Review of "HETerogeneous vectorized or Parallel (HETPv1.0): An updated inorganic heterogeneous chemistry solver for metastable state NH_4^+–Na^+–Ca^{2+}–K^+–Mg^{2+}–SO_4^{2-}–NO_3^-–Cl^- based on ISORROPIA II " for *Geoscientific Model Development*

General Comments

Miller, Makar and Lee describe the development of a novel computational model written in Fortran 90 for the thermodynamic partitioning of a total amount of inorganic species as listed in the title between the gas and liquid phases in the presence of water. Typically, they follow the algorithms of the forward solution of ISORROPIA II for the metastable cases. Additionally, they identify and correct algorithmic formulations that lead to errors in the output of ISORROPIA. Furthermore, they implement a recently developed root finding approach instead of bisection that improves the accuracy of equilibrium solutions and the speed of arriving at them in most cases. The model is thoroughly tested not only against ISORROPIA but against the analytical solution of the equilibrium equations. Additionally, the accuracy and computational costs were explored for realistic cases as derived from the regional GEM-MACH model for selected conditions summer and winter. Finally, the code for this open source model has been made publicly available through Zenodo.

The manuscript is replete with evidence of the carefulness with which this model was developed and tested. The authors are clear as to where HETP outperforms ISORROPIA in terms of accuracy or computational cost but also where it underperforms, which is less common. In most cases, the authors posit fundamental explanations for why HETP performs differently than ISORROPIA, identifying specific algorithmic changes associated with the results in most tables and figures. For instance, the third subtable in Table 2 is evidence of an excellent investigation of the algorithmic explanation for the observed improvements. The authors have helpfully structured the manuscript by selecting tests that pertain to specific algorithmic changes to avoid inundating the reader with excessive comparisons while still demonstrating the extent of the testing conducted and then building to the comprehensive timing tests, which are sufficient to demonstrate the robustness of the model development process.

The authors are helpfully straightforward about HETP solving only the metastable cases. Since most chemical transport models only use the metastable solutions from ISORROPIA, this clarity is important but not impactful for those inclined to adopt the model. The primary change I think would improve the manuscript is to be clear at least in the abstract if not in the title that only the forward solution in ISORROPIA is included. Currently, the first mention that HETP only addresses the forward solution of ISORROPIA is at the beginning of Section 2 though in the introduction ISORROPIA is referenced as being used in CMAQ, in which both the forward and reverse solutions are employed for dynamic equilibrium of coarse mode aerosol with the rest of the population. So as to not cause a reader undue hope that well-documented issues with the reverse solution have been resolved with HETP, I would urge the authors to consider being clear that only the forward solution is implemented in HETP.

With this small but important change, a few responses to specific comments aimed to add value for future readers, and a careful grammatical revision including reduction of the number of parenthetical phrases, I would expect that the publication of this

manuscript would benefit many in the atmospheric modeling community for years to come.

Specific Comments

Lines 9–12 The claims in the first two lines of the introduction are very important and are well-supported in the manuscript. The current language in the first two sentences, especially the second, obscures the value of this paper somewhat. Consider rephrasing these two sentences, possibly into three shorter ones, for the sake of clarity and impact.

Lines 80-92 The authors have reasonable explanations for the use of the metastable assumption documented. One important additional reason is that the history of the aerosols is not tracked in these models such that one cannot know whether the mutual efflorescence or deliquescence relative humidity would best characterize the conditions for crystallization.

Also, consider starting a new paragraph with the metastable state discussion since this topic is slightly distinct from the thesis of the paragraph and other content in it.

Lines 116-8 Consider making a separate sentence with the content beginning with "which" on account of the parenthetical nature of the descriptions of parallel and vector implementations not being sufficiently clear. If that restructuring is not desired, at least make the parenthetical explanations parallel in form and each properly introduced.

Lines 305-18 Many of the enumerated algorithmic improvements, such as the more robust solution of cubic equations, are documented with sufficient detail. One helpful addition to these enumerated algorithmic improvements would be to identify one example of their application by line number in the HETP.ftn90 file included in the Zenodo repository.

Grammatical Comments

Line 45 Elsewhere "N" is written as an italicized variable. Consider formalizing this expression, too.

Line 115 Please ensure that the "forward" nature of the solution is somehow mentioned in this helpful, governing sentence.

Line 119 It is not clear to me where the idea of the metastable state representing the "efflorescence branch" arose. To my knowledge, the mutual deliquescence relative humidities are used in ISORROPIA and insufficient information exists for the efflorescence relative humidities of these salt mixtures to treat efflorescence in a thermodynamic equilibrium model of inorganic aerosols. It would be sufficient to delete this parenthetical phrase and leave the reader to the other portions of the manuscript in which the metastable state is more accurately described.

Line 159 Here and in some other cases, a ";" is missing: "initial mass adjustments, however any output" should be "initial mass adjustments; however, any output". Please revise here and in other places as needed.

Line 193-4 No comma exists after the adverb at the beginning of the sentence (i.e., "Currently,") but an unnecessary comma is used before "and" in the same sentence though the conjunction is not followed by an independent clause. Elsewhere, commas are used where semicolons are needed. Please revise the use and absence of commas throughout the manuscript.

Line 195-9 Four independent clauses are joined by two semicolons and one conjunction that is not preceded by a comma. Please look for run-on sentences such as these and revise as needed.

Line 230 "the ZSR correlation" would be better as "the Zdanovskii-Stokes-Robinson (ZSR) correlation".

Line 271 Although the code-based expression "TCl = max(TCl, 1×10^{-10})" will be comprehensible to those accustomed to writing Fortran, perhaps using words to express the concepts would be more appropriate for this part of the text.

Line 706 "computations into in" was likely meant to be "computations into".