Comparing Modal and Sectional Approaches in Modeling Particulate Matter in Northern California

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1. MODEL AND EPISODE DESCRIPTIONS

1.1. Modified CMAQ

The Community Multiscale Air Quality (CMAQ) Model released by US EPA uses three log-normal modes to represent particle size distributions. The gas-particle partitioning of organic and inorganic components of PM is treated as in bulk thermodynamic equilibrium, which enables the aerosol module to run relatively fast, but could result in positive bias towards the particulate phase (Zhang et al., 2000). The USEPA CMAQ model was improved previously to incorporate more California specific information and minor bug fixes (Liang and Kaduwela, 2005).

1.2. CMAQ-UCD

The Community Multiscale Air Quality Model with the UC-Davis Aerosol Module (CMAQ-UCD) uses Models-3/CMAQ as the host gas-phase model and incorporates a mechanistic, fully dynamic, internally-mixed, sectional aerosol module, which evolved from the Aerosol Inorganic Model and related work (Wexler and Seinfeld, 1990; Wexler et al., 1994; Potukuchi and Wexler, 1995ab; Sun and Wexler, 1998ab). CMAQ-UCD uses nine discrete size bins, with Stoke's diameter ranging from 0.04 micron to 20 micron. CMAQ-UCD employs three gas-to-particle transport schemes, Replacement, Coupled and Uncoupled transport, as appropriate, avoiding substantial numerical stiffness due to rapid pH changes. It adopts a simplified aerosol thermodynamics scheme, able to predict particle phase states and water contents quickly and reasonably. The integration is accomplished using a computationally efficient asynchronous time-stepping (ATS) method, where particles in different sizes integrate with different time scales (Zhang and Wexler, 2005ab). Model improvements made previously to the USEPA CMAQ model (Liang and Kaduwela, 2005) were also implemented in CMAQ-UCD.

1.3. CRPAQS Domain and Model Episode

An extended winter PM episode (December 25, 2000-January 7, 2001) was captured in the San Joaquin Valley during the California Regional PM10/PM2.5 Air Quality Study (CRPAQS). The model period contains eight additional days, from December 17 2000 to December 24 2000, to allow for spin-up. The model domain, which was designed to include central and northern California, extends from the northern Sacramento Valley to the Tehachapi Mountains, and from the Pacific Ocean to the southeast desert. The model contains 185×185 horizontal grid of 4km × 4km size, and 15 expanding vertical layers (Liang and Kaduwela, 2005).

We inter-compare the model results for the concentrations of gaseous species and PM2.5 compositions at core stations and over the model domain. This paper mainly presents the comparisons at three anchor sites, namely, Angiola (ANGI), Bakersfield (BAC) and Fresno (FSF). ANGI is a rural site; BAC and FSF are two urban sites. Note the all concentrations presented in this paper are daily averaged values.

2. GAS-PHASE SPECIES

Figures 1 and 2 compare major gas-phase species concentrations (CO, SO_2, NO, NO_2, O_3, NH_3 and HNO_3) at site BAC between CMAQ and CMAQ-UCD. The concentrations of less reactive species such as CO and SO_2 from two models are almost identical, while agreements for more reactive species such NO, NO_2 and O_3 are less perfect, but still excellent. These results are expected since both models use the same advection, diffusion and gas-chemistry mechanisms.

It is interesting to make a peer-to-peer comparison between the NH_3 and HNO_3, the two soluble gas-phase species. First, the modeled NH_3

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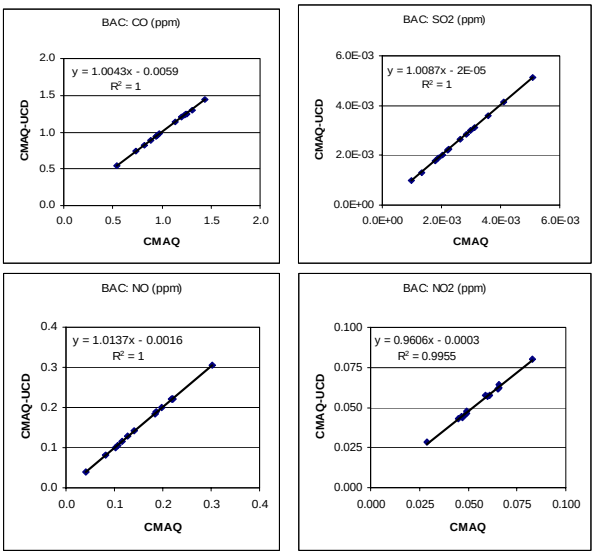
concentrations are about two magnitudes higher than HNO_3 concentrations. Second, while the agreement between NH_3 is excellent, there are large discrepancies in HNO_3 concentrations between the two models. Due to the considerable amount of ammonia emission from animal operations in the San Joaquin Valley, NH_3 was in excess during the CRPAQS episode, so condensation of NH_3 and HNO_3 to form ammonium nitrate (NH_4NO_3) was limited by the avilability of HNO_3. As a result, most HNO_3 partitioned into the particle phase, leaving only trace amount in the gas phase. Since the absolute values of HNO_3 concentrations are very low, relative errors are large but absolute errors are small.

Results for Fresno and Bakersfield are similar so we will only show results for Bakersfield as our characteristic urban site.

Since ANGI is a rural site, primary gas-phase species, such as NO and NO_2, were transported from urban areas. Since discrepancies between the two models could accumulate during transport process, the agreements at ANGI are generally not as good as those at the two urban sites. Nevertheless, as shown in Figure 3, the two models still closely match each other.



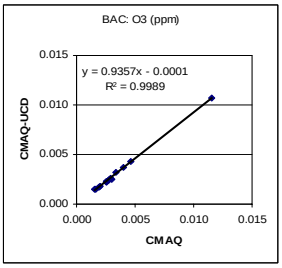


Figure 1. Concentrations of gas-phase species, CO, SO_2, NO, NO_2 and O_3, at site BAC in CMAQ vs. CMAQ-UCD.

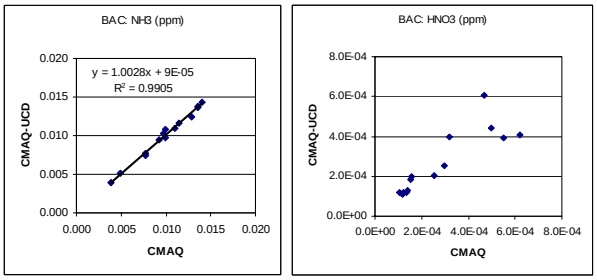


Figure 2. Concentrations of NH_3 and HNO_3 at site BAC in CMAQ vs. CMAQ-UCD.

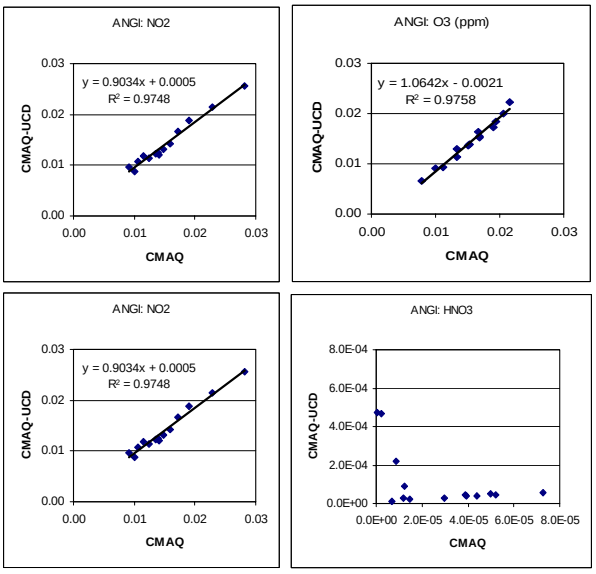


Figure 3. Concentrations of NO_2, O3, NH_3 and HNO_3 at site ANGI in CMAQ vs. CMAQ-UCD.

3. PRIMARY PARTICULATE SPECIES

Next, we compare the particulate-phase species, first evaluating the primary species and then the secondary species. During the CRPAQS

2000-2001 winter episode, photochemical activity was relatively weak and the formation of secondary organic aerosols was negligible. Therefore, the organic compositions were dominated by primary organic emissions. OC and EC are two major primary particulate species. Although sulfate is not a primary species, its vapor pressure is so low that it is partitioned completely to the particle phase. Figure 4 depicts excellent agreements for these three species between the two models at BAC, similar to the agreements at FSF and ANGI (not shown).

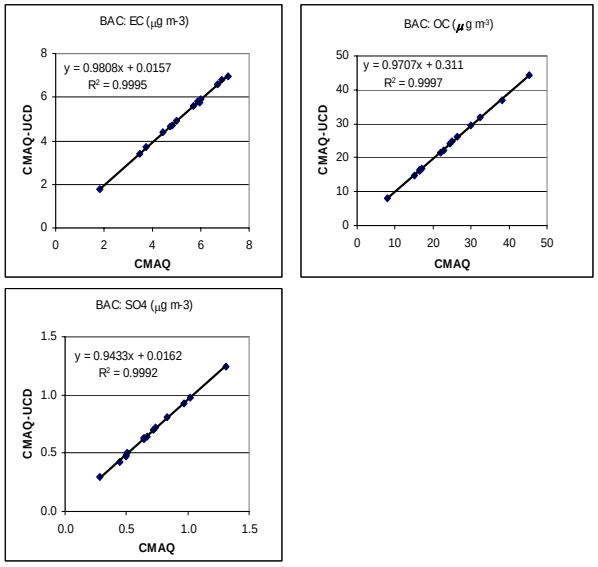


Figure 4. Concentrations of two PM2.5 primary components (OC and EC) and SO_4 at site BAC in CMAQ vs. CMAQ-UCD.

4. SECONDAY PARTICULATE SPECIES

Modeling secondary species is one of the most challenging tasks for aerosol models. Nitrate is the most complicated species to model since it is affected by photochemistry, heterogeneous chemistry, and aerosol thermodynamics. During this episode, ammonium nitrate was the dominant component of PM2.5, and modeling results suggest the heterogeneous formation via N_2O_5 to be the major formation pathway.

Figures 5 to 7 illustrate the comparisons of NH_4 and NO_3 in PM2.5 at the three anchor sites. The agreements are very good. CMAQ-UCD systematically predicts lower NH_4 and NO_3 than CMAQ does, mainly because CMAQ-UCD employs dynamic partitioning between the gas phase and particle phase. In this approach, equilibrium is sometimes limited by gas-particle transport, especially when surface area loading is low. In contrast, CMAQ assumes equilibrium under all conditions.

In contrast to the two urban sites, at ANGI CMAQ-UCD predicted slightly higher NH4 and NO3 than CMAQ, likely due to the transport nature in this rural site.

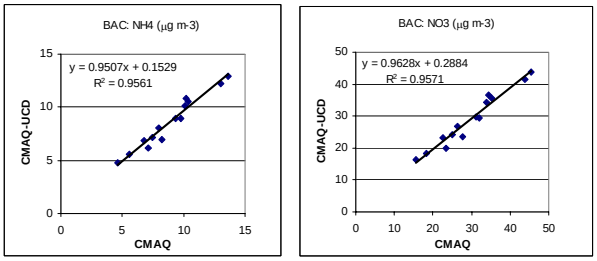


Figure 5. Concentrations of PM2.5 NH_4 and NO_3 at site BAC in CMAQ vs. CMAQ-UCD.

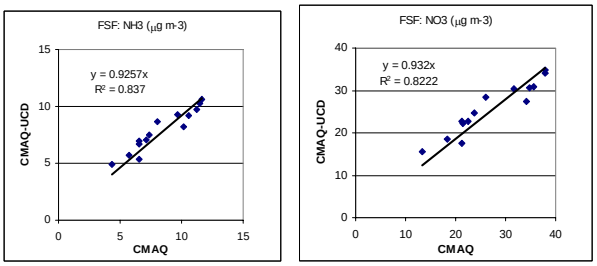


Figure 6. Concentrations of PM2.5 NH_4 and NO_3 at site FSF in CMAQ vs. CMAQ-UCD.

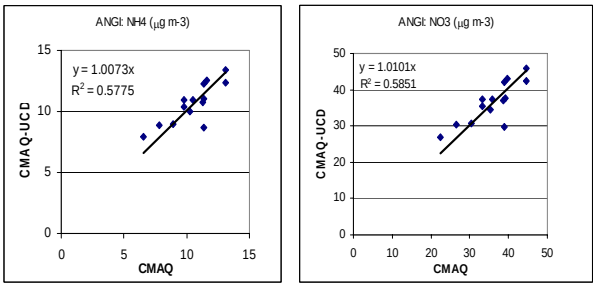


Figure 7. Concentrations of PM2.5 NH_4 and NO_3 at site ANGI in CMAQ vs. CMAQ-UCD.

5. PARTICLE SIZE DISTRIBUTIONS

Comparisons of particle size distributions between the two models pose some challenges. While CMAQ-UCD represents size distribution in

discrete size sections, a rather straightforward manner, the modal approach in CMAQ solves the changes in number and surface area for three log-normal modes. The original code released by USEPA assumed constant values for geometric mean diameter and standard deviation from emissions. A modified version of CMAQ (Liang and Kaduwela, 2005) allows these two parameters to be user inputs, which gives the model more flexibility. However, the CMAQ model assumes the three modes to be always log-normal in the updating of PM number and surface area, and the comparison is only meaningful for dry conditions. Comparison of size distributions is thus beyond the scope of this work.

6. CPU TIMES

CMAQ takes about 2.5 CPU hours on 15 3-GHz processors (of a 16-node Linux cluster) to simulate a day during the CRPAQS winter episode and CMAQ-UCD takes about 10 hours on a similar 16-node Linux cluster.

7. CONCLUSIONS

We compared the simulation results, for the CRPAQS 2000-2001 winter episode, of two air quality models, CMAQ and CMAQ-UCD. While CMAQ adopts the modal approach in representing particle size distributions, CMAQ-UCD employs a sectional approach. The preliminary comparisons showed excellent agreements in the gas-phase species concentrations. The agreements in primary PM2.5 components were also satisfactory. CMAQ-UCD systematically predicted slightly lower/higher secondary aerosol species concentrations, i.e., NH_4 and NO_3, at urban/rural sites. These phenomena may be explained by the differences in the partitioning methods used in the two models, i.e., dynamic (CMAQ-UCD) vs. equilibrium (CMAQ), together with transport processes. Further investigations are needed on how to compare the size distributions between the two models.

DISCLAIMER

This paper has been reviewed by the staff of the California Air Resources Board and has been approved for publication. Approval does not signify that the contents necessarily reflect the views and policies of the California Air Resources Board, nor does mention of trade names or commercial products constitute endorsement or recommendation for use.

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