently detailed, and with the code now accessible through a Zenodo private repository, the manuscript could be considered for publication once all reviewer comments have been addressed. It's important to note that I concur with the specific comments made by referee 1 and won't reiterate them here.

We thank the reviewer for the positive review of our manuscript and the helpful comments. Below is a point-by-point response to his/her comments.

General Comments

1. *Accuracy and Clarity: To ensure accuracy and clarity, it's essential to avoid misleading statements. While the results from ISORROPIA-lite are promising, its restriction to the metastable aerosol state renders it too limited for global atmospheric chemistry applications. This limitation could lead to errors in radiative forcing estimates, particularly in the free troposphere with low humidity. What is really needed are codes that can capture the hysteresis effect of aerosols in order to improve aerosol radiative forcing effects. Therefore, the statement that "ISORROPIA-lite can be a reliable and computationally effective replacement of the previous thermodynamic module in EMAC" should be approached with caution, pending a thorough evaluation of its suitability for global applications.*

ISORROPIA-lite should not be considered as a replacement for the ISORROPIA-II stable mode, but rather as an alternative version of the model that can be selected by the user depending on the application and the desired efficiency and/or state assumption. The aim of this study is to demonstrate that ISORROPIA-lite is equally accurate in predicting inorganic aerosol composition with improved computational efficiency and to provide insight into the conditions and regions where the results of the two versions available in EMAC might differ. However, it should be emphasized that the stable assumption should not always be considered as more accurate. During simulations, atmospheric particles are transported from one simulated cell to another by simultaneously undergoing several atmospheric processes that change their chemical composition. In many cases, they end up in computational cells with completely different RH without "carrying" their historical RH profile with them. Therefore, the choice between a stable state (e.g., following the deliquescence branch of crystallization) and a metastable state (following the efflorescence branch) should not be considered obvious. While a stable state could be considered more accurate under very low humidity conditions (e.g., over remote deserts), in regions, such as those with intermediate RH and low nitrate concentration (e.g., Northeastern US), particles are mostly in metastable state. However, the two state assumptions produce very similar results in most cases, as shown in our study. Overall, following the reviewer's comment, we have enriched our discussion in Sections 2, 4, and 5 of the revised manuscript by avoiding statements that could lead to confusion about the climatic impacts of the two model versions.

2. ***Omission of References****: The omission of references to relevant thermodynamic codes commonly used within EMAC is a notable gap in the introduction, potentially impacting the manuscript's scientific credibility. It's crucial to acknowledge and cite widely accepted models, following established conventions in scientific publishing.*

We thank the reviewer for pointing this out. Indeed, EQSAM is the other available option besides ISORROPIA in the EMAC model for aerosol thermodynamic calculations and is now described in the introduction. We also clarify that EQSAM is still an available option in EMAC.

3. ***Consistency****: Ensure consistency in the spelling of "ISORROPIA-lite" and other acronyms throughout the text to maintain clarity and professionalism.*

All the acronyms used in the manuscript have been thoroughly revised.

Specific Comments:

1. ***Discussion of Activity Coefficient****: If tabulated activity coefficients are mentioned, it's crucial to provide a clear and comprehensive explanation or reference regarding their origin and relevance. This will ensure that readers fully understand their context.*

The use of tabulated activity coefficients (by ISORROPIA II and ISORROPIA-lite) is now explained in Section 2.2. The methodology for their calculation is briefly presented, with all relevant references cited (Kusik and HP (1978); Bromley (1973); Meissner and Peppas (1973)). Further information can be found in Fountoukis and Nenes (2007).

2. ***Temporal Analysis****: In Table 2, where annual means of surface concentrations are discussed, it's worth noting that a 5% difference on an annual scale can translate to significantly higher variations when considering shorter timeframes, such as hourly averages, commonly used in air quality applications. To enhance the analysis, consider extending the statistical examination to at least daily values at a regional scale, focusing on selected networks. Relying solely on mean annual concentrations limits the scope of the analysis and its conclusion.*

Tables 1,2 and 7, in Sections 3.1, 3.2, and 4.1, respectively, which presented the statistical comparison between the model estimates of the different ISORROPIA versions, have been updated to include the daily averages. The box plots in Figures 7, S1 and S2, show the regional differences of the estimated daily average coarse and fine NO_3^- concentrations by the different ISORROPIA versions for five specific regions. The regional analysis focuses on the differences in NO_3^- concentrations since this is the aerosol component with the highest discrepancy between the different ISORROPIA versions.

3. ***Computational Speed-Up Analysis****: The metric presented in Table 6 regarding computational speed-up should ideally encompass information about load imbalances within the system or undergo a more rigorous statistical analysis. To strengthen the analysis, consider running multiple iterations for each version to draw more robust and conclusive findings. As currently presented, the analysis is relatively weak, and its conclusions are somewhat limited.*

The statistics presented in Table 6 have been updated to include not only the results of a single simulation for each version, but a total of 18 simulations (6 for each version). The revised table 6 shows the average values of the statistical metrics used, as well as their standard deviation.

4. ***Section 4 Focus on Surface Concentrations**: Section 4 predominantly concentrates on surface concentrations, which may not offer a comprehensive evaluation of the metastable effect as intended by the authors. Consider revising the analysis in Section 4 to include an assessment of the vertical integral (burden) and, at the very least, a comparison of zonal means. The current presentation may be misleading without these additional elements.*

An assessment of the tropospheric burden of total NO_3^- aerosol between the two ISORROPIA versions can be found in Section 4.1. The analysis has now been extended to include the zonal mean annual concentrations of all aerosol components and their deviation between ISORROPIA II and ISORROPIA-lite (Figures S3 and S5 in the revised supplement). We found that the deviations between the results of the two ISORROPIA versions are becoming smaller as the air masses move higher in the atmosphere, until they are practically identical at altitudes above 700hPa. The discussion in Section 4.1 has been extended accordingly.

5. ***References and Errata**: Ensure that references are not duplicated and address any missing errata. This will enhance the overall quality of the document and its accuracy.*

The reference list has been thoroughly revised.

REFERENCES

Bromley, L. A.: Thermodynamic properties of strong electrolytes in aqueous solutions. *AIChE journal*, *19*(2), 313-320, https://doi.org/10.1002/aic.690190216, 1973.

Fountoukis, C. and Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K+–Ca 2+–Mg 2+–NH 4+–Na+–SO 4 2−–NO 3−–Cl−–H 2 O aerosols. *Atmospheric Chemistry and Physics*, *7*(17), 4639-4659, https://doi.org/10.5194/acp-7-4639-2007, 2007

Kusik, C. and HP, M. 1978. Electrolyte Activity Coefficients in Inorganic Processing. AIChE Symp. Series, 173, 14-20, 1978.

Meissner, H. P. and Peppas, N. A.: Activity coefficients – aqueous solutions of polybasic acids and their salts, AIChE Journal,19(4), 806–809, https://doi.org/10.1002/aic.690190419, 1973.