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Assessing Multi-year Changes in Modeled and Observed Urban NO_X Concentrations from a Dynamic Model Evaluation Perspective

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Abstract

An investigation of the concentrations of nitrogen oxides (NO_X) from an air quality model and observations at monitoring sites was performed to assess the changes in NO_X levels attributable to changes in mobile emissions. This evaluation effort focused on weekday morning rush hours since urban NO_X concentrations are strongly influenced by the significant loading of emissions associated with heavy commuter traffic. On-road vehicle NO_X emissions generated by the MOBILE6 model revealed a steady decline with an overall decrease of 25% for 2002-2006. In this study, a dynamic model evaluation was undertaken that entails an assessment of the predicted concentration response of the Community Multiscale Air Quality (CMAQ) model due to changes in NO_X emissions as well as to meteorological variability spanning 3-month summer periods over five consecutive years (2002-2006) against observed concentration changes at NO_X monitoring sites located primarily in urban areas of the eastern United States. Both modeled and observed hourly NO_X concentrations exhibited maximum values that coincided with the morning peak NO_X emissions. The notable results, based on 3-hour average (6-9 AM local time) NO_X concentrations, derived between the 50th and 95th percentiles of cumulative concentration distributions, revealed that modeled changes at these elevated NO_X levels generally tracked the year-to-year variations in the observed concentration changes. When summer 2002 values were used as a reference, both

modeled and observed results also showed definitive decreases in weekday morning urban NO_X concentrations over this multi-year period, which can be primarily attributed to the reductions in mobile source emissions. Whereas observed NO_X concentrations have declined by about 25% over this period consistent with the decline in the modeled mobile emission sector, modeled NO_X concentration changes were close to the decreases exhibited in all (mobile+other sectors) surface NO_X emissions whose overall decline was about 15% over this multi-year period.

Keywords: dynamic model evaluation, weekday NO_X changes, mobile NO_X emissions, NO_X concentrations, air quality modeling

1. Introduction

Nitrogen oxides (NO_X = NO+NO_2) are key precursor species that are intricately involved in the photochemical production of tropospheric ozone and fine particulate matter. Consequently, numerous control programs have been implemented to reduce anthropogenic NO_X emissions from different source categories. The primary contributors to NO_X emissions are the electrical utility sector, industrial point sources, on-road mobile and nonroad sources, and area sources. While a reduction of about 40% has occurred in NO_X emissions from the electric utility sector (major point source emissions category) during 2003-04 due to the implementation of the US Environmental Protection Agency's (EPA) NO_X State Implementation Plan (SIP) Call program (USEPA, 2005), changes in NO_X emissions for the on-road mobile source sector appear to be much more gradual and results have even differed on the trend especially during the 1990's (Parrish, 2006). In fact, the multi-year results covering the1990's in Sistla et al. (2002) showed no trend in the annual average morning NO_X concentrations at sites in the New York City metropolitan area, which they attributed to increases in vehicle miles

traveled even though vehicle emissions standards were tightened. In contrast to the utility sector, a decline in mobile NO_X emissions was estimated to be close to 5% per year during 2002-04 (USEPA, 2005). The implementation of the EPA's Tier 2 program commenced with the 2004 model year for new vehicles and other mobile emissions control programs, which were initiated in recent years, have further reduced mobile NO_X emissions (USEPA, 2007).

Although the on-road mobile sector contributed about 36% to the total NO_X emissions budget in 2004 (USEPA, 2005), on the spatial scale of a highly populated urban area it is often the dominant emission source of ground-level NO_X. During a typical weekday morning high traffic period, elevated NO_X concentrations in urban areas are strongly influenced by the rapid rise in mobile emissions associated with heavy commuter traffic. This large release of NO_X and other primary species initiated in the early morning period represents the leading edge of the urban pollutant plume. During the course of the daytime period, it is transported by wind, horizontally and vertically dispersed, and chemically-transformed into secondary oxidant species, leading to elevated levels of ozone and other secondary nitrogen species in downwind areas. Therefore, it is important from an accountability standpoint to ascertain whether emission reductions will lead to concomitant improvements in air quality. In addition, since air quality models are being used in a regulatory setting to determine the impact of various emission reduction strategies on ambient concentration levels, it is critical that a model's response to emission changes are compared to observed changes as part of a model performance evaluation (Dennis et al., 2010).

This dynamic model evaluation study was undertaken to determine the amount of change in NO_X concentrations that could be related to the year-to-year declines in estimated mobile NO_X emissions. Observations from monitoring sites and modeled NO_X concentrations from the Community Multiscale Air Quality (CMAQ) model have been

analyzed for the 0600-0900 AM time period on all weekdays over 3 summer months in consecutive years spanning 2002 to 2006. This dynamic model evaluation approach, an emerging technique, which is one component of a comprehensive model evaluation effort, involves assessing modeled and observed concentration changes in relative (i.e. percentage) and absolute terms (Gilliland et al., 2008). Thus, it is complementary to the traditional operational model evaluation approach, which deals with the computation of statistical metrics from data sets of modeled and observed concentrations pairs to assess a model's ability to reproduce past observations in an overall sense. Recent operational model evaluation results have been reported on the performance of CMAQ to predict ozone (Appel et al., 2007) and various gaseous pollutant species, including NO and NO_2 (Yu et al., 2006). Another alternative evaluation approach based on a high-resolution filter technique has been applied to compare the contributions to observed and modeled NO_X concentrations (Biswas et al., 2001) and various species (Hogrefe et al., 2006) from processes on time scales ranging from intra-day, diurnal, synoptic, and longer term over an extended modeling period of a single summer and a full year, respectively.

The previous application of the dynamic model evaluation approach (Gilliland et al., 2008) focused on assessing the impact of the NO_X SIP Call point source emission reductions on the daily maximum 8-hour ozone concentrations. Their modeling effort took advantage of the hourly NO_X emissions measurements made routinely at major point sources. In this dynamic evaluation effort, the results are intended to provide information to help reconcile whether multi-year changes in estimated mobile NO_X emissions, which strongly modulate modeled concentrations in CMAQ, coincide with NO_X concentration changes derived from real-world observations. This paper discusses the changes in modeled and observed weekday morning NO_X concentrations as well as in NO_X emissions covering this recent multi-year period.

2. Study Description

2.1 Modeling Details and Inputs

The same version (CMAQ v4.7) of the chemical transport model (CTM) was applied for the simulations of all summer periods (June, July, August) during the years of 2002 through 2006. Key components of the CMAQ/CTM employed in these applications included the CB-05 (Carbon Bond 2005) photochemical mechanism, asymmetric convective mixing scheme (ACM2) for vertical dispersion, and the piece-wise parabolic method (PPM) for the horizontal advection process (Byun and Schere, 2006). Modeling was performed on a large regional domain (279 x 240 grid cells) that encompassed the eastern two-thirds of the US and southeastern Canada with a 12-km horizontal grid cell size. The vertical structure contained 24 layers with the thickness of the first layer being near 38 m. Model results from a continental domain with a 36-km grid cells provided the lateral boundary concentrations for this 12-km modeling domain. Since this modeling effort was also part of a broader study involving annual simulations, initial concentrations for the first day (June 1) of each period were provided by the results from the end of May of each year.

Meteorological fields were generated by the Penn State/NCAR fifth-generation mesoscale model (MM5; Grell et al., 1994) for each summer period. The MM5v3.6.3 model was applied in a non-hydrostatic mode and a four-dimensional data assimilation technique was also used to incorporate available observed winds, temperatures, and moisture data to provide more accurate meteorological parameter fields for air quality modeling (Otte, 2008). More information about the specific physics options applied in the MM5 simulations are described in Appel et al. (2009). In addition, the CMAQ Meteorology-Chemistry Interface Processor (MCIPv3.4) program was exercised to reformat the MM5 output into compatible input data sets containing the hourly 2-D and 3-D meteorological parameter fields for the CMAQ model simulations.

The hourly gridded emission data sets were generated by the Sparse Matrix Operator Kernel Emissions (SMOKEv2.2) processing system. Anthropogenic emissions were extracted from the U.S. EPA's 2002 NEI (National Emissions Inventory) to generate gridded surface and minor point source emissions, while the hourly pollutant emissions for elevated major point sources were specified from Continuous Emissions Monitoring System (CEMS) data sets for each year. Natural surface emissions of NO_X, isoprene, and other biogenic VOC species were computed by the Biogenic Emissions Inventory System (BEISv3.14) model for each period.

Gridded on-road vehicle emissions were generated by the MOBILE6.2 model (USEPA, 2004), hereafter referred to as MOBILE6, for each modeling period. Cook et al. (2006) provides a description of several key inputs required to exercise MOBILE6. For this study, county-specific control program information from each state was taken into account in the MOBILE6 modeling. A typical example of the hourly-averaged weekday mobile NO_X emissions during each summer period is displayed in Figure 1 from the grid cell encompassing Washington, DC. It is evident that mobile emissions exhibited a steady decrease from summer-to-summer, a common feature exhibited in all other urban grid cells. The temporal evolution of mobile NO_X emissions in Figure 1 reveals the characteristically rapid ramp-up that occurs as the morning time rush-hour period progresses with a maximum emission rate occurring between 0700-0800 AM. A slight decrease in emissions is evident afterward, which is followed by gradually higher emission rates continuing throughout the middle of the day.

It must be noted that the temporal pattern in Figure 1 represents a composite of mobile emissions from several road types contained within the grid cell since a unique temporal profile of traffic is assigned by the MOBILE6 model to an individual road type. Therefore, the temporal evolution of mobile NO_X emissions can be expected to differ somewhat among cities depending on the mix of roadway types. On the other hand, an

examination of hourly emission values on different days revealed little variation from weekday-to-weekday in the magnitudes of morning mobile NO_X emissions that is consistent with the results of Pierce et al. (2010). Thus, a particular urban grid cell received essentially the same hourly mobile emission rates on each weekday of a modeling period. Nevertheless, large differences in mobile NO_X emission rates existed among the various urban grid cells due primarily to variations in road density, differences in vehicle miles traveled (VMT) and to on-going local emission control and inspection programs that were considered in the MOBILE6 modeling. In addition, from a comparison of mobile and all NO_X emissions in Figure 1, it is evident that 2002 mobile emissions account for about 70% of total ground-level NO_X emissions during 2002 in this urban grid cell, which was found to be typical for many urban areas. Finally, non-mobile (i.e. area, low-level minor point sources, and nonroad sources) NO_X emissions from summer 2002, also displayed in Figure 1, are clearly greater than the on-road mobile emissions during nocturnal hours prior to the morning commuter rush period. These non-mobile emissions from the 2002 NEI were not projected to subsequent years, so they remained essentially constant over these periods.

2.2 Observations

Hourly NO_X measurements collected at various monitoring sites in the eastern US were analyzed in this study. NO_X monitoring sites are located in various land-use environments ranging from downtown urban areas to suburban and rural locales. The NO_X concentration data were obtained from the U.S. EPA's Air Quality System (AQS) on-line data base (http://www.epa.gov/ttn/airs/airsaqs/detaildata/). The NO_X measurements at most sites were made by chemiluminescence instruments described in McClenny et al. (2002). Although they are prone to interference from secondary

nitrogen species (Fehsenfeld et al., 1987), especially during the photochemically-active afternoon period as reported by Dunlea et al. (2007), the impact on the NO_X measurements during the early morning time period is believed to be small. This was reinforced from comparative analysis of NO_X and total nitrogen species (NO_Y= NO_X + secondary nitrogen species (NO_Z)) concentrations from the model. Results revealed NO_X to be more than 90% of NO_Y during the weekday morning period at major urban locations included in this study, which was not unexpected owing to the high NO_X mobile emissions.

The hourly NO_X observations were paired with hourly-averaged modeled values from the grid cell where each monitoring site was located. In addition, modeled and observed 3-hour average NO_X concentrations were computed for each day from the hourly values starting at 0600, 0700, and 0800 AM local time, encompassing the typical 3-hour commuter traffic period. Our analysis focused on modeled and observed results from a set of 42 urban measurement sites out of more than 180 monitoring sites because they were situated in grid cells exhibiting "urban" as the dominant land-use type. Additionally, mobile emissions in each grid cell containing these sites contributed a majority (> 60-70%) of all surface NO_X emissions. It is also recognized that these urban NO_X monitoring locations do not represent near-roadway "hot spot" sites, which during special field studies (Baldauf, et al., 2008), can be situated within a few meters to tens of meters from a major highway. The monitoring sites are found in large urban areas in the following states with the number in parentheses signifying the number of sites in each state; AR(1), CT(1), DE(1), GA(2), KS(1), LA(1), MD(1),MA(6), MI(2), NJ(3), NY(7), NC(2), OK(1), PA(8), RI(2), and Washington, DC(3).

3. Results and Discussion

The temporal behavior of hourly NO_X concentrations in Figure 2 during two different summer periods, derived from all weekday values at the urban locations, reveals that elevated modeled and observed NO_X concentrations occurred during the 6-9 AM period demonstrating that concentrations were highly correlated with the early morning peak mobile NO_X emissions signal shown in Figure 1. Median values are displayed in Figure 2 along with values at the 10th and 90th percentiles to give an indication of the spread in modeled and observed values during the 2002-2006 summer periods. The characteristic decrease in NO_X concentrations starting in the mid-morning period is attributable to a combination of meteorological processes (i.e., greater vertical mixing and stronger horizontal transport) associated with the growth of the convective mixing layer and increasing photochemical activity, which converts NO_X into secondary nitrogen species. Additionally, large elevated point source plumes are generally trapped in the residual layer aloft during the early morning period minimizing the impact of NO_X emissions from elevated major point sources on near-surface concentrations (Zhang and Rao, 1999; Parrish, 2006). These are reasons why our analyses focused on the high NO_X concentrations during the 0600 to 0900 AM period rather than later hours of the day. It is also noteworthy that the temporal pattern of the modeled concentrations tracks the observed results in Figure 2 revealing that CMAQ is closely replicating the dynamic and chemical processes governing concentrations during this time period. Furthermore, these results also provide initial evidence of noticeable decreases in hourly median values as well as at higher concentration levels (e.g., 90th percentile in Figure 2) between 2002 and 2006, which is further explored from weekday morning 3-hour average values from each summer period in the following sections.

3.1 Change in NO_X Emissions

Since morning NO_X concentrations are strongly related to emission levels, it is relevant to examine any changes in mobile versus all (mobile + other source sectors) NO_X emissions from these summer periods. Morning 3-hour NO_X emission totals were determined by summing the emissions over the 6 to 9 AM period on all weekdays and computing the weekday average 3-hour emission totals in the grid cells containing the 42 urban monitoring locations. By defining summer 2002 (i.e., 02) results as reference values, relative changes at each site were determined by $(E_{YY} - E_{02}) \bullet 100 / E_{02}$ for each subsequent summer year (YY = 03 to 06). Results from all urban locations in Figure 3 show a rather steady decline from year-to-year in mobile NO_X emissions over these summer periods, which is primarily attributable to a continuous drop in the fleet emission factor associated with vehicle turnover. A decline of 25% in mobile NO_X emissions is evident between 2002 and 2006 in Figure 3. In these box/whisker plot results, the length of the boxes is indicative of the variability in the mobile NO_X emissions change among the urban sites, which was anticipated due to differences in county-specific controls and vehicle inspection/maintenance programs among these urban locations.

Figure 3 also depicts results for all surface (mobile+other sectors) NO_X emissions found in the first model layer from the same urban grid cells. These results also depict a gradual decline in all emissions over this multi-year period. However, the changes in all NO_X emissions are somewhat less than those found in the mobile sector. Specifically, the overall decrease revealed is closer to 15% between 2002 and 2006. Nevertheless, it was apparent that the modeled change in mobile NO_X emissions is the primary contributor to the overall change in all NO_X emissions for the morning period. Since the estimated emissions from the non-mobile sectors remained nearly unchanged over this period, as noted earlier, the effect of the on-road mobile emissions reduction was somewhat diminished over this period. Hence, the fractional contribution of the on-road

mobile sector to the total NO_X emissions budget became incrementally smaller over this multi-year period.

3.2 Changes in Weekday Morning NO_X Concentrations

The 3-hour morning high traffic period on weekdays is of particular interest since relatively high NO_X concentrations are strongly related to the mobile emissions. Nevertheless, it is recognized that numerous low-level minor point sources, area sources, and the nonroad sector emissions existing in the 12-km grid cells also impacted modeled concentrations, which somewhat complicates the interpretation of modeled concentration changes attributable solely to mobile emission changes.

Observed and modeled weekday 3-hour average concentrations computed from the upper portion (50^{th} to 95^{th} percentiles) of cumulative frequency distributions (CFD) for each urban location were analyzed to assess the magnitude of changes over this concentration interval for NO_X. Relative changes were computed with the summer 2002 value (C_{02}) at each urban site serving as a reference according to $\Delta C / C_{02}$ where $\Delta C = C_{YY} - C_{02}$. C_{YY} values were the NO_X concentrations at the urban sites from summer periods in 2003 through 2006 (03-06). Therefore, a negative change is indicative of a lower concentration and signals a decrease from summer 2002 levels.

Figure 4 contains the results based on changes in the 3-hour average observed and modeled NO_X concentrations determined from the upper part of the CFD's over all urban sites. Considerable agreement in the observed and modeled concentration changes exists as demonstrated by the overlap of the boxes spanning the inter-quartiles (25^{th} to 75^{th} percentiles). It is notable that modeled concentration changes closely track the year-to-year variations displayed in the observed results. Of particular significance, these modeled and observed results also reveal that weekday morning NO_X concentrations were increasingly lower following summer 2002, particularly over the

summer periods of 2004 to 2006. However, medians for the modeled changes are somewhat less negative (i.e., smaller decreases) than observed medians. The most notable difference between modeled and observed concentration changes occurred for the summer 2006, the last simulation period. Overall, the observed median change (ΔC_{06-02}) reveals a nearly 25% decrease in NO_X concentrations. However, the corresponding modeled median change indicates a decrease of about 15%, which is in close agreement with the change in all surface NO_X emissions between 2002 and 2006 shown earlier. Although NO_X emissions from all surface source types (i.e. minor point sources, area and nonroad sources) in a 12-km grid cell impact model concentrations, all of these sources may not have necessarily influenced the observed concentrations at the particular monitoring sites since certain emission sources would not have been directly upwind of the measurement location. Additionally, greater uncertainty also exists in the magnitudes as well as the temporal variation of these non-mobile NO_X emission sectors in urban areas. Although the emissions inventory contains best available estimates, uncertainties could not be quantified.

Another interesting aspect of the modeled and observed 3-hour NO_X concentrations can be gleaned from the correlations for the concentrations between locations within the same major urban area. As expected, correlation coefficients (r) were consistently higher in modeled concentrations in adjacent grid cells than between paired observations located in those corresponding cells. For example, average r values derived for the morning weekday NO_X concentration series between a reference site and other measurement sites located within 50 km for large cities like Washington, DC, New York City, and Boston were 0.84 (0.64), 0.86 (0.44), and 0.87 (0.64) from the model and measurement sites (in parentheses), respectively, which are consistent with those presented by Biswas et al. (2001). Thus, these results illustrate the importance of local-scale influences and the spatial gradients associated with nearby emission sources on

individual site measurements that cannot be captured by the horizontal grid cell size employed in the model. With a grid cell size of 12 km, the Nyquist interval is twice the grid size, implying that spatial features on scales less than 24 km cannot be resolved by the model. In reality, those features of spatial scales less than four times the grid size cannot be resolved well by the model. Nevertheless, these dynamic evaluation results of change in NO_X concentrations indicate that the modeled results are able to capture a decreasing trend displayed in the site measurements over this 5-year period.

Figure 5 displays median values of the 3-hour morning concentrations computed from the upper portion (50^{th} – 95^{th} percentiles) of the CFDs noted previously from all urban sites. These results demonstrate that both observed and modeled NO_X concentrations have decreased over this 5-year period. There is certainly considerable overlap of the inter-quartiles; however, it is apparent that the medians of observed concentrations are consistently greater than modeled values during each summer period. These results suggest another interesting feature concerning comparisons of observed and modeled concentration pairs in complex urban environments. Observed concentrations of primary species, in this case NO_X, can exhibit higher concentrations due to the relative proximity of a local source (i.e. major roadway or other emission source) that cannot be captured at the grid resolution employed in this modeling study, for the reasons explained previously. Nevertheless, there are common features in the modeled and observed results in the box/whisker plot of Figure 5. In particular, there is a noticeable tendency of high NO_X concentrations, as represented by values at the 90^{th} percentile, becoming lower and the concentration variability diminishing steadily (i.e., narrower distributions) over this period in both modeled and observed values as a consequence of the emission reductions. Weekday morning NO_X concentrations from northeastern measurement sites covering the longer period from 1997-2005 appear to be comparable to these results (NESCAUM, 2006). Recently, Geddes et al. (2009) reported that NO_X

concentrations decreased by 30-40% at most monitoring sites located in metropolitan Toronto, Ontario, which are somewhat greater than our overall findings, but they happen to span a longer period from 2000-2007.

Since weekday morning NO_X concentrations have decreased gradually over this 5-year period, there is interest in investigating the possible effect on weekday morning ground-level ozone (O_3) concentrations. In particular, ambient ozone is strongly titrated by its well-known reaction with the high NO concentrations, associated in this case with mobile emissions, to form nitrogen dioxide (NO_2), which leads to very low O_3 concentrations during morning traffic rush hours. To determine whether there has been a change in the magnitude of the morning "ozone deficit" (ΔO3) in response to decreasing NO_X concentrations, 3-hour average weekday O_3 concentrations for the 0100-0400 AM (nocturnal) and 0600-0900 AM periods were computed from hourly observed and modeled values. Ozone concentrations were found to display small variations with time during the nocturnal hours (pre-rush period) in the same manner as the NO_X concentrations (Figure 2), so ΔO_3 was defined herein to be the difference from the rather steady O_3 value averaged over the pre-rush 0100-0400 AM period and the 0600-0900 AM average O_3 concentration. The modeled and observed ΔO_3 results based on 32 urban ozone locations collocated with NO_X monitoring sites are displayed in Figure 6, which reveals that modeled and observed ΔO_3 values were indeed quite comparable with median values between 10 and14 ppb. However, the observed ΔO_3 exhibits a more noticeable change toward smaller deficits than the modeled results over these summer periods. Interestingly, the observed percentage decline in ΔO_3 appears to be comparable to the relative change in the observed NO_X concentrations. An examination of the O_3 concentrations revealed the decline in the observed ΔO_3 was primarily due to a decrease in the 3-hour nocturnal ozone levels from year-to-year, which was not found in the modeled results. In this case, the lack of change in the non-mobile

NO_X emissions during the nocturnal period prior to the traffic rush period may be an important factor for the lack of response in the model results.

As far as the impact on daytime ozone air quality, CMAQ modeling results based on the combination of major point source (i.e. utility sector) and mobile NO_X emissions reductions (Gilliland et al., 2008) showed larger decreases in the daily maximum 8-hour ozone concentrations across the eastern US than model simulation results based solely on major point source emission reductions (Godowitch et al., 2008). The spatially-distributed decreases in mobile NO_X emissions not only lead to lower morning NO_X concentrations, but also appear to have contributed to the reductions in maximum ozone concentrations on the regional-scale over this multi-year period. This effect is borne out from an analogous dynamic evaluation analysis of modeled and observed daily maximum 8-hour ozone at the rural-based CASTNET (Clean Air Status and Trends Network, www.epa.gov/castnet) sites distributed throughout the eastern US. Figure 7 indicates notable decreases in the daily maximum 8-hour ozone levels relative to summer 2002. The modeled ozone changes in Figure 7 also track the observed changes which display an overall decline in the daily maximum 8-hour ozone levels across the eastern US over this same period. The meteorological conditions during the summer 2004 ozone season were anomalously cooler and wetter than normal with both modeled and observed changes responding by displaying a substantial drop in ozone levels during that summer period. When omitting summer 2004 values, results based on the other summer periods in Figure 7 suggest a distinctive yet steady downward trend in the daily maximum 8-hour ozone in the region, with the observed changes indicating a somewhat greater decline than found in the modeled results. Again, the fact that surface NO_X emissions, with the exception of the mobile on-road sector, remained unchanged in the model simulations is believed to be partly responsible for the discrepancy seen between these observed and modeled O_3 changes.

3.3 Variations in NO_X Concentrations due to Meteorological Parameters

Although, as previously mentioned, little difference in NO_X emissions was found from weekday-to-weekday at these urban locations, large daily differences in the modeled and observed 3-hour average morning NO_X concentrations certainly existed from weekday-to-weekday, which are believed to be attributable to the variability in key meteorological processes influencing the horizontal transport and dispersion of pollutants. In particular, variations in the wind speed and mixing height can strongly perturb NO_X concentration levels from one day to the next. Specifically, the ventilation coefficient (Vc), defined as the product of wind speed and mixing height (Rao et al., 2003) was examined. Vc is generally inversely proportional relationship with pollutant concentration levels. Consequently, conditions with light wind speeds coupled with a shallow mixing height result in small Vc values, which generally contribute to higher concentrations due to the lower dilution capacity of the atmosphere. Vc values were computed from the modeled wind speeds and modeled mixing heights for each weekday morning period at each urban location and the corresponding NO_X concentrations were grouped together into bands based on the magnitudes of Vc. The results in Figure 8 reveal that the higher NO_X concentrations tended to occur under low ventilation conditions, as anticipated, and both modeled and observed concentrations decreased at higher Vc levels. Another interesting feature in Figure 8 is that for each Vc interval, which is representative of similar meteorological conditions, both modeled and observed NO_X concentrations exhibited a notable decrease between the summer periods of 2002 and 2006. Results from summer periods during the interim years (not shown for clarity) generally fell between these values.

Concentrations at a particular site often vary with wind direction, primarily due to the relative position of a site from sources within an urban area. For example, a monitoring

site located on the northeast side of a city would exhibit generally higher NO_X concentrations under southwesterly wind flows. Both observed and modeled morning weekday NO_X concentrations displayed comparable differences among the wind direction quadrants (not shown). Since wind direction frequencies within each wind quadrant from the mornings were found to be very comparable among the summer periods at these selected sites, variations in this meteorological parameter did not appear to impact the concentration change results.

4. Summary

A dynamic model evaluation approach was applied to investigate the change in modeled and observed NO_X concentrations during weekday morning traffic rush hour periods from summer months over 5 consecutive years. Model estimates of mobile NO_X emissions revealed a 25% decline over this multi-year period. Results of modeled and observed changes in 3-hour average NO_X concentrations from the 50th to 95th percentiles of CFDs exhibited discernable overall declines at urban locations in the eastern US. The modeled changes tracked the year-to-year pattern displayed in the observed changes, although overall changes in modeled concentrations were found to be somewhat less than the observed changes because surface NO_X emissions, other than on-road mobile sources, remained unchanged in the model simulations over this period. The year-to-year pattern of concentration changes in the model results, as expected, closely followed the decline displayed in total surface NO_X emissions.

Since the mobile NO_X emission changes were solely responsible for the change in all ground-level NO_X emissions, the lack of change in non-mobile surface NO_X emission sectors moderated the overall decline in total NO_X emissions at the surface in the model to about 15% between summer 2002 and 2006. Nevertheless, a beneficial result of the decline in NO_X emissions is the marked decrease in weekday morning NO_X

concentrations, especially at higher levels, as demonstrated by the drop in model and observed values at the 90th percentile. The decline in NO_X concentrations also appeared to cause a response in the weekday morning ozone deficit, with evidence of a more perceptible decrease in the observed than modeled results. Lastly, the overall reductions in the on-road mobile emissions generated by the MOBILE6 model appear to be consistent with the overall observed NO_X concentration changes found at these eastern urban sites.

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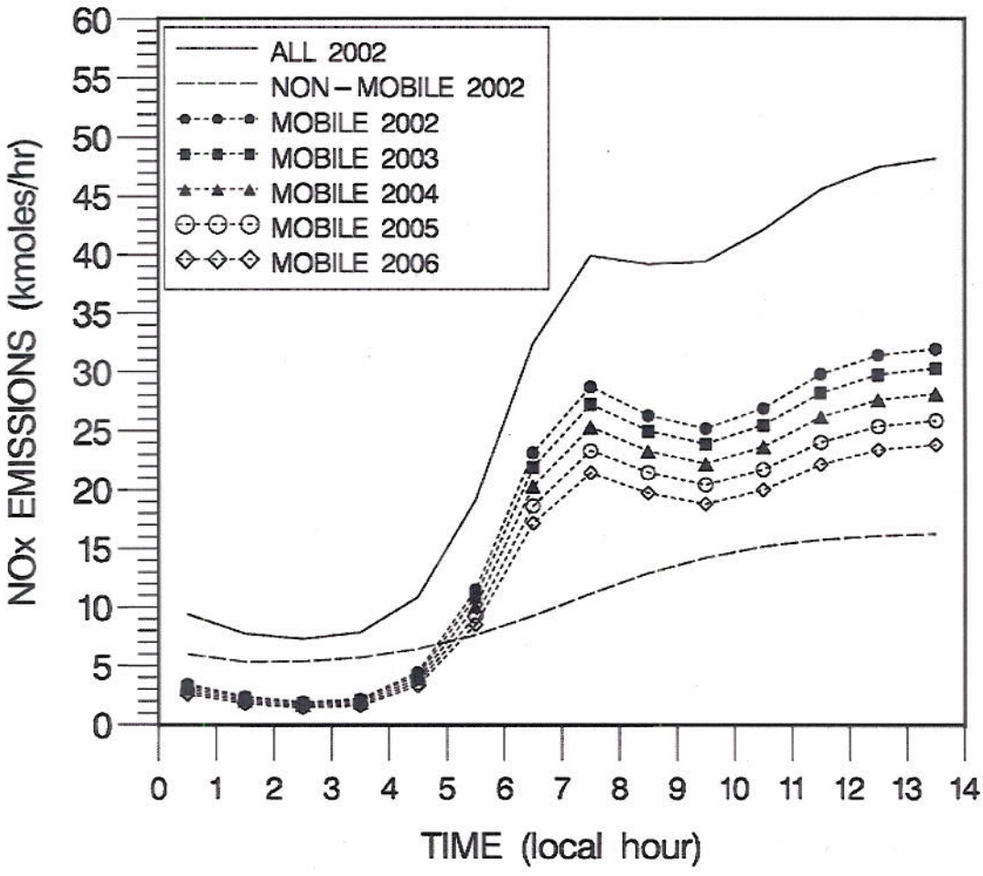

Figure 1. Time variation of hourly average weekday mobile NO_X emissions in the grid cell for Washington, DC from each summer period. All surface NO_X emissions and non-mobile (i.e., area, minor point, and nonroad sources) NO_X emissions for summer 2002 only are also displayed.

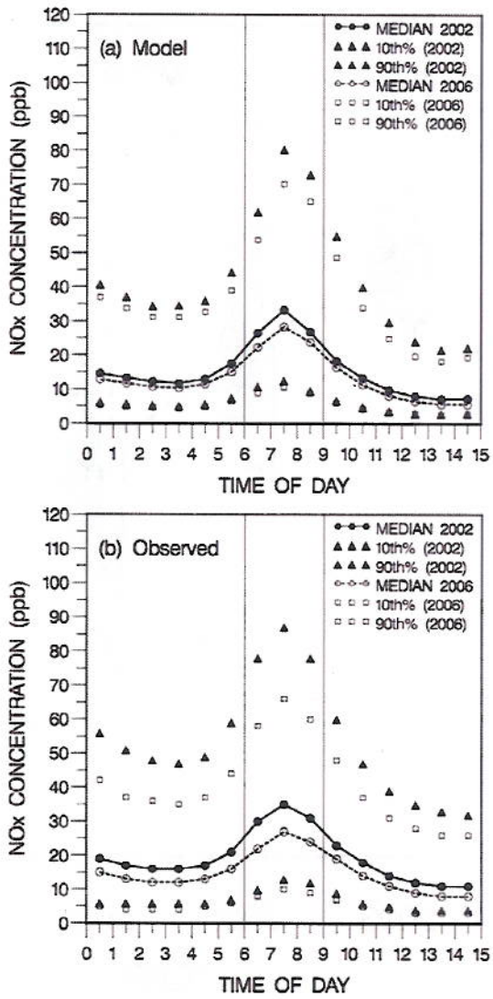

Figure 2. Temporal variation of hourly weekday median a) modeled and b) observed NO_X concentrations from summer 2002 and 2006 periods based on values at 42 urban locations. Values at the 10^{th} and 90^{th} percentiles of the cumulative frequency distributions are depicted by symbols.

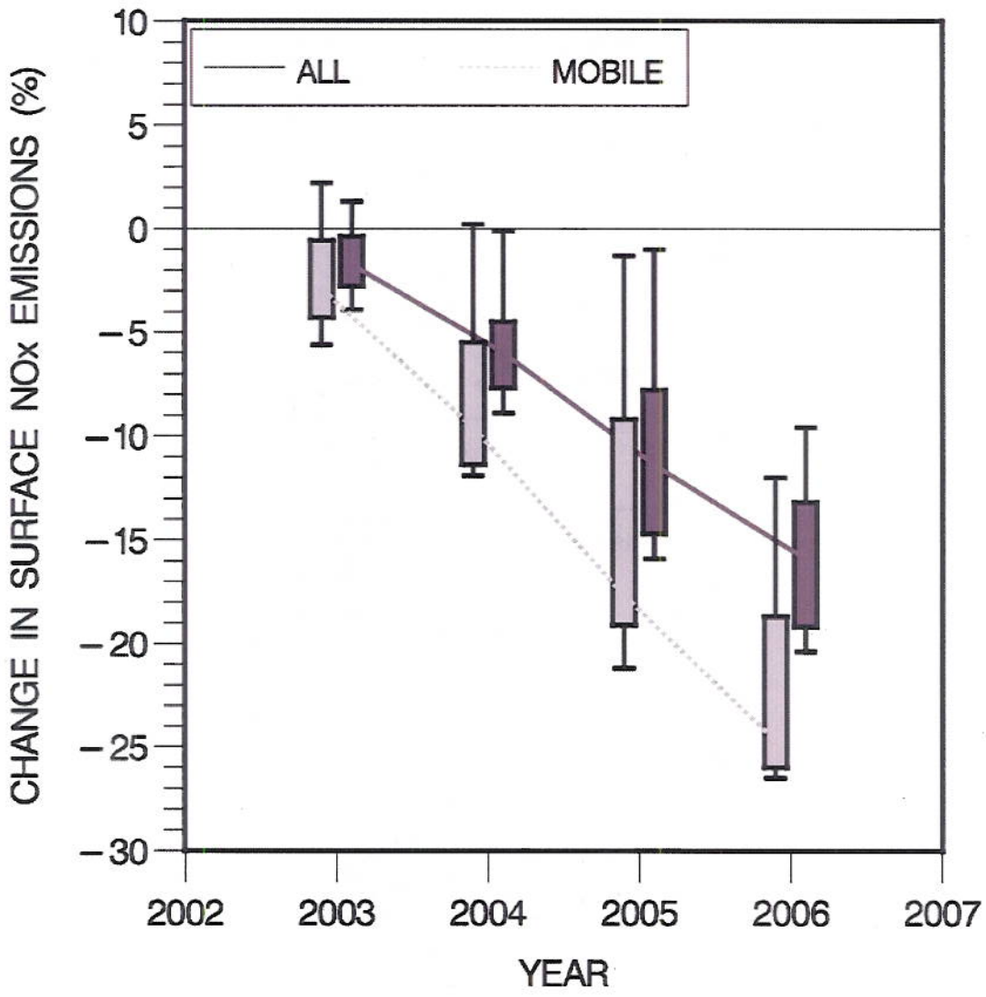

Figure 3. Percentage changes in weekday morning NO_X emissions from all (dark gray) and on-road mobile (light gray) sources relative to the summer 2002 period. The box/whisker plot displays lines connecting the median values. The boxes extend from the 25th to 75th percentiles and whiskers extend to the 10th to 90th%. Results are based on values from urban grid cells containing the 42 NO_X concentration monitoring sites.

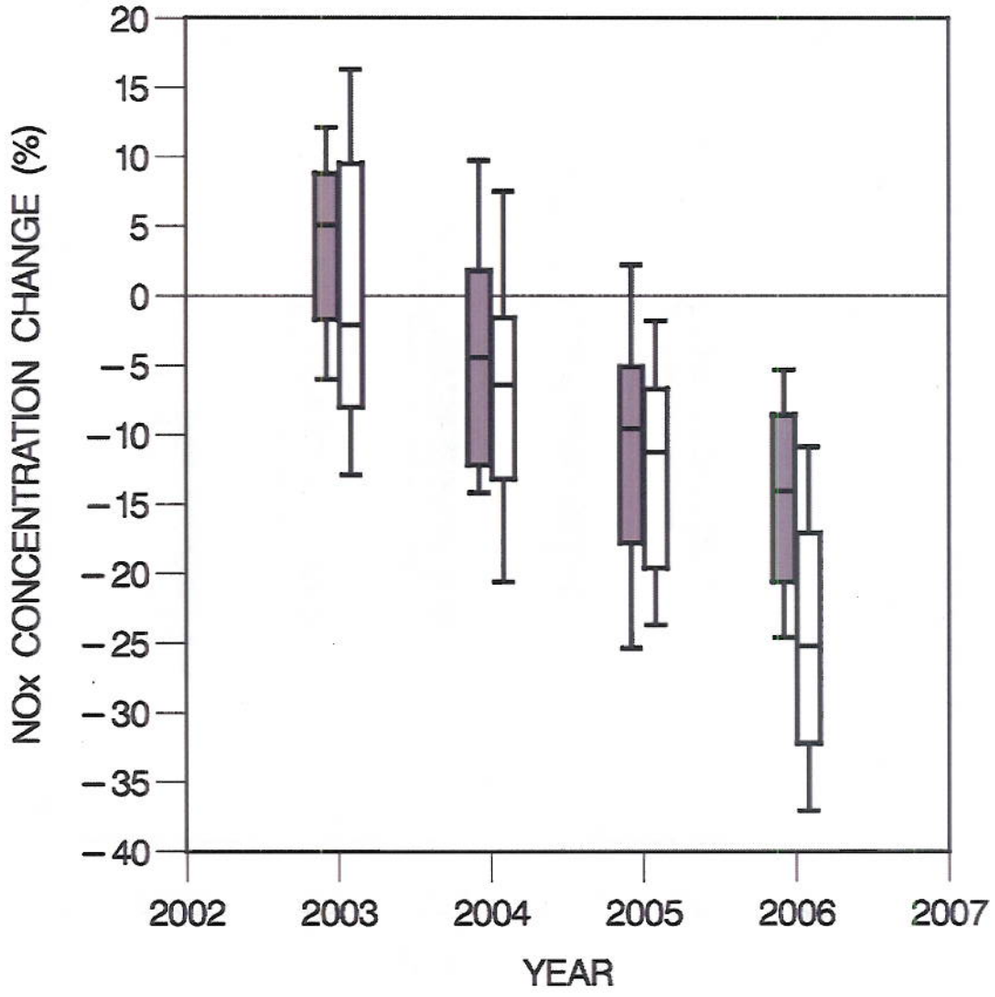

Figure 4. Change in weekday 3-hour average morning NO_X concentrations from modeled results (gray) and observations (white) based on the same set of 42 urban sites for each summer period. The box / whisker plot shows the medians denoted by the horizontal line inside each box. The boxes extend from the 25th to 75th percentiles and whiskers span from the 10th to 90th percentiles of the cumulative change distributions.

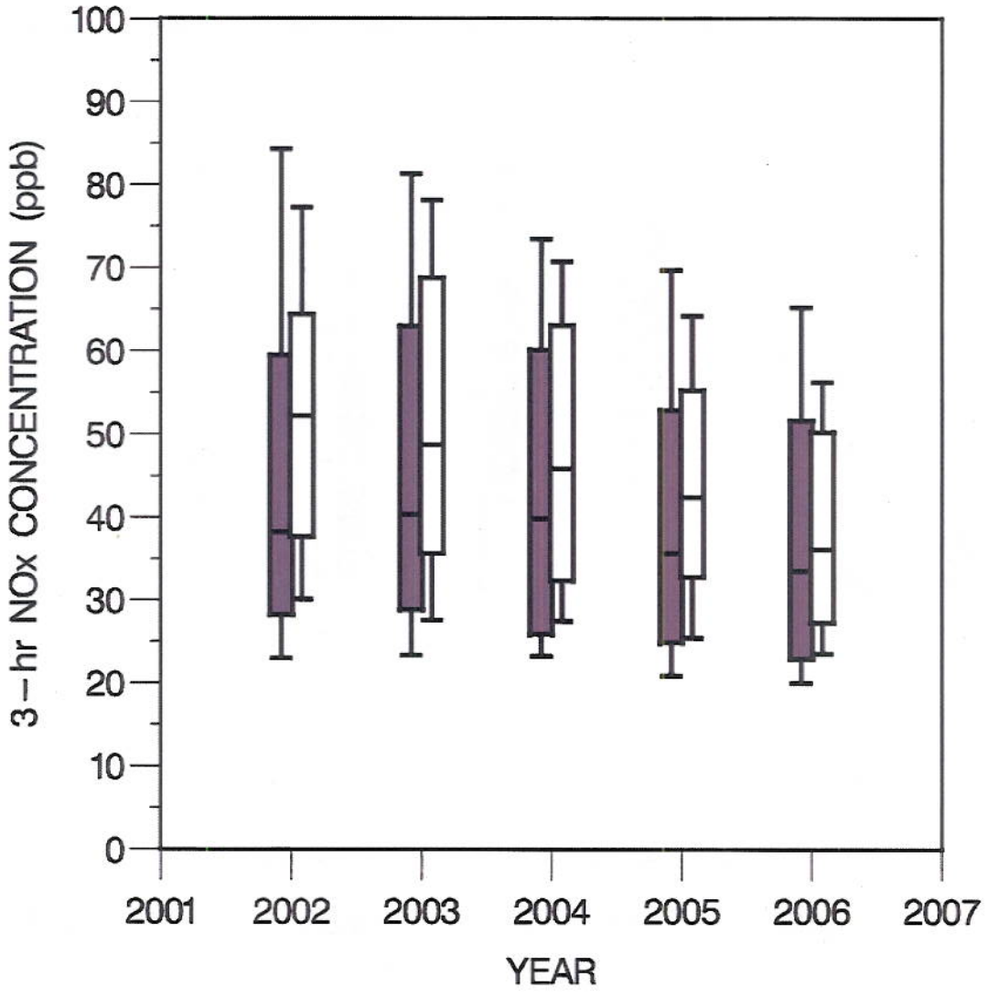

Figure 5. Weekday morning 3-hour average modeled (gray) and observed (white) NO_X concentrations during summer 2002 through 2006 from a set of 42 urban sites in the eastern US. The box/whisker plot depicts the median (horizontal line inside the boxes) in each case. Boxes extend from the 25th to 75th percentiles and the whiskers extend out to the 10th and 90thpercentiles.

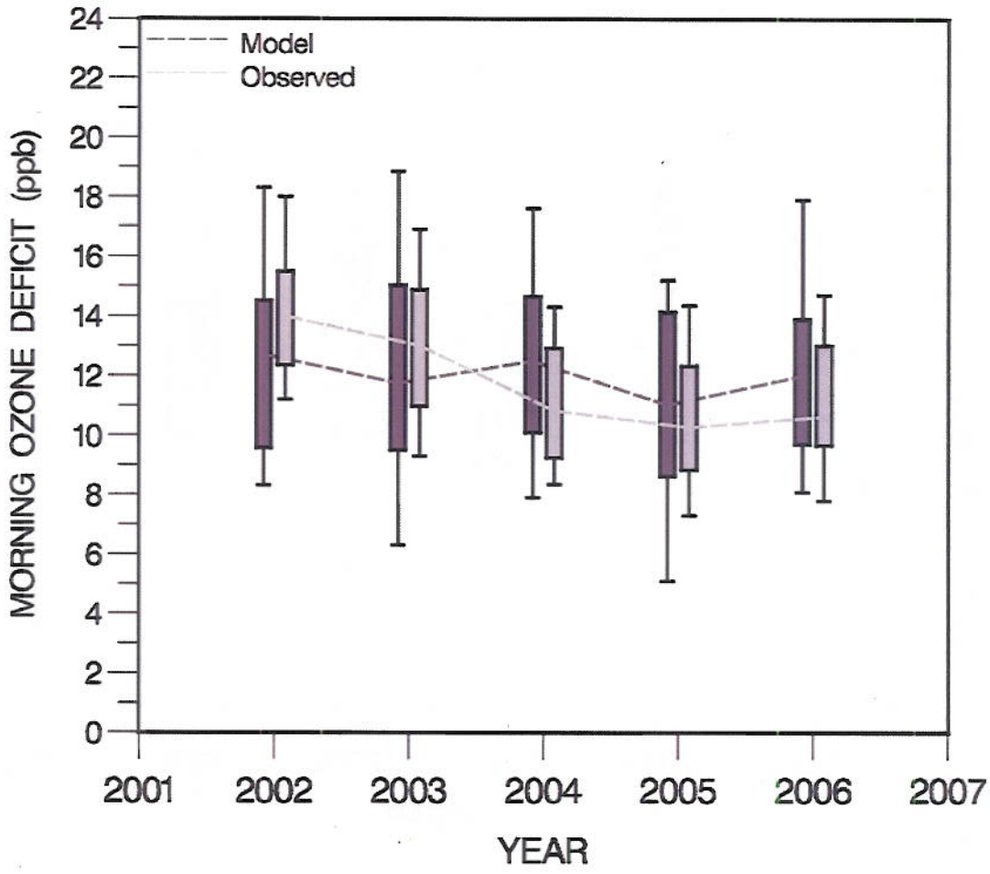

Figure 6. The morning urban ozone deficit (ΔO_3) in modeled and observed results from each summer period. Median values are connected by dashed lines. The boxes extend from the 25th to 75th percentiles and the whiskers represent values at the 10th and 90th percentiles.

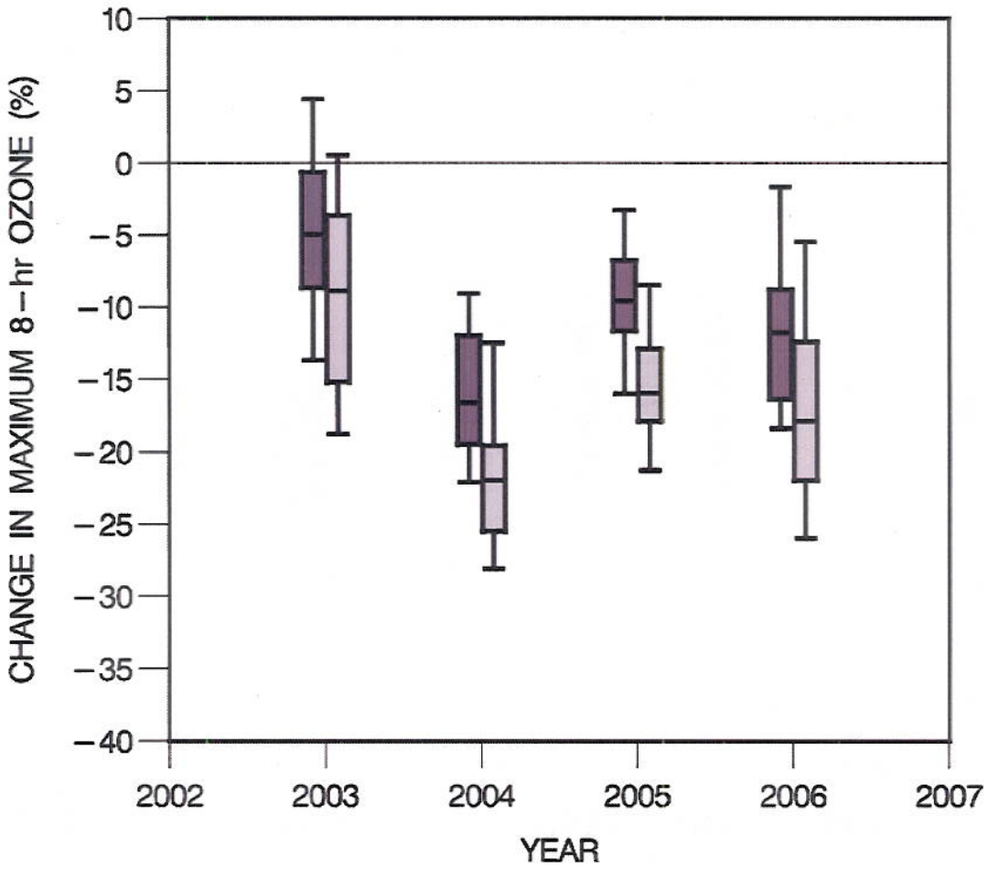

Figure 7. Percentage change in modeled (dark gray) and observed (light gray) maximum 8-hour ozone concentrations (≥ 95th percentile) at CASTNET sites in the eastern United States.

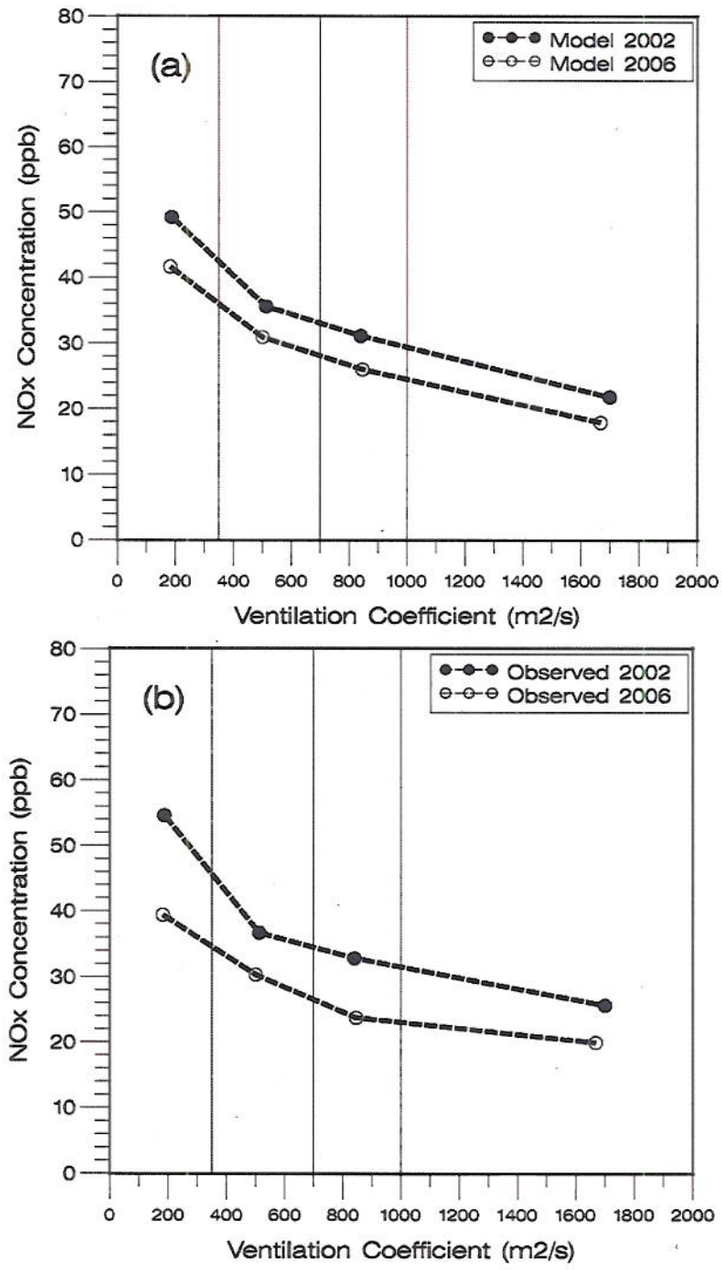

Figure 8. Variation in median weekday 3-hour morning NO_X concentrations (averaged over the 50^{th}-95^{th} percentiles) versus the ventilation coefficient (Vc) from a) model results and b) observations from summer 2002 and 2006 periods. Vertical lines denote the limits for averaging values into four Vc groups.