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A COMPARISON OF OBSERVED AND SIMULATED LONG-TERM OZONE FLUCTUATIONS AND TRENDS OVER THE NORTHEASTERN UNITED STATES

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1. INTRODUCTION

Measured ozone fluctuations occur on temporal scales ranging from hours to years and spatial scales ranging from urban to hemispheric. While regional photochemical modeling systems are commonly used for a variety of regulatory and research applications, their ability to capture the observed range of ozone fluctuations across multiple scales relevant to such applications has not been fully evaluated. We present and analyze the results of an 18-year air quality simulation over the Northeastern U.S. performed with a regional photochemical modeling system consisting of the MM5 meteorological model (Grell et al., 1994), the Sparse Matrix Operator Kernel Emissions (SMOKE) system (Houyoux et al., 2000) emissions processor, and the Community Multiscale Air Quality (CMAQ) chemistry transport model (Byun and Schere, 2006). The analysis focuses on the comparison of observed and simulated trends and variability of ground level ozone concentrations. Furthermore, we also present results based on an 18-year sensitivity simulation in which we derived boundary conditions from archived monthly mean fields of global chemistry simulations performed with the ECHAM5-MOZART modeling system rather than using the climatological time-invariant boundary conditions used in the original simulation.

2. DATABASE AND MODELING SYSTEM

The following is a brief summary of the model set-up used to perform the simulations analyzed in this study. The reader is referred to Hogrefe et al. (2009) for additional details. The MM5 model was used to simulate meteorological conditions for the time period from January 1, 1988 to December 31, 2005. The meteorological simulations were performed on two nested grids with 36km and 12km grid cell sizes covering the Northeastern U.S. Throughout the model simulation, MM5 was nudged towards NCEP reanalysis fields using four-dimensional data assimilation. All emissions processing, including mobile sources and biogenic sources, was performed within the SMOKE system. Anthropogenic emission inventories for the 1988-2005 time period were compiled from a variety of sources as described in Hogrefe et al. (2009). Biogenic sources for the entire modeling period from 1988 to 2005 were estimated with the BEIS3.12 model taking into account MM5 temperature, radiation, and precipitation. Using these meteorological and emission fields, air quality simulations were performed with CMAQ version 4.5.1 on two nested grids of 36km and 12km horizontal spacing, corresponding to the MM5 grids except for a ring of buffer cells. Gas phase chemistry was represented by the CB-IV mechanism (Gery, 1989) while aerosol chemistry was simulated with the "aero3" module. For all subsequent analyses, only results from the 12 km CMAQ simulations were utilized. For the first set of simulations, the hourly boundary conditions for the 36 km grid were derived from time-invariant climatological vertical profiles while the 36km simulation was used to create hourly boundary conditions for the 12km grid. These simulations are subsequently referred to as CMAQ/STATIC. For the second set of 1988-2005 simulations, hourly chemical boundary conditions for the 36km grid were extracted and temporally interpolated from archived monthly-mean fields of global chemistry simulations performed for the 1988-2005 time period with a ECHAM5-MOZART modeling system as part of the RETRO project (http://retro.enes.org/index.shtml). These simulations are subsequently referred to as

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CMAQ/ECHAM5-MOZART. Note that the archived ECHAM5-MOZART fields did not contain all CMAQ gas phase species and no aerosol species. For the unavailable species, including most VOC groups except isoprene, the same time-invariant climatological values used in the CMAQ/STATIC simulations were used in the CMAQ/ECHAM5-MOZART simulations. Table 1 shows the average boundary conditions for selected species and layers for both simulations, we averaged these concentrations over all boundary cells and, in the case of the CMAQ/ECHAM5-MOZART simulations, over the entire simulation time period. For the CMAQ/ECHAM5-MOZART simulations, boundary conditions for layers 14 and 15 were set to the same value as for layer 13 to avoid intrusion of stratospheric concentration values because the vertical resolution of the MM5/CMAQ simulations in the upper troposphere and lower stratosphere was not set up to properly handle stratosphere-troposphere exchange processes.

Hourly ozone observations from 1988 to 2005 were obtained from the U.S. EPA Air Quality System (AQS). All data were screened for completeness prior to analyses, and any monitor with more than 60% of missing data during any year was excluded from the analysis. This screening resulted in the selection of 90 sites. For all subsequent analyses, monitored values were assigned to the model grid cells in which the monitor was located.

For the evaluation of upper air ozone simulations, ozonesonde observations taken at two sites within the 36km CMAQ modeling domain were obtained from the World Ozone and Ultraviolet Radiation Data Center (WOUDC). These two sites are Wallops Island, Virginia and Huntsville, Alabama, where the number of available ozonesonde launches during the analysis time period was 660 and 305, respectively.

3. RESULTS

While the focus of the analysis in the paper is on the comparison of observed and simulated ozone variability and trends over 18 years, we also compiled standard statistical measures of model performance for May-September for 8-hr daily maximum ozone concentrations. The results of this analysis across the 18 years and 90 monitors reveal a similar level of model performance as reported in other studies for individual years (e.g. Eder and Yu, 2006; Appel et al., 2007) with an absolute (normalized) bias of +4.5 ppb (+8.9%) and an absolute (normalized) root mean square error of 14.5 ppb (28.2%). At the 95th percentile of May-September 8-hr daily maximum ozone concentrations, the absolute (normalized) bias is -1.3 ppb (-1.6%) and the absolute (normalized) root mean square error is 7.8 ppb (9.4%). At the 5th percentile of May-September 8-hr daily maximum ozone concentrations, the absolute (normalized) bias is +12.3 ppb (+55.8%) and the absolute (normalized) root mean square error is 13.4 ppb (59%), indicating the model tends to slightly underestimate high values and strongly overestimates low observed values.

As a first step in comparing observed and simulated variability, Figure 1 presents power spectra calculated from 18 years of hourly observed and CMAQ/STATIC ozone time series. To reduce the noise in the spectra and facilitate the comparison, we calculated the spectra at 12 selected sites and then averaged the spectral density at each frequency over these sites. Figure

Table 1: Average boundary conditions for selected species and layers. The concentrations in the “Profile” columns were used for the CMAQ/STATIC simulations while those in the “MOZART” columns were used for the CMAQ/ECHAM5-MOZART simulations.

Layer	Midpoint Height (m)	O_3 (ppb)		NO (ppt)		NO_2 (ppt)		PAN (ppt)		CO (ppb)	
		Profile	MOZART	Profile	MOZART	Profile	MOZART	Profile	MOZART	Profile	MOZART
1	18	32	49	44	153	89	1,907	68	664	77	168
8	560	38	55	38	45	76	513	62	543	77	145
10	1,403	45	57	22	17	44	177	48	434	76	131
12	3,855	56	63	4	7	8	40	25	320	69	105
13	6,139	62	69	0	8	0	31	14	308	64	98
14	9,480	69	69	0	8	0	31	11	308	56	98
15	13,004	70	69	0	8	0	31	11	308	55	98

1 illustrates that CMAQ/STATIC tends to capture the variability in the diurnal and synoptic bands but underestimates variability in the high-frequency (intra-day) and low-frequency (seasonal and longterm) bands of the spectrum. The underestimation of the intra-day variability is consistent with earlier analyses of simulations for single summers (Hogrefe et al., 2001) while an analysis of the strength of longer-term fluctuations had not been possible previously because of the limited duration of simulations.

To further study longer-term variability, we calculated inter-annual variability (IAV) of observed and CMAQ/STATIC 8-hr daily maximum ozone as follows. First, we rank-ordered each year's May-September distribution of daily maximum 8-hr ozone at each site. Next, for each rank we calculated IAV as the standard deviation of these 18 values divided by the mean of these 18 values. We performed this calculation separately for observations and the CMAQ/STATIC simulations at each site. Figure 2a shows boxplots of the observed and simulated IAV for the 5th, 25th, 50th, 75th, and 95th percentiles of May – September 8-hr daily maximum ozone; the box plots show the distribution of IAV for a given percentile across all 90 sites. It is evident that the CMAQ/STATIC IAV is lower than the observed IAV for all percentiles. This is confirmed by Figure 2b which shows the ratio of simulated to observed IAV versus all percentiles of May – September 8-hr daily maximum ozone. While this ratio is less than one for all percentiles, the underestimation is most pronounced for the lower percentiles.

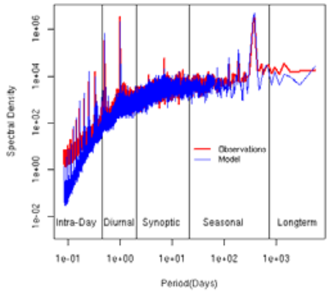

Figure 1. Power spectrum derived from time series of hourly observed and CMAQ/STATIC simulated ozone concentrations for 1988 – 2005.

In addition to comparing observed and simulated variability on interannual timescales, the extended simulation period also provides an opportunity to compare observed and simulated trends in ozone concentrations. For this analysis, linear trends were estimated at each site for each percentile of the rank-ordered May – September 8-hr daily maximum ozone concentrations over the 1988–2005 time period. Figure 3 shows the magnitude of these trends on the y-axis plotted against the percentiles on the x-axis. While trends were calculated separately at each site, the median across all sites is shown in this figure. Results indicate that the agreement between the linear trends estimated from observations and CMAQ/STATIC is better for the upper than the lower percentiles. While typical observed trends at stations in the modeling domain tend to be upward for percentiles <40 and downward for higher percentiles, CMAQ/STATIC trends tend to be downward or flat at all percentiles.

Because lower percentiles of the summertime ozone distribution tend to be more influenced by

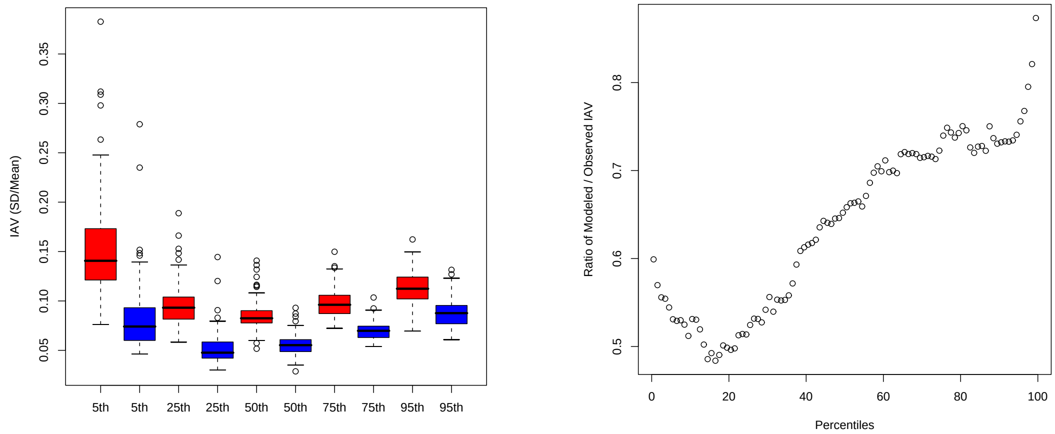

Figure 2. a) Boxplots of the observed and CMAQ/STATIC simulated IAV for the 5th, 25th, 50th, 75th, and 95th percentiles of May – September 8-hr daily maximum ozone; the box plots show the distribution of IAV for a given percentile across all 90 sites. b) ratio of CMAQ/STATIC simulated to observed IAV vs. all percentiles of the May – September 8-hr daily maximum ozone distribution; the median ratio across all 90 sites is shown.

background conditions and boundary conditions, the use of time-invariant lateral boundary conditions in the CMAQ/STATIC simulations likely contributed to the underestimation of interannual variability and the disagreement between observed and simulated ozone trends, especially for lower percentiles. To investigate this hypothesis, we repeated the analysis of IAV and trends for the 18 year CMAQ simulations that utilized chemical boundary conditions derived from monthly-mean concentrations from archived ECHAM5-MOZART simulations as described in Section 2. Figures 4a and b show the results of this analysis, with the IAV analysis (analogous to Figure 2b) displayed in Figure 4a and the trend analysis (analogous to Figure 3) displayed in Figure 4b. The results of the IAV analysis indicate that both sets of CMAQ simulations underestimate observed IAV with modeled/observed IAV ratios less than 1, but also shows that the CMAQ simulation deriving its boundary conditions from the archived ECHAM5-MOZART simulations significantly improves the representation of IAV for mid and low percentiles.

Because of these pronounced impacts of the choice of boundary conditions on variability and trends, it is of interest to further study the differences between these two simulations. Figure 5 shows differences in monthly average daily maximum ozone concentrations between the CMAQ/ECHAM5-MOZART and CMAQ/STATIC simulations for model layer 1 for January, April, July, and October, each averaged over the 18 years of the simulation period. While the impact of different boundary conditions on monthly average daily maximum ozone decreases towards the interior of the domain, it still reaches 3-9 ppb in July for the regions typically exhibiting the highest observed ozone concentrations. Figures 6 a-b display the impact of different boundary conditions on average daily maximum ozone concentrations as function of day-of-year, again averaged over 1988 – 2005. It can be seen that CMAQ/ECHAM5-MOZART generally yields higher concentrations than CMAQ/STATIC and that the differences are largest in spring and fall and can be as large as 12 ppb averaged over all sites. The higher concentrations for the CMAQ/ECHAM5-MOZART simulations are consistent with Table1 that showed higher boundary conditions for ozone as well as NOx and PAN compared to the time-invariant static profile. It is also evident that the CMAQ/STATIC concentrations are generally closer to observed concentrations than the CMAQ/ECHAM5-MOZART concentrations.

While the analysis presented thus far has

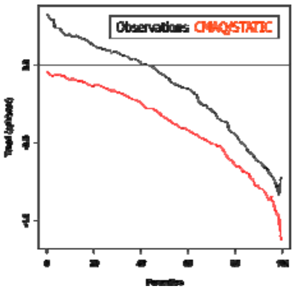

Figure 3. Linear trends (ppb/yr) of observed and CMAQ/STATIC simulated daily maximum 8-hr ozone for 1988 – 2005 for all percentiles of the May – September distribution. The median value across all 90 sites is shown.

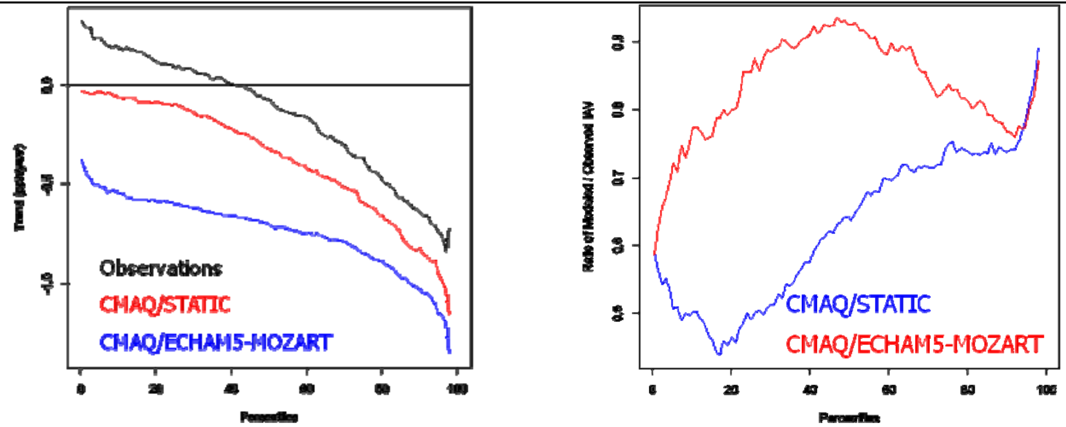

Figure 4. a) Linear trends (ppb/yr) of observed, CMAQ/STATIC, and CMAQ/ECHAM5-MOZART simulated daily maximum 8-hr zone for 1988 – 2005 for all percentiles of the May – September distribution. b) ratio of simulated to observed IAV vs. all percentiles of the May – September 8-hr daily maximum ozone for CMAQ/STATIC and CMAQ/ECHAM5-MOZART. Both panels show the median values across all 90 sites.

focused on surface observations, the choice of lateral boundary conditions also is expected to have a significant impact on simulated concentrations in the free troposphere. Figures 7a-d show a comparison of observed and modeled vertical profiles of the average and standard deviation of ozone concentrations across all available launches at the two ozonesonde sites described in Section 2. We restricted the comparison to CMAQ layers that are completely within the troposphere because of the limited vertical resolution of these simulations in the tropopause region as discussed in Section 2. The mean concentration profiles show that the CMAQ/STATIC simulations are closer to observations than the CMAQ/ECHAM5-MOZART simulations throughout the troposphere at Wallops Island. At Huntsville, the CMAQ/STATIC simulations are closer to observations in the lower and upper troposphere while the CMAQ/ECHAM5-MOZART simulations are closer to observations in the mid troposphere. The comparison of observed and simulated vertical profiles of ozone standard deviations over all available launches shows better performance for the CMAQ/ECHAM5-MOZART simulations at both sites, especially in the free troposphere. Overall, these profiles confirm that boundary conditions have a profound impact on simulated ozone concentrations throughout the troposphere, that the CMAQ/ECHAM5-MOZART simulations have a tendency for overpredictions that is less evident in the CMAQ/STATIC simulations, and that the CMAQ/ECHAM5-MOZART simulations capture more of the observed variability than the CMAQ/STATIC simulations, especially in the free troposphere.

4. SUMMARY

This study presented a comparison of observed and simulated ozone variability and trends over the Northeastern U.S. for 1988 – 2005. We performed simulations with two sets of boundary conditions, one corresponding to a time-invariant climatological vertical profile and the other derived from monthly mean concentrations extracted from archived ECHAM5-MOZART global simulations. Analysis of the CMAQ simulations

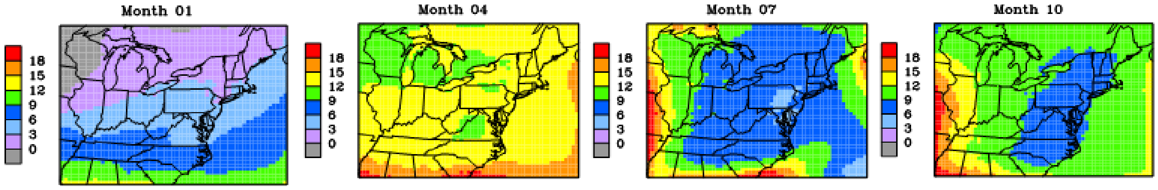

Figure 5. Differences (ppb) of monthly average daily maximum ozone concentrations between the CMAQ/ECHAM5-MOZART and CMAQ/STATIC simulations for model layer 1 for January, April, July, and October, each averaged over the 18 years of the simulation period.

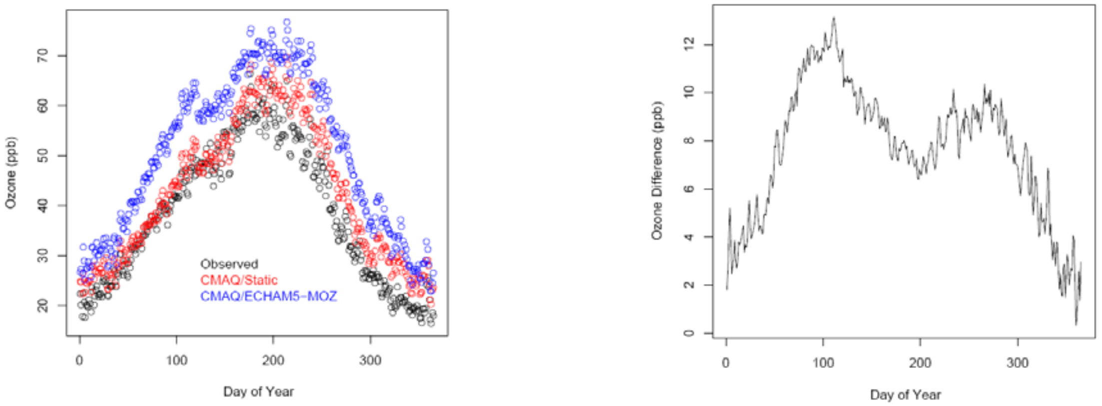

Figure 6. a) Average 1988-2005 daily maximum ozone concentrations as function of day-of-year for observations, CMAQ/STATIC, and CMAQ/ECHAM5-MOZART. b) Difference between the CMAQ/ECHAM5-MOZART and CMAQ/STATIC average 1988-2005 daily maximum ozone concentrations as function of day-of-year.

using the time-invariant boundary conditions indicates that the observed downward trend in the upper percentiles of summertime ozone concentrations is captured by the model in both directionality and magnitude. However, for lower percentiles there is a marked disagreement between observed and simulated trends. In terms of variability, the CMAQ simulations using the time-invariant boundary conditions simulations underestimate observed inter-annual variability by 30% - 50% depending on the percentiles of the distribution. The use of boundary conditions from the ECHAM5-MOZART simulations improved the representation of interannual variability. However, it was also shown that possible biases in the global simulations have the potential to significantly affect ozone simulations throughout the modeling domain, both at the surface and aloft. These results highlight the significant impact lateral boundary conditions can have on a regional air quality model's ability to simulate long-term ozone variability and trends, especially for the lower percentiles of the ozone distribution.

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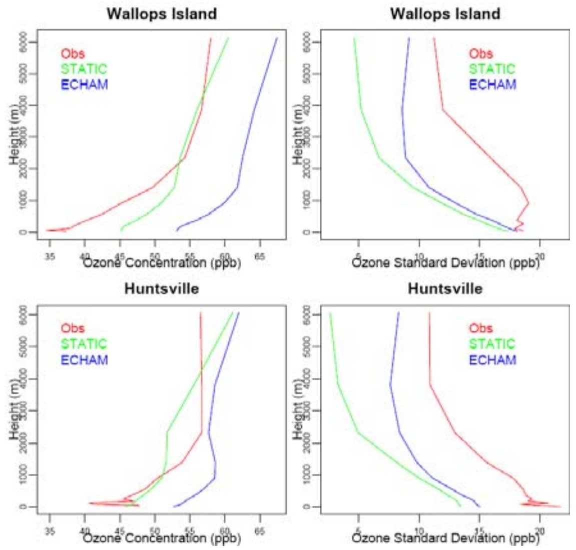

Figure 7. Observed and modeled vertical profiles of the average (a,c) and standard deviation (b,d) of ozone concentrations across all available launches at Wallops Island, VA, and Huntsville, AL.

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