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

James T. Kelly [a,b], Prakash V. Bhave [a], Uma Shankar [c], Kristen M. Foley [a], and Chris G. Nolte [a]

[a] U.S. Environmental Protection Agency, Office of Research and Development, National Exposure Research Laboratory, Atmospheric Modeling Division
[b] Now at the California Air Resources Board, Planning and Technical Support Division, Regional Air Quality Modeling Section
[c] Institute for the Environment, University of North Carolina at Chapel Hill

Address correspondence to Prakash Bhave **l** bhave.prakash@epa.gov **l** 919-541-2194

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 H_2SO_4 and NH_3, thereby reducing the availability of these gases to form fine-particle SO_4^{2-} and NH_4^+. Modeled concentrations of $PM_{2.5}$ should decline in dust-rich regions.
  - Replacement of Cl^- in coarse sea-salt particles by NO_3^- reduces the total nitrate pool available for condensation on fine particles. Thus, modeled concentrations of fine particle NO_3^- in coastal urban areas may decline.
  - Over water bodies, coarse-particle NO_3^- deposits faster than gas-phase HNO_3. Thus, treating the condensation of HNO_3 on coarse particles should increase the modeled deposition of nitrogen to coastal ecosystems.
- In future CMAQ releases,
  - … chlorine-containing molecules released from coarse sea-salt particles to the gas phase may enhance photochemical O_3 formation and Hg oxidation.
  - … the aerosol thermodynamic module will be updated to treat crustal cations such as Mg^{2+}, K^+, and Ca^{2+}. These ions can be abundant in coarse PM, so a framework will be needed through which they can interact with the gas phase.
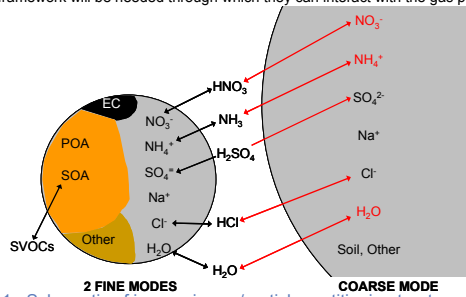

Figure 1. Schematic of inorganic gas/particle partitioning treatment in CMAQ v4.7. Newly added species and pathways are shown in RED.

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
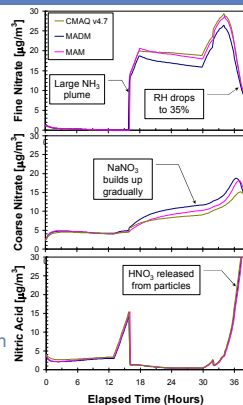 - a modal aerosol module (MAM) with fully-dynamic mass transfer

Figure 2 (right). Treatment in CMAQ v4.7 reasonably reproduces the NO_3^- distribution from reference modules. Prior versions of CMAQ yielded no coarse NO_3^-.
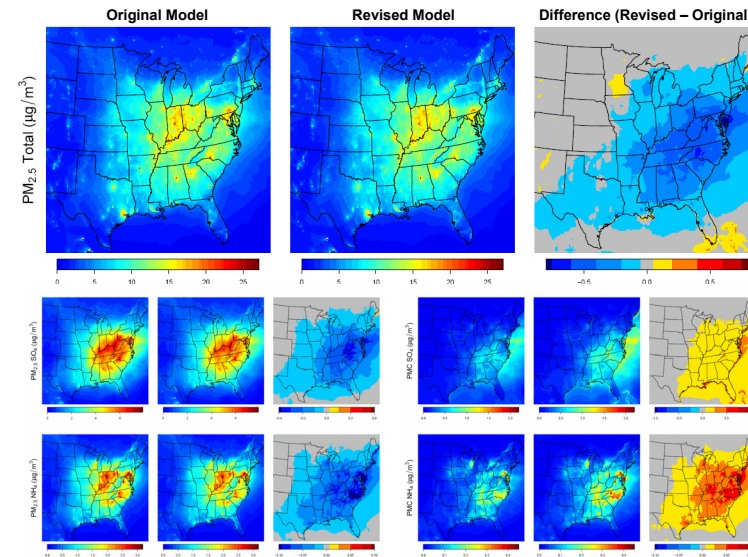
Impacts on CMAQ Model Output

Figure 3. Effect of model revisions on monthly-averaged $PM_{2.5}$ during August 2006. Lower modeled values of $PM_{2.5}$ are largely explained by changes in SO_4 and NH_4, 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 $PM_{2.5}$ are less pronounced due to lower ambient SO_4 concentrations.
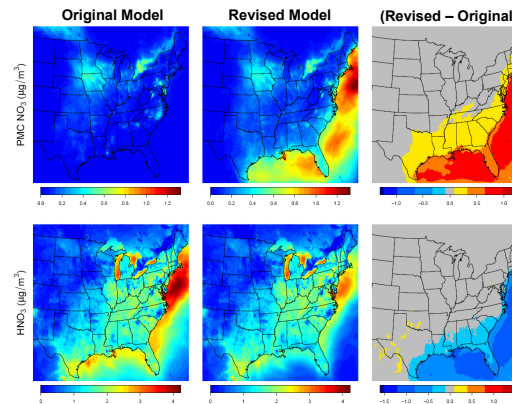

Figure 4. Effect of model revisions on monthly-averaged nitrate distribution during January 2006. Over the ocean, coarse-particle NO_3^- increases at the expense of HNO_3. Shifting nitrate from the gas phase to the coarse particles will increase dry deposition of nitrogen in coastal ecosystems. During summer (not shown), a similar effect was found.
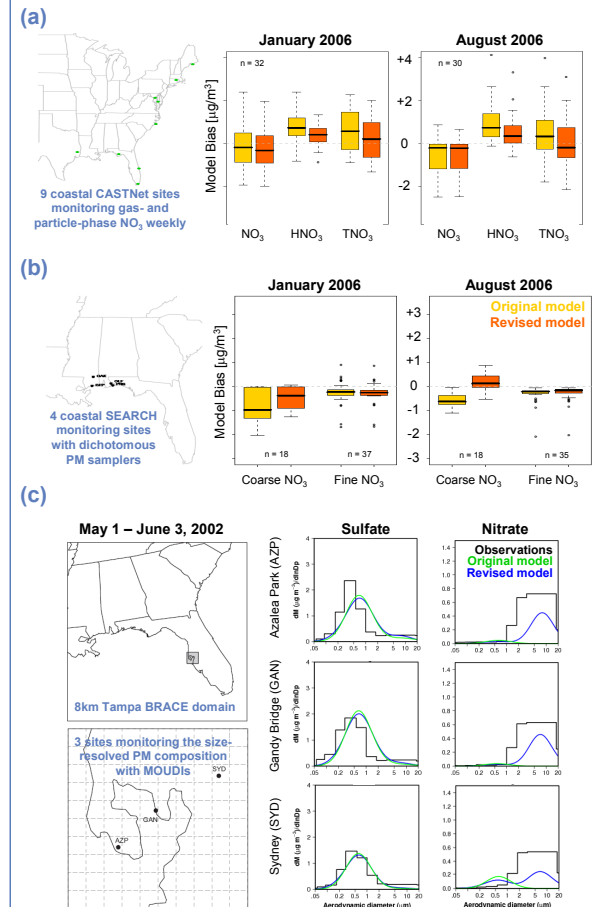
Model Evaluation at Coastal Locations

(a)

9 coastal CASTNet sites monitoring gas- and particle-phase NO_3 weekly

(b)

4 coastal SEARCH monitoring sites with dichotomous PM samplers

(c)

May 1 – June 3, 2002

8km Tampa BRACE domain

3 sites monitoring the size-resolved PM composition with MOUDIs

Figure 5. Evaluation of past and present model configurations against measurements of (a) bulk gas and particulate NO_3, (b) fine and coarse NO_3, and (c) highly size-resolved SO_4 and NO_3. Revised model treatment consistently outperforms the previous version of CMAQ.