Dynamic Evaluation of a Regional Air Quality Model: Assessing the Emissions-Induced Weekly Ozone Cycle

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Abstract

Air quality models are used to predict changes in pollutant concentrations resulting from envisioned emission control policies. Recognizing the need to assess the credibility of air quality models in a policy-relevant context, we perform a dynamic evaluation of the Community Multiscale Air Quality (CMAQ) modeling system for the "weekend ozone effect" to determine if observed changes in ozone due to weekday-to-weekend (WDWE) reductions in precursor emissions can be accurately simulated. The weekend ozone effect offers a unique opportunity for dynamic evaluation, as it is a widely documented phenomenon that has persisted since the 1970s. In many urban areas of the Unites States, higher ozone has been observed on weekends than weekdays, despite dramatically reduced emissions of ozone precursors (nitrogen oxides [NO_x] and volatile organic compounds [VOCs]) on weekends. More recent measurements, however, suggest shifts in the spatial extent or reductions in WDWE ozone differences. Using 18-years (1988-2005) of observed and modeled ozone and temperature data across the northeastern United States, we re-examine the long-term trends in the weekend effect and confounding factors that may be complicating the interpretation of this trend and explore whether CMAQ can replicate the temporal features of the observed weekend effect. The amplitudes of the weekly ozone cycle have decreased during the 18-year period in our study domain, but the year-to-year variability in weekend minus weekday ozone amplitudes is quite large. Inter-annual variability in meteorology appears to influence WEWD differences in ozone, as well as WEWD differences in VOC and NO_x emissions. Because of the large inter-annual variability, modeling strategies using a single episode lasting a few days or a few episodes in a given year may not capture the WEWD signal that exists over longer time periods. The CMAQ model showed skill in predicting the absolute values of ozone concentrations during the daytime. However, early morning NO_x concentrations were underestimated and ozone levels were overestimated. Also, the modeled response of ozone to WEWD differences in emissions was somewhat less than that observed. This study reveals that model performance may be improved by (1) properly estimating mobile source NO_x emissions and their temporal distributions, especially for diesel vehicles; (2) reducing the grid cell size in the lowest layer of CMAQ; and, (3) using time-dependent and more realistic boundary conditions for the CMAQ simulations.

Key words: dynamic model evaluation, CMAQ model, weekend ozone effect, air quality modeling, ozone

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1. Introduction

Photochemical simulation models such as the Community Multiscale Air Quality (CMAQ) modeling system (Byun and Schere, 2006) are being used as air quality management tools for predicting the change in air pollution levels resulting from proposed emission control strategies. To establish model credibility, most air quality models undergo operational evaluations, in which predictions are compared to observations (matched in time and space) for “base case conditions” (e.g., Eder et al., 2006; Tesche et al., 2006; Appel et al., 2007). In assessing model performance from a policy perspective, Hogrefe et al. (2008) and Dennis et al. (2010) strongly recommend that model evaluations be extended to include dynamic evaluation of modeled response to changes in emissions and meteorological conditions. As defined by Dennis et al. (2010), a “dynamic evaluation” explores whether a model can accurately respond to changes in emissions and meteorology and is a requirement for properly evaluating an air quality model in the context of its policy-relevant application.

Examples of dynamic evaluation include Hogrefe et al. (2001a and b) and Biswas and Rao (2001) who examined whether a model was able to capture the energy content across different spectral bands, attributable to meteorological forcings operating on different time scales. An example of a dynamic evaluation focusing on emission changes is Gilliland et al. (2008), who analyzed the response of CMAQ for modeling ozone for a summer period before and after the implementation of a major NO_x emissions reduction control strategy (referred to as the NO_x-SIP call, www.epa.gov/airmarkets/progsregs/nox/sip.html). This study is indeed motivated by the findings of Gilliland et al. (2008) and Godowitch et al. (2010) who reported that modeled ozone response to emission changes was somewhat less than that observed.

One of the challenges in performing a dynamic evaluation is to identify a time period with a well-defined change in meteorology and/or emissions. The difference in ozone precursor emissions between weekday and weekends offers an excellent opportunity for a dynamic evaluation because of the persistent and large reduction in ozone precursor emissions that occur on weekends relative to weekdays (Rao, 1988). While a few model evaluation studies have examined the weekend effect (e.g., Marr and Harley, 2002b; Yarwood et al., 2003; Jimenez et al., 2005), these studies tend to be restricted to small spatial domains and brief meteorological periods.

The so-called "weekend ozone effect", in which it was originally reported that daytime levels of ozone tended to be higher on weekends than weekdays in many urban areas, has been widely documented since at least the 1970s (e.g., Cleveland et al., 1974; Karl, 1978; Altshuler et al., 1995; Tonse et al., 2008). Analyses of more recent observations across the eastern United States (Lawson, 2003; Blanchard et al., 2008), however, reveal that the weekend enhancement of ozone has diminished or is negligible, despite greatly reduced levels of ozone precursor emissions (volatile organic compounds [VOCs] and nitric oxides [NO_x]) on weekends. In addition, Croes et al. (2003) noted shifts in the spatial extent of enhanced weekend ozone levels across many parts of California since the 1970s. Chinkin et al. (2003) estimated that VOC emissions are reduced by ~15% and NO_x emissions are reduced by ~40% on weekends relative to weekdays across southern California in the summer. In light of the spatial and temporal variability in the weekend effect and the large decrease in emissions on weekends, we view the "weekend ozone effect" as an excellent candidate for the dynamic evaluation of a regional air quality model.

With 18 years of modeled and observed ozone concentrations over the northeastern United States (Hogrefe et al., 2009), we examine the CMAQ's ability to respond to weekly changes in emissions over a large domain for a long time period. In conducting this examination, we also investigate whether there are long-term trends in the WEWD effect and whether there are confounding factors complicating the interpretation of the trends. Through this dynamic evaluation, we can assess the credibility of CMAQ in simulating the response in pollutant concentrations stemming from changes in emissions and use the results from this evaluation to guide future model improvements.

2. Database

2.1. Modeling System

The following is a brief summary of the model simulations analyzed in this study. Additional details on the model set-up can be found in Hogrefe et al. (2009). Meteorological conditions for the time period from January 1, 1988 to December 31, 2005 were simulated using the MM5 meteorological model (Grell et al., 1995). The simulations were performed on two nested model grids with 36 km and 12 km grid cell sizes. 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 Sparse Matrix Operator Kernel Emissions (SMOKE) system (Houyoux et al., 2000). Emission inventories for the 1988-2005 time period were compiled primarily from the U.S. Environmental Protection Agency (U.S. EPA) National Emission Trends database, the anthropogenic emission inventories prepared for the U.S. EPA Clean Air Interstate Rule, and the emission inventories prepared for the 2002 Ozone Transport Commission (OTC) modeling platform (OTC, 2007). The same temporal and spatial allocation and speciation profiles were used to process the anthropogenic emission inventories for all years. In particular, the allocation of annual total emission or VMT inventories to individual hours was performed by applying month-of-year, day-of-week, and hour-of-day temporal profiles to each entry in the inventory file. In this study, SMOKE utilized approximately 1,400 month-of-year, 50 day-of-week, and 200 hour-of-day profiles that were assigned to individual emission sources in the inventory based on the type of emission source and in some cases also based on the pollutant and the geographic location of the emissions source. One exception to this temporal allocation approach was the processing of 2000 – 2005 point source emissions, which utilized hourly measured data from Continuous Emission Monitors (CEM) to allocate annual total emissions to individual hours for all point source units where SMOKE could match CEM data against inventory sources. Biogenic sources for the entire modeling period from 1988 to 2005 were estimated with the BEIS3.12 model taking into account MM5 simulated temperature, radiation, and precipitation.

Using the meteorological and emission fields described above, hourly gridded fields of concentrations, wet deposition, and dry deposition for about 80 gas phase and aerosol species were simulated with the Community Multiscale Air Quality (CMAQ) model (Byun and Schere, 2006), version 4.6. This model version is a slight change from the model simulations described in Hogrefe et al. (2009) that were performed with CMAQ version 4.5.1 and were focused on $PM_{2.5}$. The simulations were performed with two nested grids of 36 km and 12 km, corresponding to the MM5 grids except for a ring of buffer cells (Figure 1a). The boundary conditions for the 36 km grid correspond to time-invariant climatological values, while the boundary conditions for the 12 km grid were derived from the 36 km grid simulation. The height of the first model layer was set at 38 m. Gas phase chemistry was represented by the CB-IV mechanism (Gery et al., 1989) while aerosol chemistry was simulated with the "aero3" module. For all subsequent analyses, only results from the 12 km CMAQ simulations were utilized.

2.2. Observations

Hourly ozone observations from 1988 to 2005 were obtained from the U.S. EPA Air Quality System (AQS) (www.epa.gov/oar/data) . In addition, hourly CO and NO_x observations from 1993 to 2005 were obtained from the U.S. EPA AQS. As stated above, only sites located within the 12 km CMAQ modeling domain, shown in Figure 1a, were included in the analysis. All data were screened for completeness prior to analyses, and data with more than 60% of missing data in any given year were excluded from this analysis. For the analysis of 1988-2005 ozone observations, this screening resulted in the selection of 90 sites with at least 40% data completeness in each year. For the May – September ozone season, all of these sites had a data completeness greater than 90%, indicating that the sites with a larger fraction of missing data on a year-round basis were those that did not operate outside the ozone season. For the analysis of co-located ozone, CO, and NO_x sites for 1993-2005, application of the data completeness criterion resulted in the selection of 8 sites. Note that the 8 co-located ozone, CO and NO_x sites for 1993-2005 are a subset of the 1988-2005 ozone sites. Finally, hourly temperature observations for 1988-2005 at sites within the 12 km CMAQ domain were retrieved from the National Center for Atmospheric Research – Data Support Section. The location of the ozone monitors selected for the 1988-2005 analysis, the location of the co-located ozone, CO, and NO_x monitors for the 1993-2005 analysis, and the location of the temperature monitors are displayed in Figure 1b. In all analyses comparing observations and model predictions, monitored values were assigned to the model grid cells in which the monitor was located. All analyses presented in this study were performed for the ozone season, lasting from May 1 through September 30 of each year.

Observations (ozone concentrations and temperature) used for the time series analysis were part of the *Clean Air Status and Trends Network* (CASTNET, www.epa.gov/castnet/data/metdata/). Here, the CASTNET site at Beltsville, MD (BEL116) was chosen for purposes of illustration. Beltsville, MD is situated in the northern outskirts of the Washington D.C. urban area.

3. Extraction of the average weekly cycle

The average weekly cycle for a given variable of interest was determined by computing mean Monday, Tuesday, etc. values over the analysis period according to

$$\overline{X_{DOW}} = \sum_{i=1}^{n} X_{DOW}(i) \qquad (1)$$

where *DOW* stands for a specific day of the week (e.g., Monday through Sunday), and the analysis period represented by *n* can range from single ozone season to all 18 ozone seasons from 1988 to 2005. The weekend-weekday (WEWD) differences were then determined by subtracting the average of the mean Monday through Friday values from the average of the mean Saturday and Sunday values according to

$$WEWD\ Difference\ (X) = \frac{1}{2} \times \left(\overline{X_{Sa}} + \overline{X_{Su}}\right) - \frac{1}{5} \times \left(\overline{X_{Mo}} + \overline{X_{Tu}} + \overline{X_{We}} + \overline{X_{Th}} + \overline{X_{Fr}}\right) \qquad (2)$$

In both equations (1) and (2), X represents the variable of interest such as daily maximum 1-hr and 8-hr average ozone concentrations, 6am-9am average ozone, CO, or NOx concentrations, and the daily maximum and 6am-9am average temperature.

Continuous local-in-time information about the strength and timing of weekly variation for individual sites was extracted with a wavelet filter. The KZFT (Zurbenko and Porter, 1998), a Fourier transform (FT) of the KZ filter (Rao et al., 1997), was used because it can function when some values are missing. The KZFT is defined as follows:

$$Y_t = \frac{1}{(2q+1)} \sum_{k=-q}^{+q} \exp\left(-\, i\, 2\, \pi\, \omega\, k\right) X_{t+k} \qquad (3)$$

The filter is a Fourier transform of a moving window of width 2q+1; ω is a frequency of interest, which in this case is 1/week, or 1/168 hours for hourly observations. The filter is iterative, meaning that the time series Y_t is also filtered in the same manner. Iteration reduces the magnitude of the ringing (artifacts or spurious signals) that characterizes simple moving average filters; we used three iterations.

The choice of the parameter q is a compromise among the width of the frequency band selected, end effects, and bias. As the window size, 2q+1, increases, the width of the frequency band that is selected decreases (a desirable feature) but end effects increase (an undesirable feature caused by incomplete filtering near the ends of a time series). Large window sizes also reduce the local nature of a wave, as values distant in time from the present are incorporated. Bias occurs when periodic processes are sampled at other than integer multiples of the period of interest (one week). For this paper we set q=84 hours, which has (50%) upper and lower cutoff frequencies of 1/133 and 1/226 hours, respectively. Each iteration with q=84 incorporates 1.006 weeks (169 hours). End effects were handled by clipping the ends of filtered time series.

The KZFT is a complex wavelet. The real part of the filtered time series, Y_t, is a wave with frequency ω, and $|Y_t|$ is the instantaneous amplitude of Y(t). The imaginary part of Y_t has information about the instantaneous phase, ph_t (Bloomfield, 2000):

$$ph_t = \Im\left[ln \frac{Y_t \, exp(-i\,2\,\pi\,t\,\omega)}{abs\left(Y_t \, exp(-i\,2\,\pi\,t\,\omega)\right)}\right] \quad (4)$$

Phase can be thought of as the time of the week that a process reaches a maximum. A stable (relatively constant) phase is evidence of periodicity, while a drifting phase indicates that variation captured by the filter is not periodic (weekly in this case).

4. Results

Figures 2 and 3 depict various aspects of observed and simulated WEWD differences for both ozone and temperature across the northeastern U.S. As described in the figure caption, panels (a) – (f) illustrate changes in the observed and simulated distributions of ozone and temperature over the 18 year time period, with Figure 2 displaying results for daily maximum 8-hr ozone and daily maximum temperature and Figure 3 displaying results for 6am-9am average ozone and temperature. Panels (a), (b), (e) and (f) indicate that the WEWD differences for ozone across the northeastern U.S. have decreased during the 18 year period for both the daily maximum 8-hr and the 6-9 a.m. local time average in both observations and model simulations. Despite the similarities in the downward trends of the WEWD differences between observations and model predictions, there are discrepancies in regard to their absolute magnitudes. For the observed daily maximum 8-hr ozone values, 92% of the early years (i.e., 1988-2000) show positive WEWD median differences, while 80% of the later years (i.e., 2001-2005) show negative differences. In contrast, the CMAQ results show that nearly one-half of the median values are close to zero for the early years, and 80% of median values for the later years are more negative than the observed values.

In addition to depicting the noticeable decrease in the WEWD difference after 2000, Figure 2 reveals substantial year-to-year variability in the observed and simulated WEWD differences over this 18-year period. Even though the model results track the year-to-year changes seen in the observations, the modeled WEWD differences tend to be smaller than those observed. To investigate the role of meteorology in contributing to the WEWD ozone differences discussed above, panels (c) – (f) depict corresponding results for temperature, and panels (g) and (h) show the relationship between WEWD differences for ozone and temperature in both observations and model predictions. As expected, Figure 2 illustrates a strong relationship between 8-hr daily maximum ozone and daily maximum temperature, indicating the meteorological influence on WEWD differences in ozone concentrations.

On the other hand, the corresponding panels in Figure 3 for 6am-9am values show little relationship between ozone and temperature, indicating that ozone WEWD differences during the early morning period are largely controlled by changes in emissions rather than in meteorology. In addition, both Figures 2 and 3 illustrate better agreement between modeled and observed temperature WEWD differences than that for ozone. This suggests that while MM5-simulated WEWD temperature variations are quite accurate, other factors in the photochemical modeling system are contributing to the discrepancy between observed and modeled WEWD differences for ozone.

We further probe how the year-to-year variability in meteorology can affect the interpretation of WEWD differences by examining the magnitude of these differences as a function of the number of ozone seasons used to compute the amplitude of the weekly cycle. Figure 4 presents the results of this analysis for 6am-9am ozone in panels (a) and (d), daily maximum 8-hr ozone in panels (b) and (e), and daily maximum temperature in panels (c) and (f) at the CASTNet site in Beltsville, MD. (Other sites in the study region were also analyzed; although not shown here for space considerations, the results were very similar to those at Beltsville.) These panels reveal that the number of ozone seasons used for the analysis can change the interpretation of the WEWD differences. Such differences based on examining a single ozone season or even 2-3 ozone seasons display a large range of values for the WEWD effect depending on the choice of the analysis period, attributable to the strong influence of year-to-year meteorological variability. For example, with only one year of data, WEWD differences of the observed 8-hr daily peak ozone range from +7 ppb in 1989 to -7 ppb in 2001, and similarly, the differences for the daily maximum temperature are positive in 1989 and negative in 2001. Despite substantial year-to-year variability, the use of multiple seasons for the analysis causes the amplitudes for all variables to converge for both observations and model predictions. Consistent with Figures 2 and 3, 6am-9am ozone WEWD differences are consistently positive, but as in the case for daily maximum 8-hr ozone, large year-to-year variability is evident (+1 to +11 ppb). Both observed and modeled amplitudes of early morning ozone converge to ~+5.5 ppb at this site as the number of years used in the analysis increases. Figure 4 suggests that at least seven years of data are needed to help reduce the effect of the inter-annual variability on the daily maximum 8-hr ozone. Of interest are the consistently lower WEWD differences for the years 2001-2005 relative to 1988-1999, suggesting that the photochemical regime, meteorological, and/or emission conditions are different between the earlier vs. later time periods.

Even at locations where the analysis of multi-year time series indicates large positive WEWD differences, there are many weeks when the maximum occurs on a day other than Saturday or Sunday. Figure 5 presents histograms of the observed and simulated weekly phase at all 90 sites for the 1988-2005 time period as defined in Equation (4) and determined by wavelet analysis from time series of hourly and daily maximum ozone concentrations in panels (a) – (b) and (d) – (e). Equation (4) estimates the continuous phase and Figure 5 indicates the frequency with which the wavelet weekly maximum falls on the day indicated. These histograms indicate that there is no clear preference for weekends in terms of which day of the week the weekly maximum occurs. Moreover, the relative ranking of the various days present in the observations is roughly captured by the model, especially the daily maximum values shown in panels (d) and (e). This is confirmed by panels (c) and (f) which show that in about 2/3 of all weeks, there is a phase difference of less than +/- 1 day between the weekly wave in the observations and model outputs, with a narrower distribution for the daily maximum values in panel (f) compared to the hourly values in panel (c).

With respect to individual sites, wavelet analysis was used to illustrate the transient nature of the weekend effect and its connection to meteorology. Figure 6a shows ozone and temperature at the Beltsville, MD site during the summer of 1999; weekly variation is displayed in Figures 6b and 6c, respectively. Small amplitude and drifting phase indicate that the weekend effect is too small to be detected. For example, while the average weekly forcing (amplitude) at Beltsville was about 22 ppb during the entire period shown, amplitude during the early part of the summer was only about 11 ppb. Large late-summer and small early-summer amplitudes are accompanied by relatively stable (+/- 0.5 days, Figure 6c), and drifting phases, respectively (note that for the roughly six week period spanning 1999.33-1999.45, the phase drifted an entire week). More effective model evaluation with respect to the weekend effect might result if times and places of strong/stable weekly forcing were selected for analysis. Space limitations prevent presentation of other sites that resemble Beltsville with respect to weekly variation.

With respect to meteorological influences on the WEWD effect, Figure 6c shows a multi-week period at Beltsville when weekly variation in ozone and temperature appear to be synchronized (constant weekly phase difference). The average weekly amplitudes are 22.4 and 12.4 ppb for the 'time of stable phase difference' and the rest of the summer season, respectively. Short periods of apparent ozone/temperature synchronization, characterized by strong weekly ozone forcing, were also observed at other sites (not shown) and seems to be a necessary condition for detection of a weekend effect.

Figure 7 explores whether CMAQ can replicate the day-of-the week changes observed in ozone concentrations and its precursors. At 8 co-located monitoring sites for the period 1993-2005, we analyzed estimated emissions of CO and NO_x, and observed and modeled concentrations of CO, NO_x, and O_3. For the early morning (6-9 a.m.), modeled CO concentrations are only about half that observed. Most of the CO monitoring sites are in the vicinity of roadways in urban areas, seeing the impacts of CO emissions in the near-field. The sub-grid scale variability in emissions is not explicitly modeled in CMAQ and modeled CO represents average concentrations for a 12 km x 12 km x 38 m grid cell. Hence, absolute levels of modeled concentrations will be smaller than those observed at individual monitoring stations. However, WEWD differences of observed and modeled CO show good agreement (-24% and -29%, respectively).

The agreement between the observed and modeled values is not as good for early morning concentrations of NO_x. Modeled NO_x concentrations are again about half the observations, and the mean WEWD difference for the model (-27%) is about half of the observed difference (-53%). Whereas the average observed NO_x concentrations are in the range of 30-35 ppb during the weekdays, the corresponding modeled NO_x concentrations are about 12 ppb. In the early morning, modeled ozone concentrations are ~10 ppb higher than the observed, while the observed WEWD difference (+6 ppb) is higher than modeled (+2 ppb). The higher modeled ozone level and reduced modeled WEWD difference is indicative of reduced early morning ozone titration in the model relative to that observed. Despite the trends and differences seen in the precursors and in the early morning values of ozone, mean values of observed and modeled daily maximum 1-hour ozone concentrations agree to within ~1 ppb, showing little evidence of a day-of-week trend in this dataset.

The 8 -- mostly small urban -- sites, which were used above to examine day-of-the-week variations above, show only a small WEWD effect for the daily maximum 1-h ozone. However, examination of all 90 surface ozone monitors across the study domain during 1988-1993 in Figure 8 (panels a and c) indicates that the means in both observed and modeled daily maximum 8-hour ozone values tend to be higher on weekends. While the spatial patterns are similar, the WEWD difference in the model is about 50% of the observed signal (see the regression in panel e). For the more recent analyzed time period (2000-2005), ozone levels on the weekends tend to be *lower* than the weekdays as shown in Figure 8 (panels b, d, and f), and again the WEWD difference in the model is 50% less than the observations as indicated by the regression equation.

In looking at the early morning ozone values in Figures 9 for the early (1988-1993) and later (2000-2005) periods (panels a, c, e and b, d, f, respectively), observations are clearly enhanced on weekends especially near urban areas, suggesting reduced NO titration than during the weekdays. The response of CMAQ is better than for the daily maximum 8-hr ozone and is only about 30% less than the observed response and better than the model response for the early morning time.

That the VOC/NO_x sensitivity associated with the peak ozone concentrations has changed during the 18-year analysis period is evident in the model simulations (Figure 10). In the left-hand panels, areas where the ozone production efficiency (i.e., the O_3/NO_z ratio) vary between 10 and 20 represent a transition from VOC-sensitive regions (low ratios) to NO_x-sensitive regions (high ratios). For the earlier period (1988-1993), nearly 25% of the modeling domain is either neutral or VOC-sensitive, especially near urban areas, but by later years (2001-2005), only the New York City urban area is clearly in the VOC-sensitive regime. Across the neutral/VOC-sensitive areas, the right-hand panel of Figure 10 shows that the ratio of the Sunday-to-Weekday daily maximum ozone decreases in later years compared to earlier years.

Further evidence of change in the chemical regime can be seen in Figure 11 with observed ratios of VOC to NO_x measured at two sites in the greater New York metropolitan area (Bronx, New York, and Rutgers, New Jersey). The average observed early morning VOC to NO_x ratio of 3-4 ppbC/ppb for weekdays is similar to other major U.S. cities (e.g., Pun et al., 2003) and is considered to represent VOC-sensitive conditions. The observations clearly show a shift to more VOC-sensitive conditions on the weekends at these sites, whereas the modeled results show only limited changes between weekdays and weekends. Changes in ratios between the earlier (1999-2001) and later (2003-2005) time periods were small during this time interval.

5. Discussion

Our analysis of the weekend-weekday (WEWD) effect has shown that the magnitudes of the differences in ozone between weekend and weekdays (i.e., the WEWD signal) have decreased during a recent 18 year period, both in observed and modeled concentrations across the northeastern United States. The downward trend in the WEWD effect over this time period is not too surprising, as there have been major decreases in precursor emissions. Our estimates, based on an analysis of the SMOKE-processed model-ready emissions for the 12 km modeling domain, indicate a 49% decrease in anthropogenic VOCs and 44% in NO_x for the 1988-2005 period. When biogenic emissions are included in the analysis, the decrease is 25% for VOC and 43% for NO_x, supporting the notion of increased VOC-to-NO_x ratios over the modeling period as discussed in the previous section. The decline in anthropogenic emissions over 18 years is comparable to the decrease in estimated VOC and NO_x weekend emissions relative to weekdays (~35%). Also, Dallman and Harley (2010) in performing a "top down" analysis of mobile source NO_x emissions using national fuel sales and published emission factors show that emissions from diesel engines have became a much greater fraction of the mobile source inventory and that the current published EPA trends may be understating the contribution of diesels to the overall total by a factor of two. Furthermore, Harley et al. (2005) point out that activity in the diesel sector is much more reduced relative to light-duty automobiles on the weekend, which may lead to shifts towards VOC-sensitivity as NO_x emissions are reduced. Considered together, these factors point to the need to accurately characterize annual and day-of-week changes in mobile NO_x emission for both diesel and gasoline engines.

Interpretation of the WEWD trend is further complicated by the presence of large year-to-year variability in meteorology. Interestingly, Forster and Solomon (2003) detect a “weekend effect” in the diurnal temperature range across many parts of the U.S., but from their analysis it is unclear what impact, if any, that changes in the diurnal temperature range might have on precursor emissions or photochemistry. Because of the high year-to-year variability in the WEWD effect, discerning a response in ozone to changes in emissions and/or meteorology requires an analysis of time periods lasting longer than just an episode or a few episodes in a year as modeled and observed weekly signals are controlled by changes in both meteorology and emissions. Since it is difficult to isolate the relative contributions of meteorology and emissions to the weekday-weekend signal, model simulations of short-term episodes may not reveal a response to the emission changes that longer-term modeling would help detect.

The differences in modeled and observed early morning NO_x and ozone concentrations may be partly attributed to CMAQ’s relatively coarse lower grid mesh (which is approximately 12 km x 12 km wide and 38 m deep) during the early morning, when ozone precursors tend to pool near the earth’s surface. Godowitch et al. (2010) analyzed changes in the observed and modeled NO_x concentrations during the 2002-2006 period and found that while NO_x observations are responding primarily to changes in near-field (mobile source) emissions during this period, modeled NO_x concentrations are reflecting the changes in all emissions in a the grid cell (low-level point, area, on-road, and off-road). Godwitch et al. (2010) further report that changes in weekday early morning NO_x concentrations from 2000-2006 in CMAQ are smaller than that observed. They also found that modeled NO_x levels in the morning are much more coherent in space than observed NO_x levels, suggesting that models tend to overdilute NO_x. The fact that most NO_x monitoring sites are in urban areas and the spatial heterogeneity is not modeled as well with 12 km grids may explain some of the discrepancy between observations and model outputs.

Another area of concern is with lateral boundary conditions. Time-dependent boundary conditions available from global scale models were not available for the 18 year time period simulated in this study. However, as noted by researchers like Tang et al. (2007), boundary conditions can impact ozone simulations in a regional scale model and can influence the temporal variations in model performance. Although the 12-km grid mesh used boundary conditions from the 36-km grid simulation (which included temporal variability in emissions), it is possible that the boundary conditions used for 36-km simulation affected modeled response inside the 12-km mesh.

While there is general acceptance that most of the U.S. is becoming more NO_x sensitive on a regional scale as large reductions in anthropogenic emissions of NO_x occur (e.g., Duncan et al., 2010), several studies have reported that reductions of NO_x on weekends relative to VOCs in urban areas that reportedly remain VOC-sensitive can lead to ozone enhancement as supported by the process-oriented studies of Tonse et al. (2008). Areas of VOC sensitivity have been identified in California (Blanchard and Fairley, 2001; and, Marr and Harley, 2002a), Cincinnati, Ohio (Torres-Jardon and Keener, 2006), Chicago, and possibly Philadelphia (Pun et al., 2003). As indicated in our modeling results, the spatial gradients of chemical sensitivity appear quite steep, and the interpretation of the trends in the WEWD effect may be confounded as chemical regimes in urban areas continue to evolve with changes in VOC and NO_x emissions.

6. Summary

This dynamic evaluation study reveals that the CMAQ modeling system has successfully captured the weekend-weekday (WEWD) signal by demonstrating an overall enhancement of ozone on weekends compared to weekdays during the early period of the 18-year simulation and a reduction in the enhancement during the 2001-2005 period. However, CMAQ's ability to capture the daily maximum 8-hr ozone WEWD amplitudes was only ~50% of that observed. We conclude from this dynamic evaluation that there are at least three areas that may yield improvements to the modeling system: (1) revising temporal estimates of mobile source NO_x emissions, especially from diesel engines, (2) decreasing the grid size (both horizontally and vertically) of layer 1 in the CMAQ modeling system, and (3) refining temporal estimates of boundary conditions for ozone and ozone precursors.

Preliminary results using the MOtor Vehicle Emissions Simulator MOVES model, which is scheduled to be used for air quality model applications later in 2010, suggest that current estimates of heavy-duty diesel emissions of NO_x will increase by as much as 50% in some urban areas (www.epa.gov/otaq/models/moves/). We recommend that sensitivity tests with alternative mobile NO_x emission estimates be performed when MOVES becomes available for multi-annual historical simulations like that performed in this paper. In addition, we encourage further work by the modeling community to develop and test finer grid structures within the CMAQ modeling system to help alleviate the likely overdilution of early morning NO_x emissions and undertitration of early morning ozone concentrations. Finally, as the availability of simulations from global models extends to the 1988-2005 time period, we recommend that the impact of temporal variations on boundary conditions be examined in the context of model response.

Because the magnitude and even the direction of ozone response to emission differences may be strongly influenced by meteorology as well as changes in the chemical regime, modeling strategies with just a few WEWD episodes in a year are unlikely to capture the WEWD signal seen over longer time periods. Long-term analysis of chemical data (that includes ozone precursors) and modeling with perturbations in ground-level emissions is needed to properly separate out the meteorologically-induced from emissions-induced signals. Because of the complex interplay between meteorology and emissions, identifying opportunities for emissions-induced signals for the dynamic evaluation of regional air quality models remains a challenge.

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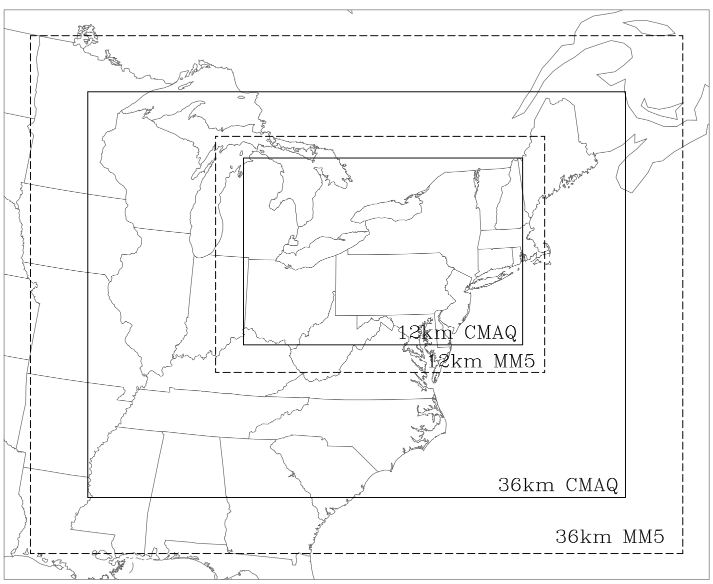
Figure 1

(a)


12km CMAQ
12km MM5
36km CMAQ
36km MM5

(b)

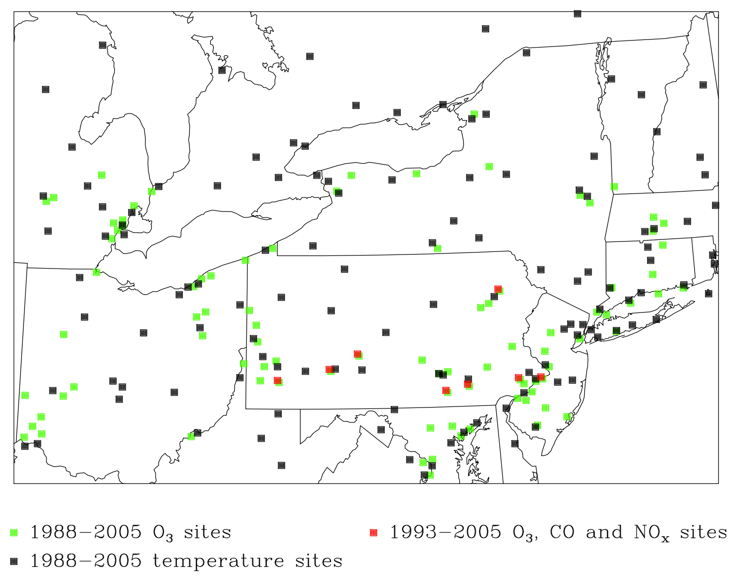
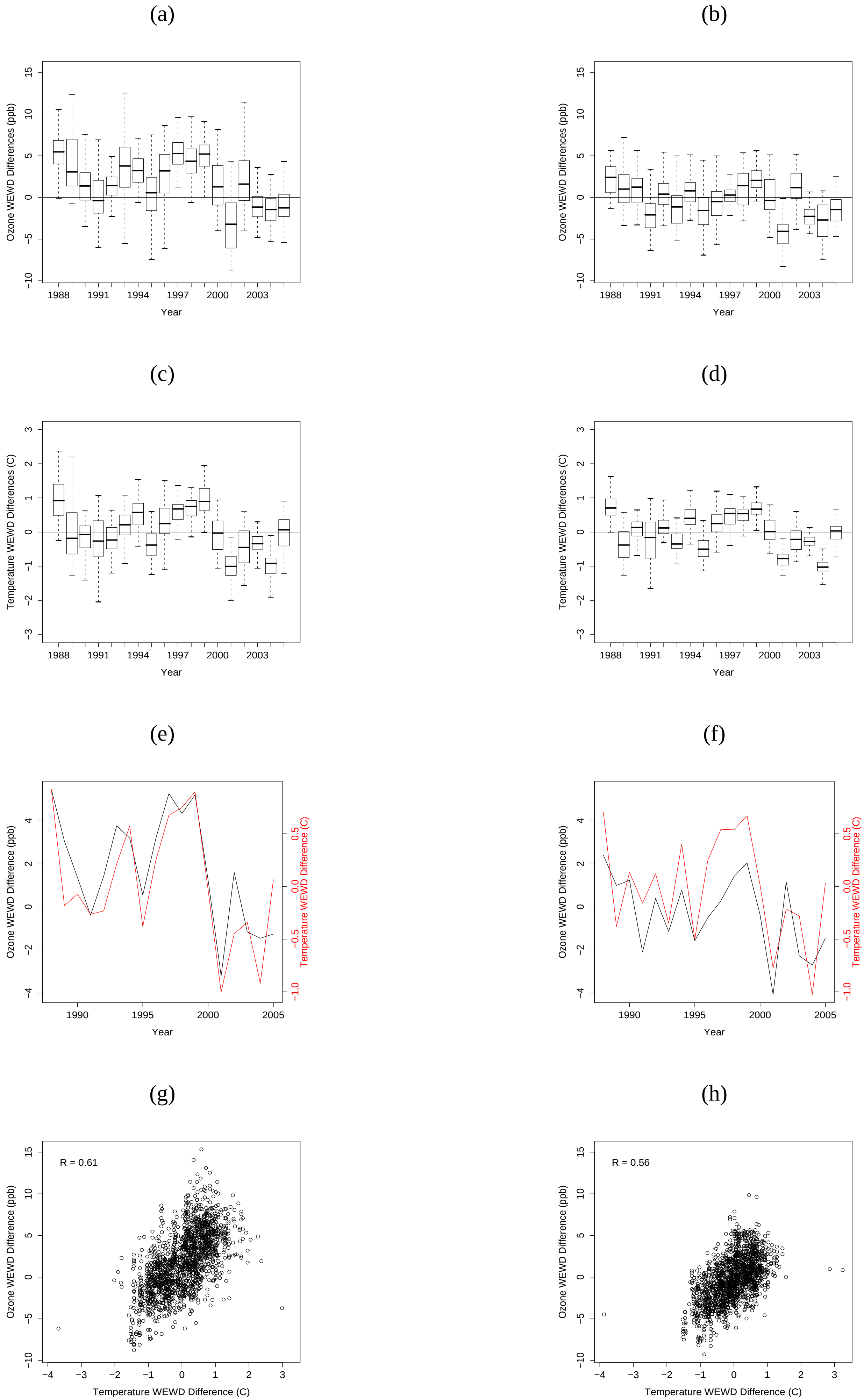
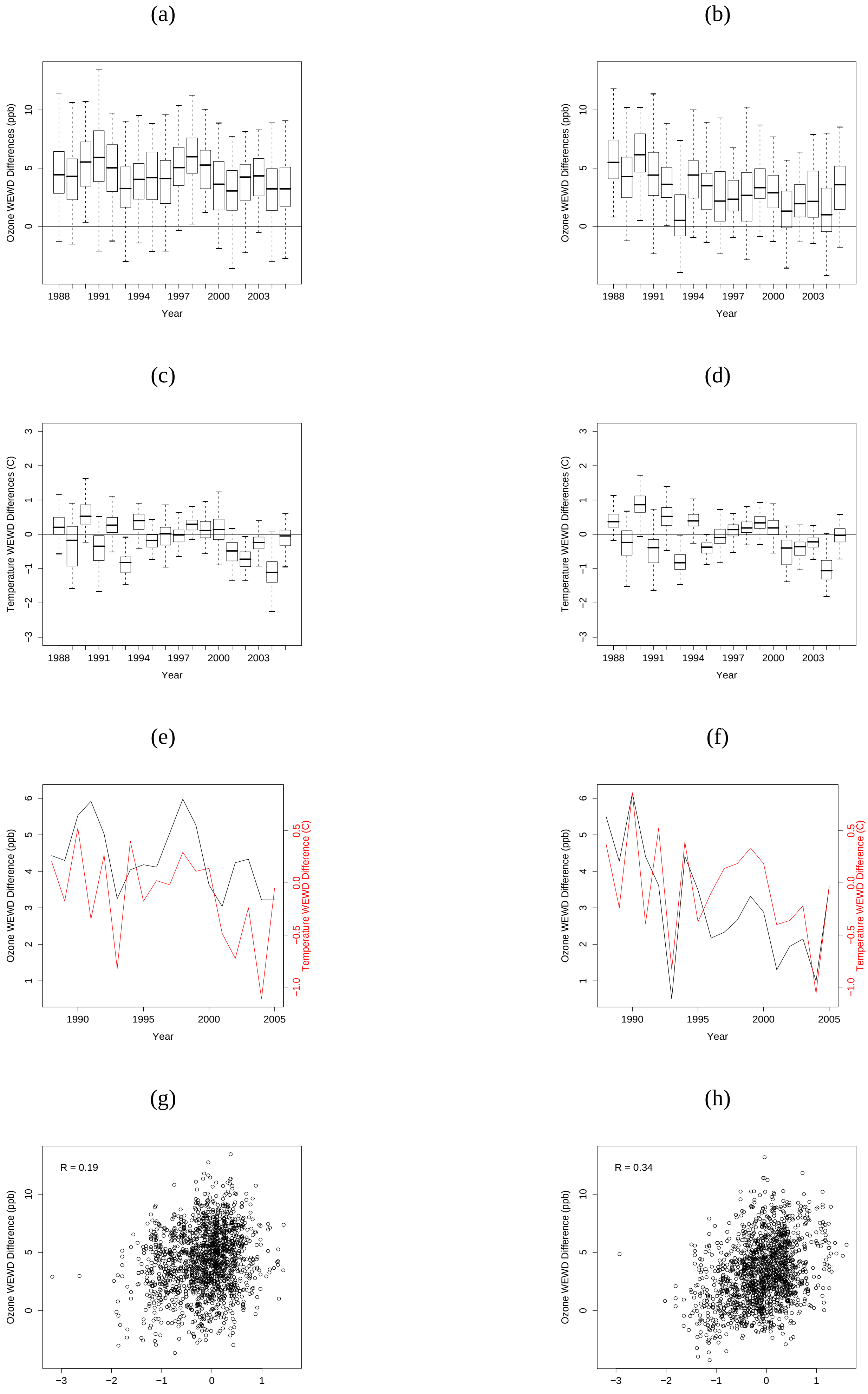

1988–2005 O3 sites
1993–2005 O3, CO and NOx sites
1988–2005 temperature sites

Figure 2



(a)
(b)
Ozone WEWD Differences (ppb)
Year
1988
1991
1994
1997
2000
2003
(c)
(d)
Temperature WEWD Differences (C)
Year
(e)
(f)
Ozone WEWD Difference (ppb)
Temperature WEWD Difference (C)
Year
1990
1995
2000
2005
(g)
(h)
R = 0.61
R = 0.56
Ozone WEWD Difference (ppb)
Temperature WEWD Difference (C)

Figure 3



(a)
(b)
Ozone WEWD Differences (ppb)
Year
1988
1991
1994
1997
2000
2003
0
5
10
(c)
(d)
Temperature WEWD Differences (C)
−3
−2
−1
0
1
2
3
(e)
(f)
Ozone WEWD Difference (ppb)
Temperature WEWD Difference (C)
1990
1995
2000
2005
1
2
3
4
5
6
−1.0
−0.5
0.0
0.5
(g)
(h)
R = 0.19
R = 0.34
Temperature WEWD Difference (C)
−3
−2
−1
0
1

Figure 4

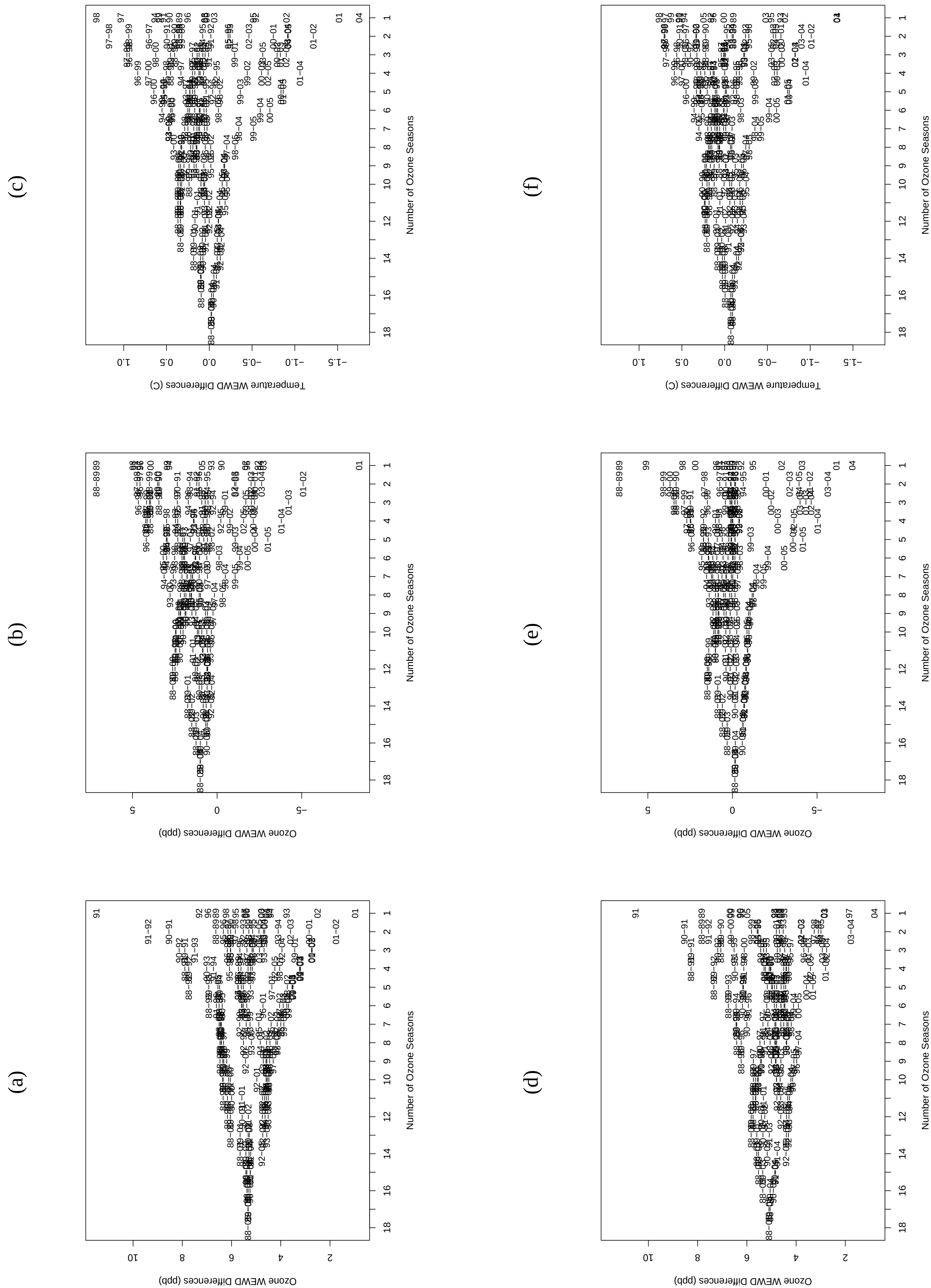
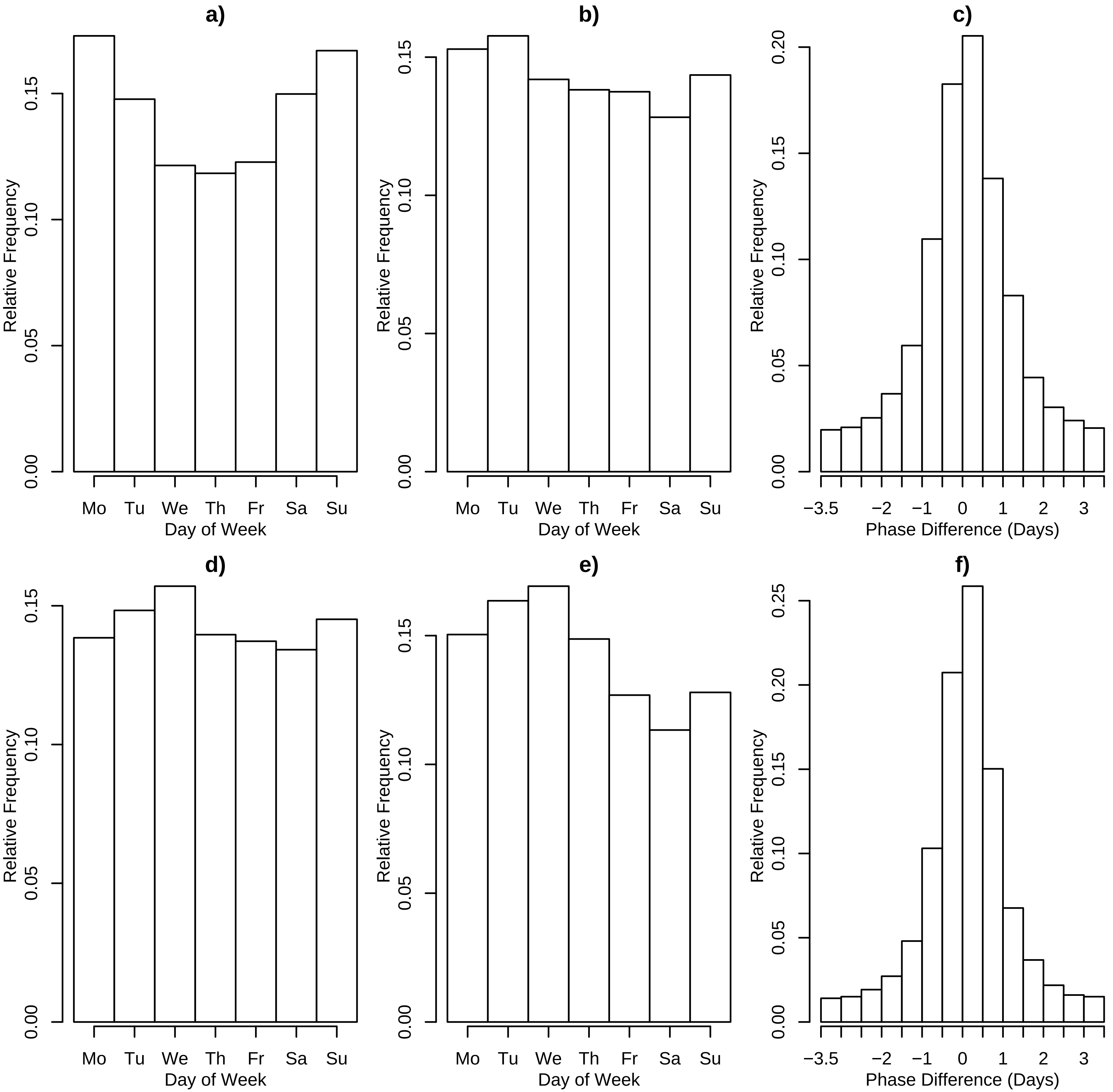

(a)
(b)
(c)
(d)
(e)
(f)
Number of Ozone Seasons
Ozone WEWD Differences (ppb)
Temperature WEWD Differences (C)

Figure 5



a)
b)
c)
d)
e)
f)
Relative Frequency
Day of Week
Mo
Tu
We
Th
Fr
Sa
Su
Phase Difference (Days)
0.00
0.05
0.10
0.15
0.20
0.25
−3.5
−2
−1
0
1
2
3

Figure 6

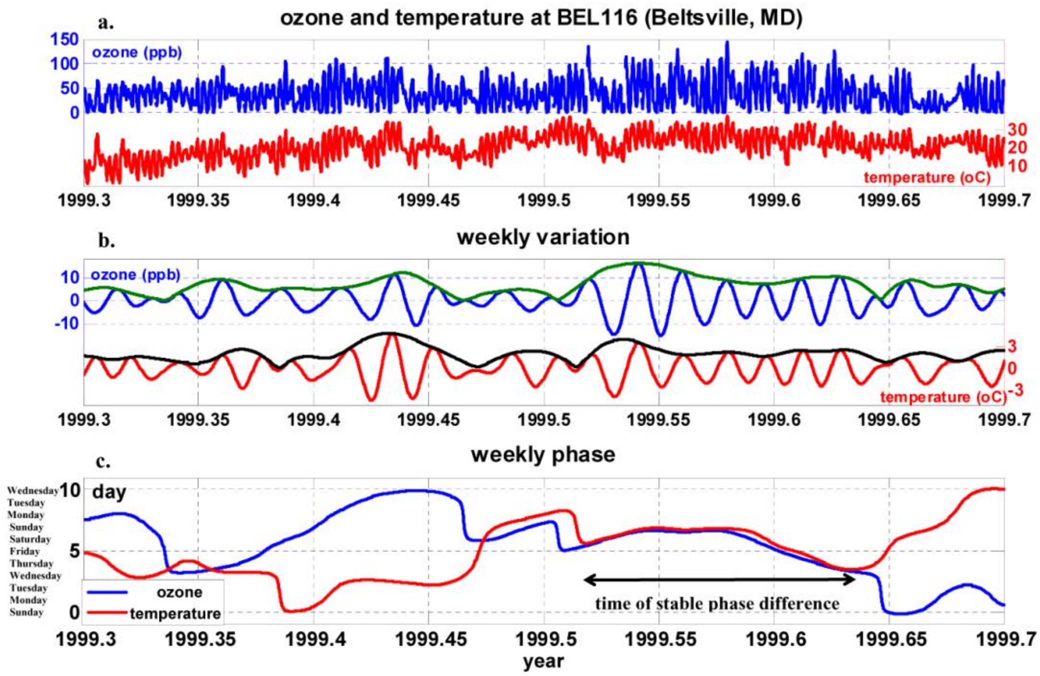

a.
ozone and temperature at BEL116 (Beltsville, MD)
ozone (ppb)
150
100
50
0
30
20
10
temperature (oC)
1999.3
1999.35
1999.4
1999.45
1999.5
1999.55
1999.6
1999.65
1999.7
b.
weekly variation
ozone (ppb)
10
0
-10
3
0
-3
temperature (oC)
c.
weekly phase
day
Wednesday
Tuesday
Monday
Sunday
Saturday
Friday
Thursday
Wednesday
Tuesday
Monday
Sunday
10
5
0
ozone
temperature
time of stable phase difference
year

Figure 7

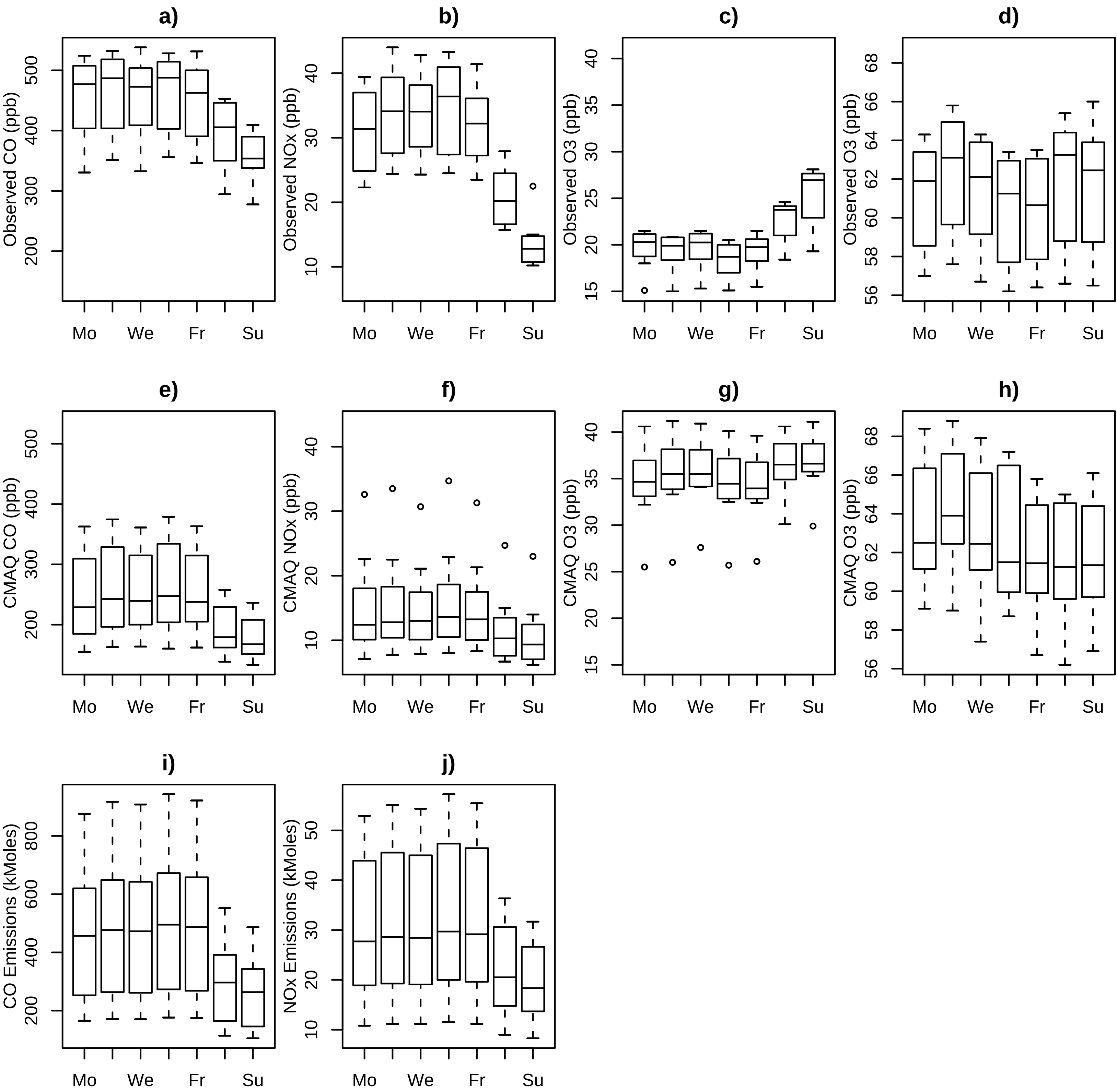

a)
b)
c)
d)
e)
f)
g)
h)
i)
j)
Observed CO (ppb)
Observed NOx (ppb)
Observed O3 (ppb)
CMAQ CO (ppb)
CMAQ NOx (ppb)
CMAQ O3 (ppb)
CO Emissions (kMoles)
NOx Emissions (kMoles)
Mo
We
Fr
Su

Figure 8

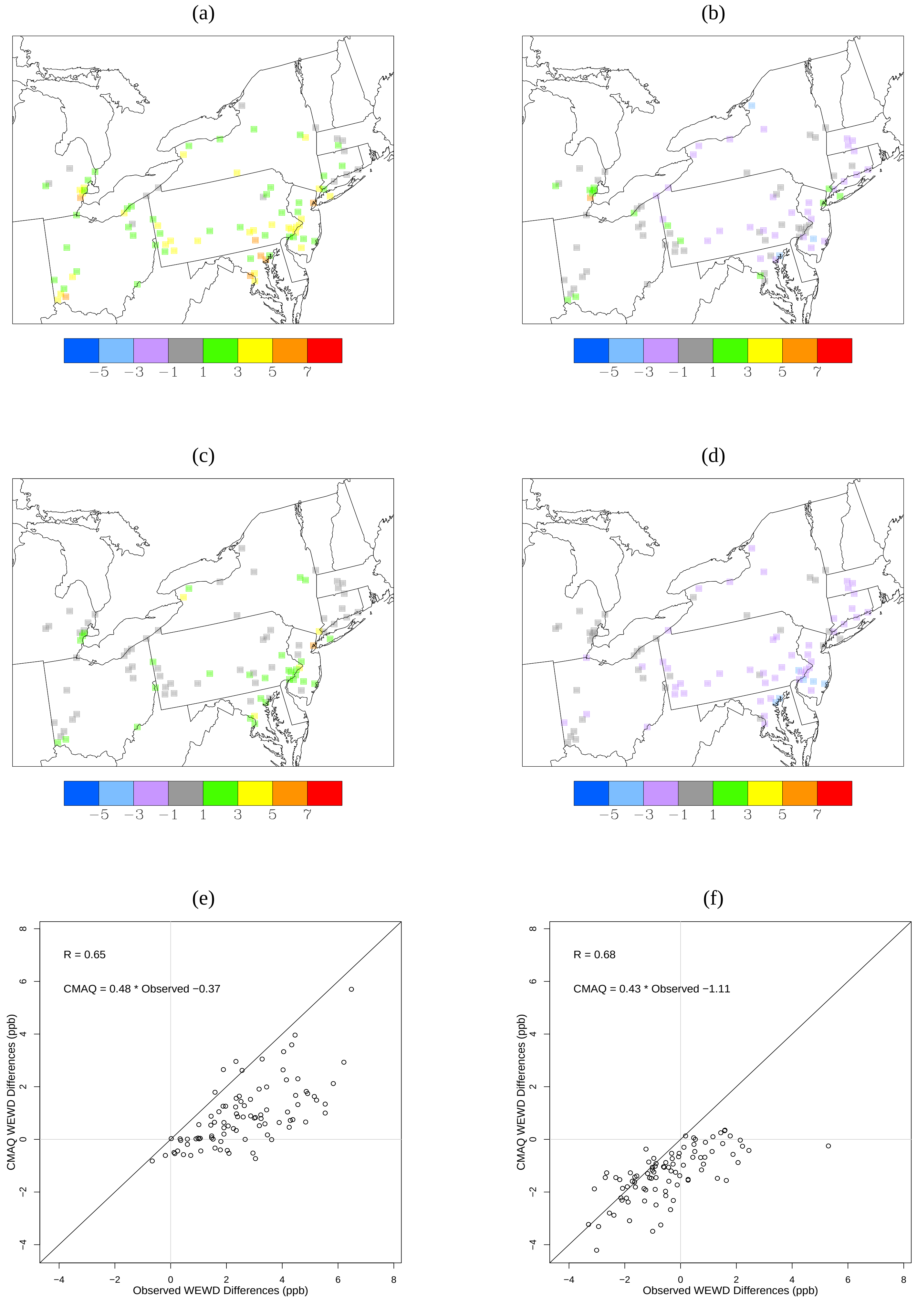
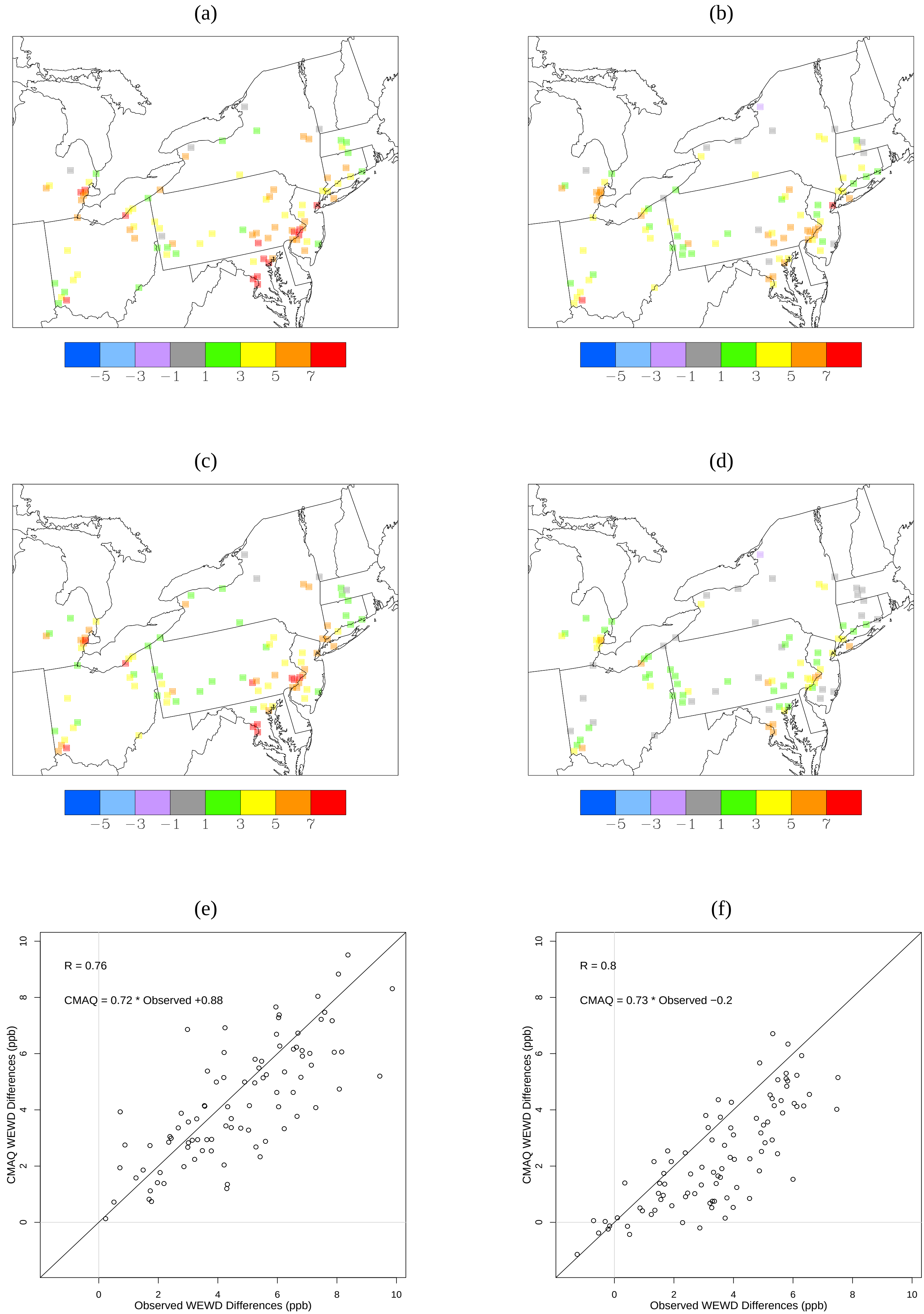

(a)
(b)
(c)
(d)
(e)
(f)
−5 −3 −1 1 3 5 7
R = 0.65
CMAQ = 0.48 * Observed −0.37
R = 0.68
CMAQ = 0.43 * Observed −1.11
CMAQ WEWD Differences (ppb)
Observed WEWD Differences (ppb)

Figure 9



(a)
(b)
−5 −3 −1 1 3 5 7
(c)
(d)
−5 −3 −1 1 3 5 7
(e)
R = 0.76
CMAQ = 0.72 * Observed +0.88
CMAQ WEWD Differences (ppb)
Observed WEWD Differences (ppb)
(f)
R = 0.8
CMAQ = 0.73 * Observed −0.2
CMAQ WEWD Differences (ppb)
Observed WEWD Differences (ppb)

Figure 10

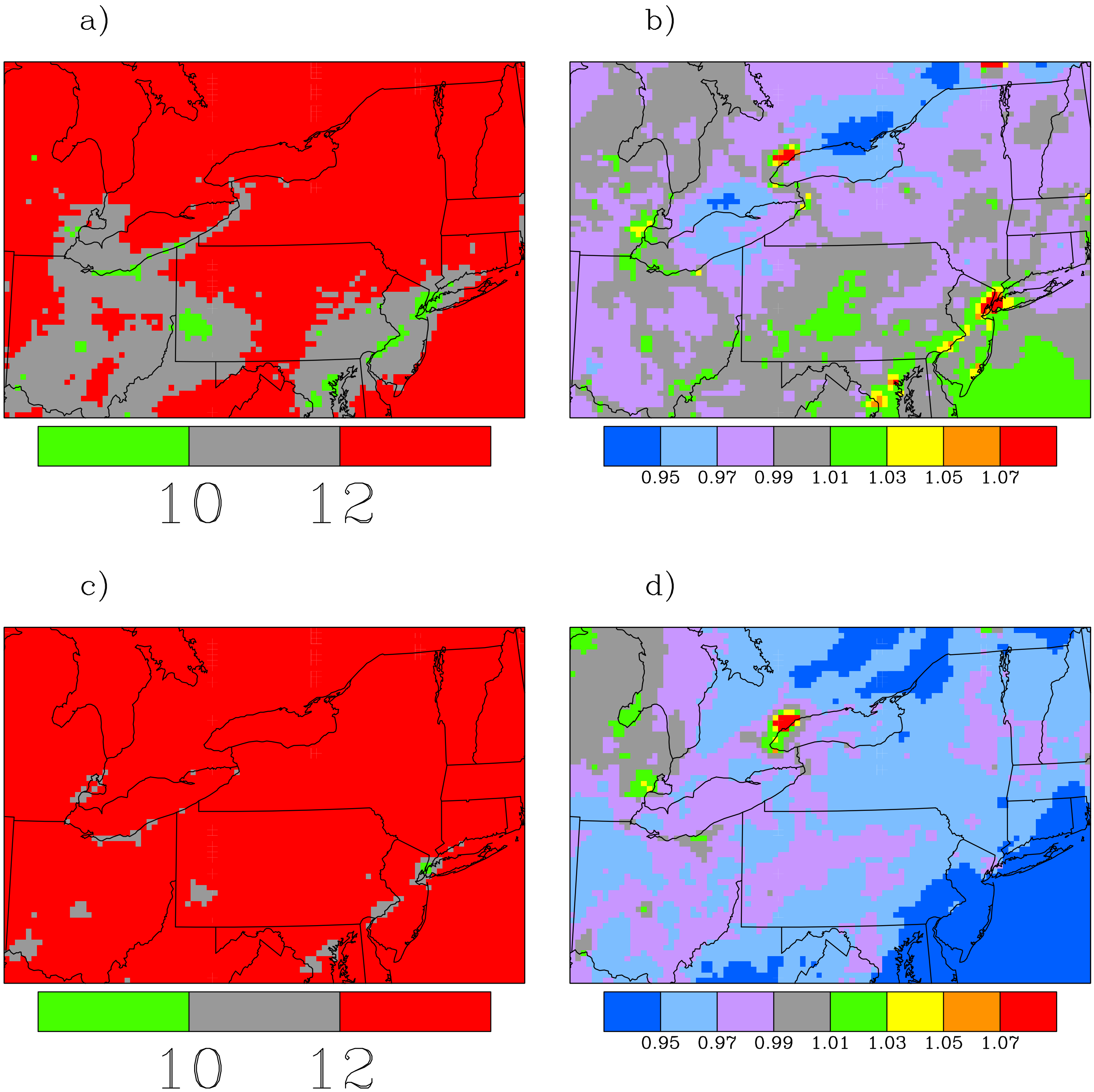


Figure 11a

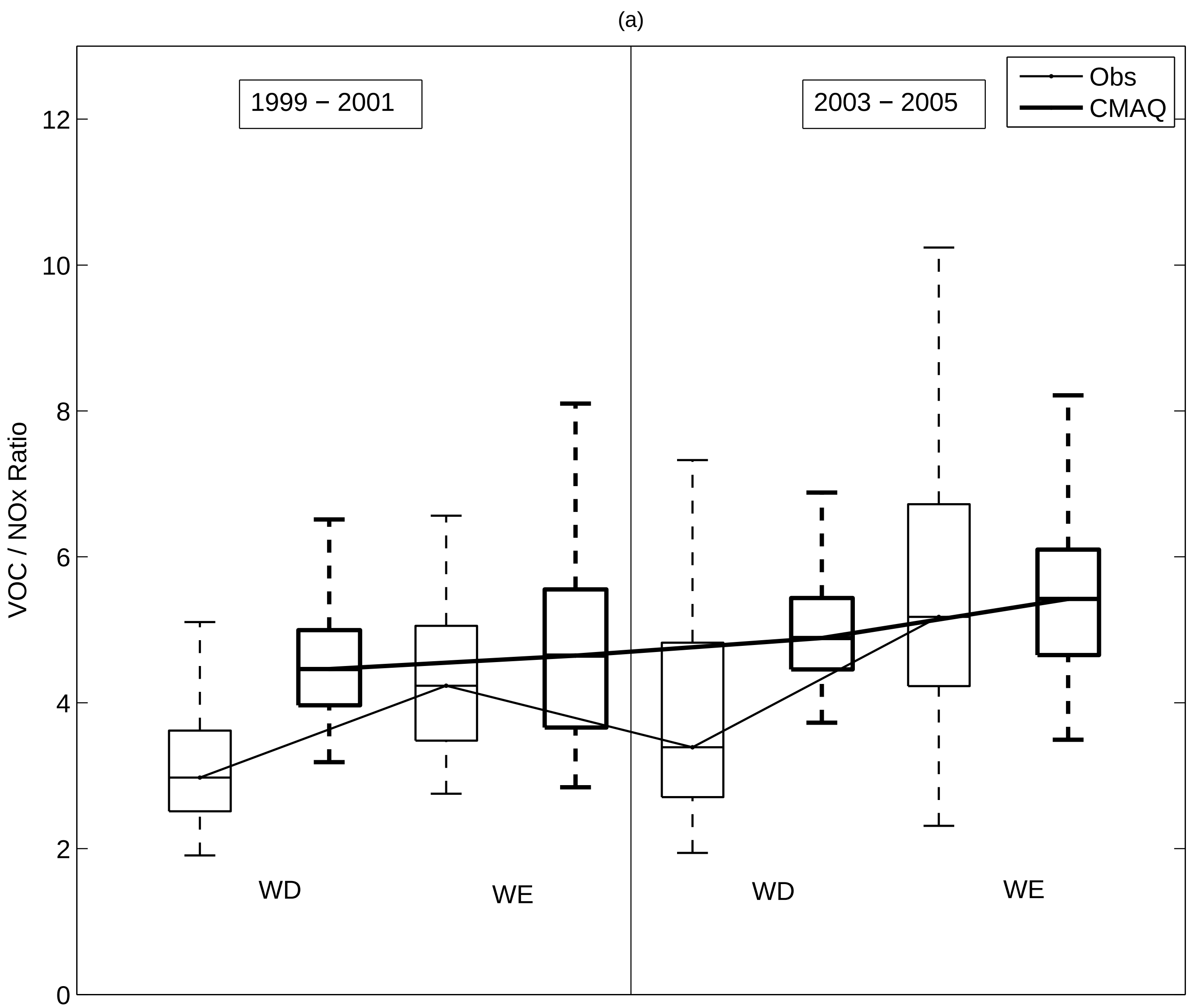
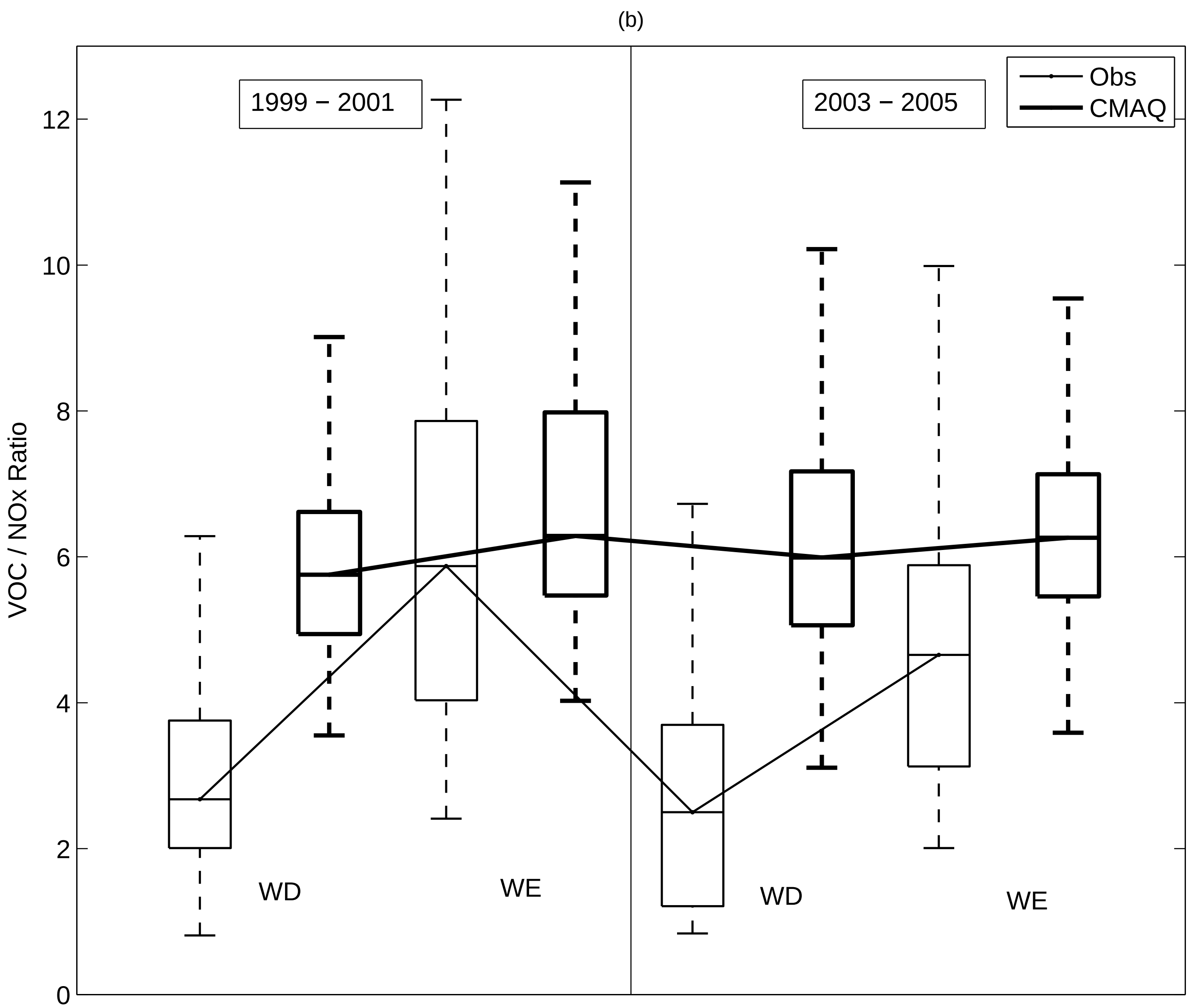

(a)
1999 − 2001
2003 − 2005
Obs
CMAQ
VOC / NOx Ratio
0
2
4
6
8
10
12
WD
WE
WD
WE

Figure 11b



(b)
1999 − 2001
2003 − 2005
Obs
CMAQ
VOC / NOx Ratio
0
2
4
6
8
10
12
WD
WE
WD
WE

Figure Captions