SPECIAL
NOAA/EPA Golden Jubilee
ISSUE

Seasonal Simulation of Tropospheric Ozone over the Midwestern and Northeastern United States: An Application of a Coupled Regional Climate and Air Quality Modeling System

HO-CHUN HUANG, XIN-ZHONG LIANG, KENNETH E. KUNKEL, MICHAEL CAUGHEY, AND ALLEN WILLIAMS

Illinois State Water Survey, Champaign, Illinois

(Manuscript received 11 January 2006, in final form 26 September 2006)

ABSTRACT

The impacts of air pollution on the environment and human health could increase as a result of potential climate change. To assess such possible changes, model simulations of pollutant concentrations need to be performed at climatic (seasonal) rather than episodic (days) time scales, using future climate projections from a general circulation model. Such a modeling system was employed here, consisting of a regional climate model (RCM), an emissions model, and an air quality model. To assess overall model performance with one-way coupling, this system was used to simulate tropospheric ozone concentrations in the midwestern and northeastern United States for summer seasons between 1995 and 2000. The RCM meteorological conditions were driven by the National Centers for Environmental Prediction/Department of Energy global reanalysis (R-2) using the same procedure that integrates future climate model projections. Based on analyses for several urban and rural areas and regional domains, fairly good agreement with observations was found for the diurnal cycle and for several multiday periods of high ozone episodes. Even better agreement occurred between monthly and seasonal mean quantities of observed and model-simulated values. This is consistent with an RCM designed primarily to produce good simulations of monthly and seasonal mean statistics of weather systems.

1. Introduction

Human activities have led to several important air quality issues, such as elevated tropospheric ozone (O_3), particulate matter, and visibility problems, which affect human health and the natural environment. Anthropogenic increases in atmospheric concentrations of greenhouse gases also are increasing concerns about future climate change (Houghton et al. 2001). Global warming could have detrimental effects on future air quality, such as increased frequency of harmful tropospheric O_3 episodes (Hogrefe et al. 2004b). Rising temperatures could increase emissions of pollutants and certain biogenic volatile organic compounds (VOCs; Guenther et al. 1994), as well as enhance rates of atmospheric chemical reactions. In addition to mean temperature changes, there also could be shifts in the frequency and intensity of weather systems, resulting in changes of transport, mixing patterns, and removal of chemical species (Berkowitz et al. 1998, 2000; Fast et al. 2000; Lu et al. 2000; Doran et al. 2003), as well as atmospheric optical properties for photolytic reactions (e.g., cloud cover and incident solar radiation). Therefore, it is important to understand the potential impact of climate change on future air quality and prepare possible solutions to alleviate and/or resolve any significant negative impacts.

To identify solutions to air quality problems, a modeling system, including meteorological conditions, emissions, and air quality components, often is used to explore the causes and help identify control strategies. Past episodes, no more than a few days in length, typically are chosen for this purpose. Such an assessment approach is valid, assuming meteorological conditions favorable for high pollution levels remain the same. However, to address the potential influence of future climate change, seasonal-to-interannual simulations (Fast et al. 2002; Fast and Heilman 2003, 2005) are needed to assess future changes in air quality (Bouchet et al. 1999b; Hogrefe et al. 2001, 2004a,b). To this end, the Illinois State Water Survey has developed a modeling system that consists of a regional climate model

Corresponding author address: Ho-Chun Huang, Illinois State Water Survey, 2204 Griffith Dr., Champaign, IL 61820.
E-mail: huang2@uiuc.edu

DOI: 10.1175/JAM2521.1

© 2007 American Meteorological Society

(RCM), an emissions model (EM), and an air quality model (AQM).

The meteorological model is the central component of the air quality modeling system. It provides not only the description of gas transport and local chemical reactivity to the AQM, but also the environmental conditions for biogenic emissions and for specifying the vertical distribution of point sources to the EM. In traditional air quality studies, the four-dimensional data assimilation (FDDA) technique often is used to reproduce the weather of past adverse air quality events within a mesoscale meteorological model (MMM). To simulate past events, the MMM normally assimilates all available measurements over the whole modeling domain and the entire episode to produce the most realistic proxy of the observed meteorological conditions. This procedure keeps the simulated episodic or seasonal meteorological behavior from drifting too far from observations (Seaman 2000).

For future air quality studies, events are not known and long simulations must be performed to produce stable climatologies of these infrequent adverse events. For long simulations, the RCM, developed from the MMM, is the more appropriate tool because it incorporates physical parameterizations better suited to simulate spatial and temporal variability at climatic time scales. For example, Liang et al. (2004) identified that the cloud–radiation scheme in the MMM needed to be improved to correct the solar radiation deficit reaching the surface, an issue of minor importance for the typical MMM simulation periods of a few days but of critical importance for RCM simulations of a month or more. This allowed the RCM to produce more realistic characteristics of geographic distributions and seasonal variations.

Furthermore, it is obvious that for modeling future climate change, the use of FDDA is not ideal. The only possible input data for the FDDA technique is from climate "projections" of a general circulation model (GCM), the use of which would incorporate all of the regional climate biases of the GCM into the MMM simulation. Most RCMs are driven by lateral boundary conditions (LBCs) at their domain edges for the entire period (Liang et al. 2001; Bouchet et al. 1999a,b). These LBCs are provided either by global reanalysis data for validation/evaluation purposes or by GCM simulations for projections of future climate. The RCM integrates the LBCs and generates its own mesoscale circulation within the modeling domain, producing much better regional climate simulations than the driving GCMs. Liang et al. (2006) demonstrated that the RCM used in this study reproduced more realistic regional precipitation and temperature patterns than the driving GCM.

The AQM integrates RCM and EM inputs to simulate the evolution of chemical species through physical and chemical processes. Biases generated in the RCM meteorological conditions and emissions ultimately affect the accuracy of AQM simulations. In this study, LBCs constructed from global reanalysis data actually possess substantial uncertainties, especially over the oceans and the Tropics (Liang et al. 2001, 2004). These uncertainties may limit the ability of RCMs to reproduce observed regional climate variations. Without independent observational data to validate the EM directly, it was beyond the scope of this study to assess the uncertainties of the emission inputs and their impacts on AQM performance. Rather, the focus of this study was to investigate how the RCM simulation affects AQM performance, by examining tropospheric O_3 formation and distribution.

2. The modeling system

The RCM is based on the fifth-generation Pennsylvania State University–National Center for Atmospheric Research Mesoscale Model (MM5), version 3 (Dudhia et al. 2000). Various MM5 versions often have been used as the meteorological model in episodic studies of air quality. For regional climate applications, the buffer zone treatment, ocean interface, and improved cloud–radiation interactions (Liang et al. 2001, 2004) have been incorporated in the RCM used in this study. As noted above, small biases that are not important in short MMM integrations can be critical in long RCM simulations. To reproduce the best proxy of climate conditions for the present and future climate, the RCM requires more a stringent and realistic configuration of model physics and dynamics as well as accurate incorporation of LBCs to reduce climate biases. As driven by the National Centers for Environmental Prediction/Department of Energy Atmospheric Model Intercomparison Project (AMIP-II) global reanalysis (R-2) data (Kanamitsu et al. 2002), the RCM has realistically simulated summer 1993 flooding rainfall in the Midwest (Liang et al. 2001). The RCM also realistically simulated surface temperature and soil moisture patterns (Zhu and Liang 2006). For a more detailed description of the RCM, refer to Liang et al. (2001, 2004). The greenhouse gases and aerosol could have important direct and indirect feedbacks to the climate system. However, this is a complicated issue subject to ongoing research. In this study, there is no feedback from chemical conditions to meteorological conditions in the RCM. The EM estimations and AQM simulations used an offline computation, that is, they were not run step by step with the RCM simulation.

The EM adopted for this study is the Sparse Matrix

Operator Kernel Emissions (SMOKE) modeling system (Houyoux et al. 2000) that processes the U.S. Environmental Protection Agency (USEPA) National Emission Inventory (NEI). It computes gridded, temporalized, speciated emissions in the following four data categories: point, area, mobile, and biogenic. A plume rise algorithm partitions stack emissions as a function of height using the NEI stack parameters and time-dependent RCM meteorological data. These meteorological data and county-level land use information are used to compute biogenic emissions. By using a geographical information system, county-level area, mobile, and biogenic emissions are partitioned to gridded emissions based on fractional county land area in each grid. The ability of the SMOKE system to simulate the emissions pattern in the Midwest realistically has been demonstrated (Williams et al. 2001).

The AQM was developed from the San Joaquin Valley Air Quality Study/Atmospheric Utilities Signatures, Predictions and Experiments Study (SJVAQS/AUSPEX) Regional Modeling Adaptation Project (SARMAP) Air Quality Model (SAQM; Chang et al. 1997), which evolved from the Regional Acid Deposition Model (RADM; Chang et al. 1987). (Ranzieri and Thuillier 1991). The SAQM has been used successfully to study the elevated O_3 problem in the Central Valley of California. The AQM, an improved version of the original SAQM, includes a faster, more accurate numerical scheme for solving gas-phase chemistry (Huang and Chang 2001) as well as an aerosol module. The photolysis rates are modified in the presence of cloud according to the fractional area of cloud coverage and cloud optical depth (Chang et al. 1987). Most of other model procedures and transport and chemical processes, such as the operator-splitting technique, are described in detail by Chang et al. (1987, 1997) and Huang and Chang (2001). Input and output modules of the AQM were redesigned for continuous, long-term climatic applications in this study. Seasonal simulations were conducted to determine model capability and identify improvements needed to increase model suitability for addressing climate impacts on air quality, including the careful estimation of biogenic emissions in air quality studies and an adequate spatial resolution to avoid overestimating the biogenic emission impact in subgrid areas (Tao et al. 2003; Huang 2002). The AQM has several options for a chemical mechanism: the RADM, version 2 (RADM2; Stockwell et al. 1990), the Statewide Air Pollution Research Center Chemical Mechanism (Carter 1990), and the Carbon Bond Mechanism IV (Gery et al. 1989). The first two mechanisms use reactivity with the hydroxyl radical (OH) to lump similar species into a species group (Middleton et al. 1990), while the last mechanism lumps similar species according to their carbon bond structure. Reaction coefficients were updated according to Sander et al. (2003).

3. Experiment design

For climate studies, multiyear simulations are needed to obtain a more robust evaluation of the climate modeling system. The period of years 1995–2000 was selected for this study as the current climate simulation. Some years have anomalously warm conditions affecting some regions of the United States, for example, the 1995 summer in the Midwest, including a very intense mid-July heat wave (Kunkel et al. 1996). Several elevated O_3 episodes occurred in the Midwest as a result of high temperatures, subsiding air masses, and clear skies associated with recurring stagnant high pressure systems. The AQM simulations started on yearday 150 of each year and were integrated continuously for 90 days (3 summer months). Each model month contained 30 days of data.

Figure 1 shows the computational domains for the modeling system. Domain 1 has 90-km grid spacing that extends from the Pacific Ocean to the Atlantic Ocean, including the Gulf of Mexico and much of Canada. This outer domain optimally integrates LBCs for the RCM and objectively resolves the transport of precursor gases from the west to the Midwest and northeast United States. Two subdomains have 30-km grid spacing that extends from just east of the Rocky Mountains to the East Coast. The inner domains were designed to resolve regional and local characteristics that govern the air quality over the northeast and Midwest.

In this study, LBCs for the RCM were constructed from the R-2 reanalysis data that assimilate all available observations and are available at 6-h intervals. The 1999 NEI is the most comprehensive emissions dataset for the present simulation period and was used in this study (available online at http://www.epa.gov/ttn/chief/net/1999inventory.html). For Canada, the area and mobile emissions from the 1995 emissions inventory of criteria air contaminants were combined with the major atmospheric point source data from the 2001 national pollutant release inventory (available online at http://www.ec.gc.ca/pdb/npri/npri_dat_rep_e.cfm). The Big Bend Regional Aerosol and Visibility Observational Study inventory was the data source for emissions from 10 states in northern Mexico (available online at http://www.epa.gov/ttn/chief/net/mexico.html). The EM used the RCM-simulated meteorological conditions in processing the surrogated emissions inventory. Given RCM meteorological conditions and EM source emissions inputs, the AQM then was continuously inte-

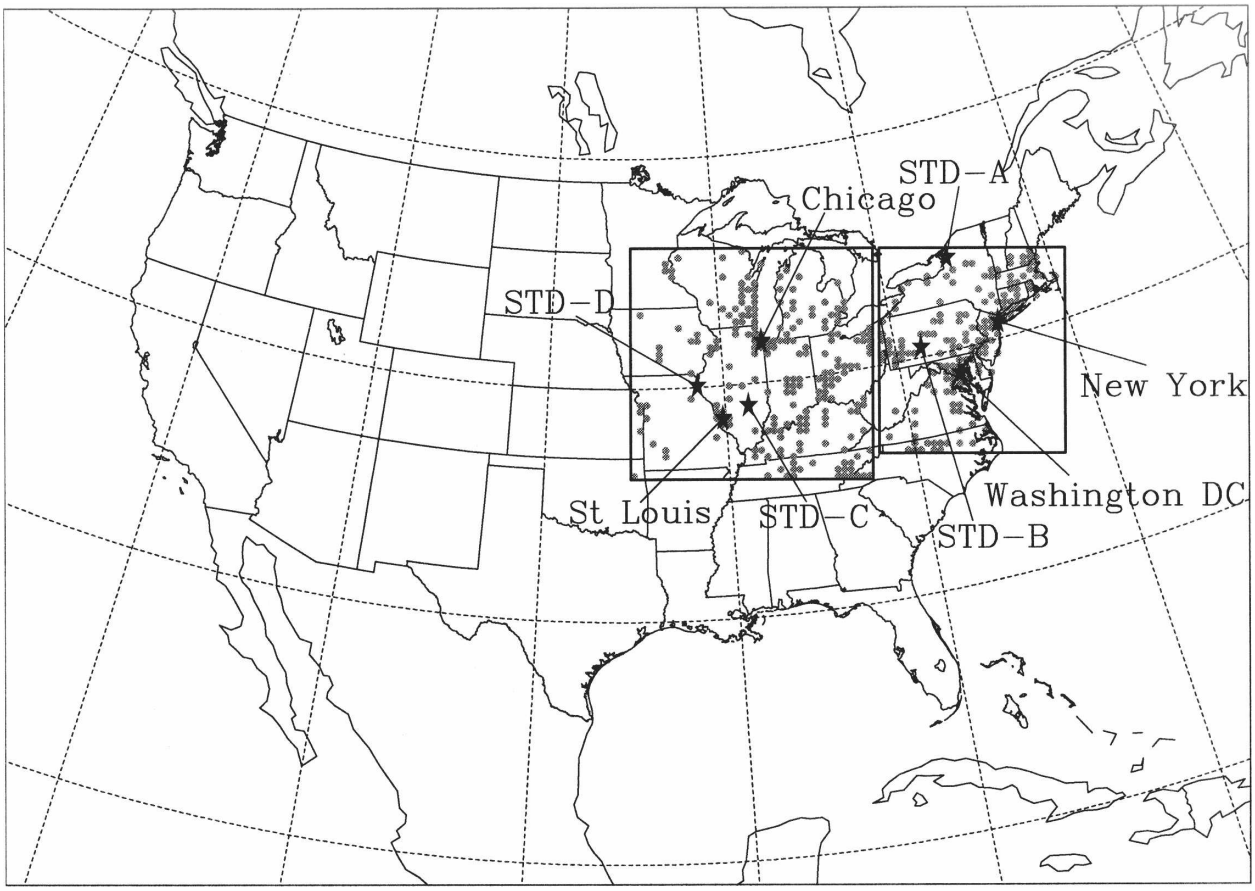

FIG. 1. Nested modeling domains with 90-km grid spacing in the outer box and 30-km grid spacing within the inner boxes. The selected target areas are marked with black star and gray dots represent selected monitoring grid cells.

grated for the study period. The AQM has 15 vertical layers as compared with the 23 vertical layers in the RCM. The lowest 10 layers (~3 km) of both models were identical. The vertical mass average method was applied to meteorological parameters for the merged RCM layers in the upper model domain. Except for point source emissions, which were placed vertically in layers according to the plume rise algorithm in SMOKE, all hourly mean emissions were placed in the lowest simulated AQM layer. A 5-min time step was used in AQM simulations to produce hourly average output in the surface layer and instantaneous hourly output for upper layers. For a reasonable comparison with observations, the surface layer submodel (SLS; Chang et al. 1997) option was selected, and the lowest SLS layer depth (~15 m) was used. The first grid point in the vertical was about 7.5 m above the surface. Invariant clean background lateral boundary conditions (Fig. 2) were used for the outer coarse grid domain (Chang et al. 1997). Considering the lengthy simulation and the chemical speciation included in the EM, this study used the RADM2 chemical mechanism. A one-way-nested method was used and, because of limited computational resources, the aerosol module was turned off. It should be noted that the tropospheric O_3 chemistry can be different without the presence of aerosol chemistry. Because the same model configuration will be used in future climate simulations, it is expected the future air quality simulations will be impacted in a similar way. As described in the introduction, the tropospheric O_3 was used to explore the potential impacts of climate on air quality and to identify key characteristics for future research. The inclusion of the aerosol chemistry is an important issue and will be studied in the future.

For evaluation purposes, surface hourly O_3 measurements were obtained from the USEPA Air Quality System (AQS; information available online at http://www.epa.gov/ttn/airs/airsaqs/detaildata/downloadaqsdata.htm). The AQM simulations produce species concentrations averaged in a grid-box volume, while the observations normally were taken at 5-m height in a finite space. To achieve a more realistic comparison with observations, model-simulated O_3 concentrations in each

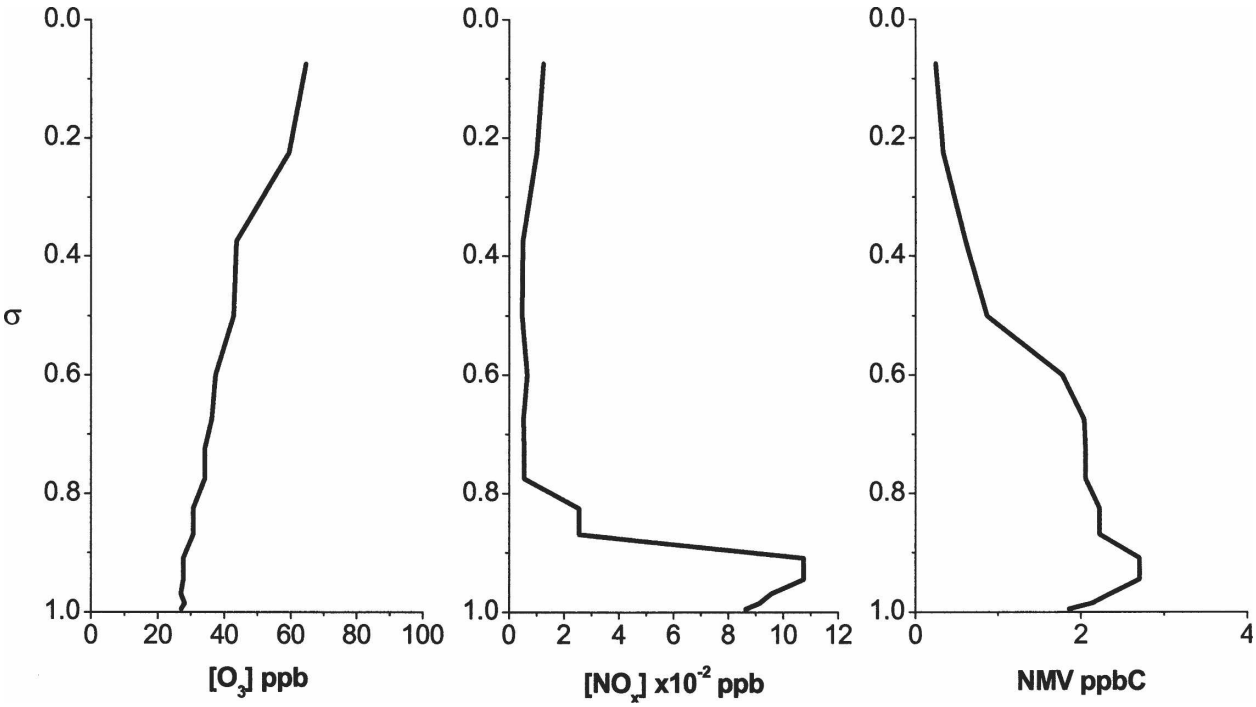

FIG. 2. The AQM lateral boundary conditions of (left) ozone (ppb), (middle) nitrogen oxides (10^{-2} ppb), and (right) total nonmethane hydrocarbon (ppbC). The vertical axis is the height in σ coordinates.

selected target area were compared with the corresponding observed values averaged for all monitoring stations in the AQM grid cell containing the target area. A criterion was applied to select monitoring grid cells for comparison. Each selected grid cell must contain at least 95% of the monthly observed records for all summer months of the year. The criterion is vital to avoid comparing biased observations with model simulations for a specific time of a day and/or specific month of a year, as well as for the specific location of modeling domain. However, selected grid cells were not required to meet the criterion for the entire simulation period, that is, to have 6 yr of data. A number of new monitoring sites were established during the period from 1995 to 2000. To take advantage of the temporally increasing spatial coverage, the qualified monitoring grid cells only needed to satisfy the criterion for each individual year. As a result, the number of qualified monitoring grid cells increases from 84 (101) in 1995 to 166 (212) in 2000 for the Northeast (Midwest) subdomain (Fig. 1). Four urban metropolitan areas were selected for analyses: New York City, New York, Washington, D.C., Chicago, Illinois, and St. Louis, Missouri. These major metropolitan areas were chosen because they frequently post high O_3 episodes, as well as experience significant health issues because of the very large population residing in the areas. Observed surface air temperatures were obtained by a Cressman (1959) objective analysis using daily measurements from 7325 stations in the U.S. National Weather Service Cooperative Observer Network.

Several statistical variables will be used in the following discussion, including variance and correlation coefficient. The difference (bias) between model-simulated (mod) and observed (obs) data based on hourly average values (x) is defined as

$$\text{bias} = \frac{1}{b}\sum_{a=1}^{b}\frac{1}{l}\sum_{k=1}^{l}\frac{1}{m}\sum_{j=1}^{m}\frac{1}{n}\sum_{i=1}^{n}(x_i^{\text{mod}} - x_i^{\text{obs}}), \quad (1)$$

and the normalized gross error (NGE) based on paired hourly average values (x) is defined as

$$\text{NGE} = \frac{1}{b}\sum_{a=1}^{b}\frac{1}{l}\sum_{k=1}^{l}\frac{1}{m}\sum_{j=1}^{m}\frac{1}{n}\sum_{i=1}^{n}\left(\frac{|x_i^{\text{mod}} - x_i^{\text{obs}}|}{x_i^{\text{obs}}}\right)100, \quad (2)$$

where n is the number of hours, m is the number of days, l is the number of months, and b is the number of years. If the variables used in Eq. (2) are diurnal hours of monthly or seasonal mean values, then $m = 1$ or $m = l = 1$, respectively. Because only the summer months were simulated in this study, the seasonal mean

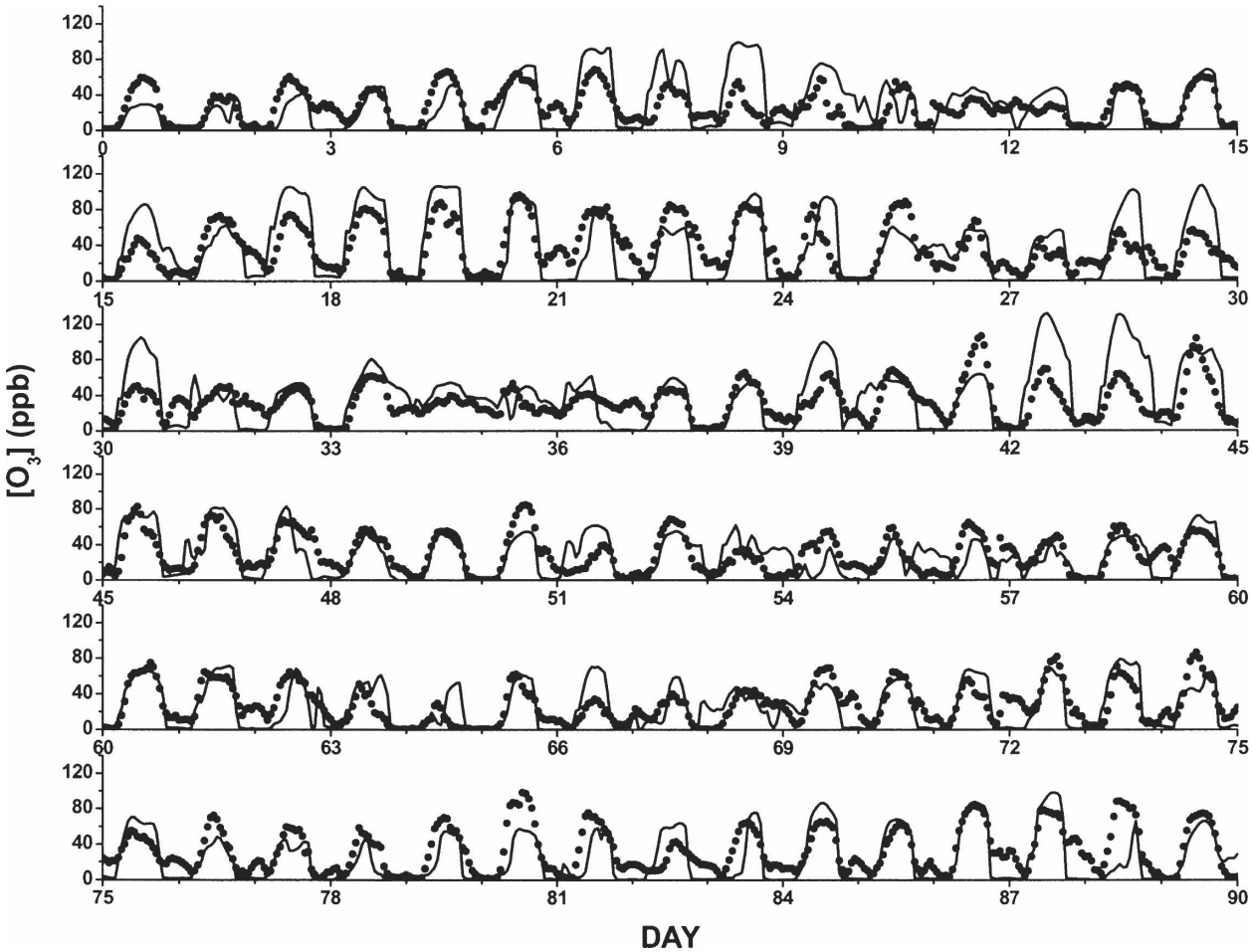

FIG. 3. The 90-day hourly averaged ozone concentration (O_3; ppb) in St. Louis for summer of 1995. Solid circles represent observations, and the line represents model simulations.

is equivalent to a summertime mean in the following discussion. To prevent an unrealistically huge NGE value in an environment of very low observed O_3 concentration, a cutoff value of 40 ppb for minimum observed O_3 concentration is applied.

4. Results

Figure 3 shows the observed and simulated time series of hourly averaged surface O_3 concentration in St. Louis for summer of 1995. There is general agreement for several features, including the strong diurnal cycle and some multiday periods of high O_3 concentrations. The AQM, however, substantially overestimated daily O_3 maxima for model days 7, 9, 16, 18, 29, 30, 31, 40, 43, and 44 in June and July, while August estimates were more realistic. The correlation coefficients between observed and model-simulated daily mean O_3 concentration for the entire simulation period are 0.31, 0.33, 0.27, and 0.19 in New York City, Washington, D.C., Chicago, and St. Louis, respectively. There were different correlations among months and years of a selected area, as well as different correlation changes for different metropolitan areas. As discussed below, the nighttime underestimates were often found in selected metropolitan areas. For the daytime model performance, the correlation coefficients between observed and model-simulated daily maximum 8-h average O_3 concentration are 0.32, 0.36, 0.30, and 0.25 for the entire simulation period in New York City, Washington, D.C., Chicago, and St. Louis, respectively. For monthly means of the daily maximum 8-h average O_3 concentration, the correlation coefficients between observed and model-simulated increase to 0.42, 0.43, and 0.46 for New York City, Washington D.C., and St. Louis, while it decreases to 0.24 in Chicago. Between the model-simulated temperature and O_3 concentration, the correlation coefficients are always high in all metropolitan areas, ranging

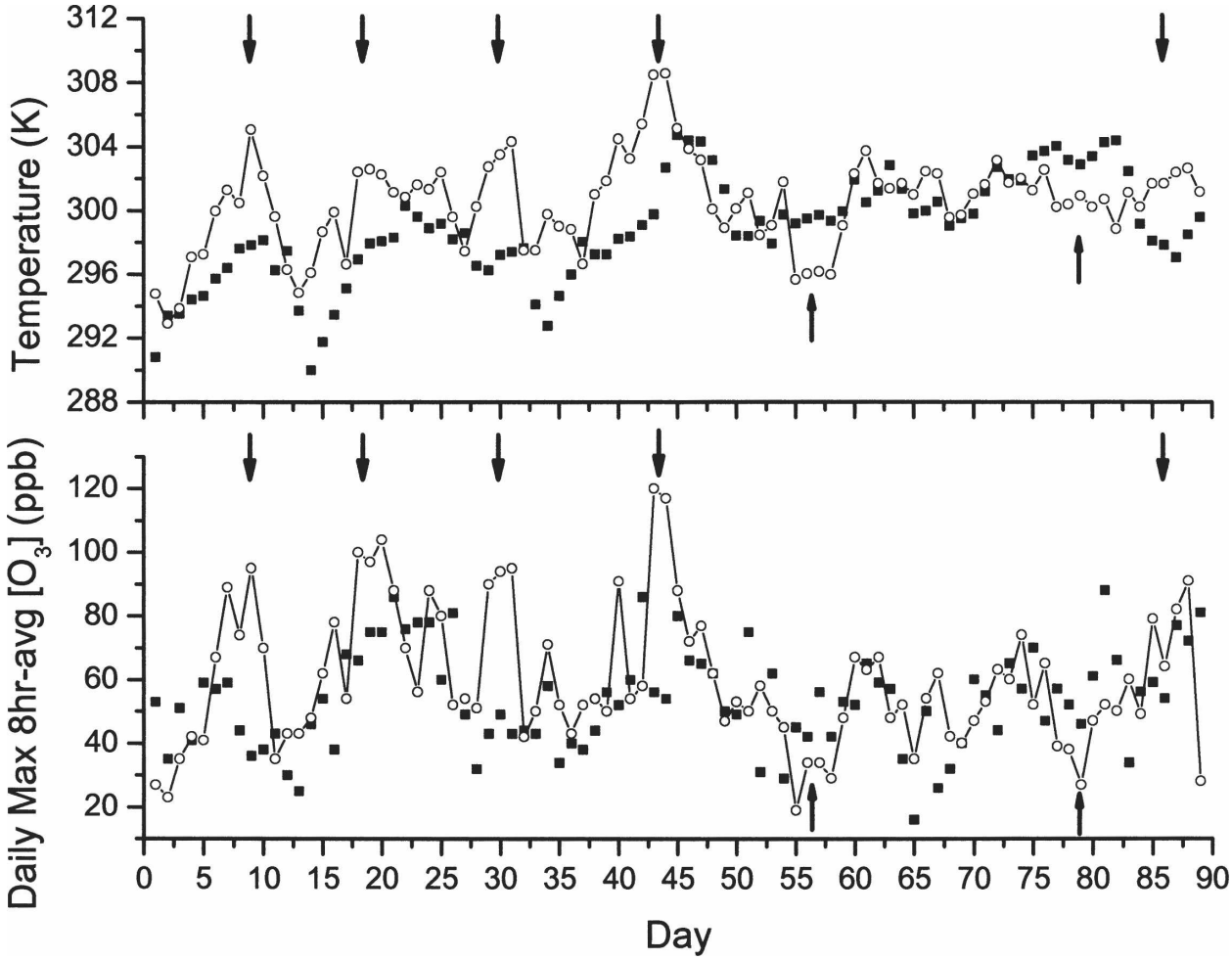

FIG. 4. (top) Daily means of model-simulated vs observed temperature and (bottom) daily maximum 8-h average ozone concentration in St. Louis for summer of 1995. Solid squares and lines with open circles represent observed and model-simulated values, respectively.

from 0.53 to 0.68 for daily values and from 0.36 to 0.77 for monthly mean values. As discussed below, the AQM-observed differences in O_3 concentration are associated with RCM-simulated meteorological conditions, especially surface temperature biases.

High O_3 episodes generally occur under weather conditions identified with stable atmosphere, clear skies, and prevailing high temperatures. Although transport and emissions also play important roles in O_3 formation, temperature is considered to be the most important factor (NRC 1991). Aw and Kleeman (2003) show that summertime ozone concentration generally increases as temperature increases. It can be viewed as the zero-order variable that represents the net effect of solar radiation, cloud cover, mixing, and other factors. Correlation coefficients between the RCM-simulated daily mean surface temperature and AQM-simulated 8-h average O_3 concentration (June, July, and August) for the simulation period were high in New York City (0.71, 0.72, and 0.68), Washington, D.C. (0.73, 0.69, and 0.69), Chicago (0.64, 0.69, and 0.71), and St. Louis (0.79, 0.84, and 0.67). Figure 4 shows the time series of observed and model-simulated daily mean temperature and daily maxima 8-h average O_3 concentration in St. Louis for the 1995 summer. Overestimates of surface temperature around model days 10, 20, 30, 45, and 85 correspond to overestimates of surface daily maximum 8-h average O_3 concentration (downward arrows in Fig. 4), while underestimates of surface temperature around model days 57 and 77 correspond to underestimates of surface daily maximum 8-h average O_3 concentration. The analyses of different model years also showed a similar result. However, the correlation coefficient between the observed daily mean surface temperature and ozone concentration is lower than that modeled, and is 0.41–0.45 for the New York City, 0.31–0.32 for

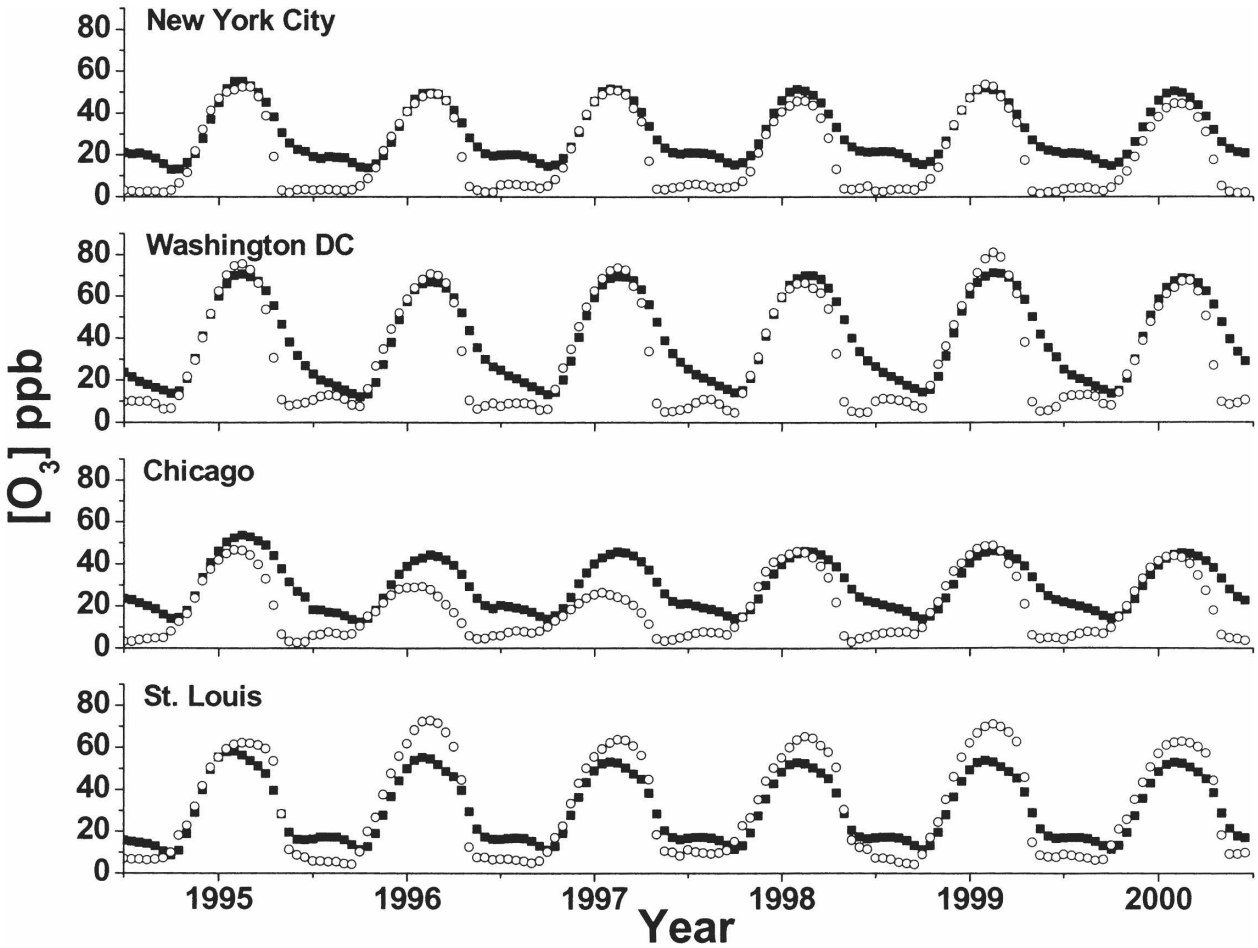

FIG. 5. Seasonal mean diurnal distribution of ozone concentration (O_3; ppb) in selected areas from 1995 to 2000. Solid squares and open circles represent observed and model-simulated values, respectively.

the Washington, D.C., 0.27–0.46 for the Chicago, and 0.12–0.31 for St. Louis. It implies that other factors, such as transport, also play an important role in ozone formation in these areas. Nevertheless, because of high correlation between modeled quantities, results indicate the RCM temperature biases explain a large portion of AQM O_3 biases.

Figure 5 show the seasonal mean diurnal distribution of O_3 concentration for the simulation period in selected metropolitan areas. The model underestimated nighttime O_3 concentration in selected metropolitan areas over the entire integrated period. Underestimates often were found to be related to the model procedure such as the choice of surface layer depth in which a shallow model surface layer is coupled with stronger nighttime nitric oxide (NO) emissions typically associated with a large metropolitan area. The enhanced nighttime O_3 titration process

$$O_3 + NO \rightarrow NO_2 + O_2 \quad (3)$$

transforms O_3 to nitrogen dioxide (NO_2). Soon after sunrise, the photochemical processes quickly transform NO_2 back to O_3 and produce fairly realistic distributions of daytime O_3 concentration. As a result, the bias of daily mean O_3 concentration using hourly averaged values, $[O_3]_{aqm} - [O_3]_{obs}$, for the entire simulation period ranged from −8.6 to −11.6 ppb for New York City, from −5.4 to −10.8 ppb for Washington, D.C., from −6.8 to −14.8 ppb for Chicago, and from −0.2 to 3.5 ppb for St. Louis (Table 1). The mean normalized gross error is 42.7%, 45.7%, 49.0%, and 38.9% in New York City, Washington, D.C., Chicago, and St. Louis, respectively. These values are much higher than the USEPA-suggested threshold of 35%. Although St. Louis had a lower bias of daily mean O_3 concentration than that of other three areas, Fig. 5 shows that the overestimation of daytime O_3 concentration in St. Louis is the worst among selected metropolitan areas. The bias of daily maximum 8-h average O_3 concentration for the entire simulation period ranged from −4.1 to 1.4 ppb for New

TABLE 1. Seasonal mean bias and normalized gross error of daily mean ozone concentration (ppb) between modeled and observed data in selected metropolitan areas for the simulation period.

	Bias (ppb)							Normalized gross error (%)						
Area/year	1995	1996	1997	1998	1999	2000	Mean	1995	1996	1997	1998	1999	2000	Mean
New York City	−9.9	−8.6	−8.7	−11.3	−9.8	−11.6	−10.0	46.1	43.3	40.1	40.0	40.6	46.2	42.7
Washington, DC	−8.0	−5.4	−8.0	−10.8	−7.3	−9.5	−8.2	45.6	43.4	45.8	43.8	45.6	50.1	45.7
Chicago	−12.6	−12	−14.8	−7.3	−6.8	−8.6	−10.4	50.7	57.1	59.0	39.8	42.6	44.7	49.0
St. Louis	0.2	2.0	0.4	3.5	2.0	1.2	1.6	36.3	41.1	33.4	37.3	41.6	43.6	38.9

York City, from −2.2 to 5.7 ppb for Washington, D.C., from −14.8 to 3.8 ppb for Chicago, and from 5.9 to 17.1 ppb for St. Louis (Table 2). The mean normalized gross error is 30.4%, 29.4%, 36.0%, and 34.4% in New York City, Washington, D.C., Chicago, and St. Louis, respectively. The model showed a better performance of daytime O_3 in New York City and Washington, D.C. The worse value in Chicago resulted mainly from the large underestimates of daytime O_3 concentration in the second and third years. The model consistently overestimated the daytime O_3 concentration in St. Louis. In fact, the daytime overestimates compensated for the nighttime underestimates found in all areas and lead to a smaller bias in Table 1. Observation stations in the USEPA AQS are often concentrated in the vicinity of urban areas, which limit the ability to compare model-simulated values with observations at rural areas. Four grid cells that contain a small township or city and have small nitrogen oxides (NO_x) emissions (Fig. 6) were selected as "rural sites" (STD_A, STD_B, STD_C, and STD_D; see Fig. 1). Figure 7 show the seasonal diurnal distribution of O_3 concentration over the simulation period for selected rural sites. Different from the diurnal distribution of O_3 concentration in selected metropolitan areas, there was no noticeable nighttime O_3 underestimation in these sites. However, common daytime overestimates were shown.

The underestimates of daytime O_3 concentration in most metropolitan areas and the daytime O_3 overestimation in rural sites may be an indication of too much NO_x emissions estimated by the emission model, which resulted in an enhanced O_3 production in rural areas that were NO_x limited and enhanced O_3 titration in metropolitan areas. It could also be the result of inefficient dynamical processes that removed the O_3 precursor gases away from the polluted boundary layer and into the free troposphere, as well as the possibility of weaker NO_x deposition simulated by the AQM. It is difficult to examine the magnitude of the emissions with nearly no available observed emissions. As described in the introduction section, the current modeling system was designed for application at climatic time scales and regional spatial scales. Thus, the temporal and spatial resolutions that can be resolved by the current modeling system also are limited by the numerical schemes incorporated. Although the resolution is an important issue that warrants further investigation, it is beyond the scope of this study to perform extensive analyses on the EM and RCM. Therefore, the results shown in this study should be viewed as the combination of uncertainties of all modeling components. As shown below, the results still indicate the significant impacts of the RCM meteorological conditions' biases on the AQM biases.

Taking a broad average of AQM output, Fig. 8 shows the seasonal mean diurnal distribution of O_3 concentration over the Midwest and northeast subdomains from 1995 to 2000. The diurnal O_3 distributions shown in Fig. 8 were the average of all qualified monitoring grid cells (see section 3) in each subdomain of each year. The modeling system actually performed a good nighttime O_3 simulation in both subdomains, but small daytime overestimates of O_3 concentration still existed over the simulation period. There are different annual

TABLE 2. Seasonal mean bias of daily maximum 8-h average ozone concentration (ppb) and normalized gross error (%) between modeled and observed data in selected metropolitan areas for the simulation period.

	Bias (ppb)							Normalized gross error (%)						
Area/year	1995	1996	1997	1998	1999	2000	Mean	1995	1996	1997	1998	1999	2000	Mean
New York City	−0.5	−0.9	1.0	−4.1	1.4	−4.0	−1.2	26.6	29.2	29.3	28.0	30.9	38.3	30.4
Washington, DC	2.5	2.7	2.8	−2.2	5.7	−2.0	1.6	27.4	27.5	31.3	25.4	29.5	35.0	29.4
Chicago	−6.9	−12.1	−14.8	1.0	3.8	0.4	−4.8	31.4	44.1	48.8	28.4	29.3	34.1	36.0
St. Louis	5.9	16.2	10.9	12.4	17.1	11.8	12.4	29.0	33.0	30.6	32.1	38.8	42.7	34.4

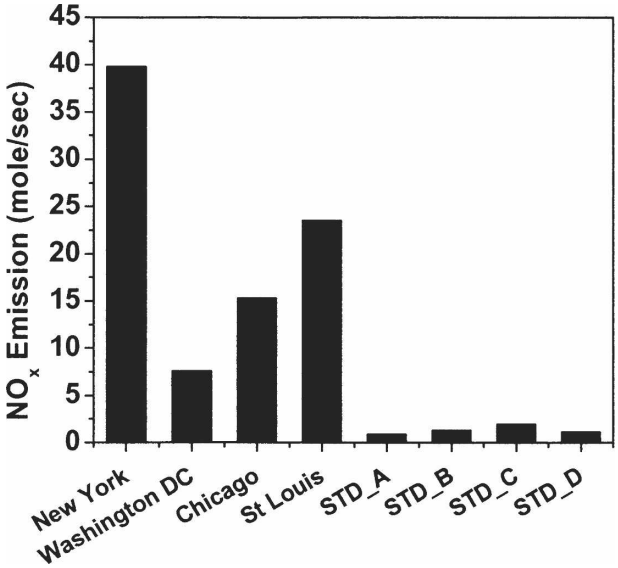

FIG. 6. Nighttime NO_x emissions in selected areas.

variations for the differences between observed and model-simulated seasonal mean diurnal O_3 distribution in two subdomains over the entire simulation period. In this study, the anthropogenic emissions did not change annually. Guenther et al. (1994) showed that the biogenic VOC emissions are a function of incident solar radiation and temperature. The variables were simulated by the RCM, thus, the biogenic emissions could change as a result of changing RCM-simulated meteorological conditions. This indicates that the variations of AQM output were significantly influenced by the annual variations of RCM-simulated meteorological conditions.

Table 3 shows the bias and normalized gross error of seasonal daily mean O_3 concentration over the entire simulation period in the two subdomains. The bias of daily mean O_3 concentration for the summer months ranged from 3.5 to 6.4 ppb in the northeast subdomain and 0.2 to 2.3 ppb in the Midwest subdomain. For traditional model evaluation involving hourly paired data,

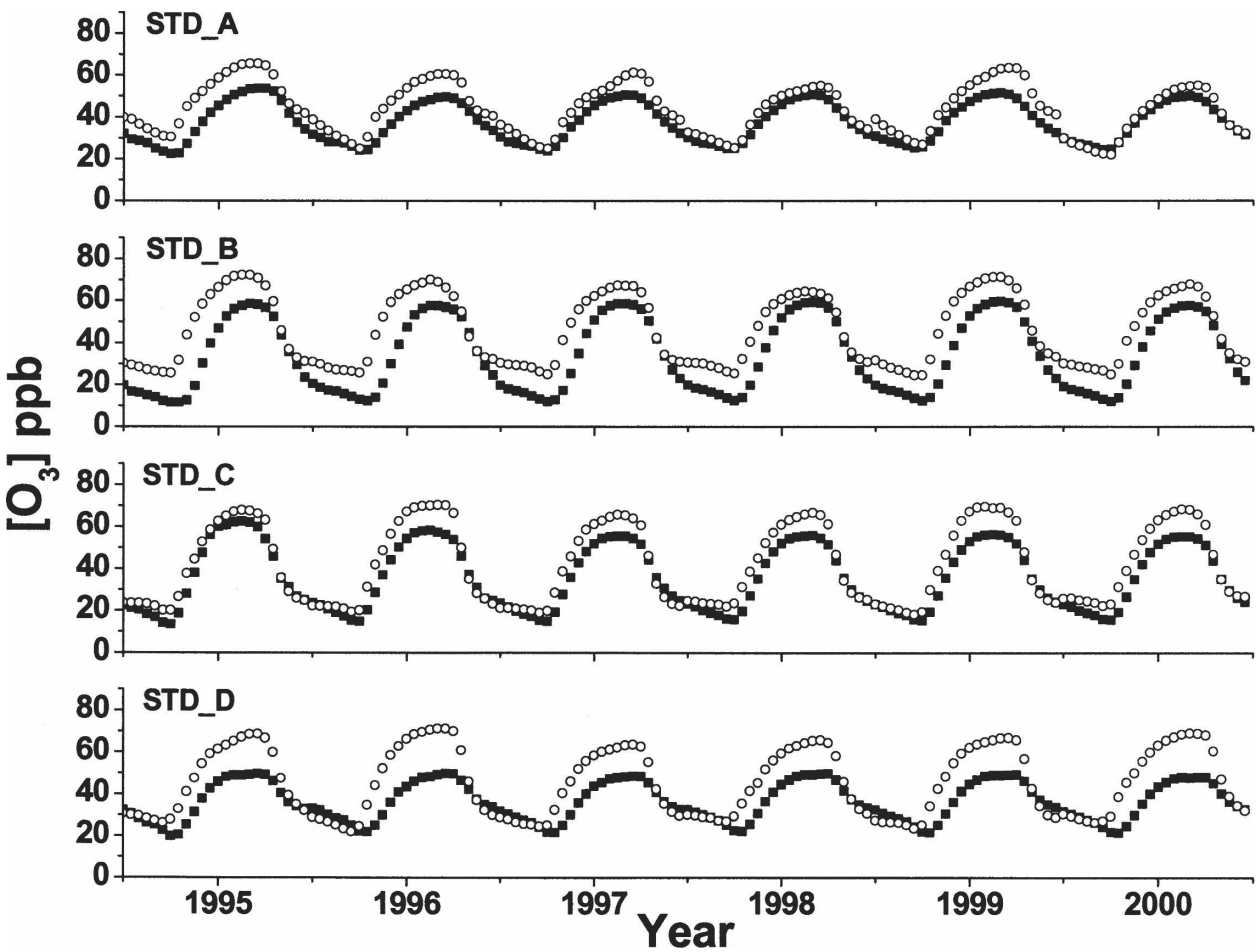

FIG. 7. Similar to Fig. 5, but for selected rural sites.

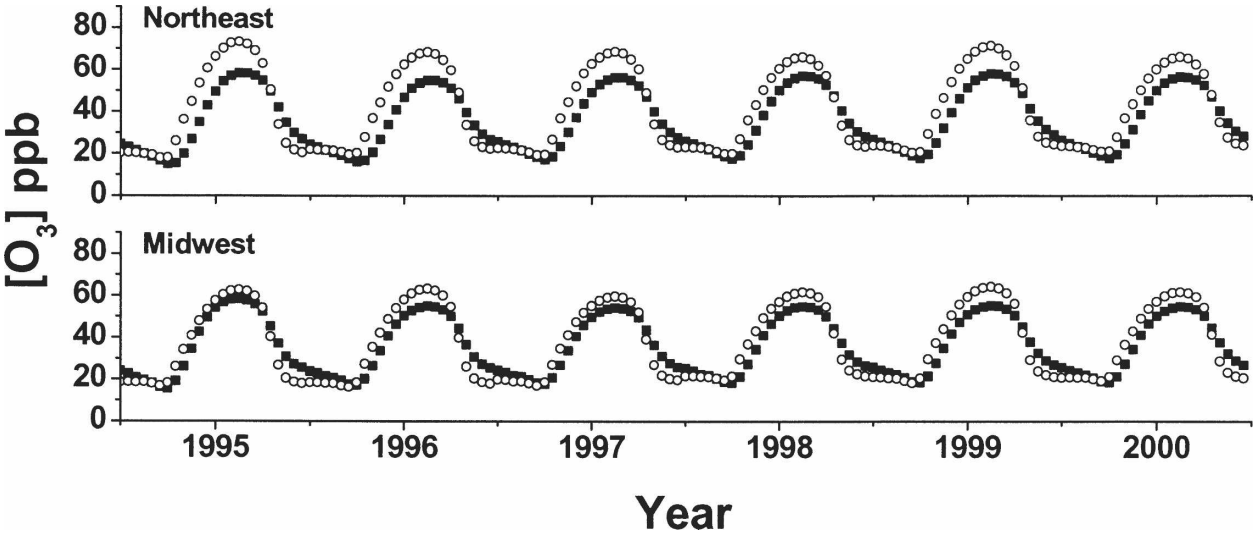

FIG. 8. Similar to Fig. 5, but for the two subdomains.

the normalized gross error of summer months ranged from 35.3% to 40.1% (33.5% to 38.0%) for the northeast (Midwest) subdomains. Most values were close and somewhat larger than the USEPA-suggested threshold of 35%. Because the FDDA technique is not applied in this type of study, it is challenging to constrain the instantaneous RCM meteorological conditions as closely to observations as in the traditional MMM studies. The threshold may have to be relaxed for climatic studies. Figure 9 shows the histograms of variance of different quantities with respect to monthly average in two subdomains. Similar results with respect to the seasonal average were found and are not shown here. For hourly averaged data (Figs. 9a and 9b) model simulations had more occurrences of larger variance values than those observed. This indicates that the modeling system tends to simulate a stronger diurnal fluctuation than that of the observations. The difference between model-simulated and observed variance distributions using daily mean values is smaller than that using hourly mean values (Figs. 9c–e), but still shows a stronger daily fluctuation than that simulated by the model.

In addition to hourly and daily mean statistics, monthly mean statistics of model-simulated values also were examined. To test whether the current modeling system actually can simulate monthly mean air quality statistics, a Student's *t* test was applied using the 0.05 significance level. For example, results show that monthly means differences between observed and model-simulated daily maximum 8-h average O_3 concentrations are not significantly different for the three individual summer months of 1995 in New York City and Washington, D.C. Although the differences are statistically significant for June 1995 in Chicago and St. Louis, they are not significant for July and August 1995. This suggests that the AQM can simulate most of the observed monthly means of daily maximum 8-h-averaged O_3 concentrations of 1995 in these areas. Applying the same analyses to the entire simulation period of 18 model months, a Student's *t* test showed that the modeling system can capture 94.4% of the monthly means in the daily maximum 8-h average O_3 concentration in New York, 83.3% in Washington, D.C., and 55.6% in Chicago, but only 22.2% in St. Louis. The Student's *t* test also showed that model-simulated seasonal means of daily maximum 8-h average O_3 concentration are not statistically different than those observed in New York City, Washington, D.C., and Chicago. The modeling system failed to explain the

TABLE 3. Seasonal mean bias and normalized gross error of daily mean ozone concentration (ppb) between modeled and observed data in two subdomains for the simulation period.

	Bias (ppb)							Normalized gross error (%)						
Subdomain/year	1995	1996	1997	1998	1999	2000	Mean	1995	1996	1997	1998	1999	2000	Mean
Northeast	5.9	6.4	5.0	3.5	5.3	4.0	5.0	40.1	39.2	39.5	35.3	38.9	37.6	38.4
Midwest	0.2	1.3	0.6	2.2	2.3	1.7	1.4	37.4	38.0	35.8	33.5	35.1	34.6	35.7

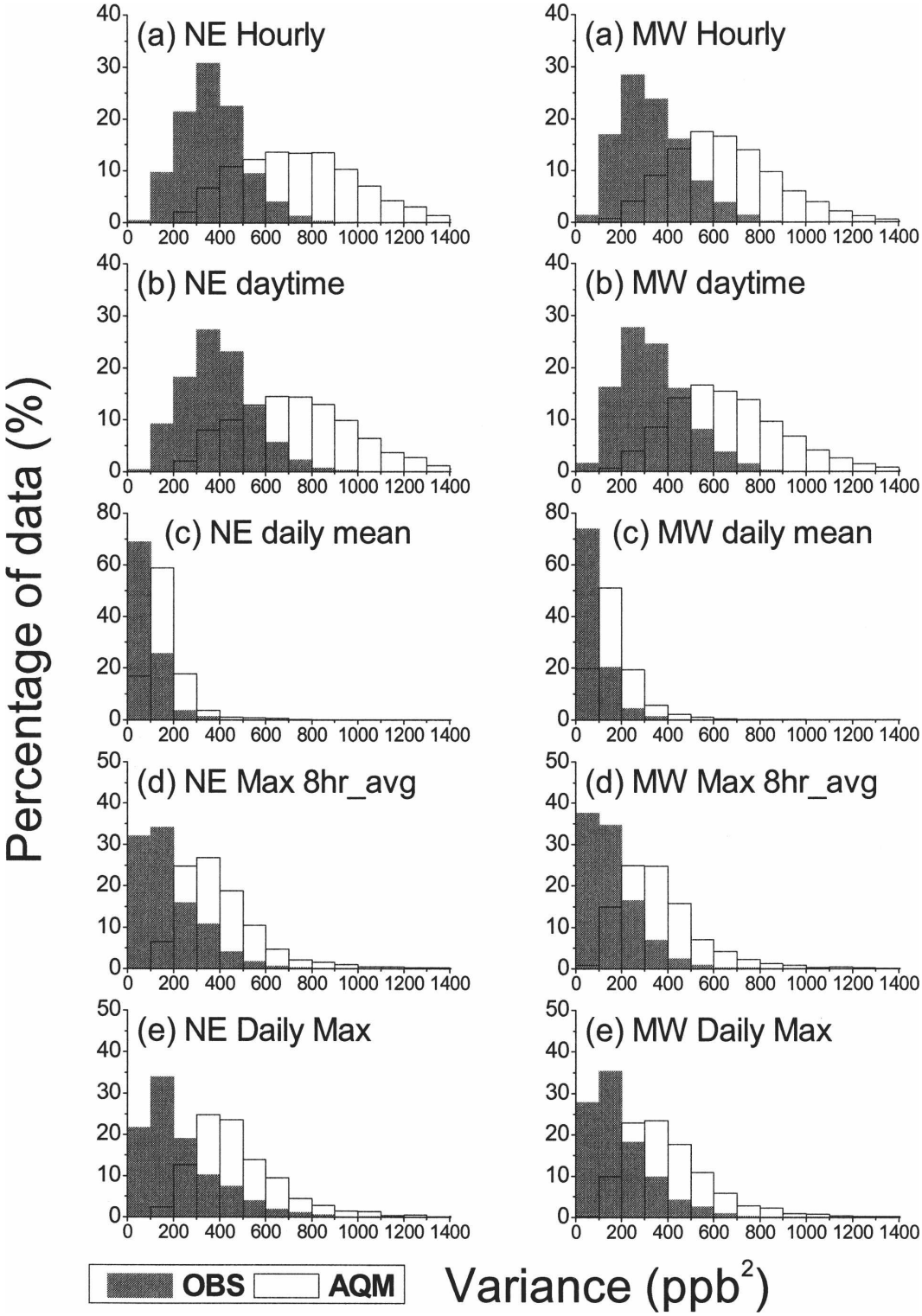

FIG. 9. Histograms of variances of (a) hourly averaged O_3, (b) daytime O_3, (c) daily mean O_3, (d) daily maximum 8-h average O_3, and (e) daily maximum O_3 in the (left) northeast (NE) and (right) Midwest (MW) subdomains from 1995 to 2000. The shaded and unshaded histograms represent the O_3 variance for the observations and model simulations.

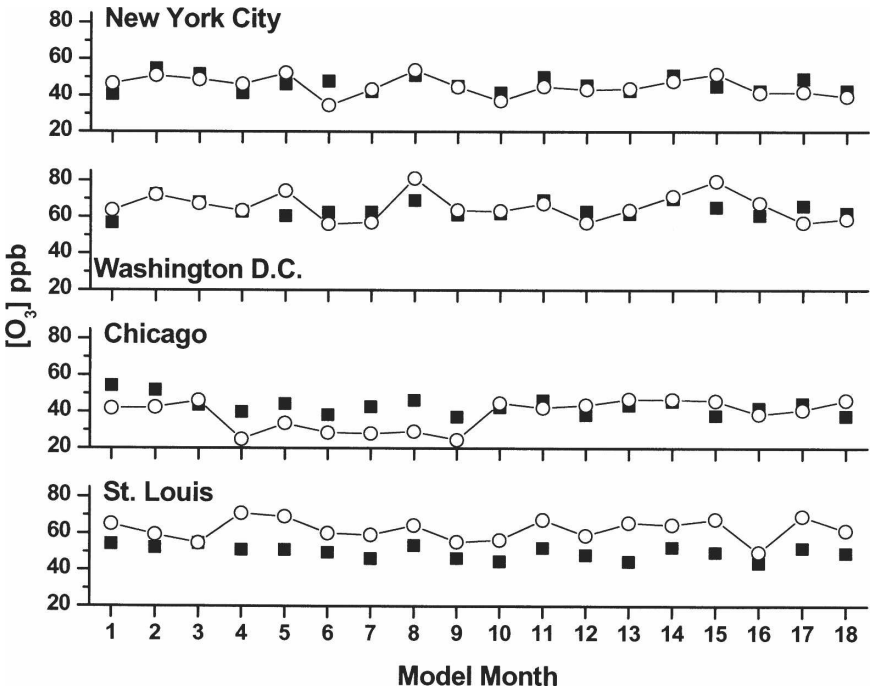

FIG. 10. Monthly mean daily maximum 8-h average ozone concentration (O_3; ppb) of summer months in selected areas for 1995–2000. Lines with solid squares and open circles represent observed and model-simulated values, respectively.

variations of the monthly and seasonal means in St. Louis, where overestimates of daytime O_3 concentration were common (Fig. 10). Overestimates of RCM-simulated seasonal mean surface temperature were 2.7° and 2.5°C in St. Louis and New York City, respectively (Fig. 11). For nearly the same magnitude of temperature overestimates, seasonal mean biases of AQM-simulated daily maximum 8-h average O_3 concentrations are +12.4 ppb in St. Louis and −1.2 ppb in New York City. It is known that greater St. Louis has a strong source of biogenic VOC emissions located to the southwest of the metropolitan area. Huang (2002) showed that without the presence of the isoprene emissions the O_3 reduction in St. Louis is 33.3% versus 17.9% in New York. It shown the O_3 production in St. Louis was greatly influenced by the isoprene concentration. When the local meteorological conditions are favorable to O_3 production, that is, there are both a higher temperature and strong incident solar radiation at the surface, the biogenic emissions also are expected to be stronger (Guenther et al. 1994). Although the reaction between the isoprene and OH radical is faster than that between the isoprene and ozone, with the lifetime of ~1.7 h versus 1.3 days (Seinfeld and Pandis 1998), the latter can efficiently recycle odd hydrogen as well as the NO_x and thus promote the ozone production. Coupled with the favorable isoprene transport that brought the isoprene concentration into the urban area, the O_3 concentration in St. Louis was more sensitive to temperature variation than that in New York.

Because the *t* test suggested that the climate modeling system can capture most of the monthly features in some areas, the modeling system was evaluated with observations for the 18 simulated months based on diurnal paired monthly and seasonal mean values (Figs. 5 and 8). This procedure resembles the common episodic AQM model evaluation on the model results using diurnal paired hourly average values for a simulation period of a few days. The NGE using the regional monthly mean values (seasonal mean values) ranged from 4.4% to 88.3% (10.3%–63.6%) in the northeast subdomain and from 1.0% to 81.3% (3.7%–53.0%) in the Midwest subdomain. The NGEs using paired monthly mean values are 11.3%, 22.2%, 24.6%, and 23.3% in New York City, Washington, D.C., Chicago, and St. Louis, respectively. The NGEs using paired seasonal mean values are 6.5%, 18.1%, 21.7%, and 22.5% in New York City, Washington, D.C., Chicago, and St. Louis, respectively. Using regional monthly mean values (seasonal mean values), the NGE is 20.1% (19.8%) in the northeast subdomain and 11.9% (12.1%) in the Midwest subdomain. These values are much better than the USEPA-suggested threshold. Although no reference threshold was established for the NGE using monthly and sea-

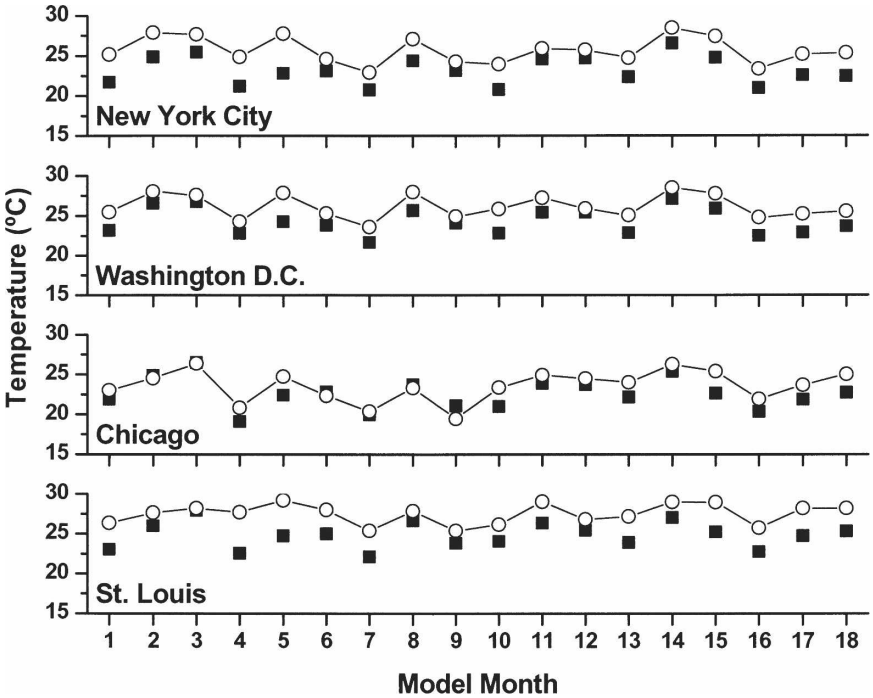

FIG. 11. Similar to Fig. 10, but for monthly mean surface temperature (°C).

sonal means, the result nonetheless suggests that the current modeling system successfully simulated the main monthly characteristics with acceptable accuracy. The results indicate that traditional model evaluation criteria based on monthly mean statistics may be more suitable in evaluating the true performance of a climatic modeling system.

The climatic air quality studies are different from the past historical air quality studies, especially the driving meteorological conditions. When using the RCM-simulated present climate for evaluation purpose, the simulated meteorological conditions are not constrained by the observations as in the past historical air quality studies. The development and movement of meteorological systems were controlled internally by numerical schemes incorporated in the RCM. The simulated meteorological systems may travel through any specific area at different times and/or with some spatial shift. The changes of transport prevented the present modeling system from capturing all air quality variations in limited space and/or of shorter time frame. Nevertheless, the variations of mean quantities in both the temporal and spatial scale are valuable to climatic studies and were captured by the present modeling system. It is reasonable to conclude that the current model system is capable of simulating air quality variation on climate time scales.

5. Summary

An integrated modeling system that consists of RCM meteorological conditions, EM source emissions, and AQM air quality was developed to study potential climate change impacts on regional and local air quality. A 90-day one-way-coupled simulation of tropospheric O_3 formation during summer months from 1995 to 2000 was performed with a focus on the Midwest and northeast and four selected metropolitan areas. The modeling system produced good qualitative simulations of several important features, including the diurnal cycle and multiday periods of high O_3 concentrations resulting from transient weather regimes. The modeling system tended to underestimate nighttime O_3 concentrations in the vicinity of metropolitan areas. However, two subdomains produced a realistic diurnal seasonal mean O_3 distribution over the simulation period. A daytime O_3 overestimate of 5.0 ppb was found in the northeast subdomain over the simulation period. A fairly good simulation was found in the Midwest subdomain, with a bias of 1.4 ppb. It is known that several meteorological factors, including transport and environmental condition, for example, temperature, can impact the atmospheric chemical reactions. The bias of the RCM meteorological conditions, as shown in surface air temperature analyses, was found to have a significant impact on the AQM bias. Because RCM me-

teorological conditions are designed to reproduce monthly and longer time-scale mean statistics, AQM results driven by RCM meteorological conditions cannot fully describe the hourly variation of observed O_3 concentration. Instead, AQM simulations successfully reproduced most of the monthly and seasonal mean values of daytime O_3 in New York, Washington, D.C., and Chicago. The monthly and seasonal mean characteristics in the two subdomains also could be captured by the modeling system. This suggests that the climate modeling system is capable of performing climate change/air quality assessments. Because different modeling systems were designed to study the issues at different temporal and spatial scales, it may be practical to use various reference values to evaluate the model performance. The results indicated that it is more realistic to use monthly mean quantities than hourly average values to evaluate a climatic modeling system. Although offline simulations were performed in this study, the aerosol and greenhouse gases could have potential impact on the climate in a long-term simulation. The impact is not included in this study but will be studied in the future.

Acknowledgments. This work was supported partially by U.S. Environmental Protection Agency Award RD-83096301-0 and the National Computational Science Alliance under ATM040000N that utilized the IBM pSeries 690 and Xeon Linux Cluster. The authors acknowledge NOAA/FSL for supercomputing support. The views expressed are those of the authors and do not necessarily reflect those of the sponsoring agencies or the Illinois State Water Survey.

REFERENCES

Aw, J., and M. J. Kleeman, 2003: Evaluating the first-order effect of intraannual temperature variability on urban air pollution. *J. Geophys. Res.,* **108,** 4365, doi:10.1029/2002JD002688.

Berkowitz, C. M., J. D. Fast, S. Springston, R. Larson, C. Spicer, and P. Doskey, 1998: Formation mechanisms and chemical characteristics of elevated photochemical layers over the United States. *J. Geophys. Res.,* **103,** 10 631–10 647.

——, ——, and R. C. Easter, 2000: Boundary-layer vertical exchange processes and the mass budget of ozone: Observations and model results. *J. Geophys. Res.,* **105,** 14 789–14 805.

Bouchet, V. S., R. Laprise, E. Torlaschi, and J. C. McConnell, 1999a: Studying ozone climatology with a regional climate model, 1, Model description and evaluation. *J. Geophys. Res.,* **104,** 30 351–30 371.

——, ——, ——, ——, and D. A. Plummer, 1999b: Studying ozone climatology with a regional climate model, 2, Climatology. *J. Geophys. Res.,* **104,** 30 373–30 385.

Carter, W. P. L., 1990: A detailed mechanism for the gas-phase atmospheric reactions of organic compounds. *Atmos. Environ.,* **24A,** 481–518.

Chang, J. S., R. A. Brost, I. S. A. Isaksen, S. Madronich, P. Middleton, W. R. Stockwell, and C. J. Walcek, 1987: A three dimensional Eulerian acid deposition model: Physical concepts and formulation. *J. Geophys. Res.,* **92,** 14 681–14 700.

——, S. Jin, Y. Li, M. Beauharnois, C.-H. Lu, H.-C. Huang, S. Tanrikulu, and J. Damassa, 1997: The SARMAP Air Quality Model final report. Air Resource Board, California Environmental Protection Agency.

Cressman, G., 1959: An operational objective analysis system. *Mon. Wea. Rev.,* **87,** 367–374.

Doran, J. C., C. M. Berkowitz, R. L. Coulter, W. J. Shaw, and C. W. Spicer, 2003: The 2001 Phoenix Sunrise experiment: Vertical mixing and chemistry during the morning transition in Phoenix. *Atmos. Environ.,* **37,** 2365–2377.

Dudhia, J., D. Gill, Y.-R. Guo, K. Manning, W. Wang, and J. Chriszar, 2000: PSU/NCAR Mesoscale Modeling System tutorial class notes and user's guide: MM5 Modeling System Version 3. [Available online at http://www.mmm.ucar.edu/mm5/documents/MM5_tut_Web_notes/tutorialTOC.htm.]

Fast, J. D., and W. E. Heilman, 2003: The effect of lake temperatures and emissions on ozone exposure in the western Great Lakes region. *J. Appl. Meteor.,* **42,** 1197–1217.

——, and ——, 2005: Simulated sensitivity of seasonal ozone exposure in the Great Lakes region to changes in anthropogenic emissions in the presence of interannual variability. *Atmos. Environ.,* **39,** 5291–5306.

——, J. C. Doran, W. J. Shaw, R. L. Coulter, and T. J. Martin, 2000: The evolution of the boundary layer and its effect on air chemistry in the Phoenix area. *J. Geophys. Res.,* **105,** 22 833–22 848.

——, R. A. Zaveri, X. Bian, E. G. Chapman, and R. C. Easter, 2002: Effect of regional-scale transport on oxidants in the vicinity of Philadelphia during the 1999 NE-OPS field campaign. *J. Geophys. Res.,* **107,** 4307, doi:10.1029/2001JD000980.

Gery, M. W., G. Z. Whitten, J. P. Killus, and M. C. Dodge, 1989: A photochemical kinetics mechanism for urban and regional scale computer modeling. *J. Geophys. Res.,* **94,** 12 925–12 956.

Guenther, A., P. Zimmerman, and M. Wildermuth, 1994: Natural volatile organic compound emissions rate estimates for U.S woodland landscapes. *Atmos. Environ.,* **28,** 1197–1210.

Hogrefe, C., S. T. Rao, P. Kasibhatla, W. Hao, G. Sistla, R. Mathur, and J. McHenry, 2001: Evaluating the performance of regional-scale photochemical modeling systems: Part II—Ozone predictions. *Atmos. Environ.,* **35,** 4175–4188.

——, and Coauthors, 2004a: Simulating regional-scale ozone climatology over the eastern United States: Model evaluation results. *Atmos. Environ.,* **38,** 2627–2638.

——, and Coauthors, 2004b: Simulating changes in regional air pollution over the eastern United States due to changes in global and regional climate and emissions. *J. Geophys. Res.,* **109,** D22301, doi:10.1029/2004JD004690.

Houghton, J. T., Y. Ding, D. J. Griggs, M. Noguer, P. J. van der Linden, X. Dai, K. Maskell, and C. A. Johnson, Eds., 2001: *Climate Change 2001: The Scientific Basis.* Cambridge University Press, 881 pp.

Houyoux, M. R., J. M. Vukovich Jr., C. J. Coats, N. J. M. Wheeler, and P. S. Kasibhatla, 2000: Emission inventory development and processing for the Seasonal Model for Regional Air Quality (SMRAQ) project. *J. Geophys. Res.,* **105,** 9079–9090.

Huang, H.-C., 2002: Modeling study on the importance of biogenic VOCs in tropospheric ozone formations over the U.S. *Proc. Int. Workshop on Biogenic Volatile Organic Compounds and Ozone Pollution,* Taipei, Taiwan, National Yunlin University of Science and Technology, 65–90.

——, and J. S. Chang, 2001: On the performance of numerical solvers for chemistry submodel in three-dimensional air quality models, 1. Box-model simulations. *J. Geophys. Res.,* **106,** 20 175–20 188.

Kanamitsu, M., W. Ebisuzaki, J. Woolen, S.-K. Yang, J. J. Hnilo, M. Fiorino, and G. L. Potter, 2002: The NCEP-DOE AMIP-II reanalysis (R-2). *Bull. Amer. Meteor. Soc.,* **83,** 1631–1643.

Kunkel, K. E., S. A. Changnon, B. C. Reinke, and R. W. Arritt, 1996: The July 1995 heat wave in the Midwest: A climatic perspective and critical weather factors. *Bull. Amer. Meteor. Soc.,* **77,** 1507–1518.

Liang, X.-Z., K. E. Kunkel, and A. N. Samel, 2001: Development of a regional climate model for U.S. Midwest applications. Part I: Sensitivity to buffer zone treatment. *J. Climate,* **14,** 4363–4378.

——, L. Li, K. E. Kunkel, M. Ting, and J. X. L. Wang, 2004: Regional climate model simulation of U.S. precipitation during 1982–2002. Part I: Annual cycle. *J. Climate,* **17,** 3510–3528.

——, J. Pan, J. Zhu, K. E. Kunkel, J. X. L. Wang, and A. Dai, 2006: Regional climate model downscaling of the U.S. summer climate and future change. *J. Geophys. Res.,* **111,** D10108, doi:10.1029/2005JD006685.

Lu, R., C. Lin, R. Turco, and A. Arakawa, 2000: Cumulus transport of chemical tracers, 1, Cloud-resolving model simulations. *J. Geophys. Res.,* **105,** 10 001–10 021.

Middleton, P., W. R. Stockwell, and W. P. L. Carter, 1990: Aggregation and analysis of volatile organic compound emissions for regional modeling. *Atmos. Environ.,* **24A,** 1107–1133.

NRC, 1991: *Rethinking the Ozone Problem in Urban and Regional Air Pollution*. National Academy Press, 500 pp.

Ranzieri, A. J., and R. Thuillier, 1991: San Joaquin Valley Air Quality Study (SJVAQS) and Atmospheric Utility Signatures, Predictions and Experiments (AUSPEX): A collaborative modeling program. *Proc. 84th Annual Meeting of the Air & Waste Management Association,* Vancouver, BC, Canada, Air and Waste Management Association, Paper 91-70.5.

Sander, S. P., and Coauthors, 2003: Chemical kinetics and photochemical data for use in atmospheric studies. Evaluation Number 14, NASA JPL Publication 02-25, 334 pp.

Seaman, N. L., 2000: Meteorological modeling for air-quality assessments. *Atmos. Environ.,* **34,** 2231–2259.

Seinfeld, J. H., and S. N. Pandis, 1998: *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*. Wiley-Interscience, 1326 pp.

Stockwell, W. R., P. Middleton, J. S. Chang, and X. Tang, 1990: The second generation Regional Acid Deposition Model chemical mechanism for regional air quality modeling. *J. Geophys. Res.,* **95** (D10), 16 343–16 367.

Tao, Z., S. M. Larson, D. J. Wuebbles, A. Williams, and M. Caughey, 2003: A summer simulation of biogenic contribution to ground-level ozone over the continental United States. *J. Geophys. Res.,* **108,** 4404, doi:10.1029/2002JD002945.

Williams, A., M. Caughey, H.-C. Huang, X.-Z. Liang, K. E. Kunkel, Z. Tao, S. Larson, and D. J. Wuebbles, 2001: Comparison of emissions processing by EMS95 and SMOKE over the Midwestern U.S. *10th Int. Emission Inventory Conf. "One Atmosphere, One Inventory, Many Challenges,"* Denver, CO, U.S. EPA.

Zhu, J., and X.-Z. Liang, 2006: Regional climate model simulation of U.S. soil temperature and moisture during 1982–2002. *J. Geophys. Res.,* **110,** D24110, doi:10.1029/2005JD006472.