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Abstract

Emissions of nitrogen oxides (NO$_x$) and, subsequently, atmospheric levels of nitrogen dioxide (NO$_2$) have decreased over the U.S. due to a combination of environmental policies and technological change. Consequently, NO$_2$ levels have decreased by 30–40% in the last decade. We quantify NO$_2$ trends (2005–2013) over the U.S. using surface measurements from the U.S. Environmental Protection Agency (EPA) Air Quality System (AQS) and an improved tropospheric NO$_2$ vertical column density (VCD) data product from the Ozone Monitoring Instrument (OMI) on the Aura satellite. We demonstrate that the current OMI NO$_2$ algorithm is of sufficient maturity to allow a favorable correspondence of trends and variations in OMI and AQS data. Our trend model accounts for the non-linear dependence of the NO$_2$ concentration on emissions associated with the seasonal variation of the chemical lifetime, including the change in the amplitude of the seasonal cycle associated with

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Preprint submitted to Atmospheric Environment February 2, 2015

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the significant change in NO\textsubscript{x} emissions that occurred over the last decade. The direct relationship between observations and emissions becomes more robust when one accounts for these non-linear dependencies. We improve the OMI NO\textsubscript{2} standard retrieval algorithm and, subsequently, the data product by using monthly vertical concentration profiles, a required algorithm input, from a high-resolution chemistry and transport model (CTM) simulation with varying emissions (2005–2013). The impact of neglecting the time-dependence of the profiles leads to errors in trend estimation, particularly in regions where emissions have changed substantially. For example, we find that by including the time-dependency there are 18% more instances of significant trends and up to 15% larger total NO\textsubscript{2} reduction. Using a CTM, we explore the theoretical relation of the trends estimated from NO\textsubscript{2} VCDs to those estimated from ground-level concentrations. The model-simulated trends in VCDs strongly correlate with those estimated from surface concentrations ($r = 0.83$, $N = 355$). We then explore the observed correspondence of trends estimated from OMI and AQS data. We find a significant, but slightly weaker, correspondence (i.e., $r = 0.68$, $N = 208$) than predicted by the model and discuss some of the important factors affecting the relationship, including known problems (e.g., NO\textsubscript{2} interferents) associated with the AQS data. This significant correspondence gives confidence in trend and surface concentration estimates from OMI VCDs for locations, such as the majority of the U.S. and globe, that are not covered by surface monitoring networks. Using our improved trend model and our enhanced OMI data product, we find that both OMI and AQS data show substantial downward trends from 2005 to 2013, with an average reduction of 38% for each over
the U.S. The annual reduction rates inferred from OMI and AQS measurements are larger (-5.0%/yr, -3.7%/yr) from 2005 to 2008 than 2010 to 2013 (-1.6%/yr, -2.8%/yr). We quantify NO\textsubscript{2} trends for major U.S. cities and power plants; the latter suggest the largest trend (-4%/yr) by 2008 and small or insignificant changes during 2010–2013.

Keywords: Nitrogen dioxide, troposphere, air quality, trend, Aura OMI

1. Introduction

Emissions of nitrogen oxides (NO\textsubscript{x} = NO + NO\textsubscript{2}) are regulated by the U.S. Environmental Protection Agency (EPA), because NO\textsubscript{x} contributes to the formation of unhealthy levels of surface ozone, a pollutant that has been long known to damage lung tissue when inhaled (Kleinfield et al., 1957; Challen et al., 1958). Though nitrogen dioxide (NO\textsubscript{2}) is a respiratory irritant, its levels in the U.S. are currently below the National Ambient Air Quality Standard (NAAQS) set by EPA (EPA, 2008). As a result of emission reductions from mobile and point sources (e.g., McDonald et al., 2012; Xing et al., 2013), surface NO\textsubscript{2} levels in the U.S. decreased by 33% between 2001 and 2010 and, subsequently, ozone concentrations decreased by 14%, when year-to-year variations in meteorology are taken into account (EPA, 2012). The temporal evolution of this substantial decrease in NO\textsubscript{2} has been recorded by the Ozone Monitoring Instrument (OMI), which was launched on the NASA Aura satellite in July 2004 (Figure 1). In this manuscript, we show that recent modifications to the OMI retrieval algorithm have sufficiently improved the quality of the data product so that trends and variations derived from OMI and EPA Air Quality System (AQS) surface data are similar for
2005 2013

0.1 0.9 1.8 2.7 3.6 4.5 5.4 6.3 \times 10^{15} [\text{molec. cm}^{-2}]

Figure 1: Annual average OMI tropospheric NO$_2$ VCDs at 1/2° latitude × 2/3° longitude spatial resolution for 2005 (left) and 2013 (right).

U.S. cities.

Tropospheric vertical column density (VCD) data of NO$_2$, as measured from space, serve as an effective proxy for surface NO$_2$ in many air quality applications. (The VCD is defined as the number of molecules of an atmospheric gas between the satellite instrument and the Earth’s surface per unit area.) For instance, VCD data are used for inferring surface NO$_x$ emissions (Leue et al., 2001; Martin et al., 2003; Jaeglé et al., 2005; Wang et al., 2007; Boersma et al., 2008a; Napelenok et al., 2008; Zhao and Wang, 2009; Lin et al., 2010; Lamsal et al., 2011; Streets et al., 2013; Tang et al., 2013; Ghude et al., 2013; Vinken et al., 2014), estimating trends and variations in atmospheric concentrations (Beirle et al., 2003; Richter et al., 2005; Frost et al., 2006; Boersma et al., 2008b; Lin and McElroy, 2011; Zhou et al., 2012; Castellanos and Boersma, 2012; Russell et al., 2012), monitoring emission changes in point sources (e.g., Kim et al., 2006; Wang et al., 2012; Duncan
et al., 2013), and inferring ozone formation sensitivities to NO$_x$ and VOCs levels (Martin et al., 2004; Duncan et al., 2010; Valin et al., 2013; Tang et al., 2014). Duncan et al. (2013) demonstrated that variations and trends in OMI NO$_2$ data near US power plants correlate well with changes in emissions reported by the Continuous Emissions Monitoring System (CEMS) for large facilities, particularly if they are located away from cities. Lamsal et al. (2008, 2010) used an early version of OMI NO$_2$ data to infer ground-level concentrations, such as those measured by the EPA AQS network, over a range of locations and seasons. They found that their OMI-derived surface concentrations, estimated using a chemistry and transport model (CTM), were significantly correlated with AQS observations, both temporally and spatially, though the correlations varied widely between monitoring stations.

The EPA AQS network of surface monitors is sparse and unevenly distributed, including in major metropolitan areas. The network lacks observations for large regions of the U.S. Because the lifetime of NO$_x$ is short, trends estimated from these monitoring data may not be spatially representative, confounding NO$_2$ trend estimates derived from these surface data. Moreover, NO$_2$ trends estimated from the commonly used chemiluminescent monitor (equipped with molybdenum oxide converter) data may differ from true NO$_2$ trends because of interference from the oxidation products of NO$_x$ (NO$_2$), such as peroxyacetyl nitrate (PAN), alkyl nitrates, and nitric acid (HNO$_3$) (Winer et al., 1974; Dunlea et al., 2007; Steinbacher et al., 2007; Lamsal et al., 2008). Satellite observations, which have the advantage of spatial coverage, provide information on NO$_2$-specific trends, which complement and enrich the AQS-observed trends. Here, we take advantage of three indepen-
dent measurements from OMI, a photolytic converter, and a molybdenum
converter to explore (1) how trends derived from OMI measurements, which
are collocated once per day, relate to trends available from high temporal res-
olution (hourly) surface data, and (2) how the interference in molybdenum
converter measurements could affect the actual observed NO\textsubscript{2} trend.

In this manuscript, we conduct a systematic investigation of the relation-
ship between AQS and OMI NO\textsubscript{2} trends; to our knowledge, such a trend
estimation and comparison have not been shown before. In Section 2, we de-
scribe our method for estimating trends, our new high-resolution OMI VCD
data product and the surface data that we use in our analysis. We also
present a model study of the expected correspondence of trends derived from
VCD and surface concentration data and discuss the various factors that
should be considered when comparing these trends. We discuss the observed
correspondence of trends derived from OMI VCD and AQS data in Section 3.
We summarize our conclusions in Section 4.

2. Observations, model and methods

2.1. Multivariate linear regression for trend estimation

We use a regression model to infer both the seasonal and the linear trend
components in OMI and AQS NO\textsubscript{2} observations. The time series of monthly
average NO\textsubscript{2} values (Ω) can be assumed to be comprised of three additive
subcomponents: a time dependent seasonal component (α), a linear trend
component (β), and residual or noise (R) component:

\[ Ω(t) = α(t) + β(t) + R(t), \quad (1) \]
where \( t \) represents time (month). The time dependent regression component \( (\alpha) \) is given by a constant plus intra-annual sine and cosine harmonic series (Randel and Cobb, 1994):

\[
\alpha(t) = c_0 + \sum_{j=1}^{3} (c_{1j} \sin\left(\frac{2\pi j t}{12}\right) + c_{2j} \cos\left(\frac{2\pi j t}{12}\right)),
\]

where \( c_0, c_{1j}, \) and \( c_{2j} \) are constant coefficients to be determined from the measurements. The major portion of the \( \text{NO}_2 \) annual cycle is explained by the seasonal variation of the \( \text{NO}_x \) lifetime. The contributions from all other factors, such as monthly variation in \( \text{NO}_x \) emissions, to the seasonal cycle are typically small. The seasonal pattern may be constant or evolve in time.

Analysis of the temporal evolution of the seasonal variation of OMI \( \text{NO}_2 \) over the eastern U.S. reveals a considerable change (decrease) in seasonal amplitude over the Aura record, suggesting the need of an approach that accounts for it when calculating the trend and estimating trends in emissions. Changes in the seasonal amplitude are also reported by Hilboll et al. (2013) and occur at places where \( \text{NO}_x \) emissions are changing rapidly due to economic growth or emission control measures. Removal of varying seasonal components in a time series can be achieved by a locally weighted regression smoothing technique (Cleveland et al., 1990) or by including a scaling factor (Hilboll et al., 2013). In line with these methods, we identify and extract seasonal and linear trend components by exploiting changes in the measured seasonal pattern (amplitude and phase) for individual years. For each year, \( Y \), we fit a regression line using monthly observations from that year itself plus 6 monthly observations from years adjacent to \( Y \). This provides a series of local regression lines, which incorporate explicit time dependence.
Comparison of local regression lines with high- and low-amplitude regression lines allows identification and isolation of two seasonal terms ($\alpha_1$, $\alpha_2$, where $\alpha = \alpha_1 + \alpha_2$ in Eq. 1 and $\alpha_1$ represents the regression line with the lowest seasonal amplitude) and the linear trend ($\beta$). A graphical illustration of applying the multiple regression analysis to OMI data is shown in Figure A1 in Appendix A.

2.2. The expected correspondence of NO$_2$ VCDs and surface concentrations

As mentioned in Section 1, the primary advantage of satellite data for estimating trends is spatial coverage, especially for areas without surface monitors or ones where the existing monitors do not provide NO$_2$ levels or trends that are representative of an urban area as discussed in Section 2.4.3. In addition, a VCD has the advantage over surface data that it also includes NO$_2$ above the surface, which is necessary for estimating variations and trends in NO$_x$ levels and emissions from all sources, including power plants where emissions are released well above the surface (i.e., tall smokestacks and plume rise).

We use the NASA Global Modeling Initiative (GMI) CTM, which is described in Appendix B, to examine how the trend in surface level NO$_2$ observed by AQS relates to the trend in OMI VCD data. We examine the observed correspondence in Section 3.1. The simulation includes annually varying anthropogenic emissions and captures both the spatial distribution and temporal changes observed by OMI over the continental U.S. (Strode et al., 2014).

We apply the multivariate linear regression analysis, described in Section 2.1, to the model output to calculate the linear trend component ($\beta$)
Figure 2: Model simulated reduction (%, 2005–2010) in surface NO$_2$ versus reduction in tropospheric NO$_2$ VCDs. Reductions in VCDs are color-coded by tropospheric NO$_2$ VCDs for 2005. The results are from a GMI simulation over the U.S., and include only areas with annual average tropospheric NO$_2$ VCDs $> 1 \times 10^{15}$ molecules cm$^{-2}$ and statistically significant trends at the 95% confidence level. The dotted line represents the 1:1 relationship.

in surface concentrations and tropospheric VCDs. Figure 2 compares the reductions (calculated from $\beta$) for 2005–2010 in surface NO$_2$ with those in tropospheric NO$_2$ VCDs over all polluted areas in the U.S. The trend in surface concentrations is well correlated ($r = 0.83$, $N = 355$) with the trend in VCDs, though, as expected, the scatter is higher for lower VCDs as the contribution of surface NO$_2$ to the tropospheric VCD is less than in polluted areas. The reductions in surface concentrations range from 6.4 to 42 %, while tropospheric VCD reductions range from 8.6 to 41 %. Reductions are higher over power plants in the tropospheric VCDs than the surface concentrations.
because of tall smokestacks and plume rise associated with these sources.
The average reductions for the two quantities are in agreement to within 4%, but their correlation improves with increasing NO\textsubscript{2} levels (Figure 2).

2.3. Description of OMI observations and recent improvements relevant for U.S. air quality applications

2.3.1. Retrievals of tropospheric NO\textsubscript{2} VCD

The OMI monitors NO\textsubscript{2} VCDs by measuring spectral variation in backscattered solar radiation in the broad visible spectral window between 405 nm and 465 nm. OMI measurements are made in early afternoon (i.e., local time of 13:00–14:45) with a spatial resolution of 13 × 24 km\textsuperscript{2} at nadir and with nearly daily global coverage.

In this work, we further improve the operational OMI NO\textsubscript{2} retrieval algorithm (NASA standard Product, version 2.1 see AppendixC), by using new a priori NO\textsubscript{2} profiles simulated by the GMI CTM with year-specific emissions. The profiles not only improve the representation of the NO\textsubscript{2} vertical distribution, but also capture the yearly changes in NO\textsubscript{2} profile shapes. The latter is critical due to rapid decline in the U.S. NO\textsubscript{x} emissions in recent years (e.g., Figure 1), as NO\textsubscript{2} retrievals and, therefore, the estimated trends are sensitive to the vertical shape of NO\textsubscript{2} profiles as discussed in Section 2.3.2.

We use individual pixel clear sky (cloud radiance fraction < 0.5) Level 2 OMI NO\textsubscript{2} data. The estimated errors in individual tropospheric NO\textsubscript{2} VCDs observed under polluted (tropospheric NO\textsubscript{2} VCD > 1 × 10\textsuperscript{15} molecules cm\textsuperscript{-2}) and clear sky conditions are ∼30% (Bucsela et al., 2013). To exclude the cross-track rows affected by the row anomaly after 2007 and to avoid inconsistent sampling, we use OMI data for rows 5–23 for the entire OMI dataset.
The largest pixels that are at swath edges (rows 1–4) were excluded to reduce spatial smearing of tropospheric NO$_2$. We then map the remaining data into a 0.1° latitude × 0.1° longitude grid by calculating an area-weighted average. The approach accounts for the overlap and size of OMI ground footprints (pixels). Finally, we compute monthly averages to estimate trends for the entire OMI period (2005–2013).

2.3.2. Impact on the OMI-derived trend of the assumption of the vertical NO$_2$ distribution in the retrieval algorithm

Satellite retrieval algorithms of NO$_2$ VCDs are sensitive to the assumed vertical distribution of NO$_2$ (Martin et al., 2002; Boersma et al., 2004; Heckel et al., 2011; Russell et al., 2011; Hains et al., 2010; Lamsal et al., 2014). Most operational NO$_2$ retrieval algorithms assume an a priori vertical distribution, such as those taken from a coarse-resolution CTM (e.g. Boersma et al., 2011; Hilboll et al., 2013; Bucsela et al., 2013). Since the model simulations often use outdated bottom-up emissions, these profiles may not capture the actual vertical distribution of NO$_2$, especially where anthropogenic NO$_x$ emissions are undergoing rapid changes (e.g., Figure 1). For instance, the operational OMI retrieval algorithm (NASA standard product, version 2.1) uses a climatology of a priori NO$_2$ profiles at 2° latitude × 2.5° longitude horizontal resolution from a GMI CTM simulation based on emissions from the 1999 National Emission Inventory (NEI). Here, we explore how the current practices of operational algorithms employing NO$_2$ profiles based on constant and/or outdated emissions affect satellite-derived NO$_2$ trends.

As discussed in Section 2.3.1, we performed two separate retrievals, one using monthly NO$_2$ vertical profiles based on emissions and meteorology of
2005 profiles
Year-specific profiles

Figure 3: Percent reduction in OMI tropospheric NO$_2$ VCDs for 2005–2010 (1/2° latitude × 2/3° longitude horizontal resolution) calculated from two separate retrievals, one using 2005 (left) and another using year-specific (middle) monthly mean NO$_2$ vertical profiles. The dark gray color represents the locations with insignificant trends at 95% confidence. (right) The difference between trends calculated from the retrievals based on 2005 profiles minus the retrievals based on year-specific profiles. Values are color-coded by average tropospheric NO$_2$ VCDs for 2005.

2005, which we refer to as “2005 profiles”, and another using monthly NO$_2$ profiles based on year-specific emissions and meteorology, which we refer to as “year-specific profiles”, to demonstrate the impact of vertical profile assumptions on the OMI-derived NO$_2$ trends. Interannual variations in the meteorological fields are expected to have a minor effect on monthly average NO$_2$ data. We use OMI pixels with cloud radiance fraction < 0.5, calculate area-weighted average VCDs on a 1/2° latitude × 2/3° longitude horizontal grid, and use Eq. 1 to derive linear trends for 2005–2010.

Figure 3 shows the spatial variation of NO$_2$ reduction for 2005–2010 calculated from the two retrievals. Although the reductions from both retrievals are highly consistent with one another ($r = 0.97$, $N = 3524$), the year-specific retrievals offer a considerably higher number (18%) of cases with significant
trends and up to 15% larger reductions. Therefore, using 2005 profiles in
the retrievals underestimates the trends, on average, by 0.6%/yr and overall
2005–2010 reduction by 3.5%. The smaller deviations for larger VCDs imply
that the trend is less sensitive to the vertical profile assumption in highly
polluted areas. Since the vertical profiles used in the generation of OMI op-
erational products are based on NEI 1999, we anticipate that annual trends
estimated using these products would be biased low.

2.4. Description of in situ surface NO\textsubscript{2} measurements and considerations for
trend estimation

2.4.1. AQS surface measurements

We use hourly NO\textsubscript{2} measurements from the EPA AQS monitoring net-
work (Demerjian, 2000). The network employs the EPA-designated NO\textsubscript{2}
chemiluminescence automated Federal Reference Method (FRM) described
in detail in EPA (1975). The NO\textsubscript{2} measurement method of these commercial
instruments relies on detecting NO by reducing NO\textsubscript{2} to NO on the surface of
a heated molybdenum oxide (MoO\textsubscript{x}) substrate at 300–400°C. However, the
reduction of NO\textsubscript{2} to NO by the MoO\textsubscript{x} substrate is not specific to NO\textsubscript{2}, but is
also sensitive to some unknown fraction of NO\textsubscript{z} (NO\textsubscript{z}′) (Winer et al., 1974;
Grosjean and Harrison, 1985; Demerjian, 2000; EPA, 2006; Dunlea et al.,
2007; Steinbacher et al., 2007; Lamsal et al., 2008). The magnitude of the
total interference is variable, and depends not only on the relative fraction of
actual NO\textsubscript{2} to total reactive nitrogen compounds (NO\textsubscript{y} = NO\textsubscript{x} + NO\textsubscript{z}), but
also on the characteristics of individual monitors. As a result, NO\textsubscript{2} measure-
ments from AQS monitors are consistently biased high (up to 50%), with
the highest biases in the afternoon as discussed in Section 2.4.2, at distant
locations from NO\textsubscript{x} sources, and during summer months when these NO\textsubscript{x} products are expected to peak (e.g., Steinbacher et al., 2007). The Aura overpass is once per day in early afternoon when this bias is near its daily maximum.

To relate OMI with AQS observations, we select measurements collocated in space and time. For overlapping OMI overpasses for a given day, we compute averages weighted by OMI ground pixel sizes. The hourly data from AQS are averaged over a 2-hour period (13:00–15:00 local time) to temporally match with the OMI measurements. We exclude stations that offer measurements only during the summer ozone season or ceased operation anytime between 2005 and 2013. These criteria retain 208 sites, including 30 in rural, 89 in suburban, and 88 in urban environments. We also use the hourly data from these sites to examine the diurnal variation in surface NO\textsubscript{2} trends in Section 4.

2.4.2. Interferences in the AQS-observed trends

Here we examine how the trend estimated from the biased AQS NO\textsubscript{2} data relates to the actual NO\textsubscript{2} trend. We use hourly NO\textsubscript{2} observations made by collocated MoO\textsubscript{x} and more accurate photolytic converter instruments at a rural site in Yorkville, Georgia, which is located 72 km to the northwest of Atlanta. The photolytic converter instrument employs broadband photolysis of ambient NO\textsubscript{2} followed by chemiluminescence detection of the product, NO, offering a true NO\textsubscript{2} measurement with accuracy better than 10\% (Kley and McFarland, 1980; Ryerson et al., 2000; Zellweger et al., 2000; Pollack et al., 2010). Long-term photolytic converter measurements are available from the SouthEastern Aerosol Research and Characterization (SEARCH)
Figure 4: (top) Hourly averaged surface NO\textsubscript{2} mixing ratios for 2005 (red) and 2010 (blue) measured by collocated AQS molybdenum (dotted line open circles) and SEARCH photolytic (solid line with closed circles) converter analyzers at Yorkville, GA. (middle) Hourly variations of the ratio of the molybdenum and photolytic measurements for 2005 and 2010. (bottom) Diurnal changes in the NO\textsubscript{2} reductions for 2005–2010 calculated from the photolytic and molybdenum converter measurements.

network (Edgerton et al., 2006). We use the data for the period 2005–2010 to calculate annual averages and monthly trends.

Figure 4 (top) shows the diurnal variation of annually averaged NO\textsubscript{2} measured by the MoO\textsubscript{x} converter and photolytic converter instruments. The two measurements agree to within 5\% between 8 PM and 8 AM in 2005. Significant differences of 8–43\% are observed during the day with the largest difference at 2–3 PM, near the OMI overpass time. The difference is a result
of the diurnal changes in the relative contribution of HNO$_3$, PAN, and alkyl nitrates to NO$_x$' (Dunlea et al., 2007; Steinbacher et al., 2007; Lamsal et al., 2008). In 2010, the biases increase, relative to 2005, not only in the early and mid afternoon hours (22–54%), but also in early morning (7–13%) and late afternoon (11–16%), suggesting that the NO$_x$' interferences in the MoO$_x$ converter measurements have grown relative to NO$_2$ as NO$_2$ concentrations have decreased in recent years. This result is not surprising given that the formation of individual NO$_x$ species is dependent on the NO$_x$ concentration (e.g., Duncan and Chameides, 1998). The bottom row of Figure 4 shows NO$_2$ reductions for 2005–2010 calculated from the two datasets using the same trend model (Eq. 1). Reductions in true NO$_2$ offered by the photolytic converter measurements are on average 8.6% higher than those calculated from the molybdenum converter data.

The magnitude of NO$_x$', as well as the deviation of AQS-derived trends from the actual NO$_2$ trends, are site specific and may not be identical to the results from the case study presented here. They depend on several factors, including local meteorological conditions, speciation of local NO$_x$, the distance between the site and emission sources (i.e., the degree of photochemical aging), and characteristics of individual monitors. As a result, the AQS-derived trends may differ from the actual NO$_2$ trends by a variable and non-negligible amount. In fact, the results shown in Figure 4 imply that any trend derived from AQS data would include a spurious trend introduced as the NO$_2$ to NO$_x$' ratio changes over time with NO$_x$ emission reductions. We anticipate that impact of the NO$_x$' interferents will be less for AQS monitors at highly polluted sites, where NO$_2$ makes up a larger fraction of NO$_y$, and
greater at background sites, where NO₂ dominates.

2.4.3. Spatial heterogeneity in NO₂ and implications for trends estimated from sparse surface stations

There are obvious difficulties in relating local measurements from AQS with satellite observations. For instance, NO₂ is a short-lived species, so it is concentrated near combustion sources and, consequently, there can be considerable spatial heterogeneity in the NO₂ field near sources that is not resolved in the relatively spatially-coarse OMI measurements. Therefore, the AQS-derived NO₂ trends may reflect highly localized trends (e.g., near a highway) that cannot be captured in OMI trends. Thus, interpretation of the differences between the AQS- and OMI-derived trends at individual AQS sites is difficult, and is not expected to always show high correlations.

To illustrate this issue, we calculate trends using OMI data at 0.1° latitude
$\times 0.1^\circ$ longitude spatial resolution for Dallas, TX. Figure 5 shows the spatial structure in the total decrease in OMI NO$_2$ levels from 2005 to 2013. In an absolute sense, the largest decreases occurred where NO$_2$ levels are highest, but in a relative sense, some of the largest changes are away from the city center. For instance, there is a large relative decrease south of Fort Worth, which is collocated with a major manufacturing area. Trends estimated from the AQS monitors and OMI are similar at the majority of sites, but they exhibit large discrepancies at some locations. These differences could arise from preferential placement of monitors (e.g., next to highways), spatial variation of source characteristics, and interference in surface NO$_2$ measurements as discussed in Section 2.4.2. Therefore, trends derived from AQS data from a specific city may not reflect area-wide average trends because the lack of representativeness of those monitors (e.g., too few for a statistically significant sample size). However, when averaged across all areas in this analysis, we expect the trends to be robust as discussed in Section 3.

3. Comparison between OMI and AQS trends over the Aura record (2005–2013)

In this section, we compare variations in the OMI tropospheric NO$_2$ VCDs and AQS surface measurements, and explore the correspondence of trends estimated from the two datasets.

3.1. Trends for individual stations

Figure 6 shows the annual average OMI tropospheric VCDs (at individual AQS sites) and AQS surface mixing ratios for 2005 and 2013. Surface measurements are sparse and unevenly distributed with most sites being located
in polluted regions. Both OMI and surface measurements exhibit broad similarities in both spatial distribution ($r = 0.76$, $N = 208$) and temporal changes.

The total reductions from 2005 to 2013 (estimated from the linear trend, $\beta$) at individual sites generally range from 7.1% to 64% for OMI tropospheric NO$_2$ VCDs, and from 3.2% to 68% for AQS measurements. However, the average reductions at all sites are 38% for both the OMI and AQS datasets.

As shown in Figure 7, the overall scatter is larger for the observed reductions in surface AQS data and VCDs as compared to what is predicted from the model simulation (Figure 2). This is not surprising given the measurement and sampling uncertainties that affect the trends derived from the OMI and AQS observations as discussed in Section 2. The correlation between the two
Figure 7: Scatter plot of the reductions derived from surface concentrations from individual AQS sites versus collocated OMI tropospheric VCDs in absolute values (left column) and percent (right column). Symbols indicate land use type: circles for urban, squares for suburban, and triangles for rural sites. Values are color coded by OMI tropospheric NO₂ VCDs for 2005. The dotted line represents the 1:1 relationship.

trends is significant \((r = 0.68, N = 208)\).

3.2. Trends by regions and EPA categories

Tables 1–3 show comparisons of trends from OMI and AQS data grouped by regions, land type, and land use. Regional reductions from both datasets are in the range of 37–47 %, with the largest reductions in southern California (Table 1). In Tables 2 and 3, we compare the trend in OMI and AQS measurements according to EPA’s classification of the monitoring sites by land type and land use, respectively. Average reductions calculated from OMI and AQS measurements for all land uses and types range from 35 % to 43 %, and agree to within 3 % with the exception of land type designated as mobile. Since there are few sites in this category and the classification
Table 1: Average Reduction by Regions.

<table>
<thead>
<tr>
<th>Region</th>
<th>Domain</th>
<th>Number of sites</th>
<th>NO₂ reduction (%)</th>
<th>AQS</th>
<th>OMI</th>
</tr>
</thead>
<tbody>
<tr>
<td>New England</td>
<td>41–45°N, 70–75°W</td>
<td>13</td>
<td>38.3</td>
<td>37.9</td>
<td></td>
</tr>
<tr>
<td>Mid-Atlantic</td>
<td>36–41°N, 72–81°W</td>
<td>19</td>
<td>41.4</td>
<td>43.1</td>
<td></td>
</tr>
<tr>
<td>S. California</td>
<td>31–36°N, 116–122°W</td>
<td>50</td>
<td>42.8</td>
<td>47.2</td>
<td></td>
</tr>
<tr>
<td>Central Valley</td>
<td>36–41°N, 118–124°W</td>
<td>30</td>
<td>37.2</td>
<td>41.2</td>
<td></td>
</tr>
</tbody>
</table>

Table 2: Average Reduction at AQS Sites Grouped by Land Types.

<table>
<thead>
<tr>
<th>Land type</th>
<th>Number of sites</th>
<th>NO₂ reduction (%)</th>
<th>AQS</th>
<th>OMI</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mobile</td>
<td>6</td>
<td>34.9</td>
<td>43.1</td>
<td></td>
</tr>
<tr>
<td>Industrial</td>
<td>15</td>
<td>37.2</td>
<td>34.7</td>
<td></td>
</tr>
<tr>
<td>Agriculture</td>
<td>19</td>
<td>35.7</td>
<td>38.7</td>
<td></td>
</tr>
<tr>
<td>Commercial</td>
<td>74</td>
<td>39.5</td>
<td>37.0</td>
<td></td>
</tr>
<tr>
<td>Residential</td>
<td>88</td>
<td>37.9</td>
<td>40.3</td>
<td></td>
</tr>
</tbody>
</table>

Table 3: Average Reduction at AQS Sites Grouped by Land Uses.

<table>
<thead>
<tr>
<th>Land use</th>
<th>Number of sites</th>
<th>NO₂ reduction (%)</th>
<th>AQS</th>
<th>OMI</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rural</td>
<td>30</td>
<td>35.1</td>
<td>35.5</td>
<td></td>
</tr>
<tr>
<td>Suburban</td>
<td>89</td>
<td>39.0</td>
<td>40.1</td>
<td></td>
</tr>
<tr>
<td>Urban and center city</td>
<td>88</td>
<td>37.6</td>
<td>37.2</td>
<td></td>
</tr>
</tbody>
</table>
Figure 8: Annual trend (%/yr) for 2005–2008 (top row) and 2010–2013 (bottom row) for OMI data (1/2° latitude × 2/3° longitude horizontal resolution; left column), OMI data over AQS sites (middle column), and AQS data (right column). The dark gray color represents locations with insignificant trends at the 95% confidence level.

...does not provide information on local sources and their characteristics, it is difficult to interpret the somewhat larger difference.

3.3. Variations in the pace of NO₂ reductions

To examine variations in the pace of NO₂ reductions, we analyze the time series for $\beta$ for two time periods, 2005–2008 and 2010–2013. The majority of the decrease in OMI NO₂ levels occurred in the earlier period with less of a decrease or even a slight increase in the latter period as discussed in Russell et al. (2012). Figure 8 shows the spatial variation of the annual trend in OMI tropospheric NO₂ VCDs for the 2005–2008 and 2010–2013 periods. There are significant decreasing trends over large areas of the U.S. during 2005–2008 with an annual reduction rate of 0–12%. Reductions are more rapid...
in many populated areas of the U.S.; trends in sparsely populated areas
are small or insignificant. The trends are substantially weaker during 2010–
2013. Figure 8 compares the trends observed by OMI and AQS monitors.
The spatial distribution as well as the magnitude of the two trends for both
periods are generally consistent, but are less so as compared to the entire
2005–2013 period. OMI observations suggest a stronger average decrease of
5%/yr as compared to 3.7%/yr from AQS data for 2005–2008, whereas the
AQS data suggest a larger reduction of 2.8%/yr as compared to 1.6%/yr

There are two main reasons for the change in the pace of reductions.
First, emission control devices (ECDs) were installed on many power plants
by 2008, reducing their NO\textsubscript{x} emissions and, subsequently, the OMI NO\textsubscript{2}
levels above the facilities dramatically (Duncan et al., 2013). The larger
decrease seen in the eastern U.S. resulted from the 2005 Clean Air Interstate
Rule (CAIR) for 27 eastern states with the goal to decrease NO\textsubscript{x} emissions
from power plants. Second, U.S. emissions decreased in 2008 due to the
global economic downturn. OMI NO\textsubscript{2} levels changed little in many areas
of the country in the latter period as the U.S. economy slowly recovered
(Bishop and Stedman, 2014; Tong et al., 2014), but, at the same time, the
fleet of light-duty vehicles continued to become more fuel efficient and less
polluting (e.g. Bishop and Stedman, 2008; Russell et al., 2012) to meet the
more stringent Tier 2 standards of the Clean Air Act Amendments of 1990.

3.4. OMI NO\textsubscript{2} trends over major U.S. metro areas and power plants

In this section, we present the reductions in OMI NO\textsubscript{2} VCDs for 20 U.S.
metropolitan areas and over 150 of the largest U.S. power plants.
3.4.1. Trends over 20 metropolitan areas

As shown in Figures 6 and 8, the largest percent decreases occurred in heavily populated, urban areas, which are collocated with the highest emissions. Table 4 shows the total reduction (%) in OMI NO$_2$ levels for 20 metropolitan areas for the period, 2005 to 2013, and the annual trends (%/yr) for the two sub-periods discussed in the previous section. Levels decreased by $>40\%$ in 8 of the 20 cities, including New York City, Los Angeles, and Philadelphia, 30–40\% in 6 cities, and 20–30\% in 6 cities. In all cities, the major decrease in levels occurred in the earlier period, 2005–2008, with small or insignificant changes in the latter period, 2010–2013.

3.4.2. Trends over 150 power plants

Over the OMI record, 2005–2013, U.S. NO$_2$ emissions from power plants decreased by about 52\% (http://www.epa.gov/ttn/chief/trends/index).
Table 4: Annual Average OMI Tropospheric NO$_2$ VCD in 2005 and 2013, Total Reduction (%) for 2005–2013, and Annual Trend (%/yr) for 20 U.S. Metropolitan Areas.

<table>
<thead>
<tr>
<th>Cities</th>
<th>Tropospheric NO$_2$ VCD (10$^{15}$ molec. cm$^{-2}$)</th>
<th>2005–2013 reduction (%)</th>
<th>Annual trend (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>New York</td>
<td>16.5</td>
<td>11.2</td>
<td>40.9</td>
</tr>
<tr>
<td>Los Angeles</td>
<td>20.5</td>
<td>11.8</td>
<td>43.6</td>
</tr>
<tr>
<td>Chicago</td>
<td>10.8</td>
<td>8.8</td>
<td>29.7</td>
</tr>
<tr>
<td>Dallas</td>
<td>5.7</td>
<td>3.8</td>
<td>38.5</td>
</tr>
<tr>
<td>Houston</td>
<td>8.5</td>
<td>5.2</td>
<td>39.4</td>
</tr>
<tr>
<td>Philadelphia</td>
<td>10.9</td>
<td>6.7</td>
<td>47.1</td>
</tr>
<tr>
<td>Washington DC</td>
<td>9.6</td>
<td>4.7</td>
<td>48.6</td>
</tr>
<tr>
<td>Miami</td>
<td>4.3</td>
<td>3.3</td>
<td>23.0</td>
</tr>
<tr>
<td>Atlanta</td>
<td>9.2</td>
<td>4.5</td>
<td>46.6</td>
</tr>
<tr>
<td>Boston</td>
<td>7.1</td>
<td>5.0</td>
<td>35.7</td>
</tr>
<tr>
<td>San Francisco</td>
<td>5.0</td>
<td>4.0</td>
<td>30.2</td>
</tr>
<tr>
<td>Phoenix</td>
<td>6.8</td>
<td>4.0</td>
<td>46.6</td>
</tr>
<tr>
<td>Detroit</td>
<td>9.4</td>
<td>5.2</td>
<td>25.1</td>
</tr>
<tr>
<td>Seattle</td>
<td>5.7</td>
<td>3.9</td>
<td>25.5</td>
</tr>
<tr>
<td>Minneapolis</td>
<td>4.9</td>
<td>3.1</td>
<td>23.4</td>
</tr>
<tr>
<td>San Diego</td>
<td>5.8</td>
<td>3.9</td>
<td>36.2</td>
</tr>
<tr>
<td>Tampa</td>
<td>5.0</td>
<td>2.8</td>
<td>47.0</td>
</tr>
<tr>
<td>St. Louis</td>
<td>5.8</td>
<td>3.9</td>
<td>31.5</td>
</tr>
<tr>
<td>Baltimore</td>
<td>8.6</td>
<td>5.4</td>
<td>40.2</td>
</tr>
<tr>
<td>Denver</td>
<td>7.0</td>
<td>5.6</td>
<td>26.3</td>
</tr>
</tbody>
</table>
Duncan et al. (2013) found that changes in OMI NO$_2$ levels near 55 power plants are consistent with changes in each facility’s emissions as reported to the CEMS and that the implementation of ECDs was clear in the OMI data for the majority of facilities. Figure 9 shows the annual change (%/yr) in OMI NO$_2$ levels over 150 of the largest power plants in the U.S. The largest decreases occurred in the eastern U.S. and by 2008, which is consistent with compliance with the provisions of the CAIR. On average, OMI NO$_2$ levels decreased by 4.0%/yr during the 2005–2008 period and by 0.6%/yr during the 2010–2013 period. The number of power plants exhibiting positive trends increased from 10 during 2005–2008 to 59 during 2010–2013, although the trends are either small or insignificant (Figure 9).

4. Discussion and conclusions

Satellite data are currently being used in many air quality applications (e.g., Duncan et al., 2014) and uniquely have the advantage of spatial coverage as compared to sparse surface networks. While the satellite data do not currently provide nose-level concentrations, they are of sufficient maturity that they do deliver important quantitative information on the trends and variations in pollutants important to the air quality community. For instance, Duncan et al. (2013) showed that trends and variations in OMI NO$_2$ data above U.S. power plants agree well with trends and variations in their emissions reported to CEMS, particularly for large facilities. In this manuscript, we show the good correspondence of trends and variations in OMI NO$_2$ VCD data to another common air quality quantity, surface concentration from the EPA AQS network of surface air quality monitors.
Linear trends (2005–2013), estimated from OMI NO$_2$ VCD data, compare well with those from AQS data when grouped by region, land use, and land type. We found that there are an insufficient number of surface NO$_2$ monitors (e.g., 0–2) in most cities to estimate an overall trend in a given metropolitan area. The under-representativeness of an individual monitor for a metropolitan region is highly dependent on site location, particularly those located near major pollution sources (e.g., highways). Nevertheless, the reductions (2005–2013) estimated from AQS data compare favorably for most metropolitan areas as discussed below.

Several issues need to be considered when comparing trends and spatial and temporal variations estimated from OMI NO$_2$ VCD data to those estimated from surface data. The VCD data are influenced by NO$_2$ levels in the free troposphere and the relatively coarse spatial resolution of the data may not allow for the VCD data to reflect the variations and trends near individual surface stations, particularly for those stations sited near major pollution sources. However, the satellite data provide greater spatial coverage as compared to the relative sparse monitoring network. In addition, AQS monitors measure species other than NO$_2$, such as some fraction of NO$_x$ ($= \text{HNO}_3 + \text{PAN} + \text{alkyl nitrates}$), where the NO$_x$ composition is typically unknown. Comparison of the trends inferred from collocated photolytic and molybdenum converter measurements suggests that the true NO$_2$ trends are likely greater than those computed from AQS data.

Despite the limitations of both satellite and surface data, they complement each other and the correlation of the variations in the datasets is relatively high for some metropolitan areas. For example, Figure 10 shows the
Figure 10: The normalized residual term, $R$, from Eq. 1 for four U.S. metropolitan areas. The correlation coefficients ($r$) and number of AQS monitors ($N$) are given for each city. The OMI data are sampled at the same locations as the AQS data.

The normalized residual term, $R$, from Eq. 1. $R$ reflects variations between the two datasets not explained by the seasonal cycle ($\alpha$) nor the linear trend ($\beta$) regression terms. It incorporates fluctuations in NO$_2$ associated with month-to-month variations in weather, but it also includes variations associated with the limitations of the two datasets (e.g., NO$_2$ interferents for AQS data). We will continue to work to understand the sources of some of the stronger deviations in $R$ between the two datasets, such as those that occurred in late 2009 in both Houston and Dallas (Figure 10).

In addition to understanding the sources of differences between the two datasets, work is ongoing to further refine and tailor the OMI NO$_2$ retrieval.
Figure 11: Diurnal changes in NO$_2$ reductions calculated from 2-hour average NO$_2$ mixing ratios (circles) in rural (blue), suburban (green), and urban (orange) EPA AQS sites. Dotted line represents average value for all land types. Values shown are the reductions from 2005 to 2013. Diamonds represent reductions in OMI tropospheric NO$_2$ VCDs. The bars represent the 1$\sigma$ variability of the average.

Algorithm for air quality applications. In this manuscript, for instance, we showed that, by capturing the general trend in the vertical profile of NO$_2$ concentrations, the comparison between the satellite and surface data improves, particularly in less polluted areas. Our ongoing retrieval algorithm development consists of improved retrievals of SCDs (Marchenko et al., 2014), use of high-resolution MODIS surface reflectivity together with explicit aerosol corrections in the AMF calculation, and coupled NO$_2$ and cloud retrievals, and these improvements should improve the quality of OMI NO$_2$ data and their scientific applications.

As a final comment, the Aura satellite overpasses a given location once a day in early afternoon local time, which limits the data’s usefulness, such as for estimating the diurnal variation of emissions from mobile and point
sources. Concentrations of NO$_2$ in urban areas undergo strong diurnal variations with higher values during the night when photolysis ceases to convert NO$_2$ to NO, a trace gas that is a member of the NO$_x$ family that cannot be measured from space, and the boundary layer height shrinks, trapping fresh NO$_x$ emissions near the surface, particularly in early morning during rushhour. For our purpose of estimating NO$_2$ trends, we found that there is little variation in the reductions estimated from AQS data during daylight hours and the reductions estimated from OMI data. Figure 11 indicates that the reductions during the night are generally lower than during the day (i.e., 27% at 5–6 AM to 38% at 3–4 PM), which should be considered when inferring changes in emissions, such as mobile sources, from the OMI data.

There are two upcoming satellite missions that are relevant for air quality applications using NO$_2$ VCD data, including the application presented in this manuscript. Both instruments will provide data similar to the OMI, but with improved capabilities. The NASA Tropospheric Emissions: Monitoring of Pollution (TEMPO) instrument (Chance et al., 2013) will be in geostationary orbit, continuously observing the U.S. during daylight hours; the satellite’s orbital period will match the Earth’s rotational period, so the satellite will appear to be motionless to an observer on the Earth’s surface. This orbit will improve the signal-to-noise ratio of the data and allow for the estimation of NO$_2$ trends and emissions throughout the day. The European Space Agency’s (ESA) Tropospheric Ozone Monitoring Instrument (TROPOMI, (Veefkind et al., 2012)) will be launched in 2016 and have a similar once daily overpass time as OMI, but its pixels will have finer spatial resolution, which will allow for better detection of smaller emissions sources and likely improve the
Appendix A. Graphical illustration of the multivariate regression analysis

Figure A1 illustrates the variation of the four subcomponents with time from OMI tropospheric NO$_2$ columns over the eastern U.S. In this example, seasonal variation and linear trend contribute, respectively, 74% and 11% of the total variance. The residual contains the impact of short-term variations (e.g. transport) in NO$_2$ columns that were not captured by the regression model, and represents about 12% of the total variance. The original and modeled time series constructed from seasonal and trend components are highly correlated ($r = 0.92$, $N = 108$), suggesting that the approach offers a simple yet robust seasonal and trend estimates.

Appendix B. The GMI model

The GMI CTM is driven by meteorology from the Modern Era Retrospective-Analysis for Research and Applications (MERRA) (Rienecker et al., 2011), which reasonably reproduces observed weather for the Aura record (2005–2013). The simulation is performed at 1° latitude × 1.25° longitude horizontal resolution and with 72 vertical levels extending from the surface to 0.01 hPa. About 33 levels are in the troposphere, including 8 levels below 1 km. The model output is sampled at the OMI overpass time for a self-consistent comparison with the observational dataset.

We use a simulation for 2005–2010 that uses monthly and annually varying emissions of CO, NO$_x$, and non-methane hydrocarbons. Anthropogenic
Figure A1: Time series of monthly OMI tropospheric NO$_2$ columns (Ω, $10^{15}$ molecules cm$^{-2}$) over the eastern U.S. (36–40°N, 70–75°W) separated into four subcomponents: seasonal cycle and change in seasonal amplitude ($α = α_1 + α_2$), long-term linear trend ($β$), and residuals (R). The line in the top row represents the summation of the $α$ and $β$ subcomponents, and the filled circles represent the actual OMI data.
emissions are based on the EDGAR3.2 Inventory (Olivier et al., 2005), over-witted by regional inventories: The EPA NEI 2005 inventory (http://www.epa.gov/ttnchie1/net/2005inventory.html) over the U.S., CAC (https://www.ec.gc.ca) over Canada, BRAVO over Mexico (Kuhns et al., 2005), the European Monitoring and Evaluation Programme (EMEP, http://www.emep.int) over Europe, and inventory from Zhang et al. (2009) over Asia. The anthropogenic emissions prior to 2006 are scaled applying annual scaling factors from the GEOS-Chem model (van Donkelaar et al., 2008). For 2007–2010, the U.S. and European emissions are scaled on a country-wide basis using the national emission totals from EPA and EMEP, respectively. The REAS inventory projections (Ohara et al., 2007) are used to scale the Asian anthropogenic emissions for 2007–2009. Soil NO\textsubscript{x} emissions are computed online with temperature and precipitation dependence ((Yienger and Levy, 1995). Lightning NO\textsubscript{x} emissions are calculated following Allen et al. (2010).

Appendix C. Details of the OMI retrieval algorithm

We use the OMI standard product, OMNO2 (version 2.1), which is public-licy available from the NASA Goddard Earth Sciences Data Active Archive Center (GES DISC, http://disc.sci.gsfc.nasa.gov). This data version includes significant updates and improvements over previous versions (Bucsela et al., 2006; Celarier et al., 2008; Lamsal et al., 2008, 2010). Detailed descriptions of the current algorithm and assessments for the standard OMI NO\textsubscript{2} product are given in Bucsela et al. (2013) and Lamsal et al. (2014), respectively. In brief, the algorithm retrieves NO\textsubscript{2} slant column densities (SCDs) with the Differential Optical Absorption Spectroscopy (DOAS) tech-
nique (Platt, 1994) in the visible region (405–465 nm). This is followed by
computation of air mass factors (AMFs) by integrating relative vertical dis-
tribution (shape factors) of NO$_2$ weighted by altitude dependent scattering
weight factors. The NO$_2$ vertical profiles are simulated by the GMI CTM
(Strahan et al., 2007; Duncan et al., 2007) at 2° latitude × 2.5° longitude hor-
izontal resolution for the time of the OMI measurement. Scattering weights
are computed as a function of reflectivity and pressure of cloud and terrain,
and viewing geometry with a radiative transfer model. Stratospheric AMFs
and retrieved SCDs from five consecutive orbits over clean regions (30°S–5°N)
are used to correct for cross-track biases (stripes) in SCDs resulting from cal-
ilbration errors. Stratospheric NO$_2$ fields needed to derive tropospheric NO$_2$
VCDs are determined by box-car smoothing and interpolation of OMI NO$_2$
VCDs estimates, after accounting for a priori tropospheric NO$_2$ VCDs over
unpolluted or cloudy areas. Comparison of OMI tropospheric NO$_2$ VCDs
with a suite of in situ and remote sensing measurements suggests agreement
within ±20% for clear-sky conditions (Lamsal et al., 2014).

Acknowledgements

The work was supported by the NASA Air Quality Applied Science Team
(AQAST) and NASA’s Earth Science Directorate Atmospheric Composition
Programs.

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