The observed response of Ozone Monitoring Instrument (OMI) NO$_2$ columns to NO$_x$ emission controls on power plants in the United States: 2005–2011

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HIGHLIGHTS

• Satellite data may be used to monitor changes in NO$_x$ emissions from power plants.
• The changes in NO$_x$ emissions and NO$_2$ columns are well correlated.
• Significant changes in regional NO$_2$ levels influence the signal of the facilities.
• We discuss factors that affect the detection of NO$_2$ from power plants.

ABSTRACT

We show that Aura Ozone Monitoring Instrument (OMI) nitrogen dioxide (NO$_2$) tropospheric column data may be used to assess changes of the emissions of nitrogen oxides (NO$_x$) from power plants in the United States, though careful interpretation of the data is necessary. There is a clear response for OMI NO$_2$ data to NO$_x$ emission reductions from power plants associated with the implementation of mandated emission control devices (ECDs) over the OMI record (2005–2011). This response is scalar for all intents and purposes, whether the reduction is rapid or incremental over several years. However, it is variable among the power plants, even for those with the greatest absolute decrease in emissions. We document the primary causes of this variability, presenting case examples for specific power plants.

1. Introduction

In response to federal and state regulations, total emissions of nitrogen oxides (NO$_x$ = NO + NO$_2$) decreased since the late 1990s by 47% in the United States (US). Emissions from electric power generation and highway vehicles, two of the largest sources, decreased by 68% and 43%, respectively (http://www.epa.gov/ttn/chief/trends/index.html). The US Environmental Protection Agency (EPA) issued the 1998 NO$_x$ State Implementation Plan (SIP) Call with the intent to reduce emissions in 22 eastern states during the summer season so as to decrease ozone. In 2005, it issued the Clean Air Interstate Rule (CAIR) for 27 eastern states with the goal to decrease NO$_x$ emissions even further from power plants. Individual state rules and court orders have also contributed to power plant emission reductions. The mobile source of NO$_x$ emissions has declined nationwide as a result of the requirements of the Clean Air Act Amendments (CAA) of 1990, specifically the Tier 1 (phased-in between 1994 and 1997) and more stringent Tier 2 (phased-in
between 2004 and 2009) standards, and the gradual turnover of the fleet of light-duty vehicles (e.g., Dallmann and Harley, 2010; McDonald et al., 2012).

Satellite observations confirm that NO\textsubscript{2} columns over power plants and urban areas in the US have declined as a result. Kim et al. (2006) used both the European Remote-sensing Satellite-2 (ERS-2) Global Ozone Monitoring Experiment (GOME) and Envisat Scanning Imaging Absorption spectroMeter for Atmospheric CHArtography (SCIAMACHY) NO\textsubscript{2} column data to infer that NO\textsubscript{2} emissions from power plants in the Ohio River Valley decreased from 1997 to 2005 by about 35%, which is consistent with reported emission changes from the Continuous Emissions Monitoring System (CEMS). Kim et al. (2009) were the first to show that NO\textsubscript{2} columns from a model of chemistry and transport (CTM) using CEMS data were consistent with columns from three retrievals (i.e., the University of Bremen Ozone Monitoring Instrument (OMI), National Aeronautics and Space Administration (NASA) OMI operational product, and SCIAMACHY) over 13 isolated power plants in the western US in 2005. Russell et al. (2012) used NO\textsubscript{2} column data from the OMI Berkeley High Resolution (BEHR) retrieval algorithm to infer that NO\textsubscript{2} emissions changes from large power plants were variable because of regionally-specific regulations, decreasing by 26 ± 12% from 2005 to 2011. They estimated an average total reduction of 32 ± 7% in NO\textsubscript{2} for US cities from 2005 to 2011 with a 34% decrease in NO\textsubscript{2} from mobile sources. They attributed part of the observed decline to the turnover in the mobile source fleet and part to the global economic recession that began in 2008.

To comply with federal and state requirements, emission control devices (ECDs) were installed on power plants, which create a natural experiment to assess the response of the satellite-observed tropospheric NO\textsubscript{2} column to a known, and oftentimes rapid and significant, change in a power plant's emissions. For instance, in Selective Catalytic Reduction systems (SCRs), ammonia is mixed with the flue gas before entering the reactor so that ammonia and NO\textsubscript{x} react to form nitrogen and water. Other techniques to reduce NO\textsubscript{x} emissions include the installation of Low NO\textsubscript{x} Burners (LBVs) and Rotating Opposed Fire Air (ROFA) devices, which may be used in combination with SCRs. ECDs remove up to 90% of NO\textsubscript{x} from the effluent.

The purpose of this study is to use Aura OMI data (2005–2011) to understand the response of the NO\textsubscript{2} column to a change in a power plant's emissions; hereafter, we refer to this as the “Response”. As we will show, the Response is scalar, as the change in the column is a linear function of the change in emissions for all intents and purposes. However, there are variations in the magnitudes of the Responses. We document the primary sources of these variations. Quantifying the Response and understanding the primary drivers of its variability for power plants in the US will allow for (1) confidence in the assessment of the impact of ECDs on air quality and 2) better estimation of NO\textsubscript{2} emissions from large point sources in other regions of the world where estimates of emissions are often highly uncertain.

2. Data and method

2.1. OMI NO\textsubscript{2} column data

The OMI is on board the Aura satellite, which was launched on July 15, 2004 into a sun-synchronous polar orbit. It measures direct and backscattered solar radiation in the UV–visible range from 264 to 504 nm (Levelt et al., 2006) and provides early afternoon (local time 1300–1430) NO\textsubscript{2} columns at a spatial resolution of up to 13 × 24 km\textsuperscript{2} with global coverage within two days. We use the OMI operational tropospheric NO\textsubscript{2} column data product (version 2.1, collection 3) from 2005 to 2011, which is available from the NASA Goddard Earth Sciences, Data and Information Services Center (GES DISC; http://disc.sci.gsfc.nasa.gov). The early releases of the two main OMI products of NO\textsubscript{2}, one from NASA and the other from the Royal Netherlands Meteorological Institute (KNMI), showed large differences for some regions (Lamsal et al., 2010). This current version represents substantial OMI retrieval algorithm improvements (Boersma et al., 2011; Bucsela et al., 2013, and references therein) from its preceding version 1.0, so that it is now feasible to derive quantitative information about NO\textsubscript{2} emissions from large point sources (Streets et al., 2013). The current, refined retrieval algorithms of both research groups, though different in their approaches, now produce very similar columns.

Retrieval of tropospheric NO\textsubscript{2} columns involves (1) retrieval of NO\textsubscript{2} abundance along the viewing path (slant column) with a Differential Optical Absorption Spectroscopy (DOAS) fit (Platt, 1994) in the 405–465 nm wavelength range, (2) computation of an air mass factor (AMF) by integrating the relative vertical distribution (shape factors) of NO\textsubscript{2} weighted by altitude-dependent scattering weights for NO\textsubscript{2} (Palmer et al., 2001), (3) removal of cross-track artifacts (stripes) resulting from insufficient calibration in the OMI back-scattered reflectances, and (4) separation of stratospheric and tropospheric NO\textsubscript{2} components (Bucsela et al., 2013).

The tropospheric AMF is sensitive to the a priori NO\textsubscript{2} profile shape. The retrieval of the operational OMI NO\textsubscript{2} product uses NO\textsubscript{2} shape factors generated from the NASA Global Modeling Initiative (GMI; http://gmi.gsfc.nasa.gov/) CTM at 2.5° longitude × 2° latitude resolution grids. In this work, we use the NO\textsubscript{2} product discussed in Lamsal et al. (2013) that was generated with high resolution (0.67° longitude × 0.5° latitude) over the US. NO\textsubscript{2} shape factors were derived from a nested-grid GEOS-Chem CTM (http://acmg.seas.harvard.edu/geos/) simulation and scattering weights for NO\textsubscript{2}. Use of NO\textsubscript{2} shape factors from the nested simulation improves the representation of vertical distributions, including those of the elevated plumes of power plants (Lamsal et al., 2013). The errors in the individual pixel tropospheric NO\textsubscript{2} columns under clear-sky conditions are estimated to be 30% (Boersma et al., 2004).

The OMI tropospheric NO\textsubscript{2} columns agree with in situ and ground-based measurements within 20% (Lamsal et al., 2013; Bucsela et al., 2013). Individual clear-sky (i.e., cloud fraction < 0.3) data not affected by the so-called “row anomaly” (Dobber and Braak, 2010) were allocated by area-weights into 0.1° longitude × 0.1° latitude grids. The row anomaly is the result of a partial blockage of the field of view of the OMI, which lengthens the time necessary to obtain global coverage from one day to two days. For consistency over the OMI record, we restricted our analysis to scan positions 10–23 as they are unaffected by the row anomaly.

There are rarely ideal conditions for assessing emissions changes from power plants from space, so we used all available data, regardless of season. Lu and Streets (2012), and references therein, recommend using data only for summer (e.g., the policy-relevant ozone season of May–September) for a variety of reasons. For instance, the chemical lifetime of NO\textsubscript{x} tends to be shortest in summer, which has the advantage that a facility’s emissions are convolved less with NO\textsubscript{2} from other sources than in other seasons. However, there are disadvantages to using data only for the ozone season, such as the stratospheric contribution to the total NO\textsubscript{2} column, and the associated error, is seasonally greatest and important.

For the purposes of this study, it was not practical to restrict our analysis to the ozone season as many facilities, including some of the largest emitters, were already operating ECDs during the ozone season at the start of our study period, especially in the eastern US. In Section 3, we show that the relationship between a facility’s NO\textsubscript{x} emissions and the OMI NO\textsubscript{2} column over the power plant is much stronger in the southern US than in the northern US, where the
seasonal variation in the chemical lifetime of NO\textsubscript{x} is more pronounced. We discuss the implications of using all available data in Section 3.2.5.

2.2. Selection of power plants

We identified the top 100 highest-emitting power plants in 2005 based on the US national NO\textsubscript{x} emissions inventory (http://www.epa.gov/ttn/chief/eiinformation.html). If there is more than one power plant within a 0.4° longitude \(\times\) 0.4° latitude gridbox, we combined and treated them as one facility. Then, we used the Emission Database for Global Atmospheric Research version 4 (EDGAR v4; http://edgar.jrc.ec.europa.eu/), which is for 2005 and is available on a 0.1° longitude \(\times\) 0.1° latitude resolution grid, to select the power plants least affected by other industrial sources within a 0.4° longitude \(\times\) 0.4° latitude area around the facility. Lu and Streets (2012) found that the agreement between NO\textsubscript{2} columns and NO\textsubscript{x} emissions improves with increasing relative contribution of the power plant’s emissions to total NO\textsubscript{x} emissions from all sources within a NO\textsubscript{2} column gridbox; they refer to this quantity as \(f_{\text{power}}\). For this study, we required that \(f_{\text{power}} > 0.90\), which eliminated 45 of the top 100 highest-emitting power plants from our analysis. The locations of the facilities used in this analysis are shown in Fig. 1. Information for each power plant is given in Table 1.

The characteristics of a plume from a power plant depend on variations in meteorology (e.g., “plume meandering”; Beirle et al., 2011) and chemistry, so we used the maximum value of the plume for each overpass whether in the gridbox containing the facility or in adjacent gridboxes (i.e., a 0.3° longitude \(\times\) 0.3° latitude area) to create monthly averages. We found that the correlation between the change in emissions from a power plant and the concomitant change in the OMI NO\textsubscript{2} column is best for this coarse grid resolution (i.e., 0.1° longitude \(\times\) 0.1° latitude) as compared to more coarse resolutions (i.e., 0.25° longitude \(\times\) 0.25° latitude).

2.3. Definition of the response (\(\rho\))

In order to reflect the relative contributions of NO\textsubscript{x} emissions from a power plant to the total NO\textsubscript{x} emissions (and column), we define the following parameters: \(E^T\), \(E^{PP}\), and \(E^O\), which represent NO\textsubscript{x} emissions from all sources within a gridbox, the power plant, and sources other than the power plant, respectively, where \(E^T = E^{PP} + E^O\). Similarly, NO\textsubscript{2} \(E^{PP}\) and NO\textsubscript{2} \(E^O\) represent, respectively, the total NO\textsubscript{2} column within a gridbox, the portion of the column associated with the power plant, and the portion of the column associated with all other sources, including NO\textsubscript{2} advected into the gridbox, where NO\textsubscript{2} \(E^{PP}\) = NO\textsubscript{2} \(E^{PP}\) + NO\textsubscript{2} \(E^O\).

As defined in the introduction, the Response (\(\rho\)) is given by:

\[
\rho = \frac{\Delta \text{NO}_{2}^{PP}}{\Delta E^{PP}} = \frac{\left(\Delta \text{NO}_{2}^T - \Delta \text{NO}_{2}^O\right)}{\left(\Delta E^T - \Delta E^O\right)}
\]

(1)

where \(\Delta\) represents the change in NO\textsubscript{2} column or NO\textsubscript{x} emissions. Rearranging Equation (1) into linear form:

\[
\Delta \text{NO}_{2}^T = \rho \left(\Delta E^T - \Delta E^O\right) + \Delta \text{NO}_{2}^O
\]

(2)

where \(\rho\) is the slope of the line and \(\Delta \text{NO}_{2}^O\) is the y-intercept. In the ideal situation where \(\Delta E^O\) and \(\Delta \text{NO}_{2}^O\) equal zero, Equation (2) simplifies to:

\[
\Delta \text{NO}_{2}^T = \rho \Delta E^{PP}
\]

(3)

In this case, \(\Delta \text{NO}_{2}^O\), such as determined from OMI data, is solely due to \(\Delta E^{PP}\). \(\Delta E^{PP}\) in all figures and Table 1 in this manuscript is the sum of emissions for those days where OMI data are available. Consequently, \(\Delta E^{PP}\) is less than the total change in a facility’s emissions over a given time period.

Though Equations (1)–(3) are rather straightforward, \(\rho\) is a complicated parameter that is a function of the chemical lifetime of NO\textsubscript{x}, meteorology, and the factors that affect the partitioning of NO\textsubscript{x} into NO and NO\textsubscript{2} (e.g., Martin et al., 2003; Stavrakou et al., 2008; Beirle et al., 2011; Lamsal et al., 2011; Walter et al., 2012; Zhou et al., 2012). The dependence of \(\rho\) on these factors is discussed in the Supplemental Material. Accounting for the complexities of \(\rho\) requires a CTM, ideally with a plume-in-grid technique, to properly treat the evolution of a power plant’s plume. In Section 3, we show that this onerous step is not necessary for our practical application, particularly given the large uncertainties associated with the OMI data discussed in Sections 2.1 and 3.2. In practice, \(\rho\) is relatively stable.

Fig. 1. The locations of the facilities listed in Table 1 with the facility identifier beside each point. The magnitude of \(\Delta E^{PP}\) (kTon) is indicated by the size of the circle around the square indicating the facility location. The color of each square corresponds to the correlation \(r^2\) shown in Table 1. The horizontal dashed line indicates 36.5° N latitude, which is the boundary between the southern and northern US in Fig. 3. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
for each site so that $\Delta NO_2$ can be treated as linearly proportional to $EPP$ (e.g., Martin et al., 2003; Kaynak et al., 2009; Kim et al., 2009).

### 3. Results

Over the Aura record, 2005–2011, NO$_2$ emissions from electric power generation decreased by 48% in the US (http://www.epa.gov/tnn/chief/trends/index.html). The CEMS data indicate that there was a large (>50%) decrease between 2005 and 2011 in annual emissions at 22 power plants that we include in our study (Table 1), presumably because of the implementation of new ECDs. Emissions at most of the facilities decreased by >20%, while emissions at seven facilities did not change or increased. At many facilities, emissions decreased rapidly, but they decreased...
incrementally at others, which we know to be associated with, for instance, the implementation over time of ECDs on specific units within a facility.

3.1. Response of OMI NO₂ to the implementation of ECDs

Fig. 2 (left column) shows monthly total EPP and monthly mean NO₂ for several facilities over the OMI record (Figure S1 shows this information for all facilities listed in Table 1.). The Crystal River facility (ID #1; Fig. 2a) in Florida had the largest ΔEPP (70%; Table 1). The CEMS data show that emissions began decreasing rapidly during the installation of ECDs that came online in June 2009 (Unit 5) and May 2010 (Unit 4). There were concomitant decreases in NO₂ with a 37% overall reduction (Table 1). The correlation (r²) of the data is shown along with the slope (m) and y-intercept (b) of a line fit to the data. m is r as shown in Table 1. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Fig. 2. (left) Monthly mean NO₂ (black line; × 10¹⁵ molecules cm⁻²) and EPP (blue dotted line; kTon) data from 2005 to 2011 for four power plants. Vertical black lines represent the standard error of the mean of the OMI data. The sample size (N) is the number of days with data used to create monthly means. The annual mean NO₂ data are represented with a green line and the monthly median data as an open red diamond. (right) Annual mean NO₂ (× 10¹⁵ molecules cm⁻²) versus annual EPP (kTon) data. The red numbers represent the years that correspond to the annual means (e.g., "09" = 2009). The correlation (r²) of the data is shown along with the slope (m) and y-intercept (b) of a line fit to the data. m is r as shown in Table 1. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
3.2. Sources of variation in the response

Fig. 3 shows $\Delta E^{pp}$ and $\Delta NO_2$ (Table 1) for all power plants. Overall, the correlation ($r^2 = 0.31$) is weak and does not improve much when only facilities are considered where $E^{pp} > 4$ kTon in 2005. The poor correlation occurs whether the absolute or relative changes are considered. The correlation for the facilities in the southern US is better ($r^2 = 0.58; n = 25$) than in the northern US ($r^2 = 0.18; n = 30$; Fig. 3), though it is important to note that $\rho$ is very high at most individual facilities are generally scalar, including in the northern US (Figure S1) (There are too few facilities in the western US with which to draw any conclusion about a possible systematic bias between eastern and western facilities.). We chose 36.5°N latitude (shown in Fig. 1) to separate the northern and southern US as the range of the standard error of the means of the OMI data for the individual facilities is 0.1–0.4 below this latitude and 0.1–0.8 above this latitude (Fig. 3; Table 1). In the following subsections, we discuss the sources of variation of $\rho$ for the power plants, including those that cause the differences between facilities in the northern and southern US.

3.2.1. Magnitude of emissions reduction

For most facilities, we found that $\rho$ is scalar for all intents and purposes and $\Delta NO_2$ is well correlated with $\Delta E^{pp}$ given that $\Delta E^{pp}$ was large (Fig. 1; Table 1), which is consistent with the findings of Lu and Streets (2012). The correlations ($r^2$) between annual NO$_2$ and $E^{pp}$ (Table 1) are $>0.5$ at 32 of the 55 facilities and, not surprisingly, rise linearly with increasing $\Delta E^{pp}$, which will be discussed further in Section 3.2.6.

As with all satellite data, it is important to consider the issue of the signal-to-noise ratio (SNR). For our purposes, this means that the SNR increases with the magnitude of $E^{pp}$. Some of the facilities in Table 1 had relatively small annual emissions in 2005 so that meteorological variations and large changes in regional NO$_2$ levels, for instance, may obscure their $\rho$'s. Not all the power plants in our study used ECDs and some had relatively small variations in annual emissions. We included these facilities to help us understand what factors influence $\rho$.

3.2.2. Retrieval issues

Errors are introduced into the retrieval during the conversion of the measured OMI slant column to a more useful vertical column using a tropospheric AMF, a complex function of information on a priori NO$_2$ profile shapes, surface albedo, clouds, aerosols (not implicitly accounted for), etc. (e.g., Boersma et al., 2011). The use of coarsely-resolved retrieval parameters (e.g., NO$_2$ profile shapes, surface albedo) could introduce large errors in retrievals at places where these parameters have large spatial variability (Zhou et al., 2009; Boersma et al., 2011), such as in mountainous and desert areas in the western US. For example, the emissions remained relatively stable over our study period at the Four Corners/San Juan facility (ID #53) in New Mexico, the facility with one of the highest annual emissions (Fig. 2c). Though new ECDs were not installed, the year-to-year variation in $E^{pp}$ was larger at this facility than $\Delta E^{pp}$.

Fig. 3. (top) $\Delta E^{pp}$ (kTon) as compared to $\Delta NO_2$ (x 10$^{15}$ molecules cm$^{-2}$) as the mean of 2005 and 2006 minus the mean of 2010 and 2011 (Table 1). The colored dots indicate the magnitude of the mean $E^{pp}$ of 2005 and 2006. The number associated with each point corresponds to a particular power plant identified in Table 1. $n$ is sample size (i.e., the number of power plants) used in the correlation statistic ($r$) and line fit, where $m$ is the slope and $b$ is the $y$-intercept. (middle) The same as (top), but for only those facilities at latitudes $>36.5$°N. The horizontal lines represent the standard error of the means of the OMI data. (bottom) The same as (middle), but for only those facilities at latitudes $<36.5$°N.

$ho$ at the Bowen facility (ID #8) in Georgia is similar (0.35) to the Crystal River facility, though the correlation is somewhat lower ($r^2 = 0.75$; Fig. 2b). ECDs were operated at Bowen during the ozone season through 2008, but year-round afterward (Fig. 2b; left column). Overall, $E^{pp}$ decreased by 70% from 2005 to 2011 with a corresponding decrease in NO$_2$ of 30%. Although this facility is generally upwind of the Atlanta metropolitan area, the NO$_2$ column is likely influenced to some degree by this urban source, depending on meteorology and season, which may explain the scatter in $E^{pp}$ and NO$_2$ in Fig. 2b (right column). Nevertheless, the impact of year-round ECDs on NO$_2$ is clear from the beginning of 2009.

We found that $\rho$'s are $<1$ at all but two of the facilities (Table 1). There is a wide range of values, but there is no clustering, such as with latitude. In the next section, we discuss sources of variation of $\rho$ among the facilities.
for some smaller facilities that implemented ECDs. However, the correlation ($r^2 = 0.18$) associated with $\rho$ is weak, which may result from the parameters used in the AMF as the facility is located in the desert (i.e., high surface reflectivities) and near mountains characterized by variable snow cover.

3.2.3. Statistical significance

The number of individual days with OMI data (i.e., sample size $N$ in Fig. 2 and S1) is typically $< 10$ month$^{-1}$, so that the standard error of the mean is oftentimes large. In these situations, the monthly average is not statistically significant. This issue is compounded for power plants at higher latitudes or elevations, such as the Big Stone (ID #33; Figure S1) facility, as OMI data are well filtered for snow cover. At this facility, 23 months have $< 3$ days of data with which to create the monthly average (Table 1), so that the annual snow cover. At this facility, 23 months have $< 3$ days of data with which to create the monthly average (Table 1), so that the annual

3.2.4. Proximity to urban sources

At the Big Bend facility (ID #10) near Tampa, Florida, ECDs were brought online in 2008 (Unit 3), 2009 (Unit 2), and 2010 (Unit 1), decreasing emissions by 77%. Similar to $\rho$ (0.28) at the Crystal River facility (ID #1), which is also in Florida, $\rho$ (0.53) at the Big Bend facility is scalar, but twice as high; $\text{NO}_2$ and $E \text{PP}$ are well correlated ($r^2 = 0.89$). $\Delta \text{NO}_2$ at the Crystal River facility is smaller than that at the Big Bend facility despite $\Delta \text{PP}$ being larger for the Crystal River facility. Due to proximity, the urban plume of Tampa influenced $\text{NO}_2$ at the Big Bend facility (not shown), particularly in the earlier years of our study period. From 2005 to 2011, the OMI data indicate that $\text{NO}_2$ over Tampa decreased by more than 50% ($\sim 2.5 \times 10^{15}$ molecules cm$^{-2}$), which, coupled with the large $\Delta \text{PP}$, explains the larger $\rho$ as compared to the Crystal River facility. That is, $\rho$ for the Big Bend facility is convoluted with the large decrease of $\text{NO}_2$ in the urban plume of Tampa (i.e., $\Delta \text{NO}_2$). For facilities near large emitters, including cities, the OMI data could be filtered by wind direction to minimize the influence of these other sources.

3.2.5. Seasonal variation

The influence of the seasonal cycle in $\text{NO}_2$ associated with variations in temperature and sunlight is readily apparent in Fig. 2b at the Bowen facility (ID #8) and at numerous other facilities (Figure S1). At facilities, such as Paradise (ID #3), New Madrid (ID #4), and Gibson (ID #9), the seasonal cycles in $\text{NO}_2$ continue even after the ECDs were routinely used year-round. It is worth noting that the correlation of monthly $\text{NO}_2$ and $E \text{PP}$ may be artifi-

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3.2.5. Seasonal variation

The influence of the seasonal cycle in $\text{NO}_2$ associated with variations in temperature and sunlight is readily apparent in Fig. 2b at the Bowen facility (ID #8) and at numerous other facilities (Figure S1). At facilities, such as Paradise (ID #3), New Madrid (ID #4), and Gibson (ID #9), the seasonal cycles in $\text{NO}_2$ continue even after the ECDs were routinely used year-round. It is worth noting that the correlation of monthly $\text{NO}_2$ and $E \text{PP}$ may be artifici-

Fig. 4. Percent change of the annual mean OMI NO2 (dashed lines) and NO2 from AQS surface sites (solid lines) relative to 2005 for the whole US ("All US") and four quadrants ("NE" = northeast; "SE" = southeast; "NW" = northwest; "SW" = southwest). In total, 517 AQS sites are included. The OMI data were sampled for the 0.1° × 0.1° gridboxes in which the AQS sites lay. We used all hourly AQS data to estimate the percent change.

To understand the seasonal variability, we calculated $\rho$ for each of the four seasons for each facility. In general, there is significant variability ($>50\%$) in the seasonal $\rho$'s for the typical facility, particularly ones at higher latitudes. At the Crystal River facility (ID #1) in Florida, the seasonal $\rho$'s are similar (i.e., within $\sim 30\%$), which is not surprising given the plant’s southerly location. The seasonal $\rho$'s show more variation ($\sim 50\%$) at the nearby Big Bend facility (ID #10), though this facility is impacted by the urban plume of Tampa as discussed in Section 3.2.4. On the other hand, there is considerably more variability in the seasonal $\rho$'s at the Cardinal/W. H. Sammis facility (ID #2) in Ohio and the New Madrid facility (ID #4) in Missouri, which are both located at higher latitudes than the Crystal River facility and in areas with higher regional NO2 levels.

In general, the seasonal $\rho$'s for spring, summer and fall tend to be more similar for a typical facility with the seasonal $\rho$ for winter being the outlier. An exception is that the seasonal $\rho$'s for summer are less meaningful at facilities in which ECDs were used during the ozone season over the entire study period because of the low emissions and, subsequently, low SNR of the OMI data. In addition, the magnitude of $\text{NO}_2$ is seasonally lowest in summer as the chemical lifetime is seasonally shortest. One would expect that seasonal $\rho$'s for spring and fall are similar at a given facility because of the similar chemical lifetimes in these two seasons. Generally, this is the case, particularly for the high-emitting facilities.

The seasonal $\rho$'s for winter tend to show considerable variability because of the latitudinal-dependence of the seasonal variation of the chemical lifetime and because of missing data as discussed in Section 3.2.3. In addition, the regional NO2 levels are seasonally highest in winter because the chemical lifetime is seasonally longest, so that the ratio of NO2 from the power plant relative to the regional level is seasonally lowest at many facilities.

The methodology that we present in this manuscript to assess the impact of the implementation of an ECD on the NO2 level above a facility may be tailored for specific applications. For instance, one
may decide to exclude the winter season from the analysis because of issues associated with NO\textsubscript{x} lifetime and statistical significance as discussed above. We repeated our analysis, excluding the winter season, and found that the conclusions of our study remain unchanged. The correlations of DEPP and D\textsubscript{NO\textsubscript{2}}T changed modestly for most facilities, though we did not find an improvement in the scatter observed in Fig. 3 as indicated by the correlation statistic (r). r\textsuperscript{2} for the northern US decreased from 0.18 to 0.07 and remained the same for the southern US. Based on the discussion in this section, it is important to understand that the value of r will depend on the choice of months used in the analysis, particularly in regions with seasonal variation in the NO\textsubscript{x} lifetime.

3.2.6. Variations in regional NO\textsubscript{2} levels (\Delta NO\textsubscript{2})

Annual mean surface concentrations of NO\textsubscript{2} decreased 33% nationally between 2001 and 2010 (EPA, 2012), primarily from decreases in emissions from fuel combustion in electrical utilities and vehicles (e.g., McDonald et al., 2012). From 2005 to 2011, the combined NO\textsubscript{2} emissions reduction from electrical utilities and vehicles was about 37\% (http://www.epa.gov/ttn/chief/trends/index.html) with two-thirds of the decrease being attributed to the reduction in the mobile source. The OMI data confirm that regional NO\textsubscript{2} levels decreased substantially in many areas of the US during our study period (e.g., Russell et al., 2012). Fig. 4 shows the percent change relative to 2005 of EPA’s Air Quality Monitoring System (AQS; http://www.epa.gov/ttn/airs/airsaqs/) NO\textsubscript{2} data and the corresponding OMI data above the AQS stations. The data are averaged over the whole US and over four quadrants. The reductions by 2009 range from 20 to 30\% for the AQS sites as grouped in the quadrants, but 35–40\% for OMI data; the northwest quadrant is an outlier. However, the overall shapes of the trends in both datasets are similar. The discrepancy in the magnitudes of the trends of the AQS and OMI data may occur as the OMI detects changes in NO\textsubscript{2} throughout the whole troposphere, while monitors at the AQS sites sample near-surface air. Thus, the OMI detects the reductions in NO\textsubscript{2} associated with both mobile and power plant sources, while the AQS surface monitors preferentially sample reductions in mobile sources as power plant plumes are located aloft predominately.

Though the chemical lifetime of NO\textsubscript{x} is relatively short, NO\textsubscript{x} emissions upwind influence NO\textsubscript{2} columns downwind (e.g., Turner et al., 2012), which can lead to elevated regional NO\textsubscript{2} levels (As discussed in Section 2.2 for our analysis, we selected facilities least affected by other sources within a 0.4° longitude × 0.4° latitude area around the facility (i.e., f\textsubscript{power} > 0.90)). Over our study period, some of the largest changes in regional levels occurred in the heavily populated region extending from Washington, DC to New York City (i.e., the Northeast Corridor), and the industrialized Ohio River Valley, where eight of the power plants selected for this study are located; the Chalk Point facility (ID #20) is the only facility that met our criterion (i.e., f\textsubscript{power} > 0.90) for selection in the Northeast Corridor. Fig. 5 shows that regional NO\textsubscript{2} levels decreased by 30–40\% from 2005 to 2011 in both of these regions, though the absolute decrease was much higher in the Northeast Corridor. Most of the power plants in Table 1 are located in areas with lower regional NO\textsubscript{2} levels in 2005 than in the Northeast Corridor and Ohio River Valley.
Fig. 6 shows the relationship between $\Delta E_{\text{PP}}$ and $\rho$. For power plants with $\Delta E_{\text{PP}} > 2$ kTon, $\Delta E_{\text{PP}}$ is generally large relative to the change in the regional NO$_2$ level. $\rho$'s for these facilities are between 0.12 and 0.62 with a mean of 0.36. For facilities with $\Delta E_{\text{PP}} < 2$ kTon, there is a wider range of $\rho$'s (i.e., between ~0.91 and 1.08), indicating that a change in the regional NO$_2$ level, if large, can influence $\rho$ in a non-negligible way. We attempted to find a method for removing the influence of a change in the regional NO$_2$ level in a general way applicable to all facilities. However, we found that the change in the regional NO$_2$ level can vary widely (e.g., with meteorological variability), requiring careful processing of the data for each facility. As an example, NO$_2$'s were high at the nearby facilities of White Bluff (ID #46) and Dolet Hills (ID #25) in the winters of 2009–10 and 2010–11 (Figure 51), which we found to be caused by stagnant meteorological conditions that allowed regional NO$_2$ levels to build.

4. Summary

We conclude that it is practical to use OMI NO$_2$ tropospheric column data to assess changes of emissions from power plants that are associated with the implementation of emission control devices (ECDs), though careful interpretation of the data is necessary. We showed that there is a clear response for OMI NO$_2$ data to NOX emission reductions from power plants associated with the implementation of ECDs on both monthly and annual time scales. This response is scalar for all intents and purposes, whether the reduction is rapid or incremental over several years. However, the response is variable among the power plants, even those with the greatest absolute decrease in emissions. We discussed some of the causes of this variability, which include the magnitude of a facility’s NOX emissions, seasonal variation of the NO$_2$ lifetime, proximity to urban areas, changes in the regional NO$_2$ levels, lack of statistical significance, and retrieval issues. Ideally, one should use a CTM to account for several of these causes of variability, though this would limit the practical application of space-based data for air quality purposes because of computational expense. However, we show that this step is not necessary if the change in the facility’s NO$_2$ emissions is large.

Using space-based NO$_2$ columns to assess changes in power plant NOX emissions will likely become more quantitative as the OMI retrieval procedure continues to evolve, such as through the use of improved and finely-resolved information of surface parameters. In addition, two planned sensors promise enhanced capabilities as compared to OMI: i) the European Space Agency Tropospheric Ozone Monitoring Instrument (TROPOMI; http://www.knmi.nl/samenw/tropomi/instrument), an OMI follow-on instrument with finer horizontal resolution, and ii) the NASA Tropospheric Emissions: Monitoring of Pollution (TEMPO; http://science.nasa.gov/missions/tempo) instrument, an OMI-like instrument that will be in geostationary orbit, collecting data throughout the day as opposed to one overpass per day as with OMI.

Many of the facilities included in our study were already using ECDs during the ozone season before the start of the OMI data record. A next step would be to repeat our analysis over the SCIA-Team (AQAST) program. We acknowledge the free use of 1) tropospheric NO$_2$ column data from the Aura OMI, 2) NO$_2$ emissions data from the US EPA, and 3) EDGAR data, which is maintained as a joint project of the European Commission Joint Research Centre (JRC) and the Netherlands Environmental Assessment Agency (PBL).

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2013.08.068.

References


Lamsal et al., 2013. Evaluation of Improved Operational Standard Tropospheric NO$_2$ Retrievals from Ozone Monitoring Instrument Using In Situ Surface-based NO$_2$ Observations in preparation.


