RESPONSES TO REVIEWERS AND X. ZHANG

Ms. Ref. No.: Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2018-556.

Title: Nitrogen oxides in the global upper troposphere: interpreting cloud-sliced NO_2 observations from the OMI satellite instrument

Journal: Atmos. Chem. Phys. Discuss.

Reviewer comments are in blue. Responses are in black and include line numbers consistent with the updated manuscript with changes tracked. The manuscript starts on the 8th page of this pdf and the supplementary material requested by Reviewer #2 starts on the 25th page.

Responses to Anonymous Referee #1:

The manuscript by Marais et al. presents an application of satellite-derived NO2 data for the upper troposphere to diagnose NOx sources with the aid of the GEOS-Chem model. Two such products from the OMI instrument on the Aura satellite are evaluated in relation to aircraft data. The NASA product compared slightly better with the aircraft data than did the KNMI product. Therefore, the NASA product is used in comparison with GEOS-Chem output. This comparison suggests that the lightning NOx production in the model is too large, and the authors have scaled it down to better match the OMI-based data. The authors determined that there was no evidence to suggest larger NOx production efficiency per flash in the mid-latitudes than in the tropics. I have some concern about the level of detail of the lightning NOx emissions that are derived as I describe below. I consider this a major revision. Otherwise, my comments are minor.

Major Comment:

Neither of the OMI-derived UT NO2 products compared well with aircraft data. The better correlation (0.64) was with the NASA 450 - 280 hPa product at very coarse (20 x 32 degree resolution). However, this means R^2 = 0.4 and that the satellite-based data only capture 40% of the variance seen in the aircraft data averaged to this resolution. Is this really good enough to constrain a global chemistry model? If one assumes there is sufficient meaning in these data, the comparison with GEOS-Chem suggests that the lightning NOx emission per flash in the mid-latitudes should be reduced from 500 to 260 moles/flash, leading to an overall lightning source strength reduction from 6.5 to 5.5 TgN/year. However, the authors go on to scale the lightning production per flash upward or downward for each 20 x 30 degree grid cell. Any discrepancy between the OMI UT data and GEOS-Chem is being attributed to differences in NOx production efficiency per flash. Given the relatively poor comparison between OMI and the aircraft data and uncertain model UT NOy chemistry, I think this is taking the analysis too far. It is a real stretch to quantitatively believe the values given in Figure 7 and in lines 259 - 269. I would suggest eliminating Figure 7 and perhaps just comparing the derived NOx production per flash values for mid-latitudes as a whole and tropics as a whole. Figure 8 could stay, as although it contains individual grid cell value of NOx production efficiency, it does not contain specific regional values that someone might quote.

Thank you for pointing this out. We have removed Figure 7 and now only discuss aggregated (tropics, midlatitudes, and global) lightning NO_x production rates derived with OMI and GEOS-Chem (lines 359-364).

Minor Comments:

Introduction section: The authors need to add some more background material on previous uses of OMI (and earlier satellites) data for diagnosing lightning NOx production. The prior literature is in 2 categories: satellite data and model analyses (Boersma et al., 2005; Martin et al., 2007) and satellite-alone analysis (Beirle et al., 2010; Bucsela et al., 2010; Pickering et al., 2016)

Thank you for your suggestion. Studies listed in the comment include other NO_2 sensors (GOME and SCIAMACHY) and so we provide a general background to the application of tropospheric column NO_2, rather than specific to OMI, to estimate lightning NO_x emissions and production rates (lines 69-72).

line 48: Add Allen et al., 2010 after Tost et al.

Added (line 56).

lines 52-53: The 100-500 mol N/flash and 3-7 TgN/yr do not match. If one assumes the OTD/LIS climatological 46 flashes per second, 100 mol N/flash is about 2 TgN/yr and 500 mol N/flash is about 10 TgN/yr.

Thank you for pointing out this contradiction. We now clarify that higher production rates are typically applied to the northern midlatitudes than the tropics (line 61).

line 80: OMI was launched in July 2004.

This has been corrected (line 106).

line 90: For what pressure range was the Choi et al. (2014) product? Are there any other differences between that product and the NASA product used here?

We now specify the range of the mid-tropospheric product (900-650 hPa) (line 81) and also list the other differences between these products: cloud radiance fraction, number of OMI pixels, effective cloud scene pressure, and the cloud pressure product (lines 143-144, lines 159-160).

lines 94-95: How well does TM4 do at these estimates?

It is apparent in Figure 1 that TM4 is not able to correct for contamination over scenes with large surface sources and intermediate cloud fraction (southern Africa biomass burning in June-August). Rather than identify flaws in TM4 specifically, we reiterate that Belmonte-Rivas et al. (2015) caution that the correction relies on accurate simulation of NO_2. (lines 177-178 and 180-181)

line 101: I don't understand how this difference yields a column for 330-450 hPa.

The assumption for both products is that the NO_2 concentration is uniform across the pressure range of interest. For clarity, we now state the pressure centre and pressure range of each product (lines 127 and 132).

line 158: ....lightning NOx emissions and convective transport of boundary layer pollution....

Added (line 216).

line 212: What percentage is this?

About 20-40% overestimate. This is now provided (line 277-278).

lines 216 - 222: Are these comparisons for the model with 6.5 Tg N?

We now clarify that GEOS-Chem simulations that follow use a single global lightning NO_x production rate of 260 mol N per flash (lines 278-279).

line 221: domain average UT NO2 is 19% lower than aircraft data. The opposite bias is present in comparing the model with OMI. Which should you believe?

We now clearly state that the model simulations from Figure 3 onwards use 260 mol N per flash everywhere (to address the previous comment).

line 242: Why would this be the case?

We reanalyzed the data to obtain a linear relationship between UT NO2 and lightning flashes to address one of the major comments by reviewer #2. This updated analysis is shown in Figure 6 in the manuscript and discuss the implications for UT NOx (lines 330-337).

line 259: OMI-derived and GEOS-Chem lightning NOx production.

Changed throughout (lines 361, 485, Figure 7 caption).

line 289: 5.6 TgN/yr doesn't match the 5.5 TgN/year mentioned in line 214.

Fixed (lines 26, 507).

Responses to Anonymous Referee #2:

In this manuscript, upper tropospheric NO2 columns from two different cloud slicing approaches applied to one year of OMI data are used to study the impact of lightning on upper tropospheric NO2. First, data from the two retrievals is compared with each other and with aircraft observations taken over North America in the period March –August of the same year. Data from the NASA algorithm are then compared to GEOS-Chem model data for two seasons. Scatter plots of NO2 columns from both model and satellite retrievals against lighting flashes from the LIS/OTD climatology are compared and the conclusion is drawn, that UT NO2 from OMI is largely dominated by lightning NOx. Finally, spatially and seasonally resolved maps of NOx production per flash are computed from the ratio of retrieved to modelled UT NO2, and the dependency of these production rates on LIS lightning properties is evaluated.

Measurements of NO2 in the UT are sparse and the use of satellite data for validation of model results in this important atmospheric region is of high scientific interest. The approach taken by the authors is interesting and the manuscript overall well written, although I would have hoped to get more details on what exactly was done in many places.

Nevertheless, the paper leaves me a bit helpless as my impression is, that combining the uncertainties of the individual steps taken in this analysis will make the results basically worthless. More specifically,

- *the two retrievals which are based on the same data and on quite similar assumptions lead to very different results on UT NO2,*

The two products follow very different retrieval steps. We now iterate that this is the case in Section 2 and point readers to retrieval details that follow (lines 115).

- *the comparison with airborne measurements shows only broad agreement, and that only if data are averaged over large areas,*

The intention in this work is to evaluate whether and at what temporal and spatial resolutions two new satellite-derived products provide useful information about global UT NO_2, and which of the two retrieval approaches leads to data that is consistent with aircraft observations. This guides future retrievals with higher resolution instruments like TROPOMI. We now state this in Conclusions (lines 501-502).

- *the conclusion that the main driver for the observed UT NO2 variability is lightning Is probably correct in general but clearly not for individual points in Fig. 6,*

We now soften this claim in the Abstract (line 24).

- *computing NOx emission rates per flash by taking ratios between model and measurement in the scattered distributions shown in Fig. 6 seems really optimistic to me.*

We have removed Figure 7 (following the recommendation of Reviewer #1) and now only evaluate aggregated (midlatitudes, tropics, global) lightning NO_x production rates.

I'm also surprised by the briefness of the discussion of the log-linear relationship found between lightning frequency and NO2. Is this a known fact, and is there an explanation for it? The fact that this relationship is not so clear in GEOS-Chem data would not lead me to the conclusion that NOx lifetime is lower at high lightning frequencies (how would that follow from the lightning parametrization used? Are non-linear effects really expected at the relatively coarse resolution of the model?) and that NO2 observations are uncertain at low concentrations (there are no observations in the model figure). I would rather suspect that other factors such as transport, vertical mixing, and chemistry are also important drivers of upper tropospheric NO2 in addition to lightning, which would explain that very large changes in lightning frequency are needed to see moderate changes in UT NO2.

Thank you for your comment. We now present the linear relationship between OMI UT NO2 and lightning flashes (Figure 6) and discuss the implication for UT NOx (lines 330-337).

The variations in NOx production per flash shown in Fig. 7 are large in many places, and would be important input for global modelling studies. However, an error bar is needed for these numbers before they can be used, and maybe this is the reason why the authors don't mention them in abstract and conclusions.

We have removed this figure (following the recommendation of Reviewer #1) and now only evaluate aggregated (midlatitudes, tropics, global) lightning NO_x production rates.

In summary, I cannot recommend this paper for publication in the current form. Before it can be published, the authors need to add more detail on the individual steps of the analysis and the data used, they need to provide uncertainty estimates and explain how they were derived, and also should add more discussion on how the fact that lightning is not the only factor affecting UT NO2 impacts on their results and conclusions.

Thank you for your comment. We have responded to major comments above and minor comments below to address these concerns.

Minor Comments:

Introduction / beginning of section 2: There is a lot of repetition here, please read again and shorten where possible.

We have deleted repetitive statements (apparent in the tracked changes on lines 30, 72, 76, 82).

page 3, line 97: Is aerosol really accounted for in the NO2 air mass factors, and if so, how?

Yes. We now reference Boersma et al. (2004) (line 122) that is in turn referenced by Belmonte-Rivas et al. (2015) when describing the air mass factor calculation.

page 3, line109: What are near-Lambertian clouds?.

We now define these clouds with uniform scatter that are optically thick and geometrically thin (line 137).

page 4, line 124: Why is the slant column offset affecting the UT NO2 data – I thought this is cancelled by the stratospheric correction?

We have removed this statement to avoid confusion (line 170).

page 4, line 133: How do the authors know which signatures in the figures are real, and which linked to misrepresentation of lower tropospheric signals?

We are guided by the location of lightning flashes to determine which features are real in Figure 1 and by prior information about large surface sources of pollution to determine which features are associated with contamination.

page 7, line 199: I understand that aircraft measurements are screened for stratospheric air masses in tropospheric applications. However, here data are compared to satellite retrievals, and these will - as far as I understand – include such stratospheric air masses if they are in the right pressure range above a cloud. I therefore wonder if this screening really makes sense here.

Both the aircraft and satellite products exclude stratospheric contributions, and so the comparison is consistent. We now clarify that the OMI satellite product is tropospheric NO_2 only (line 74). We already state that the KNMI product removes stratospheric NO_2 by subtracting the stratospheric contribution in the retrieval (lines 121-122). We now elaborate that the stratospheric contribution in the NASA product is removed when differencing two nearby partial columns, as NO_2 aloft is assumed uniform (lines 134-135 and lines 141-142).

page 7, Fig. 4. It is unfortunate, that here another time period is shown than in Fig. 1. As airborne data is collected over a period of 6 months, seasonal variability in UT NO2 could play a role in the comparison to

satellite retrievals. I'd therefore suggest to show all 4 seasons in Fig. 1 or at least to add this figure to the Appendix / Supplement.

Thank you for your suggestion. We now include the other seasons in supplemental Figure S1 (referenced in lines 163, 168, and 180).

page 8, Fig. 5. Again, I would suggest to add the other seasons as well.

We now also include these in supplemental Figure S2 (referenced in lines 291-292).

Responses to X. Zhang Interactive Comment:

Well written paper about the cloud slicing method.

I have a few comments below:

1) L120: What's the definition of correlation between coincident gridsquare (R)? How did you calculate that?

We now specify in the text that this is the Pearson's correlation to determine spatial consistency between the two products in each season (lines 165-166).

2) L205: What's the ratio between intra-cloud (IC) and cloud-to-ground (CG) lighting? This will also affect UT NO2.

The GEOS-Chem parameterization does not distinguish NO_x production from between and within clouds and cloud-to-ground lightning (now stated in lines 279-281). Different flash types likely influence the amount of NO_x generated per flash (Schumann and Huntrieser, 2007), but models have poor predictive capability for the location of lightning flashes and by extension the type of flash (Murray, 2016).

3) L208: You mentioned that the modeled lightning flashes are redistributed to match LIS/OTD HRMC. How did you redistribute these flashes? Because lightning NOx can affect chemical reactions. Is this method online?

Scaling factors based on the discrepancy between the model estimate of lightning flashes and LIS/OTD monthly climatologies are used to correct for biases in the magnitude of the spatial distribution of lightning flashes (Murray et al., 2012). We have specified this for clarity (line 267).

4) L210: It's better to explain the origin of both lightning production rates.

We have rewritten this section for clarity (lines 275-281).

5) L215: How about the result of adjusting the production rate when compared with OMI?

This line doesn't correspond to any text, so we are unsure what is being asked.

6) L237: Did you exclude contamination of UT NO2 like southern Africa and northeast China when calculating the relationship?

We now explicitly state in the text that observations over northeast China are not considered in the comparison (lines 331-333). We only consider the NASA product in Figure 6 that has no observations over southern Africa in June-August (Figure 1).

References:

Schumann and Huntrieser, doi: 10.5194/acp-7-3823-2007, 2007.

Murray et al., doi:10.1029/2012JD017934, 2012.

Murray, doi:10.1007/s40726-016-0031-7, 2016.

Belmonte-Rivas et al., doi:10.5194/acp-15-13519-2015, 2015.

Boersma et al., doi:10.1029/2003JD003962, 2004.

Nitrogen oxides in the global upper troposphere: interpreting cloud-sliced NO_2 observations from the OMI satellite instrument

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Abstract. Nitrogen oxides ($NO_x \equiv NO + NO_2$) in the upper troposphere (UT) have a large impact on global tropospheric ozone and OH (the main atmospheric oxidant). New cloud-sliced observations of UT NO_2 at 450-280 hPa (~6-9 km) from the OMI satellite instrument produced by NASA and KNMI provide global coverage to test our understanding of the factors controlling UT NO_x. We find that these products offer useful information when averaged over coarse scales (20° × 32°, seasonal), and that the NASA product is more consistent with aircraft observations of UT NO_2. Correlation with LIS/OTD satellite observations of lightning flash frequencies suggests that lightning is the dominant source of NO_x to the upper troposphere except for extratropical latitudes in winter. We infer a global mean NO_x yield of 280 moles per lightning flash, with no significant difference between the tropics and midlatitudes, and a global lightning NO_x source of 5.5 Tg N a^{-1}. There is indication that the NO_x yield per flash increases with lightning flash footprint and with flash energy.

1. Introduction

Nitrogen oxides ($NO_x \equiv NO + NO_2$) in the upper troposphere (UT) have profound effects on the oxidizing capacity of the atmosphere and on climate, but the factors controlling their concentrations are poorly understood. NO_x in the UT impacts climate by efficiently producing ozone where it is a potent greenhouse gas (Dahlmann et al., 2011; Worden et al., 2011; Rap et al., 2015) and by increasing the concentration of OH (the main tropospheric oxidant) (Murray et al., 2012; Murray et al., 2014). Primary NO_x sources in the UT include lightning, aircraft, convective injection, and downwelling from the stratosphere (Ehhalt et al., 1992; Jaeglé et al., 1998b; Bertram et al., 2007). NO_x cycles chemically with reservoir species including nitric acid (HNO_3), pernitric acid (HNO_4), dinitrogen pentoxide (N_2O_5), peroxyacetylnitrate (PAN), and other organic nitrates, thus defining the NO_y chemical family ($NO_y \equiv NO_x$ + reservoirs). Effective loss of NO_x from the UT is through subsidence of NO_y to lower altitudes where deposition of HNO_3 provides the ultimate sink. The residence time of NO_y in the UT is 10-20 days (Prather and Jacob,

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1997). The lifetime of NO_x against conversion to short-lived reservoirs varies from ~3 hours in the convective outflow of thunderstorms to 0.5-1.5 days in background air (Nault et al., 2016). Chemical recycling from these reservoirs maintains relatively high UT NO_x background concentrations (Bradshaw et al., 2000; Baehr et al., 2003; Nault et al., 2016).

Representation of lightning NO_x in chemical transport models (CTMs) is particularly uncertain. Physically-based parameterizations relating lightning frequency to deep convective cloud tops, convective mass flux, convective precipitation, or high-cloud ice content have poor predictive capability (Tost et al., 2007; Allen et al., 2010; Murray et al., 2012; Finney et al., 2014), limiting our ability to estimate the response of lightning NO_x to future climate (Finney et al., 2016; 2018). An alternative is to prescribe flash densities from space-based observations and static NO_x production rates per flash (Sauvage et al., 2007; Allen et al., 2010; Murray et al., 2012). NO_x production efficiencies per flash in the literature vary from <10 to 5000 moles nitrogen per flash (mol N fl^{-1}) (Schumann and Huntrieser, 2007; Murray, 2016). Global chemical transport models (CTMs) typically use 100-500 mol N fl^{-1}, sometimes assuming higher production rates at mid-latitudes than in the tropics (Hudman et al., 2007; Ott et al., 2010), and a global lightning NO_x source of 3-7 Tg N a^{-1} to match observations of tropospheric ozone and NO_y species (Sauvage et al., 2007).

Our understanding of UT NO_x has so far been evaluated with observations from aircraft campaigns (Drummond et al., 1988; Jacob et al., 1996; Crawford et al., 1997; Jaeglé et al., 1998a; Bradshaw et al., 2000; Hudman et al., 2007; Stratmann et al., 2016). There are also long-term NO_x measurements from instruments onboard commercial aircraft dating back to the 1990s, but these are mostly over the north Atlantic and the NO_2 measurements have low precision and interference from thermally unstable NO_y reservoir compounds (Brunner et al., 2001). A number of studies have used satellite observations of tropospheric NO_2 columns from solar backscatter to infer lightning NO_x emissions (Beirle et al., 2010; Pickering et al., 2016), including in combination with global models (Boersma et al., 2005; Martin et al., 2007; Bucsela et al., 2010; Miyazaki et al., 2014). These studies estimate global lightning NO_x emission estimates of 1 to 8 Tg N a^{-1}.

New cloud-sliced satellite products of tropospheric NO_2 mixing ratios at 280-450 hPa (~6-9 km) offer additional vertical resolution by retrieving partial NO_2 columns above clouds and exploiting differences in heights of neighboring clouds to calculate NO_2 mixing ratios (Choi et al., 2014; Belmonte-Rivas et al., 2015). There are two new products of seasonal mean UT NO_2 mixing ratios retrieved from Ozone Monitoring Instrument (OMI) partial NO_2 columns by research groups at KNMI and NASA. The KNMI product has been evaluated against UT NO_2 from the Tracer Model version 4 (TM4) CTM. Large regional differences between OMI and TM4 are attributed to model deficiencies in lightning NO_x and uplift of anthropogenic pollution (Belmonte-Rivas et al., 2015). The NASA UT product is new to this work and follows a similar retrieval approach to the mid-tropospheric (900-650 hPa) product of Choi et al. (2014). That product was evaluated with aircraft observations of NO_2 and interpreted with the Global Modeling Initiative (GMI) CTM (Choi et al., 2014). Choi et al. (2014) identified large discrepancies between modeled and observed NO_2 seasonality over regions influenced by pollution and lightning.

Here we compare the two UT NO_2 products, obtained with distinct retrieval methods, and use aircraft observations of NO_2 from multiple NASA DC8 aircraft campaigns to arbitrate and evaluate the information that can be derived from the satellite datasets. We go on to test current understanding of UT NO_x and the implications for lightning emissions using the GEOS-Chem CTM.

2. OMI observations of upper troposphere NO_2

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OMI is onboard the NASA Aura satellite launched into sun-synchronous orbit in July 2004. It has an overpass time of about 13h30 local time (LT), a swath width of 2600 km, and a horizontal resolution of 13 km × 24 km at nadir (Levelt et al., 2006). Columns of NO_2 along the instrument viewing path (slant columns) are obtained by spectral fitting of solar backscattered radiation in the 405-465 nm window (Boersma et al., 2011). Standard products of total and tropospheric column NO_2 are screened for cloudy scenes using a cloud radiance fraction threshold of 0.5. Partial columns of NO_2 above cloudy scenes can be used to estimate vertically resolved NO_2 mixing ratios, as was first demonstrated with satellite observations of ozone (Ziemke et al., 2001). This approach, so-called cloud slicing, assumes a uniform trace gas concentration between two horizontally nearby clouds at different altitudes, so that the UT NO_2 mixing ratio is proportional to the slope of the partial columns versus the corresponding cloud pressures at the optical centre of the cloud. Two products of seasonal mean UT NO_2 have been retrieved from OMI following distinct retrieval steps detailed below: a product from KNMI at 330-450 hPa for 2006 (Belmonte-Rivas et al., 2015) and from NASA at 280-450 hPa for 2005-2007 following an approach similar to that used to retrieve mid-tropospheric NO_2 (Choi et al., 2014). In what follows we distinguish the two OMI NO_2 products as KNMI and NASA.

The KNMI product uses DOMINO v2.0 slant columns (Boersma et al., 2011) and OMCLDO2 cloud fractions and altitudes (Acarreta et al., 2004) over partially to very cloudy scenes (cloud radiance fraction > 0.5). Contamination due to NO_2 from below (up to 66% over polluted land masses) is estimated using the TM4 model and removed. Stratospheric NO_2 from an assimilated product (Belmonte-Rivas et al., 2014) is also removed. An air mass factor (AMF) (detailed in Boersma et al. (2004)) that accounts for viewing geometry, surface albedo, light attenuation by gases and aerosols along the viewing path, and sensitivity to NO_2 vertical distribution is applied to the resultant UT slant columns to convert to vertical columns. Additional data filtering removes scenes with solar zenith angle (SZA) ≥ 70° and surface albedo ≥ 30%. Resultant daily vertical partial columns are aggregated on consistent pressure and horizontal (1° × 1°) grids and used to determine seasonal mean UT NO_2 mixing ratios for gridsquares with at least 30 measurements. UT NO_2 centred at 380 hPa (pressure range 330-450 hPa) is estimated as the difference between columns at 380 hPa to the tropopause and at 380-500 hPa. Biases from sampling cloudy scenes, such as the effect of clouds on photochemistry, are corrected using TM4. These are small (typically <20%) in the UT (Belmonte-Rivas et al., 2015).

The NASA UT NO_2 product for 2005-2007, centred at 350 hPa (pressure range ~280-450 hPa), uses updated version 3 slant columns (OMNO2 v3.0) (Krotkov et al., 2017) that correct for a positive bias in the DOMINO v2.0 product with improved spectral fitting (Marchenko et al., 2015; van Geffen et al., 2015). Partial columns from the cloud height to the top of the atmosphere are retrieved for individual OMI pixels above very cloudy scenes (cloud radiance fraction > 0.7) to minimize contamination from below. Cloud fraction and height is from the OMCLDO2 product (Acarreta et al., 2004). The AMF accounts for viewing path geometry and light scattering by clouds with uniform scatter that are optically thick and geometrically thin (near-Lambertian clouds). Data filtering is applied to remove scenes with SZA > 80° and snow/ice cover and severe aerosol pollution that could be misclassified as clouds. Daily UT NO_2 is estimated for neighboring partial columns with sufficient cloud variability (cloud pressure distance > 160 hPa) and well-mixed NO_2 (NO_2 vertical gradient < 0.33 pptv hPa^{-1} diagnosed with the GMI CTM). The stratospheric column is assumed uniform above neighboring clouds and so is removed when differencing two nearby partial columns. Daily values of UT NO_2 are gridded to obtain seasonal means at 5° × 8° (latitude × longitude) for scenes with at least 50 measurements. Gaussian weighting is applied to assign higher weighting to UT NO_2 closest to 350 hPa. Choi et al. (2014) used a similar approach to retrieve mid-tropospheric NO_2, except cloud fraction and height were from the OMCLDRR

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product, and a stricter cloud radiance fraction of 0.9, a minimum of 30 measurements, and a wider minimum cloud pressure distance of 200 hPa were used.

Figure 1 compares seasonal mean UT NO_2 from the two satellite products in December-February and June-August. KNMI NO_2 is gridded to the NASA coarse grid. Data for March-May and September-November are in the Supplement (Figure S1). KNMI NO_2 has greater coverage than the NASA product, due to a lower cloud fraction threshold in the retrieval. The two products exhibit very different spatial features. Spatial correlation between the two products (Pearson's correlation coefficient between coincident gridsquares) is R = 0.41 in December-February, R = 0.38 in June-August. There is marginal improvement in the correlation with further spatial averaging. At 20° × 32° we find R = 0.50 in December-February and R = 0.45 in June-August. The correlation only increases substantially in September-November from R = 0.49 at 5° × 8° (Figure S1) to R = 0.66 at 20° × 32°. KNMI is systematically lower than NASA in all seasons for coincident gridsquares, varying from 16% lower in June-August to 48% lower in December-February at 20° × 32°.

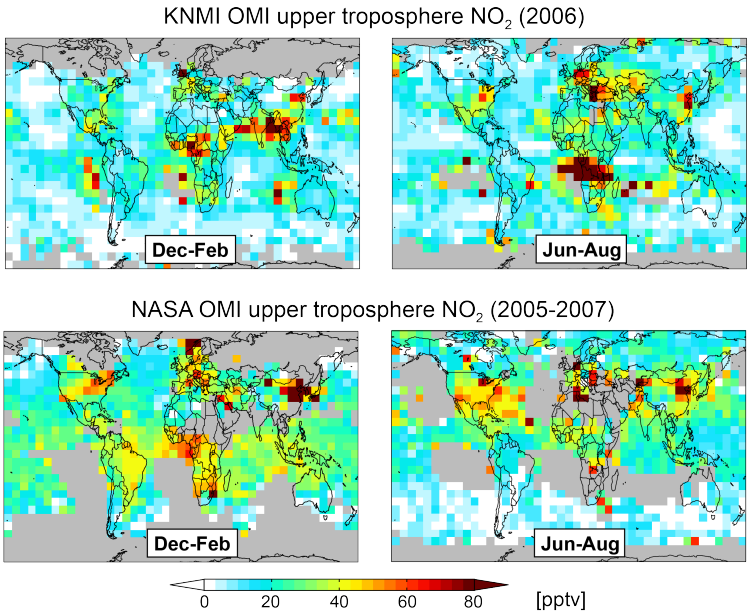

Figure 1. Upper troposphere (UT) NO_2 from the OMI satellite instrument. Seasonal mean UT NO_2 from KNMI in 2006 at 330-450 hPa (top) is compared to NASA in 2005-2007 at 280-450 hPa (bottom). Data are at 5° × 8° horizontal resolution for December-February (left) and June-August (right). Grey areas indicate no data and, for NASA, scenes with fewer than 50 measurements.

Contamination of UT NO_2 from below the cloud may still be present in the datasets despite attempts to correct for this using the TM4 model in the case of KNMI and by only considering very cloudy scenes in the case of NASA. These include a large enhancement in KNMI NO_2 (> 90 pptv) over southern Africa in June-August when there is intense biomass burning, and the NO_2 hotspot over northeast China in all seasons in both products (Figures 1, S1). Belmonte-Rivas et al. (2015) caution that the contamination correction in the KNMI product relies on accurate simulation of NO_2 vertical distribution.

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3. Evaluation of OMI upper troposphere NO_2 with aircraft observations

The aircraft observations we use to evaluate the OMI NO_2 products are from thermal-dissociation laser-induced fluorescence (TD-LIF) instruments (Day et al., 2002) for NASA DC8 aircraft campaigns over North America and Greenland in spring-summer when there is a high density of measurement campaigns. These include INTEX-A, INTEX-B, ARCTAS, DC3, and SEAC^{4}RS. Only INTEX-B is in the same year as the OMI products but we consider interannual variability to be only a small source of error. Measurements of NO_2 from TD-LIF are susceptible to interference from decomposition of thermally unstable reservoir compounds methyl peroxy nitrate ($CH_3O_2NO_2$) and HNO_4, in particular in the UT, where NO_2 concentrations are relatively low, temperature gradients between the instrument inlet and ambient air are large, and reservoir compounds are abundant (Browne et al., 2011). Publicly available DC3 and SEAC^{4}RS TD-LIF NO_2 are already corrected for this interference. We apply a correction for the other campaigns using the relationship between temperature and percentage interference from Browne et al. (2011). Observed mean ambient air temperature in the UT during INTEX-A is 246 K, corresponding to 20% interference. That for INTEX-B is 241 K (30% interference) and 236 K for ARCTAS (38% interference).

There are also NO_2 observations from the recent NASA ATom campaign, and from the In-service Aircraft for a Global Observing System (IAGOS) commercial aircraft campaign (Berkes et al., 2017). These use chemiluminescence instruments that are also susceptible to interference. Chemiluminescence and TD-LIF NO_2 are consistent during the SEAC^{4}RS campaign for the altitude range considered in this work (6-9 km) (Travis et al., 2016), but the interference from chemiluminescence is challenging to quantify, due to dependence also on the operator and instrument design that varies across campaigns (Reed et al., 2016).

Figure 2 shows the sampling extent of TD-LIF UT NO_2 over North America and Greenland in spring-summer at 450-280 hPa, around the satellite overpass (11h00-16h00 LT) for scenes not influenced by the stratosphere (diagnosed with collocated ozone/CO > 1.25 mol mol^{-1} (Hudman et al., 2007)). Concentrations of UT NO_2 exceed 80 pptv over the eastern US due to lightning NO_x emissions and convective transport of boundary layer pollution, and are < 30 pptv over the rest of the domain.

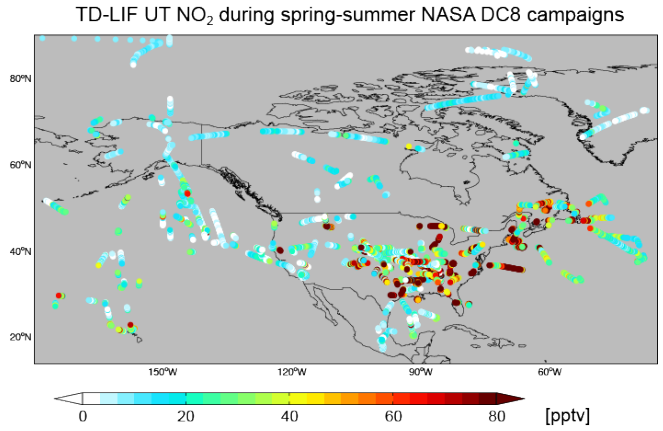

Figure 2. NASA DC8 upper troposphere NO_2 over North America in spring-summer (March-August). Observations are from the TD-LIF instrument at 450-280 hPa, 11h00-16h00 local time, and without stratospheric influence. Campaigns include INTEX-A in June-August 2004 (Singh et al., 2006), INTEX-B in March-May 2006 (Singh et al., 2009), ARCTAS

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in March-April and June-July 2008 (Jacob et al., 2010), DC3 in May-June 2012 (Barth et al., 2015), and SEAC^{4}RS in August 2013 (Toon et al., 2016).

Figure 3 shows the spatial correlation between March-August mean gridded aircraft and OMI UT NO_2 from the 2 products as a function of horizontal resolution. There is no spatial consistency between the OMI products and aircraft NO_2 at 5° × 8° (R < 0.1) and 10° × 16° (R < 0.2). The correlation improves for both products with further spatial averaging, peaking at 20° × 32° (R = 0.56 for KNMI, R = 0.64 for NASA). The satellite products are also spatially consistent at this resolution over this domain (R = 0.89), but KNMI is 43% lower than NASA.

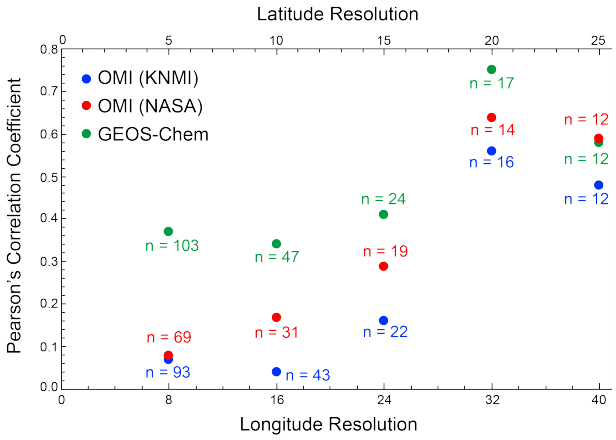

Figure 3. Evaluation of OMI and GEOS-Chem upper troposphere NO_2 with aircraft observations. Individual points are Pearson's correlation coefficients between gridded March-August mean UT NO_2 measured from the aircraft and OMI KNMI in 2006 (blue), OMI NASA in 2005-2007 (red), and GEOS-Chem in 2006 (green) at 5° × 8° (latitude × longitude), 10° × 16°, 15° × 24°, 20° × 32°, and 25° × 40°. Values inset are the number of points at each resolution. The domain sampled is shown in Figure 2.

Figure 4 compares the spatial distrbution of OMI and aircraft UT NO_2 at 20° × 32° over North America. Domain mean KNMI UT NO_2 is 38% lower than the aircraft observations, compared to 2.2% higher for NASA UT NO_2. Both products exhibit less variability (reduced major axis, RMA, regression slopes < 1) and high bias in background NO_2 compared to the aircraft observations (positive RMA intercepts of 5.9 ± 1.4 pptv for KNMI and 9.2 ± 2.7 pptv for NASA). We proceed with the NASA UT NO_2 product at 20° × 32°, as correlation peaks at this resolution and the NASA product is more consistent with domain mean aircraft UT NO_2 than the KNMI product.

4. Constraints on upper tropospheric NO_x

The NASA product provides near-global coverage of UT NO_2 to assess current understanding of regional UT NO_x sources and dynamics by comparing to UT NO_2 from the GEOS-Chem CTM (version 10-01; http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_v10-01) driven with NASA MERRA-2 reanalysis meteorology. The model horizontal resolution is 2° × 2.5° and the output is regridded to 20° × 32° for comparison with OMI. GEOS-Chem is sampled under all-sky conditions. We find that the effect on NO_2 of sampling the model under cloudy conditions is small. Isolating NO_2 under very cloudy

conditions using MERRA-2 cloud fractions decreases modeled UT NO_2 by no more than 5 pptv in the tropics/subtropics and less at higher latitudes. We use output from the model for 2006 following a one-year spin-up for chemical initialization. Interannual variability in UT NO_2, determined as the difference between modeled 2006 and multi-year (mean 2005-2007) UT NO_2, is small (< 4 pptv) everywhere except central Africa year-round (4-12 pptv), the Arctic north of 60°N (up to 25 pptv), and the Middle East in June-August and northern India in March-May (both 10-20 pptv).

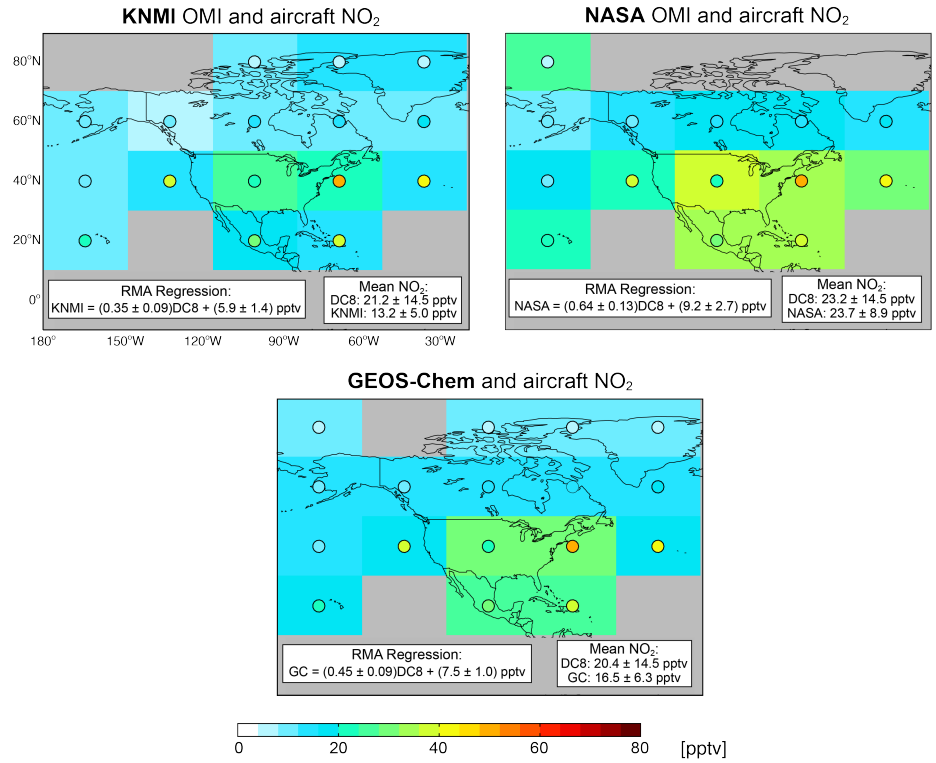

Figure 4. March-August upper troposphere NO_2 over North America. All data are at 20° × 32°. Background colors in the different panels show concentrations from KNMI, NASA, and GEOS-Chem (GC). Circles show the aircraft observations (same in all panels). Aircraft observations are for 11h00-16h00 LT. The model is sampled in the satellite overpass time window (12h00-15h00 LT). Model and aircraft data are at 280-450 hPa and screened for stratospheric influence using ozone/CO > 1.25 mol mol^{-1}. Inset boxes show reduced major axis (RMA) regression statistics and mean NO_2 for coincident gridsquares. Grey gridsquares indicate no observations.

Local GEOS-Chem emissions of NO_x in the UT include aircraft and lightning. Aircraft emissions from the AEIC inventory (Stettler et al., 2011) total 0.82 Tg N in 2006; much less than lightning in the same year (6.5 Tg N). Lightning in the model is estimated using the parameterization implemented by Murray et al. (2012). This includes an initial estimate of lightning flashes using the Price and Rind (1992, 1993, 1994) relationship between cloud-top height and lightning flashes. These are then scaled to the same annual global flash frequency (46 fl s^{-1}) and regional distribution as the climatology from the combined Lightning Imaging Sensor (LIS) and Optical Transient Detector (OTD) high-resolution monthly climatology (LIS/OTD HRMC) (Cecil et

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al., 2014). The standard GEOS-Chem model has higher NO_x yields per flash at northern mid-latitudes (north of 35°N) than in the tropics (500 mol N fl^{-1} versus 260 mol N fl^{-1}), but we find that this overestimated observed OMI UT NO_2 by 10-20 pptv (20-40%) at northern mid-latitudes in summer when the lightning source is dominant. Here we assume a NO_x yield of 260 mol N fl^{-1} everywhere. This decreases global lightning NO_x emissions by 15% from 6.5 to 5.5 Tg N a^{-1}. The lightning parameterization in GEOS-Chem does not distinguish lightning NO_x production for flashes within and between clouds (intra- or inter-cloud) and from the cloud to the Earth's surface (cloud-to-ground).

Figure 3 shows the spatial correlation between the model and aircraft observations. The model is more consistent with the aircraft observations than OMI at fine spatial resolution. Like OMI, GEOS-Chem correlation with the aircraft observations improves with spatial averaging, peaking at 20° × 32° (R = 0.75). Figure 4 also shows comparison of March-August GEOS-Chem UT NO_2 with the aircraft observations at 20° × 32°. The model is sampled over the same pressure range as NASA (280-450 hPa) around the OMI overpass (12h00-15h00 LT) and is filtered for stratospheric influence using model ozone/CO > 1.25 mol mol^{-1}. Domain average UT NO_2 from the model is 19% lower than the aircraft measurements and the model also overestimates background UT NO_2 (intercept = 7.5 ± 1.0 pptv) and underestimates the variability (slope = 0.45 ± 0.09).

Figure 5 compares seasonal mean OMI and GEOS-Chem UT NO_2 in December-February and June-August. The other seasons are shown in the Supplement (Figure S2). Formation of PAN, HNO_4 and $CH_3O_2NO_2$ accounts for over 75% of NO_x loss in the model in all seasons. Lower UT NO_2 in the northern hemisphere winter compared to summer in the model is mostly because lightning activity is at a minimum. The model underestimates UT NO_2 in the northern mid-latitudes in winter by 20-40 pptv, suggesting misrepresentation of another process in the model, such as excessive NO_x loss by N_2O_5 hydrolysis in aerosols (Kenagy et al., 2018). The particularly large bias over polluted regions in winter could also be due to contamination of the UT NO_2 retrievals by enhanced boundary layer NO_2.

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Deleted: The resultant lightning NO_x emissions are distributed vertically from the surface to the top of clouds using regional vertical profiles from Ott et al. (2010). We find that GEOS-Chem overestimates UT NO_2 in summer across the northern midlatitudes by 10-20 pptv (not shown) compared to OMI that we attribute to excessive lightning NO_x emissions. We correct for this overestimate by instead using a single global NO_x production rate of 260 mol N fl^{-1} in the model. This decreases global lightning NO_x emissions by 15% from 6.5 to 5.5 Tg N a^{-1}.

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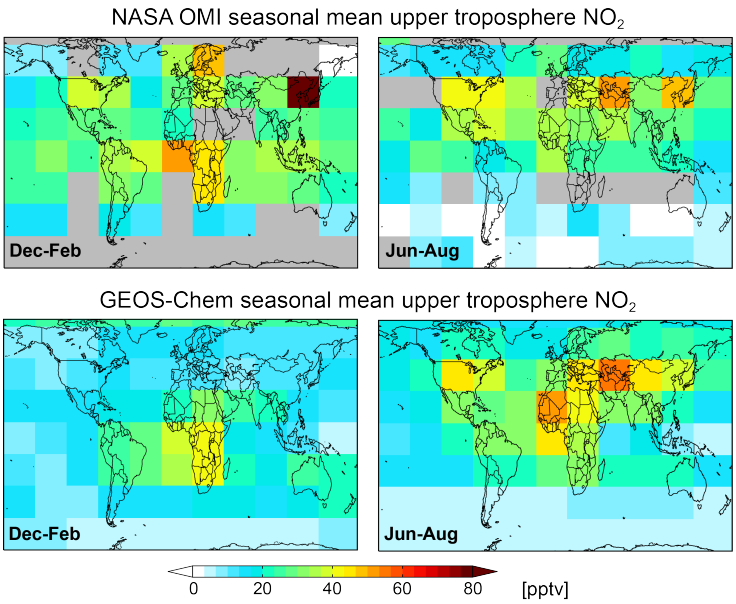

Figure 5. Observed and modelled upper troposphere NO_2. The figure shows NASA OMI seasonal mean UT NO_2 for 2005-2007 (top) and corresponding GEOS-Chem model values (bottom). The model is sampled at 280-450 hPa during the satellite overpass (12h00-15h00 LT), and filtered for stratospheric influence. Data are at 20° × 32° horizontal resolution for December-February (left) and June-August (right). Grey gridsquares in the top panel indicate no OMI data.

Figure 6 shows the linear relationship between seasonal mean LIS/OTD lightning flash climatology and seasonal mean UT NO_2 from OMI and GEOS-Chem. Data are divided into northern mid-latitudes and tropics seasonal means and exclude the contaminated observations over northeast China and wintertime northern mid-latitude gridsquares that show no correlation with lightning flashes ($R < 0.1$). Results from multi-model sensitivity studies indicate UT NO_x in winter is predominantly from surface sources, with a smaller contribution from extra-tropical lightning (Grewe et al., 2001). Background concentrations of UT NO_2 (intercepts in Figure 6) from non-lightning sources is 10-20 pptv, 3-5 pptv higher in the northern midlatitudes than the tropics. The slopes for the linear fit to the observations and model are similar for the northern mid-latitudes and the tropics, providing no support for the previously reported higher lightning NO_x production rates in the mid-latitudes than the tropics.

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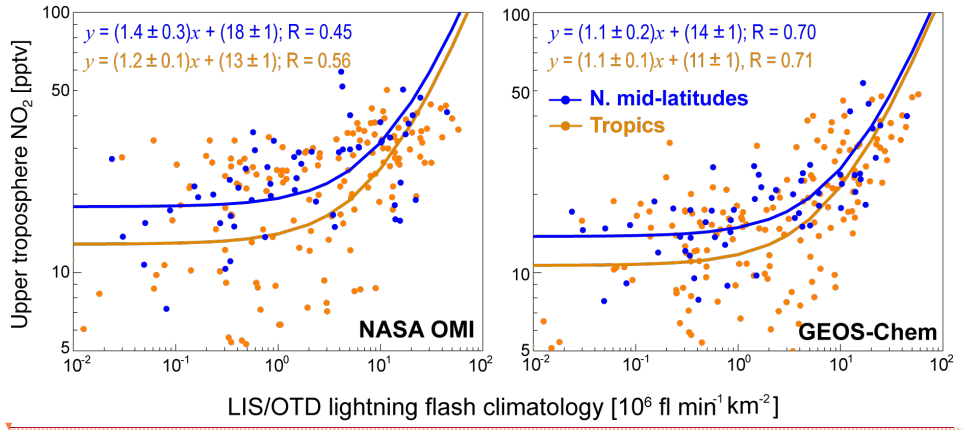


Figure 6. Linear relationship between upper troposphere NO_2 and lightning flash frequencies. Individual points are coincident seasonal mean UT NO_2 from OMI (left) and GEOS-Chem (right) versus seasonal mean LIS/OTD lightning flash climatologies for coincident 20° × 32° gridsquares in the northern mid-latitudes (> 30°N; blue) and tropics (< 30°N; orange). Northern mid-latitude points exclude December-February that show poor correlation with lightning flashes (see text for details). Lines show linear fit to data plotted on logarithmic scales. Values inset are Pearson's correlation coefficients and RMA regression statistics.

We go on to obtain OMI-constrained lightning NO_x production rates by scaling 260 mol N fl^{-1} by the ratio of OMI to GEOS-Chem UT NO_2 estimated in each 20° × 32° gridsquare (local discrepancies between the observations and model). Resultant local OMI and GEOS-Chem-derived production rates vary from 100 to 900 mol N fl^{-1} and values are higher in the tropics (300 ± 60 mol N fl^{-1}) than the northern mid-latitudes (270 ± 100 mol N fl^{-1}), but the difference is not significant. The global average (280 ± 80 mol N fl^{-1}) is similar to 310 mol N fl^{-1} obtained using multiple satellite observations of atmospheric composition (Miyazaki et al., 2014).

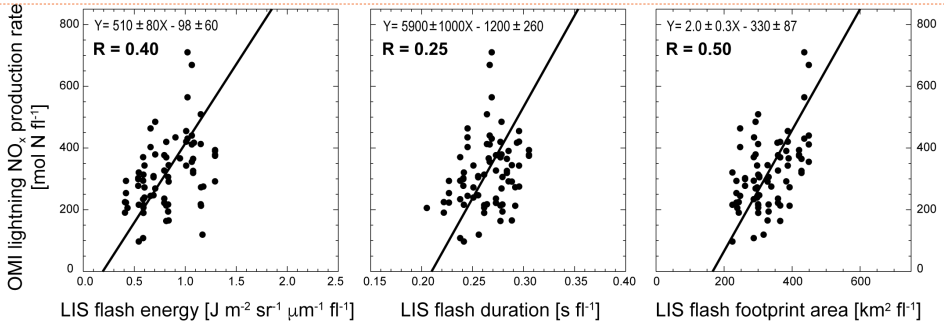
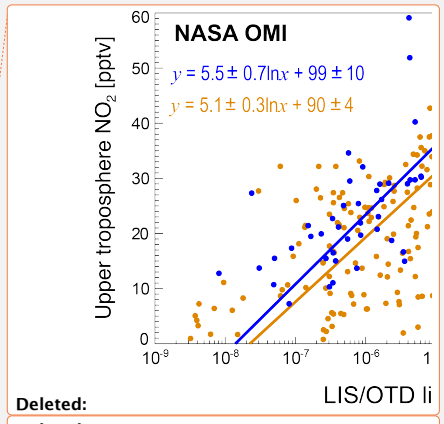


Figure 7. Relationship between OMI and GEOS-Chem derived lightning NO_x production rates and LIS lightning properties: energy (radiance), duration, and footprint area. Individual points are seasonal mean 20° × 32° gridsquares at 40°N-40°S.



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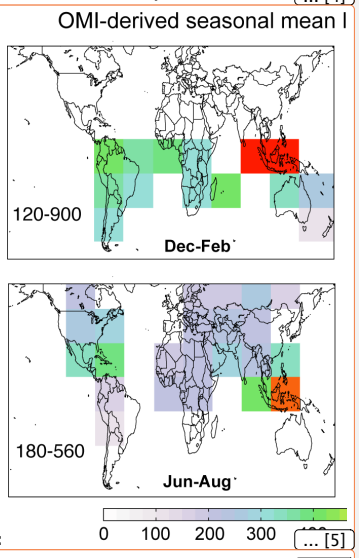

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Properties of lightning flashes including energy, duration, and footprint area, have been retrieved from the OTD and LIS sensors (Beirle et al., 2014). The flash footprint area is the spatial extent of lightning detection events contributing to the flash (collection of local events) diagnosed by the satellite data. Figure 7 shows the relationship between OMI and GEOS-Chem derived lightning NO_x production rates and LIS lightning properties from Beirle et al. (2014). The strongest correlation is with lightning extent (R = 0.50), followed by energy (R = 0.40). The correlation with flash duration is weak (R = 0.25). The relationships in Figure 7 suggest a dependence of lightning NO_x production rates on lightning flash energy of 510 ± 80 mol N (J m^{-2} sr^{-1} $\mu m^{-1})^{-1}$ and on flash footprint area of 2.0 ± 0.3 mol N km^{-2}., offering guidance for relating NO_x yields to physical properties in global models rather than the current approach of assigning static values.

5. Conclusions

The majority of measurements of NO_x in the upper troposphere (UT) are from measurement campaigns that are limited in space and time. Two new cloud-slicing UT NO_2 products from the Ozone Monitoring Instrument (OMI) produced by KNMI and NASA offer the potential to address uncertainties in our understanding of UT NO_x sources. We intercompare these products, evaluate them with aircraft observations, and use them to test and improve the GEOS-Chem model representation of UT NO_x.

The KNMI and NASA UT NO_2 products use different retrieval methods. They show weak global correlation at 5° × 8° (latitude × longitude), R = 0.4, and only marginal improvement when extended to 20° × 32° (R = 0.5-0.7). At that resolution they show correlation with in situ aircraft observations of UT NO_2 for different years (R = 0.56-0.64). The KNMI product is biased low by 38% relative to the aircraft observations while the NASA product has no significant bias. Although the OMI data can only provide coarse information on UT NO_2, measurements from the recently launched TROPOMI instrument with 7 km × 3.5 km nadir pixel resolution (compared to 13 km × 24 km for OMI) may be able to provide finer information in the future.

We find that the relationship of OMI UT NO_2 with LIS/OTD flash rates suggests most NO_x in the upper troposphere is from lightning, except in the mid-latitudes in winter. The relationship also suggests no difference in NO_x yields per flash between the mid-latitudes and the tropics, in contrast to the higher yields at mid-latitudes often assumed in models. We derive a global mean lightning NO_x production rate of 280 mol N fl^{-1} and estimate a global lightning NO_x emission of 5.5 Tg N.

Data Availability

Data from this work can be made available upon request to E. A. Marais for GEOS-Chem output, M. Belmonte-Rivas for KNMI UT NO_2, and S. Choi and J. Joiner for NASA OMI UT NO_2, and S. Beirle for LIS lightning properties.

Competing Interests

The authors declare that they have no conflicts of interest.

Author Contributions

EAM conducted model simulations, analysed and interpreted satellite, model, and aircraft data, and prepared the manuscript, DJJ provided supervisory guidance and assisted in the writing. SC, JJ, and MR-B retrieved the OMI UT NO_2 products, RCC aided in

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interpreting aircraft observations., LTM contributed LIS/OTD lightning flash observations, SB contributed lightning flash properties, LS, VS, and LJ contributed updated GEOS-Chem code.

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Tropospheric column observations of NO_2 are obtained from space-based UV/visible spectrom[JDJ1]eters that measure solar-backscattered radiation.

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The northern mid-latitudes data points in Figure 6 exclude December-February, when lightning is at a minimum and there is no correlation between NO_2 and lightning flashes is weak (= 0.33[EAM2]). Results from multi-model sensitivity studies indicate UT NO_x is then predominantly from surface sources, with a smaller contribution from extra-tropical lightning (Grewe et al., 2001). The model reproduces the observed slopes in Figure 6. Spatial correlation between OMI and LIS/OTD suggests that OMI UT NO_2 can be used to derive spatially and seasonally varying lightning NO_x production rates per flash by scaling 260 mol N fl^{-1} by the local ratio of observed-to-modelled (OMI/GEOS-Chem) UT NO_2.

Figure 7 shows the resultant seasonal mean OMI-derived [MOU3]lightning NO_x production rates at 20° × 32°.

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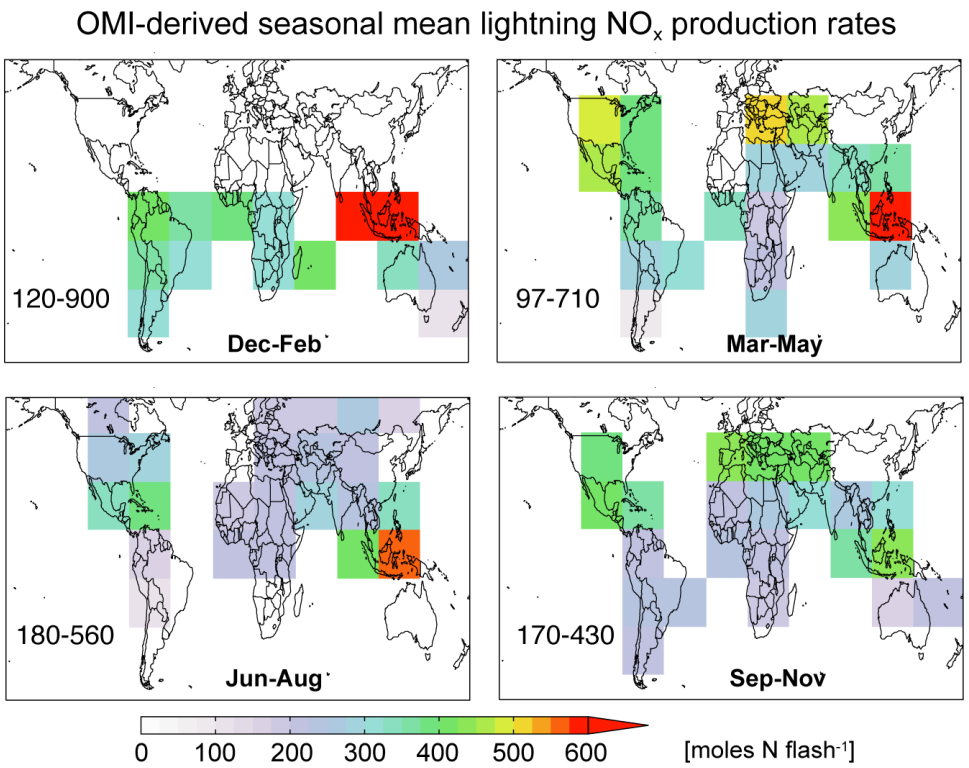

Figure 7. Lightning [MOU4]**NO_x production rates per flash estimated with OMI and GEOS-Chem. Maps show seasonal mean nitrogen (N) produced per flash at 20° × 32° for gridsquares with lightning flashes > 5 × 10^{-6} flashes km^{-2} min^{-1}. Values inset are the range in production rates for each season. White gridsquares remain unchanged (260 mol N fl^{-1}).**

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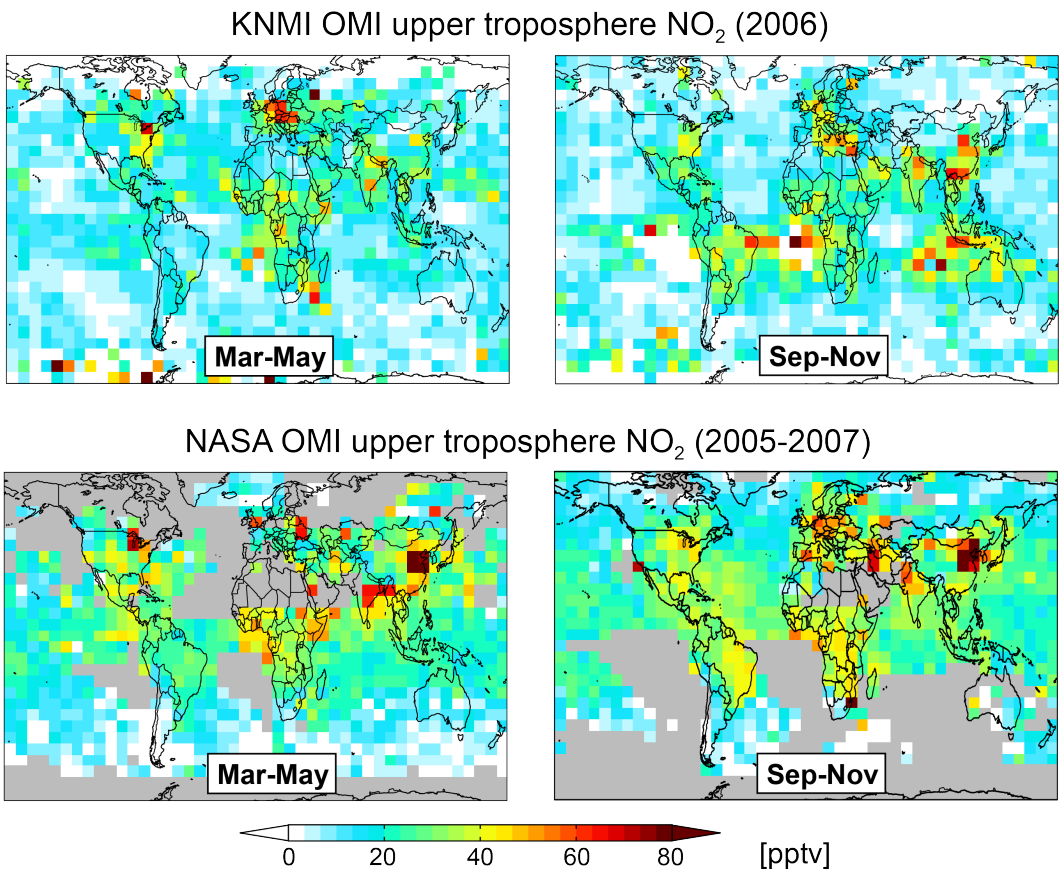

Fig. S1. Upper troposphere (UT) NO_2 from the OMI satellite instrument. Seasonal mean UT NO_2 from KNMI in 2006 at 330-450 hPa (top) is compared to NASA in 2005-2007 at 280-450 hPa (bottom). Data are at 5° × 8° horizontal resolution for March-May (left) and September-November (right). Grey areas indicate no data and, for NASA, scenes with fewer than 50 measurements.

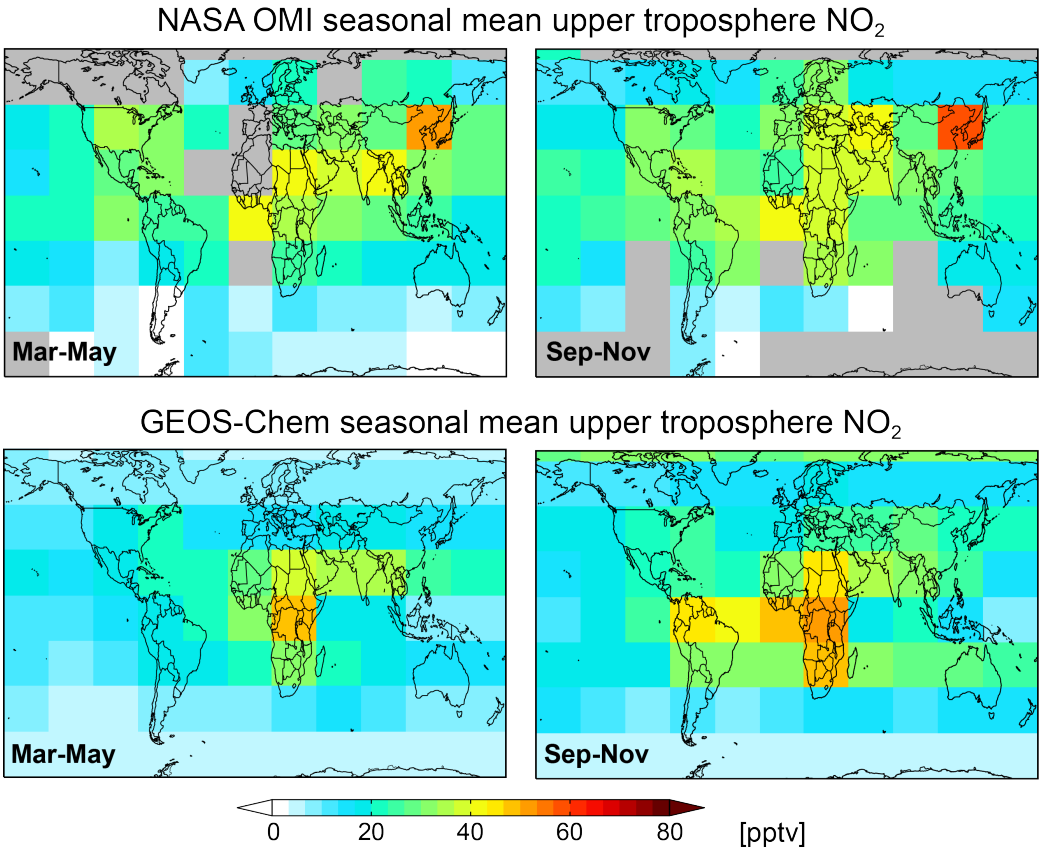

Fig. S2. Observed and modelled upper troposphere NO_2. NASA OMI for 2005-2007 (top) and GEOS-Chem (bottom) seasonal mean UT NO_2. The model is sampled at 280-450 hPa during the satellite overpass (12h00-15h00 LT), and filtered for stratospheric influence. Data are at 20° × 32° horizontal resolution for March-May (left) and September-November (right). Grey gridsquares in the top panel indicate no OMI data.