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Estimating the impact of the 2004 Alaskan forest fires on episodic particulate matter pollution over the eastern United States through assimilation of satellite-derived aerosol optical depths in a regional air quality model

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[1] During the summer of 2004, extensive wildfires burned in Alaska and western Canada; the fires were the largest on record for Alaska. Smoke from these fires was observed over the continental United States in satellite images, and a variety of chemical tracers associated with the fires were sampled by aircrafts deployed during the International Consortium for Atmospheric Research on Transport and Transformation field experiment. Several recent studies have quantified the impacts of the long-range transport of pollution associated with these fires on tropospheric CO and O_3 levels over the eastern United States. This study quantifies the episodic impact of this pollution transport event on surface-level fine particulate matter ($PM_{2.5}$) concentrations over the eastern United States during mid-July 2004, through the complementary use of remotely sensed, aloft, and surface measurements, in conjunction with a comprehensive regional atmospheric chemistry-transport model. A methodology is developed to assimilate MODIS aerosol optical depths in the model to represent the impacts of the fires. The resultant model predictions of CO and $PM_{2.5}$ distributions are compared extensively with corresponding surface and aloft measurements. On the basis of the model calculations, a 0.12Tg enhancement in tropospheric $PM_{2.5}$ mass loading over the eastern United States is estimated on 19 July 2004 due to the fires. This amount is significantly larger (approximately a factor of 8) than the total daily anthropogenic fine particulate matter emissions for the continental United States. Analysis of measured and modeled $PM_{2.5}$ surface-level concentrations suggests that the transport of particulate matter pollution associated with the fires resulted in a 24–42 % enhancement in median surface-level $PM_{2.5}$ concentrations across the eastern United States during 19–23 July 2004.

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1. Introduction

[2] The role of forest fires as large sources of reactive trace gases and aerosols in the atmosphere and their impacts on chemistry of the atmosphere has long been recognized [e.g., *Eagan et al.*, 1974, *Crutzen et al.*, 1979]. Millions of hectares of forest in North America are consumed by fires every year, generating significant emissions of air pollutants. Under favorable conditions, the large amounts of gaseous and aerosol emissions from these fires can be subject to long-range transport and can adversely impact air quality on regional to global scales [e.g., *Fishman et al.*, 1986; *Crutzen and Andreae*, 1990; *Andreae and Merlet*, 2001]. In recent years, several cases of large-scale regional pollution impacts associated with long-range transport of forest fire emissions have been documented in North America. *Wotawa and Trainer* [2000] attributed the widespread high CO concentrations observed over the eastern United States during the summer of 1995 to emissions associated with large forest fires in northern Canada. *McKeen et al.* [2002] further showed that during the period of strongest fire influence, 10–30 ppb enhancements in O_3 levels over large regions of the eastern United States could be attributed to precursor species emitted from these fires. *DeBell et al.* [2004] observed extremely high CO mixing ratios at surface monitoring sites in New Hampshire and Massachusetts during July 2002, concurrent with satellite images of smoke plume from wildfires in Quebec blanketing the United States east coast. Through airborne measurements, *Taubman et al.* [2004] detected a smoke plume between 2–3 km at locations in Virginia and Maryland

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during the same period with enhanced CO, O_3, submicrometer particle number, scattering, and absorption, while *Sapkota et al.* [2005] estimated that transport of emissions from the 2002 Quebec fires resulted in as much as a 30-fold increase in ambient fine PM concentrations in Baltimore during this period.

[3] During the summer of 2004, extensive wildfires burned in Alaska and western Canada. The fires were the largest on record for Alaska, consuming more then 6 million acres (more than 8 times the 10 year acreage average; http://www.dec.state.ak.us/air/am/2004_wf_sum.htm). Smoke from these fires was observed over the continental United States in satellite images of aerosol optical depth (AOD) from the GOES (Geostationary Operational Environmental Satellites) and the MODIS (Moderate Resolution Imaging Spectro-radiometer) aboard the Terra satellite [*Kondragunta et al.*, 2008], images of the aerosol index from the TOMS (Total Ozone Mapping Spectrometer; available from http://toms.gsfc.nasa.gov/aerosols/aerosols_v8.html), and extensive plumes of enhanced CO concentrations measured by the MOPITT (Measurements of Pollutants in the Troposphere) satellite instrument [*Pfister et al.*, 2005]. Through analysis of surface and ozonesonde data in conjunction with trajectory model calculations, *Morris et al.* [2006] attributed a 50–110% increase in ozone concentrations in the first 5 km over Houston due to the transport of NO_x and VOC precursors from these fires. *Martin et al.* [2006] further attribute enhanced aerosol black carbon levels in the North Atlantic lower free troposphere during this period to these fires. However, no assessment of the impacts of these fires on surface-level fine particulate matter (or $PM_{2.5}$; particles with diameter less than 2.5 μm) pollution has been performed to date. This study quantifies the episodic impact of the long-range transport of smoke plumes from the 2004 Alaskan and Western Canada fires on surface-level particulate matter pollution over the eastern United States during mid-July 2004, through assimilation of aerosol optical depths from the MODIS into a comprehensive regional atmospheric chemistry-transport model.

2. The Regional Atmospheric Chemistry-Transport Modeling System

[4] The Community Multiscale Air Quality (CMAQ) model [*Byun and Schere*, 2006] driven with meteorological fields from the Eta model [*Black*, 1994] is used to examine the three-dimensional atmospheric chemical conditions during July 2004. This Eta-CMAQ system had also been deployed over the eastern United States during the summer of 2004 to provide air quality forecast (AQF) guidance [*Mathur et al.*, 2004; *Mathur et al.*, 2005a]. 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 that 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 can be found in *Otte et al.* [2005].

[5] The emission inventories used by the AQF system were constructed to represent the 2004 forecast period. NO_x emissions from point sources were projected to 2004 (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/index.html). Mobile emissions were estimated using the least-squares regression approximations to the MOBILE6 model following the approach of *Pouliot and Pierce* [2003]. Area source emissions were based on the 2001 National Emissions Inventory, version 3, while BEIS3.12 [*Pierce et al.*, 2002] was used to estimate the biogenic emissions.

[6] In the applications presented here, the Carbon Bond-IV chemical mechanism is used to represent the gas-phase reaction pathways. The Eta-CMAQ model uses the same aerosol module as CMAQ described in *Binkowski and Roselle* [2003] with updates described in *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 2) 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-hour 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.

[7] The model simulated aerosol optical depth (AOD) at any location is obtained by integrating the estimated aerosol extinction coefficient β_{sp} [km^{-1}] at a given wavelength from the surface to the model top (100 mb):

$$AOD = \int \beta_{sp}(z)dz. \quad (1)$$

[8] The simulated three-dimensional aerosol characteristics such as volume concentrations, size distributions, and index of refraction were used to estimate the Mie extinction efficiency factor, Q_{ext}, using the method of *Evans and Fournier* [1990]. Three-dimensional β_{sp} fields can then be estimated following equations (23) and (24) in *Binkowski and Roselle* [2003]; these are then vertically integrated through the model column depth following equation (1) to yield the modeled AOD at each hour. It should be noted that the accuracy of the modeled AOD is dependent on the accurate simulation of aerosol composition and size distribution; however, very little observational data is available to rigorously evaluate the simulated aerosol size distributions.

3. Model Simulations

3.1. The Base Case

[9] Two sets of model simulations were conducted to assess the impact of the long-range transport of pollution

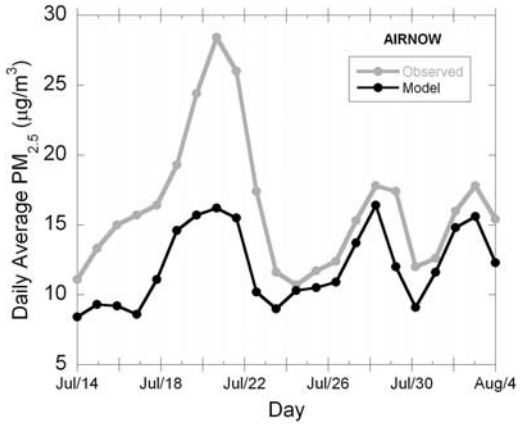


Figure 1. Time series comparison of variations in daily-average $PM_{2.5}$ forecasts from the base simulation with AIRNOW measurements during mid-July to early August 2004.

associated with emissions from the wildfires in Alaska and Western Canada on surface-level pollution over the eastern United States. Figure 1 presents comparisons of domain mean model-predicted daily-average $PM_{2.5}$ concentrations with corresponding measurements from the AIRNOW during mid-July and early August, 2004 from the Eta-CMAQ forecast runs (hereafter referred to as the base case). The AIRNOW surface network reports only 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 the 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 due to the volatilization of semi-volatile 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 in the observations. Both modeled and measured 24-hour 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. It can be seen from Figure 1 that the model is able to capture the broad synoptic signatures that shape the day-to-day variability in regional $PM_{2.5}$ levels, with a tendency to slightly under-predict the domain mean values, except during the period in mid-July when significantly larger under-predictions are noted. This is further illustrated in Figure 2 (top) which presents comparisons of spatial trends in the evolution of the observed and simulated base case surface-level daily 24-hour average $PM_{2.5}$ concentrations over the eastern United States during the 17–22 July 2004 period. Widespread enhancements in regional $PM_{2.5}$ observed concentrations are noted during 20–22 July 2004, which are not captured in these base model calculations. Detailed comparisons of $PM_{2.5}$ predictions from the base case simulations against measurements from a variety of surface networks and aircraft measurements during July–August 2004 are summarized in *Yu et al.* [2008] and analysis of model results during other seasons can be found in *Mathur et al.* [2008].

[10] Also shown in Figure 2 are comparisons of spatial distributions of modeled (base case; middle) and observed (bottom) AOD at 550nm. The observed AOD values are based on measurements from the National Aeronautics and Space Administration (NASA) satellite-borne Moderate Resolution Imaging Spectro-radiometer (MODIS) sensor aboard the Terra satellite; the MODIS AOD retrieval algorithm uses the satellite measured radiance/reflectance through the use of finite look-up tables as described in *Kaufman and Tanré* [1998]. Additional details on the algorithm and associated uncertainties in the MODIS AOD retrievals can be found in *Kaufman et al.* [2002] and *Remer et al.* [2005]. AOD retrievals for the daytime overpasses from the MODIS are used in this study. MODIS AOD data in latitude/longitude coordinates was mapped to the CMAQ 12 km grid on a Lambert conformal projection; no spatial interpolation was performed to fill in regions/grid-cells with missing data. If multiple MODIS observations were present within a single grid cell, the maximum value was used to represent the observed AOD for that grid cell for that day (the average of the multiple values within the grid cell was also examined, but it had little impact on the subsequent analysis). Similarly, the modeled spatial distributions displayed in Figure 2 were constructed on the basis of the maximum of the predicted hourly AOD values between 1500–2000 GMT, the time period over which the Terra satellite covered the continental United States. As described in *Morris et al.* [2006], the plume from the Alaskan fires moved eastward over Canada during 12–16 July 2004, before dividing on 17 July, with part moving east and part moving south. The region of high AOD observed (Figure 2, MODIS) near the north-western boundary of the modeled domain on 17 July is associated with this southern component of the divided Alaskan fire plume. Over the next few days, the troposphere over the eastern United States was impacted by the long-range transport of aerosols associated with this plume, as a front traversed the eastern United States as depicted in the evolution of the AOD fields in Figure 2. In regions ahead of the front, the modeled AOD showed reasonable agreement in spatial signatures with those inferred from the MODIS retrievals; however, large under-predictions are noted in the base case simulated AOD fields in regions behind the front. These discrepancies can be attributed to the fact that static lateral boundary conditions representative of "clean" background conditions [*Byun and Schere*, 2006] were used in the base case simulation.

[11] Taken together, the spatial distributions shown in Figure 2 suggest that the initial transport of the plume into the modeled domain was aloft above the daytime boundary layer during 17–19 July. Over the next few days as the plume was transported eastward along a front, it mixed downwards, and led to widespread impacts on regional surface-level particulate matter pollution (as indicated by the AIRNOW surface $PM_{2.5}$ measurements during 20–22 July 2004). Further evidence of the impact of the Alaskan fires plume is presented in Figure 3, which shows temporal changes in measured surface-level concentrations of total carbonaceous aerosol mass (TCM) from the U.S. EPA Speciation Trends Network (STN). Shown in Figure 3 is the distribution of

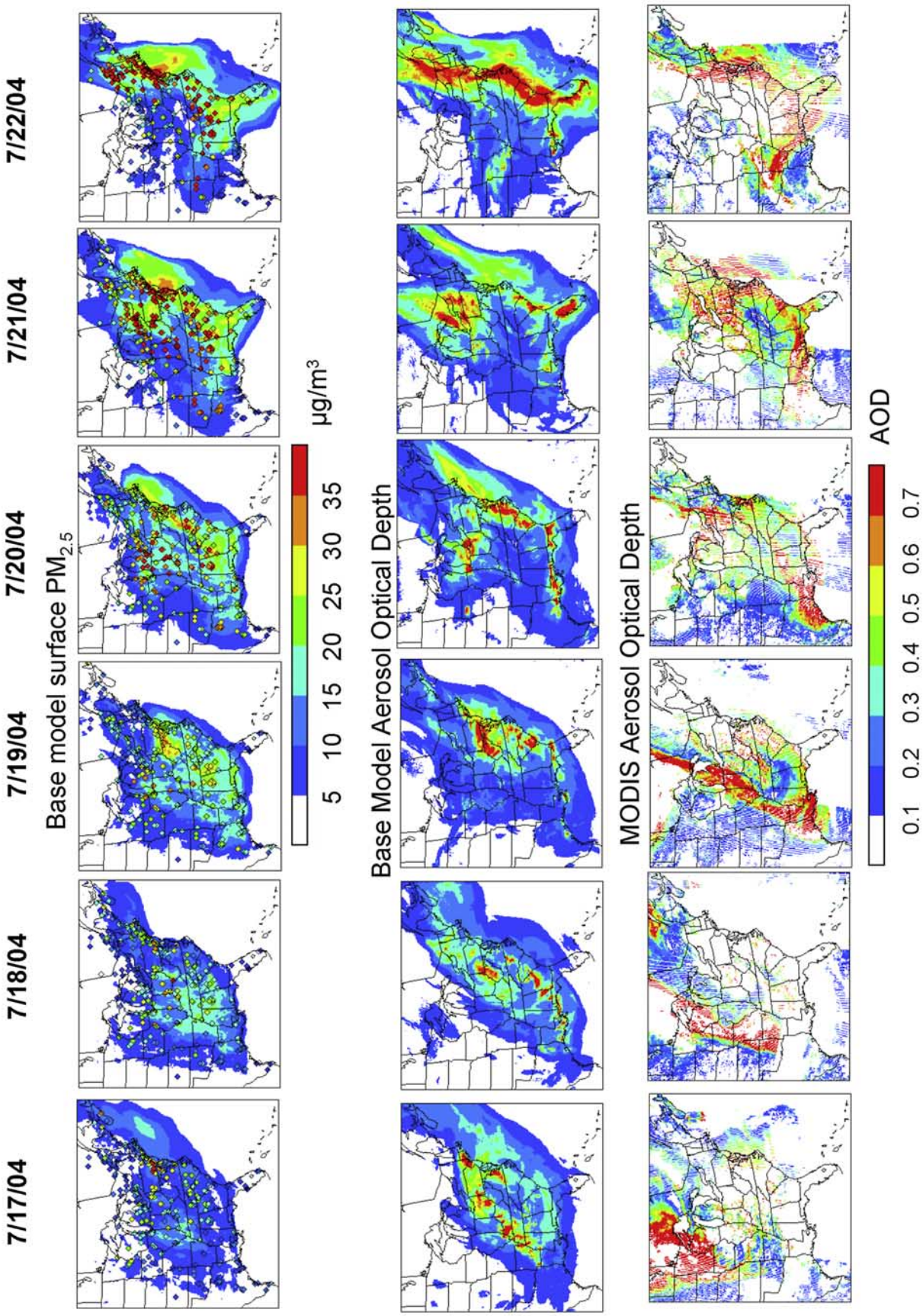

7/17/04
7/18/04
7/19/04
7/20/04
7/21/04
7/22/04
Base model surface $PM_{2.5}$
5 10 15 20 25 30 35
µg/m³
Base Model Aerosol Optical Depth
MODIS Aerosol Optical Depth
0.1 0.2 0.3 0.4 0.5 0.6 0.7
AOD

Figure 2

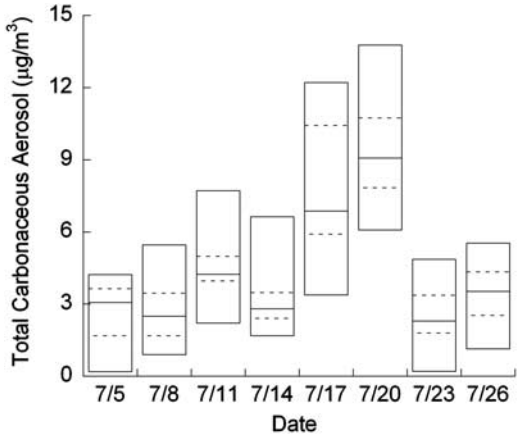


Figure 3. Distribution of observed total carbonaceous aerosol concentration at STN monitors across the eastern United States model domain. Lower and upper edges of the boxes represent the 5th and 95th percentile, respectively; the solid line in the middle is the median (or 50th percentile), while the lower and upper dashed lines represent the 25th and 75th percentile, respectively.

24-hour average TCM measurements across 178 STN sites within the eastern United States model domain on days on which measurements were available. Median observed TCM concentrations on 17 and 20 July are noted to be 2–3 times higher than those on other days. These enhancements in observed regional TCM concentrations are coincidental with enhancements in regional $PM_{2.5}$ levels noted in the AIRNOW measurements (Figures 1 and 2), further suggesting the influence of wildfires, which are a large source of carbonaceous aerosols.

3.2. The Assimilation Simulation and Methodology

[12] Similar to weather forecasting, knowledge of current and recent conditions is critical for accurate predictions of the onset, severity, and duration of episodic pollution events. The impacts of long-range pollutant transport, as in the case of pollution associated with the Alaskan fires discussed in the previous section, can potentially be modeled more accurately either through (1) improved representation of the chemical lateral boundary conditions used by the limited area regional air quality model, or (2) through assimilation of routine real-time measurements of pollutant concentrations. The former approach would require coupling with a larger scale atmospheric chemistry-transport model which, in turn, would need to include an accurate description of the sources and origin of such pollution as well as fidelity in representing the long-range three-dimensional transport, chemical processing, and potential sinks of the transported pollutants.

[13] Unfortunately, routine measurements of pollutant concentrations are only available at the surface and only for a selected set of species, and have limited spatial coverage. Further, the transport of pollution plumes over large distances often occurs aloft, undetected by surface monitors. Emerging observations of atmospheric trace constituents made from satellites provide useful information on the large-scale pollutant distributions and transport, the assimilation of which into atmospheric models could potentially improve model skill. Satellite retrievals of AOD can be used to infer integrated columnar distribution of fine particulate matter. Recent studies on comparisons of MODIS AOD with surface measurements have demonstrated its suitability for monitoring air quality events over local to global scales [e.g., *Chu et al.*, 2003; *Wang and Christopher*, 2003; *Hutchison*, 2003; *Engel-Cox et al.*, 2004; *Al-Saadi et al.*, 2005]. In this study, we explore the possible use of the MODIS AOD for assimilation in a regional atmospheric chemistry-transport model to improve model predictions of $PM_{2.5}$ with special emphasis on the period in mid-July 2004, when $PM_{2.5}$ pollution over the eastern United States was impacted by the long-range transport of the Alaskan fire plume.

[14] The AOD assimilation methodology developed in this study is based on the premise that at any given location a functional relationship exists between the AOD and the total column integrated $PM_{2.5}$. *Wang and Christopher* [2003] and *Al-Saadi et al.* [2005] found strong linear correlation between MODIS AOD and surface-level $PM_{2.5}$ measurements from the AIRNOW network at several locations. These relationships are valid only for conditions wherein the $PM_{2.5}$ pollution burden is dictated solely by boundary layer pollution and do not characterize adequately situations wherein aerosols are present aloft above the boundary layer (moderately large observed AOD, but not surface $PM_{2.5}$ concentrations) such as in cases involving long-range transport aloft. Since direct measurements of atmospheric burden of fine particulate matter are not available, the correlation between modeled AOD and model column PM burden was examined. On the basis of analysis of CMAQ-predicted AOD and column-average $PM_{2.5}$ burden (hereafter referred to as PM_{burden} and expressed in $\mu g/m^3$) at each 12-km grid cell for the 11 July–18 August 2004 period, the following model-based linear relationship (with an $R^2 = 0.9$) was found

$$PM_{burden} = 9.065 \times AOD + 0.18 \tag{2}$$

where,

$$PM_{burden} = \frac{\sum_{i=1}^{N} C_i \Delta z_i}{H}. \tag{3}$$

[15] In equation (3), C_i represents the $PM_{2.5}$ concentration at model layer i, Δz_i is the thickness (in m) of model layer i, H is the height of the vertical extent of the model from surface to 100 mb at any given location ($H = \Sigma \Delta z_i$), and

Figure 2. Evolution of spatial trends in model and observed $PM_{2.5}$ concentrations and aerosol optical depth (AOD) during 17–22 July 2004. (top) Surface-level daily-average $PM_{2.5}$ forecasts from the base simulation. Observed values are shown as color-coded diamonds. (middle) Simulated aerosol optical depth in the base simulation. (bottom) Aerosol optical depths derived from MODIS on the model grid. No spatial interpolation is performed to fill in missing values.

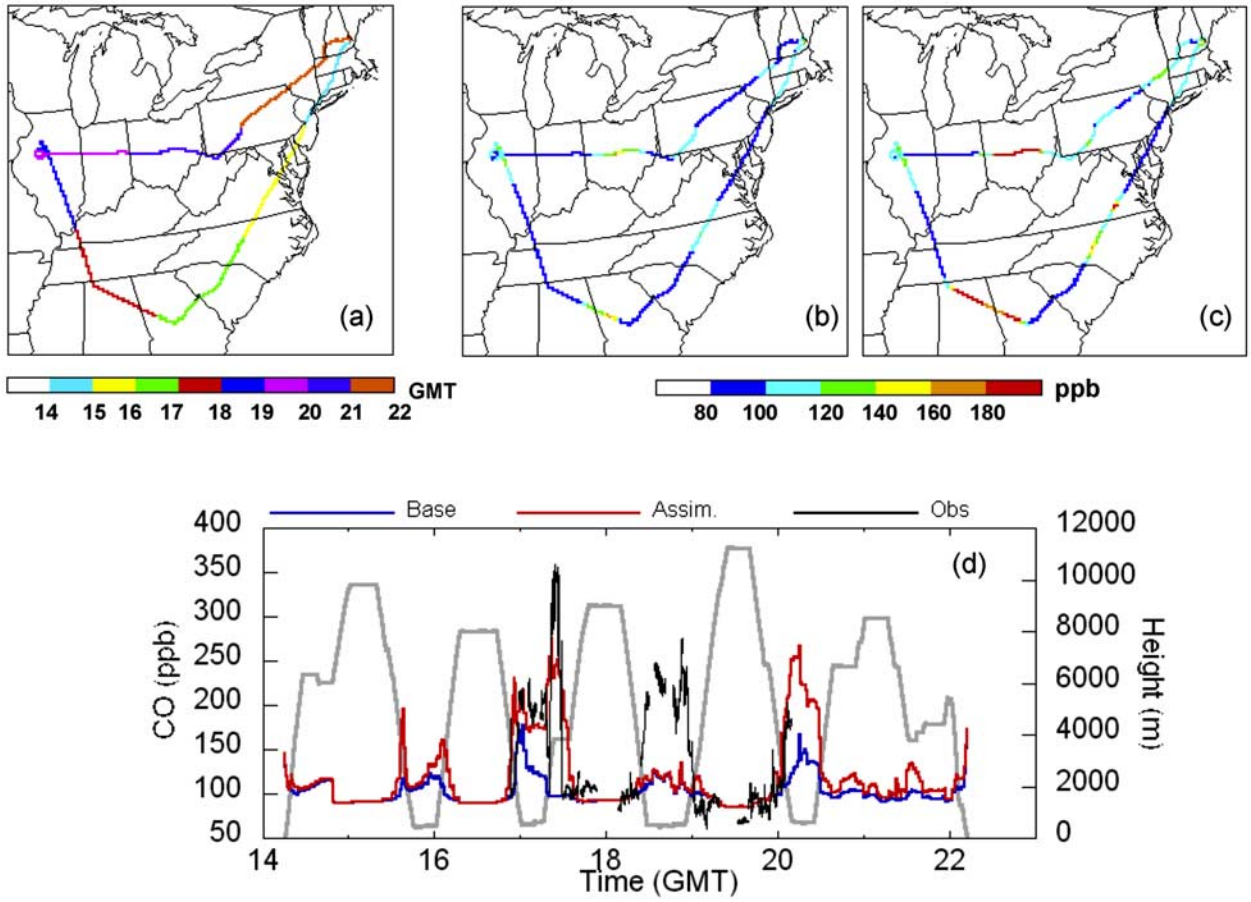


Figure 4. Comparison of modeled and observed CO mixing ratios along the NASA DC-8 flight path on 20 July 2004. (a) Flight path color coded by the GMT hour; (b) modeled concentrations in the base simulation; (c) modeled concentrations in the assimilation simulation; and (d) time series comparisons of (black) observed CO with corresponding modeled values paired in space and time from the (blue) base and (red) assimilation simulations. The grey line represents the aircraft altitude.

N is the number of layers used to discretize the vertical model extent. Substituting the gridded MODIS AOD (as in Figure 2) in equation (2) then yields an inferred column-average $PM_{2.5}$ mass burden (PM_{infer}) field. The difference between the inferred and the base case column burden then quantitatively represents the missing source of $PM_{2.5}$ mass in the base calculation. Thus, for this case the $PM_{2.5}$ mass burden as a result of advection into the domain (PM_{adv}) can then be estimated as:

$$PM_{adv} = H\left(PM_{infer} - PM_{burden,\,Base}\right) \quad (4)$$

[16] This excess mass at each location can then be allocated vertically and speciated to its constituent species to initialize the vertical $PM_{2.5}$ distributions for input to CMAQ, provided information is available on the characteristics of aerosol composition as well as its vertical distribution.

[17] Aerosol backscatter measurements from the Regional East Atmospheric Lidar Mesonet (REALM; available at http://alg.umbc.edu/REALM) indicated the presence of the Alaskan smoke plume between 2.5–4 km, above the daytime boundary layer over the mid-western United States on 19 July 2004. The methodology described in equations (2) (3) (4) was applied to MODIS AOD observations on 19 July 2004 to estimate an inferred particulate matter burden at 1600 GMT, the time of the Terra overpass over the eastern United States. The excess $PM_{2.5}$ mass estimated from equation (4) was then distributed between layers 14–16 of the CMAQ model domain corresponding approximately to model altitude of 2.2–4 km as

$$\left[PM_{2.5adj}\right]_i = \left[PM_{2.5Base}\right]_i + \left(PM_{adv}/\Delta z_i\right),\ i = 14, 16 \quad (5)$$

where, $[PM_{2.5adj}]_i$ represents the adjusted or reinitialized concentration in layer i, and $[PM_{2.5Base}]_i$ is the corresponding simulated concentration at the given time-step from the base case simulation. The $PM_{2.5}$ mass was further apportioned to its constituents following U.S. EPA AP-42 compilation of air pollutant emission factors for typical wildfire emissions [available at: http://www.epa.gov/ttn/chief/ap42] as organic carbon (OC, 77%), elemental carbon (EC, 16%), SO_4^{2-} (2%), NO_3^- (0.2%), and other unspeciated fine particulate matter mass (4.8%). Following this procedure, the three-dimensional $PM_{2.5}$ constituent fields over the eastern United States model domain were re-initialized at 1600 GMT on 19 July 2004. Additionally, using an emission

mass ratio of $CO/PM_{2.5}$ = 10 from the AP-42 wildfire emission factors, CO fields were also reinitialized to reflect the influence of CO originating from the Alaskan fires. CMAQ was then run forward through the end of 23 July 2004; these simulations are, hereafter, referred to as the assimilation case.

4. Model Results

[18] In addition to particulate matter emissions, forest fires are also known to be large sources of CO. Given its typical atmospheric lifetime, CO emitted from forest fires can be transported over long distances, making it a good indicator for forest fire emissions [cf. *Wotawa and Trainer*, 2000]. To test the ability and accuracy of the assimilation methodology described in section 3.2 in representing the impacts of the Alaskan fires on tropospheric pollution over the eastern United States, CO predictions are quantitatively analyzed first.

4.1. Comparison with Aloft and Surface-Level CO Measurements

[19] During July and August 2004, airborne measurements of a variety of trace species were made from extensively instrumented aircrafts deployed as part of the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) field study [*Fehsenfeld et al.*, 2006; *Singh et al.*, 2006]. Enhanced CO concentrations associated with the Alaskan forest fire plume were detected at several locations along transects by the NOAA WP-3 and NASA DC-8 research aircraft and were associated with concurrently enhanced concentrations of acetonirile, a chemical marker of wildfire emissions [*Holzinger et al.*, 1999]. Comparison of model predictions with these CO measurements provide an opportunity to test the ability of the assimilation methodology in representing the three-dimensional transport signature of the Alaskan plume, consequently providing a basis for the assessment of its impacts on surface-level particulate matter pollution discussed later in this section. Following *Mathur et al.* [2005b], modeled concentrations and mixing ratios along flight paths of selected NOAA WP-3 and NASA DC-8 flights during the 20–23 July 2004 analysis period, were extracted by "flying" the aircraft through the 3-D modeling domain; the spatial locations of the aircraft were mapped to the model grid, whereas hourly resolved model output were linearly interpolated to correspond to the time of the measurement.

[20] Comparisons of modeled and observed CO mixing ratios paired in both space and time along the NASA DC-8 flight path on 20 July 2004 are presented in Figure 4. Figure 4a presents the flight path of the DC-8 on this day, color coded by the GMT time; the corresponding aircraft altitude is shown in the time series plots in Figure 4d. The modeled CO mixing ratios along the flight path across the region from the base and the assimilation simulation cases are shown in Figures 4b and 4c, respectively. Also illustrated in Figure 4d are comparisons of CO mixing ratios measured by the NASA DC-8 with corresponding modeled values from the base and the assimilation simulations. In Figure 4d, significant enhancements in measured CO mixing ratios are noted along the flight segments between 1700–1800 GMT between altitudes of 2–4 km and also between 2000–2100 GMT coincident with enhancements in measured acetonitrile mixing ratios (not shown) suggesting that the aircraft sampled the biomass burning plume associated with the Alaskan fires. These enhancements were not captured in the base case simulation which used the default static lateral boundary conditions, but are more accurately represented in the assimilation simulation. In these comparisons, since the model and measured mixing ratios are paired in space and time, the much improved agreement with the measurements shown in the assimilation simulation suggests that the initialization of the model resulting from the assimilation methodology and the model dynamics were able to accurately represent the three-dimensional transport of the Alaskan fire plume over the eastern United States.

[21] The efficacy of the modeling methodology in representing the transport and placement of the Alaskan fire plume is further illustrated in Figure 5, which presents similar comparisons of modeled CO mixing ratios with measurements along selected segments of the NOAA-WP3 aircraft flights. Analysis of NOAA-WP3 measurements of CO, acetonitrile, and water soluble organic carbon by *Sullivan et al.* [2006] identified four cases in which the WP3 intercepted major biomass burning plumes during July 2004. Comparison of the modeled CO values for two of those cases within the analysis period considered here (on 20 and 21 July) are shown in Figure 5. Excellent agreement in the placement of the plume by the model is evident as illustrated by the simulated enhancements in CO mixing ratios in the assimilation case relative to both the base case simulation and the measured values; the CO mixing ratios were grossly under estimated in the base case simulation, but were more accurately captured at locations where the aircrafts sampled the Alaskan fire plume in the assimilation case. It should be noted that some discrepancy between modeled CO mixing ratios in the assimilation case and the measured values from these flights should be expected given that the model values shown in the figure are based on linear interpolation of hourly model output mixing ratios and also because the variability reflected in the measured mixing ratios during these aircraft intercepts is essentially a sub-grid scale feature when compared to the discrete 12 km horizontal resolution used in the model calculations. Nonetheless, the agreement in the location and timing of the simulated CO enhancements in the comparisons with CO measurements from both the WP-3 and the DC-8 does point to the ability of the assimilation methodology combined with the model dynamics in capturing the three-dimensional transport and structure of the Alaskan fire pollution plume.

[22] The impact of the Alaskan fire plume on surface-level CO pollution was assessed through comparisons of modeled CO mixing ratios with measurements from the South Eastern Aerosol Research and CHaracterization (SEARCH) network. The SEARCH network reports hourly averaged concentrations for several gaseous species at eight sites including three rural sites – Yorkville, GA, Oak Grove, MS, and Centreville, AL; four urban sites – Jefferson Street in Atlanta, GA, North Birmingham, AL, Gulfport, MS, and downtown Pensacola, FL; and one suburban site near Pensacola, FL (http://www.atmospheric-research.com/searchhome.htm). Figure 6 presents comparisons of daytime average (10 am–4 pm EST) modeled and measured CO

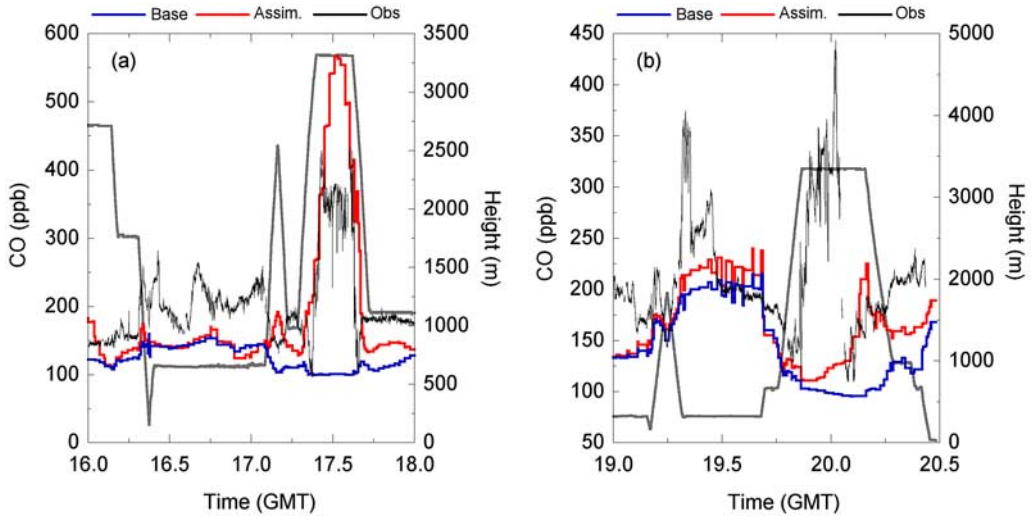


Figure 5. Comparison of modeled and observed CO mixing ratios along selected segments of the NOAA-WP3 flights identified by *Sullivan et al.* [2006] as having intercepted biomass burning plumes. (a) Comparisons with measurements from flight on 20 July 2004; (b) same as Figure 5a but for the flight on 21 July 2004. Observed mixing ratios are represented by the black line, base case simulation results by the blue line, assimilation simulation results by the red line. The grey line represents the aircraft altitude.

mixing ratios for the 19–23 July 2004 time period at seven SEARCH sites; CO measurements were not available at the North Birmingham site. There are two reasons for the choice of this averaging period. First, measured diurnal CO variations especially in urban and suburban locations are dominated by early morning and late evening peaks associated with rush hour mobile source emissions, the amplitude of which is generally not captured well with grid resolution employed in regional models. Second, since the intent of the analysis presented here is to assess the impact of the Alaskan fire plume on surface concentrations, its signal is likely to be more detectable during daytime conditions when boundary

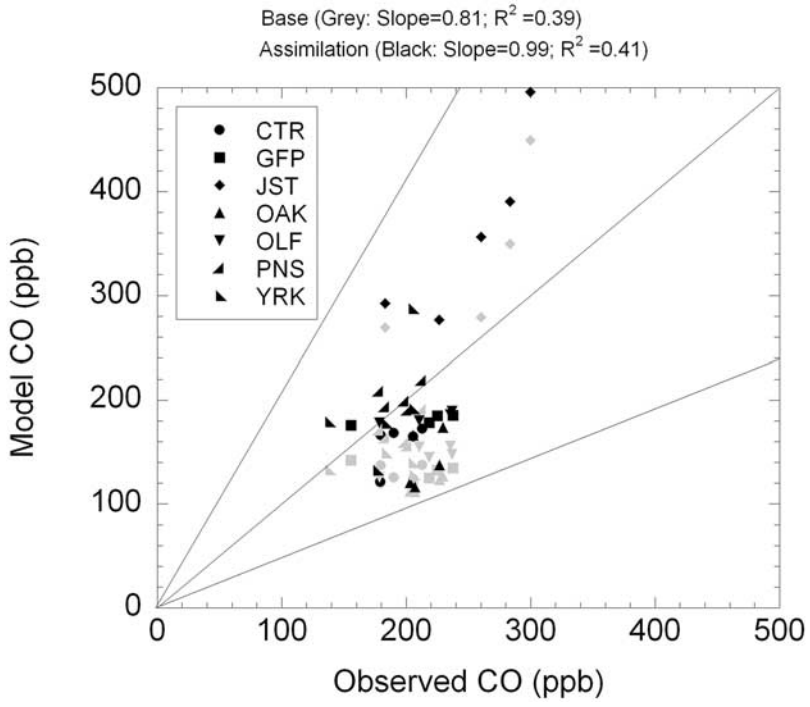


Figure 6. Comparison of model-predicted surface-level afternoon-average CO mixing ratios with measurements at the SEARCH network sites: Centreville, AL (CTR), Gulfport, MS (GFP), Jefferson Street, GA (JST), Oak Grove, MS (OAK), suburban site near Pensacola, FL (OLF), Pensacola, FL (PNS), and Yorkville, GA (YRK). Different symbols represent data at these seven locations. Grey and black symbols denote base and assimilation simulation predictions, respectively.

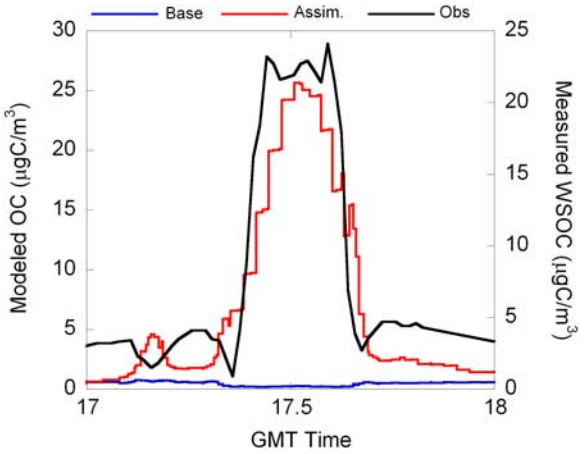


Figure 7. Comparisons of modeled organic carbon aerosol (OC) concentration prediction with water-soluble organic carbon (WSOC) measurements along the NOAA WP3 flight segment that intercepted a biomass burning plume on 20 July 2004. Observed mixing ratios are represented by the black line, base case simulation results by the blue line, and assimilation simulation results by the red line.

layer transport processes can efficiently mix the plume down to the surface. As can be seen in Figure 6, surface-level CO concentrations were generally under-predicted at all sites (except the Jefferson Street site in Atlanta) in the base simulation, which did not account for the impacts of the Alaskan fire plume. In contrast, significant enhancements in simulated surface-level daytime average CO mixing ratios at the SEARCH locations (up to 140 ppb) are noted in the assimilation case, which results in better agreement with measured daytime average values for these days.

4.2. Comparison with Aloft and Surface-Level Fine Particulate Matter Measurements

[23] The comparisons of simulated and observed surface-level and aloft CO mixing ratios discussed in the previous section established the efficacy of the assimilation methodology in capturing the three-dimensional pollution signature associated with the transport of the Alaskan smoke plume over the eastern United States troposphere and its subsequent downward mixing to the surface. In this section, the impact of the fire plume on surface-level $PM_{2.5}$ pollution is quantified through comparisons with measurements of total $PM_{2.5}$ from the AIRNOW network and with speciated fine particulate matter measurements from the Speciated Trends Network (STN). The U.S. EPA STN (http://www.epa.gov/air/data/aqsdb.html) observations are 24-hour 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 across the eastern United States. For the STN data used here, field blank measurements were used to blank correct the measured OC (Venkatesh Rao, personal communication, U.S. EPA, 2007) before comparing with the modeled values. In the analyses reported here, modeled surface-level daily (24-hour) average values are compared with corresponding measurements.

[24] Figure 7 presents comparisons of modeled organic carbon (OC) aerosol concentrations with measurements of water-soluble organic carbon (WSOC) aerosol on 20 July 2004 along the flight segment of the NOAA-WP3 that encountered the biomass burning plume. Similar to the CO comparisons in Figure 5, excellent agreement in the location and timing of the enhancements in modeled OC concentrations are noted for the assimilation case. Since the PILS (particle-into-liquid sampling) instrument only detects soluble organic carbon, the modeled OC concentrations are expected to be higher than the WSOC measurements. *Sullivan et al.* [2006] estimated a ΔWSOC/ΔCO enhancement ratio within the plume (above local background concentrations) of 69.4 μgC/m^3/ppmv for this case. A similar modeled enhancement ratio, ΔOC/ΔCO, can be computed by examining the ratio of the difference in predicted CO and OC between the assimilation and base simulations. For this case, the ratio was estimated to be 54 μgC/m^3/ppmv. This is in good agreement with the observed enhancement ratio, further suggesting consistency with the CO/$PM_{2.5}$ emission ratio used in initializing the model as described earlier in section 3.

[25] The impacts of the Alaskan fire plume on surface-level $PM_{2.5}$ pollution over the eastern United States during 19–23 July 2004 are illustrated in Figure 8, which compares daily-average modeled and measured (from the AIRNOW network) surface $PM_{2.5}$ concentrations. Figures 8a–8d present variations in domain mean metrics, while Figure 8e presents scatter plot comparisons of model-observed pairs for all sites within the modeling domain for the 19–23 July 2004 analysis period. Systematic under-predictions were noted in the base simulation which did not account for the long-range transport effects of the Alaskan fire plume. These under-predictions were rectified in the assimilation simulation at a majority of the sites as depicted in the scatter plot in Figure 8e. Additionally, for the 19–23 July 2004 period, the assimilation methodology helped improve the overall skill of the model as depicted by consistently lower regional mean bias, lower RMSE, and higher correlation coefficient.

[26] The ability of the assimilation methodology in representing the impacts of the fire plume on surface-level $PM_{2.5}$ pollution over the eastern United States is further illustrated in Figure 9, which compares model results from the base and assimilation simulations with surface measurements of $PM_{2.5}$ and carbonaceous aerosol from the STN. Illustrated in the scatter plots in Figure 9 are paired modeled and observed daily average values on days within the analysis period on which measurements were available from the STN. Similar to the comparisons with the AIRNOW measurements systematic under-predictions in $PM_{2.5}$ are also noted in the comparisons of the base simulation results with STN measurements (Figure 9a). The AOD assimilation methodology results in significant improvement in the $PM_{2.5}$ prediction skill as reflected by the improvements in slope and R^2 values. As further illustrated in Figure 9b, the under-predictions in surface $PM_{2.5}$ in the base simulation were largely associated with significant under-predictions (often greater than a factor of 2) in the total carbonaceous aerosol mass (TCM = organic + elemental), further illustrating the impacts of the fire plume on surface-level particulate matter pollution during this time period. The systematic under-predictions in the TCM were largely rectified in the assimilation simulation which not only

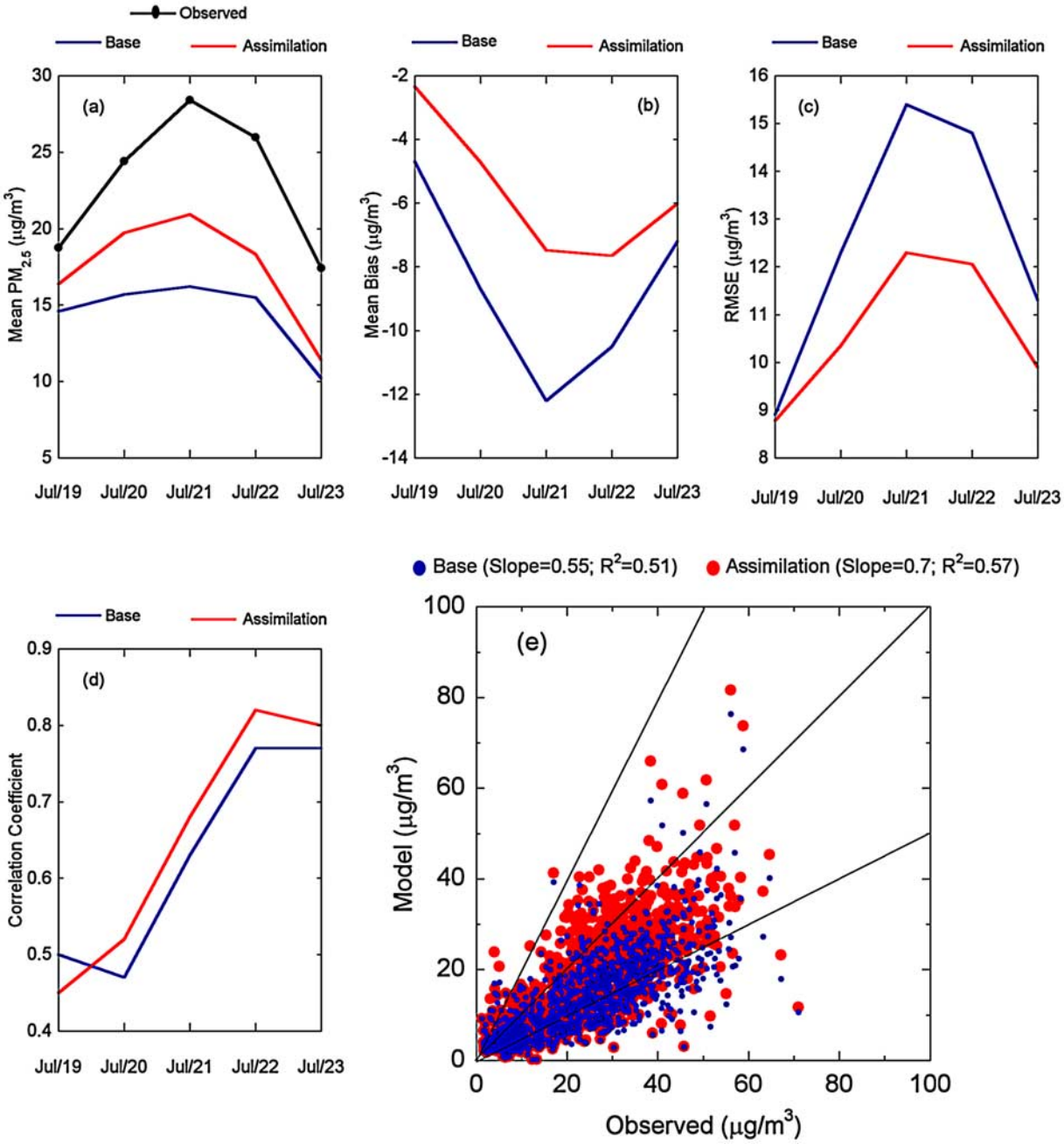
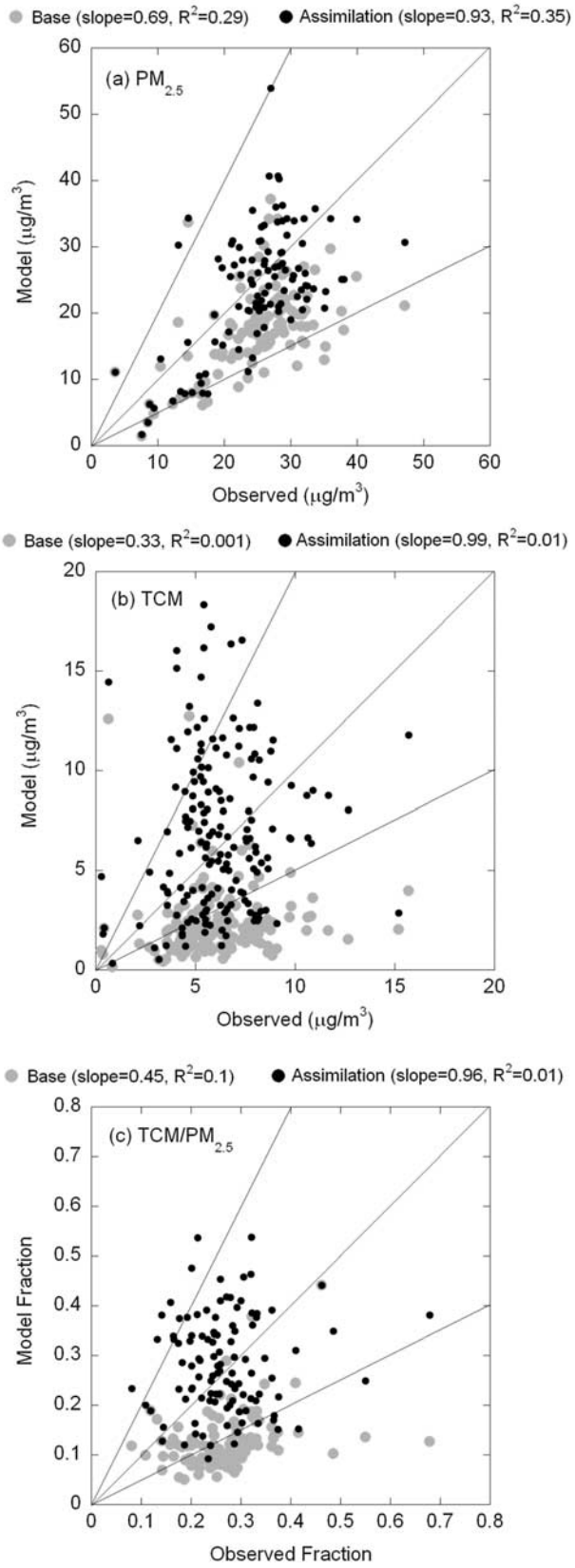


Figure 8. Simulated impact of the transport of pollution associated with the Alaskan fires on surface-level $PM_{2.5}$ pollution: comparisons of modeled surface-level daily-average $PM_{2.5}$ concentrations with measurements from the AIRNOW network. Comparisons in Figures 8a–8d based on spatial averages over all model-observed pairs for each day. (a) Comparison of domain-mean model predictions of $PM_{2.5}$ from the base simulation, assimilation simulation, and measurements (black); (b) variations in mean bias; (c) variations in RMSE; (d) variations in correlation coefficients; and (e) scatter plot of modeled and measured daily-average $PM_{2.5}$. Also shown are the 1:1, 1:2, and 2:1 lines and the slopes and R^2 values for the linear regressions.



improved the model's skill in predicting the TCM concentrations and spatial distributions but also its relative contribution to total $PM_{2.5}$ (Figure 9c).

4.3. Enhancement in Tropospheric Fine Particulate Matter Mass Loading Over the Eastern United States

[27] The cumulative $PM_{2.5}$ loading over eastern United States troposphere during the study period as a result of the long-range transport Alaskan fire plume can be estimated as the difference in the mass loading between the assimilation and base simulations at 1600 GMT on 19 July 2004 (time of re-initialization for the assimilation simulation). This represents the fine particulate mass transported to the eastern United States which then subsequently mixed down to the surface over the next few days. These estimates are tabulated in Table 1; also shown is the modeled total daily anthropogenic $PM_{2.5}$ emissions within the east United States domain and the annual fine particulate matter emissions over the continental United States estimated from the 2001 EPA National Emissions Inventory (NEI). On the basis of the model calculations presented here a cumulative tropospheric $PM_{2.5}$ enhancement of 0.12 Tg on 19 July 2004 is estimated; this amount is approximately 13 times greater than the daily fine particulate matter emissions within the modeled domain and about a factor of 8 higher than the daily fine particulate matter emission estimates for the entire continental United States.

5. Summary and Conclusions

[28] Recent studies have examined the effects of long-range transport of smoke plumes from forest fires burning in Alaska and western Canada during early July, 2004 in elevating the concentrations of CO, ozone, and ozone precursors over the continental and eastern United States [*Pfister et al.*, 2005; *Morris et al.*, 2006; *Warnecke et al.*, 2006; *de Gouw et al.*, 2006]. This study provides additional evidence of the episodic but large-scale regional impacts of this remote biomass burning event on surface-level particulate matter pollution over the eastern Unites States.

[29] The impacts of the long-range transport of pollution associated with wildfires in Alaska and Canada on episodic tropospheric fine particle pollution during the 19–24 July 2004 is estimated through the development of a methodology to assimilate remotely sensed aerosol optical depth information in a comprehensive regional chemical transport modeling system. To demonstrate the efficacy of the methodology in representing the three-dimensional structure and transport of the Alaskan fire plume, predicted CO mixing ratios were compared with both aloft measurements by the NASA DC-8 and NOAA WP-3 aircrafts, as well as surface

Figure 9. Comparison of model-predicted daily-average concentrations of $PM_{2.5}$ and constituents with measurements from the STN. (a) Comparisons for $PM_{2.5}$; (b) comparisons for total carbonaceous aerosol mass (TCM); and (c) comparisons for the TCM/$PM_{2.5}$ ratio. Grey solid circles represent base simulation results while the black solid circles represent the assimilation simulation results. Slopes and R^2 for individual species correlations are indicated. Also shown are the 1:1, 1:2, and 2:1 lines.

Table 1. Model Estimates of Tropospheric $PM_{2.5}$ Mass Loading on 19 July 2004

Emissions/Simulation Case	$PM_{2.5}$ Burden
Modeled $PM_{2.5}$ emissions (east United States on 7/19/04)[a]	0.0089 Tg
Estimated $PM_{2.5}$ emissions (continental United States)[b]	0.0137 Tg
Base case[c]	0.18 Tg
Assimilation case[c]	0.30 Tg
Difference (assimilation-base)[c,d]	0.12 Tg

[a]24-h total emissions on 19 July 2004 used in model simulations.

[b]On the basis of 5 Tg annual $PM_{2.5}$ emissions over continental U.S. from the U.S. EPA 2001 National Emissions Inventory (5/365 = 0.0137).

[c]Tropospheric $PM_{2.5}$ mass loading over the east U.S. model domain at 1600 GMT on 19 July 2004.

[d]Difference between the assimilation and base case $PM_{2.5}$ loadings represents the cumulative impact of the Alaskan fire plume on the eastern U.S. troposphere up to 19 July and represent a lower limit.

measurements from the SEARCH network. The base case simulation, which did not account for the inflow of pollution associated with the transport of the Alaskan plume to the eastern United States, significantly underestimated aloft CO concentrations when the aircraft measurements detected the presence of the fire plume. In contrast, the assimilation simulation provided excellent agreement with measurements when the model predictions and aircraft measurements were paired in space and time, even 24–48 hours after the assimilation initialization, suggesting that the model was able to accurately capture the three-dimensional transport of the fire plume after intrusion into the eastern United States troposphere. The assimilation methodology employed here also resulted in significant improvement in the model's skill in simulating surface-level CO concentrations during the study period; enhancements up to 140 ppb in surface-level daytime-average CO mixing ratios were simulated at the locations of the SEARCH sites.

[30] The impact of the Alaskan fire plume on three-dimensional fine particulate matter distributions were assessed through comparisons with both aloft and surface measurements. Limited comparison of modeled organic carbon aerosol concentrations with measurements of water soluble organic carbon measurements from the PILS instrument onboard the NOAA WP3 during segments when the aircraft intercepted the biomass burning plume on 20 July, 2004 also indicates that the assimilation methodology was able to capture the enhancements (relative to background) in organic carbon aerosol within the air-mass impacted by pollution associated with biomass burning. Further, for this case the modeled $\Delta OC/\Delta CO$ enhancement ratio of 54 $\mu gC/m^3$/ppmv, is consistent with the estimated measured enhancement ratio (69.4 $\mu gC/m^3$/ppmv from *Sullivan et al.* [2006]), suggesting agreement between the methodology for speciating the assimilated particulate mass in the model and the observed chemical composition within the fire plume.

[31] Comparison of the surface-level modeled and measured total $PM_{2.5}$ concentrations, at locations of the AIRNOW and STN monitors, also shows that the large and systematic under estimations that occur in the base case simulation were largely rectified in the assimilation case simulation which provided substantially improved model skill (higher slope and R^2) in predicting the observed spatial variability in surface-level $PM_{2.5}$. On the basis of these comparisons, the model calculations suggest that during 19–23 July 2004, because of the long range transport of pollution associated with wildfires in Alaska and Western Canada, median concentrations of surface-level $PM_{2.5}$ were enhanced by 33% at the location of the STN monitors and by 24–42% (on different days) at the locations of the AIRNOW monitors. Comparisons of modeled $PM_{2.5}$ constituent concentrations with corresponding STN measurements further show that the enhancements in $PM_{2.5}$ during this time period are largely associated with enhancements in total carbonaceous aerosol mass associated with pollution from biomass burning. The simulated impacts of this episodic long-range transport event on surface-level fine particulate matter concentrations in the eastern United States are significantly larger than those suggested by previous long-range transport assessment studies. For instance, model calculations with the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model by *Chin et al.* [2007] suggest that on an annual basis, majority of the surface-level fine particulate mass over the United States is from North American regional sources with long-range transport contributing only ~2–4% of the surface-level fine particulate matter mass in the eastern United States, with dust constituting a major fraction of the imported fine particulate matter mass. The results of this study however suggest that under certain conditions the episodic contribution of long-range transport to $PM_{2.5}$ pollution can be significantly larger than previously documented. Additionally, the transport of pollution associated with distant biomass burning sources can in such cases contribute to significant enhancement in carbonaceous aerosol content of surface-level $PM_{2.5}$.

[32] On the basis of the model calculations, a 0.12Tg enhancement in tropospheric $PM_{2.5}$ mass loading over the eastern United States is estimated on 19 July 2004 because of this long-range transport. This amount is approximately 8 times the total daily anthropogenic fine particulate matter emissions for the continental United States. It should be noted that this estimate is perhaps a lower bound since: (1) the assimilation methodology used here only initialized the model on the first day whereas in actuality the inflow of pollution associated with the Alaskan plume also occurred on subsequent days though perhaps at a lesser magnitude, and (2) enhancements in particulate matter column loading was not considered in the calculations at locations where no MODIS aerosol optical depth retrievals were available (missing data or cloud interference). Nevertheless, the results of this study indicate that large forest fires in the remote boreal regions of Alaska and Canada can have a significant impact on surface-level particulate matter pollution over large regions of the eastern United States.

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