Atmos. Chem. Phys. Discuss., 8, S1158–S1167, 2008
www.atmos-chem-phys-discuss.net/8/S1158/2008/

Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on “Online coupled meteorology and chemistry models: history, current status, and outlook” *by* Y. Zhang

Y. Zhang

yzhang9@ncsu.edu

Received and published: 1 April 2008

Reply to Comments by R. Zaveri

Interactive comment on “Online coupled meteorology and chemistry models: history, current status, and outlook” by Y. Zhang R. Zaveri

Rahul.Zaveri@pnl.gov Received and published: 31 March 2008

The author has undertaken a rather challenging task of reviewing several aerosol modules and the fully coupled 3-D models into which they are implemented. These modules and models are very complex and it is not at all straightforward to compare and contrast their approaches, strengths, and weaknesses in a manner that is fair to all the developers and at the same time be clear enough to be understood by the rest of us. Nevertheless, this important task must be done at some point and I appreciate the author’s first attempt at it.

As the lead developer of MOSAIC, my comments and suggestions are mostly focused on the author’s description of MOSAIC, its sub-modules, and their comparison with other similar modules. The objective is to rectify some factual errors and clarify some subtle but important details.

Reply: I appreciate the reviewer’s effort in reading the paper and writing the detailed comments on MOSAIC, as well as the fact that the reviewer well understood the challenging aspect of this comprehensive review paper and the fact that this is the first such review paper in the history.

Please change "CBMZ" to "CBM-Z" wherever it is used.

Reply: The suggested change has been made throughout the text and Tables in the revised version.

Page 1847, line 1: The MOSAIC reference is now in press and should be cited as: Zaveri, R. A., Easter, R. C., Fast, J. D., and Peters, L. K.: Model for Simulating Aerosol Interactions and Chemistry (MOSAIC), J. Geophys. Res., doi:10.1029/2007jd008782, 2008, in press.

Reply: The update in Zaveri et al. (2008) has been included in the revised version.

Page 1850, line 17: It should be mentioned either here that "CBM-Z extends the original CBM-IV mechanism to function properly at regional to global spatial scales and longer time periods than the typical urban air-shed simulations. The CBM-Z version implemented in WRF-chem also includes a condensed dimethylsulfide (DMS) photooxidation mechanism (Zaveri, 1997) to simulate the temperature dependent formation of SO2, H2SO4, and methanesulfonic acid (MSA) in the marine environment." Ref: Zaveri, R. A. (1997), Development and evaluation of a comprehensive tropospheric chemistry model for regional and global applications, Ph.D. thesis, Va. Polytech. Inst. and

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

State Univ., Blacksburg, Va.

Reply: The differences between CBM-Z/CB05 and the original CBM-IV have been added in the revised version.

Page 1852, line 21: WRF/Chem-MOSAIC also treats calcium, carbonate, and MSA. This information should be updated in Table 3 as well.

Reply: The additional species in WRF/Chem-MOSAIC and other models have been added in Table 3 and relevant text section.

Page 1857, lines 19-24: For a fair comparison of different modules, it should be mentioned here that EQUISOLV II and MESA have also been carefully compared with the benchmark AIM model for a number of representative test cases over the entire RH range at 298 K (Zaveri et al., 2005a). It was found that MESA was able to simulate the mutual deliquescence RH (MDRH) and the solid-liquid phase transitions much more accurately and efficiently than EQUISOLV II. The errors seen in EQUISOLV II predictions, however, were largely due to the errors associated with the activity coefficients used in EQUISOLV II. Also, because MESA diagnoses the MDRH point directly as a function of particle composition and temperature, it is significantly faster than EQUISOLV II when RH $<$ MDRH.

Reply: The intercomparison of MESA, EQUISOLV II and AIM model III is relevant here and will be added in the revised version. However, the author disagrees with the reviewer’s statement “It was found that MESA was able to simulate the mutual deliquescence RH (MDRH) and the solid-liquid phase transitions much more accurately and efficiently than EQUISOLV II”, as it did not accurately describe what was presented in Zaveri et al. (2005a). The review will include an accurate and unbiased description regarding this intercomparison. In all 16 comparative figures in Zaveri et al (2005a), MESA and EQUISOLV II give overall very close results visually to each other and to AIM Model III. As stated in paragraph 37, page 14 of Zaveri et al (2005a): “Even though EQUISOLV II is not tuned to reproduce AIM results, it is able to capture the complex growth behavior quite well. In most cases it predicted the MDRH within 4% RH points (and exactly in two cases). In two cases, the deviations in the predicted the MDRH were 8–10% RH points (cases 6 and 7), while it was as large as 15% RH point in case 14.

Given large uncertainties in activity coefficients that are currently used in inorganic aerosol thermodynamic modules, the deviation of 4% for 13 out of 16 cases and even 8-15% for 3 out of 16 cases in MDRH should not be considered to be very significant. In addition, the deviation of 8-15 occurred when aerosol water content is relatively small, which will unlikely cause any potentially large errors in simulating the phase state of aerosols. It is also important to note that AIM Model III itself does not treat the system H+ - NH4+ - Na+ - SO42- - NO3- - Cl- - H2O at temperatures other than 298.15 K and it does not treat other species that may be potentially important for thermodynamic equilibrium such as crutal species. The cases tested in Zaveri et al. (2005a) were only for a limited set of species and at one temperature (298.15 K). It is not clear to the author whether MESA is capable of treating other species or other temperatures without a re-tuning of the parameterization. This will likely limit its application for 3-D atmospheric modeling, as the equilibrium in the atmosphere may involve more species with a broad range of temperatures. On the other hand, EQUISOLV II is generalized to treat any set of species at any temperatures, it does not need to tune for the specific system at a specific temperature. Zaveri et al. (2005a) further compared the statistical performance of MESA and EQUISOLV II against AIM Model III for the above 16 cases. They relative mass growth factors (MGF) deviations they reported were 2.3% in MESA vs. 4.3% in EQUISOLV II. The relative deviations in the total water contents were 2.46% in MESA vs. 3.75%. Such differences are not statistically significant, and the number of cases tested is also too few to lead to any meaningful conclusion regarding the relative performance of EQUISOLV II and MESA. The close agreement of MESA to AIM Model II is not surprising because the activity coefficients used in MESAwere tuned to those used in AIM Model II. As stated in paragraph 29, page 10 of Zaveri et al. (2005a), they computed the multicomponent activiity coefficients with the comprehensive PSC model

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

at 298.15K. Therefore, it appears that the "observed" values were used to predict the "observed" values in their work. Since the PSC is the activity coefficient model used in AIM Model II, MESA achieved a close agreement to AIM Model II through tuning its activity coefficients to what is used in AIM Model II at a fixed temperature. Because the PSC model is limited in the number of species it can treat, the parameterization method used in Zaveri et al. (2005a) is limited to those species for the accuracy reported in their work. It is not clear to the author whether such a good agreement can be obtained when other species are included.

It should also be mentioned that ISORROPIA continues to use an approximate treatment for water content and solid-liquid equilibrium in the mutual deliquescence region. This approach is not consistent with the actual thermodynamic equilibrium solution, and can therefore introduce large errors in particle composition, size, and gas-particle partitioning at moderate and low RH values (Ansari and Pandis, 1999; Zaveri et al., 2008). Ref: Ansari, A. S., and Pandis, S. N.: An analysis of four models predicting the partitioning of semivolatile inorganic aerosol components, Aerosol Sci. Technol., 31, 129-153, 1999.

Reply: The review paper did not intend to cover all details in inorganic aerosol thermodynamics as it contains a broad review of several topics, and this inorganic aerosol thermodynamics is only a small aspect of them. More details regarding this and additional publications will be included wherever possible and appropriate.

Page 1858, line 9: It should be mentioned here that the activity coefficients used in EQUISOLV II have temperature dependence only for the NH4-Na-NO3-SO4-HSO4-Cl system, while they are fixed at 298 K for the Ca-Mg-K-SO4-NO3-Cl-CO3 system. Similarly, all the Bromley activity coefficients used in ISORROPIA are limited to 298 K as well. More importantly, the activity coefficients are not very sensitive to temperature; Zaveri et al (2005a) showed that the DRH(T) and MDRH(T) parameterizations derived using activity coefficients at 298 K are in very good agreement over 240-320 K range with those obtained with the benchmark AIM model using temperature dependent ac-

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

tivity coefficients. The MTEM activity coefficients used in MESA are therefore certainly applicable for low and mid tropospheric conditions and may even be used down to 220 K without any significant loss in accuracy of the key predicted variables. In any case, higher priority should be given to first estimating activity coefficients more accurately at 298 K than applying temperature dependence to inaccurate activity coefficients referenced at 298 K.

Reply: As indicated previously, the review paper did not intend to cover all details in inorganic aerosol thermodynamics. More details regarding this will be included wherever possible and appropriate. EQUISOLV II accounts for temperature dependence for all activity coefficients when such information are available. ISORROPIA uses KUSIK-MEISSNER METHOD to calculate binary activity coefficients and the BROLEY’s method for multiplecomponent activity coefficients. The correction factors were applied for temperatures other than 298.15 K in binary activity calculations in version 1.4 and newer versions (note that MADRID uses the latest version of ISORROPIA, version 1.7 which also accounts for temperature dependence of these activity coefficients). In addition, the temperature dependence of activity coefficients in ISORROPIA was used to correct the water uptake data for a wide range of temperatures. I The temperature dependence of activity coefficients is important. As shown in Figure 17.3 (Jacobon, Fundamentals of Atmospheric Modeling, 2nd edition, 2005), page 568, the activity coefficient of H+/HSO4- is quite sensitive to temperatures, particularly at low temperature ranges and higher molality. For example, its activity coeff. is about 2 x 105 at T = 201 K and m1/2 = 8, compared to a value of 2 x 103 at 298 K and m1/2 = 8. The differences in activity coefficient calculation can often help explain much difference among various aerosol thermodynamic modules, so different treatments on this are an important aspect of aerosol thermodynamic modules.

Page 1858, line 10: This statement is not accurate. MESA currently includes solidliquid equilibria and water content calculations for all the globally important inorganic species. It is also quite easy to include additional ions and salts in MESA. The MDRH(T) pa-

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

rameterizations for additional salt mixtures can also be easily developed. Overall, it is worth the additional one-time effort because it would result into substantial CPU time savings when applying the model.

Reply: It is factual that MESA only treats limited number of species (which are also commonly treated in most current inorganic aerosol modules), as compared to EQUISOLV II that treats significantly more species. Additional species that are currently not included in MESA may be important under some atmospheric conditions, e.g., other crustal species such as K+ and Mg2+. Expanding the current system in MESA to include more species and chemical reactions can be done, but this may require some efforts to obtain the necessary parameters for such species (and the benchmark for parameterizations may not always available) and to revise the code structure to accommodate them. A generic code is more desirable for atmospheric applications at all scales.

Page 1860, line 3: MOSAIC no longer uses the ASTEEM approach for gas-particle mass transfer as was briefly described in Fast et al. (2006). Please change the sentence to "... WRF/chem-MOSAIC uses the Adaptive Step Time-split Euler Method (ASTEM) algorithm (Zaveri et al., 2008)." Page 1860, line 18: Please change "ASTEEM" to "ASTEM" and wherever else it is mentioned, including Table 5.

Reply: The current review paper includes what has been used in WRF/Chem v2.2. The author can include the update of ASTEEM to ASTEM but need to point out that it has not been included in WRF/Chem v 2.2, consistent with updates to other model treatments that have not yet been included in WRF/Chem v 2.2.

Page 1860, lines 22-29: Please delete "ASTEEM is developed to ... ... small loss in accuracy." and suggest replacing it with the following: "The ASTEM algorithm includes a new concept of dynamic pH, a novel formulation for mass transfer to mixed-phase and solid particles, and an adaptive time-stepping scheme, which together hold the key to smooth, accurate, and efficient solutions of dynamic gas-particle partitioning

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

over the entire relative humidity range. It first analytically solves the condensation of all the non-volatile species only (H2SO4, MSA, and some NH3), and then simultaneously computes mass transfer of all the semi-volatile species (HNO3, HCl, and NH3) to all the size bins using a combination of explicit and semi-implicit Euler methods depending on the phase-state of the particles. This new approach in MOSAIC was found to be in excellent agreement with a benchmark version of the model that uses LSODES, a rigorous solver for integrating the stiff ODEs. The steady-state MOSAIC results were also in excellent agreement with those obtained with the benchmark equilibrium model AIM for monodisperse aerosol test cases. The CPU times required for fully dynamic solutions by MOSAIC per size bin per 5 min intervals (typical 3-D model time-steps) were similar to those for bulk equilibrium solutions by the computationally efficient but relatively less accurate equilibrium model ISORROPIA."

Reply: The author will include the updates that have been published (e.g., Zaveri et al., 2008). Please note that the author cannot include other updates that are not documented in peer-reviewed publications and/or conference presentations, so the reviewer is expected to provide all references for the above updates to facilitate their inclusion in the revised version.

Page 1861, line 7: It would be useful to add a couple of sentences here to describe the kinetic mass transfer approach implemented in MADRID. According to Hu et al. (2008, in press), the APC scheme of Jacobson (1997) is implemented in MADRID. However, if I understand correctly, Jacobson (2005) developed the Predictor of Nonequilibrium Growth (PNG)-EQUISOLV II scheme to overcome the oscillatory solution problem in the APC scheme and reduce the computational cost of the kinetic mass transfer treatment. So, then did Hu et al. implement PNG-EQUISOLV II in MADRID or just the 1997 APC scheme? If it is indeed just the 1997 APC approach, then what measures did Hu et al. take to overcome the numerical problems associated with it as described in Jacobson (2005)? This was not clear from reading the Hu et al. paper, which could perhaps be clarified here.

Reply: As an initial step, the APC scheme of Jacobson (1997) was implemented and tested in MADRID. For all cases tested in Hu et al. (2008), it gave a very good agreement at a time step of 15-second for kinetic/APC_MC. Our test results found that this scheme is numerically stable when a time step of 15-second or smaller is used, which is consistent with Jacobson (2005). We indeed have done testing for kinetic/APC_MC using other time steps (e.g., 0.25-30 seconds). Our test results show that even with a long time step of 30 seconds, kinetic/APC_MC with 8 sections can still give results that are reasonably close to the observations for the Hong Kong test case. The oscillation may occur for some volatile species (e.g., Cl and NH4 at a time step longer than 15 seconds for any growth scheme coupled with any equilibrium scheme due to delays in feedbacks between the equilibrium and growth calculations when the operator-splitting method is used. This has been recognized as one of the most challenging issues in resolving non-equilibrium gas/particle mass transfer (e.g., Meng et al. (1998), Sun and Wexler (1998 a, b), and Pilinis et al. (2000)). A time step of 15 seconds will be used in our 3-D WRF/Chem-MADRID testing. If it turns out that the CPU cost is too expensive to apply WRF/Chem-MADRID for long-term simulations, we will consider to incorporate the Predictor of Nonequilibrium Growth (PNG)-EQUISOLV II scheme or other similar schemes that may work more accurately for long time step.

It would be useful to add a short discussion on the CPU time requirements by various modules/models in section 3. As it stands, this section only describes the various treatments available in different models, but nothing is said about the CPU time requirements and its tradeoff with accuracy. CPU time is obviously a big concern in fully-coupled models, and so a discussion on it is warranted.

Reply: While the CPU information is useful, such information is unfortunately not always available for all modules/models reviewed in section 3. Also, the type of computers and their specifics (e.g., number of processors, memory, etc.) are typically not reported along with the CPU. So, it is not possible to include the CPU information as expected by the reviewer.

Page 1869, "Major challenges and future directions" section: Since there are (or will be) several fully-coupled chemistry and meteorology models available, it would be useful to add a few words on module and model intercomparison that needs to be done in the future to see how they perform. As has been done in the past, it would be extremely useful to intercompare some of the new aerosol modules that have been implemented in WRF-chem and other 3-D host models, for example. Wherever applicable, the intercomparison must be first done with the box-model versions of these modules so that the results can be easily understood and evaluated against the available benchmarks (e.g., AIM for thermodynamics, LSODE for mass transfer, etc.). The same modules should then be evaluated in the fully-coupled 3-D models to examine their performance and CPU time requirements. Without such a carefully designed intercomparison exercise in the future, it will become very difficult or impossible to figure out how these different modules and models are performing and what the sources and magnitudes of uncertainties in their predictions are?

Reply: This is a very good point, which will be incorporated into the revised version of the paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 1833, 2008.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper