Inferring ground-level nitrogen dioxide concentrations at fine spatial resolution applied to the TROPOMI satellite instrument

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Abstract

Satellite-based estimates of ground-level nitrogen dioxide (NO\(_2\)) concentrations are useful for understanding links between air quality and health. A longstanding question has been why prior satellite-derived surface NO\(_2\) concentrations are biased low with respect to ground-based measurements. In this work we demonstrate that these biases are due to both the coarse resolution of previous satellite NO\(_2\) products and inaccuracies in vertical mixing assumptions used to convert satellite-observed tropospheric columns to surface concentrations. We develop an algorithm that now allows for different mixing assumptions to be used based on observed NO\(_2\) conditions. We then apply this algorithm to observations from the TROPOMI satellite instrument, which has been providing NO\(_2\) column observations at an unprecedented spatial resolution for over a year. This new product achieves estimates of ground-level NO\(_2\) with greater accuracy and higher resolution compared to previous satellite-based estimates from OMI. These comparisons also show that TROPOMI-inferred surface NO\(_2\) concentrations from our updated algorithm have higher correlation and lower bias than those found using TROPOMI and the prior algorithm. TROPOMI-inferred estimates of the population exposed to NO\(_2\) conditions exceeding health standards are at least three times higher than for OMI-inferred estimates. These developments provide an exciting opportunity for air quality monitoring.

1. Introduction

Air pollution is a leading risk factor for premature mortality, as exposure to air pollution is associated with increases in heart disease, stroke, cancer, and other respiratory and cardiovascular diseases (Cohen \textit{et al} 2017). Nitrogen dioxide (NO\(_2\)) is a major contributor to poor air quality and exposure to NO\(_2\) has been associated with increased rates of asthma incidence (Anenberg \textit{et al} 2018, Achakulwisut \textit{et al} 2019), lung cancer (Hamra \textit{et al} 2015), and overall mortality (Burnett \textit{et al} 2004, Brook \textit{et al} 2007, Crouse \textit{et al} 2015). NO\(_2\) is often used as an indicator of air pollution from traffic and other combustion sources (Brook \textit{et al} 2007, Levy \textit{et al} 2014, Achakulwisut \textit{et al} 2019). Additionally, NO\(_2\) is a precursor to aerosol and ozone production, which are also important pollutants (Cohen \textit{et al} 2017, Stieb \textit{et al} 2019). Quantifying the impacts of NO\(_2\) on health requires accurate monitoring of NO\(_2\) concentrations and their spatial variability.

The distribution of NO\(_2\) has substantial spatial gradients due to its short lifetime and the spatial variability of emission sources. This inhomogeneity inhibits constructing accurate NO\(_2\) fields by \textit{in situ} monitoring alone. Satellite instruments provide NO\(_2\) observations with greater spatial coverage. Previous studies have shown that surface NO\(_2\) concentrations can be inferred from satellite-retrieved vertical column densities using a chemical transport model to
relate the retrieved column to ground-level concentrations (Lamsal et al 2010, 2013, Kharol et al 2015, Geddes et al 2016, Gu et al 2017). Recent studies have used machine learning techniques to convert column abundances to ground-level concentrations (Chen et al 2019, De Hoogh et al 2019, Beloconi and Vounatsou 2020, Di et al 2020, Qin et al 2020). These satellite-based ground-level concentration estimates have been useful in health impact studies as an estimate of pollution exposure (Anenberg et al 2018) or as an input to land use regression models used to estimate exposure at finer spatial resolution (Larkin et al 2017, Achakulwisut et al 2019).

Previous evaluations indicate that satellite-inferred surface concentrations correlate well with in situ measurements but were biased low (Geddes et al 2016, Gu et al 2017) for reasons that have not been resolved. The contribution of biases in columnar retrievals to this surface bias has largely been attributed to issues related to horizontal resolution, either from coarse resolution a priori NO2 profiles used in the retrieval (e.g. Heckel et al 2011, Russell et al 2011) or from using point ground monitors to evaluate satellite observations with coarse pixel resolution (Kharol et al 2015, Judd et al 2019). These same factors may be exacerbated when surface concentrations are inferred from columnar retrievals. Comparisons with ground monitors and aircraft indicate that there is substantial variability in NO2 concentrations within the coarse pixel size of prior satellite observations, causing coarse resolution satellite measurements to underestimate peaks in the NO2 field by a factor of two or more (Broccardo et al 2018, Judd et al 2019; Lamsal et al 2017). Fine resolution NO2 profiles have been shown to improve NO2 retrieval accuracy (Russell et al 2011, Valin et al 2011, Mclinden et al 2014, Laughner et al 2016, Goldberg et al 2017, Ialongo et al 2020, Liu et al 2020). Fine spatial resolution observations also reduce the likelihood of a satellite observation being affected by clouds that obscure surface-level NO2 from observation and hinder the ability to resolve fine-scale spatial structure of the surface NO2 field.

Application of algorithmic advances to the TROPOspheric Monitoring Instrument (TROPOMI) provides an opportunity to build upon these previous studies to re-examine the bias with surface concentrations. The spatial resolution of TROPOMI data used here (3.5 × 7 km2) is roughly a factor of 13 finer than its predecessor Ozone Monitoring Instrument (OMI) (13 × 24 km2). Preliminary studies have shown that TROPOMI is capable of observing pollution at unprecedented scales (Hu et al 2018, Alvarado et al 2020, Goldberg et al 2019, de Laat et al 2020, Theys et al 2019, Zhao et al 2020). Initial evaluations have found that TROPOMI NO2 columns are well correlated with other measurements with a small low bias (Griffin et al 2018, Ialongo et al 2020, Zhao et al 2020).

This paper presents a new algorithm for inferring ground level NO2 concentrations that improves upon previous methods by updating assumptions regarding vertical mixing in the boundary layer and allowing the satellite-retrieved column densities to inform the relation of column abundances to ground level concentrations. The high spatial resolution of TROPOMI NO2 column observations, together with these algorithmic developments, addresses the bias in satellite-inferred surface concentrations and reveals unprecedented fine-scale information in estimated ground-level NO2 concentrations.

2. Data

2.1. Satellite NO2 data
In this study we use retrieved NO2 column densities from both TROPOMI (Veekind et al 2012, Van Geffen et al 2020) and OMI (Levelt et al 2006) satellite instruments from July 2018-June 2019. Both instruments have spectrometers measuring in the UV–Vis spectral bands, and are on sun-synchronous orbits with local overpass times around 1:30 P.M. NO2 column retrievals from TROPOMI use the DOMINO retrieval method (Boersma et al 2011, 2018). NO2 retrievals from OMI are from the NASA Standard Product version 3 (Krotkov et al 2017). Only observations with retrieved cloud fractions less than 0.1 are used. The important difference between the two satellite products for this study is their spatial resolution; while both TROPOMI and OMI have a 2600 km swath width, TROPOMI (3.5 × 7 km2) has 450 across-track pixels while OMI (13 × 24 km2) has 60. Additionally, a partial blockage of the OMI field of view, known as the row anomaly, prevents quality observations from many pixels and further hinders OMI’s observation density (http://projects.knmi.nl/omi/research/product/rowanomaly-background.php).

2.2. GEOS-Chem
The GEOS-Chem chemical transport model version 12.3.2 (www.geos-chem.org) is used here. GEOS-Chem contains a detailed HOx–NOx–VOC–O3–aerosol chemical mechanism (Bey et al 2001, Park et al 2004) and is driven by Goddard Earth Observing System Forward Processing (GEOS-FP) assimilated meteorological data. We use a nested grid with 0.25° × 0.3125° resolution over North America (9.75°–60° N, 60°–130° W) with dynamic boundary conditions at 2° × 2.5° resolution. The nested grid values are averaged to coarser resolutions for testing the effect of resolution on inferred surface concentrations.

Anthropogenic NOx emissions are from the Community Emissions Data System (Hooshy et al 2018) with regional overwrites where more detailed information is available over Canada (Air Pollution Emission Inventory) and the US, (National...
Emissions Inventory 2011, Travis et al (2016). Biomass burning emissions are from the GFED4.1 inventory (Van der Werf et al 2010). Lightning NOx emissions are described by Murray et al (2012). Soil NOx emissions are described by Hudman et al (2012). Of importance to this work is the capability of GEOS-Chem to translate column abundances to surface concentrations, which requires accurate representation of the NO2 profile; past evaluations demonstrate that GEOS-Chem simulated NO2 profiles are consistent with aircraft observations (Lin and Mcelroy 2010, Travis et al 2016).

2.3. In situ NO2 observations
We use hourly surface NO2 measurements from the US Environmental Protection Agency Air Quality System (US-EPA AQS, https://aqs.epa.gov/aqsweb/documents/data_mart_welcome.html) over the continental US and Environment and Climate Change Canada’s National Air Pollution Surveillance Program (NAPS, http://maps-cartes.ec.gc.ca/rnspa-naps/data.aspx) over Canada to evaluate the satellite products. We calculate annual mean concentrations at each of the 625 sites by averaging observations between 13:00 h and 15:00 h corresponding to the satellite overpass times. NAPS measurements from 2017 are used as these are the most recent available dates. Following Lamsal et al (2008) we use a correction factor derived from GEOS-Chem to correct for the known overestimate in regulatory measurements of NO2 concentrations due to interference of other reactive nitrogen species (i.e. peroxyacetyl nitrate (PAN), nitric acid (HNO3), and organic nitrates):

\[
\text{NO}_2 = \frac{\text{NO}_2}{\text{NO}_2 + \text{alkyl nitrates} + 0.95 \times \text{PAN} + 0.15 \times \text{HNO}_3}.
\]

(1)

3. Methods

3.1. Improving the air mass factor calculation
A significant source of systematic error in satellite NO2 retrievals is the a priori assumed NO2 profile used in calculating the air mass factor (AMF) that converts line-of-sight slant columns to vertical column densities (Lorente et al 2017; Boersma et al 2018). Replacing the a priori used in the retrievals with profiles from fine-resolution models has been shown to improve retrieved vertical column densities (Russell et al 2011, Valin et al 2011, McLinden et al 2014, Laughner et al 2016, Goldberg et al 2017, Ialongo et al 2020, Liu et al 2020). Inconsistencies between a priori profiles may introduce spurious differences when comparing two different satellite products, or when using a model to relate surface concentrations to vertical columns (Boersma et al 2016). For these reasons we remove the influence of the a priori profile in both the TROPOMI and OMI AMFs and replace them with GEOS-Chem vertical profiles using the method outlined in Lamsal et al (2010):

\[
\omega^o = \frac{\Omega^o \sum_k n_k^G}{M \sum_k A_k n_k^G},
\]

(2)

where \(\omega^o\) is the updated vertical column, \(\Omega^o\) is the observed slant column, \(M\) is the retrieval AMF, \(A_k\) is the retrieval averaging kernel, and \(n_k^G\) is the normalized tropospheric NO2 profile from the GEOS-Chem simulation. We use the Interactive Multi-sensor Snow and Ice Mapping System (IMS) (Helfrich et al 2007) 4 km product to identify snow-covered pixels, which are then omitted from this analysis. IMS is used here in addition to the snow identification used in the TROPOMI and OMI retrievals as it has been shown to more accurately identify snow cover (Cooper et al 2018).

3.2. Surface estimation
We estimate satellite-based surface NO2 concentrations at both the 0.25\(^\circ\) × 0.3125\(^\circ\) (≈28 × 28 km\(^2\)) at 35\(^\circ\)N resolution of the GEOS-Chem nested simulation (i.e. ‘moderate’ resolution) and at a sub-model-grid resolution of 0.025\(^\circ\) × 0.03125\(^\circ\) (i.e. ‘fine’ resolution, ≈2.8 × 2.8 km\(^2\)) at 35\(^\circ\)N. Satellite vertical column densities are gridded daily using an area-weighted oversampling approach (Spurr 2003). We infer fine-resolution surface concentrations (\(S^o\)) by further developing the method outlined in Kharol et al (2015) and Lamsal et al (2008) that accounts for subgrid variation in the column to surface relationship to correct for biases related to vertical mixing assumptions.

Surface concentrations (\(S\)) can be inferred from satellite-retrieved vertical column abundances (\(\Omega\)) using a simulated surface-to-column conversion factor from a chemical transport model:

\[
S^o = \frac{S}{\Omega^o} \Omega^o.
\]

(3)

The superscript o signifies quantities based on satellite observations (otherwise quantity is modeled). Satellite-based surface concentrations at a sub-model-grid resolution could be similarly calculated:

\[
S^o_i = \frac{S_i}{\Omega^o_i} \Omega^o_i.
\]

(4)

where the subscript i represents sub-grid resolution values (model resolution values otherwise), however \(S_i\) and \(\Omega_i\) are unknown. Following Lamsal et al (2008) we use the satellite-observed sub-grid variability to infer sub-grid variability in the simulated tropospheric column:

\[
\nu = \frac{\Omega^o_i}{\Omega^o} = \frac{\Omega_i}{\Omega}.
\]

(5)
where $\Omega^F$ is the average satellite-retrieved tropospheric column within the model grid box.

We assume that the tropospheric column consists of two parts:

$$\Omega = \Omega^{\text{low}} + \Omega^{\text{up}},$$

(6)

where $\Omega^{\text{up}}$ is the upper portion of the column that is well mixed both vertically and horizontally across the model grid box (such that $\Omega^{\text{up}} = \Omega^{\text{up},0}$), and $\Omega^{\text{low}}$ is the lower portion of the column that is well mixed vertically but has sub-grid horizontal variability due to local surface emissions. Using these definitions, Equation (6) can be expressed for sub-pixel columns as:

$$\Omega_s = \Omega^{\text{low}}_s + \Omega^{\text{up}}_s = \Omega^{\text{low}}_s + \Omega^{\text{up}}.$$

(7)

Applying the sub-pixel tropospheric column variability from the satellite observations via equation (5) into equation (7) gives:

$$\Omega^{\text{low}}_s = \nu \Omega - \Omega^{\text{up}}.$$

(8)

We assume that air density is constant within a grid box and treat the ratio of surface concentration to lower partial column as the same at sub-grid and grid resolution, i.e.

$$\frac{S_s}{\Omega^{\text{low}}_s} = \frac{S}{\Omega^{\text{low}}}. $$

(9)

Substituting equations (5), (8), and (9) into equation (4) gives

$$S_s = \left(\nu \Omega - \Omega^{\text{up}}\right) \left(\frac{S}{\Omega}\right) \Omega^F. $$

(10)

An important decision is selecting how to distinguish the horizontally well-mixed upper partial column from the lower partial column (which has sub-grid variability). Previous studies have defined the lower partial column as the being within the boundary layer and the upper column as the free troposphere (e.g. Lamsal et al 2010, Kharol et al 2015). This has been shown to well represent the spatial distribution as the free troposphere is generally well mixed and has less horizontal variability compared to the boundary layer. However, this definition assumes that the boundary layer NO$_2$ profile is uniform vertically, which is not the case when there are large emission sources that produce significant surface enhancements, and leads to an underestimation of surface NO$_2$ concentrations (Zhang et al 2016). This bias has been noted in evaluations of previous satellite-inferred surface concentration estimates (Kharol et al 2015).

Alternatively, one could define the lower column as the surface-level partial column (i.e. the lowest model grid box, centered at $\sim$50 m altitude) and the upper column as the remaining column above (i.e. model level 2 to tropopause). This is likely a better assumption over large emission sources and can better attribute local peaks in NO$_2$ columns to surface enhancements, reducing the underestimations in the previous method. However, assuming all the sub-grid variability occurs at the surface ignores potential sub-grid variability in the lower boundary layer away from the emission source, artificially inflating the surface level variability and producing an unrealistic sub-grid spatial distribution.

Of these two possibilities, assuming a uniform free troposphere and uniform boundary layer NO$_2$ ($S_{\text{BL},BL}$) is more reasonable in most cases and produces a more realistic spatial distribution, while assuming a well mixed surface-level partial column ($S_{\text{surf}}$) is more accurate over large emission sources. Therefore, we calculate satellite-inferred surface concentrations using both methods, and define a scaling factor $\chi$ in each model grid box as the ratio of the maximum values from each method:

$$\chi = \frac{\max \left(S_{\text{surf}}\right)}{\max \left(S_{\text{BL},BL}\right)}. $$

(11)

The maximum value in each grid box represents the value where assuming a uniform surface-level partial NO$_2$ column is likely the more accurate assumption. Therefore, this scaling factor quantifies the magnitude of the bias that occurs from incorrectly assuming a uniform boundary layer NO$_2$ over large emission sources. One can then apply this scaling factor to the original method over polluted regions where a surface enhancement is expected:

$$S_s^{(\text{polluted})} = \chi \left(\frac{\nu \Omega - \Omega^{\text{free troposphere}}}{\Omega^{\text{boundary layer}}}\right) \left(\frac{S}{\Omega}\right) \Omega^F = \chi S_{\text{BL},BL}. $$

(12)

In relatively clean regions, the mixed boundary layer assumption without the scaling factor is preferred:

$$S_s^{(\text{clean})} = \left(\frac{\nu \Omega - \Omega^{\text{free troposphere}}}{\Omega^{\text{boundary layer}}}\right) \left(\frac{S}{\Omega}\right) \Omega^F = S_{\text{BL},BL}. $$

(13)

We define clean regions as those with annual mean TROPOMI tropospheric columns less than $10^{15}$ mol cm$^{-2}$ and polluted regions as those with annual mean columns greater than $11 \times 10^{15}$ mol cm$^{-2}$. This allows the satellite-retrieved column abundance to inform the shape of the sub-grid vertical profile. We use a linear interpolation between the two methods for regions that fall between the clean and polluted thresholds.

To summarize, sub-grid surface concentrations are given by:

$$S_s = \kappa \left(\frac{\nu \Omega - \Omega^{\text{free troposphere}}}{\Omega^{\text{boundary layer}}}\right) \left(\frac{S}{\Omega}\right) \Omega^F $$

(14)
where
\[
\kappa = \begin{cases} 
1 & \text{if } \Omega_o < 10 \times 10^{15} \text{ molec cm}^{-2} \\
(1 - w) + \chi w & \text{if } 10 \times 10^{15} < \Omega_o < 11 \times 10^{15} \text{ molec cm}^{-2} \\
\chi & \text{if } \Omega_o > 11 \times 10^{15} \text{ molec cm}^{-2}
\end{cases}
\]
(15)

and
\[
w = \frac{\Omega_o - 1 \times 10^{15}}{10 \times 10^{15}}.
\]
(16)

The $10^{15}$ and $11 \times 10^{15} \text{ mol cm}^{-2}$ cut-off values were chosen following a sensitivity test where values ranging from 1 to $20 \times 10^{15} \text{ mol cm}^{-2}$ were tested. The selected values yielded the best agreement between satellite-inferred concentrations and the in situ surface measurements.

Typical values of $\kappa$ are 1 in clean regions, 1.5–2 in most cities, and 2.5–3 in major cities like New York and Los Angeles. Setting $\kappa$ equal to unity makes equation (14) equivalent to the method used in Lam sal et al (2008) and Kharol et al (2015).

We use GEOS-Chem to correct for sampling biases in the satellite records due to persistent cloudy periods or surface snow cover by sampling the GEOS-Chem simulated surface concentrations to match the satellite ($S_{\text{sampled}}$), and using the ratio of the sampled mean to the true annual mean ($S_{\text{annual}}$),
\[
S_{\text{annual}}^{\text{true}} = \frac{S_{\text{annual}}}{S_{\text{sampled}}}. 
\]
(17)

4. Results

Figure 1 shows surface concentrations inferred from OMI and TROPOMI at both moderate ($\approx 28 \times 28 \text{ km}^2$ at $35^\circ \text{N}$) and fine ($\approx 2.8 \times 2.8 \text{ km}^2$ at $35^\circ \text{N}$) resolution. NO$_2$ concentrations from both instruments exhibit enhancements over urban and industrial areas. TROPOMI-inferred concentrations are typically larger than those from OMI at moderate resolution (mean fractional bias 13%). At fine resolution, TROPOMI-inferred concentrations are higher over cities and large emission sources. The advantage of the smaller TROPOMI pixel size is apparent at fine resolution, as the TROPOMI-inferred concentrations at this resolution display less noise and reveal more fine-scale features in the NO$_2$ field than those from OMI. Area-weighted and population-weighted mean NO$_2$ concentrations over North America for both
Figure 2. Annual mean surface NO$_2$ concentrations inferred from OMI and TROPOMI vertical column densities from July 2018 to June 2019 at 0.025$^\circ$ × 0.025$^\circ$ resolution. Features mentioned in text are labeled as (A) Indianapolis, (B) Columbus, (C) Orlando, (D) Interstates-75 and -81, (E) Edmonton-Red Deer-Calgary, (F) Saskatoon, (G) Winnipeg, (H) Interstate-35, (I) Canadian oil sands region, (J) eastern Wyoming oil fields.

Instruments are inset in figure 1. TROPOMI-inferred population-weighed concentrations are 41%–91% higher than those inferred from OMI, with potential implications for health impact assessments.

Figure 2 shows fine resolution surface concentrations inferred from TROPOMI and OMI on a regional scale. While OMI can observe enhancements surrounding major cities, the TROPOMI-inferred surface concentrations show greater fine-detail structure (i.e. more well-defined enhancements over Indianapolis and Columbus, distinguishing Edmonton/Red Deer/Calgary) as well as signatures from smaller cities not captured by OMI (i.e. Orlando, Saskatoon, Winnipeg). TROPOMI-inferred surface concentrations also show signatures from major industrial corridors that cannot be observed in the OMI-inferred NO$_2$ field (i.e. I-75 and -81 from Atlanta through eastern Tennessee, I-35 from Dallas to Austin). TROPOMI-inferred surface concentrations also show a greater enhancement over regions with significant oil and gas production (i.e. the Canadian oil sands region, eastern Wyoming oil fields).

Table 1 summarizes the impact that resolution has on the likelihood of observing clear-sky conditions that are ideal for surface NO$_2$ sensitivity. We define clear pixels as those with retrieved cloud fractions less than 0.1, and the fraction of observed area that is clear as the fraction of observed area consisting of pixels with cloud fractions <0.1. Pixels affected by the OMI row anomaly are not included. The fine resolution of TROPOMI allows for a greater fraction of clear pixels and a greater observed clear area overall. These clear-sky conditions have greater sensitivity to
near-surface NO$_2$ and thus improve the quality of surface NO$_2$ inferences.

Figure 3 shows a scatterplot comparing satellite-inferred surface concentrations to the ground-based measurements over the US and Canada. TROPOMI has a higher correlation and lower bias compared to the in situ observations at both resolutions. The new algorithm developed here (figure 3(c)) improves upon the prior algorithm of Lamsal et al. (2008) (figure 3(b)). The supplementary material (available online at stacks.iop.org/ERL/15/104013/mmedia) shows that the quality of these estimates persists at seasonal scales.

Figure 4 examines the effect of a priori resolution on TROPOMI-inferred surface concentrations. The prior method of Lamsal et al. (2008) is sensitive to model resolution, with agreement with in situ observations deteriorating as profile resolution increases. The method developed here allows for the TROPOMI column observations to inform sub-grid profile information, including adjusting limits on w at each resolution (upper w limit = 11, 10, 9, and $4 \times 10^{15}$ mol cm$^{-2}$ for panels (A) through (D)). This allows for consistency with surface measurements at all resolutions tested here.

Figure 5 shows estimates of the population exposed to different NO$_2$ concentrations across North America as a function of different instruments and algorithms. TROPOMI-inferred surface concentrations are higher than those inferred from OMI, with TROPOMI indicating nearly three times as many people across North America experience exposures that exceed the Canadian Ambient Air Quality Standard annual mean NO$_2$ exposure of 17 ppb than indicated by OMI. TROPOMI-based estimates indicate that 9 million people live...
in North American regions that exceed the World Health Organisation annual mean guideline of 40 ppb and 3 million exceeding the US EPA guideline of 53 ppb, while OMI-inferred concentrations suggest no exceedances at these levels. In contrast, exposures calculated using the prior algorithm of Lamsal et al. (2008) suggest no exceedances for either TROPOMI or OMI.

5. Discussion and conclusion

A longstanding question has been why previous satellite-derived surface NO$_2$ concentrations underestimated ground level measurements. Here we developed improved algorithms to derive surface NO$_2$ from columnar satellite observations and find that the biases observed in previous studies cannot be corrected by increasing satellite resolution alone, as they were partially due to inaccurate assumptions regarding vertical mixing within the boundary layer.

The new algorithm developed here allows for different vertical mixing assumptions to be made based on satellite-observed NO$_2$ conditions, substantially reducing these biases. Improvements in spatial resolution achieved by TROPOMI have greatly improved the ability to infer surface concentrations from satellite NO$_2$ columns compared to its predecessor OMI. TROPOMI provides a more detailed surface NO$_2$ field across North America, allowing for identification of features that were previously difficult to observe, such as smaller cities and industrial corridors along highways. TROPOMI-inferred surface concentrations also have better agreement with surface observations at both resolutions. Fine resolution estimates from TROPOMI have significantly better agreement than those from OMI found here as well as in previous studies, owing both to TROPOMI’s greater resolution and improvements to the method of inferring surface concentration from column abundances developed here. The finer spatial resolution of TROPOMI also

Table 1. Frequency of clear-sky conditions for North America. Clear pixels are those with retrieved cloud fractions less than 0.1.

| Satellite | Fraction of pixels that are clear | Fraction of observed area that is clear |
|-----------|----------------------------------|--------------------------------------|
| OMI       | 0.38                             | 0.34                                 |
| TROPOMI   | 0.44                             | 0.42                                 |

Figure 4. Comparison between annual mean surface observations and TROPOMI-inferred surface concentration using the method developed here ($S^o_s$) and the original method of Lamsal et al. (2008) ($S^o_{s,BL}$) using model information at different horizontal resolutions. The slope for line of best fit and correlation coefficients are inset. The dashed line represents a one-to-one relationship.
allows for more clear-sky observations and a larger percentage of clear-sky observed area compared to OMI, which allows for increased sampling of surface NO$_2$ concentrations. Population-weighted concentrations estimated here from TROPOMI at 0.025° × 0.03125° resolution are up to twice as high as similar estimates from OMI, demonstrating how fine resolution maps of NO$_2$ that more accurately describe surface NO$_2$ variability can impact exposure estimates and health studies. Land use regression models used to produce very high (sub-km) resolution surface NO$_2$ distributions for epidemiological studies will also likely benefit from having a higher resolution satellite-inferred surface concentration data set as an input.

Past studies have shown the importance of using a priori NO$_2$ profiles from high resolution models in AMF calculations for improving both the magnitude and spatial distribution of satellite retrieved vertical column densities (Russell et al. 2011, Valin et al. 2011, McLinden et al. 2014, Laughner et al. 2016, Goldberg et al. 2017, Ialongo et al. 2020, Liu et al. 2020). High resolution model information is also important when interpreting these satellite observations, particularly for NO$_2$ due to the nonlinearity of NO$_x$ chemistry (Valin et al. 2013, Goldberg et al. 2019). Model resolutions of 4–12 km$^2$ are likely needed to fully resolve nonlinear chemistry effects on NO$_2$ concentrations when estimating NO$_x$ emissions (Valin et al. 2011). However, we find good agreement between in situ measurements and our TROPOMI-inferred surface concentrations based on simulated information at ∼28 × 28 km$^2$, and tests indicate that the algorithm developed here is largely insensitive to simulation resolution. This robustness to model resolution is due to the new development of using TROPOMI columns to inform the vertical mixing assumptions used in the algorithm.

Comparisons performed here indicate that fine resolution TROPOMI-inferred surface concentrations agree well with surface observations for annual means and in winter, spring, and autumn. Monitor placement within the grid used to average satellite values combined with increased sub-grid spatial variability in the NO$_2$ field during summer can also contribute to this bias, even at the relatively fine resolution of TROPOMI (Kharol et al. 2015, Judd et al. 2019).

![Figure 5](https://example.com/figure5.png)

Figure 5. Estimates of population exposures to ambient NO$_2$ across North America. Solid lines represent satellite-inferred concentrations derived using the algorithm developed here. Dashed lines indicate concentrations derived by assuming a uniform boundary layer following Lamsal et al. (2008). Number of people exposed to concentrations exceeding Canadian Ambient Air Quality Standards (CAAQS), US National Ambient Air Quality Standards (NAAQS), and World Health Organization (WHO) guidelines are inset.
Changes to TROPOMI instrument settings made in August 2019 have improved its spatial resolution for future observations, with along-track pixel size reduced from 7 km to 5.5 km. This will allow for even greater detail in observing NO$_2$ plumes. Further improvement to satellite-inferred surface NO$_2$ concentrations can be expected from future satellite instruments that promise even higher spatial resolutions, such as the geostationary constellation of TEMPO (Zoogman et al. 2017), Sentinel-4/UVN (Ingmann et al. 2012), and GEMS (Kim 2012). These fine resolution satellite products can greatly improve air quality monitoring from space and will allow for inferring surface NO$_2$ concentrations at the fine resolution needed for improving studies on health impacts.

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Data availability

Satellite data used here are available from the NASA Goddard Earth Sciences Data and Information Services Center (TROPOMI DOI: 10.5270/S5P-s4lig54; OMI DOI: 10.567/Aura/OMI/DATA2017). The GEOS-Chem model version used here is available at DOI: 10.5281/zenodo.2658178. Population distribution data available is from the Center for International Earth Science Information Network, DOI: 10.7927/H4JW8BX5.

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References

Achakulsrisut P, Brauer M, Hystad P and Anenberg S C 2019 Global, national, and urban burdens of paediatric asthma incidence attributable to ambient NO$_2$ pollution: estimates from global datasets Lancet Planet. Health 3 E166–78

Alvarado L M A, Richter A, Vrekoussis M, Hildob A, Kalisz Hedegaard A B, Schneising O and Burrows J P 2020 Unexpected long-range transport of glyoxal and formaldehyde observed from the Copernicus Sentinel-5 Precursor satellite during the 2018 Canadian wildfires Atmos. Chem. Phys. 20 2057–72

Anenberg S C et al 2018 Estimates of the global burden of ambient PM$_{2.5}$, ozone, and NO$_2$ on asthma incidence and emergency room visits Environ. Health Perspect. 126 107004

Beloconi A and Vounatsou P 2020 Bayesian geostatistical modelling of high-resolution NO$_2$ exposure in Europe combining data from monitors, satellites and chemical transport models Environ. Int. 138 105578

Bey I, Jacob D J, Yantosca R M, Logan J A, Field B D, Fiore A M, Li Q, Liu H Y, Mckley L J and Schultz M G 2001 Global modeling of tropospheric chemistry with assimilated meteorology: model description and evaluation J. Geophys. Res. Atmos. 106 23073–95

Boersma K, Braak R and van der A R J (2011). Dutch OMI NO$_2$ (DOMINO) data product v2.0, tropospheric emissions monitoring Internet service on-line documentation (www.temis.nl/docs/OMI_NO2_HE5_2.0_2011.pdf)

Boersma K F et al 2018 Improving algorithms and uncertainty estimates for satellite NO$_2$ retrievals: results from the quality assurance for the essential climate variables (QA4ECV) project Atmos. Meas. Tech. 11 6651–78

Boersma K F, Vinken G C M and Eakes H J 2016 Representativeness errors in comparing chemistry transport and chemistry climate models with satellite UV–Vis tropospheric column retrievals Geosci. Model Dev. 9 875–98

Broccardo S et al 2018 Intra-pixel variability in satellite tropospheric NO$_2$ column densities derived from simultaneous space-borne and airborne observations over the South African Highveld Atmos. Meas. Tech. 11 2797–819

Brook J R, Burnett R T, Dann T E, Cakmak S, Goldberg M S, Fan X and Wheeler A J 2007 Further interpretation of the acute effect of nitrogen dioxide observed in Canadian time-series studies J. Expo. Sci. Environ. Epidemiol. 17 36–44

Burnett R T, Stieb D, Brook J R, Cakmak S, Dales R, Raizenne M, Vincent R and Dann T 2004 Associations between short-term changes in nitrogen dioxide and mortality in Canadian cities Can. Environ. Health 59 228–36

Chen Z-Y, Zhang R, Zhang T-H, Ou C-Q and Guo Y 2019 A kriging-calibrated machine learning method for estimating daily ground-level NO$_2$ in mainland China Sci. Total Environ. 690 556–64

Cohen A J et al 2017 Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015 The Lancet 389 1907–18

Cooper M J, Martin R V, Lyapustin A I and McLinden C A 2018 Assessing snow extent data sets over North America to inform and improve trace gas retrievals from solar backscatter Atmos. Meas. Tech. 11 2983–94

Crouse D L et al 2015 Within-and between-city contrasts in nitrogen dioxide and mortality in 10 Canadian cities: a subset of the Canadian Census Health and Environment Cohort (CanCHEC) J. Expo. Sci. Environ. Epidemiol. 25 482–9

De Hoogh K et al 2019 Predicting Fine-Scale Daily NO$_2$ for 2005-2016 Incorporating OMI Satellite Data Across Switzerland. Environ. Sci. Technol. 53 10279–87

de Laat A, Vazquez-Navarro M, Theys N and Stammes P 2020 Analysis of properties of the 19 February 2018 volcanic eruption of Mount Sinabung in S5P/TROPOMI and Himawari-8 satellite data Nat. Hazards Earth Syst. Sci. 20 1203–17

Di Q et al 2020 Assessing NO$_2$ concentration and model uncertainty with high spatiotemporal resolution across the Contiguous United States using ensemble model averaging Environ. Sci. Technol. 54 1372–84

Geddes J A, Martin R V, Boys B L and van Donkelaar A 2016 Long-term trends worldwide in ambient NO$_2$ concentrations inferred from satellite observations Environ. Health Perspect. 124 3

Goldberg D L, Lansal L N, Lougner C P, Swartz W H, Lu Z and Streets D G 2017 A high-resolution and observationally constrained OMI NO2 satellite retrieval Atmos. Chem. Phys. 17 11403–21
Goldberg D L et al 2019 Enhanced capabilities of TROPOMI NO₂: estimating NO₂ from North American cities and power plants Environ. Sci. Technol. 53 12594–601
Griffin D et al 2018 High resolution mapping of nitrogen dioxide with TROPOMI: first results and validation over the Canadian oil sands Geophys. Res. Lett. 46 1049–60
Gu J et al 2017 Ground-level NO₂ concentrations over China inferred from the Satellite OMI and CMAQ model simulations Remote Sens. 9 819
Hamra G B, Laden F, Cohen A J, Raaschou-Nielsen O, Brauer M and Loomis D 2015 Lung cancer and exposure to nitrogen dioxide and traffic: a systematic review and meta-analysis Environ. Health Perspect. 123 1107–12
Heckel A, Kim S-W, Frost G J, Richter A, Trainer M and Burrows J P 2011 Influence of low spatial resolution a priori data on tropospheric NO₂ satellite retrievals Atmos. Meas. Tech. 4 1895–20
Helfrich S R, Mcompam D, Ramsay B H, Baldwin T and Kasha C 2007 Enhancements to, and forthcoming developments in the Interactive Multisensor Snow and Ice Mapping System (IMS) HydroL. Process. 21 1576–86
Hoosly R M et al 2018 Historical (1750-2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS) Geosci. Model Dev. 11 369–408
Hu H et al 2018 toward global mapping of methane with TROPOMI: first results and inter-satellite comparison to GOSAT Geophys. Res. Lett. 45 3682–9
Hudman R C, Moore N E, Mebust A K, Martin R V, Russell A R, Valin L C and Cohen R C 2012 Steps towards a mechanistic model of global soil nitric oxide emissions: implementation and space-based constraints Atmos. Chem. Phys. 12 7779–95
Ilvalo J, Virta H, Ekes H, Holvi J and Dourou J 2020 Comparison of TROPOMI/Sentinel 5 Precursor NO₂ observations with ground-based measurements in Helsinki Atmos. Meas. Tech. 13 205–18
Ingmann P, Veihelmann B, Langen J, Lamarre D, Stark H and Courrèges-Lacoste G B 2012 Requirements for the GMES Atmosphere Service and ESA’s implementation concept: sentinel-6/5 and -5P Remote Sens. Environ. 120 58–69
Judd L M et al 2019 Evaluating the impact of spatial resolution on tropospheric NO₂ column comparisons within urban areas using high-resolution airborne data Atmos. Meas. Tech. 12 6991–111
Khalor S K et al 2015 Assessment of the magnitude and recent trends in satellite-derived ground-level nitrogen dioxide over North America Atmos. Environ. 118 236–45
Kim J 2012 GEMS (Geostationary Environment Monitoring Spectrometer) onboard the GeosKOMPASAT to monitor air quality in North Korea high temporal and spatial resolution over Asia-Pacific Region EGU General Assembly Conf. AbstractsVol. 14 p 4051
Krotkov N A et al 2017 The version 3 OMI NO₂ standard product Atmos. Meas. Tech. 10 3133–49
Lamsal L N et al 2017 High-resolution NO₂ observations from the Airborne Compact Atmospheric Mapper: retrieval and validation J. Geophys. Res. Atmos. 122 1953–70
Lamsal L N, Martin R V, Parrish D D and Krotkov N A 2013 Scaling relationship for NO₂ pollution and urban population size: a satellite perspective Environ. Sci. Technol. 47 7855–61
Lamsal L N et al 2010 Indirect validation of tropospheric nitrogen dioxide retrieved from the OMI satellite instrument: insight into the seasonal variation of nitrogen oxides at northern midlatitudes J. Geophys. Res. Atmos. 115 D20
Lamsal L N et al 2006 Ground-level nitrogen dioxide concentrations inferred from the satellite-borne Ozone Monitoring Instrument J. Geophys. Res. 111 D16308
Larkin A et al 2017 Global land use regression model for nitrogen dioxide air pollution Environ. Sci. Technol. 51 6957–64
Laughner J L, Zare A and Cohen R C 2016 Effects of daily meteorology on the interpretation of space-based remote sensing of NO₂ Atmos. Chem. Phys. 16 15247–60
Levelt P F et al 2006 The ozone monitoring instrument IEEE Trans. Geosci. Remote Sens. 44 1093–100
Levy L, Mihele C, Lu G, Narayan J J and Brook J R 2014 Evaluating multipollutant exposure and urban air quality: pollutant interrelationships, neighborhood variability, and nitrogen dioxide as a proxy pollutant Environ. Health Perspect. 122 65–72
Lin J-T and Mcelroy M B 2010 Impacts of boundary layer mixing on pollutant vertical profiles in the lower troposphere: implications to satellite remote sensing Atmos. Environ. 44 1726–39
Liu S et al 2020 An improved air mass factor calculation for nitrogen dioxide measurements from the Global Ozone Monitoring Experiment-2 (GOME-2) Atmos. Meas. Tech. 13 753–87
Lorente A et al 2017 Structural uncertainty in the air mass factor calculation for NO₂ and HCHO satellite retrievals Atmos. Meas. Tech. 10 759–82
Mcinleden C A et al 2014 Improved satellite retrievals of NO₂ and SO₂ over the Canadian oil sands and comparisons with surface measurements Atmos. Chem. Phys. 14 3637–56
Murphy T L, Jacob D J, Logan J A, Hudman R C and Koshak W J 2012 Optimized regional and interannual variability of lightning in a global chemical transport model constrained by LIS/OTD satellite data J. Geophys. Res. Atmos. Res. 117 D20
Park R J, Jacob D J, Field B D, Yantosca R M and Chin M 2004 Natural and transboundary pollution influences on sulfate-nitrate-ammonium aerosols in the United States: implications for policy J. Geophys. Res. Atmos. 109 D115
Qin K et al 2020 Satellite-based estimation of surface NO₂ concentrations over east-central China: a comparison of POMINO and OMNO2d data Atmos. Environ. 224 117522
Russell A R, Perring A E, Valin L C, Bucsela E J, Browne E C, Wooldridge P J and Cohen R C 2011 A high spatial resolution retrieval of NO₂ column densities from OMI: method and evaluation Atmos. Chem. Phys. 11 8543–54
Spurr R 2003 Area-weighting tessellation for nadir-viewing spectrometers Internal Technical Note, Harvard-Smithsonian Centre for Astrophysics, Cambridge, MA, USA
Stieb D M et al 2019 Variability in ambient ozone and fine particle concentrations and population susceptibility among Canadian health regions Can. J. Public Health 110 149–58
Thys N et al 2019 Global monitoring of volcanic SO₂ degassing with unprecedented resolution from TROPOMI onboard Sentinel-5 Precursor Sci. Rep. 9 2643
Travis K R et al 2016 Why do models overestimate surface ozone in the Southeast United States? Atmos. Chem. Phys. 16 13561–77
Valin L C, Russell A R and Cohen R C 2013 Variations of OH radical in an urban plume inferred from NO₂ column measurements Geophys. Res. Lett. 40 856–60
Valin L C, Russell A R, Hudman R C and Cohen R C 2011 Effects of model resolution on the interpretation of satellite NO₂ observations Atmos. Chem. Phys. 11 11647–55
Van der Werf G R, Randerson J T, Giglio L, Collatz G J, Mu M, Kasibhatla P S, Morton D C, Defres R S, Jin Y and Van Leeuwen T T 2010 Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009) Atmos. Chem. Phys. 10 11707–35
Van Geffen J, Folkert Boersma K, Ekes H, Smeets M, Ter I M, Zara M and Peppin Veefkind J 2020 Sentinel-5P TROPOMI NO₂ slant column retrieval: method, stability, uncertainties, and comparisons against OMI Atmos. Meas. Tech. 13 1315–35
Veefkind J P et al 2012 TROPOMI on the ESA Sentinel-5 Precursor: A GMES mission for global observations of the atmosphere and air quality EGU General Assembly Conf. AbstractsVol. 14 p 4051
atmospheric composition for climate, air quality and ozone layer applications Remote Sens. Environ. 120 70–83
Zhang Y et al 2016 Large vertical gradient of reactive nitrogen oxides in the boundary layer: modeling analysis of DISCOVER-AQ 2011 observations J. Geophys. Res. Atmos. 121 1922–34
Zhao X et al 2020 Assessment of the quality of TROPOMI high-spatial-resolution NO2 data products Atmos. Meas. Tech. 13 2131–59
Zoogman P et al 2017 Tropospheric emissions: monitoring of pollution (TEMPO) J. Quant. Spectrosc. Radiat. Transfer 186 17–39