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Assessment of the wintertime performance of developmental particulate matter forecasts with the Eta-Community Multiscale Air Quality modeling system

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[1] It is desirable for local air quality agencies to accurately forecast tropospheric $PM_{2.5}$ concentrations to alert the sensitive population of the onset, severity, and duration of unhealthy air and to encourage the public and industry to reduce emissions-producing activities. Since elevated particulate matter concentrations are encountered throughout the year, the accurate forecast of the day-to-day variability in $PM_{2.5}$ and constituent concentrations over annual cycles poses considerable challenges. In efforts to characterize forecast model performance during different seasons, $PM_{2.5}$ forecast simulations with the Eta-Community Multiscale Air Quality system are compared with measurements from a variety of regional surface networks, with special emphasis on performance during the winter period. The analysis suggests that while the model can capture the average spatial trends and dynamic range in $PM_{2.5}$ and constituent concentrations measured at individual sites, significant variability occurs on a day-to-day basis both in the measurements and the model predictions, which are generally not well correlated when paired both in space and time. Systematic overpredictions in regional $PM_{2.5}$ forecasts during the cool season are noted through comparisons with measurements from different networks. The overpredictions are typically more pronounced at urban locations, with larger errors at the higher concentration range. Variability in aerosol sulfate concentrations were captured well, as well as the relative amounts of sulfur (IV) and sulfur (VI). The mix of carbon sources as represented by the ratio of organic to elemental carbon is captured well in the southeastern United States, but the total carbonaceous aerosol mass is underestimated. On average, during the wintertime the largest overpredictions among individual $PM_{2.5}$ constituents were noted for the ‘‘other’’ category which predominantly represents primary-emitted trace elements in the current model configuration. The systematic errors in model predictions of both total $PM_{2.5}$ and its constituents during the winter period are found to arise from a combination of uncertainties in the magnitude and spatial and temporal allocation of primary $PM_{2.5}$ emissions, current uncertainties in the estimation of chemical production pathways for secondary constituents (e.g., NO_3^-), and the representation of the impacts of boundary layer mixing on simulated concentrations, especially during nighttime conditions.

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1. Introduction

[2] The ability to forecast local and regional air pollution events is challenging since the processes governing the production and accumulation of ozone and fine particulate matter are complex and nonlinear. Comprehensive atmospheric models, by representing in as much detail as possible the various dynamical, physical, and chemical processes regulating the atmospheric fate of pollutants, provide a scientifically sound tool for providing air quality forecast guidance. The availability of increased computational power coupled with advances in the computational

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structure of the models have now enabled their use in real-time air quality forecasting. In recent years, such efforts have been used to provide daily guidance to state and local air quality forecasters [e.g., *McHenry et al.*, 2004; *Otte et al.*, 2005], as well as to aid in the design of field experiments [e.g., *Flatøy et al.*, 2000; *Uno et al.*, 2003; *Lawrence et al.*, 2003; *McKeen et al.*, 2005].

[3] Recently, the NOAA and the U.S. Environmental Protection Agency (EPA) partnered to develop a real-time air quality forecasting (AQF) system that is based on the National Weather Service (NWS) National Centers for Environmental Prediction's (NCEP) operational North American Mesoscale (NAM) model (previously the Eta model [*Black*, 1994]) and the U.S. EPA's Community Multiscale Air Quality (CMAQ) modeling system [*Byun and Schere*, 2006]. An initial version of the system was deployed to forecast surface-level O_3 pollution over the northeast United States during the summer of 2003 [*Otte et al.*, 2005]; the surface-level O_3 forecast system is currently operational over a domain covering much of the continental United States and is being evaluated continuously [e.g., *Yu et al.*, 2006, 2007a; *McKeen et al.*, 2005].

[4] An increasing number of clinical and epidemiological studies [e.g., *National Research Council*, 1998] have associated adverse health effects in humans with exposure to fine particulate matter (or $PM_{2.5}$; particles with diameter less than 2.5 μm). It is thus desirable for local air quality agencies to accurately forecast $PM_{2.5}$ concentrations to alert the sensitive population of the onset, severity, and duration of unhealthy air and to encourage the public and industry to reduce emissions-producing activities. In addition, optical characteristics of ambient $PM_{2.5}$ can also result in atmospheric visibility impairment and play an important role in local and global climate change through their impact on direct and indirect radiative forcing. This enhanced interest in ambient particulate matter has provided an impetus to develop and deploy modeling capabilities that can characterize the local to global distributions of particulate matter mass and its chemical composition from daily to annual temporal scales. While model-based forecasts for ozone have publicly been available for some years, forecasts for $PM_{2.5}$ are currently in a developmental stage.

[5] Significant scientific and technical issues surround the characterization of ambient $PM_{2.5}$ distributions both through modeling and measurements [e.g., *McMurry*, 2000]. $PM_{2.5}$ pollution results from both primary emissions and secondary formation through complex photochemical and heterogeneous chemical pathways. The emissions, chemical, and removal processes controlling the day-to-day levels of ambient $PM_{2.5}$ and precursor concentrations also exhibit seasonal variability resulting in significant spatial and temporal variability in ambient $PM_{2.5}$ mass and its chemical composition. Unlike O_3 pollution which occurs primarily during the warm season, elevated $PM_{2.5}$ concentrations are observed throughout the year. Consequently, the challenges for a $PM_{2.5}$ forecast modeling system are quite enormous, i.e., accurately predicting the day-to-day variability in ambient $PM_{2.5}$ mass and composition over daily to annual cycles.

[6] In efforts to address these challenges, developmental model-based forecasts of $PM_{2.5}$ distributions with the Eta-CMAQ modeling system were initiated during the summer of 2004. Eta-CMAQ model predictions of gas and aerosol species during the summer of 2004 have been extensively compared with surface network measurements, as well as measurements aloft collected during the 2004 International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) field experiment [*Mathur*, 2006; *Yu et al.*, 2007a], and have also been compared with predictions from other forecast models [*McKeen et al.*, 2005, 2007]. To develop further insights on the performance of the model during other seasons, in this study we compare predictions from the Eta-CMAQ developmental forecast runs during the winter periods with measurements from a variety of surface networks. The specific study objectives are (1) to assess the ability of the Eta-CMAQ AQF system to provide forecasts of particulate matter during the winter season, (2) to identify and contrast seasonal (summer versus winter) biases in the predictions of both total $PM_{2.5}$ mass as well as its compositional characteristics, and (3) to identify strengths and weaknesses of the overall system and develop guidance for further model development to improve $PM_{2.5}$ forecast capability.

2. Eta-CMAQ AQF System

[7] In this study, we analyze the results from the developmental simulations of the Eta-CMAQ AQF system over the eastern United States during the year 2005. The Eta-CMAQ AQF system consists of three primary components: (1) the Eta meteorological model that simulates the atmospheric dynamic conditions for the forecast period; (2) the CMAQ model which simulates the transport, chemical evolution, and deposition of atmospheric pollutants; and (3) an interface component (PREMAQ) that facilitates the transformation of Eta derived meteorological fields to conform with the CMAQ grid structure, coordinate system, and input data format. Since both the Eta and CMAQ models use significantly different coordinate systems and grid structures, the interface component has been carefully designed to minimize the effects associated with horizontal and vertical interpolation of dynamical fields in this initial implementation. Details on the methods employed and the impacts of assumptions invoked are given by *Otte et al.* [2005]. In the model applications presented here, vertical turbulent mixing of modeled pollutants within the atmospheric boundary layer is based on K theory [*Byun and Schere*, 2006], following the eddy diffusivity formulation described by *Chang et al.* [1987] and extensions by *Byun and Dennis* [1995]. The planetary boundary layer (PBL) heights used in CMAQ in turn are based on the turbulent kinetic energy scheme of *Janic* [1996].

[8] The emission inventories used by the AQF system were updated to represent the 2005 forecast period. NO_x emissions from point sources were projected to 2005 (relative to a 2001 base inventory) using estimates derived from the annual energy outlook by the Department of Energy (http://www.eia.doe.gov/oiaf/aeo). Since the vehicle emissions modeling software MOBILE6 is computationally expensive and inefficient for real-time applications, mobile source emissions were estimated using approximations to the MOBILE6 model. In this approach, Sparse Matrix Operator Kernel Emissions (SMOKE)/MOBILE6 was used to create retrospective emissions over an 8-week period over

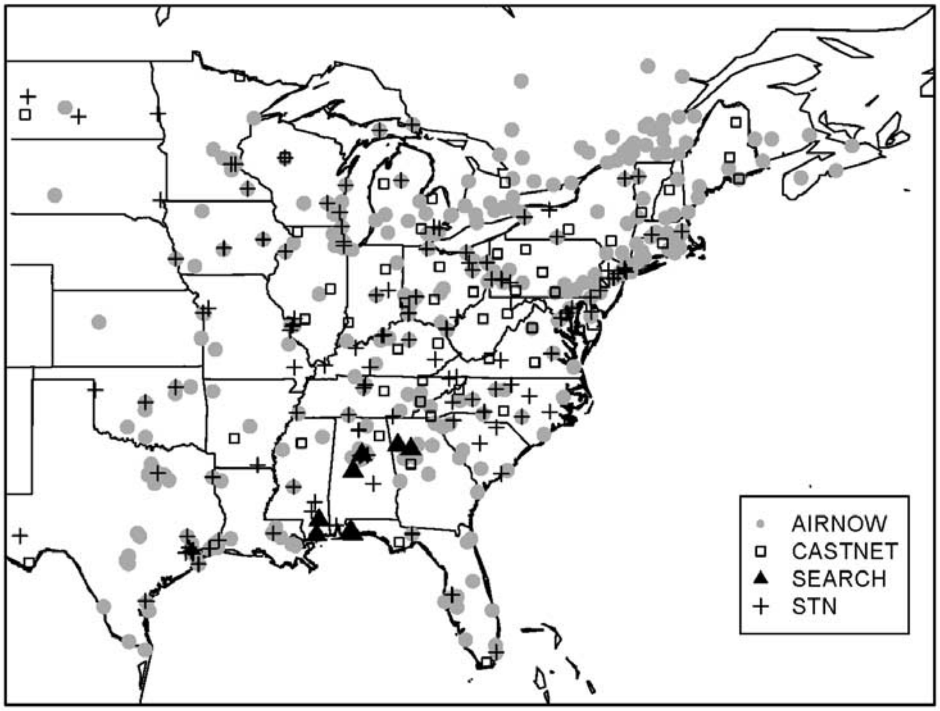

Figure 1. Modeling domain and location of measurement sites from different networks.

the AQF grid using the 2005 (projected from 2001) vehicle miles traveled (VMT) data and 2005 vehicle fleet information. Least squares regressions relating the emissions to variations in temperature were then developed for each grid cell at each hour of the week and for each emitted species. Consequently, mobile emissions could then be readily estimated in the forecast system using the temperature fields from the Eta model [*Pouliot and Pierce*, 2003]. Area source emissions were based on the 2001 National Emissions Inventory (NEI), version 3, while biogenic emissions inventory system version 3.12 (BEIS3.12) [*Pierce et al.*, 2002] was used to estimate the biogenic emissions.

[9] The Eta-CMAQ air quality forecasting system, created by linking the Eta model and the U.S. EPA's CMAQ modeling system was deployed over the domain of the eastern United States during 2005. The Carbon Bond Mechanism (version 4.2) is used to represent the gas phase reaction pathways. The developmental Eta-CMAQ model uses the same aerosol module as CMAQ described by *Binkowski and Roselle* [2003] with updates described by *Bhave et al.* [2004]. The aerosol distribution is modeled as a superposition of three lognormal modes that correspond nominally to the Aitken (diameter (D_p) < 0.1 μm), accumulation (0.1 < D_p < 2.5 μm), and coarse (D_p > 2.5 μm) modes. The model results for $PM_{2.5}$ concentrations are obtained by summing species concentrations over the first two modes. The horizontal domain (geographical extent depicted in Figure 1) was discretized using grid cell sizes of 12 km. Twenty-two layers of variable thickness set on a sigma-type coordinate were used to resolve the vertical extent from the surface to 100 hPa. Daily 24-h duration model simulations were conducted using the meteorological output from the 12 UTC Eta cycle. Three-dimensional chemical fields for all modeled gas and aerosol species were initialized from the ending hour of the previous day's run, thereby building a continuous archive of $PM_{2.5}$ forecast results for the calendar year of 2005.

3. Measurements From Surface Networks

[10] Measurements of particulate matter concentrations from four monitoring networks were employed in this study: AIRNOW, Speciated Trends Network (STN), the Clean Air Status Trends Network (CASTNET), and the Southeastern Aerosol Research and Characterization (SEARCH) network. The spatial distribution of the sites is illustrated in Figure 1. AIRNOW reports continually measured values of total $PM_{2.5}$ mass, CASTNET reports measurements for selected inorganic constituents of $PM_{2.5}$ (SO_4^{2-}, NO_3^-, and NH_4^+), while the STN and the SEARCH network provide detailed speciated data. Collectively, these networks provide good spatial coverage to assess the model's ability in predicting spatial, seasonal, and compositional trends in $PM_{2.5}$ distributions. It should however be noted that each network employs different measurement protocols and these are summarized below.

[11] The AIRNOW only reports total $PM_{2.5}$ mass concentration observations as hourly averages. All measurements are made using tapered element oscillating microbalance (TEOM) instruments averaged over hourly intervals from the top of one hour to the next. It should be recognized that TEOM measurements are somewhat uncertain and are believed to be lower limits to a true measure because of volatilization of semivolatile material (ammonium nitrate and organic carbon) in the drying stages of the measurement [*Eatough et al.*, 2003; *Grover et al.*, 2005]. No attempt is made here to account for this uncertainty.

[12] The U.S. EPA STN (http://www.epa.gov/air/data/aqsdb.html) observations are 24-h filter samples that are collected every third day. Measurements of $PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+, and carbonaceous aerosol are available at 178 STN sites over the eastern United States. For the STN data used here, field blank measurements were used to blank correct the measured aerosol organic carbon (OC) (V. Rao, Environmental Protection Agency, personal communication, 2007) before comparing with the modeled values. The majority of the STN sites are located in urban areas. In contrast, the CASTNET (www.epa.gov/castnet/) sites are predominately located in rural areas. The CASTNET measurements are made using filter packs that are exposed for 1-week intervals (i.e., Tuesday to Tuesday). CASTNET measured species used in this evaluation include SO_4^{2-}, NO_3^-, and NH_4^+.

[13] The SEARCH network reports daily average $PM_{2.5}$ and constituent (SO_4^{2-}, NO_3^-, NH_4^+, OC, and elemental carbon (EC)) concentrations at eight sites including three rural sites (Yorkville, Georgia, Oak Grove, Mississippi, and Centreville, Alabama), four urban sites (Jefferson Street in Atlanta, Georgia, North Birmingham, Alabama, Gulfport, Mississippi, and downtown Pensacola, Florida), and one suburban site (near Pensacola, Florida) (http://www.atmospheric-research.com/searchhome.htm).

[14] It should be noted that the STN and SEARCH network employ different protocols to distinguish OC from EC. The STN uses the thermo-optical transmittance (TOT) protocol, while the SEARCH network employs a thermo-optical reflectance (TOR) protocol which is consistent with the protocol used to estimate OC and EC emissions in the NEI used in the modeling study presented here. Thus only the determination of the total carbon or total aerosol carbon (TC) (TC = OC + EC) is comparable between these two analysis protocols [*Yu et al.*, 2007b]. Since the sampling protocol of the SEARCH network is consistent with the modeled emissions for these species, in the subsequent analysis we examine comparisons of modeled and measured OC and EC mass at only the SEARCH locations and TC at the STN monitor locations. Further, since organic compounds comprising ambient particulate organic mass are largely unknown, an average multiplier is frequently used to convert measurements of OC (typically reported as μg C/m^3) to organic carbonaceous aerosol mass (OCM). The value of 1.4 has been widely used to estimate particulate organic mass [e.g., *Turpin and Lim*, 2001] from measured OC and is also used in our analysis. In subsequent discussions, TC represents the total carbon mass (TC = OC + EC and expressed in units of μg C/m^3), while TCM (expressed in units of μg/m^3) represents the total carbonaceous aerosol mass.

[15] Latitudes and longitudes of each monitoring station from each of the networks falling within the domain of analysis are mapped into the model grid coordinates and observed $PM_{2.5}$ and constituent concentrations are compared against model grid values that contain a monitor for the time period of the observations; thus no spatial interpolation is performed on the model results. In the analyses reported here we compare model and observed daily (24-h) average values for the AIRNOW, STN, and SEARCH networks and weekly averages for comparisons with CASTNET. In each case, however, modeled hourly data were averaged to the times corresponding to the individual measurements.

4. Comparisons With AIRNOW $PM_{2.5}$ Measurements

[16] Figure 2 presents comparisons of domain mean model-predicted daily average $PM_{2.5}$ concentrations with corresponding AIRNOW measurements over the entire calendar year of 2005. There are two reasons for focusing our analysis on daily or 24-h average concentrations which are (1) because air quality forecast guidance often focuses on the exceedance of the 24-hour National Ambient Air Quality Standard, which for $PM_{2.5}$ is currently set at a 35 μg/m^3 daily average concentration, and (2) because it is the finest resolution at which measurements of $PM_{2.5}$ composition are routinely available from regional surface networks (discussed in section 5). Both modeled and measured 24-h average concentrations are calculated from the respective hourly values. Domain mean concentrations are then computed on the basis of the available model-observed pairs for each day; typically there are approximately 300 such pairs over the eastern United States model domain. Also shown in Figure 2 are the variations in model predicted $PM_{2.5}$ mean bias and root mean square error (RMSE). It can be seen from Figure 2 that the model is able to capture the broad synoptic patterns that shape the day-to-day variability in regional $PM_{2.5}$ levels. Examination of the trends in daily mean bias and RMSE over the entire year reveal two interesting features in model performance. First, though the magnitude of the bias and error in the regionally averaged predictions show significant day-to-day variability, the model performance for total $PM_{2.5}$ shows distinct seasonal variations with noticeable overpredictions during the cool seasons and underpredictions during the warm seasons. Second, days with relatively larger errors (RMSE) also generally correspond to days with relatively larger regional $PM_{2.5}$ concentrations.

[17] To examine the model's ability in capturing spatial variability in the predicted $PM_{2.5}$, we examined the frequency distributions of both model and measured concentrations during two seasons at each AIRNOW site. Thus at each site the time series of both model and measured daily average $PM_{2.5}$ over a season was examined and percentiles of the distribution over the season were computed for both the model and the measurements. Scatterplots of specific percentiles (e.g., median) of the model and observed time series are then examined to assess the ability of the model to capture the spatial variability in frequency distributions of $PM_{2.5}$ concentrations across the sites. These are illustrated in Figure 3 for both the winter (January–February 2005) and summer (June–August 2005) periods. At each monitoring site we examined the 5th, 25th, 50th, 75th, and 95th percentile of $PM_{2.5}$ concentration prediction over a season and compared them with the corresponding percentiles from the measured time series. Examining the data in this space-frequency context not only provides indications of the model's ability in capturing the dynamic range of the observations [cf. *Kasibhatla and Chameides*, 2000] but also helps identify trends in biases and errors over different concentration ranges. Taken together, Figures 2 and 3 provide an assessment of the model skill in capturing both

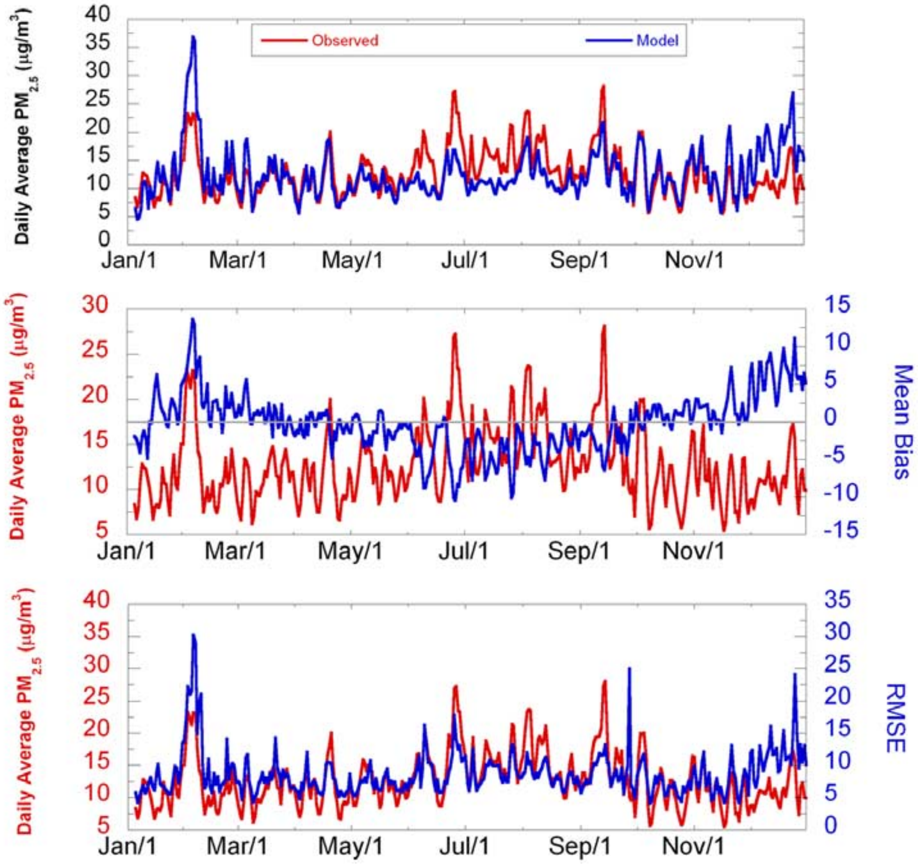

Figure 2. Time series comparison of variations in daily average $PM_{2.5}$ forecasts with AIRNOW measurements over the entire year of 2005. Comparisons are based on spatial averages over all model-observed pairs for each day: (top) domain-mean concentrations, (middle) variations in concentrations and mean bias, and (bottom) variations in concentrations and RMSE.

the temporal and spatial trends of the observations. The distributions in Figure 3 illustrate that though the model captures the dynamic range of the observations, the high bias during the wintertime originates from overpredictions at the higher concentration range (75th and 95th percentiles). In contrast, during the warm season, systematic underpredictions are noted across the entire concentration range. It should be noted that the AIRNOW TEOM-based $PM_{2.5}$ concentration measurements are expected to be biased low, especially during the cool seasons because of volatilization effects during the heating stage of the sample. This heating evaporates volatile compounds such as NH_4NO_3 and organic compounds. During winter in nitrate-rich environments, colder temperatures shift the NH_3-HNO_3- NH_4NO_3 equilibrium to the particulate phase, resulting in larger cool season bias with the TEOM [e.g., *Chow et al.*, 2006]. Additionally, the measurement bias could vary spatially from site to site. While these may impact the magnitude of the noted wintertime bias, a systematic overprediction nevertheless still exists.

[18] To further investigate the wintertime overpredictions, we examined spatial trends in modeled and observed $PM_{2.5}$ for a specific episode spanning 1–6 February 2005 that was characterized by regionally elevated $PM_{2.5}$ concentrations and also relatively large model errors. Figure 4 presents comparisons of the evolution of model and observed concentrations during this episode. These comparisons indicate that though the model was able to spatially capture the elevated $PM_{2.5}$ "hot spots" and their spatial evolution over the episode, it had a tendency to overpredict the concentrations. The midwest and northeast United States were characterized by relatively cool conditions with snow on the ground during this period. Consequently, systematic errors in boundary layer heights and mixing therein could potentially also contribute to the noted wintertime high bias.

5. Comparisons With Aerosol Composition Measurements

[19] To gain additional insights into possible reasons for the seasonal biases in predicted total $PM_{2.5}$, we examined the performance of the model for various $PM_{2.5}$ chemical constituents against available measurements from surface networks. The STN monitors are predominantly located in urban areas, but the network has good spatial coverage across the eastern United States and thus provides an opportunity to assess the ability of the model in capturing regional $PM_{2.5}$ distributions and compositional characteristics. Figure 5 displays the spatial correlations of modeled $PM_{2.5}$ and its constituents with measurements from the STN for the January–February 2005 period. The concentrations illustrated in these comparisons are averaged over all days

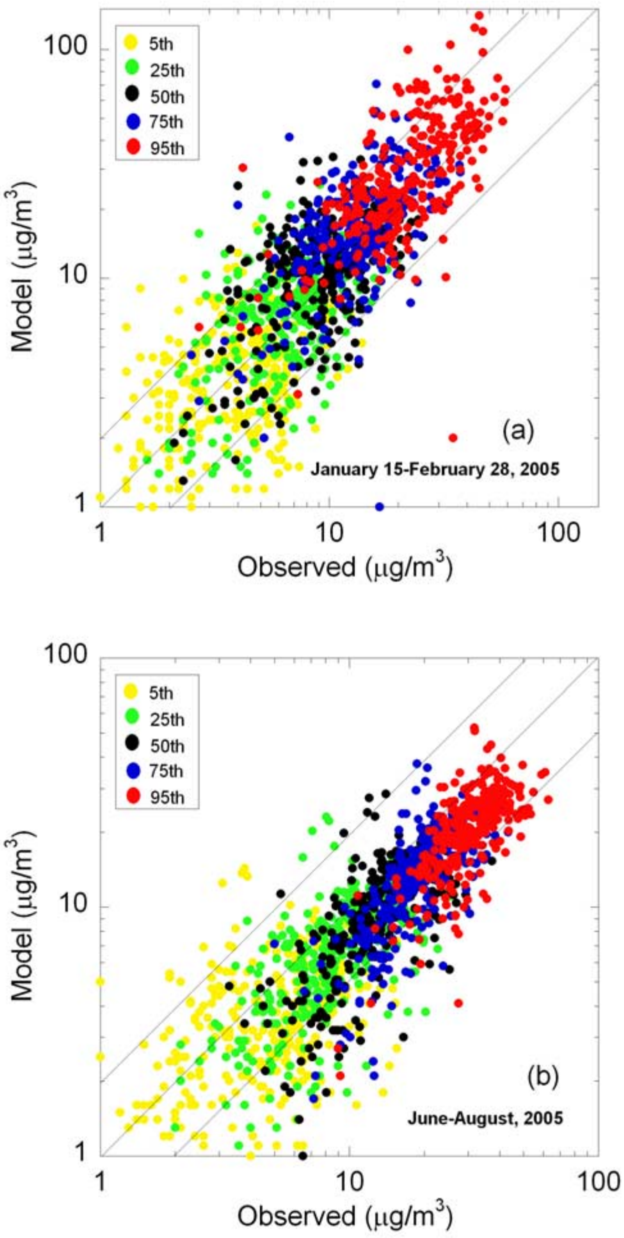

Figure 3. Comparison of model forecast and observed daily average $PM_{2.5}$ distributions for (top) winter (January–February 2005) and (bottom) summer (June–August 2005). Observed $PM_{2.5}$ concentrations are from the AIRNOW network. At each observation location the time series of modeled and observed daily average $PM_{2.5}$ is examined and percentiles of the distributions are computed. The relationship between the modeled and observed percentiles (denoted by different colors) at each location is illustrated. Also shown are the 1:1, 1:2, and 2:1 lines.

on which measurements were available during the January–February 2005 period. Also shown in Figure 5 are the minimum and maximum modeled and observed values at each site during this period. Similar to performance trends noted in wintertime comparisons with the AIRNOW $PM_{2.5}$ measurements, overpredictions in $PM_{2.5}$ mass are also noted across a large majority of STN sites. Though the model exhibits reasonable skill in capturing the spatial variability of the fine PM mass, SO_4^{2-}, NO_3^-, and NH_4^+ as indicated by correlations between the average concentrations, significant differences are noted between dynamic ranges in the modeled and measured concentrations (depicting the day-to-day variability). These comparisons nevertheless show several noteworthy features. In contrast to summer months when $PM_{2.5}$ concentrations in the eastern United States are dominated by sulfate and carbonaceous mass both the measurements and model show that during the cool season nitrate concentrations are comparable and often larger. This is consistent with seasonal trends in $PM_{2.5}$ composition over the eastern United States reported previously [e.g., *Malm et al.*, 2004]. The most noticeable model error is the overprediction of the mass of the "other" constituents. Additionally, the positive bias in $PM_{2.5}$ mass appears to be dominated by the bias in the other constituents. This category represents unspeciated primary fine PM mass in the model and represents the difference between gravimetric mass and the sum of SO_4^{2-} + NO_3^- + NH_4^+ + TCM in the observations. Significant scatter is also noted for the total aerosol carbon (TC, elemental plus organic). During winter months, since secondary aerosol formation is suppressed, the TC mass is expected to be dominated by primary emissions [*Yu et al.*, 2007b]. The discrepancies between modeled and measured TC values could thus be related to a combination of uncertainties both in the spatial representation of primary organic emissions as well as those in estimation of emissions from individual source sectors (e.g., wood burning). Also apparent in these comparisons is the large discrepancy in the dynamic range of individual species concentrations between the model and measurements. In particular, for several constituents (SO_4^{2-}, TC, other), larger overpredictions are noted at the higher end of the concentration ranges and may arise partly from inadequacies in representing the interactions between chemical and dynamical process for discrete events with a 12-km horizontal grid resolution. This, nevertheless, highlights the current challenges in predicting the spatial variability of these individual constituents on a day-to-day basis.

[20] Figure 6a presents similar comparisons of the spatial variability in weekly average concentrations for the inorganic constituents (SO_4^{2-}, NO_3^-, and NH_4^+) against CASTNET measurements. Since the CASTNET monitors are primarily located in rural areas across the eastern United States, these comparisons provide an indication of the model's ability in capturing regional distributions of the inorganic constituents, which in turn are impacted by regional transport events. These comparisons show that the model is able to capture both the dynamic range of the measurements as well as the relative magnitude of the various inorganic fine PM constituents. The better agreement with CASTNET measurements (relative to STN) can in part be attributed to the longer averaging time (weekly as opposed to daily) of the data. Also shown are comparisons of total sulfur concentrations (Figure 6b) and the SO_2:total sulfur ratio (Figure 6c). At the majority of the CASTNET sites, SO_2 constitutes a large fraction (>70%) of the total sulfur. This is captured by the model, providing an indirect assessment of the models ability in adequately capturing the spatial variability in oxidation of sulfur (IV) to sulfur (VI) and the total sulfur

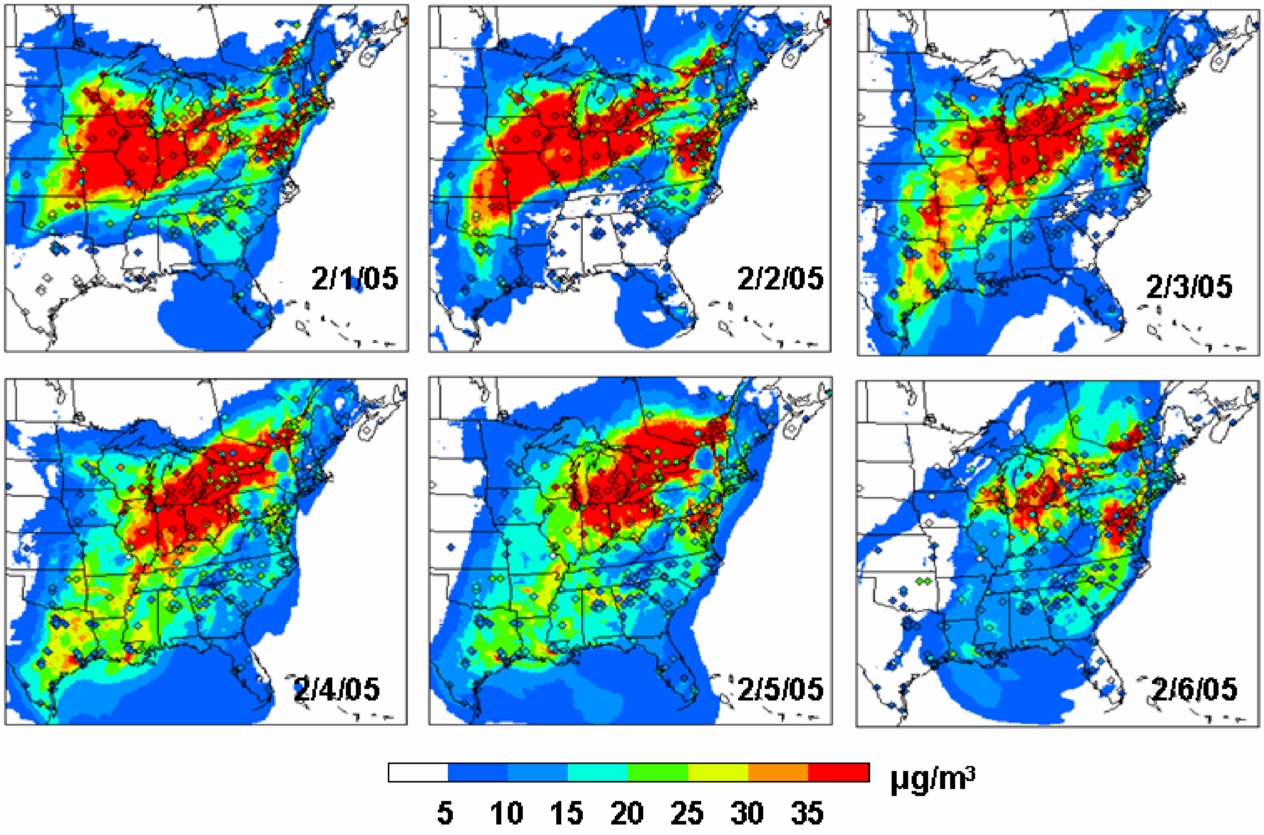

Figure 4. Evolution of model forecast and observed daily average $PM_{2.5}$ spatial distributions during the 1–6 February 2005 time period. Observed values at the AIRNOW monitor locations are shown as color-coded diamonds.

budget. An interesting feature noticeable in Figure 6c is the slight but systematic model overestimation of the SO_2:total sulfur ratio relative to the measurements. This is related to model overestimation of ambient SO_2 levels and may be in turn related to slight overestimation of SO_2 emissions from electric generation units using the emission projection methodology discussed earlier in section 2 and highlights the challenges associated with emission specification in real-time air quality forecasting.

[21] The model's ability to capture the spatial variability in compositional characteristics is further illustrated in Figures 7 and 8 which present comparisons of modeled and observed fractional contributions of various constituents to the total fine particulate mass. Period-averaged fractional contributions (based on constituent concentrations averaged over all days on which measurements are available) at STN monitor locations are shown in Figure 7a, while Figures 8a and 8b illustrate similar comparisons but for daily average values at the urban and rural SEARCH monitors, respectively. Figure 7b illustrates the relationship between the biases in total $PM_{2.5}$ with those in the individual constituents at the STN monitor locations. The rationale for using different averaging times in these comparisons is twofold. First, the number of sites within the SEARCH network is relatively small and the trends are indicative of performance and conditions in a specific geographic area, i.e., the southeastern United States. Consequently, to maintain reasonable sample size for the comparisons, we examined all the model-observed points paired in both space and time. Second, as illustrated in comparisons of constituent concentrations in Figure 5, significant day-to-day variability can occur both in modeled and measured data at the STN monitor locations, which usually were not well correlated. Thus, to isolate average trends in the spatial distributions of the constituents, we present average values over the January–February 2005 period in Figure 7. Several interesting features are apparent in these comparisons. As illustrated in Figure 7, significant spatial heterogeneity exists in the relative fractions of the various constituents in the total $PM_{2.5}$ (Figure 7a), as well as in the relative contribution of different constituents to the overall bias in $PM_{2.5}$ (Figure 7b). Though the relative proportions of the inorganic constituents (SO_4^{2-}, NO_3^-, and NH_4^+) to total $PM_{2.5}$ are captured reasonably well, there is a tendency to overpredict the contributions from SO_4^{2-} and NO_3^- at both the SEARCH and STN sites. In contrast, underpredictions are evident for the carbonaceous mass fraction in comparisons with measurements from both networks. At both the urban and rural SEARCH sites, underpredictions can be noted in the relative fractions of both organic and elemental carbon contributions. Additionally, as illustrated in Figure 7b, biases in all species are correlated with biases in total $PM_{2.5}$, suggesting a noncompositional meteorological related source of the bias.

[22] To further investigate the underprediction in carbonaceous mass contributions to the total $PM_{2.5}$, Figure 9

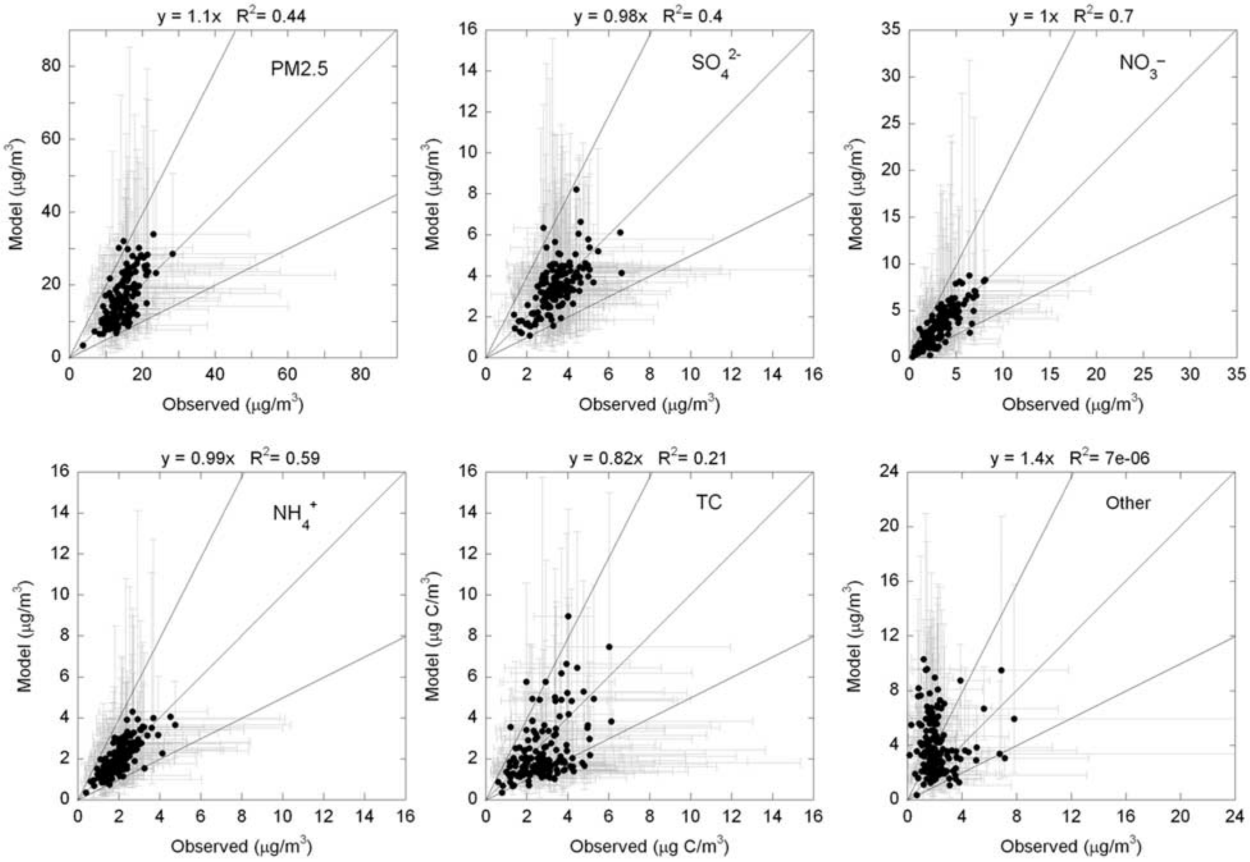

Figure 5. Comparison of model predicted daily average concentrations of $PM_{2.5}$ and its constituents with measurements from the STN for January–February 2005. Solid circles represent average concentrations over the period. Also shown as gray bars are the minimum and maximum daily average concentration at each site. Slopes and R^2 for individual species correlations are indicated. Also shown are the 1:1, 1:2, and 2:1 lines.

examines correlation between the modeled and observed OC and EC at the SEARCH urban and rural sites. The slopes of the linear regression in these correlations are indicative of the OC:EC ratio at these locations. As can be seen, the model predicts OC:EC ratios that are in agreement with the observed values with a ratio of 2.3–2.5 at the urban sites and an OC:EC ratio of 4.2–4.3 at the rural locations. Since secondary organic aerosol formation is suppressed during the wintertime, the modeled and measured OC values can be assumed to originate mostly from primary source emissions. Consequently, the wintertime ambient OC:EC ratio is indicative of the ratio of their respective emissions. Both the measurements and the model show relatively lower-EC concentrations at the rural sites compared to those at the urban sites indicating the lower impact of vehicular emissions at these locations. The difference in slopes between the urban and rural location arises from differences in the source mix contributing to the OC and EC emissions in these regions. Wintertime rural ratios are primarily dictated by biomass-burning emissions while the ratio in the urban areas is influenced by vehicular emissions which have a strong diesel signature [e.g., *Yu et al.*, 2007b]. Since diesel emissions have characteristically high EC, the wintertime OC:EC ratio is expected to be lower in the urban regions and these characteristics in urban versus rural OC:EC ratios are well captured by the model. Also noticeable in the comparisons is the underprediction in the magnitude of the predicted concentrations of both OC and EC at both the urban and rural locations. The accurate prediction of the relative values of the OC:EC ratio at the urban and rural sites, but the systematic underprediction of their concentrations, could arise either from systematic underprediction in the emissions from a source sector (e.g., combustion) or from systematic influence of dynamical processes such as mixing and dilution of the source emissions, or potentially a combination of effects.

[23] Another noticeable feature in the comparisons shown in Figures 7 and 8 is the gross overprediction of the mass fractions of the other constituent category. Since this category represents unspeciated (predominantly, trace elements) fine particulate emissions, the systematic overpredictions are likely related to either an overestimation of these emissions during the cool season or potentially errors in the representation of the prevalent dynamical conditions that regulate atmospheric dilution of these emissions. However, the overprediction of the other constituent category is opposite to the trend noted for the carbonaceous components, which are also of primary nature, at both the

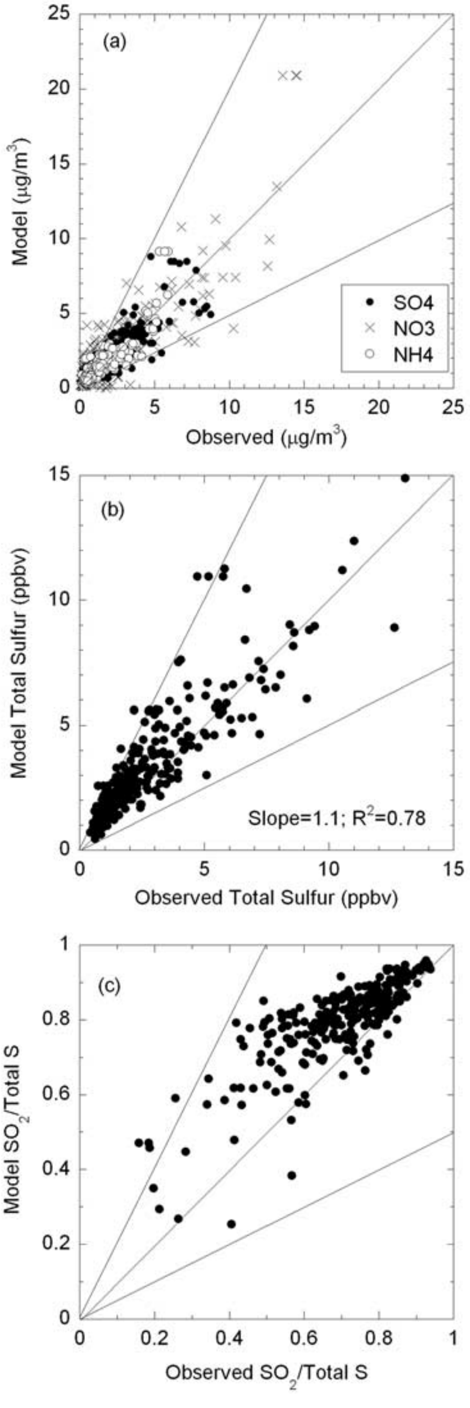

Figure 6. Comparison of model predicted weekly average concentrations of $PM_{2.5}$ constituents with measurements from CASTNET for January–February 2005. (a) Comparisons for SO_4^{2-} (slope = 0.88 and R^2 = 0.7), NO_3^- (slope = 0.99 and R^2 = 0.75), and NH_4^+ (slope = 1 and R^2 = 0.78); (b) comparison of modeled and measured total sulfur ($SO_2 + SO_4^{2-}$); and (c) comparison of modeled and measured SO_2:total sulfur ratios. Also shown are the 1:1, 1:2, and 2:1 lines.

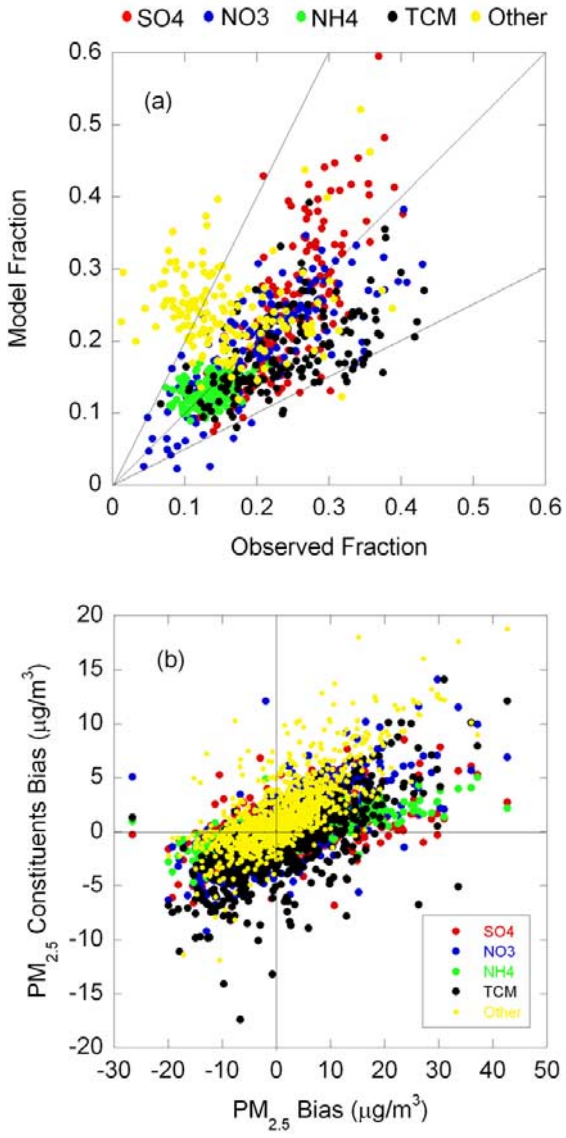

Figure 7. Modeled and measured fine particle mass compositional characteristics at the location of STN monitors for the January–February 2005 period. (a) Comparisons of period-average modeled and measured fractions of SO_4^{2-}, NO_3^-, NH_4^+, TCM, and other constituents at locations of STN monitors. Also shown are the 1:1, 1:2, and 2:1 lines. (b) Relationship between predicted daily average $PM_{2.5}$ bias and bias in individual constituents. Note that the different number of points in Figures 7a and 7b arise because of different averaging periods.

SEARCH and STN monitor locations. Another plausible reason for these discrepancies could be related to both the uncertainties in the magnitude as well as in the specification of the diurnal variation in the emissions of these primary-emitted components; excess emissions during the night can have a disproportionately large impact on the 24-h average concentration because of shallow mixing heights.

[24] To further examine the overpredictions in modeled $PM_{2.5}$ mass during the 1–6 February 2006 period (noted in

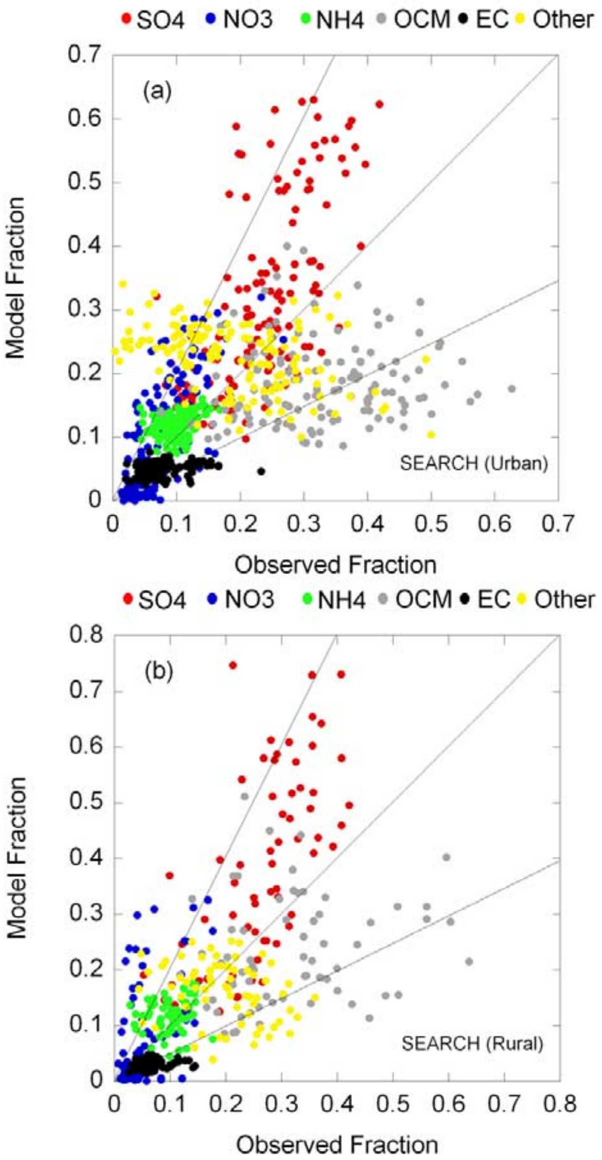

Figure 8. Modeled and measured fine particle mass compositional characteristics during January–February 2005 at SEARCH sites. Comparisons of daily average modeled and measured fractions of SO_4^{2-}, NO_3^-, NH_4^+, OCM, EC, and other at locations of SEARCH monitors. Also shown are the 1:1, 1:2, and 2:1 lines.

Figure 4), we examined the average compositional characteristics over a selection of STN sites located in the north central portions of the modeled domain which was characterized by elevated $PM_{2.5}$ pollution during this period. Comparisons of these modeled and observed mean compositional characteristics are shown in Figure 10. To contrast the performance with another time period when model errors were smaller, similar plots are also shown for the 8–15 March 2006 period (note the change in scales between the two plots). These comparisons illustrate that overestimation of NO_3^- in addition to the overestimation of the other category contribute to the overprediction in total $PM_{2.5}$ during the winter time. The NO_3^- overprediction is also apparent at the SEARCH monitor locations as further illustrated in Figure 11. The over-predictions in NO_3^- are in part related to the uncertainties associated with HNO_3 production via heterogeneous pathways involving N_2O_5 hydrolysis. In the current model implementation the reaction rate of the hydrolysis of N_2O_5 on atmospheric aerosols is quantified by the reaction probability γ_{N2O5} following *Riemer et al.* [2003]. Recent studies [e.g., *Evans and Jacob*, 2005; *Brown et al.*, 2006], however, suggest that the γ_{N2O5} could be considerably lower (perhaps by as much as a factor of 10) than those based on the *Riemer et al.* [2003] parameterization used in the model calculations presented here. Consequently, total ambient nitrate concentrations are likely overestimated and may contribute to the noted overprediction in particulate NO_3^-.

6. Diurnal Variations in Modeled $PM_{2.5}$ and Its Composition

[25] The analyses presented in section 5 indicated that the overestimation in daily average $PM_{2.5}$ concentrations during the cool season was related to the overprediction of primary-emitted constituents. To further investigate these overpredictions, we examined the diurnal variations in modeled

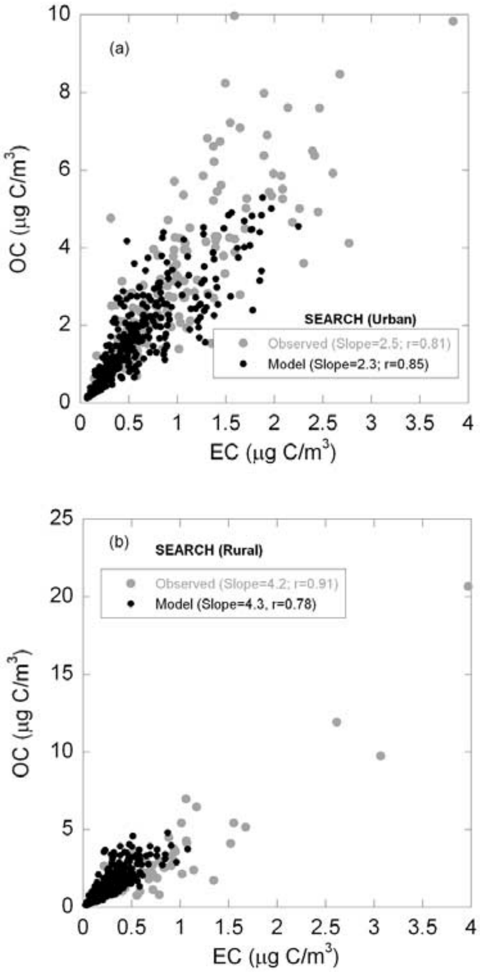

Figure 9. Model and observed correlations between OC and EC at SEARCH sites during January–February 2005.

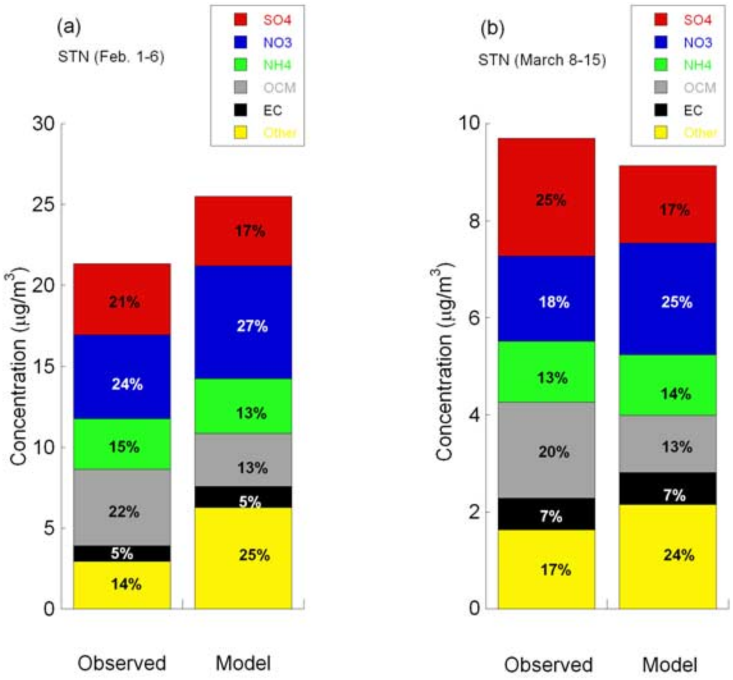

Figure 10. Comparison of observed and modeled $PM_{2.5}$ species concentrations at STN monitors for the periods of (a) 1–6 February 2005 and (b) 8–15 March 2005. The concentrations were averaged over all days during which measurements were available in the respective periods and over all STN monitors located in the northwestern part of the domain where overpredictions in $PM_{2.5}$ were noted in Figure 4.

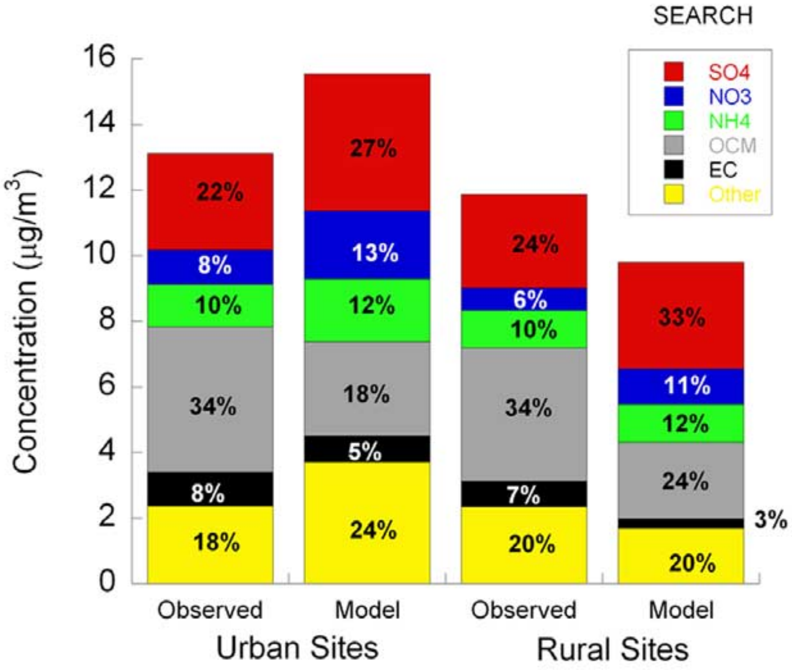

Figure 11. Comparison of observed and modeled $PM_{2.5}$ species concentrations at urban and rural SEARCH monitor sites. The concentrations were averaged over all days during January–February 2005 on which measurements were available.

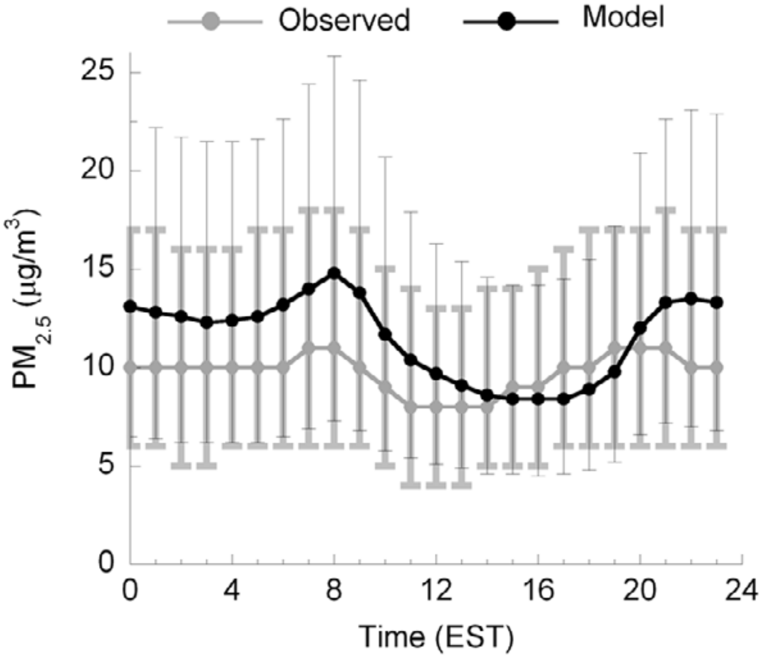

Figure 12. Comparison of diurnal variations of median modeled and observed $PM_{2.5}$ concentrations across all AIRNOW monitor locations and for the January–February 2005 period. Also shown by the upper and lower bars are the 25th and 75th percentiles for both modeled and measured values.

and observed $PM_{2.5}$ concentrations. Figure 12 presents comparisons of modeled and observed (AIRNOW) $PM_{2.5}$ levels over a diurnal cycle. In constructing this figure we examined hourly concentrations at all AIRNOW monitoring locations for the entire January–February 2005 analysis period and computed the median modeled and measured hourly concentrations, which are compared in Figure 12. Also shown are the respective 25th and 75th percentiles at each hour. Systematic overpredictions in the modeled median values (by ~30%) are noted during the nighttime and early morning hours and these appear to contribute primarily to the overpredictions of the daily average values examined in sections 4 and 5.

[26] Figure 13 presents average diurnal profiles of modeled $PM_{2.5}$ composition at the AIRNOW urban and rural monitor locations. When examined in this regional context, during the cool season the modeled relative proportions of the various constituents remain relatively constant through a diurnal cycle, with about half the $PM_{2.5}$ mass composed of the inorganic constituents (SO_4^{2-}, NO_3^-, and NH_4^+) and other half from primary-emitted constituents. Of the latter category, the other constitutes the major fraction with as much as 30% of the total-modeled $PM_{2.5}$ at urban locations and 25% at the rural locations. While these trends in modeled diurnal composition cannot be verified due to the lack of hourly speciated measurements, the noted modeled diurnal trends combined with the fact that daily average concentrations for the other category were also overestimated relative to the STN and SEARCH measurements does suggest that possible overprediction of its nighttime and early morning concentrations contribute to the noted model high bias. This suggests that uncertainties associated with the specification of diurnal variations in emissions of primary fine particulate emissions could be contributing to some of the overpredictions for these primary-emitted constituents.

7. Summary and Recommendations

[27] The analysis of wintertime (January–February 2005) forecast simulations of $PM_{2.5}$ and its constituents obtained using developmental forecast simulations with the Eta-CMAQ forecast guidance modeling system against measurements from a variety of surface monitoring networks suggests that while the model can capture the average spatial trends in $PM_{2.5}$ and constituent concentrations, significant variability occurs on a day-to-day basis both in the measurements and the model predictions, which are generally not well correlated when paired both in space and time. These discrepancies in part arise from comparing discrete events represented by point measurements with spatial average conditions simulated by the grid model and are not necessarily unique only to forecasting applications; similar trends are also noted in retrospective air quality simulations with Eulerian grid models [e.g., *Russell and Dennis*, 2000]. The model, however, does have reasonable skill in simulating the dynamic range of concentrations measured at individual sites. The challenge is in reproducing individual observed events, arising from complex interactions between dynamical and chemical conditions, with grid-based models. Systematic overpredictions in regional $PM_{2.5}$ forecasts during the cool season are noted through comparisons with measurements from different networks. The overpredictions are typically more pronounced at urban locations, with larger errors at the higher concentration range.

[28] Comparisons of model-predicted surface-level fine PM chemical composition with corresponding measure-

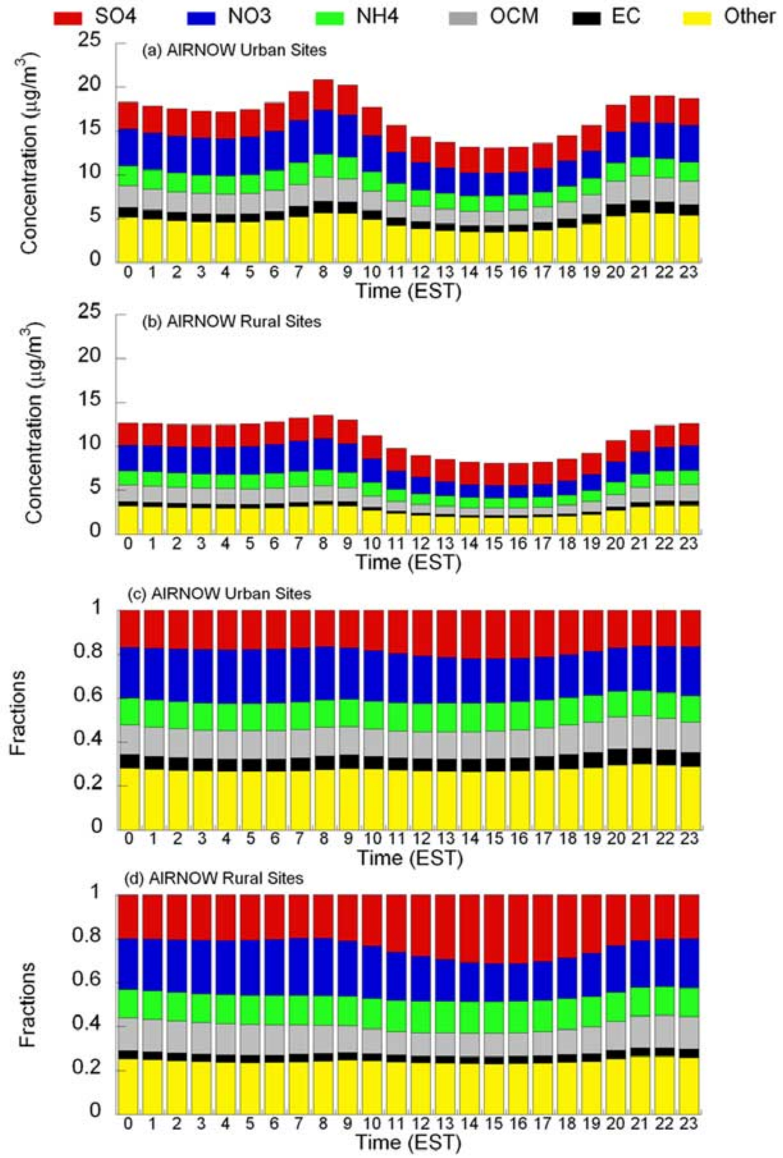

Figure 13. Diurnal variations in modeled compositional characteristics averaged over all AIRNOW monitor locations and over all days during January–February 2005: (a) average diurnal variations in $PM_{2.5}$ composition at AIRNOW urban locations, (b) average diurnal variations in $PM_{2.5}$ composition at AIRNOW rural locations, (c) diurnal variations in constituent fractions at AIRNOW urban locations, and (d) diurnal variations in constituent fractions at AIRNOW rural locations.

ments indicate significant spatial variability in the relative performance for various constituents. On average, during the wintertime the largest overpredictions in individual $PM_{2.5}$ constituents were noted for the other category which predominantly represents trace elements in the current CMAQ configuration; these trends were consistent both at the STN and urban SEARCH monitor locations. It should be noted that the overpredictions for the other category are not unique to the forecast applications discussed here; similar trends are also noted in retrospective applications of the CMAQ model (K. W. Appel et al., Evaluation of the Community Multiscale Air Quality (CMAQ) model version 4.5: Uncertainties and sensitivities impacting model performance. part II: Particulate matter and wet deposition, submitted to *Atmospheric Environment*, 2007). Ambient levels of these constituents are primarily dictated by source emissions, suggesting either an overestimation of emissions of these species in urban regions or systematic underrepresentation of processes leading to their atmospheric dilution. In contrast, systematic underpredictions were noted both in predictions of carbonaceous aerosol mass as well as their relative fractions in total $PM_{2.5}$ in comparisons with both the STN and SEARCH measurements. The model was, however, able to accurately capture the urban-rural transition in the observed OC:EC ratios at the SEARCH sites. The accurate prediction of the relative values of the OC:EC ratio at the urban and rural sites but the systematic underprediction of their concentrations also suggests either systematic underprediction in the emissions from a source sector (e.g., combustion) or systematic influence of dynamical processes such as mixing and dilution of the source emissions. However, since we note overpredictions for the other category and underpredictions for carbonaceous components and since during the winter time both are of primary origin, systematic errors in dynamical processes alone cannot be reasoned to be the cause for the noted discrepancies which likely arise from a combination of errors associated with estimation of their individual source strengths, the specified diurnal allocation, and variation in their emissions and its interaction with dynamical features regulating the dilution of emissions over a diurnal cycle. Unfortunately, very limited information on observed PBL heights and its diurnal structure are available during the study period analyzed here to adequately quantify its impacts on the noted discrepancies in modeled $PM_{2.5}$ levels. Measurements of wintertime PBL heights at several sites across the study domain are needed to assist in the assessment of the role of mixing on model results.

[29] Also noticeable in comparisons with speciated measurements is the overprediction in both the amounts and fractions of NO_3^- during the winter months. These are related to current uncertainties in the representation of total nitrate budgets during the cool season and arise from possible overestimation of the HNO_3 yields from heterogeneous N_2O_5 hydrolysis. Recent measurement [*Brown et al.*, 2006] and modeling [*Evans and Jacob*, 2005] studies indicate that the HNO_3 yields from this pathway could be overestimated using the current parameterization in the model which then contributes to overestimation of particulate nitrate. These overpredictions can then also be further magnified in certain regions from either overestimation of NH_3 emissions and/or from inadequate representation of the intensity of mixing during the cool seasons. Revisions, based on the recent studies, to the estimation of the reaction probability of the heterogeneous N_2O_5 hydrolysis pathway (γ_{N2O5}) in CMAQ are currently underway (J. M. Davis et al., Parameterization of N_2O_5 reaction probabilities on the surface of particles containing ammonium, sulfate, and nitrate, submitted to *Atmospheric Chemistry and Physics*, 2007); these updates will be included in future forecast applications.

[30] The systematic errors in model predictions of both total $PM_{2.5}$ and its constituents noted in the forecast applications presented here arise from a combination of uncertainties in representation of the magnitude and spatial and temporal allocation of primary $PM_{2.5}$ emissions, current uncertainties in representation of chemical production pathways for secondary constituents (e.g., NO_3^- during the cool seasons, secondary organic aerosols during the warm seasons), possible inadequate representation of the interactions between prevalent meteorology and chemical processes (e.g., mixing/dilution). Future research on these aspects of the aerosol processes, their model formulation, and improved linkage between dynamical and chemical model calculations will likely reduce the associated errors identified here. In spite of these existing drawbacks, the utility of $PM_{2.5}$ forecast guidance obtained from comprehensive atmospheric models can in the short-term be improved through the application of postprocess bias correction methods. Preliminary results from the applications of two such methods, which combine observations at discrete locations with model predictions, indicate significant improvement in the model's skill in forecasting $PM_{2.5}$ pollution trends at discrete monitor locations [*Kang et al.*, 2006]. Additional work is, however, needed to further refine these methods so they can be used routinely in model-based air quality forecasting.

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