Atmos. Chem. Phys. Discuss., 8, S1078–S1083, 2008
www.atmos-chem-phys-discuss.net/8/S1078/2008/

Atmospheric
Chemistry
and Physics
Discussions

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

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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

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subtle but important details.

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

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.

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 State Univ., Blacksburg, Va.

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

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 EQUI-

SOLV II when RH $<$ MDRH.

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.

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 activity 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.

Page 1858, line 10: This statement is not accurate. MESA currently includes solid-liquid 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

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MDRH(T) parameterizations 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.

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.

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 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."

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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.

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.

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?

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

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