

Interactive  
Comment

***Interactive comment on “ISORROPIA II: a computationally efficient thermodynamic equilibrium model for  $\text{K}^+$ - $\text{Ca}^{2+}$ - $\text{Mg}^{2+}$ - $\text{NH}_4^+$ - $\text{Na}^+$ - $\text{SO}_4^{2-}$ - $\text{NO}_3^-$ - $\text{Cl}^-$ - $\text{H}_2\text{O}$  aerosols” by C. Fountoukis and A. Nenes***

**Anonymous Referee #2**

Received and published: 21 March 2007

**General Comments**

This paper presents an improvement of the state-of-the-art thermodynamic module ISORROPIA for inorganic aerosols. The new version is extended in order to take into account crustal species and relies on the same decomposition approach as the first version. Results are compared between ISORROPIA II and SCAPE2 for a series of relevant aerosols types. Comparison of CPU times with the original ISORROPIA are performed.

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The manuscript is well-written and its content is relevant for publication in ACP. Its importance in terms of modeling of inorganic aerosols is obvious. The references are well-chosen. The presentation of the results is adequate, although some results are missing (see below). The results are compared and benchmarked with SCAPE2.

I will accept its publication after the completion of the following points by the authors.

- The division of the aerosols into various modes, as a function of the ratios  $R_1$ ,  $R_2$ ,  $R_3$  or RH is artificial and probably source of non-negligible approximations. The authors do not discuss the sensitivity of the results with respect to the proposed thresholds. Since salts are expected to crystallize at equilibrium depending on the definition of the threshold, some examples, sitting at the limit between two different regimes should be provided to observe if discrepancies appear in the calculations (for instance between sulfate rich and sulfate poor aerosols) when considering two different set of equilibrium salts.
- ISORROPIA II assumes as an initial guess that all salts are crystallized. This initial guess is not very "physical". The authors pointed out that all the others models start from the opposite assumption. What is the advantage of taking this reverse approach in terms of modeling and computation?
- Comparison with ISORROPIA. The extension in terms of incorporation of crustal species is clear. Although the authors compare the CPU performance of ISORROPIA II vs its previous version, there is no comparison of actual results for specific cases. These have to be added: does the second version give a more accurate solution or the same? are there differences in the iterations process? are the solutions consistent?
- Throughout the paper, the model uses precomputed data vs is real-time computations. A more precise summary of what is done in real time and what is precomputed would be interesting to estimate both computational costs.

Finally I share the opinion of a previous referee: for one given system, is the computational result accurate and how does it compare with actual data? This point has to be addressed.

## Specific Comments

1. In the introduction, the authors mention the global optimization of a nonlinear convex problem. In fact, I think the function is not necessarily convex, but only locally convex. Could you comment?
2. In the same paragraph, the nonlinearity is assumed to be stronger for low RH and activity coefficients are used only in that case. When neglecting activity coefficients for high RH, are there inconsistencies in the results between low and high RH? The authors should precise how to define in a robust way the threshold between low and high RH (in others terms, when do activity coefficients have to be incorporated)?
3. In Section 2.4, the authors propose a ZSR approach for the calculation of the water uptake, the reason being that the direct computation of water activity is computationally expensive (although the reference they mention is pretty old). However, some of the methods appearing in the references have proved that this computation is tractable. Can the authors comment on the implementation of this additional computation? Did they try and if not do they plan to? What would be an estimation of the difference in CPU times?
4. On page 1906, the authors make some assumptions on the behavior of potassium for instance. Do these assumptions come from a lack of precision of the activity model or are they required for computational simplifications? In the latter case, what would be the additional computational effort if they are relaxed?

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5. An option is used to "force" ISORROPIA II to conserve mass. Does it mean that, in general, that the method is not mass conservative? In that case, can you quantify and justify the loss of mass.
6. On page 1913, the authors mentioned the difference in ordering the associations with K. The two models are using a different ordering. Could you develop to justify why the ISORROPIA ordering is more appropriate. Is there a way to get rid of these assumptions on the ordering of species?

### Technical Corrections

1. page 1899 : Van't Hoff equation ?
2. page 1899 - lines 4 and 6 : spaces are missing.
3. page 1902 - line 22 : sentence is not clear to me.
4. page 1904 ; last line : sentence is not clear to me.
5. page 1908 - line 14 : pKa ?

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Interactive comment on Atmos. Chem. Phys. Discuss., 7, 1893, 2007.

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