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Trends in OMI NO_2 observations over the United States: effects of emission control technology and the economic recession

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Abstract. Observations of tropospheric NO_2 vertical column densities over the United States (US) for 2005–2011 are evaluated using the OMI Berkeley High Resolution (BEHR) retrieval algorithm. We assess changes in NO_2 on day-of-week and interannual timescales to assess the impact of changes in emissions from mobile and non-mobile sources on the observed trends. We observe consistent decreases in cities across the US, with an average total reduction of 32 ± 7 % across the 7 yr. Changes for large power plants have been more variable (-26 ± 12 %) due to regionally-specific regulation policies. An increasing trend of 10–20 % in background NO_2 columns in the northwestern US is observed. We examine the impact of the economic recession on emissions and find that decreases in NO_2 column densities over cities were moderate prior to the recession (-6 ± 5 % yr^{-1}), larger during the recession (-8 ± 5 % yr^{-1}), and then smaller after the recession (-3 ± 4 % yr^{-1}). Differences in the trends observed on weekdays and weekends indicate that prior to the economic recession, NO_2 reductions were dominated by technological improvements to the light-duty vehicle fleet but that a decrease in diesel truck activity has contributed to emission reductions since the recession. We use the satellite observations to estimate a 34 % decrease in NO_2 from mobile sources in cities for 2005–2011 and use that value to infer changes in non-mobile sources. We find that reductions in NO_2 from non-mobile sources in cities have been both more modest and more variable than NO_2 reductions from mobile sources (-10 ± 13 %).

1 Introduction

Nitrogen oxides ($NO + NO_2 \equiv NO_x$) play an important role in tropospheric chemistry. On a regional scale, NO_x degrades local air quality due to its dual role in regulating the production of ozone and its contribution to particle formation. NO_x is emitted into the troposphere by both anthropogenic (transportation, power generation) and natural (lightning, soil, fires) sources with anthropogenic sources dominating in densely-populated regions.

Efforts to reduce anthropogenic NO_x emissions in North America and Europe have led to dramatic reductions in urban NO_x concentrations (Richter et al., 2005; Kim et al., 2006, 2009; Stavrakou et al., 2008; van der A et al., 2008; Konovalov et al., 2010; Russell et al., 2010; Castellanos and Boersma., 2012; Zhou et al., 2012). These reductions are largely attributed to emission control measures; however, despite the improvements, concentrations in the United States (US) continue to reach levels that promote high ozone and that are detrimental to human health (US Environmental Protection Agency (EPA), 2010). Regulatory efforts implemented over the past decade have aimed at reducing emissions by establishing stricter emissions standards for both light- and heavy-duty vehicles and by imposing stricter controls on electric power generation, sources which taken together account for approximately 55 % of the anthropogenic NO_x budget in the US (National Emissions Inventory (NEI) 2008). Examples include the Clean Air Interstate Rule (CAIR), which requires reductions in the emissions of NO_x produced from power generation in the eastern US, and the Tier II Tailpipe NO_x Emissions Standard, which called

for a step-wise reduction (77–86 % total for cars) in emissions per year for all vehicles in the US produced between 2004 and 2009 (US EPA, 2004).

Satellite observations of NO_2 have been widely used to identify NO_x source regions and evaluate changes in emissions over time (Irie et al., 2005; Richter et al., 2005; Kim et al, 2006, 2009; van der A et al., 2006, 2008; He et al., 2007; Stavrakou et al., 2008; Kaynak et al., 2009; Mijling et al., 2009; Konovalov et al., 2010; Russell et al., 2010; Castellanos and Boersma, 2012; Zhou et al., 2012). This is possible due to the short chemical lifetime of NO_x which results in relatively small background NO_2 column densities against which source regions have high contrast. Some of this work has focused on using satellite observations to identify the causes of the observed reductions; for example, Kim et al. (2006) showed that in the eastern US, where a large proportion of NO_x emissions can be attributed to power generation, controls on power plant NO_x emissions have greatly reduced NO_x concentrations. Kaynak et al. (2009) showed that the weekly cycle in NO_2 column densities can be used to distinguish between urban, rural, and isolated point sources so that the contribution of mobile sources can be quantified. Russell et al. (2010) showed that trends in satellite-based NO_2 column densities agree well with trends in ground-based observations and a regional emissions inventory in four urban regions in California, confirming the ability of the satellite to capture changes in emissions over time. Together, these studies demonstrate the potential for using trends in satellite observations to discriminate between source contributions and thereby to provide timely updates to emission inventories.

In this work, satellite observations from the Ozone Monitoring Instrument (OMI) are used to evaluate trends in NO_2 across the US from 2005–2011. We employ the Berkeley High-Resolution retrieval (BEHR) in our analysis, which has been shown to minimize biases in the retrieved NO_2 column densities by using terrain pressure, albedo, and NO_2 vertical profiles with higher resolution than those used in the operational products (Russell et al., 2011). We describe changes in tropospheric NO_2 column densities for major cities, power plants, and remote regions of the US on day-of-week and interannual timescales. These temporal patterns are used to infer how changes in source categories have influenced trends in NO_2 concentrations.

2 Data description

2.1 OMI Tropospheric NO_2 column

The Ozone Monitoring Instrument (OMI) resides on the Aura satellite, launched in July 2004. The UV/Visible spectrometer observes solar irradiance and sunlight reflected from the surface of the Earth in the 270–500 nm wavelength range at approximately 0.5 nm resolution (Levelt et al., 2006). The 114° field of view yields a 2600 km swath of data collected continuously in the direction perpendicular to the flight path. These data are binned in order to yield observations ranging from $13 \times 24\,km^2$ at nadir to $24 \times 128\,km^2$ at the edge of the swath. The instrument achieves near global coverage daily, with overpasses occurring at approximately 01:45 p.m. local time at the equator. A Differential Optical Absorption Spectroscopy (DOAS) algorithm is applied to derive NO_2 slant column densities from the radiance measurements.

Over the years, several row anomalies have developed on the OMI detectors resulting in unreliable detection in a number of cross-track scenes (http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/OMI/index.shtml#info). In order to prevent biases arising from the variable availability of reliable pixels over the 2005–2011 time period, we uniformly exclude all affected cross-track scenes for all years of evaluation. Additionally, we exclude pixels otherwise flagged as unreliable and those with an effective cloud fraction exceeding 20 % according to the cloud retrieval described in Accareta et al. (2004).

Operational retrieval of NO_2 vertical column density from the global dataset is performed by both the National Aeronautics and Space Administration Goddard Space and Flight Center (NASA-GSFC) and the Royal Netherlands Meteorological Institute (KNMI). The resulting publicly available datasets are known as NASA's standard product (Bucsela et al., 2006; Celarier et al., 2008) and KNMI's DOMINO product (Dutch OMI NO_2; Boersma et al., 2007, 2011). Both products use the same general approach; the DOAS spectral analysis technique is used to determine NO_2 slant column densities and then the stratospheric portion of the column is subtracted to yield a tropospheric slant column. The air mass factor (AMF), a multiplicative factor that is used to convert the slant column into a vertical column, is determined by interpolating the output from a radiative transfer model according to terrain, profile, cloud, and viewing parameters, and is then applied to determine the tropospheric NO_2 vertical column density. Significant differences between the two products include different methods for inferring the stratospheric contribution to the total column and the use of different terrain and profile datasets for conversion of the tropospheric slant column into a vertical column.

2.2 BEHR Retrieval of Tropospheric NO_2 column

The BEHR retrieval of tropospheric NO_2 vertical column densities, developed at the University of California, Berkeley and available at http://behr.cchem.berkeley.edu, is an adaptation of the NASA standard product and is described by Russell et al. (2011). In the high-resolution BEHR product, AMFs are determined using terrain pressure information from the GLOBE (Global Land One-kilometer Base Elevation) $1 \times 1\,km^2$ topographical database and albedo is from the MODIS (Moderate Resolution Imaging Spectroradiometer) $0.05° \times 0.05°$ 16-day average albedo (MCD43C3,

produced every 8 days). To extend BEHR from the California domain described in Russell et al. (2011) to the rest of the contiguous US, we have made a minor change to the procedure, using profiles from WRF-Chem (Weather Research Forecasting Chemistry) monthly-averaged simulations at $12 \times 12\,km^2$. Details of the WRF-Chem simulations are included in the Appendix.

Russell et al. (2011) found that the BEHR tropospheric NO_2 columns are approximately 30 % lower in unpolluted regions and 8 % higher in polluted regions than in the NASA standard product (v.1). They observed improved agreement with aircraft observations collected during the ARCTAS-CA flight campaign compared with both the NASA standard product (v.1) and KNMI DOMINO product (v.1.0.3), and determined that significant variance between the operational retrievals and in situ observations was due to the low spatial and temporal resolution of the terrain and profile inputs, particularly the albedo and the NO_2 profile. Additional studies have also shown that low spatial resolution input parameters compromise the accuracy of OMI retrievals (Schaub et al., 2007; Zhou et al., 2009; Hains et al., 2010; Zhou et al., 2010; Boersma et al., 2011; Heckel et al., 2011). More recently, updates to the standard (version 3.2; Bucsela, 2013) and DOMINO (version 2; Boersma et al., 2011) products have included higher resolution profile and terrain parameters; however, with the exception of the temporal resolution of the a priori profiles that are resolved on a monthly basis in BEHR compared to a daily basis in DOMINO, the BEHR product continues to provide NO_2 column densities over the United States using the highest spatial and temporal resolution inputs of all OMI NO_2 retrievals that we are aware of.

2.3 Continuous Emission Monitoring System

The Continuous Emissions Monitoring System (CEMS) is a system of instruments for the measurement of trace gases and particulates from industrial combustion sources including power plants. Data is recorded hourly and made publicly available for download at ftp://ftp.epa.gov/dmdnload/. Here, we use CEMS data to evaluate the consistency of trends in emissions with trends inferred from OMI observations for isolated power plants as well as for cities in close proximity to major power generation sources.

3 Methods

We evaluated trends in OMI BEHR tropospheric NO_2 column densities for 2005–2011. Seasonal averages were determined by performing an area-weighted averaging, binning to $0.05° \times 0.05°$, as in Russell et al. (2010). Figure 1a shows the average OMI NO_2 column retrieved using the BEHR algorithm during summer (April–September) 2005. Elevated NO_2 column densities allow dozens of urban and industrial emission sources to be identified. A total of 47 cities and

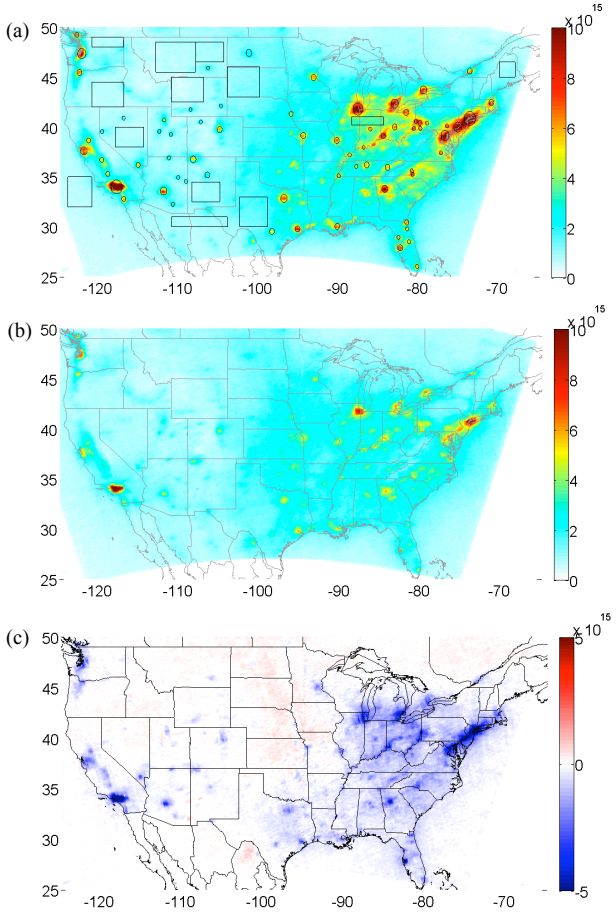

Fig. 1. Average summertime (April–September) OMI BEHR NO_2 column densities (molecules cm^{-2}) for **(a)** 2005, **(b)** 2011, and **(c)** the difference, 2011–2005.

23 power plants across the US and Canada were selected for the analyses that follow. Circular regions surrounding each location were chosen with radii large enough to capture NO_2 plumes from each source during summer 2005 (concentrations exceeding $\sim 4 \times 10^{15}$ molecules cm^{-2}), while minimizing a dampening of the signal from averaging in the low surrounding concentrations (black circles in Fig. 1a). Additionally, 12 rectangular "background" regions, defined as regions isolated from major cities and power plants, were analyzed (black rectangles in Fig. 1a). More information about the regions chosen is given in the Appendix (Table A1). Averages were computed for both winters (October–March) and summers (April–September). Weekday (Tuesday–Friday) and weekend (Saturday–Sunday) seasonal averages were also computed. Percent changes for each time period, $Y_1 - Y_2$, were determined by $(Y_2 - Y_1)/Y_1 \times 100$. Trends for cities and power plants are reported as $A \pm B$ %, where A is the average change among all locations and B is the standard deviation describing the variability. We focus most of our analysis on summertime observations when the

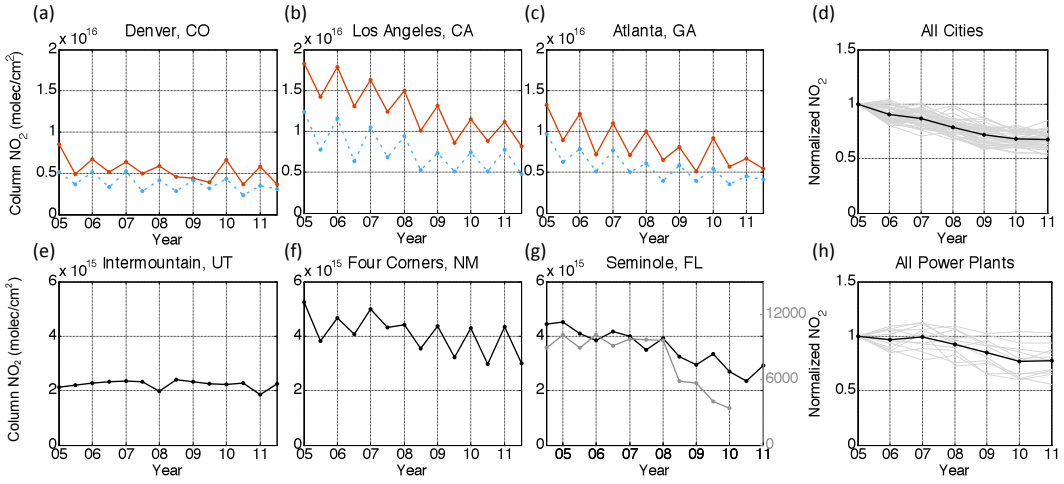

Fig. 2. Time series of average summertime (April–September) and wintertime (October–March) OMI NO_2 column densities for 2005–2011 for three US cities **(a–c)** and three power plants **(e–g)**. Average NO_2 column densities in winter are higher than those in summer due to the longer NO_x lifetime in winter. For the cities, weekday (red) and weekend (blue) time series are shown, while for power plants the combined weekday and weekend average time series are shown. CEMS power plant emissions (lbs/mmBtu) are shown in gray in **(h)**. The time series of average summertime columns normalized to 2005 concentrations for each US city and power plant (gray) and the average for all cities and power plants (black) are shown in **(d)** and **(h)**, respectively.

NO_x lifetime is short and both transport and carryover from the previous day are minimized such that variation in concentrations and columns is more closely related to variation in emissions.

4 Results and discussion

4.1 Observed trends in OMI BEHR NO_2 columns

Figure 1 shows summertime OMI BEHR NO_2 column densities from 2005 (Fig. 1a), 2011 (Fig. 1b), and their difference, 2011–2005 (Fig. 1c). Average wintertime column densities for all the sites we have analyzed are included in Appendix A (Fig. A1). From 2005 to 2011, NO_2 reductions of as much as 5×10^{15} molecules cm^{-2} (up to 47 %) are observed. Absolute reductions are largest in the most populous regions of the country, including the eastern seaboard (from Boston to Washington DC), Chicago, and Los Angeles. In contrast, small increases in column NO_2 are observed over rural regions of the central US. Comparison with the average 2005–2011 columns (not shown) indicates that NO_2 in the central US was anomalously high in 2011 and that there has been no significant trend in concentrations. Interannual variations in soil NO_x emissions as described in Hudman et al. (2010, 2012) are likely a major factor contributing to the variability of the NO_2 columns observed in the central US.

Figure 2a–c show the time series of seasonally averaged OMI NO_2 column densities on weekdays and weekends for select US cities. Sample cities shown here were chosen from different regions of the US to highlight common features. A seasonal cycle in NO_2 column densities, with larger columns in winter than in summer, is observed in each of the cities due to the longer lifetime of NO_x in winter. Additionally, we observe the well-documented weekend effect, characterized by larger NO_x concentrations on weekdays (red) than weekends (blue). The weekend effect was first observed using space-based observations by Beirle et al. (2003) and in the US, is caused by a larger proportion of emissions from mobile sources that vary according to the weekly cycle in human activity. Figure 2d shows the time series of summertime averages for all 47 cities studied here, with concentrations for each city normalized to their 2005 values. Figure 2a–d show that, in general, reductions in urban regions across the US were relatively large and linear from 2005 to 2009, but decreases in columns slowed from 2009 to 2011. Percent reductions from 2005 to 2011 are similar for US cities with an average change of −32 %. The variance among cities is approximately Gaussian with a standard deviation of ±7 %. A map showing the reductions in all cities studied here for 2005–2011 is shown in Fig. 3 (tabulated reductions are available in Appendix Table A1). In general, smaller decreases are observed in urban areas of the central US compared with urban regions located closer to the coasts. This may, in part, be due to a masking of anthropogenic reductions by NO_2 from agricultural emissions in the central US. The smallest decrease we observe is 15 % in Omaha and the largest decrease is 47 % in New York City.

Trends in NO_2 column density observed over power plants are more variable than those observed over cities. Figure 2e–g show the time series of seasonally averaged OMI NO_2 column densities for three power plants located in different regions of the US. Combined weekday and weekend averages

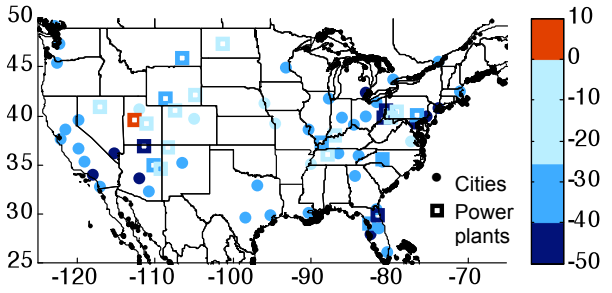

Fig. 3. Percent change in average summertime (April–September) OMI BEHR NO_2 column densities for 2005–2011 for major cities (dots) and power plants (squares). The medium blue color shows locations where reductions are within a standard deviation of the mean reduction for all cities, while the darker blue shows where reductions were larger, the light blue shows where reductions were smaller, and red shows regions where we observe increases in NO_2 for 2005–2011.

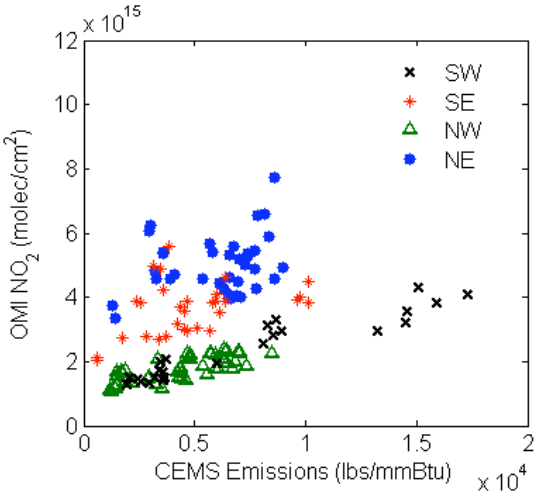

Fig. 4. Average summertime CEMS emissions versus OMI NO_2 column density for 2005–2011 for each power plant analyzed here. Symbols show the locations of the power plants by quadrant of the US (NW: northwest, SW: southwest, NE: northeast, SE: southeast).

are shown since a weekly cycle is not observed (not shown) over power plants. Figure 2h shows the time series for all power plants studied here, each normalized to their 2005 values. Trends at power plants are generally negative (Fig. 2e–h); however while large decreases are observed at some locations (e.g. Seminole, FL, Fig. 2g), little change is observed at others (e.g. Intermountain, UT, Fig. 2e). An average change of -26 ± 12 % is observed. OMI observations suggest that power plant emission reductions have been highly variable in the western US where there has been little effort to regulate emissions. In the eastern US, we observe more consistent decreases in NO_2 near power plants; however, it is much more difficult to isolate eastern power plant plumes from the nearby urban areas and therefore observed changes are likely influenced by both regulatory efforts to reduce power plant emissions (CAIR) and reductions from other sources in nearby urban centers. At these power plants, we tend to observe decreases that are linear (not shown), likely demonstrating the influence of urban reductions that are occurring roughly linearly over time. In contrast, at the more isolated power plants, the change is more variable. In some cases, we observe reductions that are step-wise (e.g. Fig. 2g), as expected if plants have installed new technology at discrete intervals over time. Trends in CEMS emissions measurements show similar features and generally good agreement with OMI observations. As an example, comparison with CEMS measurements recorded at the Seminole power plant in Florida (Fig. 2g) verifies that a step-wise reduction in emissions has occurred at the power plant (large reductions in 2007 and 2009), although the magnitude of the reduction is larger in the CEMS observations than in the satellite observations. Figure 4 shows the average summertime CEMS emissions versus OMI NO_2 column densities for each power plant analyzed in this study. There is a strong linear relationship between emissions and OMI observations at the more isolated plants in the western US. While there is a similar relationship for the eastern US power plants, the OMI measurements show that both a higher and more variable background concentration and closer proximity to urban sources complicates the direct comparison of trends in emissions from eastern power plants as measured by CEMS with columns as measured by OMI. Tablulated reductions at power plants as measured by CEMS and OMI are included in the Appendix (Table A1).

4.2 Economic impact on trends

Late 2008 marked the beginning of a significant global recession. Castellanos and Boersma (2012) showed that changes in the economy of Europe due to the recession significantly impacted regional NO_x concentrations. They observed decreases in NO_2 column densities of 10–25 % yr^{-1} for the recession years 2008–2009 compared to the long-term trend of 2–5 % yr^{-1} prior to 2008.

Here we examine the effects of the global recession on NO_2 concentrations in the US by evaluating trends in summertime OMI BEHR NO_2 during three time periods: before the recession (2005–2007), during the recession (2007–2009), and after the recession (2009–2011). The years 2007 and 2009 are included in the recession and post-recession time periods, respectively, as references against which to measure changes in subsequent years. Tabulated summertime reductions for each city and power plant are included in the Appendix (Table A1). Maps comparing the observed trends for each of the three time periods are shown in Fig. 5. In the majority of US cities, we observe moderate decreases (similar to those observed by Russell et al., 2010) for 2005–2007 (-6 ± 5 % yr^{-1}), larger decreases for 2007–

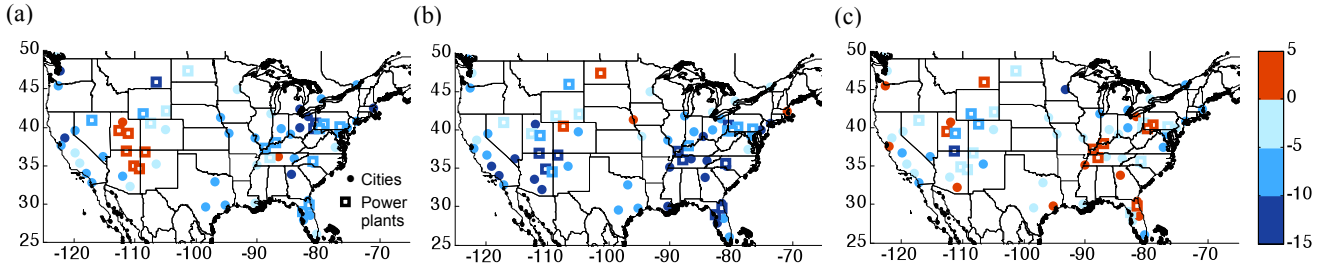

Fig. 5. Percent change per year in OMI BEHR NO_2 column for **(a)** 2005–2007, **(b)** 2007–2009, and **(c)** 2009–2011 for major cities (dots) and power plants (squares) in the US.

2009 ($-8 \pm 5\,\%\,yr^{-1}$), and smaller decreases for 2009–2011 ($-3 \pm 4\,\%\,yr^{-1}$). Reductions for cities in the western US tend to be larger than those observed previously by Kim et al. (2009) using OMI observations, likely due to both the use of high-resolution BEHR product and the manner in which regions were chosen to minimize the contribution from the low background NO_2 in this work. In general, urban areas in the southwestern and southeastern US saw the largest reductions in NO_2 during the peak of the recession. In the southeast, these same cities have seen the largest increase in NO_2 concentrations during 2009–2011. Large reductions observed for 2007–2009 are consistent with heavy duty diesel activity reports showing a reduction of 10–14 % in nationwide freight transport (California Air Resources Board, 2009), an increasing trend in total vehicle miles traveled until late 2007 followed by a steady decrease through 2009 as reported by the Department of Transportation (http://www.fhwa.dot.gov/policyinformation/travel/tvt/history/), and recent fuel consumption and NO_x emissions trends for light- and heavy-duty vehicles reported by McDonald et al. (2012). Similar trends in the NO_2 column densities during the recession are observed for power plants. The results are particularly striking for plants in the western US (Utah, Arizona, and New Mexico) where satellite observations and CEMS data show that NO_x emissions were increasing prior to 2007 but then dropped dramatically during 2007–2009. These observations are consistent with the 13 % reduction in coal-powered energy generation in the US between 2007 and 2009 reported by the US Energy Information Administration (http://www.eia.gov/electricity/annual/pdf/tablees1.pdf).

4.3 Trends on weekdays and weekends

A pronounced weekly pattern in NO_x emissions – and consequently NO_2 concentrations – has previously been observed in urban regions across the globe (Cleveland et al., 1974; Marr et al., 2002a, b; Beirle et al., 2003; Harley et al., 2005; Murphy et al., 2006, 2007; Kaynak et al., 2009; Kim et al., 2009; Russell et al., 2010). Harley et al. (2005) showed that in the US, total daily emissions from light-duty vehicles are similar throughout the week whereas on-road heavy-duty diesel emissions are reduced by roughly 60–80 % on weekends relative to weekdays. Mobile sources account for 77 % of anthropogenic emissions in the US cities analyzed here (NEI 2008) and heavy-duty diesel emissions are predicted to be the dominant mobile source of NO_x in the US in recent years (Dallmann and Harley, 2010).

Figure 6a–b shows the percent change in the OMI BEHR NO_2 column for weekdays and weekends in cities across the US for 2005–2011. Significant decreases are observed on weekdays and weekends alike; however, the magnitudes of the reductions vary, with larger reductions observed in a majority of cities on weekdays (-34 ± 8 %) than weekends (-27 ± 10 %). This result is different than observations reported by Russell et al. (2010), which showed larger reductions on weekends than on weekdays in four urban regions of California over 2005 to 2008. The largest percentage changes in NO_2 column densities – generally ranging from 40–50 % – are observed on weekdays in California and in the eastern US (Fig. 6a), coinciding with cities where differences in weekday and weekend NO_2 columns are largest. Trends on weekends in these regions are significantly lower ($\sim$ 20–30 %). In contrast, similar trends on weekdays and weekends are observed in the central US. Evaluating the changes in these trends over pre-, during, and post-recession periods, we find that for 2005–2007, we observe similar decreases on weekdays ($-6 \pm 4\,\%\,yr^{-1}$) and weekends ($-7 \pm 5\,\%\,yr^{-1}$) (consistent with Russell et al., 2010), while during and after the recession we generally observe larger decreases in NO_x emissions on weekdays (2007–2009: $-9 \pm 4\,\%\,yr^{-1}$, 2009–2011: $-4 \pm 4\,\%\,yr^{-1}$) than weekends (2007–2009: $-6 \pm 7\,\%\,yr^{-1}$, 2009–2011: $-1 \pm 7\,\%\,yr^{-1}$).

We suggest that the difference in the observed trends on weekdays and weekends between the pre-recessionary period and the recession and post recession periods has contributions from two factors: improved vehicle emission control technology and the reduced activity related to the economic downturn. Dallmann and Harley (2010) showed that there was a reduction in emission factors of about a factor of two for on-road gasoline engines between 1996 and 2006, while the reduction for on-road diesel engines was small. Coupled with steady increases in fuel consumption for both the light-duty gasoline and heavy-duty diesel fleets, McDonald

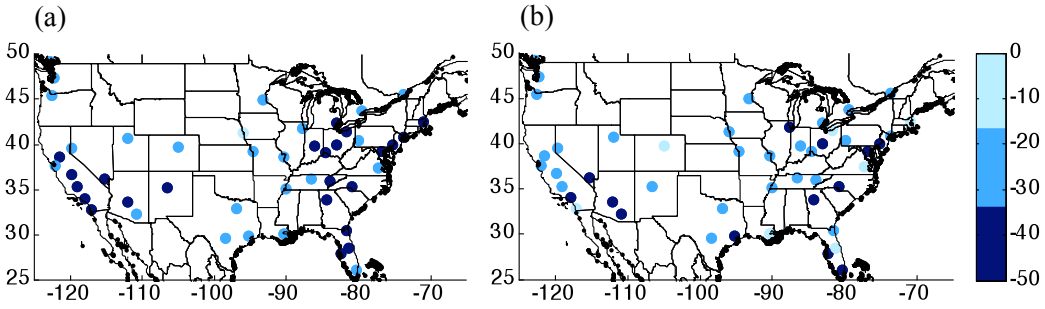

Fig. 6. Percent change in average summertime (April–September) OMI-observed NO_2 column for 2005–2011 for major cities in the US for **(a)** weekdays and **(b)** weekends.

et al. (2012) showed that NO_x emissions for gasoline vehicles decreased while emissions from the diesel fleet increased between 2005 and 2007. Russell et al. (2010) showed that, as a result of the improvements to light-duty gasoline engines, there were large decreases in NO_2 column densities in four urban regions in California from 2005 to 2008 and that the reductions were linear. This linear trend in NO_x reductions is expected to extend to cities across the US because, as Bishop and Stedman (2008) showed, reductions in fuel-specific emissions for 1999–2006 have been consistently linear in cities across the US. Since diesel trucks have been shown to contribute a larger fraction of emissions on weekdays than weekends, and their emissions increased from 2005 to 2007, we suggest that the reduction of NO_2 on weekdays prior to the recession was dampened by the increasing diesel fleet emissions, yielding reductions that are similar to or smaller than those on weekends. Here, we find that across the US, reductions on weekdays are generally smaller than decreases on weekends, as previously observed by Russell et al. (2010) for cities in California. During the recession, mobile source emissions decreased both due to improved emission control technology on light-duty gasoline vehicles and due to a 10–14 % nationwide reduction in heavy-duty diesel vehicle activity (California Air Resources Board, 2009). Recent work by McDonald et al. (2012) shows that while diesel NO_x emissions had steadily increased between 1990 and 2007, the decrease in diesel fuel consumption since 2007 has resulted in a decrease in diesel NO_x emissions. Since diesel trucks are primarily on the road on weekdays, the reduction in diesel activity has resulted in larger reductions in NO_x concentrations on weekdays compared to weekends during the 2007–2011 time period. We note that on-road diesel emissions in cities make up a relatively small fraction of the total on-road diesel emissions since a significant fraction of activity occurs outside of city centers. Unfortunately, we are unable to detect changes in on-road diesel emissions in rural regions and therefore results reported here are specific to changes in urban areas.

4.4 Trends in non-mobile sources

We estimate the percent change in non-mobile sources in each city by assuming a consistent rate of decrease for mobile NO_x emissions among cities in the US, as observed in Dallmann and Harley (2010). First, we approximate the reduction in NO_2 due to mobile sources by considering the change in NO_2 column densities for cities that are both located far from large power plant NO_x emission sources and where mobile sources are expected to contribute a large fraction ($> 85\,\%$) of total NO_x emissions according to the NEI 2008. In the 16 cities meeting these criteria, we observe a decrease of $34 \pm 5\,\%$ in NO_2 for 2005 to 2011. We apply this value to all cities and calculate the trend in non-mobile sources using observed trends in OMI BEHR NO_2 and the fractions of mobile and non-mobile sources from NEI 2008:

$$C_{\mathrm{BEHR}} = C_{\mathrm{m}} \times F_{\mathrm{m}} + C_{\mathrm{n}} \times F_{\mathrm{n}} \quad (1)$$

where C_{m} and C_{n} are the percent change in emissions from 2005–2011 for mobile and non-mobile sources, respectively, F_{m} and F_{n} are the fractions of the total NO_x emissions from mobile and non-mobile sources according to NEI 2008, and C_{BEHR} is the 2005–2011 percent change in column NO_2. Figure 7 shows the changes in non-mobile emissions for 2005–2011 inferred for each city using Eq. (1), assuming a 34 % reduction in mobile sources and using the mobile/non-mobile ratio for each city according to NEI2008. Open circles show cities where the change is $< \pm 5\,\%$ while closed circles show where the change is $\geq \pm 5\,\%$. Changes in non-mobile sources are generally negative ($-10\,\%$) with significant variability ($\pm 13\,\%$) between locations. The largest reductions in non-mobile emissions are inferred for cities in the Ohio River Valley (Indianapolis, IN, Columbus, OH, Pittsburg, PA), in Jacksonville, FL, and in Las Vegas, NV. Observations from CEMS (not shown) confirm that power plants in close proximity to these cities experienced large emission reductions during 2005–2011 that likely influenced the observed NO_2 in these cities.

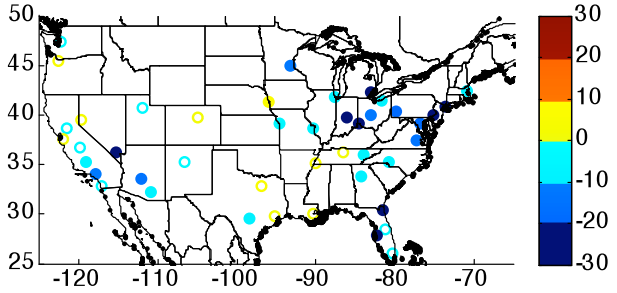

Fig. 7. Percent change in NO_2 from non-mobile sources for 2005–2011 (derived from OMI observations) in major cities of the US. Open circles show where the change is $< \pm 5\,\%$ while the solid dots show where the change is $\geq \pm 5\,\%$.

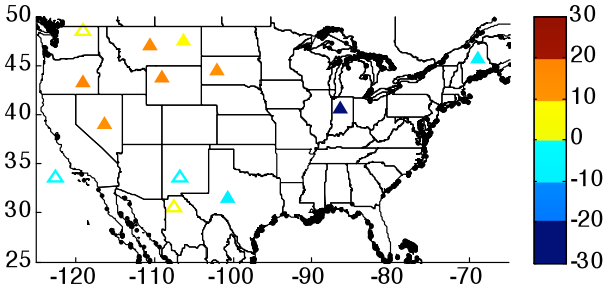

Fig. 8. Percent change in OMI-observed background NO_2 columns. Open triangles show where the change is $< \pm 5\,\%$ while the solid triangles show where the change is $\geq \pm 5\,\%$.

4.5 Trends in background NO_2

Figure 8 shows the percent change in average NO_2 column over 12 remote regions that do not contain any major cities or power plants. Increases of 10–20 % (0.8–1.4×10^{14} molecules cm^{-2}) are observed over remote areas of the northwestern US while decreases (10–20 %) are observed over regions more heavily influenced by nearby, densely populated urban centers (Texas, Maine, and Indiana). Both increasing and decreasing trends observed by OMI are linear for 2005–2011. The largest percentage enhancements are observed in the far northwestern corner of the US, with smaller percentage enhancements observed for regions further south and further east. Average weekday and weekend concentrations (not shown) are similar for remote regions in the northwest.

The spatial pattern of the NO_2 trends observed over remote locations in this work is similar to modeled spatial patterns in O_3 and PAN enhancements predicted for trans-Pacific transport of rising Asian anthropogenic emissions (Jacob et al., 1999; Jaffe et al., 2003, 2007; Hudman et al., 2004; Zhang et al., 2008; Cooper et al., 2010). GEOS-Chem simulations described in Zhang et al. (2008) for 2000 and 2006 showed that a doubling of anthropogenic NO_x emissions in Asia produced only a 3 pptv increase in NO_x concentrations, an increase that is too small to detect from space. Since trans-Pacific transport displays a spring maximum, we tested seasonal cycles of NO_2 column densities for the remote regions; however, no clear or consistent seasonal cycle was observed among the background regions to support the idea that trans-Pacific transport could be contributing to the increasing trend. Additionally, there are no ground-based monitoring sites in the regions to verify the trends. We suspect that the low concentrations observed over these regions may be largely influenced by uncertainties related to the subtraction of the seasonally varying stratospheric component of the observed column. We propose that the constant weekly cycle suggests that local mobile source emissions are not responsible and that the linearity of the trends implies that biomass burning is not the cause. Changes in agricultural NO_2 emissions could be responsible and should be explored further.

4.6 Effects of OH feedback on NO_x lifetime on observed trends

A non-linear relationship between NO_x concentrations and the predominant NO_x sink, OH, is well known. A maximum in OH concentrations in the planetary boundary layer occurs at NO_x concentrations on the order of a few ppb (e.g. Murphy et al., 2006; Lamsal et al., 2011; Valin et al., 2011). Most, if not all US cities have NO_x concentrations that exceed the peak in the OH versus NO_x curve (e.g. Pusede et al., 2012). In this regime, decreases in NO_x result in nearly equal percentage increases in OH. The increased OH is a feedback that results in NO_x concentrations decreasing faster than reductions in emissions. As a result of this lifetime feedback, the observed reductions in NO_2 column reported in this manuscript are almost certainly larger than the emissions reductions that are primarily responsible for the trends. The feedback effect is likely no more than 50 % of the observed trend. In contrast, the relative changes in emissions between different source categories are likely more accurate.

5 Conclusions

Using satellite observations of NO_2 vertical column density from the OMI instrument, we detect large decreases in NO_2 concentrations over urban regions across the US between the years 2005 and 2011. Percentage reductions in urban areas are similar across the entire US, with an average change of $-32 \pm 7\,\%$. These results indicate that improved emission control technology for on-road mobile sources has been successful in reducing NO_2 concentrations in cities across the US. Spatially, we see that reductions have been the largest in the eastern and western parts of the country and smaller in the central US. Trends over power plants are more variable, with patterns that reflect regional regulations.

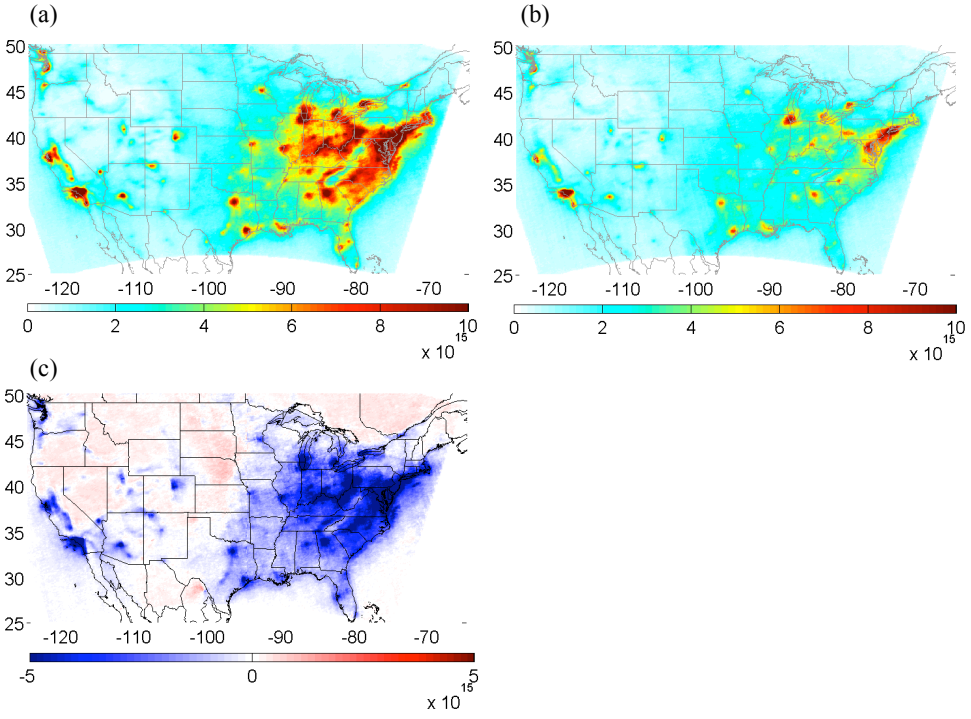

Fig. A1. Average wintertime (October–March) OMI NO_2 column densities for **(a)** 2005, **(b)** 2011, and **(c)** the difference, 2011–2005.

In order to explore the impact that the economic recession has had on NO_2 concentrations, we examined trends in OMI BEHR NO_2 prior to, during, and after the economic recession, or 2005–2007, 2007–2009, and 2009–2011, respectively. We find that NO_2 column densities in cities declined at a rate of $-6 \pm 5\,\%\,yr^{-1}$ before the recession, $-8 \pm 5\,\%\,yr^{-1}$ during the recession, and $-3 \pm 4\,\%\,yr^{-1}$ after the recession. Similarly, we also find that NO_2 concentrations over power plants generally decreased during the recession. Together, these results indicate that the economic recession had a significant impact on NO_x emissions, resulting in substantially larger reductions in NO_2 concentrations in cities and power plants across the US for 2007–2009, and that as the economy has recovered, emissions have decreased more slowly or in some cases increased. By analyzing weekday and weekend trends before, during, and after the recession, we show that emission reductions from light-duty vehicles dominated overall decreases prior to the recession but that a reduction in diesel truck activity has had a larger impact on emission reductions since the start of the recession.

Additionally, we estimate changes in non-mobile sources for 2005–2011 in cities across the US using OMI observations and the fraction of emissions from on-road mobile sources (NEI2008). In regions dominated by mobile source emissions, we infer a reduction in NO_2 from mobile source emissions of approximately $-34\,\%$. This reduction was applied uniformly to mobile sources across the US and, using the observed trends from OMI, we estimated the change in NO_2 from non-mobile sources at each location. We find that NO_2 from non-mobile sources has decreased in a majority of cities across the US for 2005–2011, but to a much lesser extent than for mobile sources. Reductions are, on average, $-10 \pm 13\,\%$ for 2005–2011, with the largest reductions likely influenced by nearby power generation according to measurements from CEMS.

Finally, we find that satellite observations suggest that NO_2 has increased by 10–20 % over remote regions in the northwestern US, despite substantially large decreases in anthropogenic emissions in urban regions across the country. This is the first time this feature has been observed. No satisfactory explanation for this observation was identified. Although it is possible an Asian influence or change in agricultural management practices is responsible, further research is needed to assess the viability of either hypothesis.

Appendix A

WRF-Chem simulation

Monthly NO_2 profiles used in the Berkeley High-Resolution retrieval (BEHR) were derived using the WRF-Chem model (Weather Research and Forecasting Chemistry), a state-of-the-art, multi-scale regional 3-D air quality chemical transport model (Grell et al., 2005). Our domain covers $3480 \times 8040\,km^2$ centered on the United States

Table A1. Summary of regions evaluated in this study and the percent change in NO_2 observed by OMI and at CEMS monitors.

Location	Latitude	Longitude	Radius	OMI 2005–2007 (% yr^{-1})	OMI 2007–2009 (% yr^{-1})	OMI 2009–2011 (% yr^{-1})	OMI 2005–2011 (%)	OMI 2005–2011 (% yr^{-1})	CEMS 2005–2010 (% yr^{-1})
Albuquerque, NM	35.20	−106.55	30	−3.22	−6.97	−6.80	−30.43	−5.07	
Atlanta, GA	33.80	−84.35	35	−10.07	−13.53	4.05	−37.04	−6.17	
Bakersfield, CA	35.30	−119.00	25	−1.16	−12.53	−3.84	−32.43	−5.41	
Boston, MA	42.45	−71.00	30	−14.70	0.26	−5.46	−36.78	−6.13	
Charlotte, NC	35.25	−80.85	20	−7.18	−12.41	−0.81	−36.67	−6.11	
Chicago, IL	41.80	−87.70	60	−4.14	−5.91	−8.09	−32.21	−5.37	
Cincinnati, OH	39.10	−84.55	25	−9.20	−12.08	−0.85	−39.18	−6.53	
Cleveland, OH	41.45	−81.67	30	−11.85	−8.39	3.66	−31.85	−5.31	
Columbus, OH	40.00	−83.10	30	−11.63	−5.38	−5.26	−38.72	−6.45	
Dallas, TX	32.85	−96.95	40	−5.89	−6.94	−1.68	−26.57	−4.43	
Denver, CO	39.75	−105.00	30	−2.89	−5.44	−4.91	−24.28	−4.05	
Detroit, MI	42.35	−83.10	45	−11.17	−2.96	−9.29	−40.50	−6.75	
Fresno, CA	36.70	−119.75	25	−4.34	−6.60	−7.20	−32.15	−5.36	
Houston, TX	29.80	95.25	30	−7.65	−7.74	0.30	−27.99	−4.67	
Indianapolis, IN	39.80	−86.15	20	−5.71	−9.57	−4.58	−34.92	−5.82	
Jacksonville, FL	30.45	−81.60	25	−6.42	−14.62	0.99	−37.12	−6.19	
Kansas City, MO	39.15	−94.55	30	−6.75	−4.18	−1.13	−22.52	−3.75	
Knoxville, TN	35.95	−84.00	30	−6.58	−11.86	−0.50	−34.42	−5.74	
Las Vegas, NV	36.20	−115.20	25	−7.62	−13.78	−5.64	−45.53	−7.59	
Los Angeles, CA	34.00	−117.90	70	−5.47	−14.99	−2.15	−40.33	−6.72	
Memphis, TN	35.10	−90.10	20	−1.19	−12.46	3.88	−21.02	−3.50	
Miami, FL	26.05	−80.30	20	−3.67	−9.87	−5.21	−33.38	−5.56	
Minneapolis, MN	44.95	−93.25	30	−4.14	−2.37	−10.10	−30.29	−5.05	
Montreal, QC	45.60	−73.70	30	−9.40	−0.81	−5.18	−28.37	−4.73	
Nashville, TN	36.20	−86.60	30	0.44	−11.67	−2.77	−26.96	−4.49	
New Orleans, LA	30.05	−90.30	30	−1.83	−11.23	−1.18	−27.06	−4.51	
New York, NY	40.85	−73.70	70	−9.28	−10.45	−9.20	−47.43	−7.91	
Omaha, NE	41.30	−96.05	25	−9.81	4.71	−1.55	−14.79	−2.47	
Orlando, FL	28.50	-81.30	20	-6.05	-9.51	0.67	-27.86	-4.64	
Philadelphia, PA	40.00	−75.20	50	−9.39	−9.86	−9.29	−46.92	−7.82	
Phoenix, AZ	33.60	−112.00	40	−5.56	−16.44	−1.24	−41.81	−6.97	
Pittsburg, PA	40.40	−79.95	25	−10.58	−11.48	3.75	−34.72	−5.79	
Portland, OR	45.45	−122.55	30	−7.04	−8.32	0.54	−27.61	−4.60	
Reno, NV	39.55	−119.70	20	−6.16	−6.55	−3.19	−28.65	−4.78	
Richmond, VA	37.40	−77.30	20	−4.30	−2.97	−5.11	−22.82	−3.80	
Sacramento, CA	38.65	−121.40	25	−11.93	−4.18	−3.04	−34.46	−5.74	
Salt Lake City, UT	40.70	−111.95	20	0.05	−11.81	0.36	−22.99	−3.83	
San Antonio, TX	29.55	−98.45	30	−5.74	−5.54	−4.10	−27.75	−4.63	
San Diego, CA	32.80	−117.00	25	−6.22	−5.90	−7.23	−33.94	−5.66	
San Francisco, CA	37.60	−122.00	40	−7.79	−7.76	0.89	−27.42	−4.57	
Seattle, WA	47.35	−122.25	50	−11.51	−3.88	−2.05	−31.90	−5.32	
St Louis, MO	38.65	−90.35	30	−8.05	−7.05	−2.83	−32.01	−5.34	
Tampa, FL	27.90	−82.40	30	−9.62	−12.38	−4.82	−45.10	−7.52	
Toronto, ON	43.70	−79.50	40	−6.80	−4.46	−4.48	−28.37	−4.73	
Tucson, AZ	32.25	−110.85	20	−2.42	−14.52	0.03	−32.44	−5.41	
Vancouver, BC	49.25	−122.85	25	−3.98	−9.97	−0.99	−27.75	−4.63	
Washington DC	39.15	−76.80	50	−9.02	−9.86	−5.06	−40.85	−6.81	
Bruce Mansfield, PA	40.40	−79.05	20	−8.07	−5.75	0.31	−25.34	−4.22	−0.73
Cholla, AZ	34.95	−110.20	20	1.44	−13.55	−3.65	−30.47	−5.08	−9.85
Colstrip, MT	45.90	−106.45	20	−10.84	−8.20	0.75	−33.54	−5.59	−11.93
Conemaugh, PA	40.10	−76.55	20	−6.46	−5.57	−3.42	−27.91	−4.65	5.66
Coronado Generating Station, AZ	34.55	−109.20	20	5.58	−8.85	−3.96	−15.76	−2.63	−4.05
Craig, CO	40.50	−107.35	20	−3.38	1.72	−7.92	−18.85	−3.14	−1.37
Crystal River, FL	28.95	−82.65	20	−5.74	−13.18	−0.01	−34.83	−5.81	−11.69
Four Corners/San Juan, NM	36.75	−108.30	35	6.42	−12.71	−3.23	−21.30	−3.55	−3.79
Gen J M Gavin, OH	40.55	−80.45	20	−12.00	−10.95	−2.63	−43.77	−7.30	−6.99
Gibson, IN	37.95	−87.00	20	−6.86	−6.77	0.17	−25.15	−4.19	−2.09
Hatsfields Ferry Power Station, PA	39.85	−79.90	20	−6.98	−8.30	8.84	−15.56	−2.59	7.35
Huntington, UT	39.25	−111.10	20	5.76	−8.66	−8.41	−23.31	−3.89	−1.13
Intermountain, UT	39.55	−112.55	20	2.97	−1.82	0.41	2.92	0.49	1.60
Jim Bridger, WY	41.75	−108.65	20	−7.23	−4.58	−5.69	−31.14	−5.19	−10.05
JM Stuart, OH	39.85	−80.70	20	−15.02	−9.13	−3.21	−46.48	−7.75	−18.32
Johnsonville, TN	36.05	−87.90	20	−2.07	−13.14	4.95	−22.34	−3.72	−10.49
Joppa Steam, IL	37.20	−88.75	20	−5.68	−14.25	6.31	−28.62	−4.77	4.48
Laramie River, WY	42.10	−104.85	20	−0.75	−4.04	−1.66	−12.47	−2.08	−5.49
Leland Olds, ND	47.40	−101.35	35	−3.06	3.95	−3.72	−6.23	−1.04	−6.15
Marshall, NC	35.60	−80.95	20	−6.06	−10.30	−3.58	−35.22	−5.87	−6.34
Navajo Generating Station, AZ	36.90	−111.35	20	2.65	−11.61	−15.52	−44.24	−7.37	−3.95
Seminole, FL	29.80	−81.55	20	−8.55	−16.70	4.47	−39.84	−6.64	−18.62
Valmy, NV	40.90	−117.00	20	−5.16	−3.66	−2.75	−21.46	−3.58	−8.66

($\sim$ 25–50° N, 125–65° W). A 10 day simulation was run for each month from the 11th through the 20th, with the first two days used to spin-up and the last eight days included in the average. Chemistry is computed using the Regional Acid Deposition Model 2 (RADM2; Stockwell et al., 1990) and anthropogenic emissions from on-road and off-road mobile sources are from the National Emission Inventory (NEI) 2005 at native $12 \times 12\,km^2$ resolution. More information concerning NEI2005 emissions is publicly available online (ftp://aftp.fsl.noaa.gov/divisions/taq/emissions_data_2005/Weekday_emissions/readme.txt). Initial and boundary conditions were derived from the North American Regional Reanalysis for 2005 (NARR – http://nomads.ncdc.noaa.gov/dods/NCEPNARR_DAILY).

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