To both reviewers:

Thank you for your constructive comments. They have helped to improve this manuscript. Here is a short description of the changes to the Figures and Tables. More in-depth comments are in the responses to each reviewer.

Figure 1 has been modified to include the UC-12 aircraft flight path.

Figure 3 has been added based on Reviewer #2's suggestion.

Figure 6 (previously Figure 5) has been modified to include NO_2 vertical profiles from GMI.

Figure 7 (previously Figure 6) has been modified to include only Pandora measurements. Figure 8a in the original manuscript is now Figure 7b. We also made some cosmetic changes, which include adding the standard deviation.

Figure 8 has been added based on Reviewer #1's suggestion.

Figure 9 (previously Figure 8) has been modified to include only EPA measurements. Figure 6b in the original manuscript is now Figure 9a. We also made some cosmetic changes.

Table 1 has been added. It compares the OMI NO_2 retrieval developed during this study to other OMI NO_2 retrievals.

Anonymous Referee #1

Review of "A High-Resolution and Observationally Constrained OMI NO2 Satellite Retrieval", ACPD, by Daniel L. Goldberg, Lok N. Lamsal, Christopher P. Loughner, Zifeng Lu and David G. Streets

The current paper presents a modified product of tropospheric NO2 columns during the Maryland 2011 DISCOVER-AQ. One of the major uncertainties of retrieving the vertical NO2 columns originate from the calculation of AMFs. The uncertainty of AMF might resulted from several inputs, but here, the main focus is on the shape factors. The authors claimed that a high resolution model output can potentially distinguish highly polluted regions from others, because they are roughly mixed in the original OMI product. The authors further brought up an important issue in CMAQ that is its large underprediction of NO2 in the free troposphere. This issue has nicely been addressed by using P3-B measurements to constrain the CMAQ NO2 profiles. Finally, the authors made use of CMAQ to downscale OMI tropospheric NO2 columns to provide a very high resolution "map" using the method of Kim et al., [2016]. Although there is not a great deal of effort to advance the retrieval process, I believe that this will be interesting for environmental agencies who are looking for a very fine product, particularly for the use of health impacts. This manuscript in its present form requires significant improvement before being acceptable for publication in ACP.

*The first major problem with this study is the lack of adequate comparison in terms of magnitude. I am aware that the authors used the Pandora measurements, but the comparison might have been influenced by errors in OMI stratospheric NO2 columns (which had been added to the tropospheric ones to conduct an apples-to-apples comparison with Pandora). Since you are using a more spatially detailed model to estimate shape factors, the improvement in correlation is expected. However, it is imperative to use other products such as ACAM [Lamsal et al., 2017, JGR] to show if the new product will get closer to observations with small footprints. My suggestion will become more serious for your last product (OMI_CMAQ_OD). This is a pseudo observation based on integrating a model and an observation. Thus, its accuracy should be more carefully verified.

We conducted a comparison with the ACAM data. The ACAM NO_2 product is introduced in section 2.2.3 and the analysis is described in section 3.4.2. Figure 8 describes the requested comparison: ACAM NO_2 vs. the OMI_GMI and our new OMI_CMAQ_OD NO2 product. The flight path of the UC-12 aircraft carrying the ACAM has been added to Figure 1.

*My second major concern is about overlooking the impact of accurate NO2 (and other gases) during estimating the scattering weights. Two parts that the priori NO2 values from model are used during the retrieval are i) estimating the jacobian values from a radiative transfer model and ii) calculating the shape factors. Why the first step was not performed or not even mentioned in this paper? Would considering a more accurate a NO2 priori profile lead to a better estimation of scattering weights? This may be investigated using VLIDORT.

For this study, we follow previous studies and assume that scattering weights are a function of a single a priori NO_2 profile (e.g., Palmer et al., 2001, Martin et al., 2002, Boersma et al., 2011, Bucsela et al., 2013). Therefore, we assume scattering weights and NO_2 shape factors are independent. We are aware that at very high NO_2 concentrations this assumption may not be fully valid. However, this is a novel and emerging research topic that is beyond the scope of this paper.

We now clarify this in Section 2.1 to say:

"The optical atmospheric/surface properties are characterized by the scattering weight (SW) and are calculated by a forward radiative transfer model (TOMRAD) which are output as a look-up table (LUT). The SWs are then adjusted real-time depending on observed viewing angle, surface albedo, cloud fraction, and cloud height. For this study, we follow previous studies (e.g., Palmer et al., 2001, Martin et al., 2002, Boersma et al., 2011, Bucsela et al., 2013) and assume that SWs and NO_2 profile shapes are independent."

My specific comments follow:

*P2, Line 18. "to generate tropospheric air mass factors..." might be changed to "to re-calculate" or "to modify" OMI tropospheric air mass factors.

This has been changed as suggested.

***P2, Line 24.** How about the bias compared to ACAM or other air-borne observations?

This has been added to the abstract and addressed more fully in Section 3.4.2

***P3, Line 13.** "RO2" instead of "HO2" would be broader. You might need to define it in parentheses.

RO_2 is now included and defined. HO_2 is now also defined.

***P4, Line10.** I suggest the authors mention about the recent changes in China in 2011- 2012 [Souri et al., 2017] mostly due to using SCR for power plants. Souri, A.H., Choi, Y., Jeon, W., Woo, J.H., Zhang, Q. and Kurokawa, J.I., 2017. Remote sensing evidence of decadal changes in major tropospheric ozone precursors over East Asia. Journal of Geophysical Research: Atmospheres, 122(4), pp.2474-2492.

A short clarification on the increasing OMI NO_2 trend in China before 2011, stabilization in 2011-2012, and decreases since 2012 has been added. The Souri et al. reference has also been added.

We now state: "Over this 10-year period China has seen a reversal of its trends: during 2005-2010 OMI NO2 tropospheric columns were increasing (Verstraeten et al., 2015), in 2011-2012 they had stabilized (Souri et al., 2017), and since 2012 they have subsequently decreased as the country enforces its Twelfth 5-year plan (de Foy et al., 2016b)."

***P4, Line 14.** How about the nadir-spectrometers like TES? This sentence might be revised. The footprint of surface concentrations exists in OMI signal. But it is not easy to separate it. The current sentence leaves readers with an impression that the radiance has not been impacted by the surface concentrations at all.

We have clarified this sentence to say:

"Remote sensing instruments typically measure the entire column content instead of in situ concentrations at individual vertical levels. Being able to derive surface concentrations from column content information would be very useful for the policy-making and health-assessment communities."

***P4, Line35.** You might want to elaborate their works in the introduction. ▪

We have added a full paragraph at the end of the Introduction explaining these three retrievals and their important findings. We have also now included this information in Table 1.

***P5, Line 22.** How was the stratospheric slant column subtracted from total column in OMI? CTMs or based on the OMI radiance?

The stratospheric slant column was subtracted based on OMI radiance (Bucsela et al., 2013). This has been clarified in the text:

"Stratospheric SCD… is inferred using a local analysis of the stratospheric field (Bucsela et al., 2013)"

***P5, Line 26. I would suggest adding Martin et al., 2002 for NO2 shape profile.

This reference has been added as suggested.

***P5, Line 30. Please provide references. I am assuming that scattering weights are already stored in a six-dimension LUT, and for partially cloudy pixels, a lambertian surface with albedo equal to 0.8 is assumed, then they combine the results (cloudy and clear) using the IPA. Is the new product different from this?

Yes, the scattering weights are stored in a look-up table.

This has been clarified in the text to say: "The optical atmospheric/surface properties are characterized by the scattering weight (SW) and are calculated by a forward radiative transfer model (TOMRAD), which are output as a look-up table (LUT). The SWs are then adjusted real-time depending on observed viewing angle, surface albedo, cloud fraction, and cloud pressure. For this study, we follow previous studies (e.g., Palmer et al., 2001, Martin et al., 2002, Boersma et al., 2011, Bucsela et al., 2013) and assume that SWs and NO2 profile shapes are independent."

***P6, Line 1. Please clarify whether the profile from GMI model is constant over time. You may need to mention: "It should be noted that a blue light converted which selectively photolyzes NO2 was used for P3-B. As a result, there was no need to modify NO2 concentration by applying an empirical equation from [Lamsal et al., 2008]."

This has been clarified to state a "monthly-averaged and year-specific" simulation was used.

The second part of this comment appears to be in reference to P6, Line 27. In this section, we have added a clarification that the Cohen group instrument does not suffer from the same positive bias as chemiluminescence detectors, as suggested.

***P7, Line 4. NO2 varies quickly by time, and using a short duration is more appropriate, because OMI captures NO2 just in a matter of milliseconds. Please check whether reducing the time average will make the comparisons better.

We have found that +/- 1 hour is the "sweet spot", in which there is minimal compromise in temporal matching, while also maintaining a large enough sample size. Typically, winds are 10 – 20 km/hr, so in essence, a pixel 20 – 40 km wide will be captured within a 2 hour window (assuming NO_2 remains relatively constant over the 2-hour window). If we shorten the averaging time, then we severely limit the number of samples, which are already small. This means that if the Pandora instrument is sampling a local plume (or lack thereof) not representative of the nearby environment, then this will cause an unfair comparison.

***P8, Line3. Please specifically mention which scheme was used for the biogenic emissions. Did the authors consider the soil NOx emissions?

BEISv3.14 was used for the biogenic emissions. The soil NOx emissions parameterization was not released until after completion of this CMAQ simulation, so no soil NOx emissions are included in this CMAQ simulation. Both are clarified in the text.

*****Figure 2. Why no observations were shown? In the text, you claimed that CMAQ has a better performance compared to observations (P9, line1).

Surface observations are shown on Figure 2 and are denoted by the black triangle. This is the original basis of the claim. However, we realized that this claim was very tenuous at best, so we have now compared CMAQ, GMI, and the P3-B aircraft observations on Figure 5. This figure demonstrably illustrates that CMAQ is better at simulating NO_2 in urban areas than GMI.

*****P9, Line 10. I am not sure both model used the same lightning NOx option. The way they treat lightning might differ. There exit myriad of reasons for the underprediction of CMAQ NO2. It can be related to vertical mixing, uncertainty from NOx aviation emissions, stratospheric sources, or lightning. The vertical allocation of emissions are also different. If you had used the GMI for the chemical boundary conditions of CMAQ, it would have been easier to compare them.

We have no re-phased the section to say:

"To determine whether lightning NO is the primary driver of this difference, we compare lightning NO emissions from both model simulations in Figure 3. The CMAQ model ingests lightning NO emissions that are an order of magnitude larger than the GMI simulation at most altitudes. This is likely due to WRF simulating more convective precipitation and higher cloud-top heights, both input variables to the lightning NO parameterization, than GMI. Therefore, the smaller magnitude of free tropospheric NO2 in CMAQ does not arise from the lightning NOx parameterization, but instead from a combination of the chemistry, aviation emissions, vertical mixing, long-range transport, and stratospheric-tropospheric exchange."

*****P9, Line 19. Poor grammar.

This has been corrected.

*****P9, Line23. This is a very important message. It means the poor performance of CMAQ in simulating NO2 in free troposphere will make a challenge for the retrieval purposes. We may have to use the aircraft to constrain it, or to use GMI models at those specific altitudes. You may want to highlight it in the conclusion.

Yes, this is an extremely important point, and perhaps was not highlighted enough in the original manuscript. This is now mentioned in the abstract and is highlighted again in the conclusion. In the conclusion we state: "… the poor performance of CMAQ (or any model used for a satellite retrieval) will manifest itself in the retrieval. This will be a difficult challenge going forward, and emphasizes the need to use state-of-the science models for satellite retrievals."

*P11, Line 18. Please explain why it is rudimentary (i.e., not considering the errors in observations, model and the priori).

We have revised to say "simplified" instead of "rudimentary".

*P11, Line 30. The discussion is not enough. Please explain the possible reasons of large differences between OMI_CMAQ and OMI_CMAQ_O. Were AMFs enhanced largely due to larger shape factors in the free troposphere?

A short discussion has now been included at the end of Section 3.3.2.

"The large reduction in NO2 tropospheric vertical columns between OMI_CMAQ and OMI_CMAQ_O is an outcome of using larger AMFs. The larger AMFs are a result of the original overestimate within the boundary layer and underestimate in the free troposphere. This is a particularly important finding because it means that a model with large biases in the simulation of NO2 can yield poor tropospheric vertical column contents, despite high spatial resolution. This emphasizes the need to evaluate the emissions and chemistry of a model before it should be used for satellite retrievals."

*P12, Line 28. I don't agree with your sentence "OMI can now "see" ...". This is an illusion. You used the model to downscale the values. This is not the OMI; it is the model that provided a tool to concentrate the observations. I would call it a pseudo observation or simply a "map". We have to clarify that OMI footprint is too coarse to see these plumes. That's why we need TEMPO and TROPOMI. This paragraph should be revised or be dropped.

This has been revised to say the "spatially downscaled OMI product" instead of OMI

*P15, Line 30. It is not only about the emissions, but also the meteorological fields. Simulating surface winds in many situations is not straightforward. So the winds may be off in the model resulting in wrong distribution of final product.

This has been clarified in the text to say: "… if the area is affected by a mesoscale meteorological feature that is simulated incorrectly by the model, such as a thunderstorm, valley breeze, or sea breeze, the model will be similarly deficient. Therefore, we do not recommend using the downscaling technique in areas where the emission inventory or meteorology is very uncertain."

Anonymous Referee #2

The paper by Goldberg et al. provides an interesting study of using high-resolution CMAQ, vertical profile observations and data sampling techniques to better estimate NO2 VCDs at small scales. The paper is well written, and I have a few suggestions below.

*Recent studies have revealed NO2 retrieval uncertainties related to structural errors (Lorente et al., 2017 and references therein), including treatments of clouds and aerosols (Lin et al., 2015). These errors are relevant to explanations of errors even in OMI_CMAQ_OD. A review of such works is necessary.

This has been addressed in the paragraph added to the end of the Introduction. We've also now included a table (Table 1), which compares all OMI NO_2 retrievals in the literature.

*The spatial and temporal matching between CMAQ and OMI is discussed in many places, and sometimes there appears inconsistency [For example, Sect. 2.1 says 'The satellite product was oversampled for June & July over a 5-year period (2008-2012) by re-gridding to the CMAQ 1.33 km model grid and then averaging the data over the 10-month (two months × five years) period.', but Sect. 2.4 says ' To ensure a fair comparison, we average model information to the pixel size.'] Please provide a paragraph in the method section dedicated to data mapping/sampling, including proportioning of pixel-based SWs to CMAQ grid, and refer to this section when mentioning in later sections.

We have added a new section (Section 2.5), in which this is clarified.

*Please describe the model setup (e.g., soil and lightning emissions, vertical layers, model PBL scheme, convection, upper boundary) in Sect. 2.4. This will much help understand the model vertical profiles. The missing soil emissions are not discussed until the line (P14, L1) embedded in Section 3.5.

All of these have now been included in Section 2.4

*Can you compare CMAQ and GMI lightning emissions? I wonder how much of the vertical profile differences are due to lightning (convection) parameterization rather than due to resolution.

We have now added a figure (Figure 3) comparing the lightning NO emissions from the two models. Although GMI simulates larger free tropospheric NO_2, lightning NO emissions are smaller in GMI. Therefore the differences in free tropospheric NO_2 must arise from an alternative mechanism. Further analysis is beyond the scope of this paper. The end of Section 3.1 is re-phrased as such:

"To determine whether lightning NO is the primary driver of this difference, we compare lightning NO emissions from both model simulations in Figure 3. The CMAQ model ingests lightning NO emissions that are an order of magnitude larger than the GMI simulation at most altitudes. This is likely due to WRF simulating more convective precipitation and higher cloud-top heights, both input variables to the lightning NO parameterization, than GMI. Therefore, the differences in free tropospheric NO2 between the two models likely arise from a combination of the chemistry, aviation emissions, vertical mixing, long-range transport, and stratospheric-tropospheric exchange."

*****That model profiles in June/July 2011 are applied to all years needs to be described more clearly in Sect. 2.4. The writing is vague at its current form. Some of the writing on relevant method in the first paragraph of Sect. 3.3.1 should be included in Sect. 2.4. The uncertainty due to interannual variability needs to be discussed.

This has been clarified in our new section (Section 2.5) to state:

"For years other than 2011, we used 2011 monthly mean values of NO2, temperature, and tropopause pressures for the calculation of the AMF."

*****At the end of 'Introduction', a summary paragraph showing the novelty of the present study will be very useful.

This has now been included at the end of the Introduction. We have also now included a new table (Table 1), which succinctly describes our study in relation to the previous studies.

At the end of the Introduction, we now state: "We build upon these studies by using an even higher resolution regional air quality model (1.33 km) to generate air mass factors. We use the mid-Atlantic region in the eastern United States as a case study in developing high resolution NO2 tropospheric columns for urban metropolitan areas. Furthermore we utilize a technique for constraining the NO2 shape profiles to aircraft observations and invoke a new downscaling method developed by Kim et al., (2016)."

*****P3, L17 – NO2 is a weak absorber.

This has been modified to say: "NO2 has strong absorption features within the 400 – 450 nm wavelength region…"

*****P5, L1 – POMINO does not just provide a higher-resolution retrieval, but it also includes various improvements such as explicit treatment of aerosols and re-calculation of cloud parameters.

This has now been included in the Introduction. We have also now included a new table (Table 1), which describes POMINO in relation to our study."

*****P5, L19-20 – SCD represents light path from the sun to surface/atmosphere and to the instrument.

This has been modified as suggested.

*****P5, L25 – the effects of aerosols are also important.

Aerosol optical depth has been added here.

*****P8, L34 – how to determine the 'best estimate'. I appears that if a 65% overestimate is assumed, the OMI_GMI result would be closer to EPA values.

The "best estimate" was determined based on the Dunlea et al. 2007 study referenced in Section 2.2.4, which suggests actual NO_2 is 22% lower than chemiluminescence measurements in an urban environment . Lamsal et al., 2008 suggests this number can be up to 65%. Thus we include a range of 3.7 – 10.5 ppb. The CMAQ estimate is within this range, but the GMI estimate is not.

This has been clarified in the text to state: "… the corrected surface NO_2 mixing ratio is approximately 22% lower (but may be up to 65% lower) than observed NO_2*"

*****P11, L9 – should be 'consistently larger'

Thank you for catching this. It has been modified.

*****P12, L19-26 – much of the discussion on 'rural' and 'urban' definitions here applies also to discussion in previous sections (i.e., Sect. 3.1) on these environments.

This paragraph has been shortened and moved to the Discussion section.

References: Lorente, A., Folkert Boersma, K., Yu, H., Dörner, S., Hilboll, A., Richter, A., Liu, M.-Y., Lamsal, L. N., Barkley, M., De Smedt, I., Van Roozendael, M., Wang, Y., Wagner, T., Beirle, S., Lin, J.-T., Krotkov, N., Stammes, P., Wang, P., Eskes, H. J., and Krol, M.: Structural uncertainty in air mass factor calculation for NO2 and HCHO satellite retrievals, Atmospheric Measurement Techniques, 10, 759-782, doi:10.5194/amt- 10-759-2017, 2017.

This reference has been added to the text.

A HIGH-RESOLUTION AND OBSERVATIONALLY CONSTRAINED OMI NO_2 SATELLITE RETRIEVAL

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A high-resolution and observationally constrained OMI NO_2 satellite retrieval

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Abstract. This work presents a new high-~~ ~~resolution NO_2 dataset derived from the ~~standard~~ NASA Ozone Monitoring Instrument (OMI) NO_2 version 3.0 retrieval that can be used to estimate surface level concentrations. The standard NASA product uses NO_2 vertical profile shape factors from a 1.25° × 1° (~110 × 110 km) resolution Global Model Initiative (GMI) model simulation to calculate air mass factors, a critical value used to determine observed tropospheric NO_2 vertical columns. To better estimate vertical profile shape factors, we use a high-resolution (1.33 × 1.33 km) Community Multi-scale Air Quality (CMAQ) model simulation ~~(1.33 × 1.33 km)~~ constrained by in situ aircraft observations to ~~generate re-tropospheric~~calculate tropospheric air mass factors and tropospheric NO_2 vertical columns during summertime in the eastern United States. ~~Results show~~ In this new product, OMI NO_2 tropospheric columns ~~in this new product~~ increase by up to 160 % in city centers, and decrease by 20 – 50 % in the rural areas outside of urban areas when compared to the operational NASA product. ~~This~~ Our new product shows much better agreement with the Pandora NO_2 and Airborne Compact Atmospheric Mapper (ACAM) NO_2 spectrometer measurements acquired during the DISCOVER-AQ Maryland field campaign. Furthermore, the correlation between ~~this~~ our satellite product and EPA NO_2 monitors in urban areas has improved dramatically: r^2 = 0.60 in new product, r^2 = 0.39 in operational product, signifying that this new product is a better indicator of surface concentrations than the operational product. Our work emphasizes the need to use both ~~high resolution~~high-resolution ~~models~~ and high-fidelity models in order to re-calculate satellite data in areas with large spatial heterogeneities in NO_x emissions. Although the current work is focused on the eastern United States, the methodology developed in this work can be applied to other world regions to produce high-quality region-specific NO_2 satellite retrievals.

1 Introduction

Tropospheric NO_2 is a trace gas toxic to human health and during ideal atmospheric conditions can photolyze to create O_3 another toxic air pollutant with a longer atmospheric lifetime. The eventual fate of tropospheric NO_2 is often HNO_3, a chemical species easily dissolved in water and responsible for acid rain. HNO_3 can also react with ammonia to create nitrate aerosols, which contribute to haze and are harmful to human health.

There are some natural sources of nitrogen oxides ($NO_x \equiv NO+NO_2$), such as from soil through microbial nitrification and denitrification (Conrad, 1996), lightning (Ridley et al., 1996), and natural wildfires (Val Martin et al., 2006), but the majority of the NO_2 in our atmosphere today originates from anthropogenic sources (van Vuuren et al., 2011). When temperatures are greater than 1500 K, such as in fuel combustion, nitrogen (N_2) and oxygen (O_2) spontaneously react to create NO via the endothermic Zeldovich mechanism. The nitrogen in fuels are also converted to NO during combustion making fuels rich in nitrogen, such as coal, more efficient ~~at~~ in creating NO. NO is quickly oxidized to NO_2 in the atmosphere, most often by ozone, in a matter of seconds. Thus the NO and NO_2 species are often grouped into a single species called NO_x. In the presence of hydroperoxy (HO_2) or ~~,~~ organic peroxy radicals (RO_2, where R is any organic group), NO can also be oxidized to NO_2 without consuming ozone. This is the rate-limiting step in the chemical chain reaction producing tropospheric ozone.

NO_2 ~~is a strong absorber of radiation~~has strong absorption features within the 400 – 450 nm wavelength region (Vandaele et al., 1998), which approximately corresponds to violet visible light. Satellite instruments measure the absorption of solar backscatter in the UV-visible spectral range, enabling estimation of the amount of NO_2 in the atmosphere between the instrument and the surface. By comparing observed spectra with a reference spectrum, we can derive total column amounts; this technique is called differential optical absorption spectroscopy (DOAS) (Platt, 1994).

NO_2 has been continuously measured from satellites for over two decades now. The first instrument to remotely measure NO_2 was the Global Ozone Monitoring Experiment (GOME) launched aboard the European Remote Sensing 2 (ERS-2) satellite in April 1995 (Burrows et al., 1999). Despite its coarse temporal and~~-~~ spatial resolution (global coverage once every three days and pixel size of 40 × 320 km), it was the first remotely sensed instrument to characterize NO_2 columns from space, showing enhanced tropospheric NO_2 over North America and Europe (Martin et al., 2002; Martin et al., 2003). In the early 2000s, Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) (Bovensmann et al., 1999) and Ozone Monitoring Instrument (OMI) (Levelt et al., 2006; Bucsela at el., 2006; Boersma et al., 2007) became additional space-based instruments to measure NO_2. These instruments were designed to achieve better spatial resolution (SCIAMACHY: 30 × 60 km, OMI: 13 × 24 km) than GOME. Boersma et al. (2008a) documented the differences between the two retrievals. In early 2012, ground operators lost contact with SCIAMACHY, but OMI is still operational as of 2017. There are two operational OMI NO_2 retrievals: the KNMI DOMINO v2.0 product (Boersma et al., 2007) and the NASA OMNO2 v3.0 product (Krotkov et al., 2017).

OMI NO_2 has been used to estimate NO_x emissions from various areas around the globe (Streets et al., 2013) including North America (Boersma et al., 2008b; Lu et al., 2015), Asia (Zhang et al., 2008; Han et al, 2015; Kuhlmann et al., 2015), the Middle East (Beirle et al., 2011), and Europe (Huijnen et al., 2010; Curier et al., 2014). It has also been used to generate and validate NO_x emission estimates from source sectors such as soil (Hudman et al., 2010; Vinken et al., 2014a; Rasool et al., 2016), lightning (Allen et al., 2012; Liaskos et al., 2015; Pickering et al., 2016), power plants (de Foy et al., 2015), aircraft (Pujadas et al., 2011), marine vessels (Vinken et al., 2014b; Boersma et al., 2015), and urban centers (Lu et al., 2015; Canty et al., 2015; Souri et al., 2016). More recently, there has been an emphasis on analyzing emission trends because OMI has been retrieving high-quality tropospheric NO_2 data for over ten years. Over this decade, some areas have seen increases, such as India (Lu and Streets, 2012), ~~China (Verstraeten et al., 2015),~~ the Canadian oil sands region (McLinden et al., 2015), and other oil extraction regions (Duncan et al., 2016), while areas such as the eastern United States (Russell et al., 2012; Lamsal et al., 2015; Krotkov et al., 2016; de Foy et al., 2016a) and Europe (Curier et al., 2014; Duncan et al., 2016) have seen large decreases due to a switch to cleaner fuels and the implementation of emission control technologies. Over this 10-year period, China has seen a reversal of its trends: during 2005-2010 OMI NO_2 tropospheric columns were increasing (Verstraeten et al., 2015), in 2011-2012 they had stabilized (Souri et al., 2017), and since 2012 they have subsequently decreased as the country enforces its Twelfth 5-year plan (de Foy et al., 2016b).

Remote sensing instruments typically measure the entire column content instead of in situ concentrations at individual vertical levels. Being able to derive surface concentrations from column content information would be very useful for the policy-making and health-assessment communities. ~~One of the reasons why the usage of satellite data remains tepid in policy-making communities is due to their inability to detect surface concentrations; improving this ability may spur their use~~In particular, detecting the spatial heterogeneities of NO_2 in and around city centers are of strong interest as many people are exposed to NO_2 or co-located pollutants exceeding policy thresholds in these areas. Satellite measurements with spatial resolution > 13 km, such as OMI, have difficulty observing the fine structure of NO_2 plumes at or near the surface (e.g., highways, power plants, factories, etc.) (Chen et al., 2009; Ma et al., 2013; Flynn et al., 2014), which are often less than 10 km in width (Heue et al., 2008). This can lead to a spatial smoothing of pollution, which does not exist in reality (Hilboll et al., 2013). Remote sensing instruments with finer spatial resolution, such as TROPOMI (Veefkind et al., 2012) and TEMPO (Zoogman et al., 2017), may be able to resolve this issue.

Until the next generation of satellites is launched, there have been several techniques to modify OMI NO_2 data a posteriori. Kim et al. (2016) developed a technique in which users can utilize regional air quality model information to spatially downscale OMI NO_2 measurements. This technique has shown to increase the variability of OMI NO_2 within urban areas, which is in better agreement with observations in these regions. In another effort to merge model and satellite data, Lamsal et al. (2008) was able to infer surface level NO_2 concentrations from OMI NO_2 by applying local scaling factors from a global model. There has also been an emergence of a technique that combines land-use regression techniques with satellite information to infer ground-level NO_2 concentrations (Novotny et al., 2011; Vienneau et al., 2013; Lee et al., 2014; Bechle et al., 2015; Young et al., 2016). While each individual technique is useful, all of the aforementioned techniques use model data to adjust existing satellite data, but do not address issues inherent with the satellite retrieval methodology.

Previous studies have shown that the air mass factor, a value needed to convert the slant column measurement into a vertical column amount, is one of the largest source of uncertainty in the OMI NO_2 retrieval, contributing up to half of the total error (Boersma et al., 2004; Lorente et al., 2017). There are ~~three~~two existing OMI NO_2 products that ~~follow a similar procedure~~use information from a regional chemical transport model to re-calculate the air mass factor: Berkeley High-Resolution (BeHR) NO_2 ~~for the United States~~ (Russell et al., 2011; Laughner et al., 2016)~~, POMINO for China (Lin et al., 2015),~~ and City University of Hong Kong OMI (HKOMI) ~~OMI~~ NO_2 ~~for the Pearl River Delta region of China~~ (Kuhlmann et al., 2015). BeHR NO_2 uses a monthly averaged 12 × 12 km Weather Research and Forecasting coupled with Chemistry (WRF-Chem) model simulation with higher resolution terrain pressure and Moderate-resolution Imaging Spectroradiometer (MODIS) black-sky albedo to re-calculate the air-mass factors for the United States. This study found that by updating the air mass factors with a high-resolution simulation, NO_2 tropospheric vertical columns increased in urban areas and decreased in rural areas when compared to typical satellite products processed with global model simulations (Russell et al., 2011). More recently, the BeHR NO_2 product has been updated (summer 2013 only) to account for daily variations in shape profiles and terrain pressure, which modifies daily retrievals by as much as 40% (Laughner et al., 2016). The HKOMI product uses NO_2 shape profile, terrain elevation, and meteorological information from a 3 × 3 km a Community Multiscale Air Quality (CMAQ) simulation coupled offline to a Weather Research and Forecasting (WRF) simulation to re-calculate the air mass factor for the Pearl River Delta region of China. Similarly, they found that the tropospheric vertical NO_2 columns increased in an urban area; this improved agreement between satellite and ground observations (Kuhlmann et al., 2015). One critical limitation of the BeHR and HKOMI products is the lack of lightning NO_x emissions in the model simulations used to derive the air mass factor. The POMINO product takes a slightly different approach. This study improves the air mass factor for China (Lin et al., 2015) by (1) using improved information on surface reflectivity (MODIS Bidirectional Reflectance Distribution Function (BRDF)), (2) improving the treatment of aerosols and cloud pressure/fraction, and (3) using a nested (0.667° × 0.5°) GEOS-Chem simulation for the NO_2 shape profiles. These three changes increase annual mean NO_2 tropospheric vertical columns by 15 – 40%. A summary of all available OMI NO_2 retrievals are listed in Table 1.

We build upon these studies by using an even higher resolution (1.33 km) regional air quality model ~~(1.33 km)~~ to generate air mass factors for urban metropolitan areas in the mid-Atlantic region of the eastern United States~~, a value needed for calculation of tropospheric vertical column NO_2 amounts.~~ Use of such resolution allows calculation of air mass factors representing OMI ground pixels. The new air mass factors are then used to re-calculate NO_2 tropospheric vertical columns. ~~We use a small region in the eastern United States as a case study in developing high resolution NO_2 tropospheric columns for urban metropolitan areas (200 × 200 km).~~Furthermore we utilize a technique for constraining the NO_2 shape profiles to aircraft observations and invoke a new downscaling method developed by Kim et al., (2016) to enhance the content of the satellite observations.

2 Methods

~~We use a method to re-calculate satellite data in lieu of using the operational product as is. There are three existing NO_2 products that follow a similar procedure: BeHR for the United States (Russell et al., 2011; Laughner et al., 2016), POMINO for China (Lin et al., 2015), and HKOMI for the Pearl River Delta region of China (Kuhlmann et al., 2015). We build upon these studies by using an even higher resolution regional air quality model (1.33 km) to generate air mass factors, a value needed for calculation of tropospheric vertical column NO_2 amounts. We use a small region in the eastern United States as a case study in developing high resolution NO_2 tropospheric columns for urban metropolitan areas (200 × 200 km).~~

2.1 OMI NO_2

The Ozone Monitoring Instrument (OMI) has been operational on NASA's Earth Observing System (EOS) Aura satellite since October 2004 (Levelt et al., 2006). The satellite follows a sun-synchronous, low-earth (705 km) orbit with an equator overpass time of approximately 13:45 local time. OMI measures total column amounts in a 2600 km swath divided into 60 unequal area "field-of-views", or pixels. At nadir (center of the swath), pixel size is 13 × 24 km, but at the swath edges, pixels can be as large as 26 × 128 km. In a single orbit, OMI measures approximately 1650 swaths and achieves daily global coverage over 14 – 15 orbits (99 minutes per orbit). OMI measures solar backscatter within the 270-500 nm wavelength range. For this paper, we focus on the NO_2 retrieval which is derived from measurements in the 400 – 450 nm range. Since June 2007, there has been a partial blockage of the detector's full field of view, which has limited the number of valid measurements; this is known in the community as the row anomaly ~~(RA)~~: http://projects.knmi.nl/omi/research/product/rowanomaly-background.php.

OMI measures radiance data between the instrument's detector and the Earth's surface. Comparison of these measurements with a reference spectrum (i.e., DOAS technique), allows for calculation of the total slant column density (SCD), which represents the integrated NO_2 abundance from the sun to the surface, through the atmosphere, to the instrument's detector. For tropospheric air quality studies, vertical column density (VCD) NO_2 data are more appropriate. This is done by subtracting the stratospheric slant column from the total (tropospheric + stratospheric) slant column and dividing by the tropospheric air mass factor (AMF), which is defined as the ratio of the SCD to the VCD, as shown in Eq. (1):

$$VCD_{trop} = \frac{SCD_{total} - SCD_{strat}}{AMF_{trop}} \quad \text{, where } AMF_{trop} = \frac{SCD_{trop}}{VCD_{trop}} \qquad (1)$$

The tropospheric AMF has been derived to be a function of the optical atmospheric/surface properties (surface ~~albedo~~reflectivity, aerosol optical depth, cloud fraction, and cloud height) and a priori NO_2 shape profile (Palmer et al., 2001; Martin et al., 2002) and can be calculated as follows (Lamsal et al., 2014) in Eq. (2):

$$AMF_{trop} = \frac{\sum_{surface}^{tropopause} SW \times x_a}{\sum_{surface}^{tropopause} x_a} \qquad (2)$$

Where x_a is the partial column NO_2. The optical atmospheric/surface properties are characterized by the scattering weight (SW) and are calculated by a forward radiative transfer model (TOMRAD~~) in the NASA product~~), which are output as a look-up table (LUT). The SWs are then adjusted real-time ~~by NASA~~ depending on observed viewing angles, surface ~~albedo~~reflectivity, cloud fraction, and cloud ~~height~~pressure. For this study, we follow previous studies (e.g., Palmer et al., 2001, Martin et al., 2002, Boersma et al., 2011, Bucsela et al., 2013) and assume that SWs and NO_2 profile shapes are independent. The a priori NO_2 ~~shape~~ profile shapes (x_a) must be provided by a model simulation. In an operational setting, NASA uses ~~the~~ a monthly-averaged and year-specific Global Model Initiative (GMI) model (1.25° lon × 1° lat; ~110 km × 110 km in the mid-latitudes) simulation to provide the a priori NO_2 shape profiles. ~~Instead of using a global model~~For this study, we derive tropospheric VCDs using a priori NO_2 shape profiles from a regional CMAQ simulation. A description of this methodology is included in Section 2.5. All other parameters from the NASA Level 2 product including the total SCD, stratospheric SCD (which is inferred using a local analysis of the stratospheric field (Bucsela et al., 2013)), surface reflectivity (which is derived from OMI Lambert Equivalent Reflectance (LER) (Kleipool et al., 2008)), and SW remain unchanged.

~~For this study, w~~We filter the Level 2 OMI NO_2 data to ensure only valid pixels are used. ~~We remove d~~Daily pixels with solar zenith angles ≥ 80°, cloud radiance fractions ≥ 0.5, or surface ~~albedo~~ reflectivity ≥ 0.3 are removed as well as the~~. Furthermore, we remove the~~ five largest pixels at the swath edges (i.e., pixel numbers 1 – 5 and 56 – 60). Finally, we remove any pixel flagged by NASA including pixels with NaN values, 'XTrackQualityFlags' ≠ 0 or 255 (RA flag), or 'VcdQualityFlags' > 0 and least significant bit ≠ 0 (ground pixel flag). ~~The satellite product was oversampled for June & July over a 5-year period (2008-2012) by re-gridding to the CMAQ 1.33 km model grid and then averaging the data over the 10-month (two months × five years) period. We have chosen the June & July timeframe because the CMAQ simulation and DISOCVER-AQ Maryland data are only available during these two months.~~

2.2 DISCOVER-AQ NO_2 observations

In the validation of our new satellite product, we use in situ NO_2 observations from the DISCOVER-AQ Maryland field campaign. DISCOVER-AQ was a four-part field experiment designed to probe the atmosphere near urban areas in excruciating detail from aircrafts, ground station networks, and satellites. The first experimental campaign took place in Maryland (Baltimore, MD - Washington D.C. area) in July 2011. This campaign was particularly unique for an aircraft field campaign in that the focus was limited to single metropolitan area, whereas in other aircraft campaigns, spatial coverage is often over a larger domain. We utilize data acquired by ~~three~~ four sources during this campaign: the P3-B aircraft, the ground-based Pandora spectrometer network, the Airborne Compact Atmospheric Mapper on the UC-12 aircraft, and the long-term EPA ground monitoring network. A typical P3-B aircraft and UC-12 flight path, Pandora NO_2 spectrometer locations, and ground monitor locations are shown in Figure 1. DISCOVER-AQ observations were retrieved from the online data archive: http://www-air.larc.nasa.gov/cgi-bin/ArcView/discover-aq.dc-2011. A further description of DISCOVER-AQ Maryland can be found in Crawford et al. (2014).

2.2.1 P3-B aircraft data

We use P3-B aircraft NO_2 data gathered by the Cohen group (instrument reference: (Day et al, 2002)) to assess the accuracy of our model simulation. This instrument does not have the same positive bias as chemiluminescence NO_2 detectors, so there is no need to modify NO_2 concentrations by applying an empirical equation (e.g., Lamsal et al., 2008). We utilize one-minute averaged P3-B data from all fourteen flights during July 2011. One-minute averaged data is already pre-generated in the data archive. Hourly output from our model simulation is spatially and temporally matched to the observations. We then bin the data into different altitude ranges for our comparison.

2.2.2 Pandora NO_2 data

Measurements of total column NO_2 from the Pandora spectrometer (instrument reference: (Herman et al., 2009)) are used to evaluate the OMI NO_2 satellite products. Valid OMI NO_2 pixels are matched spatially and temporally to Pandora total column NO_2 observations. To smooth the data and eliminate brief small-scale plumes or anomalies, we average the Pandora observations over a two hour period (± one hour of the overpass time) before matching to the OMI NO_2 data. During July 2011, there were twelve Pandora NO_2 spectrometers operating during the experiment; this corresponded to only seventy-eight nine instances in which valid Pandora NO_2 observations matched valid OMI NO_2 column data.

2.2.3 Airborne Compact Atmospheric Mapper (ACAM) NO_2 data

The UC-12 aircraft was outfitted with a downward looking spectrometer called the Airbone Compact Atmospheric Mapper (ACAM) during the DISCOVER-AQ Maryland campaign (instrument reference: (Kowalewski and Janz, 2009)). The instrument collects hyperspectral measurements in the UV, visible, and near-infrared range from an altitude of approximately 8 km. From these measurements, tropospheric column NO_2 underneath the aircraft can be calculated (Lamsal et al., 2017). An ACAM pixel is considered valid, if there are no clouds between the instrument's detector and the surface. Valid OMI NO_2 pixels are matched spatially and temporally (± one hour of the satellite overpass time) to the ACAM column NO_2 observations. During July 2011, there were only six days in which the UC-12 flight paths overlapped an OMI NO_2 swath; this corresponded to only 107 OMI NO_2 pixels which could be compared to the ACAM NO_2.

2.2.3 4 EPA ground monitor data

There are eighteen EPA NO_2 monitoring sites within our study area of interest that were operational during the 5-year period of interest. We gathered this data from the EPA AQS Data Mart (EPA, 2016). Monitoring data were filtered so that only days with valid satellite data were included. To smooth the data, we average all valid ground observations between 12 – 4 PM local time. All EPA monitors measure NO_2 by the chemiluminescence method which has a high bias when compared to other techniques (Dunlea et al., 2007; Lamsal et al., 2008; Lamsal et al., 2015). Dunlea et al. (2007) has shown the high bias to be 22 % in a polluted urban environment and as large as 50

% during the mid-afternoon. Lamsal et al. (2008) suggests the bias may be even higher, 50 – 65 %, in the eastern U.S. during the summertime. For this reason, we refer to NO_2 from these monitors as NO_2*.

2.3 GMI model simulation

The operational NASA OMI NO_2 product uses a Global Modeling Initiative (GMI) (Strahan et al., 2007) model simulation with a horizontal resolution of 1° × 1.25° (~110 × 110 km) sampled at the OMI overpass time to calculate a priori NO_2 shape factors. The model is driven by assimilated meteorological fields from the Goddard Earth Observing System (GEOS) at the NASA Global Modeling and Assimilation Office (GMAO, ttp://gmao.gsfc.nasa.gov/). The GEOS-5 meteorological data are provided every 3–6 h (3 h for surface fields and mixing depths) at 72 pressure levels in the vertical, extending from surface to 0.01 hPa. The model includes the latest available inventories for anthropogenic emissions as discussed in Strode et al. (2015) and Krotkov et al. (2017). These emissions are updated annually with annual scale factor estimates provided by individual countries (van Donkelaar et al., 2008). The GMI model also includes NO_x emissions from soil, lightning, biomass burning, biofuel, and aircraft sources, as described in Duncan et al. (2007) with updates as discussed in Krotkov et al. (2017). The GMI simulation is conducted for 2004-2014, sampling hourly model output at the OMI overpass time. The standard operational retrieval is based on yearly-varying monthly average NO_2 profiles derived from the GMI simulation.

2.4 CMAQ model simulation

For the ~~high resolution~~high-resolution OMI NO_2 product, we use a CMAQ regional model simulation initially prepared for use in Loughner et al. (2014). CMAQ v5.0 is driven off-line by meteorological inputs from the WRF model v3.3 for June and July 2011. Horizontal spatial resolution of both WRF and CMAQ is at 1.33 km. Both models also include 34 vertical levels between the surface and 100 hPa, with 16 layers within the lowest 2 km. The ACM2 drives the boundary layer parametrization in WRF, while ACM computes the convective mixing in CMAQ. Anthropogenic emissions are projected to 2012 from the 2005 EPA National Emissions Inventory (NEI); the 2011 NEI was unavailable when this model simulation was originally completed. Biogenic and lightning emissions are calculated online; biogenic emissions are calculated using BEIS v3.14. Soil NO_x emissions are not included here because the CMAQ soil NO_x parametrization was implemented in a newer version of the model (Rasool et al., 2016). This model simulation utilizes CB05 gas-phase chemistry. The 1.33 km simulation, which we use exclusively in this study, is nested inside three larger domains: 36 km, 12 km, and 4 km. Boundary conditions for the 36 km domain are provided by the MOZART-4 global model. The top of the model assumes "zero gradient", which means the top boundary has concentrations equal to the top model layer. ~~–~~The CMAQ 1.33 km model domain is shown in Figure 1. For additional details, including a discussion on the uncertainty of the meteorological and chemical fields in this simulation, please reference Loughner et al. (2014). ~~This~~Our study is particularly unique in that we use a 1.33 km simulation in lieu of a model with a horizontal resolution more typical of OMI (>13 km). We do this so that we can capture the fine-scale variability within urban areas that cannot be simulated by coarser models and observations.

2.5 Air Mass Factor Re-Calculation using CMAQ

This study is particularly unique in that we use a 1.33 km simulation in lieu of a model with a horizontal resolution more typical of OMI (>13 km). We do this so that we can capture the fine-scale variability within urban areas that cannot be simulated by coarser models and observations. To ensure a fair comparison, we average model information to the pixel sizeTo re-calculate the air mass factor for each OMI pixel, we first compute interim air mass factors for each CMAQ model grid cell. The interim air mass factor for each CMAQ grid cell is a function of the NO_2 shape factor from the model grid cell and scattering weight from the OMI pixel that overlaps it. We then average all interim air mass factors within an OMI pixel (usually 100's) to generate a single tropospheric air mass factor for each individual OMI pixel. This new air mass factor is used to convert the total slant column into a tropospheric vertical column using Equation 1. –Model outputs were sampled at the local time of OMI overpass. Since monthly mean values capture the seasonal variation, we derived monthly mean values for NO_2, and temperature profiles and tropopause pressures needed for the calculation of the AMF. The exception is fFor June & and July 2011, in whichwe use daily NO_2 profiles and terrain pressures (e.g., (Laughner et al., 2016)) were used to re-calculate the AMF. For years other than 2011, Since monthly mean values capture the seasonal variation, we derivedused 2011 monthly mean values forof NO_2, and temperature, profiles and tropopause pressures needed for the calculation of the AMF. The exception is Once the tropospheric vertical column of each OMI pixel was re-calculated, the product was oversampled for June and July over a 5-year period (2008-2012; 10 months total). The satellite product was oversampled for June & July over a 5-year period (2008-2012) by re-gridding to the CMAQ 1.33 km model grid and then averaging the data over the 10-month (two months × five years) period. We have chosen the June & July timeframe because the CMAQ simulation and DISOCVER-AQ Maryland data are only available during these two months.

3 Results

In this section, we describe the process to develop a new high resolutionhigh-resolution satellite product and our validation efforts. Unless otherwise noted, all OMI NO_2 results presented here are vertical column densities. First, we compare a priori NO_2 shape profiles simulated by GMI (global model) and CMAQ (regional model). Next we develop an initial OMI NO_2 satellite product (OMI_CMAQ) using AMFs generated from the CMAQ a priori NO_2 profiles. We introduce two additional steps: improving a priori NO_2 shape profiles using aircraft observations and applying a spatial weighting kernel to further improve the spatial distribution of NO_2. We then evaluate our new product by comparing to DISCOVER-AQ observations. And finally, we compare the new OMI NO_2 product with NO_2 VCDs from the original CMAQ simulation.

3.1 Evaluating modeled NO_2 shape profiles: GMI vs. CMAQ

Trace gas shape profiles provided by model simulations are a critical input to satellite retrievals. To understand the effects of model choice on the a priori NO_2 shape profile, we compare the mean 2 PM local time tropospheric NO_2 vertical profiles from CMAQ and GMI at several locations in the mid-Atlantic during June & July 2011. In the left panels of Figure 2, we show the mean NO_2 mixing ratio as function of altitude for three locations: downtown Baltimore Maryland (an urban area), the Morgantown Power Plant located in Newburg, Maryland 60 km south of the District of Columbia (D.C.), and Arendtsville, Pennsylvania (rural), a location 100 km northwest of Baltimore and upwind of major metropolitan areas during days with climatologically westerly winds. All three locations are shown on Figure 1. In Baltimore, GMI simulates a mean 2 PM surface NO_2 mixing ratio of 2.2 ppbv, while CMAQ simulates 9.6 ppbv at the same location. The "Oldtown" monitoring site in Baltimore registered a surface NO_2* mixing ratio of 10.5 ppbv within +/- 2 hours of valid co-located satellite overpasses. As discussed in Sect. 2.2.~~3~~4, the corrected surface NO_2 mixing ratio is approximately 22% ~~may be up to 65 %~~ lower (but may be up to 65% lower) than observed NO_2*; our best surface estimate of 8.2 ppbv with error bars [3.7, 10.~~6~~5] is shown by the black triangle on Figure 2. The surface value simulated by CMAQ (9.6 ppbv) is much closer to the observed value than GMI (2.2 ppbv). In the second row of panels, the panels representing the Morgantown power plant, CMAQ simulates a plume of NO_2 above the surface; the max value is 11.8 ppb corresponding to an altitude of 460 m. The GMI simulation cannot resolve power plant plumes. This yields significant errors in the NO_2 shape profiles simulated by GMI near observed large point sources. In the bottom row of panels, we show a location in rural Pennsylvania. CMAQ, once again, does better in simulating the surface concentration than GMI.

However, in the free troposphere (i.e., above 3 km and below the tropopause) CMAQ consistently simulates smaller NO_2 than GMI. CMAQ simulates NO_2 mixing ratios between 0.01- 0.04 ppbv, while GMI simulates NO_2 mixing ratios between 0.06 – 0.09 ppbv over the same altitudes; GMI simulates values which are a factor of three higher than CMAQ. To determine whether lightning NO is the primary driver of this difference~~Both simulations include lightning NO_x. –~~, we compare lightning NO emissions from both model simulations in Figure 3. The CMAQ model ingests lightning NO emissions that are an order of magnitude larger than the GMI simulation at most altitudes. This is likely due to WRF simulating more convective precipitation and higher cloud-top heights, both input variables to the lightning NO parameterization, than GMI. Therefore, the ~~–~~differences in free tropospheric NO_2 between the two models ~~likely~~ do not arise from the lightning NO_x parameterizations, but instead from a combination of the ~~treatment of~~ chemistry, aviation emissions, vertical mixing, long-range transport, and stratospheric-tropospheric exchange~~transport, which has a dominating effect at these altitudes~~.

3.2 Calculation of air mass factors: GMI vs. CMAQ

A normalization of the NO_2 as a function of altitude (i.e., x_a / Σx_a in Eq. (2)) is the next step in the calculation of the AMF; these values are defined in the literature as shape factors. The center column panels show NO_2 shape factors for three locations. In Figure 2b (Baltimore), the GMI and CMAQ shape profiles (i.e., shape factors as a function of altitude) appear to be similar, but there are noticeable differences within the boundary layer and free troposphere. In Figure 2e, ~~there are large differences in the shape profile within the boundary layer due to~~ CMAQ captures~~ing~~ a localized power plant plume, while GMI does not; this yields large differences in the shape profile within the boundary layer. And in Figure 2h, CMAQ suggests that the NO_2 gradient near the surface is not as sharp.

Since the AMF is also a function of the SW, small differences in NO_2 shape profiles can manifest very different AMFs. For example, small differences in the shape profile at 7.5 km, where the SW is a maximum (SW = 2.9), have an order of magnitude larger effect than differences at the surface (SW = 0.4).

To fully understand the differences caused by the new NO_2 shape factors, we multiply the two shape factors by the satellite scattering weights. Here we define the shape factors × scattering weight ((i.e., $(x_a \times SW)/\Sigma x_a$ in Eq. (2)) as the adjusted shape factors. This is analogous to the values used for calculation of the air mass factor. The AMF is the integral of the adjusted shape factors with respect to height. In Figure 2c, the CMAQ adjusted shape profile shows values much closer to zero above 3 km than GMI. By using a priori shape profiles from CMAQ, we are enhancing the sensitivity of satellite observations to NO_2 concentrations within the boundary layer in Baltimore. In Figure 2f, the adjusted shape profiles are even more dramatic. At this location, adjusted shape profile values from CMAQ are relatively large below 1 km, and close to zero above 1 km, while GMI shows nearly an order of magnitude larger sensitivity above 1 km. In Figure 2i, CMAQ shows larger values above the surface, but within the boundary layer, while GMI shows larger values directly at the surface. In areas, such as these, the adjusted shape factors yield only small changes. In Figures 2c and 2f, the area underneath the red curve is smaller than the area underneath the blue curve. This will yield smaller AMFs when using CMAQ at these locations. As a result, we should expect the new OMI tropospheric NO_2 columns to be larger near urban areas and point sources which cannot be resolved by global models. At the rural location, the areas underneath the two curves are roughly the same, yielding similar AMFs and NO_2 columns.

3.3 Calculation of OMI tropospheric column NO_2

3.3.1 Using CMAQ profiles

We use the AMFs based off the CMAQ simulation to generate NO_2 tropospheric column amounts; we call this the OMI_CMAQ product. For this product, tropospheric NO_2 columns are calculated from the NASA Level 2 OMI NO_2 total slant column using Eq. (1). For AMFs calculated in the months of June and July 2011, we use AMFs derived from daily NO_2 shape factors as described by Laughner et al. (2016), resulting in more day-to-day variability in the AMF. Daily CMAQ NO_2 shape profiles from the hourly output are matched temporally and spatially to the OMI pixel. For years other than 2011, we use a two-month (June and July) average of the 2011 NO_2 shape factors to derive "summertime" AMFs. Since the resolution of CMAQ is finer than the resolution of OMI, we average all CMAQ AMFs across each individual pixel. Often there are over two-hundred CMAQ AMFs within a single pixel. Since CMAQ is capturing the spatial heterogeneities in urban areas, using it in lieu of GMI to provide NO_2 shape profiles can yield large variability in the AMF between adjacent OMI pixels.

Figures 43a and 3b 4b depict the OMI NO_2 tropospheric columns using a priori shape profile information from GMI (OMI_GMI; Figure3aFigure 4a) and CMAQ (OMI_CMAQ; Figure 3b4b) in calculating the AMF. Both products are oversampled to 1.33 km for June & July over a 5-year period (2008-2012) by re-gridding to the CMAQ model grid and then averaging the data over the 10-month (two months × five years) period. We have chosen the June &

July timeframe because the CMAQ simulation is only available during these two months. For the OMI_GMI product, the tropospheric NO_2 columns were taken directly from the NASA OMI NO_2 v3.0 Level 2 product. Figure ~~4a~~ 5a shows the ratio between the two products.

In the new product (OMI_CMAQ), there are large increases of the NO_2 VCDs in city centers. In the operational OMI_GMI product, over the 5-year period, the maximum tropospheric NO_2 column within Baltimore city limits is 3.9×10^{15} molecules per cm^2. By contrast, in the OMI_CMAQ product, the maximum tropospheric NO_2 column within Baltimore city limits is 7.2×10^{15} molecules per cm^2. These results indicate that by using a regional model, the tropospheric NO_2 VCDs in urban areas rise dramatically. This is due, in part, to the regional model being able to better capture NO_2 concentrations in the lower-most part of the troposphere (i.e., Figure 2). In suburban and rural locations, NO_2 tropospheric VCDs are roughly the same. For example, at the rural Pennsylvania (Arendtsville) location, the NO_2 tropospheric ~~vertical~~ column in the operational product is 2.8×10^{15} molecules per cm^2 and 2.7×10^{15} molecules per cm^2 in the new OMI_CMAQ product.

3.3.2 Improving modeled vertical profile information with in situ observations

While using CMAQ to calculate AMFs yields a marked improvement in simulating profile shape when compared to using GMI, this CMAQ simulation has a high bias in the calculation of total reactive nitrogen oxides (NO_y) (Goldberg et al., 2014; Anderson et al., 2014), which must be accounted for. Many literature sources, including others using different model set-ups (all are based on the NEI), also show a high bias in simulating summertime column NO_2 (Canty et al., 2015; Souri et al., 2016), NO_x (Travis et al., 2016), and NO_y (Goldberg et al., 2016).

In Figure ~~5~~6, we show NO_2 observations acquired by the P3-B aircraft in the early afternoon between 300 m and 3 km during DISCOVER-AQ Maryland ~~and~~ matched to CMAQ and GMI output. NO_2 mixing ratios simulated by CMAQ are consistently ~~smaller~~ larger throughout the mid- and upper-boundary layer and lower free troposphere (1 – 3 km) by up to a factor of three, but there is fairly good agreement below 1 km; similar results were found by Flynn et al. (2016). The NO_2 mixing ratios simulated by GMI below 1 km are a factor of two lower than the P3-B observations. Furthermore, the variability is an order of magnitude smaller than the observations. These shortcomings of GMI are a result of using a monthly mean (the same value used for the satellite retrieval) and coarse resolution model.

Since the P3-B aircraft has limited measurements above 3 km, we have to use estimates from other literature sources to determine the validity of CMAQ in the free troposphere. Lamsal et al. (2017) used measurements from the Airborne Compact Atmospheric Mapper (ACAM) to deduce that GMI is better than CMAQ at simulating NO_2 in the free troposphere. In the upper free troposphere, above 10.5 km, Travis et al. (2016) note that NO_2 is significantly underestimated by global models, such as GMI. As shown in Figure 2, CMAQ simulates even lower NO_2 concentrations than GMI at these altitudes.

We apply a scaling factor inferred from in situ aircraft observations to account for the high model bias below 3 km, and low model bias above 3 km; this is a rudimentary simplified form of data assimilation. Below 3 km, the model was scaled to observations from the P3-B by multiplying the original values at these altitudes by the fraction of NO_2 actually observed. For example, modeled NO_2 between 1000 – 1500 m was multiplied by 0.63 to account for the model high bias within this altitude bin. This procedure was repeated for all altitude bins in 500-m intervals from the surface up to 3 km. It should be noted that aircraft measurements from the DISCOVER-AQ Maryland campaign took place only within the Baltimore metropolitan region, and thus these scaling factors may not be fully applicable to upwind rural regions, and certainly cannot be applied to locations outside the eastern United States. Between the altitudes of 3 km – 10.5 km, we switched out the NO_2 mixing ratios from CMAQ for NO_2 mixing ratios from GMI. Between 10.5 km and the tropopause, we use GMI NO_2 mixing ratios multiplied by a factor of three; this scaling factor is based on summertime NO_2 observations during the SEAC^{4}RS field campaign as described by Travis et al. (2016).

Using these scaled mixing ratios, we then re-calculate the AMFs and corresponding tropospheric NO_2 columns. Figure 3c 4c shows observationally-constrained OMI_CMAQ (OMI_CMAQ_O) tropospheric NO_2 columns during the same 5-year summertime period. NO_2 tropospheric columns in this product are smaller in magnitude than OMI_CMAQ, and yet still noticeably larger in urban areas than the operational OMI_GMI retrieval (i.e., in Baltimore OMI_GMI: 3.9×10^{15} , OMI_CMAQ: 7.2×10^{15}, OMI_CMAQ_O: 5.0×10^{15}). Retrievals in upwind rural areas in this new product are now lower than the operational product (i.e., in Arendtsville OMI_GMI: 2.8×10^{15} , OMI_CMAQ: 2.7×10^{15}, OMI_CMAQ_O: 1.7×10^{15}).

The large reduction in NO_2 tropospheric columns between OMI_CMAQ and OMI_CMAQ_O is an outcome of using larger AMFs. The larger AMFs are a result of the original overestimate within the boundary layer and underestimate in the free troposphere. This is a particularly important finding because it means that a model with large biases in the simulation of NO_2 can yield poor tropospheric vertical column contents, despite high spatial resolution. This emphasizes the need to evaluate the emissions and chemistry of a model before it is used for satellite retrievals.

3.3.3 Enhancing spatial resolution with spatial weighting kernels

Finally in a last step, we apply the method described by Kim et al. (2016) to downscale the OMI retrieval. This method applies a spatial-weighting kernel to portions of each pixel based on the estimated influence from each locality within the pixel. For example, if one side of a pixel overlaps a polluted region, while the other side of the pixel overlaps a cleaner area, the operational OMI product will denote that the entire area is moderately polluted. Instead, we weight portions of the individual pixel based on the variability simulated in CMAQ. Using this method, the quantity of the satellite data is numerically preserved. This yields a higher resolution snapshot of tropospheric column NO_2 that is still constrained by satellite data. Please reference Kim et al. (2016) for a visual representation of this method.

We call this product OMI_CMAQ observationally-constrained + downscaled (OMI_CMAQ_OD). Figure 3d 4d shows OMI_CMAQ_OD tropospheric NO_2 columns. There is now large variability throughout the region, which is typical of a pollutant with a short lifetime (< 1 day) such as NO_2 in the summertime. NO_2 tropospheric columns in urban cores are now significantly larger than the operational product, (i.e., in Baltimore OMI_GMI: 3.9×10^{15}, OMI_CMAQ_OD: 10.2×10^{15}). The largest increases occur near power plants, cement kilns, and major highways. OMI_CMAQ_OD in upwind rural areas are 20 – 50 % lower than the operational product (i.e., in Arendtsville OMI_GMI: 2.8×10^{15}, OMI_CMAQ_OD: 1.6×10^{15}).

We must clarify, however, that these results are only applicable for our region of interest. While we find that rural areas within our mid-Atlantic model domain now have tropospheric NO_2 columns which are 20 – 50 % lower than the operational product, we cannot conclude that this would be the same elsewhere. The "rural" locations within our model domain are situated in a particularly precarious spot because they are close, but not too close to major urban areas. These sites are only "rural" in the sense that there are no major metropolitan areas within 200 km upwind of them. Because Arendtsville lies within 50 km of Harrisburg and 100 km of Baltimore, a GMI simulation with a resolution of 1.25° × 1° (~110 × 110 km) will group this location into a grid cell also including Harrisburg and portions of Baltimore; both of which are not rural. Therefore a location that is 100's of kilometers from the nearest city and with spatial homogeneity may be simulated with fidelity by GMI and therefore the operational OMI product may be an accurate representation of reality in these cases.

While this new product shows power plant plumes that are two to three times larger, we are not suggesting that emissions from power plants are larger than we thought. Instead we are suggesting that the spatially downscaled OMI product can now "see" these individual plumes, where-as in the operational product, these plumes are blended into an average across the entire OMI pixel. In rare cases, oversampling the operational product in and around very large rural point sources, can denote large power plant plumes (deFoy et al., 2015), but up until this point, smaller point sources or localized sources near major urban areas could not be seen by in an OMI NO_2 product.

3.4 Comparison of satellite products to in situ observations

To determine the accuracy of the new products, we compare the products to independent NO_2 observations. **3.4.1 Comparison to the Pandora NO_2 spectrometer network**

During DISCOVER-AQ Maryland, total column NO_2 was measured by a network of twelve Pandora instruments (Herman et al., 2009). We match daily valid Pandora NO_2 and valid satellite overpass information, and plot the information in Figure 6a7a. To calculate total OMI columns, we add the vertical stratospheric column information, a variable in the NASA OMI NO_2 Level 2 files, to the OMI tropospheric retrievals. While the operational product (OMI_GMI) shows some agreement at low values, it has poor agreement when observed NO_2 column amounts are greater than 10×10^{15} cm^{-2}. This is due to coarse resolution of OMI pixels (24 × 13 km at nadir) and the AMFs computed with GMI a priori NO_2 profiles, among potential other factors. The slope of the OMI_GMI best-fit line is

0.~~45~~44, representing a striking low bias at high values, and the r^2 = 0.~~09~~ 10 denoting almost no correlation; similar results were found by Ialongo et al. (2016).

Table 2~~1~~ shows the statistical comparison between the satellite products and observations. All OMI_CMAQ products yield slopes closer to one indicating that they are better at capturing the variability between low and high values observed by the ground monitors. The OMI_CMAQ_OD product eliminates the bias altogether. The slope of the OMI_CMAQ_OD best-fit is ~~1.02~~0.99 and the r^2 increases. An improved but still low r^2-value in the newest product may indicate that a 1.33 km CMAQ simulation provides an improvement, but not an identical match, of daily NO_x emissions and fine-scale winds responsible for plume dispersion. Furthermore, we cannot expect the satellite to match the exact spatial heterogeneity observed by the point measurements from Pandora~~.~~ because these instruments observe a very narrow fraction of the atmosphere and measure column NO_2 in a fundamentally different manner.

3.4.2 Comparison to the Airborne Compact Atmospheric Mapper (ACAM) spectrometer NO_2

The ACAM NO_2 instrument acquired measurements of tropospheric column NO_2 below altitudes of 8 km during DISCOVER-AQ Maryland. We match ACAM NO_2 measurements within ± one hour of the OMI overpass time to valid OMI NO_2 measurements. The comparison is plotted in Figure 8. Both the slope and r^2-value of the new OMI_CMAQ_OD product are closer to one when compared to the OMI_GMI product indicating that the OMI_CMAQ_OD product yields better agreement with ACAM NO_2. The low r^2-values may be related to the ACAM instrument random error, one of which is the use of unpolluted background spectra instead of reference spectra to process the data (Lamsal et al., 2017). In Figure 8a, we shade the points based on date. There were only six days in which valid OMI NO_2 spatially and temporally overlapped with the ACAM NO_2 data. In Figure 8b, we shade based on percentage coverage. Since the ACAM field of view is very small compared to OMI, pixel coverage from the ACAM would often only overlap a very small subset of the OMI pixel (median: 12 % of the OMI pixel). Since the ACAM is only measuring the portion of the tropospheric column below 8 km, there should be a consistent high bias in the OMI NO_2; instead there is a consistent low bias. This may be due to an artifact of the flight path of the UC-12 which preferentially sampled over the densest urban locations: OMI pixels are much larger in size and are capturing a more regional, and thus lower, value.

3.4.3 Comparison to the EPA NO_2 ground monitor network

The long-term EPA monitoring network provides surface observations outside the July 2011 timeframe. In Figure ~~6b~~9a, we compare mean NO_2* at each monitoring site to the two satellite products. All valid NO_2* data at each monitoring site over a 5-year (2008-2012) 2-month (June & July) period are averaged into a single point (up to 305 data entries) and ~~is~~ matched to an average of satellite data over the same time period. The correlation between OMI_GMI and surface observations is r^2 = 0.39, while the correlation between OMI_CMAQ_OD and surface observations is r^2 = 0.60, a substantial improvement. This suggests that a ~~high resolution~~high-resolution satellite product with improved AMFs, ~~will be able to~~can detect surface NO_2 concentrations with more accuracy. As shown in Table ~~1~~2, OMI_CMAQ without observational constraints performs almost as ~~good~~ well (r^2 = 0.55); this is especially encouraging since comprehensive field measurements, such as those from DISCOVER-AQ, are limited in spatial and temporal scope.

3.5 OMI_CMAQ vs. CMAQ

We can now more fairly assess the NO_2 columns simulated by CMAQ using a ~~high resolution~~high-resolution OMI NO_2 product. In Figure ~~7~~10, we show a comparison between CMAQ and OMI_CMAQ_OD. Only model data within +/- 1 hour of and co-located with valid overpass data ~~is~~are shown in order to remove biases during cloudy days or days with invalid data. We see a consistent model low bias in rural areas, and consistent model high bias in urban areas. Interestingly the high bias is larger in the immediate Baltimore metropolitan area compared to the D.C. metropolitan region.

We attribute the model low bias in rural regions to several shortcomings of this model simulation. This simulation did not include NO_x emissions from soils. Rasool et al. (2016) has shown soil NO_x emissions to be particularly important in the central United States, with a lesser role in the eastern United States. Excluding these emissions may have resulted in less NO_x being transported from upwind regions. This model simulation utilized CB05 gas-phase chemistry, which is known to underestimate the recycling of alkyl nitrates back to NO_2 (Hildebrandt-Ruiz and Yarwood, 2013; Canty et al., 2015). CB05e51 gas-phase chemistry, released in a newer version of CMAQ (https://www.airqualitymodeling.org/in-dex.php/CMAQ_v5.1_CB05_updates), better handles alkyl nitrates and employs faster recycling of short-lived alkyl nitrate species. Faster recycling of alkyl nitrates would yield higher NO_2 concentrations throughout the modeling domain. Travis et al. (2016) found that upper tropospheric NO_x is too low when compared to observations from aircraft during $SEAC^4RS$. This is possibly due to downward stratospheric transport, outflow from convection, or OH chemistry that is not characterized correctly by models. Lightning NO_x is still a very active area of research (Pickering et al., 2016). Although this model simulation did include lightning NO_x emissions, there is a possibility these emissions are underestimated.

We attribute the model high bias in urban regions within our domain to an overestimate of anthropogenic NO_x emissions (Anderson et al., 2014; Souri et al., 2016). This may be due to an improper allocation of area and mobile (on-road and off-road) source emissions which are spatially distributed based on population and number of cars respectively, or quite simply an overestimate of these sector emissions. Quantifying the uncertainty in MOVES, the mobile emissions software, is an active area of research.

3.6 Comparison of model to satellite and in situ observations

To further evaluate the model, we compare the model simulation to DISCOVER-AQ and EPA observations. In Figure ~~8a~~7b, we show a Pandora NO_2 comparison in the same manner as Figure ~~6a~~7a. In addition to showing CMAQ, we also show OMI_CMAQ_OD. We add the stratospheric VCD information from the OMI NO_2 Level 2 product to the CMAQ tropospheric columns to ensure a fair comparison. Both the model and new OMI NO_2

product have a slope close to unity indicating that both are able to match the variability in NO_2 columns. There is, however, a consistent low offset. This may indicate that the stratospheric VCD in the NASA Level 2 retrieval may be too low during this two-month timeframe. The r^2 of CMAQ is higher than OMI_CMAQ_OD. This is not particularly surprising because the resolution of the satellite is coarse, despite it being processed with new air mass factors.

In Figure 9~~8~~b, we show a comparison between CMAQ, OMI_CMAQ_OD and ground monitors for June & July 2011. The r^2 between CMAQ and ground monitors ~~to be~~is 0.70, while the correlation with the new satellite product is 0.73. The OMI_CMAQ_OD product has a better correlation with ground NO_2 monitors than the 1.33 km CMAQ simulation alone indicating that there is added utility in the satellite data.

4 Summary and conclusions

This study demonstrates the critical importance of using ~~high resolution~~high-resolution a priori NO_2 shape factors to develop AMFs in and around metropolitan areas. We develop three new OMI NO_2 products: using high spatial resolution NO_2 profiles from a 1.33 km CMAQ model simulation (OMI_CMAQ), using CMAQ profiles constrained by in-situ observations (OMI_CMAQ_O), and applying model-derived spatial information (downscaling) to OMI_CMAQ_O (OMI_CMAQ_OD). When using high spatial resolution models to develop the AMF, the mean AMF in urban areas decreases by up to 50 % causing the tropospheric VCDs in urban areas to increase by up to a factor of two. This is because ~~high resolution~~high-resolution models simulate larger concentrations near the surface in urban areas. In essence, we are reprocessing the satellite to look for NO_2 closer to the surface than in the original product. We believe this finding extends to other urban areas since coarse global models will consistently merge rural and urban pollution, and subsequently overestimate the AMF in city centers.

Another novel step in our re-processing technique is using in situ observations to enhance modeled NO_2 profile shapes. CMAQ NO_2 values in the Baltimore-Washington metropolitan region are generally too large within the boundary layer, too small in the mid-troposphere, and a factor of three too small in the uppermost troposphere. These particular biases~~is~~ may not be fully applicable to rural regions, since the DISCOVER-AQ field campaign was only focused in the urban corridor. As a result, our adjusted satellite product in rural regions ~~must be taken with some uncertainty~~may have higher uncertainty than urban areas. With that said, constraining model simulations to observations yields an improved satellite product over the non-constrained product when comparing to Pandora NO_2. Furthermore, by constraining to observations, we reduce the dependence on a priori emission inventories (e.g., NEI) used in model simulations, which can have deficiencies (Anderson et al., 2014; Souri et al., 2016; Travis et al., 2016). For example, in the constraint-based product, VCDs in Baltimore are 30 % lower than the OMI_CMAQ product. The tropospheric VCDs in rural Mid-Atlantic areas are 20 – 50 % lower than both the OMI_CMAQ and operational products. This is a particularly important finding because it means that the poor performance of CMAQ (or any model used for a satellite retrieval) will manifest itself in the retrieval. This will be a difficult challenge going forward, and emphasizes the need to use state-of-the science models for satellite retrievals.

Lastly, we apply a technique developed by Kim et al. (2016) to downscale OMI NO_2 data. This technique is especially valuable for pollutant exposure health studies, which require ~~high resolution~~high-resolution long-term pollutant estimates. The downscaling procedure provides a higher spatial resolution snapshot of NO_2, while not altering the observed satellite pixel values. Instead, this technique re-allocates values across the pixel based on the variability within the ~~high resolution~~high-resolution model. As a result, the new satellite product (OMI_CMAQ_OD) shows higher values in urban, polluted areas and lower values in rural, unpolluted areas than the operational OMI_GMI product. This new product better captures the urban-scale variability of NO_2 and has a much better correlation with ground monitors. A deficiency with this technique is that if a localized source, such as a power plant plume or wildfire, is not simulated at all by the model, then this error will be passed on to the product. Furthermore, if the area is affected by a mesoscale meteorological feature that is simulated incorrectly by the model, such as a thunderstorm, valley breeze, or sea breeze, the model will be similarly deficient. Therefore, we do not recommend using the downscaling technique in areas where the emission inventory or meteorology is very uncertain.

We must clarify, however, that the~~se~~ results in this paper are only applicable ~~for~~to our region of interest. While we find that rural areas within our mid-Atlantic model domain now have tropospheric NO_2 columns which are 20 – 50 % lower than the operational product, we cannot conclude that this would be the same elsewhere. The "rural" locations within our model domain are situated in a particularly ~~precarious~~tricky spot because they are close, but not too close to major urban areas~~. These sites are only "rural" in the sense that there are no major metropolitan areas within 200 km upwind of them. Because Arendtsville lies within 50 km of Harrisburg and 100 km of Baltimore,~~; a GMI simulation with a resolution of 1.25° × 1° (~110 × 110 km) will group ~~this location~~rural areas into a grid cell also including ~~Harrisburg and portions of Baltimore~~a large city~~; both of which are not rural~~. Therefore a location that is 100's of kilometers from the nearest city and with spatial homogeneity may be simulated with consistency ~~fidelity~~ by GMI and therefore the operational OMI product may be an accurate representation of reality in these cases.

The refined OMI_CMAQ_OD product provides a better NO_2 column measurement when compared to Pandora column NO_2: the slope is near unity and the r^2 increases over the operational OMI NO_2 product. An important finding of this work is that using a ~~high resolution~~high-resolution model, not the constraining to observations, provides the majority of the improvement, when comparing to ground monitors. This suggests that a ~~high resolution~~high-resolution model with reasonable fidelity can be used anywhere in the world, and is not tied into an area in which a field experiment is located.

This technique can be used as a bridge until newer instruments such as TROPOMI are instituted. Future instruments will have increased spatial resolution, but comparison to OMI without using this technique may yield large differences around urban areas. At the same time, we demonstrate the importance of using ~~a high resolution~~high-resolution and high-fidelity model simulations for retrievals from future satellite missions. A combination of both increased satellite resolution and model resolution are needed in order to improve NO_2 satellite retrievals. We urge other community members to generate ~~high resolution~~high-resolution OMI NO_2 data using this technique if it is to be used for small-scale (< 100 km length scale) studies as it provides a better alternative for urban areas than standard satellite products.

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Figures

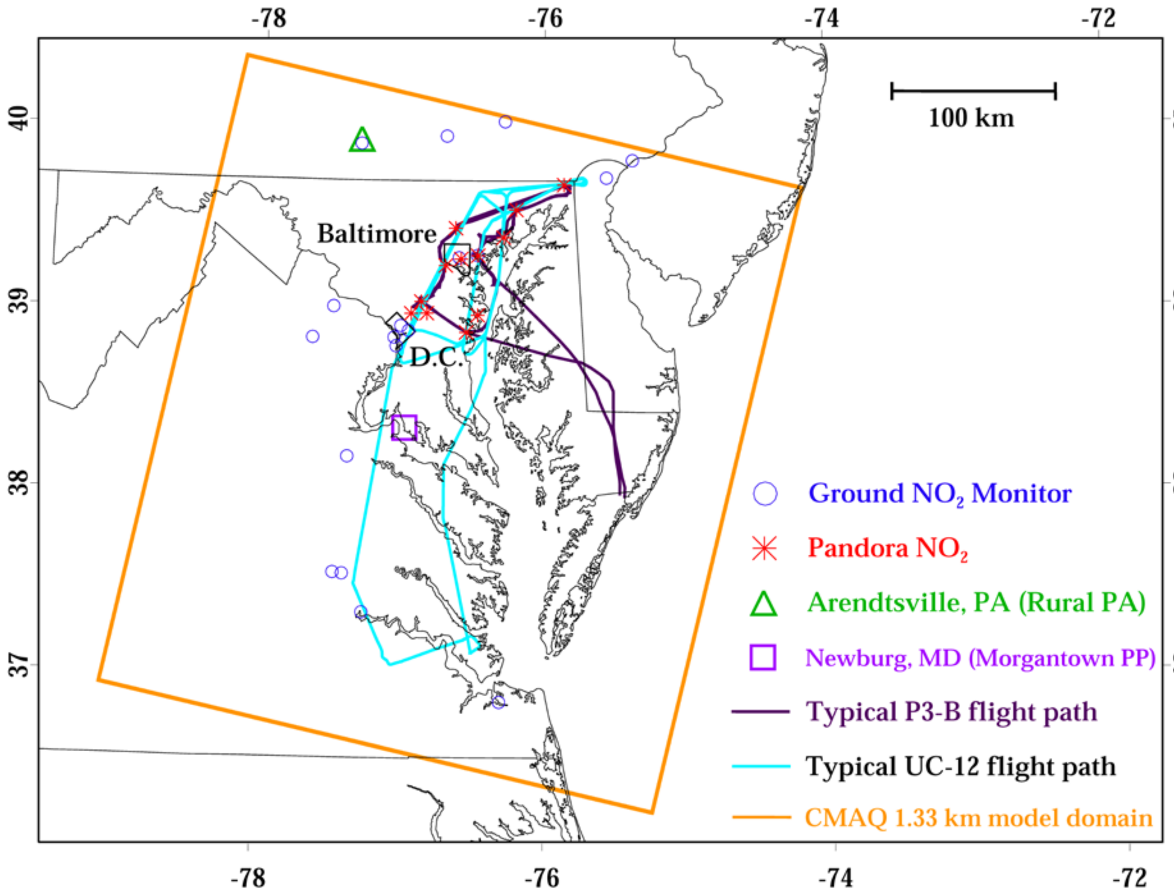

Figure 1. The Mid-Atlantic United States: the area of interest for this research project. Model domain and observation locations are depicted. There are eighteen EPA chemiluminescence NO_2 monitors and twelve Pandora NO_2 measurement sites.

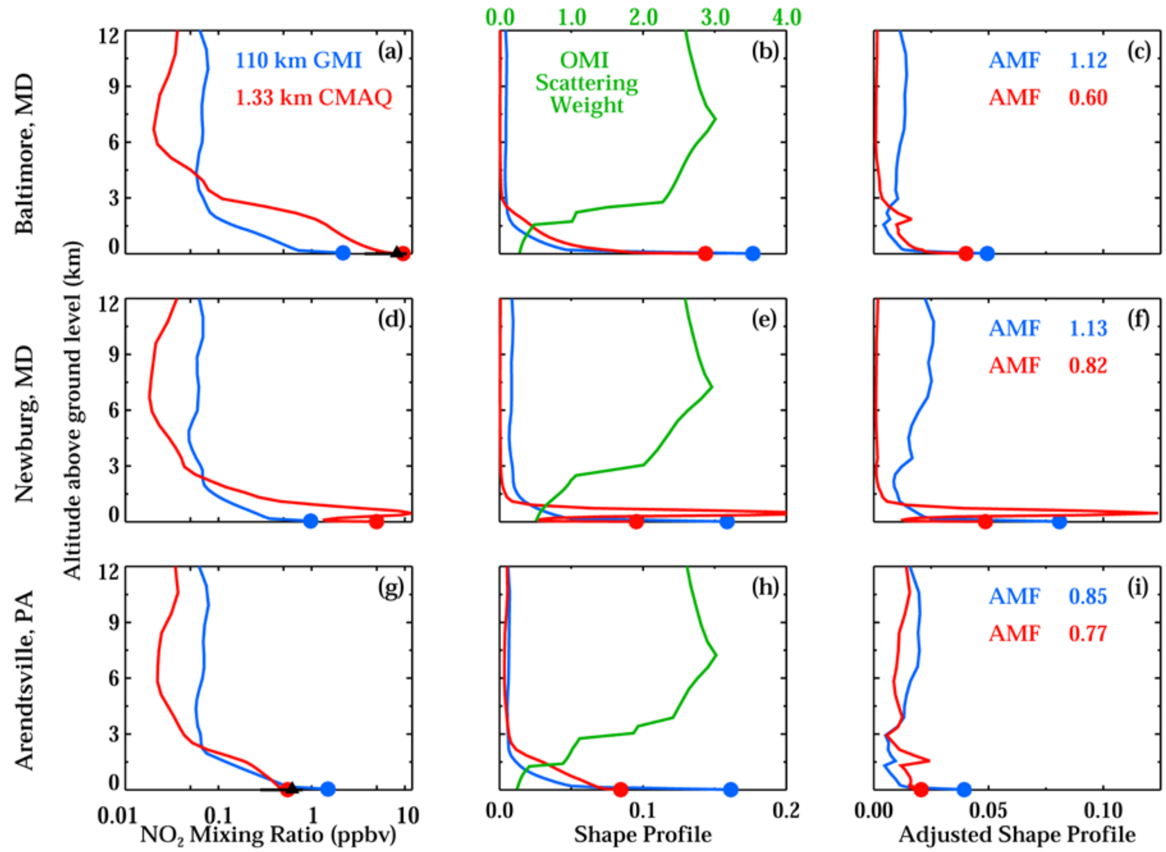

Figure 2. **(a, d, g)** Mean 2 PM local time June and July 2011 NO_2 mixing ratio as a function of altitude from a GMI (1.25° × 1°; ~110 ×110 km) model simulation and CMAQ (1.33 × 1.33 km) model simulation for three locations: **(a)** downtown Baltimore, **(d)** Morgantown power plant in Newburg, MD and **(g)** Arendtsville in rural Pennsylvania. Black triangles with error bars as discussed in the text represent co-located surface observations from the EPA monitoring network. **(b, e, h)** NO_2 shape profiles (partial NO_2 columns divided by total NO_2 column) as function of altitude for the same timeframe and locations; green line denotes co-located OMI scattering weight. **(c, f, i)** "Adjusted" shape profiles, partial NO_2 columns divided by total NO_2 columns multiplied by OMI scattering weight, as function of altitude for the same timeframe and locations.

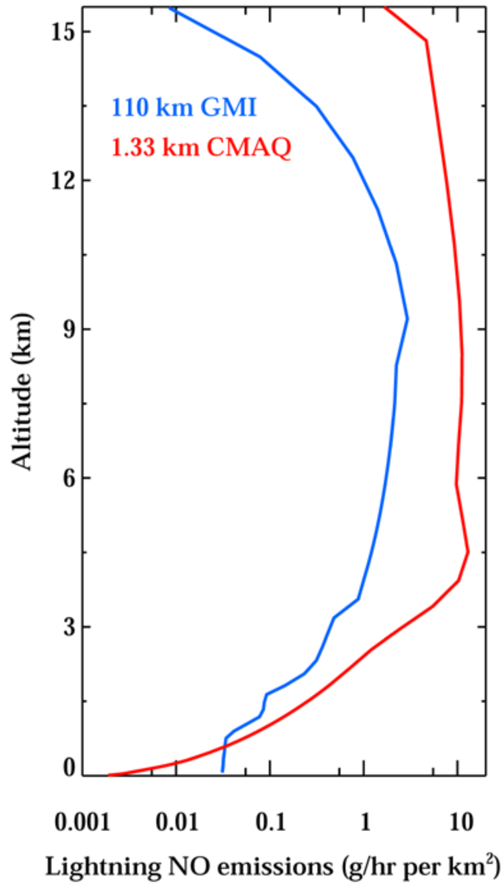

Figure 3. Mean June and July 2011 lightning NO emissions as a function of altitude from the GMI (1.25° × 1°; ~110 ×110 km) and CMAQ (1.33 × 1.33 km) model simulations.

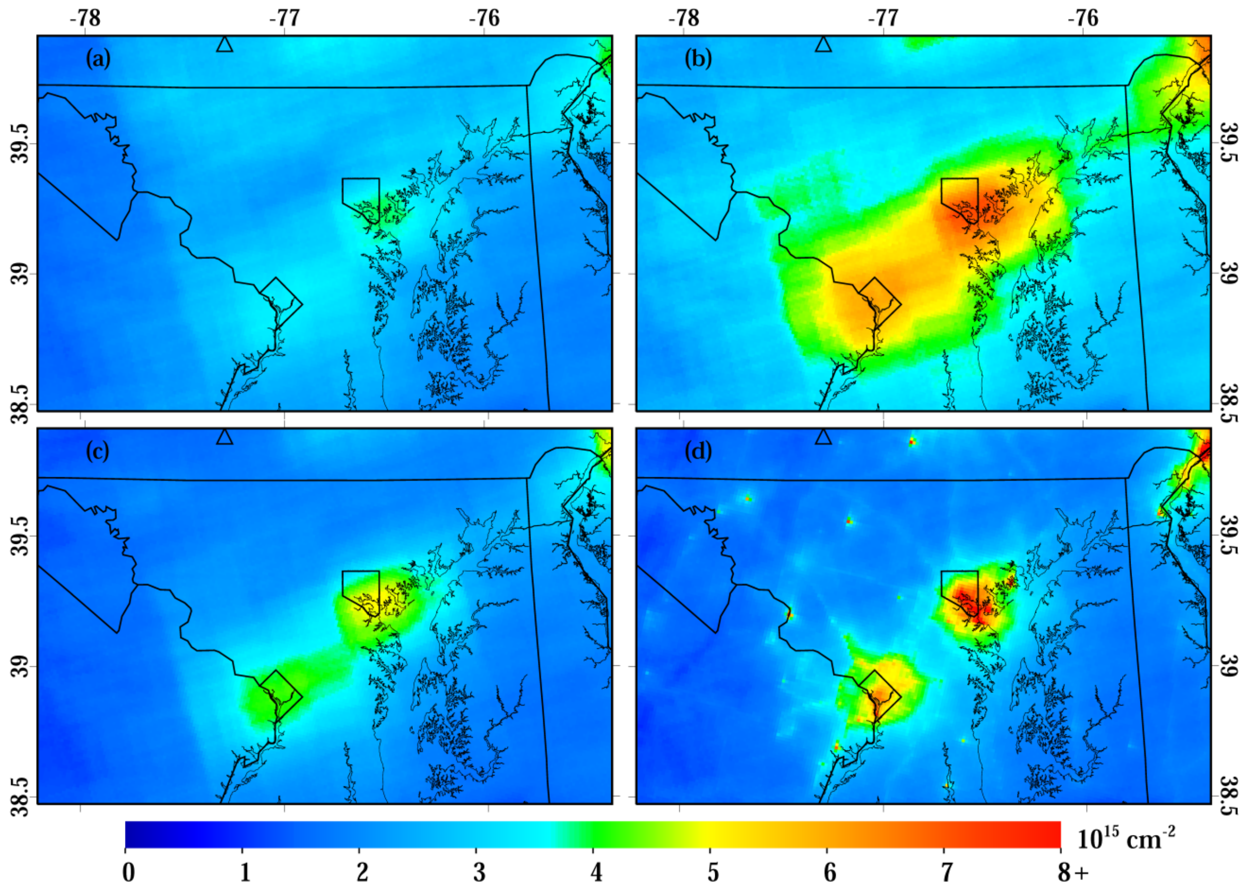

Figure 4. Oversampled OMI NO_2 tropospheric columns at 1.33 km resolution in the Baltimore-Washington metropolitan area for June & July 2008 – 2012 (2 months × 5 years; 10 months total). **(a)** The NASA version 3.0 operational OMI NO_2 product using GMI NO_2 shape profiles (OMI_GMI). **(b)** OMI NO_2 using CMAQ a priori NO_2 shape profiles (OMI_CMAQ). **(c)** OMI NO_2 using CMAQ a priori NO_2 shape profiles constrained by observations (OMI_CMAQ _O). **(d)** OMI NO_2 using CMAQ a priori NO_2 shape profiles constrained by observations and spatial weighting downscaling kernels (OMI_CMAQ_OD). In all plots, Arendtsville, PA is denoted by the triangle in the top left corner.

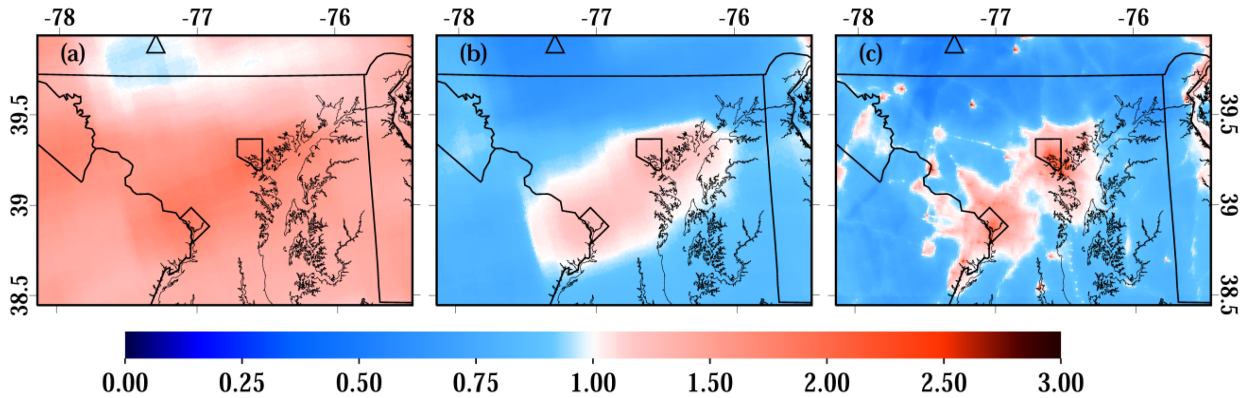

Figure 5. Ratio between the three OMI_CMAQ tropospheric NO_2 retrievals and the operational NASA v3.0 OMI tropospheric NO_2 retrieval for June & July 2008 – 2012 (2 months × 5 years; 10 months total). **(a)** OMI_CMAQ / OMI_GMI. **(b)** OMI_CMAQ_O/ OMI_GMI. **(c)** OMI_CMAQ_OD/ OMI_GMI.

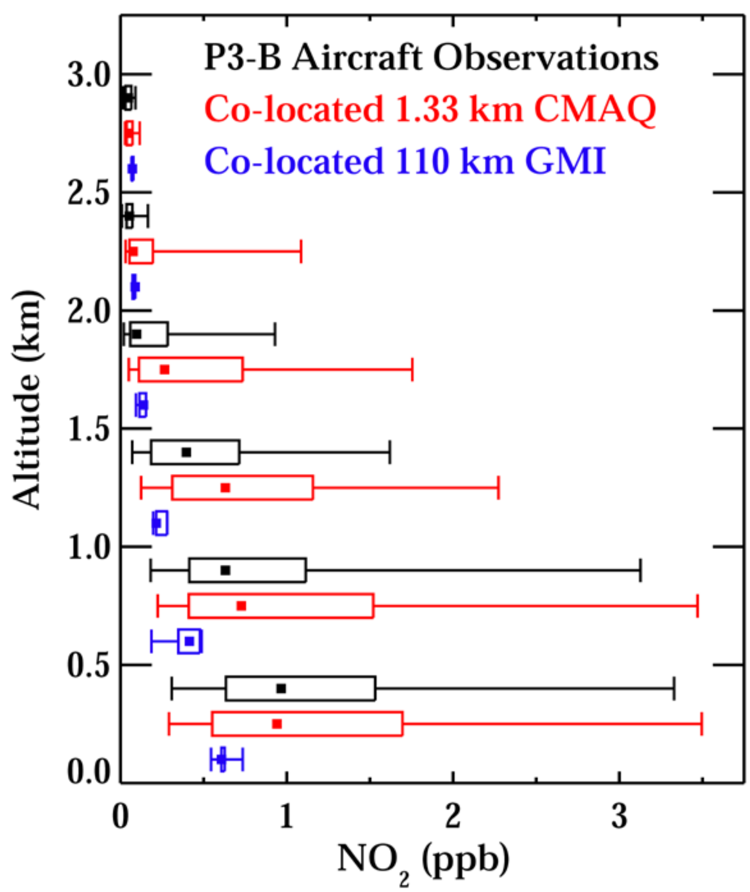

Figure 6. Vertical profiles of NO_2 binned in 500 m intervals (0 – 0.5 km, 0.5 – 1km, etc.) showing the 5th, 25th, 50th, 75th, and 95th percentiles within ± 2 hours of the OMI overpass time. **(Black)** One minute averaged data from the P3-B aircraft during July 2011 DISCOVER-AQ Maryland. **(Red)** Model output from CMAQ matched spatially and temporally to the P3-B measurements at 1 min intervals. **(Blue)** July 2011 monthly mean model output from GMI matched spatially to the P3-B measurements at 1 min intervals. In ~~both~~ all cases, the squares indicate the median values.

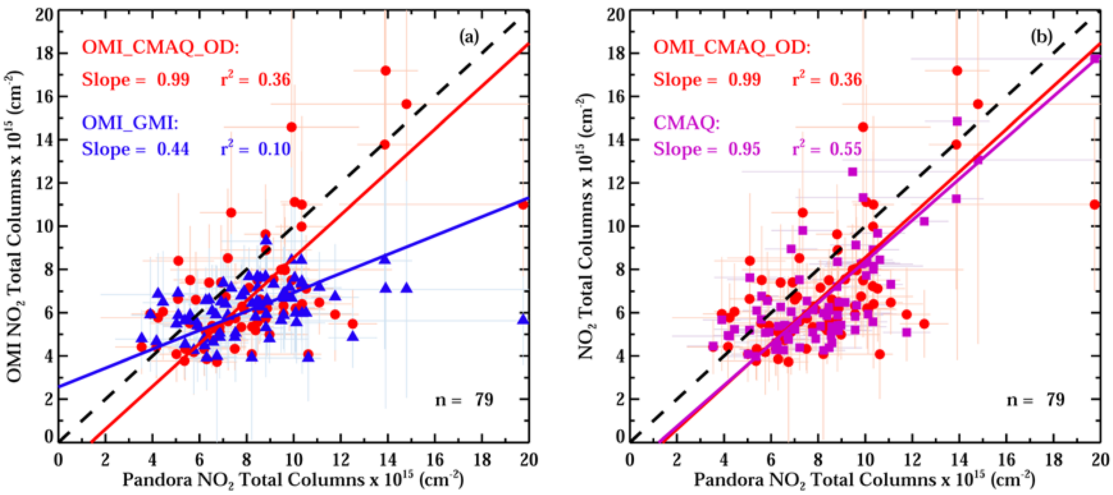

Figure 7. (a) Total column NO_2 OMI_GMI and OMI_CMAQ_OD versus co-located spatially and temporally Pandora NO_2 total column measurements within ± 1 hour of a valid satellite overpass during July 2011. (b) Same but now showing CMAQ instead of OMI_GMI; the stratospheric vertical column from NASA Level 2 product has been added to CMAQ to ensure a fair comparison. Error bars on both plots represent ± one standard deviation away from the mean.**(b)** Tropospheric column NO_2 OMI_GMI and OMI_CMAQ_OD versus co-located ground NO_2* chemiluminescence measurements within ± 2 hours of a valid satellite overpass during June & July 2008 through 2012; all ~300 daily ground monitor values are merged into a mean single value and compared to the satellite mean over the same corresponding time period.

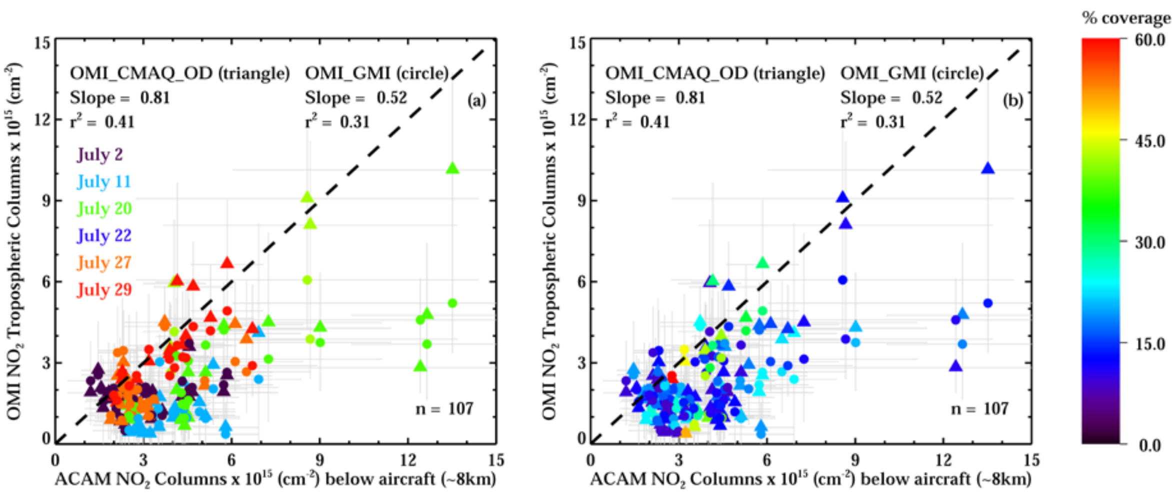

Figure 8. Tropospheric column NO_2 OMI_GMI and OMI_CMAQ_OD versus co-located spatially and temporally matched ACAM NO_2 column measurements within ± 1 hour of a valid satellite overpass during July 2011. **(a)** Color-coded based on date. **(b)** Color-coded based on percent coverage. Error bars on both plots represent ± one standard deviation away from the mean. **(b)** Tropospheric column NO_2 OMI_GMI and OMI_CMAQ_OD versus co-located ground NO_2* chemiluminescence measurements within ± 2 hours of a valid satellite overpass during June & July 2008 through 2012; all ~300 daily ground monitor values are merged into a mean single value and compared to the satellite mean over the same corresponding time period.

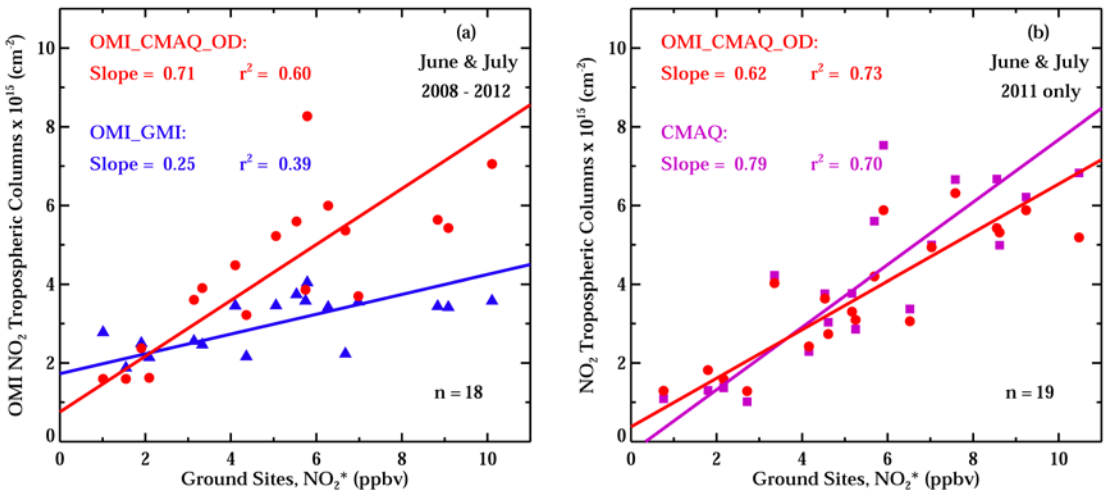

Figure 9. (a) ~~(b)~~ Tropospheric column NO_2 OMI_GMI and OMI_CMAQ_OD versus co-located ground NO_2* chemiluminescence measurements within ± 2 hours of a valid satellite overpass during June & July 2008 through 2012; ~~during June & July 2008 through 2012;~~ all ~300 daily ground monitor values are merged into a mean single value and compared to the satellite mean over the same corresponding time period. **(b)** Same as (a) but now comparing CMAQ and OMI_CMAQ_OD for June & July 2011 only. ~~**(a,b)** Same as Figure 6(a,b), but now showing CMAQ instead of OMI_GMI. **(b)** Showing June & July 2011 only due to model availability during this timeframe~~

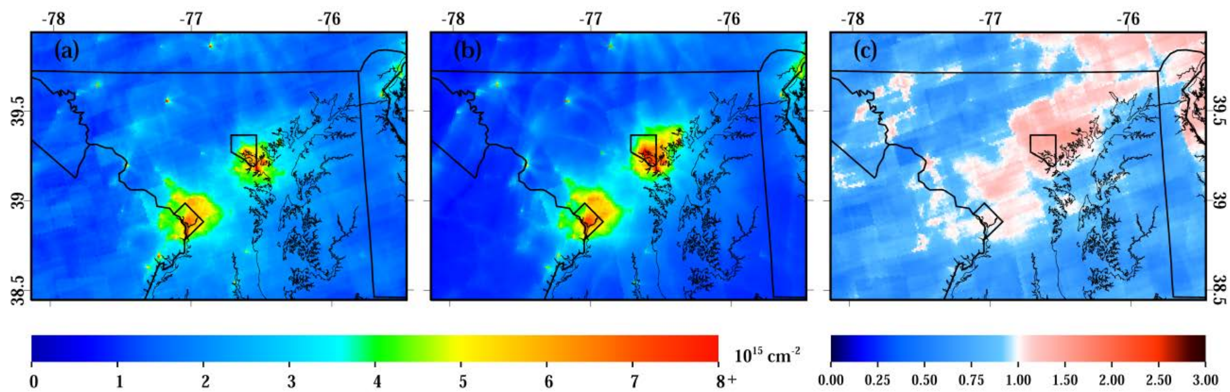

Figure ~~7~~10. Oversampled tropospheric column NO_2 at 1.33 km in the Baltimore-Washington metropolitan area for June & July 2011 only. **(a)** OMI_CMAQ_OD. **(b)** CMAQ NO_2 corresponding to valid overpass times. **(c)** Ratio between the two plots CMAQ / OMI_CMAQ_OD.

Table 1. Summary of the current OMI NO_2 retrievals in the literature.

	NASA OMNO2 v3	**DOMINO v2**	**BeHR NO_2**	**POMINO**	**HKOMI NO_2**	**This study**
CTM	GMI Global 1° × 1.25°	TM4 Global 2° × 3°	WRF-Chem U.S. 12 × 12 km	GEOS-Chem China 0.667° × 0.5°	WRF-CMAQ PRD China 3 × 3 km	WRF-CMAQ East U.S. 1.33 × 1.33 km
RTM	TOMRAD	DAK	TOMRAD	VLIDORT	SCIATRAN	TOMRAD
A priori NO_2 profile	Monthly mean profiles	Monthly mean profiles	Daily profiles when it exists. Monthly mean profiles elsewhere.	Monthly mean profiles	Daily profiles	Daily profiles when it exists. Monthly mean profiles elsewhere. All profiles constrained to aircraft observations.
Surface pressure	MERRA downscaled to 90 arcsec DEM	TM4 downscaled to 3 × 3 km	WRF downscaled to 1 × 1 km using GLOBE	GEOS-5 0.667° × 0.5°	WRF 3 × 3 km	WRF 1.33 × 1.33 km
Surface reflectivity	OMI LER climatology 0.5° × 0.5° taken from Kleipool et al., 2008	OMI LER climatology 0.5° × 0.5° taken from Kleipool et al., 2008	MODIS black-sky albedo MCD43C2 at 0.05° × 0.05°	MODIS BRDF MCD43C2 at 0.05° × 0.05°	MODIS MCD43C2 at 0.01° × 0.01°	OMI LER climatology 0.5° × 0.5° taken from Kleipool et al., 2008
Aerosol correction	Implicitly corrected through cloud products	Implicitly corrected through cloud products	Implicitly corrected through cloud products	Explicit treatment of aerosols	Correction for the aerosol effect	Implicitly corrected through cloud products

Table 21. Slope and r^2 for all four OMI satellite products compared to Pandora NO_2 from July 2011 and EPA ground monitor NO_2* observations from June & July 2008 – 2012. Pandora NO_2 is compared to the OMI NO_2 total column products, while the EPA ground monitors are compared to OMI NO_2 tropospheric column products. Figures 7a and 9a6 shows values for OMI_GMI and OMI_CMAQ_OD only.

	Pandora NO_2		EPA NO_2*	
	Slope	r^2	Slope	r^2
OMI_GMI	0.44	0.10	0.25	0.39
OMI_CMAQ	1.23	0.12	0.54	0.55
OMI_CMAQ_O	0.64	0.18	0.41	0.57
OMI_CMAQ_OD	0.99	0.36	0.71	0.60