Implementation of a Dynamically Interactive Coarse Particle Mode in CMAQv4.7

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Background & Motivation

- Chemical interactions between coarse particles and gas-phase pollutants were not treated in previous versions of CMAQ.
- The time scale for coarse particles to reach equilibrium with the gas phase is on the order of several hours, so a dynamic treatment of gas-to-particle mass transfer is necessary. An assumption of instantaneous equilibrium would be invalid.
- There are several motives to simulate such interactions in CMAQ v4.7.
  - Coarse particles provide additional surface area for condensation of H2SO4 and NH3, thereby reducing the availability of these gases to form fine-particle SO4 and NH4. Modelled concentrations of PM2.5 should decline in dual-rich regions.
  - Replacement of Cl in coarse sea-salt particles by NO3 reduces the total nitrate pool available for condensation on fine particles. Thus, modelled concentrations of fine particle NO3 in coastal urban areas may decline.
  - Over water bodies, coarse-particle NO3 deposits faster than gas-phase HNO3. Thus, treating the condensation of HNO3 on coarse particles should increase the modeled deposition of nitrogen to coastal ecosystems.
- In future CMAQ releases, changes in sea-salt Cl may enhance photochemical O3 formation and Hg oxidation. An assumption of instantaneous equilibrium would be invalid.

Box Model Testing

- Test conditions:
  - Simulate transport of a marine air mass into a polluted urban area. Over a 38-hour period, air mass encounters range of RH, aerosol pH, and pollutant concentrations.
  - Adapted from conditions used to develop sectional aerosol modules in CMAQ-MADRID and PM-CAMx.
- Compare CMAQ v4.7 treatment with 2 "reference" modules described in the literature:
  - A multi-component aerosol dynamics module (MADM) with 10 sections
  - A modular aerosol module (MAM) with fully-dynamic mass transfer

Model Evaluation at Coastal Locations

Figure 1. Schematic of inorganic gas/particle partitioning treatment in CMAQ v4.7. Newly added species and pathways are shown in RED.

Figure 2 (right). Treatment in CMAQ v4.7 reasonably reproduces the NO3 distribution from reference modules. Prior versions of CMAQ yielded no coarse NO3.

Figure 3. Effect of model revisions on monthly-averaged PM2.5 during August 2006. Lower modeled values of PM2.5 are largely explained by changes in SO4 and NH4 which now are allowed to condense on the coarse mode. Shifting mass to the coarse particles increases the dry deposition of sulfate and reduced nitrogen. During winter (not shown), impacts on PM2.5 are less pronounced due to lower ambient SO4 concentrations.

Figure 4. Effect of model revisions on monthly-averaged nitrate distribution during January 2006. Over the ocean, coarse-particle NO3 increases at the expense of HNO3. Shifting nitrate from the gas phase to the coarse particles will increase dry deposition of nitrogen in coastal ecosystems. During summer (not shown), a similar effect was found.

Figure 5. Evaluation of past and present model configurations against measurements of (a) bulk gas and particulate NO3, (b) fine and coarse NO3, and (c) highly size-resolved SO4 and NO3. Revised model treatment consistently outperforms the previous version of CMAQ.

Acknowledgements

Special thanks to Eric Edgerton for providing the SEARCH measurements and to Sharon Roselle, Wyatt Pinder, Rohit Mathur, Alice Gilliland, Jon Pleim, Ken Schere, and staff at the Computer Sciences Corporation, for helping conduct simulations and analyze output from incremental model tests preceding the CMAQ v4.7 release.