AGU PUBLICATIONS

Geophysical Research Letters

RESEARCH LETTER

10.1002/2017GL074436

Lightning NO_x Emissions: Reconciling Measured and Modeled Estimates With Updated NO_x Chemistry

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Key Points:
- The upper tropospheric NO_x lifetime is shorter than typically assumed
- A shorter NO_x lifetime improves agreement among various methods used to calculate mean lightning NO_x emission rates per flash
- Global lightning NO_x emission rates are estimated to be ~9 Tg N yr^{-1}

Supporting Information:
- Supporting Information S1

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Citation:
Nault, B. A., Laughner, J. L., Wooldridge, P. J., Crounse, J. D., Dibb, J., Diskin, G., … Cohen, R. C. (2017). Lightning NO_x emissions: reconciling measured and modeled estimates with updated NO_x chemistry. *Geophysical Research Letters*, *44*, 9479–9488. https://doi.org/10.1002/2017GL074436

Received 2 JUN 2017
Accepted 29 AUG 2017
Accepted article online 5 SEP 2017
Published online 18 SEP 2017

Abstract Lightning is one of the most important sources of upper tropospheric NO_x; however, there is a large spread in estimates of the global emission rates (2–8 Tg N yr^{-1}). We combine upper tropospheric in situ observations from the Deep Convective Clouds and Chemistry (DC3) experiment and global satellite-retrieved NO_2 tropospheric column densities to constrain mean lightning NO_x (LNO_x) emissions per flash. Insights from DC3 indicate that the NO_x lifetime is ~3 h in the region of outflow of thunderstorms, mainly due to production of methyl peroxy nitrate and alkyl and multifunctional nitrates. The lifetime then increases farther downwind from the region of outflow. Reinterpreting previous analyses using the 3 h lifetime reduces the spread among various methods that have been used to calculate mean LNO_x emissions per flash and indicates a global LNO_x emission rate of ~9 Tg N yr^{-1}, a flux larger than the high end of recent estimates.

Plain Language Summary Lightning is an important source of upper troposphere nitrogen oxides; however, there is high uncertainty in the amount of nitrogen oxides produced from lightning. Using recent updates in upper tropospheric nitrogen oxides chemistry, this study decreases this uncertainty from a factor of 4 to less than a factor of 2 and shows that the amount of nitrogen oxides produced from lightning should be higher.

1. Introduction

In recent analyses, emissions of NO_x ($NO_x \equiv NO + NO_2$) from lightning have been estimated to be in the range of 2–8 Tg N yr^{-1}, representing ~10% of the global and ~80% of the middle to upper tropospheric NO_x source (Murray, 2016; Schumann & Huntrieser, 2007). Systematic variations in convectively available potential energy (CAPE), wind shear, and flash length have been suggested as mechanisms driving variability in emissions (Schumann & Huntrieser, 2007). However, observations have not established which of these mechanisms control mean lightning NO_x (LNO_x) emission rates per flash (Hudman et al., 2007; Martin et al., 2007; Schumann & Huntrieser, 2007). The LNO_x emission range can lead to at least a 15% difference in globally modeled tropospheric O_3, as estimated by Labrador et al. (2005), and up to a 60% increase in tropical O_3 (Liaskos et al., 2015).

Two methods are typically employed to estimate mean LNO_x emission rates per flash. The first method emphasizes the near field of convection and uses observations from either in situ or space-based platforms. These studies are typically based on the idea that the NO_x lifetime in the upper troposphere (UT) is long (2–8 days); using this assumption, the observations are directly converted to total NO_x emitted without an adjustment for chemical loss prior to measurement. The NO_x measurements are combined with the number of lightning flashes observed in each of these individual events to yield the emission rate in units of mol NO flash^{-1} (Beirle et al., 2010, 2004; Bucsela et al., 2010; Huntrieser et al., 2007, 2009, 2011; Miyazaki et al., 2014; Pickering et al., 2016; Pollack et al., 2016). The second method uses in situ or space-based measurements in the far field to constrain a chemical transport model. The emission rate of NO from lightning is varied until the simulations broadly agree with measurements of NO_2, HNO_3, and/or NO_y (NO_y = NO + NO_2 + PAN + $CH_3O_2NO_2$ + HO_2NO_2 + alkyl and multifunctional nitrates + HNO_3 +…). Estimates of mol NO flash^{-1} are typically higher in far-field studies compared to near field, and both methods fail to accurately simulate the measured ratio of NO_x to HNO_3 (Allen et al., 2010, 2012; Fang et al., 2010; Hudman et al., 2007; Jourdain et al., 2010; Martini et al., 2011).

Here we largely resolve these conflicting interpretations of LNO_x. Recent improvements in our understanding of UT chemistry alter the relationships between near- and far-field concentrations of nitrogen oxides. We find that (1) the lifetime of NO_x in the near field of convection is much shorter than previously assumed (~2–3 h), reducing the downwind burden of NO_x relative to the magnitude of the emissions and, consequently, (2) that global LNO_x emissions are approximately 9 Tg N yr^{-1}—a value above the higher end of recent estimates.

2. Methods

We briefly describe the model and observations used in this study, and further details about the model and observations can be found in the supporting information.

Airborne observations from the Deep Convective Clouds and Chemistry (DC3) experiment were used in the analysis (Archive, 2014; Barth et al., 2015). Briefly, the NASA DC-8 aircraft sampled fresh and chemically aged LNO_x emissions over the continental United States, between May and June 2012. The details of the observations from the NASA DC-8 aircraft observations are described in the supporting information and include NO, NO_2, methyl peroxy nitrate ($CH_3O_2NO_2$), alkyl and multifunctional nitrates, HNO_3, O_3, and water vapor (Crounse et al., 2006; Day et al., 2002; Diskin et al., 2002; Nault et al., 2015; Ryerson et al., 1999; Talbot et al., 1997; Thornton et al., 2000). Descriptions of how the observations were filtered to match the chemical transport model can be found in the supporting information (Bertram et al., 2007; Cooper et al., 2014; Henderson et al., 2011; Hudman et al., 2007). Also, a discussion of the positive interference of thermally decomposed $CH_3O_2NO_2$ and pernitric acid on in situ UT NO_x observations from prior studies can be found in the supporting information (Browne et al., 2011; Nault et al., 2015).

The Ozone Monitoring Instrument (OMI) aboard the NASA Aura satellite is used to extend the observations, globally, for the year 2012 (Levelt et al., 2006). There are currently two products for global NO_2 column densities for OMI—the NASA Standard Product 2 (SP v2) (Bucsela et al., 2013) and the Dutch OMI NO_2 (DOMINO) Product (Boersma et al., 2011a, 2011b). For both, the Level 2 products were used to constrain LNO_x emissions around the world; the observations are matched to GEOS-Chem grid cells as described in the supporting information. Description of the air mass factor can be found in the supporting information (Boersma et al., 2002).

Finally, GEOS-Chem version 9-02 (http://geos-chem.org) (Bey et al., 2001) was used at 2° × 2.5° resolution. The standard chemistry, e.g., Sander et al. (2011), and model is described by Mao et al. (2013). Various changes to the chemical kinetics were analyzed (Table S1), and further details about GEOS-Chem can be found in the supporting information.

As a special note, we used the vertical distribution of LNO_x emissions recommended by Pickering et al. (1998). This profile was chosen since recent studies (Allen et al., 2012; Seltzer et al., 2015) conclude that the Ott et al. (2010) profile places too much NO_x in the middle troposphere and not enough NO_x in the UT.

3. Results

3.1. The Lifetime of NO_x

Our analysis begins by analyzing the impact of advances in our understanding of $CH_3O_2NO_2$ (Browne et al., 2011; Nault et al., 2015), HO_2NO_2 (Bacak et al., 2011; Nault et al., 2016), and dinitrogen pentoxide, N_2O_5 (Evans & Jacobs, 2005; Brown et al., 2009) and the reaction of OH with NO_2 to produce HNO_3 at the temperatures and pressures characteristic of convective outflow and the UT (Henderson et al., 2012; Nault et al., 2016) in the GEOS-Chem chemical transport model (Bey et al., 2001). Although, alkyl and multifunctional nitrates (ANs) are known to play an important role in removing NO_x in the outflow of convective systems (Apel et al., 2012; Nault et al., 2016), no updates to AN chemistry were added because the impact of ANs is subgrid scale in this modeling framework. As discussed later, we evaluate the impact of neglecting their formation post hoc.

GEOS-Chem is used to calculate NO_x and HNO_3 mixing ratios and tropospheric NO_2 column densities ($tVCD_{NO2}$). Evaluations of the changes to the chemical kinetics individually (Figures S1–S3) show that $CH_3O_2NO_2$ chemistry creates an additional sink for NO_x, reducing the chemical lifetime of UT NO_x and reducing the $tVCD_{NO2}$ and the UT HNO_3. The effect of $CH_3O_2NO_2$ on NO_x concentrations and $tVCD_{NO2}$ is partly balanced by the revised low temperature and low-pressure rate of the reaction of NO_2 with OH, which results in an increase in the NO_x lifetime and, thus, higher NO_x concentrations and $tVCD_{NO2}$. This change to the low temperature HNO_3 source reinforces the effect of $CH_3O_2NO_2$ on UT HNO_3, as it also results in reduced HNO_3. The changes to HO_2NO_2 and N_2O_5 chemistry are included for completeness, but they had little effect on this analysis. Overall, even with the slower formation rate of HNO_3, the UT NO_x lifetime is reduced compared to the previously assumed 2–8 days (Schumann & Huntrieser, 2007).

These model results are consistent with observations, as further discussed in section 3.2. For example, in the near field, Nault et al. (2016) found that $CH_3O_2NO_2$ and ANs accounted for ~70% of the NO_x loss, whereas HNO_3 accounted for ~15% of the NO_x loss. Combining all the recent updates, the UT NO_x day time lifetime, inferred from the photostationary steady state described in Nault et al. (2016) and observations, is ~3 h near thunderstorms (i.e., < 6 h chemical aging) (Nault et al., 2016). After ~6 h chemical aging and dilution, the highly reactive hydrocarbons and HO_x precursors are reduced below other factors controlling the NO_x lifetime (Apel et al., 2012; Bertram et al., 2007; Fried et al., 2008), with the effect that the UT NO_x lifetime away from thunderstorms is ~0.5–1.5 days (Nault et al., 2016). Thus, LNO_x is rapidly lost to chemical removal in the near field of thunderstorms, but remnant NO_x plumes can still be observed up to 1–2 days after a storm (Figure S4).

3.2. Effects of Updated Chemistry on Interpretation of Near-Field Studies

Determination of the mol NO $flash^{-1}$, using near-field analyses of NO_x or NO_2 concentrations, requires that the loss of NO_x between emission and measurement be accounted for. These analyses typically assume that the UT NO_x lifetime is controlled by dilution and production of HNO_3 through the reaction of OH with NO_2, with values for the lifetime of NO_x in the range 2–8 days (Schumann & Huntrieser, 2007). We find that a much shorter lifetime, ~2–3 h, is consistent with in situ observations and the revised model (Nault et al., 2016). As a result, much more NO_x is likely being converted to higher oxides between emission and measurement than was previously recognized, leading to underestimation of the mol NO $flash^{-1}$.

Prior to applying this analysis to previous studies, we investigate the impact of the thermal decomposition of $CH_3O_2NO_2$ and HO_2NO_2 on near-field, in situ LNO_x measurements. We evaluate the maximum bias to NO_x from the thermal decomposition of these species during in situ sampling, using observations from the DC3 21 June 2012 flight. The chemical evolution of the near-field LNO_x emissions observed during this flight has been discussed in detail by Nault et al. (2016). For the UC Berkeley instrument and instruments with similar inlet design and exposure to warm cabin temperatures before measurement, at maximum, 5% of HO_2NO_2 would thermally decompose (Nault et al., 2015), leading to a positive bias of 3 parts per trillion by volume (pptv) in NO_2 and less than 1% bias in NO_x; thus, this compound is ignored for this discussion. In contrast, we assume full decomposition of $CH_3O_2NO_2$, as this molecule rapidly and completely dissociates to NO_2 and $CH_3O_2^{\cdot}$. Figure 1a shows that the thermal decomposition of $CH_3O_2NO_2$ would lead to an average ~ 7% positive bias in NO_x for the 21 June flight.

To demonstrate the impact of the shorter lifetime, we apply a post hoc correction to previous studies around the globe (Huntrieser et al., 2009; Schumann & Huntrieser, 2007). We assume that first-order kinetics (equation (1)) apply to the chemical conversion of lightning-emitted NO_x to higher oxides. We use this equation to extrapolate the moles of NO emitted at time $t = 0$, the time the lightning flashes were recorded, by the lightning flash from the measurement downwind at time t. The lifetime, τ, inferred from GEOS-Chem and observations by Nault et al. (2016) of ~3 h is used to solve for the initial value of emissions.

$$\text{mol NO flash}^{-1}(t) = \text{mol NO flash}^{-1}(0) \times e^{-t/\tau} \tag{1}$$

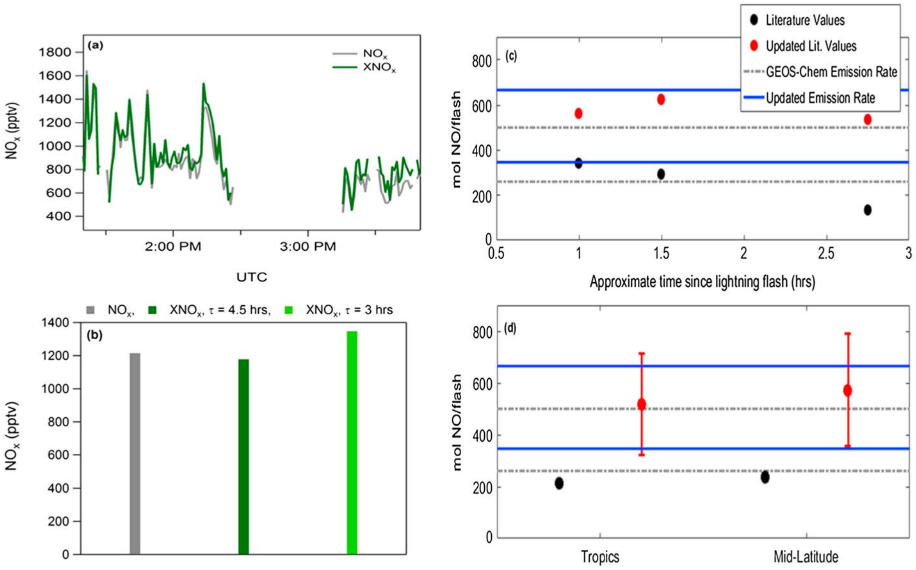

Figure 1. (a) Time series of NO$_x$ (grey) and XNO$_x$ (NO$_x$ + $CH_3O_2NO_2$, green) during DC3 21 June 2012 flight. (b) Average calculated NO$_x$ (grey) and XNO$_x$ (NO$_x$ + $CH_3O_2NO_2$, both green) at $t = 0$ at 1:35 PM using equation (1). The difference between dark green and light green is assuming $\tau = 4.5$ h (accounting for $CH_3O_2NO_2$ interference) and $\tau = 3$ h (not accounting for $CH_3O_2NO_2$ interference). (c) Mol NO flash^{-1} calculated by Huntrieser et al. (2009) (black) using an assumed lifetime of ~2 days. The mol NO flash^{-1} (red) corresponds to the measured values calculated using a 3 h UT NO$_x$ chemical lifetime. (d) The median tropical and midlatitude mol NO flash^{-1} (Huntrieser et al., 2009; Schumann & Huntrieser, 2007). The values calculated using an assumed ~ 2 day lifetime (black) and a 3 h chemical UT NO$_x$ lifetime (red), and the error bars represent the range of time between emission and measurements from prior studies used to calculate the median mol NO flash^{-1} (Huntrieser et al., 2009; Schumann & Huntrieser, 2007). Error bars for the uncorrected values in Figure 1d are not shown, as they are less than 5 mol NO flash^{-1} when assuming a 2 day lifetime. For both Figures 1c and 1d, the dash dotted lines mark the tropical (260 mol NO flash^{-1}) and midlatitude (500 mol NO flash^{-1}) values used in the standard GEOS-Chem model and the solid blue lines are the values we recommend from this study.

Applying equation (1) to the DC3 flight from 21 June 2012, the average NO$_x$ calculated for $t = 0$ is ~1200 pptv (Figure 1b). For other prior studies that measured in situ NO_2 along with NO, instead of calculating photostationary steady state NO_2, the apparent lifetime of NO$_x$ could increase from 3 h to ~4.5 h if decomposed $CH_3O_2NO_2$ is not subtracted after measuring NO_2. This increase in apparent NO$_x$ lifetime is due to measuring a NO$_x$ sink, $CH_3O_2NO_2$, as NO_2 and $CH_3O_2NO_2$ accounts for ~50% of the NO$_x$ loss during the 21st June flight. For this flight, if $CH_3O_2NO_2$ was measured as NO_2, but a 3 h lifetime is still assumed, the $t = 0$ NO$_x$ is ~1350 pptv, ~13% too high (Figure 1b). Assuming a 4.5 h lifetime instead, to account for the inclusion of $CH_3O_2NO_2$ in the measurement, gives a $t = 0$ NO$_x$ nearly identical to the case where only NO$_x$, and not $CH_3O_2NO_2$ + NO$_x$, is measured (Figure 1b). However, most studies, including the ones that are corrected post hoc here (Huntrieser et al., 2007, 2009, 2011; Schumann & Huntrieser, 2007), use NO measurements, which do not exhibit interference from $CH_3O_2NO_2$, and calculate the NO_2 mixing ratios; thus, we use the 3 h lifetime for the following analysis.

These prior studies, reviewed by Schumann and Huntrieser (2007), using a 2–8 day lifetime, report an average value of 250 mol NO flash^{-1} but with a significant range of values, including some as low as 20 mol flash^{-1}. If we reinterpret these studies' observations, using equation (1) with a 3 h chemical lifetime, the emission rate derived is approximately independent of time between lightning and the observation of the associated NO$_x$ (Figures 1c–1d) and is more consistent with methods using far-field observations to constrain models. Emission rates derived from this revised analysis are in the range of 510 and 550 mol NO flash^{-1}, for tropics and midlatitudes, respectively (Figure 1d and Table S4). This is approximately a factor of 2 increase in values normally determined for mean LNO$_x$ emission rates per flash determined in the near field of thunderstorms (Table S4) (Schumann & Huntrieser, 2007). This reduces the occurrence of very low mol NO flash^{-1} values, and thus the spread in global LNO$_x$ emission estimates, as the lower bound of 2 Tg N yr^{-1} is mainly derived from these near-field analyses. Given the spread and uncertainty in the time since LNO$_x$ emissions in the various

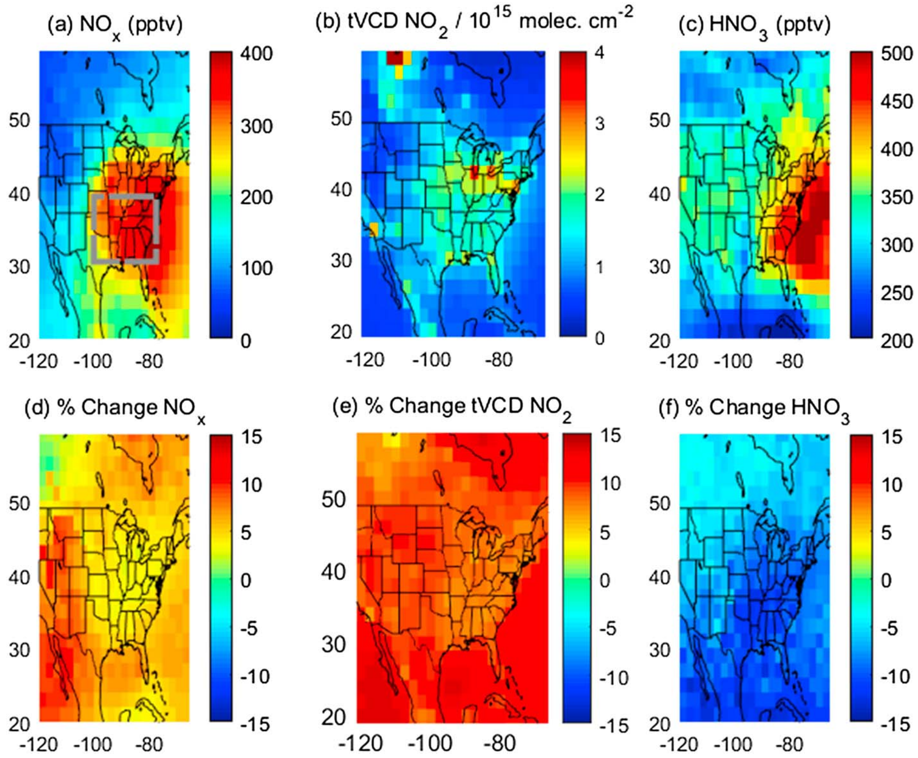

Figure 2. All results are from the simulations. Base case (a) UT NO$_x$, (b) tropospheric NO$_2$ column density (tVCD NO$_2$), and (c) UT HNO$_3$, where UT is defined as 200–350 hPa (defined in Table S1) between May and June 2012. Percent changes (updated chemistry - base)/updated chemistry (d) UT NO$_x$, (e) tVCD NO$_2$, and (f) UT HNO$_3$. The tVCD$_{NO2}$ is averaged between 12:00 and 14:00 local time, and the UT NO$_x$ and HNO$_3$ are averaged between 16:00 and 20:00 local time. Grey box in Figure 2a highlights the region with high LNO$_x$ emissions.

studies, as indicated by the uncertainty in Figure 1d, the emission rate may be even higher (as high as 800 mol NO flash^{-1}).

3.3. Effects of Updated Chemistry on Interpretation of Far-Field Studies

Prior studies (Allen et al., 2010, 2012; Fang et al., 2010; Hudman et al., 2007; Martin et al., 2007; Martini et al., 2011), focusing on far-field concentrations, have recognized that it is difficult to simultaneously match both UT NO$_x$ and HNO$_3$ observations. These studies chose an optimal value of 500 mol NO flash^{-1} for mean LNO$_x$ per flash (Table S4), which yields model concentrations at the lower end of the observed NO$_x$ and the upper end of the observed HNO$_3$ (Figure S5).

In the southeastern United States (Figure 2), the region of the model most strongly affected by lightning (Hudman et al., 2007), our revised model has 11% lower UT HNO$_3$ mixing ratios than the base case, while the UT NO$_x$ mixing ratios and tVCD$_{NO2}$ increase by 5% and 10%, respectively. With the revised chemistry (Figures 3 and S6), we find that increasing the mean LNO$_x$ emissions per flash by 33% to 665 mol NO flash^{-1} in the model leads to better agreement between the modeled and measured NO$_x$ and similar agreement to HNO$_3$ as in prior studies. We find that UT HNO$_3$ observations are now predicted to be 54% higher than the observations, while NO$_x$ is predicted to be within 10% of in situ.

Using this analysis in the southeast U.S., where we have in situ observations as a base for optimizing the model, we then compare to the global products available for OMI, the NASA SP v2 and the KNMI DOMINO product. Our model-satellite comparisons make use of averaging kernels (Boersma et al., 2016; Eskes & Boersma, 2003), so the comparisons are insensitive to the choice of a priori profiles in the retrieval. While a number of studies validating OMI tVCD$_{NO2}$ products using aircraft- or ground-based measurements or models have been done (Bucsela et al., 2008; Hains et al., 2010; Ialongo et al., 2016; Lamsal et al., 2010, 2014; Oetjen et al., 2013; Russell et al., 2011), they have mixed results on which product is more accurate, and no

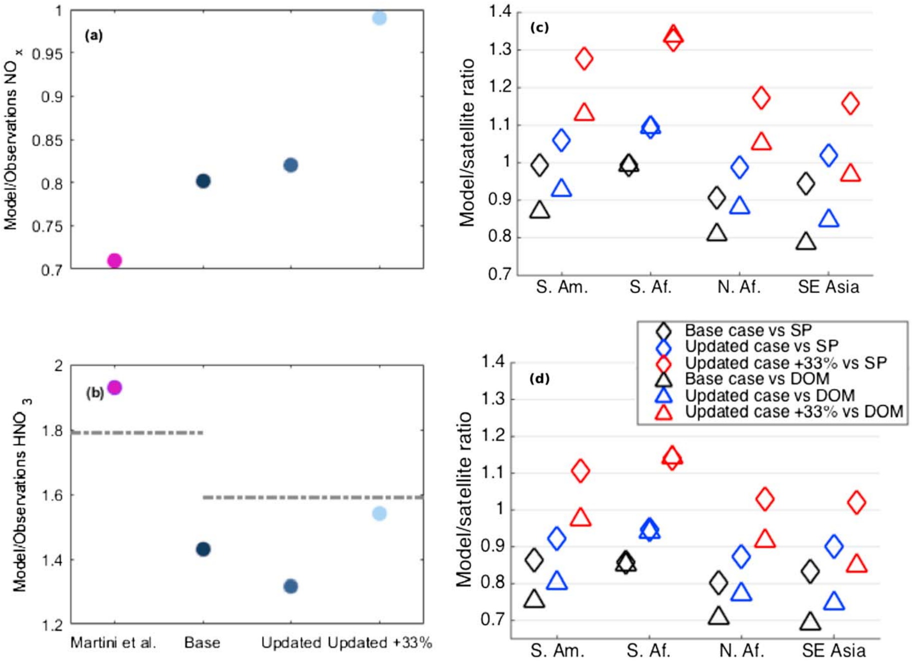

Figure 3. (a) Ratio of modeled and observed UT NO_x from Martini et al. (2011), base case, updated chemistry case, and updated chemistry with +33% LNO_x case (Table S1). (b) Ratio of modeled and observed UT HNO_3 from Martini et al. (2011), base case, updated case, and updated +33% case. Dash-dotted line represents the upper 1σ limit of observations in Martini et al. (2011) or during DC3. The NO_x and HNO_3 observations from Martini et al. (2011) have been corrected for thermal decomposition of $CH_3O_2NO_2$ and HO_2NO_2, as suggested by Bertram et al. (2007) and Browne et al. (2011). The comparison with observations for the base, updated, and updated +33% case is from DC3. (c) Comparison of tropospheric NO_2 column density versus base case (black), updated case (blue), and updated +33% case (red) for South America (S. Am.), Southern Africa (S. Af.), Northern Africa (N. Af), and Southeast Asia (SE Asia). Diamonds and triangles are the NASA SPv2 (Bucsela et al., 2013) (SP) and DOMINOv2 (Boersma et al., 2011b) (DOM) NO_2 retrievals, respectively. The regions and time periods are defined in Table S2. The different cases are defined in Table S1. (d) As in Figure 3c but with GEOS-Chem profiles adjusted to NO_2:NO_x ratios observed during DC3.

single study evaluates the versions of both the SP and DOMINO product used in this work; therefore, we use both products. The NASA SPv3 based on the revised slant columns described in Marchenko et al. (2015) was released after this analysis was completed. Krotkov et al. (2017) indicates that estimates of lightning NO_x production efficiency will not change significantly with the new product.

Figure 3c shows that, in a direct comparison, the updated chemistry improves model-satellite agreement in three of four regions, although the +33% emission increase worsens the agreement in all regions. Previous studies (e.g., Travis et al., 2016) have identified differences between modeled and observed NO_2:NO_x ratios in the UT. We therefore adjust the GEOS-Chem profiles to the average NO_2:NO_x ratio observed during DC3 (Figure S7) for Figure 3d; with this, the two retrievals bracket a model/satellite ratio of one in three of four regions. In general, the satellite data support a 33% increase in lightning NO_x emissions, especially in the Northern Hemisphere, though a smaller increase in the Southern Hemisphere may be indicated. This highlights the importance of further investigation into regionally specific lightning emissions. Assuming similar NO_2:NO_x ratios in all regions, satellite-model comparisons support an annual production of 8.3–9.0 Tg N yr^{-1} from lightning; the lower value assumes the increase only in the Northern Hemisphere, the upper one globally.

The prior results were for grid-scale chemistry, and as previously mentioned, AN chemistry is subgrid. We take this chemistry into consideration, here, post hoc. Nault et al. (2016) observed ~40 pptv of ANs produced ~2 h downwind from deep convection; GEOS-Chem captures ~10 pptv of this production. We apply a post hoc correction here by removing 30 pptv from the modeled NO_x and HNO_3 concentrations in pixels affected by lightning to represent possible repartitioning to ANs. We find that this post hoc correction, to the updated chemistry case (without adjusting the lightning emissions), decreases NO_x, resulting in concentrations that are ~30% lower than DC3 observations and HNO_3 that is within 20% of DC3 observations (Figure S6). Applying this correction to the updated chemistry case with increased lighting emissions, we find that NO_x is ~10% lower than observations, while HNO_3 is still within the 1σ spread of the observations. Thus, if this estimate of the effect of ANs chemistry is representative of regional phenomena, then an additional 5–10% increase in emissions to at least 700 mol NO $flash^{-1}$ would be required to yield modeled NO_x concentrations within 5% of DC3 observations.

4. Discussion

We identify that the UT NO_x lifetime near lightning is a key aspect contributing to current discrepancies between the near- and far-field studies of mean LNO_x emission rates per flash and a major contributor to the factor of 4 range in the emission rates that has been observed using different methods of data analysis (Schumann & Huntrieser, 2007). As demonstrated in Figure 1, once the UT NO_x lifetime is accounted for, two different approaches produce similar results for the mol NO $flash^{-1}$. After reevaluating the mean LNO_x emission rates per flash for the near-field studies and assuming 44 lightning flashes per second (Huntrieser et al., 2009), this would lead to ~9–10 Tg N yr^{-1}, which is similar to the total value calculated in the updated GEOS-Chem model run constrained by far-field studies (Table S4), assuming a global increase. This demonstrates that both the greater mol $flash^{-1}$ and the updated chemistry are necessary to optimize the agreement between models and observations. Additionally, this reduced spread in emission rates should greatly decrease the current differences among models and between models and measurements (Table S4).

Measurements of NO_y can also be utilized to circumvent the rapid UT NO_x chemistry to determine lightning N emission rate. In the near field of LNO_x emissions, NO_x is a large fraction of the total NO_y budget—up to 70–80% (Huntrieser et al., 2009, 2011). As NO_x rapidly oxidizes, total NO_y remains conserved, meaning that a measurement of total NO_y would provide a more useful constraint for lightning N emissions without needing prior knowledge or calculations of the NO_x chemistry and lifetime. For example, Nault et al. (2016) observed nearly complete closure between the rapid loss of NO_x (2.6×10^5 molecules/cm^3/s) and production of its higher oxide species (2.3×10^5 molecules/cm^3/s), demonstrating both the aforementioned rapid loss of NO_x in the first 3 h downwind from LNO_x emissions (as discussed throughout this paper) and the conservation of NO_y. Using the NO_y measurements in these two studies, we calculate 400 (Huntrieser et al., 2009) and 479 (Huntrieser et al., 2011) NO mol $flash^{-1}$. This would correspond to 7–9 Tg N yr^{-1} from lightning. This further indicates that the mean LNO_x emission rates per flash have been underpredicted and NO_y, when available, is a better measurement to calculate mean LNO_x emission rates per flash. However, measurement of total NO_y cannot be achieved from space-based platforms; therefore, accurate knowledge of the partitioning of NO_x to its higher oxides remains important for space-based near-field studies.

Another compound impacted by the increased LNO_x emission rates is UT O_3, an important greenhouse gas (Myre et al., 2013). The increased LNO_x emissions increase modeled UT O_3, which increases the differences between observed and modeled UT O_3 (Figure S8). Though investigating all the mechanisms that lead to this discrepancy between model and observations is beyond the scope of this paper, we speculate that the bias is due to subgrid effects of water vapor transport (Sauvage et al., 2007). The modeled water vapor mixing ratios are higher (by as much as 100%) than observed during DC3 (Figure S9). Since water is a key term in O_3 production, correcting this bias would likely improve the representation of UT O_3.

5. Conclusions

Utilizing the recent updates, including rapid $CH_3O_2NO_2$ and ANs production, in UT NO_x lifetime and fate improves our understanding of mean lightning NO_x (LNO_x) emission rates per flash. Incorporating these updates into a global chemical transport model, we find that the mean LNO_x emission rates per flash should increase by 23–33% to better match in situ measurements of NO_x and HNO_3 and satellite measurements of $tVCD_{NO2}$. The increase in inferred emissions stems from (a) the updated chemistry that decreases UT HNO_3, preventing the prior model overestimate of UT HNO_3 from limiting mean LNO_x emission rates per flash, and (b) from using a consistent, short NO_x lifetime instead of the one assumed in prior studies. This reduces the discrepancy of LNO_x among previous analyses.

Acknowledgments

Data used are available at https://doi:10.5067/Aircraft/DC3/DC8/Aerosol-TraceGas (DC3), https://mirador.gsfc.nasa.gov/ (NASA SP), http://www.temis.nl/airpollution/no2.html (DOMINO), and https://doi.org/10.6078/D10P4P (GEOS-Chem). Relevant analysis code is hosted at https://doi.org/10.5281/zenodo.801885. B. A. N. was supported by the National Science Foundation Graduate Research Fellowship under grant DGE 1106400. J. L. L. was supported by the NASA ESS Fellowship NNX14AK89H. B. A. N., P. J. W., and R. C. C. acknowledge funding support from NASA (NNX12AB79G). J. D. C. and P. O. W. acknowledge funding support from NASA (NNX12AC06G and NNX14AP46G). J. D. and E. S. acknowledge funding from NASA (NNX12AB80G). This research used the Savio computational cluster resource provided by the Berkeley Research Computing program at the University of California, Berkeley (supported by the UC Berkeley Chancellor, Vice Chancellor of Research, and Office of the CIO). The authors also want to thank the ground and flight crews of the DC-8 and the DC3 science team.

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