Simulations of fine particulate matter (PM$_{2.5}$) in Houston, Texas

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Simulations of fine particulate matter (PM$_{2.5}$) during an eight-day episode (24 to 31 August 2000) is conducted in association with the 2000 Texas Air Quality Study (TexAQS 2000) and the Houston Supersite Project using the EPA’s Models-3 Community Multiscale Air Quality model (CMAQ). The mass concentrations of PM$_{2.5}$ and major chemical constituents during the episode are calculated and compared with available field measurements. The predicted daily PM$_{2.5}$ mass concentrations are about 8.5–13.0 $\mu$g/m$^3$, consistent with the observed values. The diurnal patterns of PM$_{2.5}$ mass concentrations are similar throughout the region, with a strong morning peak and a weak peak in the late afternoon to the early evening. High primary emissions, high formation rates of the secondary fine particulate matter, and low planetary boundary layer (PBL) heights contribute to the morning peak. The major components of the fine particulate matter in this region are sulfate, organic carbon, elemental carbon and ammonium. The model predicts about 30% sulfate, 32% organics (including elemental carbon (EC)), and 10% ammonium of the total PM$_{2.5}$ mass. The balance of the primary cations and anions indicates that fine particulate matter in this region is acidic. Comparison with field observation reveals that CMAQ produces good simulations of averaged daily mass concentrations of major components such as sulfate, organic carbon, elemental carbon and ammonium with normalized mean biases (NMB) of less than ±25%. Uncertainties in the aerosol precursor emissions, the aerosol chemistry especially about secondary organic aerosol (SOA) formation and aqueous reactions, and the calculated PBL heights are likely responsible for the differences.

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1. Introduction

[2] Aerosols or fine particulate matter, PM$_{2.5}$, in the atmosphere have been of great interest in recent years [Environmental Protection Agency (EPA), 1996, 2000; Zhang et al., 2004a]. Concerns over human health effects of fine particulate matter constitute the most important element in formulating the national ambient air quality standard [EPA, 1996]. High levels of particulate matter are believed to be associated with adverse human effects, including increased morbidity and mortality arising from altered respiratory and cardiovascular function. Aerosols directly or indirectly affect the Earth’s energy balance [Charlson et al., 1987; Intergovernmental Panel on Climate Change, 2001]. The aerosol direct effect occurs under clear-sky conditions by scattering or absorbing solar radiation. The aerosol indirect effect takes place by increasing cloud particle concentrations and enhancing cloud reflection of solar radiation. The indirect effect on the energy budget is also related to the earth hydrological cycle, since a smaller particle size is unfavorable for growth of cloud particles to raindrops and hence inhibits precipitation and prolongs the lifetime of the cloud [Orville et al., 2001]. Modification of cloud and precipitation by aerosols may enhance lightning activity and thus influence tropospheric chemistry [Orville et al., 2001; Zhang et al., 2003]. Furthermore, aerosols contribute to regional acid deposition and visibility degradation [EPA, 1996, 2000], and promote multiphase chemical processes [Zhang et al., 1993a, 1993b; Zhang and Leu, 1997; Zhao et al., 2005].

[3] Unlike ozone which is being treated a single pollutant [Lei et al., 2004; Zhang et al., 2004b], PM$_{2.5}$ consists of mixtures of various chemical components and over a wide size distribution resulting from physical and chemical processes that make PM$_{2.5}$ a difficult pollutant to model. Modeling of particulate matter is complicated because of the lack of adequate measurements for comparison. Recently, hourly PM$_{2.5}$ measurements with Tapered Element Oscillating Microbalance (TEOM) and Federal Reference Method (FRM) during the Houston Supersite Project provide valuable data to evaluate modeling results in
Measurements also show that the average PM2.5 is close to 15 μg/m^3 for the annual average concentration. Analyses of TEOM and FRM measurements also show that the average PM2.5 is close to NAAQS and the major components of PM2.5 are organic aerosol of both anthropogenic and biogenic origins, elemental carbon, and sulfate, both comprising ~30% of the fine particulate matter mass in southeast Texas. The mass concentrations and compositions of the fine particulate matter are spatially homogeneous in the region. A strong morning peak in PM2.5 mass concentrations is observed throughout the region and a weak and slightly less consistent peak is observed in the late afternoon to early evening. The study also reveals that the aerosol is slightly acidic and much of the carbonaceous material is due to secondary organic aerosol formation.

Air quality models such as the EPA's Models-3 Community Multiscale Air Quality model (CMAQ), are useful tools to assess the current and further air quality regulations to protect human health and welfare. The CMAQ aerosol component, or module, designed to simulate the complex processes involving PM, is derived from the Regional Particulate Model (RPM). The aerosol species considered within the CMAQ aerosol module include sulfate, nitrate, ammonium, water, primary organic aerosol, secondary organic aerosol of both anthropogenic and biogenic origins, elemental carbon, primary material not otherwise specified and wind-blown dust. Two different modes, the Aitken and accumulation modes, each having variable standard deviations, represent PM2.5 particles in the CMAQ aerosol module. Conceptually, the Aitken mode represents fresh particles resulting from nucleation and/or direct emissions. The model performance for visibility and SOA formation in this region.

2. Methods

The model domain was comprised of 60 × 60 grid cells encompassing an area of 57,600 km^2 with a 4-km resolution centered at 29.83N and 95.05W. The horizontal grid specification of the domain followed that of the 4-km MM5 meteorological grids, except removing the outer 35 horizontal rows of MM5 grid points to reduce the boundary effects. There were 21 layers in the vertical direction from the surface to the top, identical to those of gridded emission data. Initial and boundary conditions were generated from the CMAQ simulation results with 12-km horizontal resolution and 45 × 45 grid cells centered at 30.04N and 95.72W, using CMAQ initial condition (ICON) and boundary condition (BCON) processors.

The meteorological fields for chemical transport simulations were generated during TexAQS 2000 by J.W. Nielsen-Gammon (Meteorological modeling files, 2003, available at http://www.tnrcc.state.tx.us/air/asp/airquality_photom.html#met) using MM5, which incorporated wind profiler and GOES satellite data. The MM5 output files were post-processed for the model domain by EPA's Models-3 Meteorology-Chemistry Interface Processor Version 2.2 (MCIP2). During the processing, vertical layer collapsing was performed to reduce the vertical layers from 43 sigma levels (42 vertical layers) to 22 sigma levels (21 vertical layers) for alleviating the computational costs associated with a larger number of vertical layers.

The emission inventory used in this study was from EPA's National Emission Inventory (NEI99) final version 3. The raw NEI99 data were converted into the Inventory Data Analyzer (IDA) and processed using Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System Version 1.5 by Lin et al. (2002) to obtain the gridded emission inventory data sets, ready for CMAQ runs. The Houston region contains a mix of major sources of ozone and SOA precursors. Houston is the tenth largest metro-plitic area in the United States, and hosts one of the world largest petrochemical complexes and several large fossil-fueled electric power plants. Transportation and industry emit a great quantity of VOCs, NOx, and SO2. In particular, the Houston Ship Channel region, east of the city center, represents major high emission sources of VOCs from petrochemical plants [Lei et al., 2004; Zhang et al., 2004b], which are believed to the major contributor to ozone and SOA formation in this region [Daum et al., 2003]. It is known that NEI99 inventory underestimated emissions of VOCs [Lin et al., 2002]. Compared with the emission inventory data with modified olefin emissions used for CAMx (Comprehensive Air Quality Model with extension) simulations provided by Texas Commission for Environmental Quality (TCEQ), the emission quantities of olefins in NEI99 data are about 3–10 times smaller in the southeast of Texas. Therefore, we increased the olefin emissions to 8 times of the original quantities in this study in order to more accurately represent VOC emissions in this area. Also, because of the very low aromatic hydrocarbon emissions reported in NEI99 data and much uncertainty during the Houston Supersite Project [Allen, 2002; Russell et al., 2004].
about the emissions of aromatics in Houston [Karl et al., 2003], the emissions of aromatics such as toluene and xylene were also boosted by 8 times.

[9] CMAQ Version 4.3 was employed in this study, which has major modifications on the treatment of secondary organic aerosol (SOA) formation and the heterogeneous N2O5 reaction probability [EPA, 2003]. The chemical mechanism chosen in this study was RADM2 with aqueous chemistry extension. The aerosol module AERO3 was used, representing the newest version released in 2002. Schell algorithm was used to calculate the secondary organic aerosol (SOA) formation rate [Schell et al., 2001]. Both the Aitken and accumulation modes contributed to the PM2.5 number concentrations. We presented in this work only the mass concentration of PM2.5, which is mainly contributed by the accumulation mode. Also, there were few measurements of new particle formation during this episode. The Modified Euler Backward Interactive (MEBI) gas-phase chemistry solver was used for the RADM2 chemical mechanism. The Models-3/CMAQ model system was built on Linux platform using Portland Group FORTRAN 90 (PGF90) compilers.

[10] The selected simulation period runs from 24 to 31 August 2000, corresponding to an 8-day episode associated with the TexAQS 2000 and Houston Supersite Project. This period corresponds to typical summertime emissions and meteorological conditions in this region. This episode had two distinct meteorological regimes in the Houston area. From 25 to 29 August, the wind was predominantly southeasterly. In contrast, the wind was predominantly westerly on 30 and 31 August. During 25 August, a large-scale wind of close to 3 m s⁻¹ led to a few hours in which coastal circulation caused the wind to become calm and gradually reverse. No stagnation events were seen during the next 4 days since the large-scale wind was slightly stronger. The temperature distribution during 25 to 29 August did not change significantly. The Houston area was essentially cloud-free with the exception of a few isolated showers on 26 and 29 August. On 28 August the sky was cloudy but without showers. The composite of winds showed little vertical wind shear during this period. On 30 and 31 August, the Houston area was characterized with high temperatures and light winds in the afternoon (near-stagnation). Instead of the similar wind variations with height, the diurnal cycle decayed to near zero by a height of 1 km; and instead of wind variation of a few meters per second, the wind changed by more than 10 m/s on average between sunset and sunrise [Nielsen-Gammon, 2001, 2002]. The MM5 results employed in this study reasonably reproduced the sea breeze circulation and urban heat island effect characteristic of this region, with an overall root-mean square accuracy of about 15 m/s [Lei et al., 2004; Nielsen-Gammon et al., 2005].

[11] The study domain in Houston covered the latitude from 29.60°N to 29.96°N and longitude from 95.62°W to 95.02°W (the box in Figure 1), since the relatively higher PM2.5 mass concentrations above the annual average standard frequently occurred in this area. In addition, PM monitoring sites were concentrated in this area, which was ideal for the model evaluation by comparing with the observations. Figure 1 shows the locations of the sites for TEOM and FRM measurements within the study domain. The site descriptions are summarized in Table 1: Except for two urban sites (HALC and BAYP), most of the aerosol monitoring stations are located near the industrial Ship Channel region.

3. Results and Discussion

[12] The diurnal variation of ground-level PM2.5 mass concentrations averaged over the study domain and over eight simulated days is depicted in Figure 2. A strong maximum

![Figure 2. Simulated PM2.5 mass concentrations averaged over the study domain and the episode.](image-url)
with the highest concentration about 21.0 µg/m³ occurred around 8:00 am, and a weaker maximum (13.0 µg/m³) took place around 8:00 pm. The average hourly PM$_{2.5}$ mass concentrations from 6:00 am to 9:00 am (morning maximum) were slightly above the NAAQS annual average standard, 15.0 µg/m³. The temporal variation of PM$_{2.5}$ at different locations was consistent throughout the region. A slight difference (less than 10\%) of PM$_{2.5}$ mass concentrations between the weekdays and weekend indicated that the traffic sources did not significantly contribute to the aerosol concentrations in Houston.

[13] The geographic distributions of PM$_{2.5}$ mass concentrations at 8:00 am and 3:00 pm (local time) averaged over the episode in Houston are displayed in Figure 3a and Figure 3b, respectively. At 8:00 am, a morning peak occurred with the PM$_{2.5}$ mass concentration exceeding 15.0 µg/m³ (Figure 3a). Especially in and around the Houston urban center and the Ship Channel, the PM$_{2.5}$ mass concentrations were higher than 20.0 µg/m³. Along the periphery of Houston, tongues of relatively high PM$_{2.5}$ concentrations were located within major roadways. The model captured localized events with the PM$_{2.5}$ mass concentrations greater than 40.0 µg/m³ in and around the Ship Channel. Those localized events have been observed frequently at the monitoring sites located in this area and they were important in determining compliance with the NAAQS. At about 3:00 pm, the PM$_{2.5}$ concentration reached the lowest value during the day. The geographic distribution at this time showed similar patterns to those of the morning maximum, except the absolute PM$_{2.5}$ mass concentrations were much lower (see Figure 3b). The entire area in Houston had PM$_{2.5}$ mass concentrations of about 5.0–8.0 µg/m³. PM$_{2.5}$ concentrations were relatively higher (8.0–15.0 µg/m³) in the locations near the Ship Channel plumes, owing to formation of secondary fine particulate matter such as sulfate and organics. Therefore the emission sources from the power plants and petrochemical plants contributed significantly to the high PM$_{2.5}$ concentrations in this area. It is clear from Figure 3 that the ratio of PM$_{2.5}$ mass concentrations in the urban to the surrounding rural area is generally greater than 10, indicating that regional transport of PM$_{2.5}$ from the surrounding region is minimal.

Figure 3. Geographical distribution of simulated PM$_{2.5}$ concentrations at (a) 8:00 am and (b) 3:00 pm averaged during the episode. The time corresponds to the local time or central daylight time (CDT).

Figure 4. Similar to Figure 3 except for geographical distribution of averaged OC concentrations.
compared to the PM$_{2.5}$ production from urban and industrial sources in Houston.

[14] Organic carbon (OC) and sulfate are the most major components of fine particulate matter in the Houston area [Allen, 2002; Russell et al., 2004]. The time-averaged geographic distributions of OC and sulfate are shown in Figures 4 and 5, respectively. OC and sulfate have similar geographic distributions as those of PM$_{2.5}$ shown in Figure 3. At 8:00 am, the entire region encountered a relatively high concentration of organic carbon over 5.0 mg/m$^3$. Higher organic carbon concentrations (greater than 8.0 mg/m$^3$) were predicted in and around the Houston urban center and Ship Channel (Figure 4a). At 3:00 pm, the concentration of organic carbon was low with the value of about 1.0–2.5 mg/m$^3$ (Figure 4b). Compared to organic carbon, sulfate was less spatially homogeneous. High sulfate mass concentrations occurred in and around Ship Channel, with the values of over 8.0 mg/m$^3$ at 8:00 am and over 3.5 mg/m$^3$ at 3:00 pm (Figures 5a and 5b). Point sources burning sulfur-containing fuels around this area were likely responsible for this inhomogeneity.

[15] The averaged PM$_{2.5}$ mass composition is presented in Figure 6. Sulfate, organic carbon, elemental carbon and ammonium are the major constituents of PM$_{2.5}$ in the Houston area. Sulfate consists of about 30% of the total average PM$_{2.5}$ mass. Organic carbon and elemental carbon makes up about 32% of the total PM$_{2.5}$ mass. Ammonium and nitrate account for about 10% and 1% of the total PM$_{2.5}$ mass, respectively. The unspecified component is predicted to be 27% by the model.

[16] The comparisons against FRM measurements were made for the sites where the composition data were available. Table 2 shows that the comparisons between modeled and observed mass concentrations of the major constituents of PM$_{2.5}$ for six sites. The mass concentrations of each component shown in this table are averaged over the days when the measurements are available. Table 2 reveals that the simulated mass concentrations of the components are in good agreement with the observations except for nitrate. For organic carbon, sulfate, elemental carbon, and ammonium, the mean biases are $-0.51$, $-0.87$, $0.04$, and $0.23$ mg/m$^3$, respectively; the corresponding normalized mean biases (NMB) are $-17.4\%$, $-20.76\%$, $9.06\%$, and $24.20\%$. Those values are less than $\pm 25\%$, indicating good agreement. The simulated concentrations of organic carbon and sulfate are 0.51 and 0.87 mg/m$^3$ less than the measurements, respectively. The mean bias for nitrate is $-0.21$ mg/m$^3$ and the NMB value is $-61.46\%$, indicating a significant underestimation for nitrate. The underprediction by the model with respect to nitrate may be explained by low ammonia emissions in the summer conditions based on the NEI99 [Mebust et al., 2003].

[17] Averaged over the seven monitoring sites in Houston, the daily PM$_{2.5}$ mass concentrations are about 8.5–13.0 mg/m$^3$, in agreement with the TEOM observed values, 8.0–14.0 mg/m$^3$. The modeled hourly PM$_{2.5}$ mass concentrations were compared with the available TEOM measurements. There were only three sites with the hourly PM$_{2.5}$ data within the study domain, i.e., CNNC, DRPK and HOEA. Figure 7 presents the comparisons of modeled hourly PM$_{2.5}$ mass concentrations averaged over the simulation period against the measurements for the three sites. Generally, the model simulations of hourly PM$_{2.5}$ concentrations were reasonable compared with the observations. The model reproduced the diurnal varia-

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**Figure 5.** Similar to Figure 3 except for geographical distribution of averaged sulfate concentrations.

**Figure 6.** Chemical compositions of simulated PM$_{2.5}$ in the Houston area averaged over the study domain and the episode.
tions, although the predicted peak time was about 1–2 hour later than the observed. The model overpredicted the morning peaks and underestimated the PM$_{2.5}$ concentrations in the afternoon for all sites. Figure 8 presents the diurnal variations of the primary emitted PM$_{2.5}$ and the calculated planetary boundary layer (PBL) heights averaged over the monitoring sites and simulation period. In the early morning (about 8:00 am), the primary emitted PM$_{2.5}$ concentration was

Table 2. Site-by-Site Comparisons of the Major Components of PM$_{2.5}$ Between Modeled and Observed Values

<table>
<thead>
<tr>
<th>Sites</th>
<th>OC, µg/m$^3$</th>
<th>Sulfate, µg/m$^3$</th>
<th>Nitrate, µg/m$^3$</th>
<th>EC, µg/m$^3$</th>
<th>Ammonium, µg/m$^3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>CNNC</td>
<td>3.00</td>
<td>2.20</td>
<td>4.05</td>
<td>5.02</td>
<td>0.22</td>
</tr>
<tr>
<td>BAYP</td>
<td>2.50</td>
<td>3.03</td>
<td>3.59</td>
<td>1.79</td>
<td>0.34</td>
</tr>
<tr>
<td>DRPK</td>
<td>2.50</td>
<td>2.02</td>
<td>4.08</td>
<td>2.33</td>
<td>0.36</td>
</tr>
<tr>
<td>H03H</td>
<td>3.74</td>
<td>3.04</td>
<td>4.95</td>
<td>5.37</td>
<td>0.40</td>
</tr>
<tr>
<td>LAPO</td>
<td>2.45</td>
<td>1.70</td>
<td>4.33</td>
<td>2.70</td>
<td>0.39</td>
</tr>
<tr>
<td>HALC</td>
<td>3.88</td>
<td>3.00</td>
<td>4.10</td>
<td>2.68</td>
<td>0.30</td>
</tr>
<tr>
<td>Mean</td>
<td>3.01</td>
<td>2.50</td>
<td>4.18</td>
<td>3.32</td>
<td>0.34</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>Mean bias</th>
<th>Sulfate, µg/m$^3$</th>
<th>Nitrate, µg/m$^3$</th>
<th>EC, µg/m$^3$</th>
<th>Ammonium, µg/m$^3$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>−0.51</td>
<td>−0.87</td>
<td>−0.21</td>
<td>0.04</td>
<td>0.23</td>
</tr>
<tr>
<td></td>
<td>−17.40</td>
<td>−20.76</td>
<td>−61.46</td>
<td>9.06</td>
<td>24.20</td>
</tr>
</tbody>
</table>

Figure 7. Site-by-site comparisons of the simulated and measured hourly PM$_{2.5}$ mass concentrations. The simulated and measured values are averaged over the time period when the observations are available.
very high because of automobile emissions. The calculated PBL height also reached the lowest value around sunrise. Hence the simulated strong morning peak is apparently associated with the PBL height.

The ratio of organic carbon to elemental carbon is often used to distinguish the relative importance of primary and secondary organics. If the predicted ratios of OC/EC are higher than those assumed to occur from primary emissions (a ratio between 2 and 5 is generally assumed for OC/EC from primary emissions [Strader et al., 1999], the excess OC is assumed to be due to secondary organic aerosol (SOA) formation. Figure 9 shows the scatterplots of OC/EC ratios in the study domain on 26 and 27 August. About 42% of the OC/EC ratios are above 5.0, suggesting that most of the OC is not due to the secondary organic aerosol (SOA) formation in our simulations. Since the measurements indicate that much of organic carbon is from SOA formation in this region [Allen, 2002; Russell et al., 2004], the modeled SOA formation is likely underpredicted.

Figure 10 shows the temporal variations of organic carbon, sulfate and SOA averaged over the monitoring sites and the simulation days. The formation of the secondary fine particulate matter such as sulfate and SOA reaches the highest in the morning because of strong emissions, low calculated PBL heights, and rapid photochemical activity after sunrise, contributing to the strong morning peak. Most of organic carbon arises from the primary sources. The SOA formation is rather low, with the highest value of about 1.7 μg/m³. SOA accounts for about 28% of the organic carbon. Field measurements, however, imply that SOA accounts mainly for the organic carbon mass [Allen, 2002; Russell et al., 2004]. The model simulations suggest that
about 40% of SOA is formed from anthropogenic precursors and 60% is formed from biogenic precursors. The underprediction in SOA is likely caused by underprediction of the emissions of anthropogenic SOA precursors. A recent measurement [Karl et al., 2003] shows that the emissions of aromatic compounds such as benzene and toluene are dramatically underestimated in the previous emission inventories in Houston. It is likely that the increase in the emission inventory for SOA precursors in our simulations may still be inadequate. Also, the SOA formation chemistry especially related to aqueous reactions within the CMAQ aerosol module might be inadequate, since the SOA production was not very sensitive to the increase of the SOA precursor emissions.

The overall acidity of the fine particulate matter can be assessed through a cation-anion balance, based on the chemical composition data. Since the nitrate mass is low and sulfuric acid is the primary source of the acidity in Houston, the majority of the ammonium is expected to neutralize sulfate to form ammonium sulfate (\((\text{NH}_4)_2\text{SO}_4\)) or ammonium bisulfate (\(\text{NH}_4\text{HSO}_4\)). The scatterplot of sulfate concentrations to ammonium concentrations in equivalents over the study domain on 26 and 27 August 2000 is shown in Figure 11 (assuming all equivalents from the model predictions for the study domain on 26 and 27 August 2000).

The diurnal patterns of PM\(_{2.5}\) mass concentrations are consistent nearly throughout the region, with a strong morning peak and a weaker peak in the late afternoon to the early evening. High primary emissions, high formation rates of the secondary fine particulate matter such as sulfate and SOA, and low PBL heights contribute to the morning peak. The geographical distribution indicates that the PM\(_{2.5}\) mass concentration is rather spatially homogeneous throughout the Houston area. The major components of the fine particulate matter in the area are sulfate, organic carbon, elemental carbon and ammonium. Their predicted contents are about 30%, 27%, 5%, and 10% of total PM\(_{2.5}\) mass, respectively. The balance of the primary cations and anions indicates that fine particulate matter in this region is acidic. The model yields good simulations of the averaged daily mass concentrations of the major components such as sulfate, organic carbon, elemental carbon and ammonium with the NMBs less than ±25%. However, the model underpredicts nitrate with the NMB value of about 61%. Uncertainties in the aerosol precursor emissions, the aerosol chemistry especially about SOA formation and aqueous reactions, and the PBL heights are likely responsible for the differences.

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