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Seasonal modeling of regional ozone pollution in the eastern United States

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Abstract. In this study, we report on the results from a comprehensive model simulation of regional ozone (O_3) throughout a season in the eastern United States. The model is shown to perform better in terms of simulating seasonal, rather than episodic, characteristics of the regional surface O_3 distribution. This finding suggests that it may be more appropriate to use models of this type to assess seasonal O_3 patterns rather than following the current paradigm of developing regional control strategies based on event-specific simulations.

Introduction

Ground-level ozone (O_3) pollution continues to be a serious problem in the United States despite three decades of air quality management efforts. In recent years, there has been a growing awareness of the need to consider the transport of O_3 and its precursors across state boundaries in developing state implementation plans for reducing O_3 pollution in the eastern United States. A related development is the recently proposed change to the national ambient air quality standard (NAAQS) for O_3 in the United States from one based on a maximum one-hour average concentration to one based on a maximum eight-hour average concentration [*United States Environmental Protection Agency*, 1997].

Regional-scale air quality models (AQMs), which aid in the analysis and assessment of the impact of regional processes such as long-range transport of O_3 and its precursors, are key components of O_3 pollution management efforts. Typically, AQMs have been used to study extreme O_3 events, thereby significantly limiting the range of chemical conditions under which these models have been evaluated. Furthermore, model evaluation procedures have typically focused on episodic metrics such as the ability to predict day-specific peak O_3 concentrations. Most recently, the Ozone Transport Assessment Group (OTAG) has evaluated the role of transport in O_3 non-attainment in the eastern United States [*LeClair*, 1997]. Again, the OTAG simulations followed an episodic modeling and evaluation paradigm [*Lurmann and Kumar*, 1997].

Schere and Wayland [1989] made an early attempt at a longer-term model evaluation by using a regional model to simulate the O_3 distribution in the northeastern U.S. over a six-week period in 1980. More recently, Winner and Cass performed a simulation of O_3 extending over a whole year for a 600 km x 200 km domain in southern California [*Winner and Cass*, 1999]. Both these studies used models with a relatively coarse vertical structure, and were driven using meteorological fields derived from a limited number of observations over the respective model domains.

In this paper, we report on the results of a comprehensive seasonal simulation of the O_3 distribution in the eastern United States using a regional AQM driven using output from a sophisticated meteorological modeling system. The focus here is on characterizing how well the model simulates the spatial distribution of ground-level O_3 over the course of a season.

Other groups have recently performed simulations for periods of one month or longer using comprehensive AQMs. *Sirois et al.* [1999] have used a time-series approach to evaluate a one month simulation of surface O_3 over eastern North America. More relevant to our study are two longer-term studies over eastern North America, one by *Bouchet et al.* [1999] in which O_3 simulations for five consecutive Julys are evaluated, and the other by Hogrefe et al. (C. Hogrefe, personal communication, 1999) in which a time-series approach is used to analyze simulated O_3 concentrations over a 3 month period. We discuss our results in the context of the results from these two studies in the last section of this paper.

Air quality model description

The modeling system used in this study has been described by *Wheeler et al.* [1998]. The AQM is the non-hydrostatic version of the Multiscale Air Quality Simulation Platform (MAQSIP) [*Odman and Ingram*, 1996]. In the current application, 22 sigma levels were used in the vertical direction up to 100 mb, and the horizontal grid spanned the eastern United States with a resolution of 36 km (see Figure 1). The AQM was driven using meteorological fields from the Mesoscale Meteorological 5 (MM5) model [*Grell et al.*, 1995], which was exercised in a four-dimensional data assimilation mode using analysis nudging with re-initialization every 5 days. The MM5 model was run in a nested mode with an inner grid (of the same resolution as the AQM grid) nested within a coarser 108 km grid which covered a larger domain as shown in Figure 1.

The chemical mechanism used in the AQM was the Carbon Bond Mechanism (Version 4.2) [*Gery et al.*, 1989; *Kasibhatla et al.*, 1997] in order to retain consistency with models which are typically used in the air quality planning process in the United States (such as the OTAG models). Nominal

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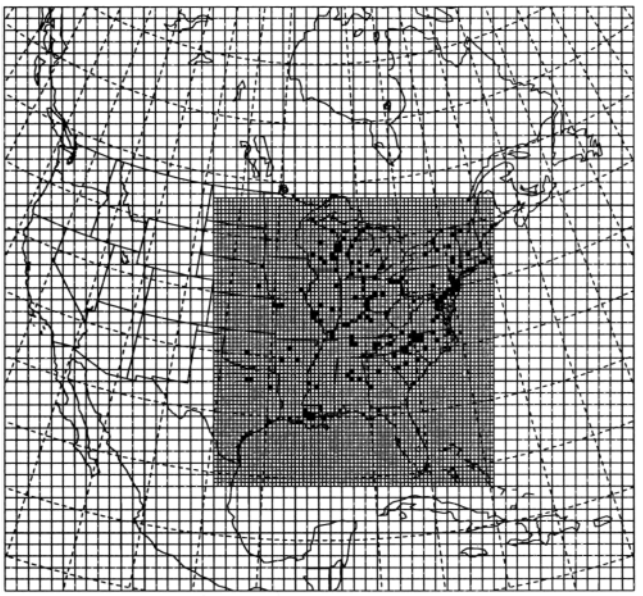

Figure 1. Maps of the outer 108 km MM5 model grid and the nested 36 km MM5 and MAQSIP model grid. Also shown are the locations (black circles) at which model surface O_3 concentrations are compared to gridded observations.

clean background concentrations of O_3 and related species were prescribed at the AQM boundaries. In particular, a time-invariant boundary concentration of 35 ppbv was specified for O_3 to retain consistency with the OTAG simulations. The emission inventories used in this study have been described in detail by *Houyoux et al.* [1999]. Temporally- and spatially-varying anthropogenic emissions of carbon monoxide, volatile organic compounds (VOC), and nitrogen oxides (NO_x) were prescribed in the AQM on the basis of the most current national emission inventories, and biogenic emissions were calculated as a function of model meteorology using the Biogenic Emission Inventory System 2 (BEIS2).

The AQM was used to simulate the distribution of O_3 over a 120-day period from May 15 to September 11, 1995. Model evaluation focused on the last 113 days of the simulation, allowing 7 days for model spin-up.

Model evaluation procedure

The extent to which an AQM captures the space-time variation in the regional O_3 distribution is a critical test of its overall accuracy, and this test must serve as a necessary step before the model can be used to evaluate alternative O_3 control strategies in the presence of real meteorological variability. Furthermore, since the focus of this paper is on a comparison of episodic and seasonal model performance, we restrict our attention to ground-level O_3 which is the only quantity for which measurements of the appropriate spatial and temporal scope are available. The aforementioned non-episodic AQM studies of relevance to our study have had a similar focus on ground-level O_3.

In this paper, we focus on comparisons of model predictions with gridded daytime (10am–5pm EST) surface O_3 concentrations derived from the Aerometric Information Retrieval System (AIRS) monitoring network. In particular, we use the Pearson correlation coefficient (r) as a convenient metric for characterizing the extent to which the model captures the observed structure in surface O_3. We also note that while the analysis presented here is based on 10am–5pm concentrations, similar results are obtained when only afternoon (1pm–5 pm EST) concentrations are used in the analysis.

A multi-step procedure was used to screen the AIRS O_3 measurements in an attempt to minimize biased comparisons with the grid-average model results (e.g., to avoid periods during which local processes such as O_3 titration by fresh NO_x emissions may have influenced the measurements, to minimize the effect of boundary-conditions, and to minimize biases due to missing data). The following sequential procedure was used to develop the gridded dataset for model evaluation: (i) only sites which were classified as rural AIRS sites were selected for analysis; (ii) from the rural subset, only sites which were located at least 5 grid boxes (180 km) from the nearest model boundary and for which measurements were available for each hour from 10 am to 5 pm were selected for analysis; (iii) the data from the selected sites were averaged within the corresponding model grid cells to create a gridded daytime-average dataset for each day during the season; (iv) grid boxes in which the daytime-average O_3 concentration was less than 20 ppbv on any particular day were removed from the gridded daily dataset for that day; and (v) grid boxes which had less than 100 days (out of a maximum of 113 days) of gridded measurements were removed from the gridded dataset. This procedure resulted in a set of 137 grid locations (shown in Figure 1) at which model results were compared to gridded measurements.

Episodic and seasonal perspectives on model performance

We first evaluate the model from an episodic perspective by considering the values of r on a day-to-day basis as shown in Figure 2. It is readily evident that the daily r values are quite variable during the course of the simulation, and are often quite low. These features do not appear to be unique to our model. Reported values of r^2 for daily maximum O_3 concentrations for four episodic model runs by OTAG range from 0.0 to 0.7, depending on the particular day and

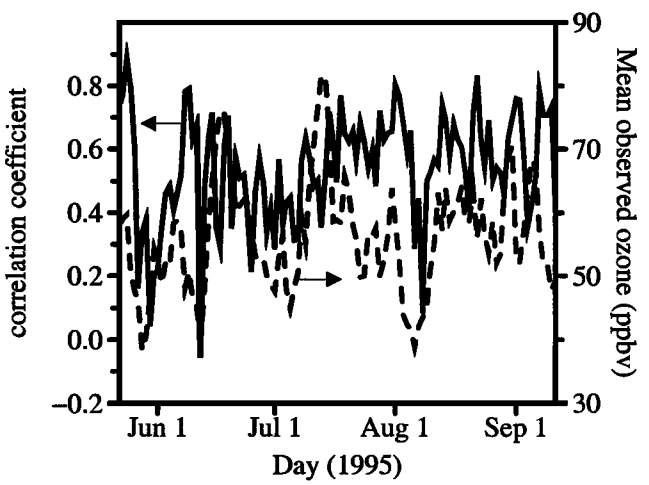

Figure 2. Day-to-day variation in the Pearson correlation coefficient (solid line) and the observed domain-mean 10am–5pm average surface O_3 concentration (dashed line).

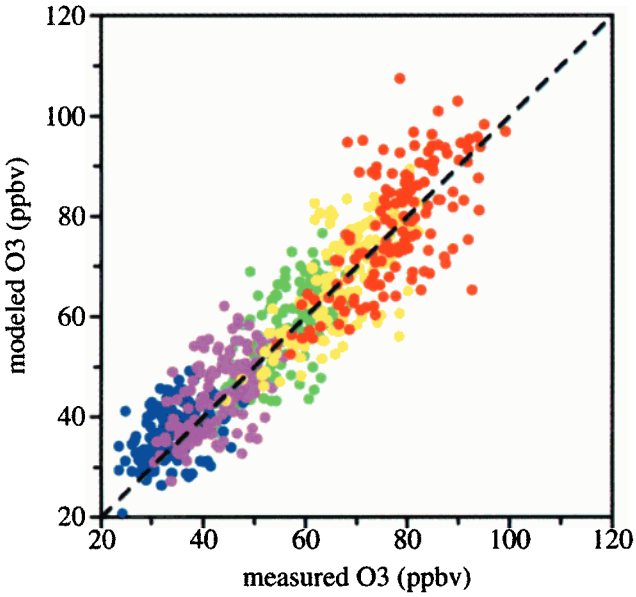

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Plate 1. Scatter plots of modeled versus observed 10am–5pm average surface O_3 concentrations at various percentiles (blue: 10th percentile; magenta: 25th percentile; green: 50th percentile; yellow: 75th percentile; red: 90th percentile). The dashed line is the 1:1 line.

subdomain considered. Figure 2 also shows that there is little correspondence between the observed domain-mean O_3 concentrations and the corresponding r values. While the lowest r values occur on days when the domain-mean O_3 is less than 50 parts per billion by volume (ppbv), there are a number of instances when the domain-mean O_3 concentration is relatively high and the corresponding r value is low and vice versa. For example, on August 5 the r value is 0.66 while the observed domain-mean O_3 is 42 ppbv. By contrast, on June 16 the r values is 0.35 while the domain-mean O_3 concentration is 72 ppbv. We also note that the low values of r on certain days are not simply due to a few outliers.

We next examine the model from a seasonal perspective by calculating r on a percentile basis [i.e. at each location a specific percentile (e.g., the median) of the observed and modeled time-series is selected and the value of r associated with this percentile is calculated]. Calculated values of r for the 10th, 25th, 50th, 75th, and 90th percentiles are 0.43, 0.56, 0.70, 0.72, and 0.66, respectively. The scatter plots of observed versus modeled concentrations corresponding to these percentiles are shown in Plate 1. It can be seen that, when viewed in this space-frequency context, the model captures the dynamic range of the observations quite well. The lower r values for the 10th and 25th percentiles are probably due to boundary conditions which cannot be accurately prescribed and the effects of which cannot be fully eliminated by our data filtering scheme.

The scatter around the 1:1 line in Plate 1 illustrates the difficulty in simulating O_3 concentrations at specific locations with a regional AQM. However, what is significant is that the r values for the 50th, 75th, and 90th percentiles are relatively high, and are in fact similar in magnitude to the highest daily r values during the course of the simulation. Of the 113 daily r values plotted in Figure 2, only 33 are higher than 0.65. Perhaps just as significant is the fact that consistently high r values are obtained for the 50th, 75th, and 90th percentiles, which is in sharp contrast to the variability in the daily r values shown in Figure 2. While this situation could arise in an extreme situation in which the model simulated O_3 is in the right range but is consistently out of phase with the measurements, Figure 1 shows that this is not the case.

Discussion and summary

A number of inherent uncertainties may account for the differences between observations and models including, but not limited to, differences between the prescribed and actual emissions, differences between the modeled and actual synoptic and sub-synoptic scale distribution of clouds, and uncertainties associated with the chemical mechanism and boundary conditions used in the model. In addition, the impact of these inherent uncertainties on model predictions can vary from day to day over the course of a typical season.

Given these inherent uncertainties which are common to both episodic and seasonal simulations, our results suggest that simulations of the ensemble of events that shape the spatial distribution of O_3 on a seasonal scale may be more robust than simulations of particular episodic events. While it may be possible to tune certain model parameters to simulate specific events, the day-to-day variability in model performance suggests that tuning a model to reproduce characteristics of a particular episode may have the consequence of degrading model performance during other periods.

Our conclusions, while based on a specific modeling system, are consistent with two recent studies of similar scope. The study by Hogrefe et al. (C. Hogrefe, private communication, 1999), based on an entirely different AQM and a different analysis approach, also finds that short-term O_3 variations are not well characterized by a model of the type used here. Our findings are also consistent with the results of *Bouchet et al.* [1999] who find that the climatology of O_3 during July over a 5-year period is better simulated than the O_3 distribution during a specific time period. We also emphasize that our analysis focuses on regional O_3, and not on short-term urban O_3 episodes, in keeping with the intended use of these types of models.

While episodic O_3 pollution modeling in concert with targeted field missions will continue to provide useful process-level insights, the seasonal analysis presented here suggests that an alternative paradigm may be worth exploring in the context of using AQMs for designing regionally-based O_3 pollution control strategies. Rather than developing regional emission control strategies based on a limited number of extreme O_3 simulations, it may be more appropriate to examine the effects of control strategies on the frequency distribution of O_3 concentrations. In this context, enhanced regional networks for monitoring O_3 and important O_3-related species such as reactive nitrogen compounds, VOC, and peroxides will be particularly useful in terms of evaluating the ability of models to accurately simulate the regional distribution and temporal variability of ground-level O_3 and O_3-precursor relationships. An evaluation of this sort is a necessary and critical first step in designing credible emission control strategies for solving the O_3 pollution problem in the eastern United States.

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