EVALUATION AND IMPROVEMENT OF GAS/PARTICLE MASS TRANSFER TREATMENTS FOR THREE-DIMENSIONAL AEROSOL SIMULATION AND FORECAST

Xiaoming Hu and Yang Zhang*
North Carolina State University, Raleigh, NC 27695, USA

Mark Z. Jacobson
Stanford University, Stanford, CA 94305, USA

1. INTRODUCTION

Modeling the size/composition distribution of atmospheric aerosols is important in assessing the impacts of human activities on air quality and climate change. Simulating gas/particle mass transfer is essential for accurately predicting the aerosol size/composition distribution since secondary aerosol accounts for a significant amount in the total aerosol mass, in some cases more than 90% to the $PM_{2.5}$ fraction (Plessow et al., 2005). Its treatment in three-dimensional (3-D) air quality models (AQMs), however, represents one of the major challenges in AQMs.

Three main approaches (i.e., equilibrium, kinetic, and hybrid) have been used to describe gas/particle mass transfer in AQMs. The equilibrium approach is commonly used in the first generation aerosol models (e.g., Russell et al. 1983; Pilinis et al. 1987; Binkowski and Shankar, 1995; Lurmann et al., 1997) because of its computational efficiency. These models either ignore the size by assuming the equilibrium for the bulk aerosol phase or redistribute the mass changes over the size sections using weighting factors (e.g., Pandis et al., 1993; Lumann et al., 1997; Capaldo et al., 2000; Zhang et al., 2004) following the bulk equilibrium treatment to diagnoze size/composition distribution. Both methods neglect the differences in the chemical driving forces for different aerosol sections by assuming the same chemical composition for all the particles (i.e., internal mixture). The mixing of acidic particles with alkaline particles in the bulk equilibrium approach may introduce errors (Ansari and Pandis, 1999; Moya et al., 2001). In addition, the bulk equilibrium approach relies on the instantaneous bulk equilibrium assumption that may be invalid under some atmospheric conditions. The kinetic approach simulates explicit mass transfer for each section (e.g., Meng and

Seinfeld 1996; Jacobson 1997a, b; Meng et al., 1998; Sun and Wexler 1998; Zhang et al., 1999; Pilinis et al., 2000). Since no assumptions are made and the chemical driving force can be differentiated for each section, this approach provides the most accurate solution when an appropriate numerical solver is used. However, its computational demands limit its application in 3-D AQMs. The hybrid approach compromises the accuracy and efficiency by using the equilibrium approach for fine particles and the kinetic approach for coarse particles but it has only been tested with limited episodes (Capoldo et al., 2000; Koo et al., 2003; Gaydos et al., 2003). The lack of an efficient yet accurate gas/particle kinetic mass transfer treatment in 3-D AQMs warrants its development, improvement, and evaluation.

In this work, the aforementioned three gas/particle mass transfer approaches are developed, tested, and improved in a box model (i.e., the model of aerosol dynamics, reaction, ionization and dissolution (MADRID) (Zhang et al., 2004)) and then incorporated into WRF/Chem. Some preliminary results from the 3-D applications of the improved aerosol treatments in WRF/Chem are presented.

2. SUMMARY

The box MADRID tests have shown that the CIT bulk equilibrium approaches failed to predict the distribution of semi-volatile species (e.g., ammonium, nitrate) when the compositions of different sections are different due to its assumptions of internal mixture and bulk equilibrium and the larger weighting factor for fine sections for mass redistribution. The CMU hybrid approach also has the same problem since it assumes bulk equilibrium for the first 6 sections. The kinetic approaches (including the analytical predictor of condensation (APC) scheme, the variable coefficient ODE (DVODE) solver, and Walcek schemes for the condensation/ evaporation equations) predict similar solutions.

Among all approaches tested, the CIT bulk equilibrium approach is the most computationally

---

*Corresponding author: Yang Zhang, Department Of Marine, Earth and Atmospheric Sciences, Campus Box 8208, NCSU, Raleigh, NC 27606 e-mail: yang_zhang@ncsu.edu

efficient one. The kinetic approach using the APC scheme is very attractive for 3-D applications in terms of both accuracy and computational efficiency.

The three mass transfer approaches are implemented into WRF/Chem-MADRID and further tested for the 3-D application over the TEXAQS-2000 testbed. The preliminary results have shown that WRF/Chem-MADRID with the kinetic mass transfer approach predicts less fine mode nitrate and more coarse mode nitrate than equilibrium mass transfer approach for the TEXAQS-2000 episode.

3. ACKNOWLEDGEMENTS

4. REFERENCES

Ansari, A. S., and S. N. Pandis (1999), An analysis of four models predicting the partitioning of semi-volatile inorganic aerosol components. *Aerosol Sci. Technol.,* 31, 129-153.

Binkowski, F. S., and U. Shankar (1995), The Regional Particulate Matter Model. 1: Model Description and Preliminary Results, *J. Geophys. Res.,* 100, 26,191-26,209.

Capaldo, K. P., Pilinis, C. and S. N. Pandis (2000), A computationally efficient hybrid approach for dynamic gas/aerosol transfer in air quality models. *Atmos. Environ.,* 34, 3617–3627.

Jacobson, M. Z. (1997a), Development and application of a new air pollution modeling system, part II, Aerosol module structure and design, *Atmos. Environ.*, 31, 131-144.

Jacobson, M. Z. (1997b), Numerical techniques to solve condensational and dissolutional growth equations when growth is coupled to reversible reactions, *Aerosol Sci. Technol.*, 27, 491-498.

Lurmann, F. W., A. S. Wexler, S. N. Pandis, S. Musarra, N. Kumar, and J. H. Seinfeld (1997), Modelling Urban and Regional Aerosols-II. Application to California's South Coast Air Basin, *Atmos. Environ.,* 31, 2695-2715.

Meng, Z., and J. H. Seinfeld (1996), Time scales to achieve atmospheric gas–aerosol equilibrium for volatile species. *Atmos. Environ.,* 30, 2889–2900.

Meng, Z., D. Dabdub, and J. H. Seinfeld (1998), Size-Resolved and Chemically Resolved Model of Atmospheric Aerosol Dynamics, *J. Geophys. Res.,* 103, 3419-3435.

Moya, M., A. S. Ansari, and S. N. Pandis (2001), Partitioning of nitrate and ammonium between the gas and particulate phases during the 1997 IMADA-AVER study in Mexico City, *Atmos. Environ.,* 35, 1791–1804.

Pandis, S. N., A. S. Wexler, and J. H. Seinfeld (1993), Secondary organic aerosol formation and transport—II Predicting the ambient secondary organic aerosol size distribution. *Atmos. Environ.,* 27A, 2403–2416.

Pilinis, C., J. H. Seinfeld, and C. Seigneur (1987), Mathematical Modeling of the Dynamics of Multicomponent Atmospheric Aerosols, *Atmos. Environ.,* 21, 943–955.

Pilinis, C., K. P. Capaldo, A. Nenes, and S. N. Pandis (2000), MADM-A new multicomponent aerosol dynamics model. *Aerosol Sci. Technol.*, 32, 482-502.

Plessow, K., G. Spindler, F. Zimmermann, and J. Matschullat (2005), Seasonal variations and interactions of N-containing gases and particles over a coniferous forest, Saxony, Germany, *Atmos. Environ.*, 39, 6995–7007.

Russell, A. G., G. J. McRae, and G. R. Cass (1983), Mathematical modeling of the formation and transport of ammonium nitrate aerosol. *Atmos. Environ.,* 17, 949-965.

Sun, Q., and A. S. Wexler (1998), Modeling Urban and Regional Aerosols-Condensation and Evaporation near Acid Neutrality, *Atmos. Environ.,* 32, 3527-3531.

Zhang, Y., B. Pun, K. Vijayaraghavan, S.-Y. Wu, C. Seigneur, S. Pandis, M. Jacobson, A. Nenes, and J. H. Seinfeld (2004), Development and application of the model of aerosol dynamics, reaction, ionization and dissolution (MADRID), *J. Geophys. Res.*, 109, D01202, doi:10.1029/2003JD003501.

Zhang, Y., C. Seigneur, J. H. Seinfeld, M. Jacobson, and F. S. Binkowski (1999),

Simulation of aerosol dynamics: a comparative review of algorithms used in air quality models, *Aerosol Sci. Technol.,* 31(6), 487-514.