



OMI NO₂ column densities over North American urban cities

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OMI NO₂ column densities over North American urban cities: the effect of satellite footprint resolution

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Abstract

Nitrogen dioxide vertical column density (NO_2 VCD) measurements via satellite are compared with a fine-scale regional chemistry transport model, using a new approach that considers varying satellite footprint sizes. Space-borne NO_2 VCD measurement has been used as a proxy for surface nitrogen oxide (NO_x) emission, especially for anthropogenic urban emission, so accurate comparison of satellite and modeled NO_2 VCD is important in determining the future direction of NO_x emission policy. The National Aeronautics and Space Administration Ozone Monitoring Instrument (OMI) NO_2 VCD measurements, retrieved by the Royal Netherlands Meteorological Institute (KNMI), are compared with a 12 km Community Multi-scale Air Quality (CMAQ) simulation from the National Oceanic and Atmospheric Administration. We found that OMI footprint pixel sizes are too coarse to resolve urban NO_2 plumes, resulting in a possible underestimation in the urban core and overestimation outside. In order to quantify this effect of resolution geometry, we have made two estimates. First, we constructed pseudo-OMI data using fine-scale outputs of the model simulation. Assuming the fine-scale model output is a true measurement, we then collected real OMI footprint coverages and performed conservative spatial regridding to generate a set of fake OMI pixels out of fine-scale model outputs. When compared to the original data, the pseudo-OMI data clearly showed smoothed signals over urban locations, resulting in roughly 20–30% underestimation over major cities. Second, we further conducted conservative downscaling of OMI NO_2 VCD using spatial information from the fine-scale model to adjust the spatial distribution, and also applied Averaging Kernel (AK) information to adjust the vertical structure. Four-way comparisons were conducted between OMI with and without downscaling and CMAQ with and without AK information. Results show that OMI and CMAQ NO_2 VCDs show the best agreement when both downscaling and AK methods are applied, with correlation coefficient $R = 0.89$. This study suggests that satellite footprint sizes might have a considerable effect on the measurement of fine-scale urban NO_2 plumes. The impact of satellite footprint resolution should be

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considered when using satellite observations in emission policy making, and the new downscaling approach can provide a reference uncertainty for the use of satellite NO₂ measurements over most cities.

1 Introduction

Tropospheric nitrogen dioxide, NO₂, is an important component of urban atmospheric chemistry. It is one of the major pollutants affecting humans and the biosphere (Chauhan et al., 2003; Kampa and Castanas, 2008), and works as an important precursor in tropospheric ozone chemistry and aerosol formation. Continuous monitoring of tropospheric NO₂ is important to understand urban air quality and changes in anthropogenic emissions. NO₂ is also used as an important indicator for traffic and urbanization (Rijnders et al., 2001; Ross et al., 2006; Studinicka et al., 1997).

Tropospheric NO₂ has been measured from space since the mid-1990s; the Global Ozone Monitoring Experiment (GOME, 1996–2003, onboard the European Remote Sensing-2), Scanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY, 2002–2012, onboard ENVISAT), Ozone Monitoring Instrument (OMI, 2004–present, onboard Aura), and GOME-2 (2007–present, onboard MetOp-A and 2013–present on MetOp-B) have all been used for the detection of NO_x emission from natural and anthropogenic sources (Beirle et al., 2004; Boersma et al., 2007; Kim et al., 2006, 2009; Konovalov et al., 2006; Lamsal et al., 2008; Martin et al., 2003; Napelenok et al., 2008; Richter et al., 2005; van der A et al., 2006, 2008)

NO₂ plumes from urban anthropogenic sources, especially from point and mobile sources, usually have a fine structure, as small as a few hundred meters and as large as 10–20 km, as reported in comparisons of column NO₂ based on in situ observations and modeled calculations (Heue et al., 2008; Valin et al., 2011; Ryerson et al., 2001). Heue et al. (2008) used an airborne instrument based on imaging Differential Optical Absorption Spectroscopy (iDOAS) to build a two-dimensional distribution model of urban plumes. By comparing NO₂ column densities over the industrialized South African

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data in order to quantify the impact from pure differences in geometry. Second, we extend the basic concept of Hilboll et al. (2013) to apply spatial-distribution information from the fine-scale model to the OMI measurements, and demonstrate how the new approach adjusts the original OMI measurements. Satellite and model data are described in Sect. 2. Construction of pseudo-OMI data and the quantification of the impact of pixel geometry are discussed in Sect. 3. In Sect. 4, the downscaling approach is discussed; Sect. 5 concludes and discusses the implications of findings for emission policy decision-making.

2 Data

OMI: We utilized OMI tropospheric NO₂ VCD data, retrieved by the Royal Netherlands Meteorological Institute (KNMI). The OMI instrument, onboard NASA's Earth Observing System Aura satellite, is a nadir-viewing imaging spectrograph measuring backscattered solar radiation with a measuring wavelength ranging from 270 to 500 nm and with a spectral resolution of about 0.5 nm. Its telescope has a 114° viewing angle, which corresponds to a 2600 km-wide swath on the surface. In its normal global operation mode, its pixel size is 13 km (along) × 24 km (across) at nadir, which can be reduced to 13 km × 12 km in zoom mode (Levelt et al., 2006). Data were downloaded from the European Space Agency's (ESA) Tropospheric Emission Monitoring Internet Service (TEMIS; <http://www.temis.nl/airpollution/no2.html>). DOMINO version 2.0 retrieval based on the Differential Optical Absorption Spectroscopy (DOAS) technique was used for the study. We disregarded data pixels with cloud fractions over 40% or other contaminated pixels using quality flags. Details on the NO₂ column retrieval algorithms and error analysis are described in Boersma et al. (2004, 2007).

NAQFC: The US National Air Quality Forecast Capability (NAQFC) provides daily ground-level ozone predictions using the Weather Forecasting and Research non-hydrostatic mesoscale model (WRF-NMM) and CMAQ framework across the CONUS with a 12 km resolution domain (Chai et al., 2013; Eder et al., 2009). In our analy-

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sis, we used the experimental version of NAQFC, which uses WRF-NMM with B-grid (NMMB) as a meteorological driver and the CB05 chemical mechanism. Meteorological data is processed using the PREMAQ, which is a special version of the Meteorology–Chemistry Interface Processor (MCIP) designed for the NAQFC system.

3 Construction of pseudo-OMI data

OMI footprint pixel size increases as the viewing angle deviates from the nadir direction to the edge of swaths. Figure 1 shows the actual size distributions of OMI pixels collected during September 2013. The blue line indicates size distribution counts for each 50 km^2 bin, while the red line indicates the cumulative distribution of the OMI pixel sizes. The size distribution has high occurrences near 300 km^2 , as expected from the OMI's resolution at the nadir (that is, $13 \times 24 = 312$). However, many pixels still have larger sizes; around half of total pixels are larger than 500 km^2 , and 20 % of total pixels are larger even than 1000 km^2 . Geographical coverage rapidly increases with pixel size, so deciding a threshold for footprint pixel sizes and available coverage may present a serious dilemma.

Figure 2 shows the relationship between OMI footprint pixel size and actual geographical coverage over the Contiguous United States (CONUS). With 1 July 2011 data, 25 % of OMI pixel sizes are less than 342 km^2 , and they cover 1.4 % of the CONUS domain. CONUS coverage changes to 11.5, 24.0, and 58.8 % when 50, 75, and 100 % of OMI pixels are used, respectively. Using only finer data may provide detailed information, but they represent only a small part of all the data. If we also use coarser-resolution data, they provide more coverage but tend to be biased over areas with spatial gradient, as discussed in the previously mentioned studies (Hilboll et al., 2013). We therefore estimated the theoretical range of biases deriving from this geometric effect by constructing a pseudo-OMI data set out of a fine-scale model. Using the fine-scale regional CMAQ simulations and assuming this model represents a true

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areas. This effect is also prominent in locations with small but strong NO_x emission sources, such as power plants or small cities such as Norfolk, VA. It should be noted that these discrepancies result from purely geometric effects deriving from OMI's designed pixel sizes and are around $\pm 5\text{--}10 \times 10^{15} \text{ \# cm}^{-2}$, with 20–30 % underestimation or overestimation biases for major cities and more than 100 % under- or overestimation for local cities like Norfolk and Richmond, VA. In the next section, we introduce a new approach – the conservative downscaling method – to reduce this effect of resolution due to varying OMI footprint pixel sizes.

4 OMI NO_2 VCD downscaling

As described in the previous section, urban NO_2 plumes usually have too fine of a spatial structure compared to OMI's measuring footprints. In this section, we introduce a new approach for adjusting those geometric effects. Downscaling is a common concept in meteorological simulations, used especially in global circulation models to provide initial and boundary conditions for regional models. We use a similar concept, describing a downscaling method in data processing as a special case of spatial re-gridding that provides further details through the incorporation of additional information into a set of coarse-resolution data. This approach differs from simply increasing the resolution, as the raw, coarse data are restructured using a set of logics, analogous to a regional meteorological model that downscales global meteorology using its own set of physical and thermal field balances. Conceptually, we use a calculation process reversed from that used to construct the pseudo-OMI data set.

Figure 5 graphically depicts the steps of conservative downscaling from OMI pixels. Figure 5a shows actual OMI NO_2 VCD measurements over Los Angeles on 4 May 2010, and Fig. 5b shows the corresponding CMAQ NO_2 VCD calculated from NAQFC modeling outputs at the same time and location. As readers can easily see, OMI footprint pixels are much bigger ($\sim 650 \text{ km}^2$) than are CMAQ grid cells ($12 \times 12 = 144 \text{ km}^2$). As a result, an OMI pixel can overlay more than 10 CMAQ grid

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cells, as demonstrated in Fig. 5b (black box representing the OMI pixel). We collected those CMAQ pixel values and then normalized them so that the total value of each grid cell sums to one. We call this a spatial-weighting kernel (Fig. 5c), and we apply this weighting kernel to the original OMI measurement. As a result, we generate a reconstructed OMI pixel with finer structure but without any loss of original quantity. Summing the reconstructed pixels gives the original OMI pixel measurement. It should be noted that we strictly apply this method conservatively; theoretically, if there are no missing or duplicated pixels, the quantity of the original data is numerically preserved. This method can be summarized as fusing a satellite-measured “quantity” with modeled “spatial information”; the strength of the modeled NO₂ field does not at all affect the result.

As expected, the accuracy of this method indeed depends on the model’s performance, especially regarding its wind-field simulation and inputs of emission source locations. Considering the uncertainties resulting from emission source locations, the air-quality community has had an excellent archive of geographical information about the geophysical locations of emission sources thanks to the efforts of US EPA, although the strengths of these sources are somewhat highly uncertain. As just described, however, the downscaling method is not affected by emission strength, so we do not think that the uncertainty associated with emission is very high. Wind field is important for simulating NO₂ plume transport. With the short lifetime of NO₂, especially during summer, the spatial distribution of NO₂ plumes is strongly determined by the location of emission sources. Improving information about emission-source locations would somewhat improve the model, but it is more important to note that the downscaling method tends to convert the error characteristics. Near urban cores, OMI’s coarse footprint resolution always causes unidirectional, systematic biases, with underestimation near urban cores and overestimation at the urban boundary. Using the downscaling method, these systematic biases from resolution are converted to random bias from wind-field error. Since these biases are random, they may be corrected by averaging over a certain time period, unlike the systematic bias resulting from resolution.

4.1 2010 CalNex campaign case

We applied the downscaling technique to compare OMI and downscaled OMI with aircraft-borne measurements from the California Research at the Nexus of Air Quality and Climate (CalNex) campaign. The CalNex field study was conducted in California from May to July 2010 and focused on atmospheric-pollution and climate-change issues, including an emission inventory, atmospheric transport and dispersion, atmospheric chemical processing, cloud-aerosol interaction, and aerosol radiative effects (Ryerson et al., 2013). Here, we compared NO₂ VCD observations from the campaign's P3 flight with corresponding OMI measurements using both the standard and downscaling methods. More detailed descriptions regarding data preparation and a discussion of the influence of environmental inhomogeneity and urban NO₂ plumes are provided by Judd et al. (2015).

Figure 6 shows scatter-plot comparisons between the P3 measurements and OMI NASA standard product (Fig. 6a), OMI KNMI product (Fig. 6b), and OMI KNMI downscaled (Fig. 6c) for three days: 4, 7, and 16 May 2010. As reported, the OMI NO₂ VCD tends to underestimate near the Los Angeles urban area. The KNMI retrieval showed a slightly better comparison with slope = 0.73 and $R = 0.85$, while the downscaled product clearly showed the best agreement with the P3 measurements, $R = 0.88$ and slope = 1.0. Deviations still remain from a true one-to-one line even with the downscaling method; these are possibly caused by errors in wind field simulation. We expect these random errors to average out as the amount of available data increases. The downscaling method seems to work even with daily time-scale data sets.

Figure 7 compares OMI NO₂ VCD spatial distributions for the original KNMI products with downscaled products for 4 May 2010, the day when the downscaling method gave the most dramatic changes in the spatial distribution. In the original retrieval, OMI pixels were coarse and mostly smoothed out over Los Angeles. However, by applying the downscaling technique, the adjusted OMI data show a shape much closer to the urban boundary and enhanced NO₂ VCD values at the center of Los Angeles, agreeing

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very well with the P3 aircraft measurements. On 7 May (not shown), the downscaling method reproduced several peak values very well but failed to generate a clean spot at the edge of Los Angeles. For 16 May (not shown), the changes from downscaling are not dramatic due to generally low NO₂ concentrations, but the downscaling method still showed slight enhancement.

4.2 Comparison with NAQFC

Comparing modeled NO₂ VCD to satellite-observed NO₂ VCD has been a popular way to evaluate the NO_x emission inventory. Since modeled NO₂ VCD and satellite NO₂ VCD have different optical and vertical properties, some researchers have used additional processing to fairly compare satellite and modeled column densities. In this section, we performed vertical and spatial adjustment by applying Averaging Kernel (AK) information in conjunction with the downscaling technique. First, we compared NAQFC NO₂ VCD with and without AK to OMI NO₂ VCD with and without downscaling processing.

The sensitivity of the instrument to tropospheric tracer density is highly height-dependent. Since the measured tracer profile may have large systematic errors as a result, the retrieved tracer columns should be interpreted with proper additional information (Eskes and Boersma, 2003). An AK stores an instrument's relative sensitivity to the abundance of the target species for each layer throughout the atmospheric column (Bucsela et al., 2008) and can be applied to a modeled atmospheric column for a fair comparison with satellite retrievals. For each OMI DOMINO product pixel, 34 layers of AKs are provided. We first converted total AK to tropospheric AK, AK_{trop}, by applying the total air mass factor (AMF) and tropospheric AMF, and we then applied AK_{trop} to model layers before vertically integrating, as described by Herron-Thorpe et al. (2010). When multiple OMI pixels overlaid a model grid cell, we conducted the conservative spatial remapping method explained above.

Figure 8 compares the monthly averaged NO₂ VCD distributions for CMAQ without and with AK (Fig. 8a and b, respectively) and for OMI NO₂ VCD without and with

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downscaling (Fig. 8c and d, respectively). In general, AK-applied CMAQ NO₂ VCD tends to be slightly lower than CMAQ NO₂ VCD without AK information. On the other hand, while OMI NO₂ VCD without DS shows a much smoother pattern, the DS-applied OMI reconstructs the sharp spatial structures near urban areas. DS-applied OMI NO₂ VCD is evidently able to construct sharp gradients near cities, and especially near middle-size cities.

Figure 9 compares CMAQ and OMI NO₂ VCDs using AK and DS methods together. Figure 9a shows a scatter-plot comparison between CMAQ and OMI NO₂ VCDs at US Environmental Protection Agency Air Quality System (AQS) surface-monitoring site locations. In this comparison, CMAQ NO₂ VCDs are much higher compared to OMI NO₂ VCDs, implying that the CMAQ simulation possibly overestimates NO_x emissions. Figure 9c compares OMI and CMAQ NO₂ VCD with AK information applied; estimated CMAQ NO₂ VCD is reduced, showing better agreement with OMI NO₂ VCD. Readers may notice that high CMAQ pixels are shifted to the left. On the other hand, applying the DS method to OMI shifts OMI pixels vertically (Fig. 9b). Finally, in Fig. 9d, both AK and DS methods are applied; this comparison shows the best agreement between OMI and CMAQ NO₂ VCD pixels. Its correlation coefficient $R = 0.89$ and the slope of line fit is 0.59. Clearly, the application of the AK and DS methods not only improved the satellite-model comparison in the high NO₂ concentration range but also significantly improved the comparison in the low NO₂ range (i.e., $0-10 \times 10^{15}$ molecules cm⁻²), implying that this method can help interpret NO_x emission in major and mid-size cities.

The differences in spatial distributions between monthly averaged OMI and CMAQ NO₂ VCDs during September 2013 are shown in Fig. 10. Positive values indicate that CMAQ NO₂ VCD is higher than OMI VCD, which should likely be interpreted as an overestimation of the NO_x emission inventory used in the CMAQ modeling. The difference between the original OMI and CMAQ NO₂ VCDs show strong positive values over most urban locations (Fig. 10a). Applying AK (Fig. 10b) and DS (Fig. 10c) reduce positive biases for major and middle-to-small cities, showing the best agreement when both AK and DS are included. NO₂ VCD is still overestimated over major cities – New

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tical structure. Four-way comparisons were conducted between OMI with and without downscaling and CMAQ with and without AK information. Results show that OMI and CMAQ NO₂ VCDs show the best agreement when both downscaling and AK methods are applied, with correlation coefficient $R = 0.89$.

5 These results should be considered when using satellite data in the evaluation of emission inventories and translating these data into decision-making around emission policy. Table 1 shows a summary of the comparisons between OMI and CMAQ NO₂ VCDs described in Figs. 8 and 9. When CMAQ without AK and OMI with DS are compared, the percentage difference is $(6.43 - 3.61)/3.61 \times 100 = 78\%$, implying that the current emission inventory likely overestimates NO₂ VCD. Comparing between OMI with DS and CMAQ without AK or between OMI without DS and CMAQ with AK still implies that the current emission inventory is possibly overestimating. However, when both vertical and spatial profiles are adjusted using the AK and DS methods, a slight underestimation is found, -7% , in modeled NO₂ VCD over AQS monitoring locations, 10 implying that the current inventory possibly underestimates emissions. This may represent an important implication for how spatial information should be considered when investigating fine-scale phenomena such as urban NO₂ plumes.

Without question, satellite observations are very useful with their large coverage supplementing sparse surface-monitoring sites. Interpretation of satellite-based measurement, however, should be performed cautiously with consideration of the instrument's characteristics, especially when translating results into policy-making. We expect our current study to provide a reference for the uncertainty of satellite-based information regarding local or regional pollutants, especially until we have the measurement data at more enhanced resolution that will be provided by future satellites, such as TEMPO, 20 TROPOMI, and GEMS.

25 *Acknowledgements.* We acknowledge the free use of tropospheric NO₂ column data from the OMI sensor from www.temis.nl.

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Table 1. Comparison of OMI and CMAQ NO₂ VCD monthly averages (September 2013) at AQS sites.

	OMI/xDS (mean = 3.61)	OMI/DS (mean = 5.00)
CMAQ/xAK (mean = 6.43)	$S = 0.28$ $R = 0.79$ $(6.43 - 3.61)/3.61 \times 100 = 78.1\%$	$S = 0.45$ $R = 0.87$ $(6.43 - 5)/5 \times 100 = 28.6\%$
CMAQ/AK (mean = 4.65)	$S = 0.39$ $R = 0.87$ $(4.65 - 3.61)/3.61 \times 100 = 28.8\%$	$S = 0.59$ $R = 0.89$ $(4.65 - 5)/5 \times 100 = -7.0\%$

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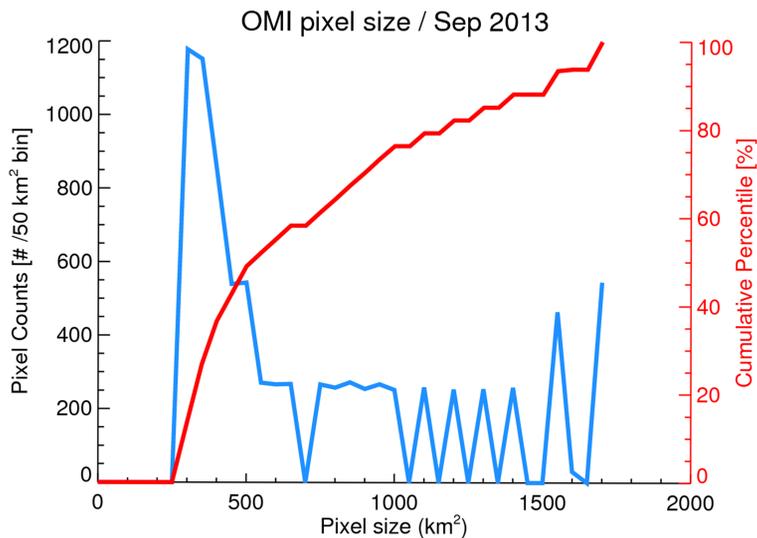


Figure 1. Size distribution of OMI pixel footprint (blue) and its cumulative percentile (red).

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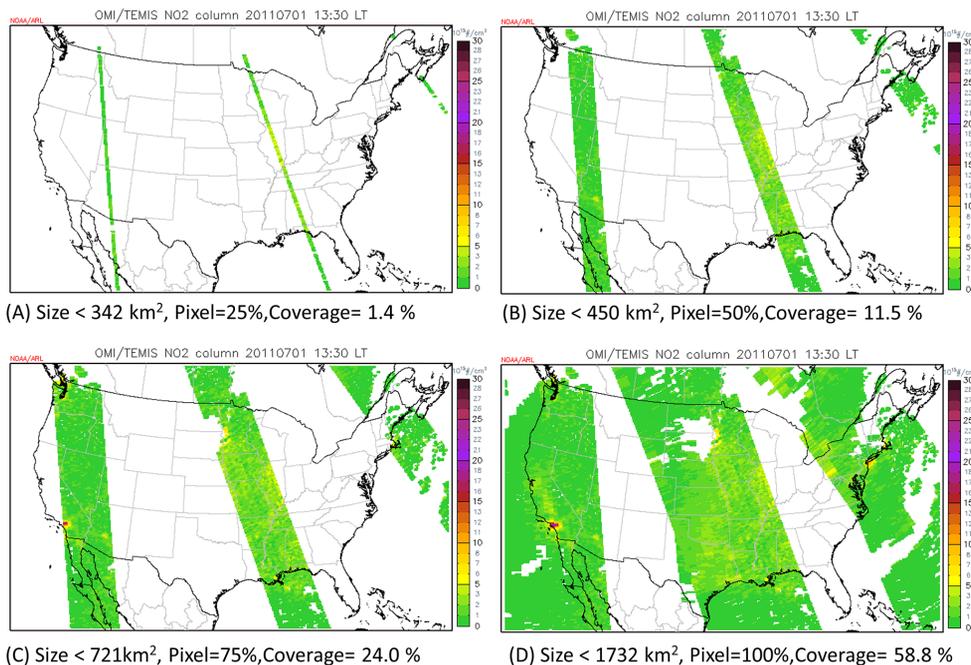


Figure 2. Comparison of OMI footprint-pixel size and actual coverage using (a) 25 %, (b) 50 %, (c) 75 %, and (d) 100 % of available pixels.

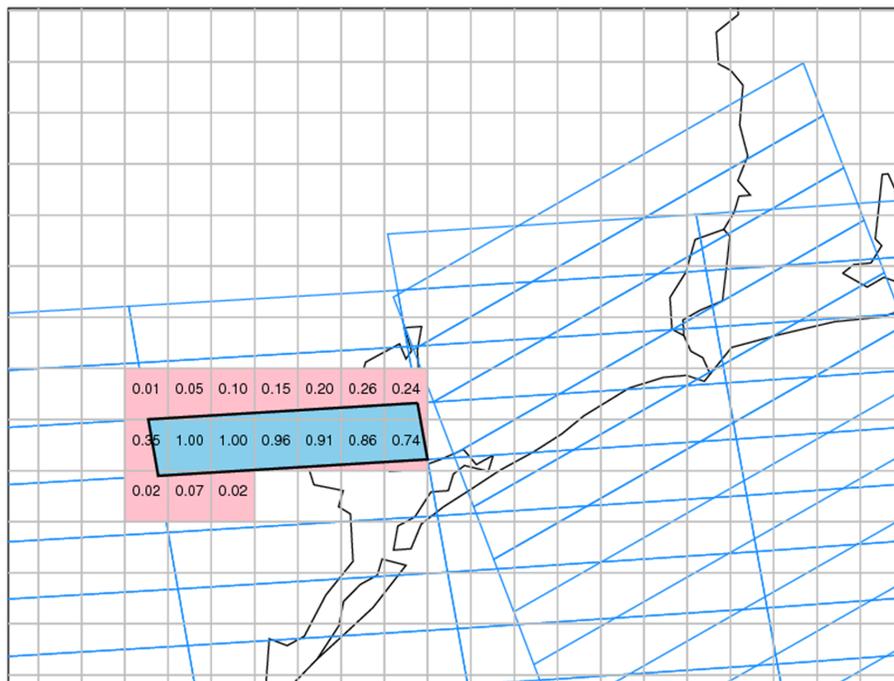


Figure 3. Calculation of pseudo-OMI (pOMI) data. Blue boxes are actual OMI pixel footprints and the gray cells are 12 km grid cells. Fraction of cells overlapped by an OMI pixel are shown, and pOMI (sky blue) data are estimated by a weighted average of the corresponding grid cells (pink).

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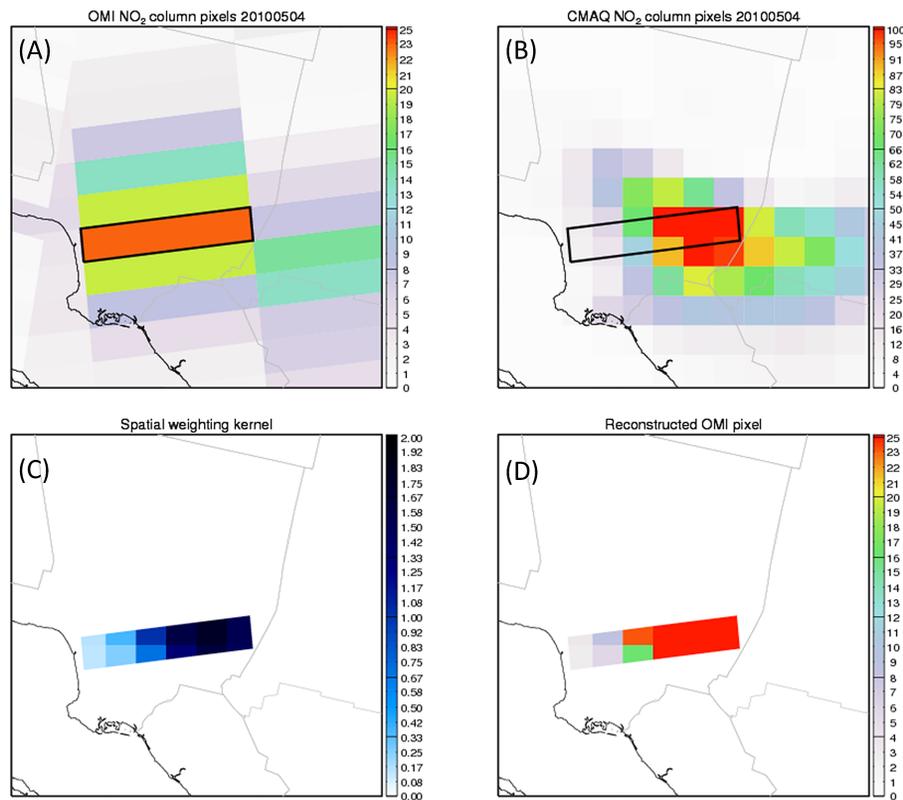


Figure 5. Example of downscaling method. **(a)** Original OMI NO₂ VCD, **(b)** 12 km CMAQ NO₂ VCD, **(c)** spatial weighting kernel, and **(d)** adjusted OMI NO₂ VCD using spatial weighting kernel.

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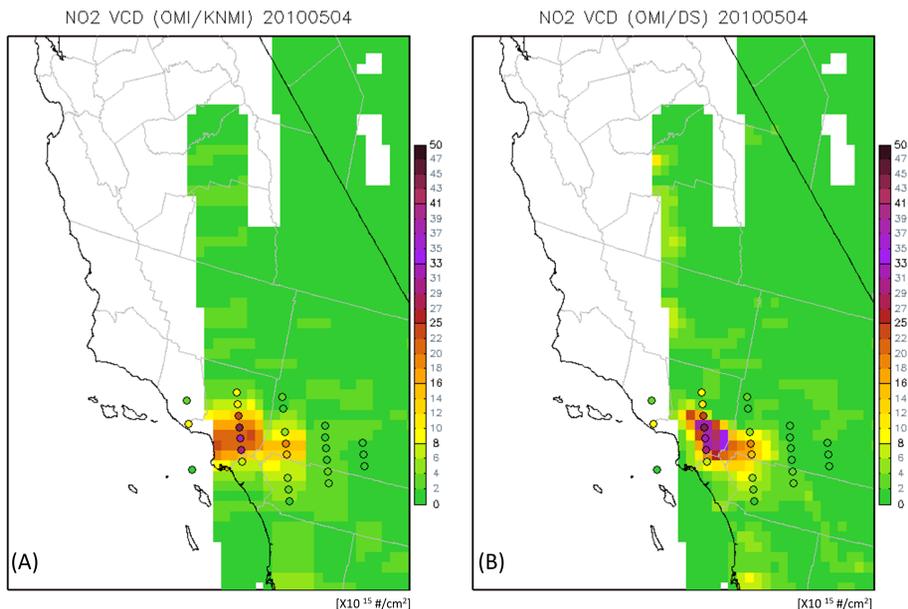


Figure 7. Spatial distribution of P3 NO₂ VCDs (circles) and OMI NO₂ VCDs for original KNMI product **(a)**, and downscaled OMI **(b)** for 4 May 2010.

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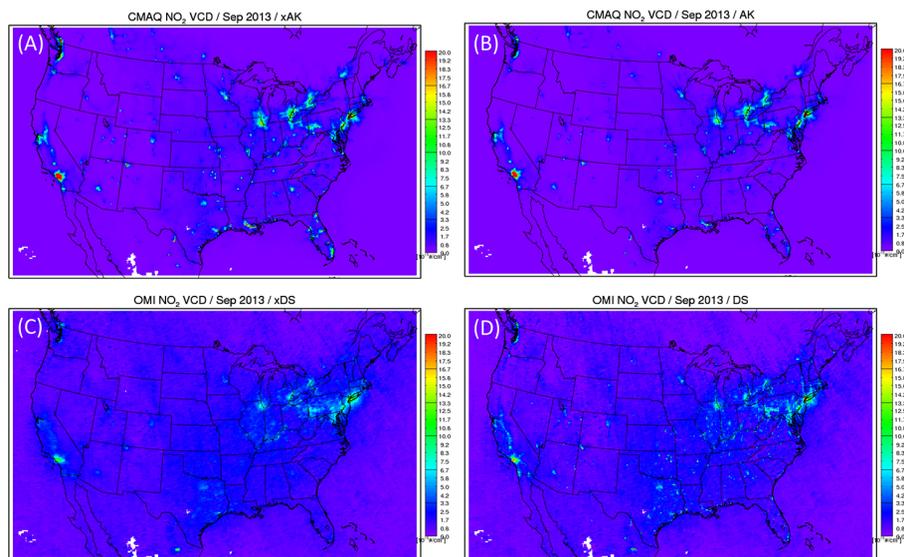


Figure 8. Spatial distributions of (a) CMAQ NO₂ VCD without AK and (b) with AK; (c) OMI NO₂ VCD without downscaling and (d) with downscaling.

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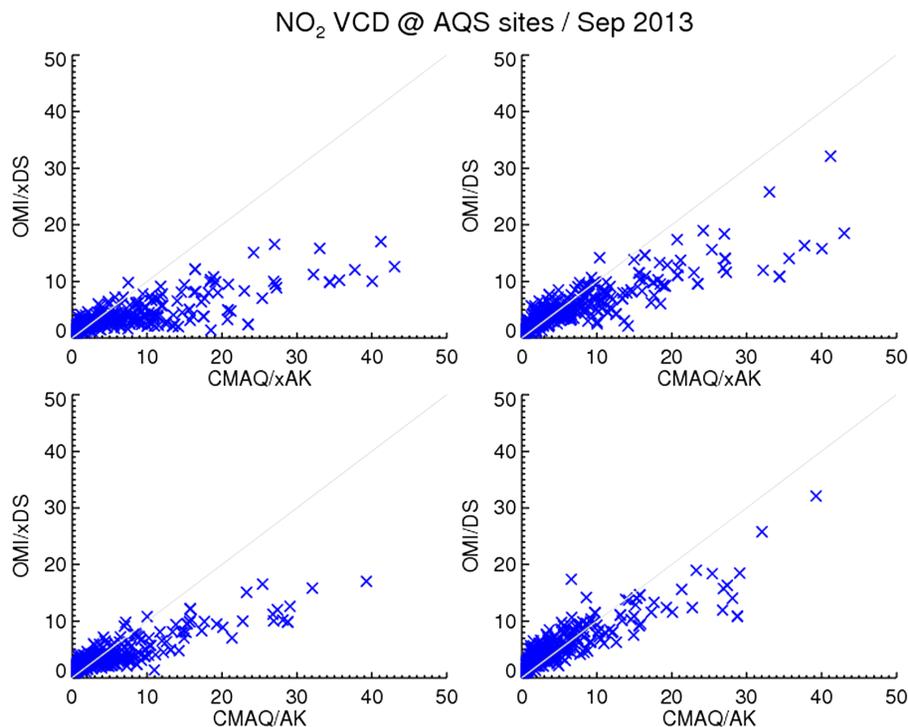


Figure 9. Comparison of OMI and CMAQ NO₂ VCD for **(a)** OMI and CMAQ with AK, **(b)** down-scaled OMI and CMAQ with AK, **(c)** OMI and CMAQ with AK, and **(d)** down-scaled OMI and CMAQ with AK.

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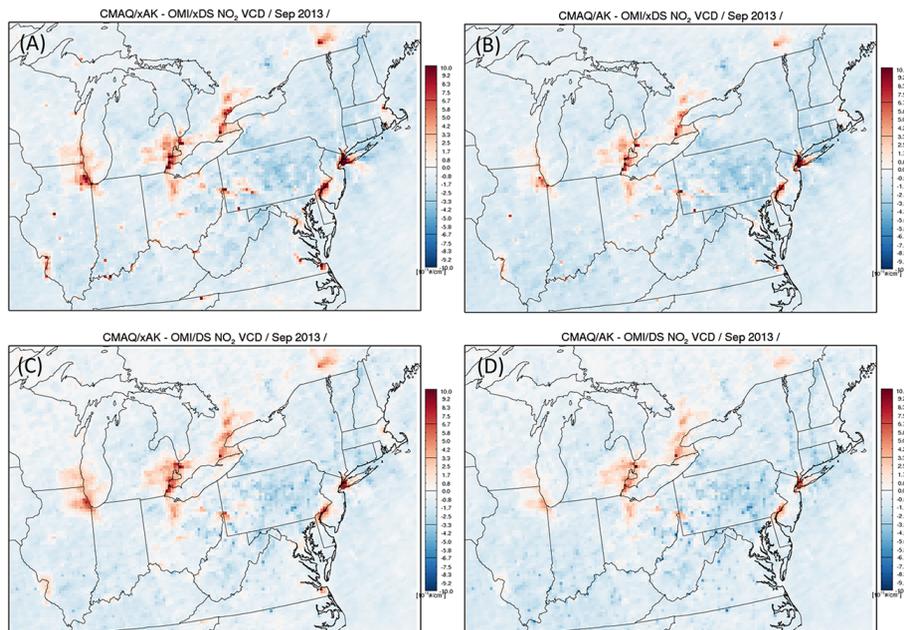


Figure 10. Comparisons of OMI and CMAQ NO₂ VCD spatial distributions in the northeast US region.

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