5.3 EVALUATION OF MODELS-3/CMAQ PERFORMANCE DURING A WINTER HIGH PARTICULATE MATTER EPISODE IN FEBRUARY 1998

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1. INTRODUCTION

Winter episodes of high particulate matter (PM) concentrations over the Eastern North America are about four times less frequent than summer episodes (Brook et al. 2001) and possibly for this reason have not attracted much attention from researchers and policy makers. The meteorology and chemistry of these episodes is much different from those of the summer episodes, where long-range transport usually plays a more significant role. Also, secondary aerosols are dominated by sulfates in the summer, but by nitrates in the winter. In this study we evaluate the performance of the U.S. Environmental Protection Agency's (EPA's) model, Models-3, in simulating a winter episode which occurred between February 8 and February 12 1998. For a more comprehensive assessment of the performance of the chemical model we extended the evaluation to include the period from February 5 to February 14 1998. During this time an extensive set of measurements was gathered from a variety of networks.

2. DESCRIPTION OF THE EPISODE

2.1 METEOROLOGY

The episode occurred during stagnant atmospheric conditions caused by an extensive high pressure system over Ontario, Quebec and the North-Eastern U.S. During the episode, the surface temperatures remained above $0°$C and no precipitation was observed. Increased static stability in the lower troposphere was enhanced by the advection of warm air over the continent above the boundary layer. The episode ended with the passage of a cold front on February 13.

2.1 AEROSOL CHEMISTRY

Hourly and daily concentrations of PM2.5 and PM10 (i.e. aerosols with diameters smaller than 2.5 μm and 10 μm respectively) as well as speciated aerosols (daily data only) were measured by an extensive network of stations including AIRS, NAPS, GAVM, CAPMON and IMPROVE. In the U.S., 24-hour average concentrations in the Syracuse reached 107 μg m^{-3} and 135 μg m^{-3} in Cleveland area. In Canada, a site near Montreal recorded 138 μg m^{-3} for PM10 and 28 μg m^{-3} for NO_3^- on February 11. Analysis of the data showed that high concentrations of the aerosols existed throughout the eastern part of the continent, but very low concentrations often occurred in the proximity of high values. As winds were weak this would suggest that locally emitted pollutants dominated those from long range transport.

3. MODELING

In this study we use the Penn. State/NCAR meteorological model, MM5, and the U.S. EPA's Models-3 system. The latter consists of two components: the MEPPS (or SMOKE) emissions processor and the chemical transport model CMAQ. The design of the numerical simulations is described below.

3.1 METEOROLOGY

Full MM5 documentation can be found at www.ncar.edu/mm5/mm5-home.html. The modeling domain (Fig. 1) included 64×58 grid points spaced at 36 km with 30 vertical levels stretching from the surface to 100 hPa. Parameterization of the boundary layer physics was done using the Blackadar scheme (Blackadar 1979). The meteorological input to the air quality model for each day was based on a 36-hour model simulation with the first 12 hours being used as a spin-up period. NCEP reanalyses were used to specify boundary and initial conditions.

3.2 CHEMISTRY

Descriptions of emission processing with MEPPS and the chemical transport model CMAQ can be found in Byun and Ching (1999). Prior to carrying out the

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modeling study, the Canadian portion of the emission inventory originally supplied in the Models-3 database was significantly improved. The improvement included:

- annual average inventories for point, area and mobile sources for 1995, obtained from Environment Canada,
- Canadian temporal allocation factors,
- information on Canadian distribution of crop and forest species,
- province-specific data in eastern Canada used in MOBILE5a,
- higher resolution population and dwelling data by census enumeration,
- PM10, PM2.5 and SO_2 mobile emissions calculated using province specific information.

It should be noted that the three major uncertainties in the emissions calculations exist in Models-3. They are:

- partitioning of primary particulate matter into sizes and chemical components such as elemental carbon, organic carbon, soil-related and sea salt are oversimplified,
- fugitive dust emissions are overestimated in the inventory; as recommended by Watson and Chow (1999), the emissions could be reduced by 75% from its current values (this was not attempted in this study),
- ammonia emission rates have large uncertainties. This is especially important in winter when nitrates constitute a large portion of the aerosol mass and are sensitive to the ammonia available. Since the seasonal variation of ammonia emissions rates is not accounted for in the model, it is likely that the emissions are significantly overestimated in winter.

Compared to the MM5 domain, the domain of the chemical model was reduced by 6 points on each side. 15 vertical levels were employed. The RADM2 chemical mechanism with aerosol and aqueous chemistry and a 4-product isoprene reaction set was used. No modifications to the original CMAQ setup with respect to boundary and initial conditions were attempted. QSSA solver was used.

4. CMAQ PERFORMANCE EVALUATION

4.1 METHODOLOGY

As previously noted, extensive measurements of aerosol concentration were made by several networks in the US and Canada during both episodes. Different measurement techniques of the species resulted in an inhomogeneous dataset that required re-processing for comparison with the model. Those concentrations that were obtained for standard atmospheric conditions (i.e. at pressure 1013.25 hPa and temperature 0° C) were converted to concentrations at local ambient conditions using the local air density from the meteorological model output. Concentrations of all gas species were converted to ppb (ppm for CO) where required.

Evaluation of the model was performed in a domain reduced by three cells from each lateral boundary. Locations of the sites where daily PM10 concentrations were measured and which were used in the model evaluation are shown in Fig. 1. The model was allowed a "spin-up" period of 2 days, during which the results were not analyzed or used in the evaluation. For comparison with daily measurements, model output was averaged over 24 hour period. Gridded model output was interpolated bi-linearly to the locations where measurements were taken.

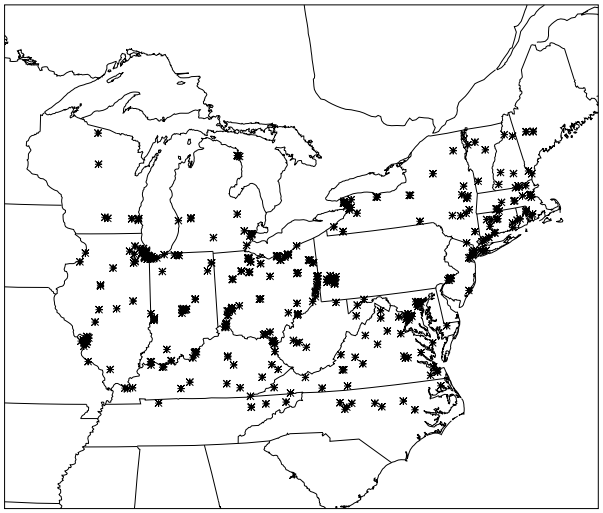

Figure 1: Approximate MM5 domain with locations where daily PM10 concentrations were measured.

4.2 RESULTS AND DISCUSSION

Scatter plots of total carbon (elemental plus organic), HNO_3, NH_4^-, NO_3^-, PM10, PM2.5, SO_2 and SO_4^{-2} are shown in Figure 2. It can be seen that the model overestimates the amount of nitrate ion; underestimates the concentration of the sulfate ion and simulates the concentration of the ammonium ion reasonably well. It should be noted that the model does not have a mechanism for the reaction of nitric acid with soil dust, which would normally reduce the concentration of the nitric acid and hence lower the nitrate concentration. This could account for the above bias in NO_3^-. Other factors causing the nitrates overestimation could be linked to overestimation of ammonia emissions. As for aerosol, total PM10 and PM2.5 are slightly overestimated. Further sensitivity test runs indicate that improved statistics for all the species were obtained when the emission of ammonia was reduced by 50% (Fig. 3). Time series of PM10 and PM2.5 concentrations plotted for randomly chosen locations (50% ammonia, Fig. 4) show that the model was able to simulate correctly the temporal variability present in the measurements.

5. CONCLUSIONS

The application of Models-3 to simulate aerosol con-

centrations during a high particulate episode in February 1998 and its comparison with an extensive set of measurements elucidated several strengths and deficiencies of the system. The simulations showed that while the total PM10 and PM2.5 mass appears to be predicted accurately, the modeled aerosol species show biases compared to the observations. This would suggest that the model requires further improvement before being used for regulatory purposes.

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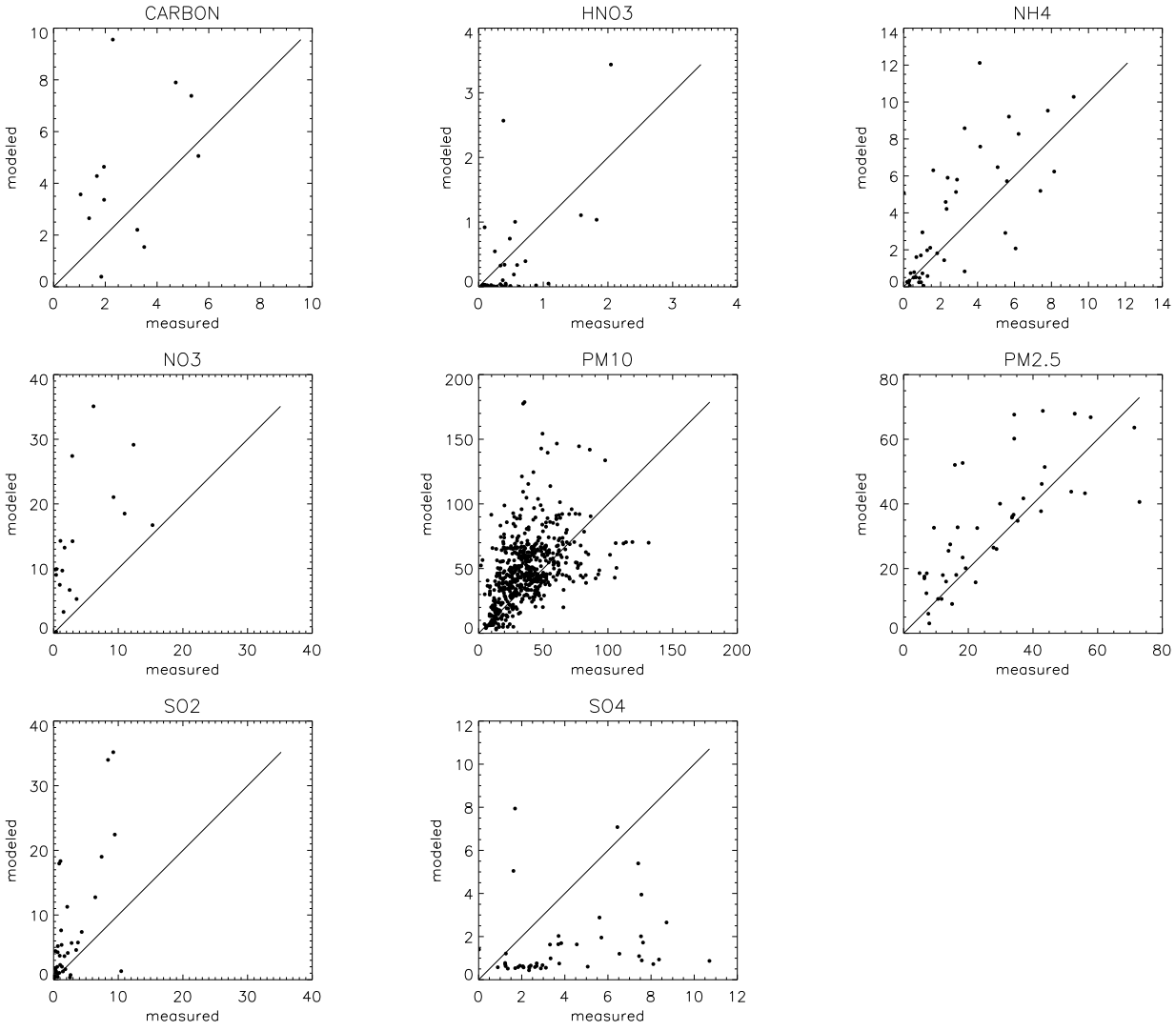

Figure 2 Scatter plots of observed and modeled concentrations. Aerosols in μg m^{-3}, gases in ppb.

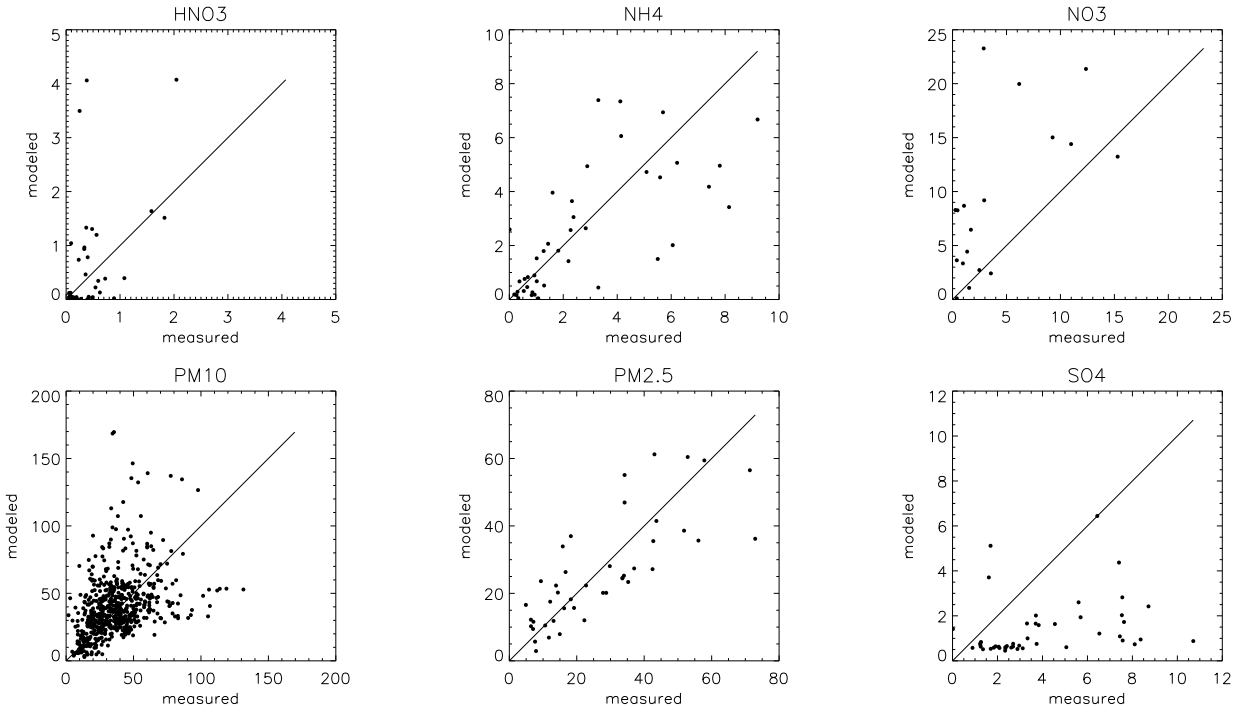

Figure 3: As in Figure 2 but for 50% ammonia emissions.

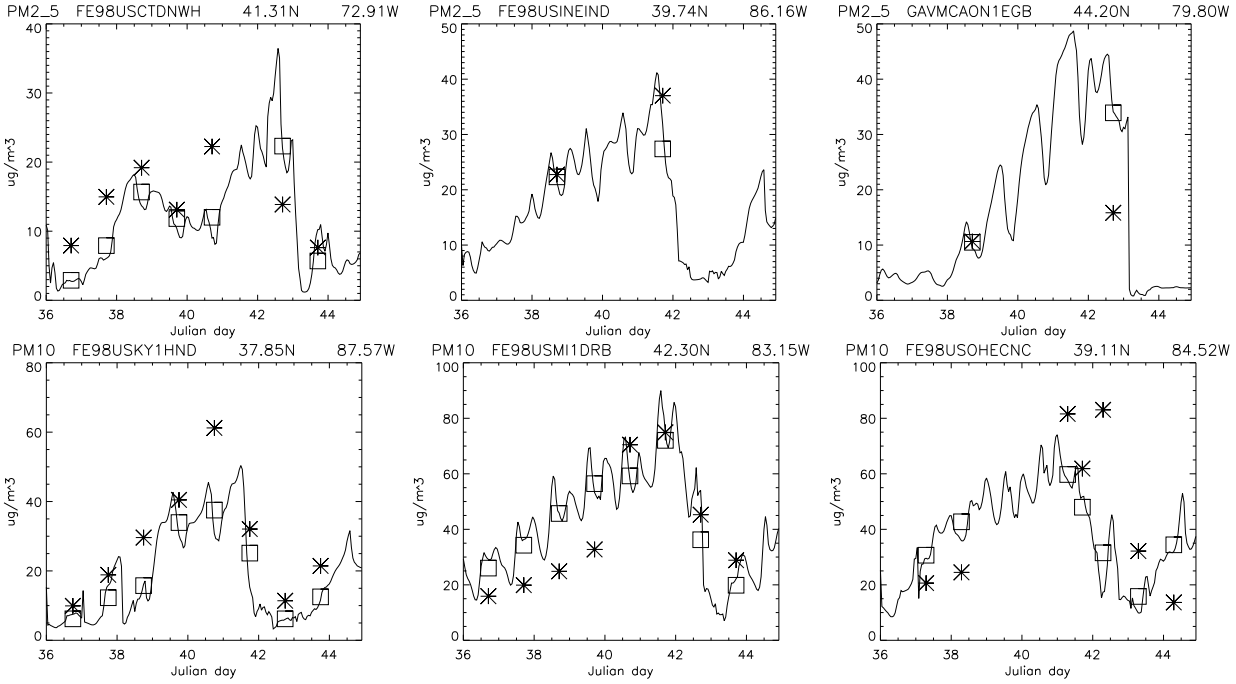

Figure 4: Comparison of modeled and observed PM2.5 and PM10 concentrations at specific locations. Stars denote measurements, squares 24-hour model averages.