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Estimates of Lightning NO_x Production based on High Resolution OMI NO_2 Retrievals over the Continental US

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Abstract. Lightning serves as the dominant source of nitrogen oxides ($NO_x = NO + NO_2$) in the upper troposphere (UT), with strong impact on ozone chemistry and the hydroxyl radical production. However, the production efficiency (PE) of lightning nitrogen oxides (LNO_x) is still quite uncertain (32 – 1100 mol NO per flash). Satellites measurements are a powerful tool to estimate LNO_x directly as compared to conventional platforms. To apply satellite data in both clean and polluted regions, a new algorithm for calculating LNO_x has been developed based on the program of new Berkeley High Resolution (BEHR) v3.0B NO_2 product and the Weather Research and Forecasting-Chemistry (WRF-Chem) model. LNO_x PE over the continental US is estimated using the NO_2 product of the Ozone Monitoring Instrument (OMI) satellite and the Earth Networks Total Lightning Network (ENTLN) data. Focusing on the summer season during 2014, we find that the lightning NO_2 (LNO_2) PE is 44 ± 16 mol NO_2 flash^{-1} and 8 ± 3 mol NO_2 stroke^{-1} while LNO_x PE is 120 ± 52 mol NO_x flash^{-1} and 22 ± 9 mol NO_x stroke^{-1}. Results reveal that former methods are more sensitive to background NO_2 and neglect much of the below-cloud LNO_2. As the LNO_x parameterization varies in studies, the sensitivity of our calculations to the setting of the amount of lightning NO (LNO) is evaluated. Careful consideration of the ratio of LNO_2 to NO_2 is also needed, given its large influence on the estimation of LNO_2 PE.

1 Introduction

Nitrogen oxides (NO_x) near the Earth's surface is mainly produced by soil, biomass burning and fossil fuel combustion, while NO_x in the middle and upper troposphere originate largely from lightning and aircraft emissions. NO_x plays an important role in the production of ozone (O_3) and the hydroxyl radical (OH). While the anthropogenic sources of NO_x are largely known, lightning nitrogen oxides (LNO_x) are still the source with the greatest uncertainty, though they are estimated to range between 2 and 8 Tg N yr^{-1} (Schumann and Huntrieser, 2007). LNO_x is produced in the upper troposphere (UT) by O_2 and N_2 dissociation in the hot lightning channel as described by the Zel'dovich mechanism (Zel'dovich and Raizer, 1967). With the recent updates

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of UT NO_x chemistry, the day time lifetime of UT NO_x is evaluated to be $\sim$ 3 h near thunderstorms and $\sim$ 0.5 – 1.5 days away from thunderstorms (Nault et al., 2016, 2017). This results in enhanced O_3 production in the cloud outflow of active convection (Pickering et al., 1996; Hauglustaine et al., 2001; DeCaria et al., 2005; Ott et al., 2007; Dobber et al., 2008; Allen et al., 2010; Finney et al., 2016). As O_3 is known as a greenhouse gas, strong oxidant and absorber of ultraviolet radiation (Myhre et al., 2013), the contributions of LNO_x to O_3 production also have an effect on climate forcing. Finney et al. (2018) found different impacts on atmospheric composition and radiative forcing when simulating future lightning using a new upward cloud ice flux (IFLUX) method and the commonly used the widely used cloud-top height (CTH) approach. As lightning with the CTH approach have reported 5 — 16% increases over the next century (Clark et al., 2017; Banerjee et al., 2014; Krause et al., 2014), a 15% decrease was estimated with IFLUX in 2100 under a strong global warming scenario (Finney et al., 2018). As a result of the different effects on compositions, a net positive radiative forcing was found with the CTH approach while there is little net radiative forcing with the IFLUX approach.

In the view of the region dependent lifetime of NO_x and the difficulty of measuring LNO_x directly, a better understanding of the LNO_x production is required, especially in the tropical and mid-latitude regions in summer. Using its distinct spectral absorption lines in the near-ultraviolet (UV) and visible (VIS) range (Platt and Perner, 1983), NO_2 can be measured by satellite instruments like the Global Ozone Monitoring Experiment (GOME; Burrows et al., 1999; Richter et al., 2005), Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY; Bovensmann et al., 1999), the Second Global Ozone Monitoring Experiment (GOME-2; Callies et al., 2000) and the Ozone Monitoring Instrument (OMI; Levelt et al., 2006). OMI has the highest spatial resolution, least instrument degradation and longest record among these satellites (Krotkov et al., 2017). Satellites measurements of NO_2 are a powerful tool compared to conventional platforms, because of its global coverage, constant instrument features and temporal continuity.

Recent studies have determined and qualified LNO_x using satellite observations. Beirle et al. (2004) constrained the LNO_x production to 2.8 (0.8 – 14) Tg N yr^{-1} by combining GOME NO_2 data and flash counts from the Lightning Imaging Sensor (LIS) aboard the Tropical Rainfall Measurement Mission (TRMM) over Australia. Boersma et al. (2005) estimated the global LNO_x production of 1.1 – 6.4 Tg N yr^{-1} by comparing GOME NO_2 with distributions of LNO_2 modeled by Tracer Model 3 (TM3). Martin et al. (2007) analyzed SCIAMACHY NO_2 columns with Goddard Earth Observing System chemistry model (GEOS-Chem) simulations to identify LNO_x production amounting to 6 $\pm$ 2 Tg N yr^{-1}.

As these methods focus on monthly or yearly mean NO_2 column densities, more recent studies applied specific approaches to investigate LNO_x directly over active convections. Beirle et al. (2006) estimated LNO_x as 1.7 (0.6 – 4.7) Tg N yr^{-1} based on a convective system over the Gulf of Mexico, using National Lightning Detection Network (NLDN) observations and GOME NO_2 column densities. However, it is assumed that all the enhanced NO_2 originated from lightning without the contribution of anthropogenic emissions. Beirle et al. (2010) analyzed LNO_x production systematically using the global dataset of SCIAMACHY NO_2 observations combined with flash data from the World Wide Lightning Location Network (WWLLN). The threshold of high flash rates is that the summation of the corrected flashes within the satellite pixel (30×60 km^2) in the last one hour must be greater than 1 flashes/km^2 /h. But the results of LNO_x production are highly variable and correlations between flash rate densities and LNO_x production are low in some cases. Bucsela et al. (2010) estimate LNO_x production as $\sim$ 100 –

250 mol NO_x/flash for four cases, using the DC-8 and OMI data during NASA's Tropical Composition, Cloud and Climate Coupling Experiment (TC^4).

Based on the approach used by Bucsela et al. (2010), a special algorithm was developed by Pickering et al. (2016) to retrieve LNO_x from OMI and the WWLLN. The algorithm takes the OMI tropospheric slant column density (SCD) of NO_2 (S_{NO_2}) as the tropospheric slant column density of LNO_2 (S_{LNO_2}) by using cloud radiance fraction (CRF) greater than 0.9 to minimize or screen the lower tropospheric background. To convert the S_{LNO_2} to the tropospheric vertical density (VCD) of LNO_x (V_{LNO_x}), an air mass factor (AMF) is calculated by dividing the a priori S_{LNO_2} by the a priori V_{LNO_x}. Since they assumed NO_2 above the cloud are all LNO_2, their AMF and derived VCD of LNO_x (LNO_2) is named as $AMF_{LNO_x Clean}$ and LNO_xClean (LNO_2Clean), respectively. Unless otherwise specified, abbreviations S and V are respectively defined as the tropospheric SCD and VCD in this paper. The a priori S_{LNO_2} is calculated using a radiative transfer model and a profile of LNO_2 simulated by the NASA Global Modeling Initiative (GMI) chemical transport model. The a priori V_{LNO_x} is also obtained from the GMI model. Results for the Gulf of Mexico during 2007 – 2011 summer yield LNO_x production of 80 $\pm$ 45 mol NO_x per flash. Among several substantial sources of uncertainty, significant uncertainty (3% $\sim$ >30%) exists in characterizing background NO_x in this region (Pickering et al., 2016).

More recently Bucsela et al. (2019) obtained an average production efficiency (PE) of 180 $\pm$ 100 mol per flash over East Asia, Europe and North America based on the method used in Pickering et al. (2016). The tropospheric NO_x background was removed by the weighted temporal average of NO_x at each box which meets the optical cloud pressure and CRF criteria but has 0 — 1 flashes instead. The lofted pollution was corrected by 15% according to the estimation from DeCaria et al. (2000, 2005) and the average chemical delay was adjusted by 15% following the 3-hour LNO_x lifetime in the nearby field of convection (Nault et al., 2017). However, there were negative LNO_x values caused by the overestimation of the tropospheric background at those locations.

On another hand, Lapierre et al. (2019) constrained LNO_2 to 1.1 $\pm$ 0.6 mol NO_2/stroke for intracloud (IC) strokes and 10.0 $\pm$ 4.9 mol NO_2/stroke for cloud-to-ground (CG) strokes over the continental US (CONUS). LNO_2 per stroke was scaled to 54.4 mol NO_x/flash by strokes per flash and the ratio of NO to NO_2 in the UT. They used the regridded Berkeley High-Resolution (BEHR) V3.0A 0.05° $\times$ 0.05° "visible only" NO_2 VCD (V_{vis}) product which includes two parts of NO_2 can be "seen" by the satellite. The first part is the NO_2 above clouds and the second part is the NO_2 detected from cloud free areas. A threshold of 3 $\times$ 10^{15} molecules cm^{-2}, the typical urban NO_2 concentration, was applied to mask the contaminated grid cells (Beirle et al., 2010; Laughner and Cohen, 2017). The main difference between Lapierre et al. (2019) and Pickering et al. (2016) is the air mass factor for lightning (AMF_{LNO_x}) implemented in the basic algorithm. In Lapierre et al. (2019), the air mass factor was used to convert S_{NO_2} to V_{vis}, while in Pickering et al. (2016) it was used to convert S_{LNO_2} to V_{LNO_x}, assuming that all S_{NO_2} is generated by lightning.

To apply the approach used by Bucsela et al. (2010), Pickering et al. (2016), Bucsela et al. (2019) and Lapierre et al. (2019) without geographic restrictions, contamination of anthropogenic emissions must be taken into account in detail. The Weather Research and Forecasting (WRF) model coupled with chemistry (WRF-Chem) has been employed to evaluate the convective transport and chemistry in many studies (Barth et al., 2012; Wong et al., 2013; Fried et al., 2016; Li et al., 2017). Meanwhile,

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Laughner and Cohen (2017) showed that the OMI AMF is increased by ∼35% for summertime when LNO_2 simulated by WRF-Chem is included in the a priori profiles to match aircraft observations. The simulation agrees with observed NO_2 profiles and the bias of AMF is reduced to $< \pm 4\%$ for OMI viewing geometries.

In this paper, we focused on the estimation of LNO_2 production per flash (LNO_2/flash), LNO_x production per flash (LNO_x/flash), LNO_2 production per stroke (LNO_2/stroke) and LNO_x production per stroke (LNO_x/stroke) in May–August (MJJA) 2014 by developing an algorithm similar to Pickering et al. (2016) based on the BEHR NO_2 retrieval algorithm (Laughner et al., 2018a, b), but it performs better over background NO_2 sources. Section 2 describes the satellite, lightning data, model settings and the algorithm in detail. Section 3 explores the suitable data criteria, compares different methods and evaluates the effect of cloud and LNO_x parameterization on LNO_x production estimation. Section 4 examines different sources of the uncertainty of the results. Conclusions are summarized in Section 5.

2 Data and Methods

2.1 Ozone Monitoring Instrument (OMI)

OMI is carried on the Aura satellite (launched in 2004), a member of A-train satellite group (Levelt et al., 2006, 2018). OMI passes over the equator at ∼ 13:45 LT (ascending node) and has a swath width of 2600 km, with a nadir field-of-view resolution of 13×24 km^2. Since the beginning of 2007, some of the measurements have become useless as a result of anomalous radiances called the "row anomaly" (Dobber et al., 2008). For the current study, we used the NASA standard product v3 (Krotkov et al., 2017) as input to the LNO_x retrieval algorithm.

The main steps of calculating the NO_2 tropospheric VCD (V_{NO_2}) in the NASA product include:

1. SCDs are determined by the OMI-optimized differential optical absorption spectroscopy (DOAS) spectral fit;

2. A corrected ("de-striped") SCD is obtained by subtracting the bias from the measured slant column;

3. The AMF for stratospheric (AMF_{strat}) or tropospheric column (AMF_{trop}) is calculated from the NO_2 profile integrated vertically using weighted scattering weights with the a priori profiles. These profiles are obtained from GMI monthly mean profiles using four years (2004 – 2007) simulation;

4. The stratospheric NO_2 VCD (V_{strat}) is calculated from the subtraction of a priori contribution from tropospheric NO_2 and a three-step (interpolation, filtering, and smoothing) algorithm (Bucsela et al., 2013);

5. V_{strat} is converted to the slant column using AMF_{strat} and subtracted from the measured SCDs to yield S_{NO_2}, leading to $V_{NO_2} = S_{NO_2}/AMF_{trop}$.

Based on this method, we developed a new AMF_{LNO_x} to obtain the desired V_{LNO_x} ($V_{LNO_x} = S_{NO_2}/AMF_{LNO_x}$) to replace the original step 5. Details of this algorithm are discussed in section 2.4.

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2.2 The Earth Networks Total Lightning Detection Network (ENTLN)

The Earth Networks Total Lightning Network (ENTLN) operates a system of over 1500 ground-based stations around the world with more than 900 sensors installed in the CONUS (Zhu et al., 2017). Both IC and CG lightning flashes are located by the sensors with detection frequency ranging from 1 Hz to 12 MHz based on the electric field pulse polarity and wave shapes. Groups of pulses are classified as a flash if they are within 700 ms and 10 km. In the preprocessed data obtained from the ENTLN, both strokes and lightning flashes composed of one or more strokes are included.

Rudlosky (2015) compared ENTLN with LIS and found that the relative flash detection efficiency of ENTLN increases from 21.6% during 2011 to 31.4% during 2013. Lapierre et al. (2019) also compared combined ENTLN and the NLDN dataset with data from the LIS and the detection efficiencies of IC flashes and strokes are 88% and 45%, respectively. Since we use the ENTLN data in 2014 as Lapierre et al. (2019) and NLDN detection efficiency of IC pulses should be lower than 33% which is calculated by the data in 2016 (Zhu et al., 2016), only the IC flashes and strokes are corrected by 88% and 45%, respectively, while CG flashes and strokes are unchanged because of the high detection efficiency.

2.3 Model Description

The present study uses WRF-Chem version 3.5.1 (Grell et al., 2005) with a horizontal grid size of 12 $\times$ 12 km and 29 vertical levels (Fig. 1). The initial and boundary conditions of meteorological parameters are provided by the North American Regional Reanalysis (NARR) dataset with a 3 hourly time resolution. Based on Laughner et al. (2018b), 3D wind fields, temperature and water vapor are nudged towards the NARR data. Outputs from the version 4 of Model for Ozone and Related chemical Tracers (MOZART-4; Emmons et al., 2010) was used to generate the initial and boundary conditions of chemical species. Anthropogenic emissions are driven by the 2011 National Emissions Inventory (NEI), scaled to model years by the Environmental Protection Agency annual total emissions (EPA and OAR, 2015). The Model of Emissions of Gases and Aerosol from Nature (MEGAN; Guenther et al., 2006) is used for biogenic emissions. The chemical mechanism is the version 2 of Regional Atmospheric Chemistry Mechanism (RACM2; Goliff et al., 2013) with updates from Browne et al. (2014) and Schwantes et al. (2015). In addition, lightning flash rate and LNO_x parameterizations are activated (200 mol NO flash^{-1}, the factor to adjust the predicted number of flashes is set to 1; hereinafter referred to as "1$\times$200 mol NO flash^{-1}"). The bimodal profile modified from the standard Ott et al. (2010) profile (Laughner and Cohen, 2017) is employed as the vertical distribution of lightning NO (LNO) in WRF-Chem, while LNO and LNO_2 profiles are defined as the difference of vertical profiles between simulations with and without lightning.

2.4 Method for Deriving AMF

The V_{LNO_x} near convection is calculated according:

$$V_{\mathrm{LNO}_x} = \frac{S_{\mathrm{NO}_2}}{AMF_{\mathrm{LNO}_x}} \tag{1}$$

where $S_{\mathrm{NO_2}}$ is the OMI-measured tropospheric slant column NO_2 and $\mathrm{AMF}_{\mathrm{LNO}_x}$ is a customized lightning air mass factor. $\mathrm{AMF}_{\mathrm{LNO}_x}$ is defined as the ratio of the "visible" modeled NO_2 slant column to the total modeled tropospheric LNO_x vertical column (derived from the a priori NO and NO_2 profiles, scattering weights, and radiance cloud fraction):

$$AMF_{\mathrm{LNO}_x} = \frac{(1-f_r)\int_{p_{surf}}^{p_{tp}} w_{clear}(p)NO_2(p)\,dp + f_r\int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p)NO_2(p)\,dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p)\,dp} \tag{2}$$

where f_{r} is the radiance cloud fraction, p_{surf} is the surface pressure, p_{tp} is the tropopause pressure, p_{cloud} is the cloud optical pressure (CP), w_{clear} and w_{cloudy} are respectively the pressure dependent scattering weights from the TOMRAD lookup table (Bucsela et al., 2013) for clear and cloudy parts, and $NO_2(p)$ is the modeled NO_2 vertical profile. Details of these standard parameters and calculation methods are given in Laughner et al. (2018a). $LNO_x(p)$ is the LNO_x vertical profile calculated by the difference of vertical profiles between WRF-Chem simulations with and without lightning.

To compare our results with those of Pickering et al. (2016) and Lapierre et al. (2019), we calculate their $\mathrm{AMF}_{\mathrm{LNO}_x\mathrm{Clean}}$ and $\mathrm{AMF}_{\mathrm{NO_2Vis}}$ respectively:

$$AMF_{\mathrm{LNO}_x\mathrm{Clean}} = \frac{(1-f_r)\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} w_{\mathrm{clear}}(p)LNO_2(p)\,dp + f_r\int_{p_{\mathrm{cloud}}}^{p_{\mathrm{tp}}} w_{\mathrm{cloudy}}(p)LNO_2(p)\,dp}{\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} LNO_x(p)\,dp} \tag{3}$$

$$AMF_{\mathrm{NO_2Vis}} = \frac{(1-f_r)\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} w_{\mathrm{clear}}(p)NO_2(p)\,dp + f_r\int_{p_{\mathrm{cloud}}}^{p_{\mathrm{tp}}} w_{\mathrm{cloudy}}(p)NO_2(p)\,dp}{(1-f_g)\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} NO_2(p)\,dp + f_g\int_{p_{\mathrm{cloud}}}^{p_{\mathrm{tp}}} NO_2(p)\,dp} \tag{4}$$

where f_g is the geometric cloud fraction and $LNO_2(p)$ is the modeled LNO_2 vertical profile. Besides these AMFs, another AMF called $\mathrm{AMF}_{\mathrm{LNO_2Vis}}$ is developed for comparison later. A full list of definitions of the used AMFs is shown in Appendix A.

$$AMF_{\mathrm{LNO_2Vis}} = \frac{(1-f_r)\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} w_{\mathrm{clear}}(p)NO_2(p)\,dp + f_r\int_{p_{\mathrm{cloud}}}^{p_{\mathrm{tp}}} w_{\mathrm{cloudy}}(p)NO_2(p)\,dp}{(1-f_g)\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} LNO_2(p)\,dp + f_g\int_{p_{\mathrm{cloud}}}^{p_{\mathrm{tp}}} LNO_2(p)\,dp} \tag{5}$$

Additionally, Vasilkov et al. (2008) found that p_{cloud}, retrieved by the OMI O_2-O_2 algorithm (Bucsela et al., 2013), is often significantly larger than the IR-derived cloud top. This means that the back-scattered UV-VIS light observed by OMI penetrates deeper into the cloud. As a result, part of the NO_2 originated from lightning can be detected by the OMI satellite. As discussed in Pickering et al. (2016), the ratio of $V_{\mathrm{LNO_2}}$ seen by OMI to V_{LNO_x} is partly influenced by p_{cloud}. The effects of LNO_2 below the cloud will be discussed in section 3.3.

2.5 Procedures for Deriving LNO_x

LNO_x is re-gridded to 0.05° × 0.05° pixels like the BEHR product and is analyzed in 1° × 1° grid boxes with a minimum of fifty valid pixels which is equivalent to five satellite pixels in Pickering et al. (2016). The main procedures used to derive LNO_x are as follows:

CRFs (CRFs ≥ 70%, CRFs ≥ 90% and CRFs = 100%) and CP ≤ 650 hPa are various criteria of deep convective clouds for OMI pixels (Ziemke et al., 2009; Choi et al., 2014; Pickering et al., 2016). The effect of different CRFs on the retrieved

LNO_x is explored in section 3.2. Furthermore, another criterion of cloud fractions (CFs) is applied to the WRF-Chem results for the successful simulation of convections. The CFs are defined as the maximum cloud fraction calculated by the Xu-Randall method between 350 and 400 hPa (Xu and Randall, 1996). This atmospheric layer (between 350 and 400 hPa) avoids any biases in the simulation of high clouds. We choose CFs $\geq$ 40% suggested by Strode et al. (2017) to determine cloudy or clear for each simulation grid.

Besides properties of cloud, the time period and sufficient flashes (or strokes) are required for fresh LNO_x detected by OMI. The time window (t_{window}) is the hours prior to the OMI overpass time. t_{window} is limited to 2.4 h by the mean wind speed at pressure levels 500 – 100 hPa during OMI overpass time and the square root of the 1° × 1° box over the CONUS (Lapierre et al., 2019). Meanwhile, 2400 flashes box^{-1} and 8160 strokes box^{-1} are chosen as sufficient for detecting LNO_x (Lapierre et al., 2019).

To ensure that lightning flashes are simulated successfully by WRF-Chem, the threshold of simulated total lightning flashes (TL) per box is set to 1000, which is fewer than that used by the ENTLN lightning observation, considering the uncertainty of lightning parameterization. In view of other NO_2 sources except LNO_2, the ratio of modeled lightning NO_2 above cloud (LNO_2Vis) to modeled NO_2 above cloud (NO_2Vis) is defined to check whether enough LNO_2 can be detected by OMI. The ratio $\geq$ 50% indicates that LNO_2 is not polluted much above the cloud.

Finally, the NO_2 lifetime against oxidation should be taken into account. As estimated by Nault et al. (2016), the lifetime (τ) of NO_2 in the near field of convections is $\sim$ 3 h. The initial value of NO_2 is solved by Eq. 6 as

$$NO_2(0) = NO_2(OMI) \times e^{0.5t/\tau} \tag{6}$$

where $NO_2(0)$ is the moles of NO_2 emitted at time t = 0, $NO_2(OMI)$ is the moles of NO_2 measured at the OMI overpass time and $0.5t$ is the half cross grid time which is 1.2 h, assuming that lightning occurred at the center of each 1° × 1° box. For each grid box, the mean LNO_x vertical column is obtained by averaging V_{LNO_x} values from all regridded 0.05° × 0.05° pixels in the box. This mean value is converted to moles LNO_x using the dimensions of the grid box. Two methods are applied to estimate the seasonal mean LNO_2/flash, LNO_x/flash, LNO_2/stroke and LNO_x/stroke:

(1) summation method: dividing the sum of LNO_x by the sum of flashes (or strokes) in each 1° × 1° box in MJJA 2014;

(2) linear regression method: applying the linear regression to daily summations of LNO_x and flashes (or strokes).

3 Results

3.1 Criteria Determination

To determine the suitable criteria from conditions defined in section 2.5, six different combinations are defined (Table 1) and applied to the original data with a linear regression method (Fig. 2).

A daily search of NO_2 product for coincident ENTLN flash (stroke) data results in 99 (102) valid days under the condition of CRF $\geq$ 90%. Taking the flashes type ENTLN data as an example, the number of valid days decreases from 99 to 81 under the basic condition coupled with TL $\geq$ 1000 and ratio $\geq$ 50%, while LNO_x/flash increases from 86.0 ± 14.0 mol/flash to 114.8

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± 18.2 mol/flash. The result is almost the same as the one without ratio ≥ 50%. Although this indicates the criterion of TL works well, it is better to include the ratio in case of some exceptions in the different AMF methods. Since CF ≥ 40% leads to a sharp loss of valid numbers and production, therefore, it is not a suitable criterion. Instead the CRF criteria are used. Finally, coincident ENTLN data, TL ≥ 1000 and ratio ≥ 50% are chosen as the thresholds to explore the effects of three different CRF conditions (CRF ≥ 70%, CRF ≥ 90% and CRF = 100%) on LNO_x production (Table 2).

Apart from the fewer valid days under higher CRF conditions (CRF ≥ 90% and CRF = 100%), LNO_x/flash increases from 109.0 ± 15.3 mol/flash to 114.8 ± 18.2 mol/flash and decreases again to 99.4 ± 15.3 mol/flash while LNO_x/stroke enhances from 16.7 ± 2.6 mol/stroke to 17.8 ± 2.9 mol/stroke and drops again to 15.6 ± 3.1 mol/stroke (Table 2). Although enhanced NO_x is often observed in regions with CRF > 70% (Pickering et al., 2016), the following analysis will be based on the criterion of CRF ≥ 90% considering the contamination by low and mid-level NO_2 and comparisons with former studies.

3.2 Comparison of LNO_x Production based on Different AMFs

Lapierre et al. (2019) derived LNO_2 production based on the BEHR NO_2 product. In order for our results to be comparable with those of Pickering et al. (2016) and Lapierre et al. (2019), we choose NO_2 instead of NO_x to derive productions. In Fig. 3, time series of NO_2Vis, LNO_2Vis, LNO_2 and LNO_2Clean production per day over CONUS are plotted for MJJA 2014 with the criterion of CRF ≥ 90% and a flash threshold of 2400 flashes per 2.4 h. LNO_2 productions are mostly in the range from 20 to 80 mol/flash. LNO_2Vis productions are smaller than LNO_2 productions which contain LNO_2 below clouds. The simulation of GMI in Pickering et al. (2016) indicated that 25% – 30% of the LNO_x column lies below the CP, while the ratio in our WRF-Chem simulation is 10% – 80%. The effect of clouds properties on LNO_x production will be discussed in more detail in section 3.3. Generally, the order of estimated daily production efficiencies (PEs) is LNO_2Clean > LNO_2 > NO_2Vis > LNO_2Vis. The percent difference in the estimated PE (ΔPE) between NO_2Vis and LNO_2Vis indicates a certain amount of background NO_2 exists above clouds. Overall, the tendency of that ΔPE is consistent with another ΔPE between NO_2Vis and LNO_2Clean. When the region is highly polluted (ΔPE between NO_2Vis and LNO_2 is larger than 200%), PEs based on NO_2Vis and LNO_2Clean are significantly overestimated. In other words, NO_2Vis and LNO_2Clean are more sensitive to background NO_2. The extent of the overestimation of NO_2Vis is larger than that of LNO_2Clean in highly polluted regions, while it is usually opposite in most regions.

Figure 4 shows the linear regression for ENTLN data versus NO_2Vis, LNO_2Vis, LNO_2 and LNO_2Clean with the same criteria as shown in Fig. 3. LNO_2Clean production (the largest slope) is 49.1 ± 8.4 mol NO_2/flash with a correlation of 0.79 and 7.5 ± 1.3 mol NO_2/stroke with a correlation of 0.77. As shown in Fig. 3, the number of positive percent differences between NO_2Vis and LNO_2Clean production is much fewer than that of negative differences. As a result, NO_2Vis production (19.3 ± 2.7 mol NO_2/flash and 3.6 ± 0.5 mol NO_2/stroke) is smaller than LNO_2Clean production using the linear regression method.

If the CP ≤ 650 hPa, TL ≥ 1000 and ratio ≥ 50% are removed from criteria, our result based on NO_2Vis (3.8 ± 0.5 mol/stroke) is still larger than the value of 1.6 ± 0.1 mol/stroke mentioned in Lapierre et al. (2019). This may be caused by the different version of BEHR algorithm, as Lapierre et al. (2019) used BEHR V3.0A and our algorithm is based on BEHR V3.0B

(Laughner et al., 2019). The input of S_{NO_2} in both versions is from the NASA standard product V3 and the major improvements of BEHR V3.0B are listed below:

1. The profile (V3.0B) closest to the OMI overpass time was selected instead of the last profile (V3.0A) before the OMI overpass.

2. The AMF uses a variable tropopause height as opposed to the fixed 200 hPa tropopause.

3. The surface pressure is now calculated according to Zhou et al. (2009).

The detailed log of changes is available at https://github.com/CohenBerkeleyLab/BEHR-core (last access: November 26, 2019). Note that Lapierre et al. (2019) used the monthly NO_2 profile, while the daily profile is used in our study and the interval of our outputs from WRF-Chem is 30 min which is more frequent than 1 h in the BEHR daily product, the AMF could be affected by different NO_2 profiles. In view of these factors, we compare different methods based on our data to minimize these effects.

Meanwhile, LNO_2 production (41.6 ± 6.9 mol/flash and 6.3 ± 1.1 mol/stroke) is between LNO_2Clean production and NO_2Vis production, which coincides with the daily results in Fig. 3. Furthermore, the calculated LNO_x production is 114.8 ± 18.2 mol/flash (or 17.8 ± 2.9 mol/stroke) which is larger than 91 mol/flash from the linear regression result of Pickering et al. (2016), possibly due to the differences in geographic location, lightning data, chemistry model and the ratio of CG to IC considered by Pickering et al. (2016) and this study.

The mean and standard deviation of LNO_2 production under CRF $\geq$ 90% using the summation method is 46.2 ± 35.1 mol/flash and 9.9 ± 8.1 mol/stroke, while LNO_x production is 125.6 ± 95.9 mol/flash and 26.7 ± 21.6 mol/stroke (Fig. 5). The LNO_2 and LNO_x production are both higher in the South Central U.S. (88°W – 103°W, 28.5°N – 39°N) and Southeast U.S. (79°W – 85°W, 25°N – 35°N), consistent with Lapierre et al. (2019) and Bucsela et al. (2019). Compared with Fig. 3, Fig. 6 (a) and (b) present some large differences between NO_2Vis production and LNO_2Vis production, which are consistent with what we expect for polluted regions. Meanwhile, the differences between LNO_2 production and NO_2Vis production depend on background NO_2, the strength of updraft and the profile. The negative differences are caused by background NO_2 carried by the updraft while parts of the below-cloud LNO_2 results in more LNO_2 production than NO_2Vis production (Fig. 6 c). Figure 6 (d) shows that the ratio of LNO_2Vis to LNO_2 ranges from 10% – 80%. This may be caused by the height of the clouds and the profile of LNO_2. If the CP is near 300 hPa, the ratio should be smaller because of the coverage of clouds. The ratio would present the same trend while the peaks of LNO_2 profile is below the CP. Therefore, a better understanding of LNO_2 and LNO_x below clouds is required.

3.3 Effects of Cloud and LNO_x Parameterization on LNO_x Production

Figure 7 presents the daily distribution of CP and the ratio of LNO_2Vis to LNO_2 during MJJA 2014 with the criteria defined in section 3.1 under CRF $\geq$ 90%. Since the ratio of LNO_2Vis to LNO_2 decreases from 0.8 to 0.2 while the cloud is higher (smaller pressure value), NO_2Vis production is smaller than LNO_2 in relatively clean areas as shown in Fig. 4. Apart from LNO_2Vis, the LNO_2 production is also affected by CP. For LNO_2 production larger than 30 mol/stroke, the CPs are all smaller than 550 hPa (Fig. 8). However, smaller LNO_2 productions (< 30 mol/stroke) occur on all levels between 650 hPa and 200 hPa. Because

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of the limited amount of large LNO_2 production and lightning data, we can not derive that higher LNO_2 production relates to higher clouds or different lightning properties at this stage. Because CP only represents the development of clouds, the vertical structure of flashes can not be derived from the CP values only. As discussed in several previous studies, flash channel length varies and depends on the environmental conditions (Carey et al., 2016; Mecikalski and Carey, 2017; Fuchs and Rutledge, 2018). Davis et al. (2019) compared two kinds of flashes: normal flashes and anomalous flashes. Because updrafts are stronger and flash rates are larger in anomalous storms, UT LNO_x production is larger than normal polarity storms. In general, normal flashes are coupled with an upper-level positive charge region and a mid-level negative charge region, while anomalous flashes are opposite (Williams, 1989). It is not straightforward to estimate the error resulting from the vertical distribution of LNO_x. There are mainly two methods of distributing LNO_x in models: LNO_x profiles (postconvection) are simulated after LNO_x is redistributed by convective transport, while the other one (preconvection) uses LNO_x profiles made before the redistribution of convective transport (Luo et al., 2017). However, given the similarity of results compared to other LNO_x studies, we believe that our $1^\circ \times 1^\circ$ results based on postconvective LNO_x profile are sufficient for estimating average LNO_x production.

The LNO production settings in WRF-Chem varied in different studies. Zhao et al. (2009) set a NO_x production rate of 250 mol NO per flash in a regional-scale model, while Bela et al. (2016) chose the same method (330 mol NO per flash) that was used by Barth et al. (2012). Wang et al. (2015) assumed approximately 500 mol NO per flash which was derived by a cloud-scale chemical transport model and in-cloud aircraft observations (Ott et al., 2010). To illustrate the impact of LNO_x parameterization on LNO_x estimation, we apply another WRF-Chem NO_2 profile setting ($2\times$base flashrate, 500 mol NO $flash^{-1}$; hereinafter referred to as "2×500 mol NO $flash^{-1}$") to a priori profiles and evaluate the changes in AMF_{LNO_2}, AMF_{LNO_x}, LNO_2 and LNO_x productions. For the linear regression method (Fig. 9), LNO_2 production is 46.4 ± 7.8 mol/flash which is 11.5% larger than the basic one (41.6 ± 6.9 mol/flash). However, LNO_x production (increasing from 114.8 ± 18.2 mol/flash to 143.4 ± 24.0 mol/flash) depends to a large extent on the configuration of LNO production in WRF-Chem (Fig. 10). It remains unclear as to whether the NO-NO_2-O_3 cycle or other LNO_x reservoirs accounts for the increment of LNO_x production. This would need detailed source analysis in WRF-Chem and is beyond the scope of this study.

Figure 11 shows the average percentage changes in AMF_{LNO_2}, AMF_{LNO_x}, LNO_2 and LNO_x between retrievals using profiles based on 1×200 mol NO $flash^{-1}$ and 2×500 mol NO $flash^{-1}$. These results were obtained by averaging data over MJJA 2014 based on the method described in Sect. 2.5 with the criterion of CRF $\geq$ 90%. The effects on LNO_2 and LNO_x retrieval from increasing LNO profile values show mostly the same tendency: smaller AMF_{LNO_2} and AMF_{LNO_x} leads to larger LNO_2 and LNO_x, but the changes are region dependent. This is caused by the nonlinear calculation of AMF_{LNO_2} and AMF_{LNO_x}. As the contribution of LNO_2 increases, both the numerator and denominator of Eq. (2) increase. Note that the LNO_2 accounts for a fraction of NO_2 above the clouds, the magnitude of increasing denominator could be different in that of increasing numerator, resulting in a different effect on the AMF_{LNO_2} and AMF_{LNO_x}.

Figure 12 shows the comparison of the mean LNO and LNO_2 profiles in two specific regions where the 2×500 mol NO $flash^{-1}$ setting leads to both lower and higher LNO_2 production. The first one (Fig. 12a) is the region (36°N – 37°N, 89°W – 90°W) containing the minimal negative percent change in LNO_2 (Fig. 11c). The second one (31°N – 32°N, 97°W – 98°W), Figure 12b, has the largest positive percent change in LNO_2 (Fig. 11c). Although the relative distribution of mean LNO and LNO_2 profiles is similar in both regions, the magnitude differs with a factor of 10. This phenomenon implies that the performance of lightning parameterization in WRF-Chem is region dependent and the unrealistic profile could appear in the UT. Although this sensitivity analysis is false in some regions, it allows the calculation of an upper limit on the NO_2 due to LNO and LNO_2 profiles. As discussed in Laughner and Cohen (2017), the scattering weights are uniform under cloudy conditions and the sensitivity of NO_2 is nearly constant with different pressure levels because of the high albedo. However, the relative distribution of LNO_2 within the UT should be taken carefully in our research. If the LNO_2/NO_2 above the cloud is large enough (Fig. 12a), the AMF_{LNO_2} is largely determined by the ratio of LNO_2Vis to LNO_2 which is related to the relative distribution. When the condition of high LNO_2/NO_2 is not met, both relative distribution and ratio are involved (Fig. 12b).

To clarify this, we applied the same sensitivity test of different simulating LNO amounts for all four methods mentioned in Sect. 2.4: LNO_2, LNO_2Vis, LNO_2Clean and NO_2Vis (Fig. 13). Note that the threshold for CRF is set to 100% to simplify Eq. (2) to:

$$AMF_{\mathrm{LNO}_x} = \frac{\int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p) NO_2(p)\, dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p)\, dp} \tag{7}$$

The overall differences of LNO_2Clean and NO_2Vis are smaller than those of LNO_2 and LNO_2Vis. Comparing the composition of numerator and denominator in the equations, it is clear why the impact of different simulating LNO amounts is smaller in Fig. 13 (c) and (d). For LNO_2Clean and NO_2Vis, both the SCD and VCD will increase (decrease) when more (less) LNO_2 or NO_2 presents. The difference between Fig. 13(a) and Fig. 13(b) is the denominator: the total tropospheric LNO_2 vertical column and visible LNO_2 vertical column respectively. As a result, the negative values in Fig. 13(a) is caused by the part of LNO_2 below the cloud. The comparison between Figure 4 and Figure 9 shows that LNO_2Clean and LNO_2 values are more similar while LNO_2 and NO_2Vis values are same. The uncertainty of retrieved LNO_2 and LNO_x productions is driven by this error, and we conservatively estimate this to be ± 13% and ± 26% respectively.

4 Uncertainties Analysis

The uncertainties of the LNO_2 and LNO_x production are estimated following Pickering et al. (2016), Lapierre et al. (2019) and Laughner et al. (2019). We determine the uncertainty due to BEHR tropopause pressure, cloud radiance fraction, surface pressure, surface reflectivity, profile shape, profile location, V_{strat}, the detection efficiency of lightning, t_{window} and LNO_2 lifetime numerically by perturbing each parameter in turn and re-retrieval of the LNO_2 and LNO_x with the perturbed values (Table 3).

The GEOS-5 monthly tropopause pressure, which is consistent with the NASA Standard Product, is applied instead of the variable WRF tropopause height to evaluate the uncertainty (6% for LNO_2 and 4% for LNO_x) caused by the BEHR tropopause pressure. The resolution of GLOBE terrain height data is much higher than the OMI pixel and a fixed scale height is assumed in the BEHR algorithm. As a result, Laughner et al. (2019) compared the average WRF surface pressures to the GLOBE surface pressures and arrived at the largest bias of 1.5%. Based on the largest bias, we vary the surface pressure (limited to less than 1020 hPa) and the uncertainty can be neglected.

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The error in cloud radiance fraction is transformed from cloud fraction using:

$$\sigma = 0.05 \cdot \left.\frac{\partial f_r}{\partial f_g}\right|_{f_{g,pix}} \tag{8}$$

where f_r is the cloud radiance fraction, f_g is the cloud fraction and $f_{g,pix}$ is the cloud fraction of a specific pixel. We calculate $\partial f_r/\partial f_g$ under $f_{g,pix}$ by the relationship between all binned f_r and f_g with the increment of 0.05 for the each specific OMI orbit. Considering the relationship, the error in cloud fraction is converted to an error in cloud radiance fraction of 2% for both LNO_2 and LNO_x.

The accuracy of the 500 m MODIS albedo product is usually within 5% of albedo observations at the validation sites and those exceptions with low quality flags have been found to be primarily within 10% of the field data (Schaaf et al., 2011). Since we use the bidirectional reflectance distribution function (BRDF) data directly, rather than including a radiative transfer model, 14% Lambertian equivalent reflectivity (LER) error and 10% uncertainty are combined to get a perturbation of 17% (Laughner et al., 2019). The uncertainty due to surface reflectivity can be neglected with the 17% perturbation.

As discussed at the end of Sect. 3.3, another setting of LNO_2 (2×500 mol NO flash^{-1}) is applied to determine the uncertainty of the lightning parameterization and the vertical distribution of LNO in WRF-Chem. Differences between the two profiles lead to an uncertainty of 13% and 26%in the resulting LNO_2 and LNO_x production. Another sensitivity test allows each pixel to shift by - 0.2, 0, or + 0.2 degrees in the directions of longitude and latitude, taking advantage of the high-resolution profile location in WRF-Chem. The resulting uncertainty of LNO_x production is 1% including the error of transport and chemistry by shifting pixels.

Compared to the NASA standard product v2, Krotkov et al. (2017) demonstrated that the noise in V_{strat} is 1×10^{14} cm^{-2}. Errors in polluted regions can be slightly larger than this value, while errors in the cleanest areas are typically significantly smaller (Bucsela et al., 2013). We estimated the uncertainty of V_{strat} component and the slant column errors to be 15% and 5%, respectively, following Allen et al. (2019).

Based on the standard deviation of the detection efficiency estimation over the CONUS relative to LIS, ENTLN detection efficiency uncertainties are $\pm$ 16% for total and IC flashes/strokes. Due to the high detection efficiency of CG over the CONUS, the uncertainty is estimated to be $\pm$ 5% (Lapierre et al., 2019). It is found that the resulting uncertainty of detection efficiency is 15% in the production analysis. We have used the t_{window} of 2.4 h for counting ENTLN flashes and strokes to analyze LNO_2 and LNO_x production. Because t_{window} derived from the ERA5 reanalysis can not represent the variable wind speeds, a sensitivity test is performed which yields an uncertainty of 10% for production per flash and 8% for production per stroke using t_{window} of 2 h and 4 h. Meanwhile, the lifetime of UT NO_x ranges from 2 hours to 12 hours depending on the convective location, the methyl peroxy nitrate and alkyl and multifunctional nitrates (Nault et al., 2017). The lifetime (τ) of NO_2 in Eq. (6) is replaced by 2 and 12 hours to determine the uncertainty as 24% due to lifetime. The lifetime is the most likely uncertainty in the production analysis of LNO_2 while the uncertainty caused by lightning parameterization is comparable with that for the LNO_x type.

The overall uncertainty is estimated as the square root of the sum of the squares of all individual uncertainties in Table 3. The net uncertainty is 37% and 43% for LNO_2 type and LNO_x type respectively. The mean LNO_2/flash, LNO_x/flash, LNO_2/stroke,

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LNO_x/stroke based on the linear regression and summation method are 44 mol/flash, 120 mol/flash, 8 mol/stroke and 22 mol/stroke. Applying the corresponding uncertainty to these mean values, we arrive at 44 $\pm$ 16 mol LNO_2/flash, 120 $\pm$ 52 mol LNO_x/flash, 8 $\pm$ 3 mol LNO_2/stroke and 22 $\pm$ 9 mol LNO_x/stroke. This is in the range of current literature estimate ranging from 33 to 500 mol LNO_x/flash (Schumann and Huntrieser, 2007; Beirle et al., 2010; Bucsela et al., 2010). Bucsela et al. (2010) estimated LNO_x production of 100 – 250 mol/flash which is similar to our flash-based results. Pickering et al. (2016) estimated LNO_x production to be 80 $\pm$ 45 mol per flash for the Gulf of Mexico, which is 50% smaller than our flash-based results over the CONUS. Note that the criteria defined in Sect. 3.1 lead to many missing data over the Gulf of Mexico, thus it is actually a comparison between different regions. For the stroke-based results, Lapierre et al. (2019) yields lower LNO_2 production of 1.6 $\pm$ 0.1 mol per stroke, the difference is caused by the different version of BEHR algorithm and several settings as mentioned in Sect. 3.2. Bucsela et al. (2019) inferred an average value of 200 $\pm$ 110 moles (67% larger than our results) LNO_x produced per flash over the North America, this is related to the different algorithm and lightning data.

5 Conclusions

In this study, a new algorithm for retrieving LNO_2 (LNO_x) from OMI, including LNO_2 (LNO_x) below cloud, has been developed for application over active convection, whether in clean or polluted regions. It uses specific criteria combining with several other conditions (sufficient CRF, coincident ENTLN data, TL $\geq$ 1000 and ratio $\geq$ 50%) to ensure that the electrically active regions are detected by OMI and simulated by WRF-Chem successfully. We conducted an analysis on 1° $\times$ 1° daily boxes in MJJA 2014 and obtained the seasonal mean LNO_2 and LNO_x PEs over the CONUS. Considering all the uncertainties (Table 3) and applying the summation and regression method, the final mean PEs are estimated to be 44 $\pm$ 16 mol LNO_2/flash, 120 $\pm$ 52 mol LNO_x/flash, 8 $\pm$ 3 mol LNO_2/stroke and 22 $\pm$ 9 mol LNO_x/stroke.

Compared with former methods, we find that NO_2Vis and LNO_2Clean are more sensitive to background NO_2, while NO_2Vis underestimates PE because of the neglected below-cloud LNO_2 and LNO_2Clean overestimates LNO_2 production due to the over-cloud background NO_2. Finally, implementing profiles generated with 1$\times$200 mol NO flash^{-1} and 2$\times$500 mol NO flash^{-1}, we find that the regionally dependent effect. Both the relative distribution of LNO_2 and the ratio of LNO_2 to NO_2 would take the comprehensive effect for differences by the nonlinear calculation of AMF_{LNO_2} and AMF_{LNO_x}.

Since other regions, like China and India, have much more NO_2 pollutions than the CONUS, it is necessary to consider the background NO_2 in detail. These analyses will be complemented by the recently launched satellite instrument (TROPOspheric Monitoring Instrument [TROPOMI]) (Veefkind et al., 2012; Boersma et al., 2018; Griffin et al., 2019) and Lightning Mapping Imager (LMI) on the new generation Chinese geostationary meteorological satellites Fengyun-4 (Min et al., 2017; Yang et al., 2017; Zhang et al., 2019). Future work investigating the flash channel length and more detailed lightning parameterization in WRF-Chem would greatly benefit LNO_x estimation. Applying current method in future studies may enhance the accuracy of LNO_x production at both local and global level.

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Code and data availability. The retrieval algorithm used in Sect. 2.4 is available at https://github.com/zxdawn/BEHR-LNOx (last access: November 26, 2019; Zhang and Laughner, 2019). The WRF-Chem model output and LNO_x product are available upon request to Xin Zhang (xinzhang1215@gmail.com).

Appendix A: AMF Definitions used in this Study

$$AMF_{\mathrm{LNO_2}} = \frac{(1-f_r)\int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p)\,dp + f_r \int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p) NO_2(p)\,dp}{\int_{p_{surf}}^{p_{tp}} LNO_2(p)\,dp} \tag{A1}$$

$$AMF_{\mathrm{LNO}_x} = \frac{(1-f_r)\int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p)\,dp + f_r \int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p) NO_2(p)\,dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p)\,dp} \tag{A2}$$

where f_{r} is the radiance cloud fraction, p_{surf} is the surface pressure, p_{tp} is the tropopause pressure, p_{cloud} is the cloud optical pressure (CP), w_{clear} and w_{cloudy} are respectively the pressure dependent scattering weights from the TOMRAD lookup table (Bucsela et al., 2013) for clear and cloudy parts, and $NO_2(p)$ is the modeled NO_2 vertical profile. $LNO_2(p)$ and $LNO_x(p)$ are respectively the LNO_2 and LNO_x vertical profile calculated by the difference of vertical profiles between WRF-Chem simulations with and without lightning.

$$AMF_{\mathrm{LNO_2 Clean}} = \frac{(1-f_r)\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} w_{\mathrm{clear}}(p) LNO_2(p)\,dp + f_r \int_{p_{\mathrm{cloud}}}^{p_{\mathrm{tp}}} w_{\mathrm{cloudy}}(p) LNO_2(p)\,dp}{\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} LNO_2(p)\,dp} \tag{A3}$$

$$AMF_{\mathrm{NO_2 Vis}} = \frac{(1-f_r)\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} w_{\mathrm{clear}}(p) NO_2(p)\,dp + f_r \int_{p_{\mathrm{cloud}}}^{p_{\mathrm{tp}}} w_{\mathrm{cloudy}}(p) NO_2(p)\,dp}{(1-f_g)\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} NO_2(p)\,dp + f_g \int_{p_{\mathrm{cloud}}}^{p_{\mathrm{tp}}} NO_2(p)\,dp} \tag{A4}$$

$$AMF_{\mathrm{NO}_x \mathrm{Vis}} = \frac{(1-f_r)\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} w_{\mathrm{clear}}(p) NO_2(p)\,dp + f_r \int_{p_{\mathrm{cloud}}}^{p_{\mathrm{tp}}} w_{\mathrm{cloudy}}(p) NO_2(p)\,dp}{(1-f_g)\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} NO_x(p)\,dp + f_g \int_{p_{\mathrm{cloud}}}^{p_{\mathrm{tp}}} NO_x(p)\,dp} \tag{A5}$$

$$AMF_{\mathrm{LNO_2 Vis}} = \frac{(1-f_r)\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} w_{\mathrm{clear}}(p) NO_2(p)\,dp + f_r \int_{p_{\mathrm{cloud}}}^{p_{\mathrm{tp}}} w_{\mathrm{cloudy}}(p) NO_2(p)\,dp}{(1-f_g)\int_{p_{\mathrm{surf}}}^{p_{\mathrm{tp}}} LNO_2(p)\,dp + f_g \int_{p_{\mathrm{cloud}}}^{p_{\mathrm{tp}}} LNO_2(p)\,dp} \tag{A6}$$

where f_g is the geometric cloud fraction and $NO_x(p)$ is the modeled NO_x vertical profile.

Author contributions. YY directed the research and RJvdA, XZ and YY designed the research with feedback from the other co-authors; RJvdA and XZ developed the algorithm; JLL provided guidance and supporting data on the ENTLN data; XZ performed simulations and analysis with the help of YY, RJvdA, QC, XK, SY, JC, CH and RS; YY, RJvdA, JLL and XZ interpreted the data and discussed the results. XZ drafted the manuscript with comments from the co-authors; JLL, RJvdA and YY edited the manuscript.

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Competing interests. The authors declare that they have no conflict of interest.

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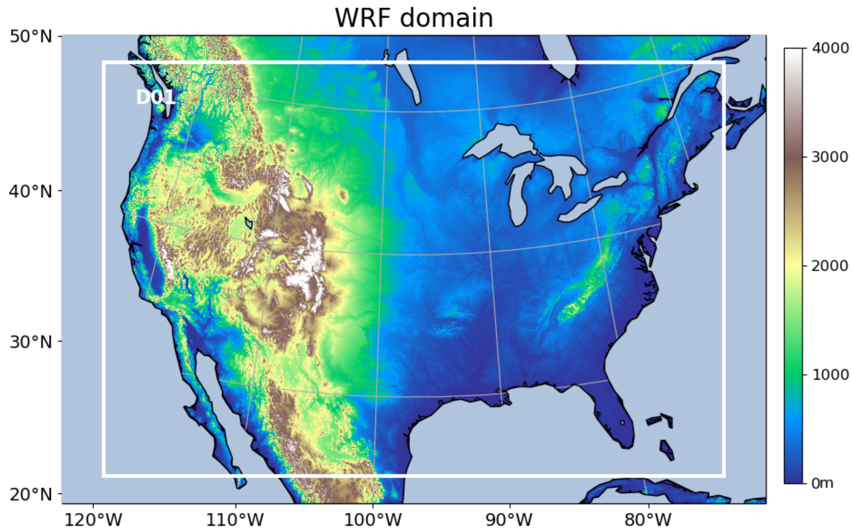

Figure 1. The 12-km resolution domain for WRF-Chem simulations.

Table 1. Definitions of the abbreviations for the criteria used in this study.

Abbreviations	Full form [source]
CRF	Cloud radiance fraction [OMI]
CP	Cloud optical pressure [OMI]
CF	Cloud fraction [WRF-Chem]
TL	Total lightning flashes [WRF-Chem]
ratio	modeled LNO_2 Vis / modeled NO_2 Vis [WRF-Chem]
crfα_entln	CRF $\geq \alpha$ + entln flashes(strokes) $\geq$ 2400(8160) [ENTLN]
crfα_cf40_entln	CRF $\geq \alpha$ + entln flashes(strokes) $\geq$ 2400(8160) + CF $\geq$ 40%
crfα_entln_tl1000	CRF $\geq \alpha$ + entln flashes(strokes) $\geq$ 2400(8160) + TL $\geq$ 1000
crfα_cf40_entln_tl1000	CRF $\geq \alpha$ + entln flashes(strokes) $\geq$ 2400(8160) + CF $\geq$ 40% + TL $\geq$ 1000
crfα_entln_tl1000_ratio50	CRF $\geq \alpha$ + entln flashes(strokes) $\geq$ 2400(8160) + TL $\geq$ 1000 + ratio $\geq$ 50%
crfα_cf40_entln_tl1000_ratio50	CRF $\geq \alpha$ + entln flashes(strokes) $\geq$ 2400(8160) + CF $\geq$ 40% + TL $\geq$ 1000 + ratio $\geq$ 50%

α has three options: 70%, 90% and 100%

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Table 2. LNO_x production under different conditions of CRF with coincident ENTLN data, TL ≥ 1000 and ratio ≥ 50%.

CRF (%)	ENTLN data type[1]	LNO_x/flash or LNO_x/stroke	R value	Intercept (10^6mol)	Days[2]
70	Flash	109.0 ± 15.3	0.84	0.23	85
90	Flash	114.8 ± 18.2	0.82	0.15	81
100	Flash	99.4 ± 15.3	0.84	0.10	71
70	Stroke	16.7 ± 2.6	0.79	0.58	96
90	Stroke	17.8 ± 2.9	0.78	0.29	93
100	Stroke	15.6 ± 3.1	0.75	0.16	82

[1]The threshold of ENTLN data is 2400 flashes box^{-1} and 8160 strokes box^{-1} during the period of 2.4 h before OMI overpass time. [2]The number of valid days with specific criteria in MJJA 2014.

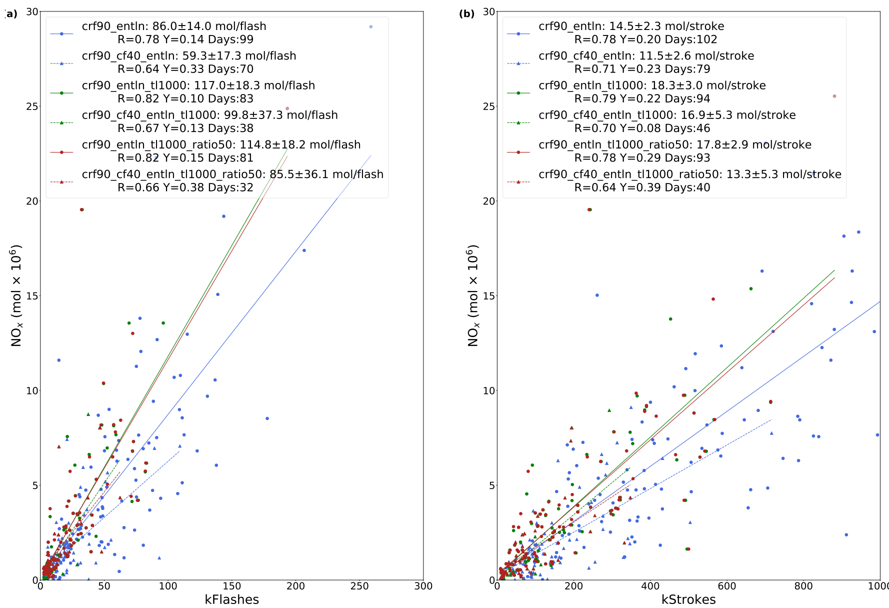

Figure 2. Linear regression of daily total LNO_x summed over boxes with lightning 2.4 h prior to OMI overpass for MJJA 2014. (a) The comparison of LNO_x production by six different combinations for CRF ≥ 90% with flash threshold of 2400 flashes. (b) Same as (a) except with a stroke threshold of 8160 strokes.

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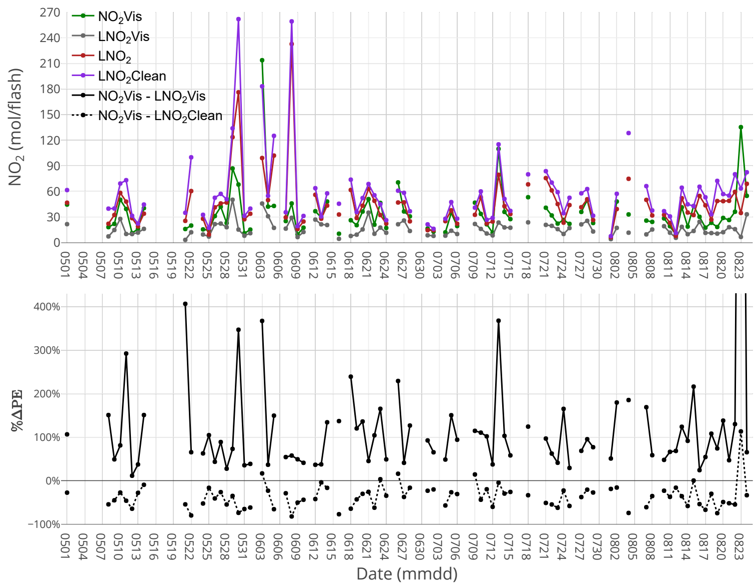

Figure 3. (top) Time series of NO_2Vis, LNO_2Vis, LNO_2 and LNO_2Clean production per day over the CONUS for MJJA 2014 with CRF ≥ 90% and a flash threshold of 2400 flashes per 2.4 h. (bottom) Time series of the percent differences between NO_2Vis and LNO_2Vis and the percent differences between NO_2Vis and LNO_2Clean with CRF ≥ 90%. The value of black dot on August 23 (not shown) is 1958%.

Table 3. Uncertainties for the estimation of LNO_2/flash, LNO_x/flash, LNO_2/stroke and LNO_x/stroke.

Type	Perturbation	LNO_2/flash[4]	LNO_x/flash[4]	LNO_2/stroke[4]	LNO_x/stroke[4]
BEHR tropopause pressure[1]	NASA product tropopause	6	4	6	4
Cloud radiance fraction[1]	± 5%	2	2	2	2
Surface pressure[1]	± 1.5%	0	0	0	0
Surface reflectivity[1]	± 17%	0	0	0	0
LNO_2 Profile[1]	2×500 mol NO $flash^{-1}$	13	26	13	26
Profile location[1]	Quasi-Monte Carlo	0	1	0	1
Lightning detection efficiency[2]	IC: ± 16%, CG: ± 5%	15	15	15	15
t_{window}[2]	2 – 4 hours	10	10	8	8
LNO_x lifetime[2]	2 – 12 hours	24	24	24	24
V_{strat}[3]	-	15	15	15	15
Systematic errors in slant column[3]	-	5	5	5	5
Net	-	37	43	36	43

[1]Laughner et al. (2019) [2]Lapierre et al. (2019) [3]Allen et al. (2019) [4]Uncertainty (%)

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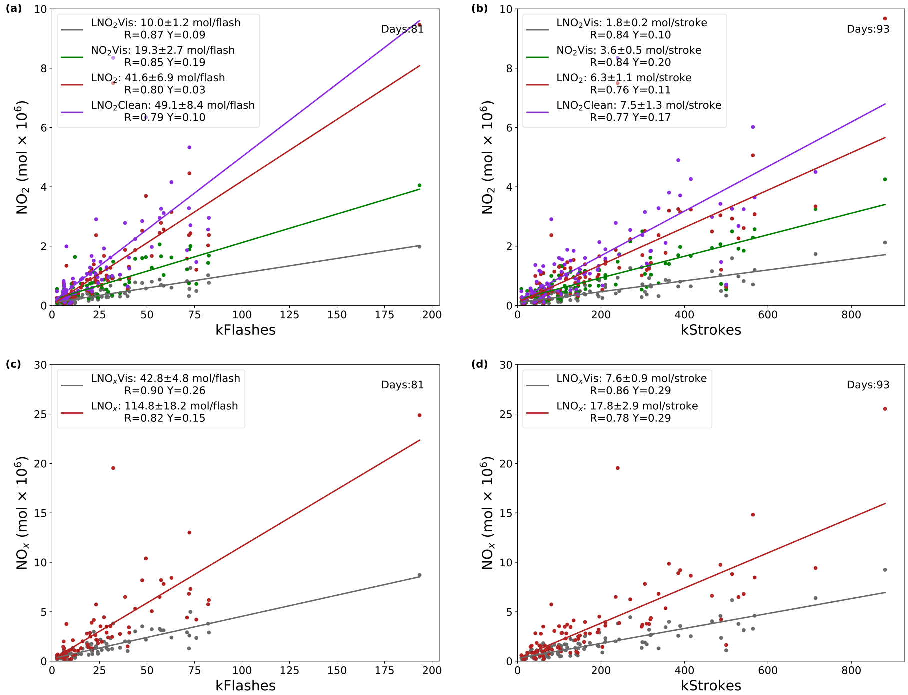

Figure 4. (a) Daily NO_2Vis, LNO_2Vis, LNO_2 and LNO_2Clean versus ENTLN total flashes data. (b) Same as (a) but for strokes. (c) Daily LNO_xVis and LNO_x versus total flashes. (d) Same as (c) but for strokes.

Table A1. Simple forms of abbreviations for AMFs.

Abbreviations	Numerator[1]	Denominator[2]
AMF_{LNO_2}	S_{NO_2}	V_{LNO_2}
$AMF_{LNO_2\,Vis}$	S_{NO_2}	$V_{LNO_2\,Vis}$
$AMF_{LNO_2\,Clean}$	S_{LNO_2}	V_{LNO_2}
$AMF_{NO_2\,Vis}$	S_{NO_2}	$V_{NO_2\,Vis}$
AMF_{LNO_x}	S_{NO_2}	V_{LNO_x}
$AMF_{NO_x\,Vis}$	S_{NO_2}	$V_{NO_x\,Vis}$

[1]The part of simulated VCD seen by OMI [2]The simulated VCD

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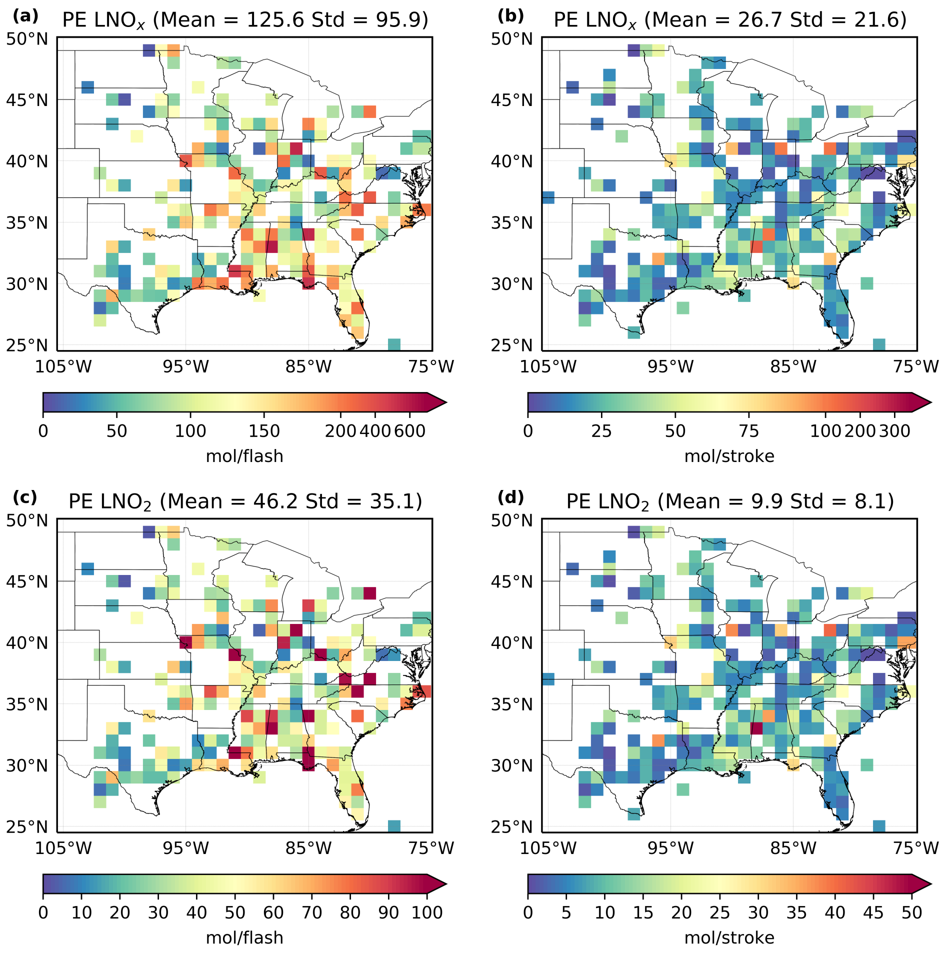

Figure 5. (a) and (c) Maps of 1°× 1°gridded values of mean LNO_x and LNO_2 production per flash with CRF ≥ 90% for MJJA 2014. (b) and (d) Same as (a) and (c) except for strokes.

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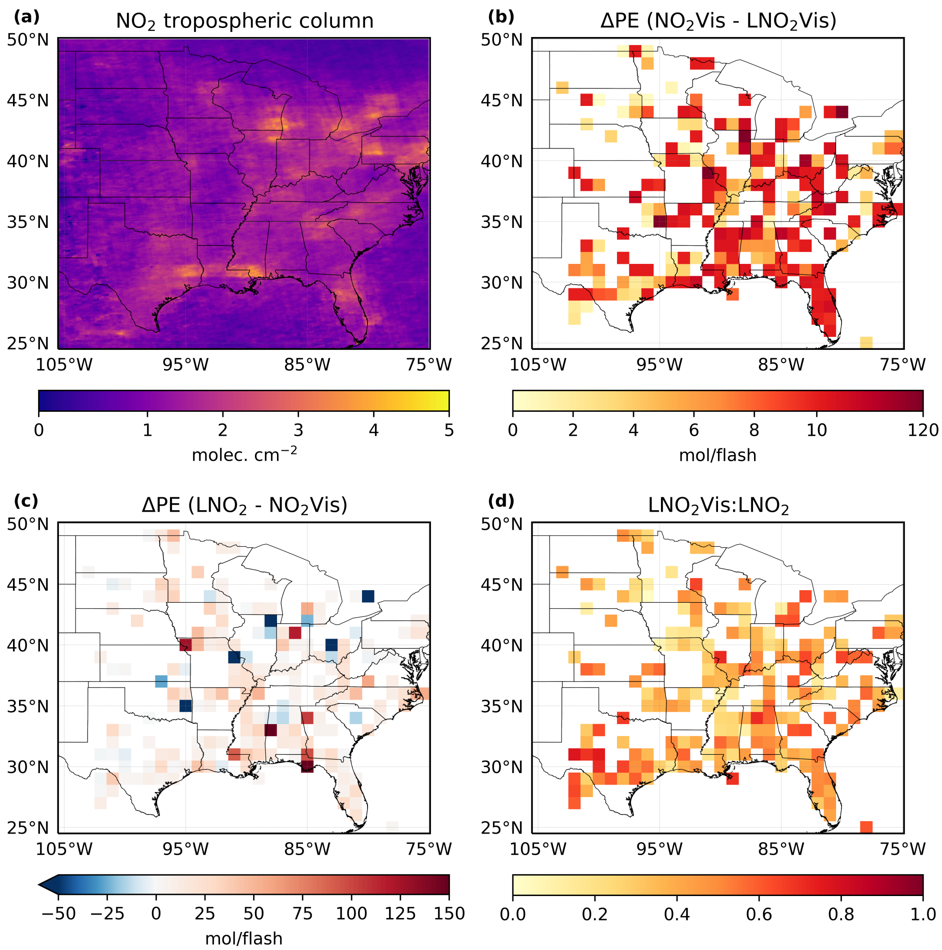

Figure 6. (a) Mean (MJJA 2014) NO_2 tropospheric column. (b) The differences of the estimated mean production efficiency between NO_2Vis and LNO_2Vis with CRF $\geq$ 90%. (c) The same differences as (b) but between LNO_2 and NO_2Vis. (d) The ratio of LNO_2Vis to LNO_2.

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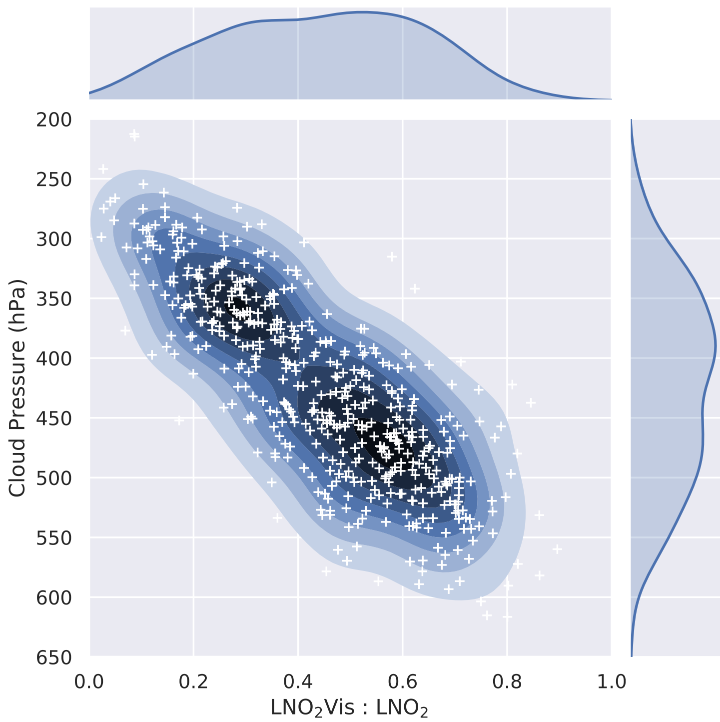

Figure 7. Scatter plots of the daily ratio of LNO_2Vis to LNO_2 versus the daily cloud pressure measured by OMI with CRF ≥ 90% for MJJA 2014.

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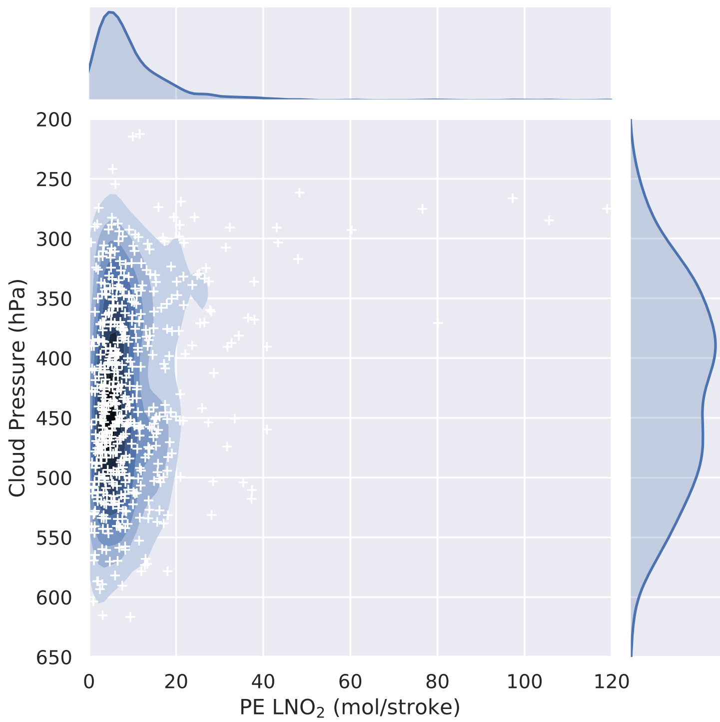

Figure 8. Scatter plots of the daily LNO_2 production efficiency versus the daily cloud pressure measured by OMI with CRF $\geq$ 90% for MJJA 2014.

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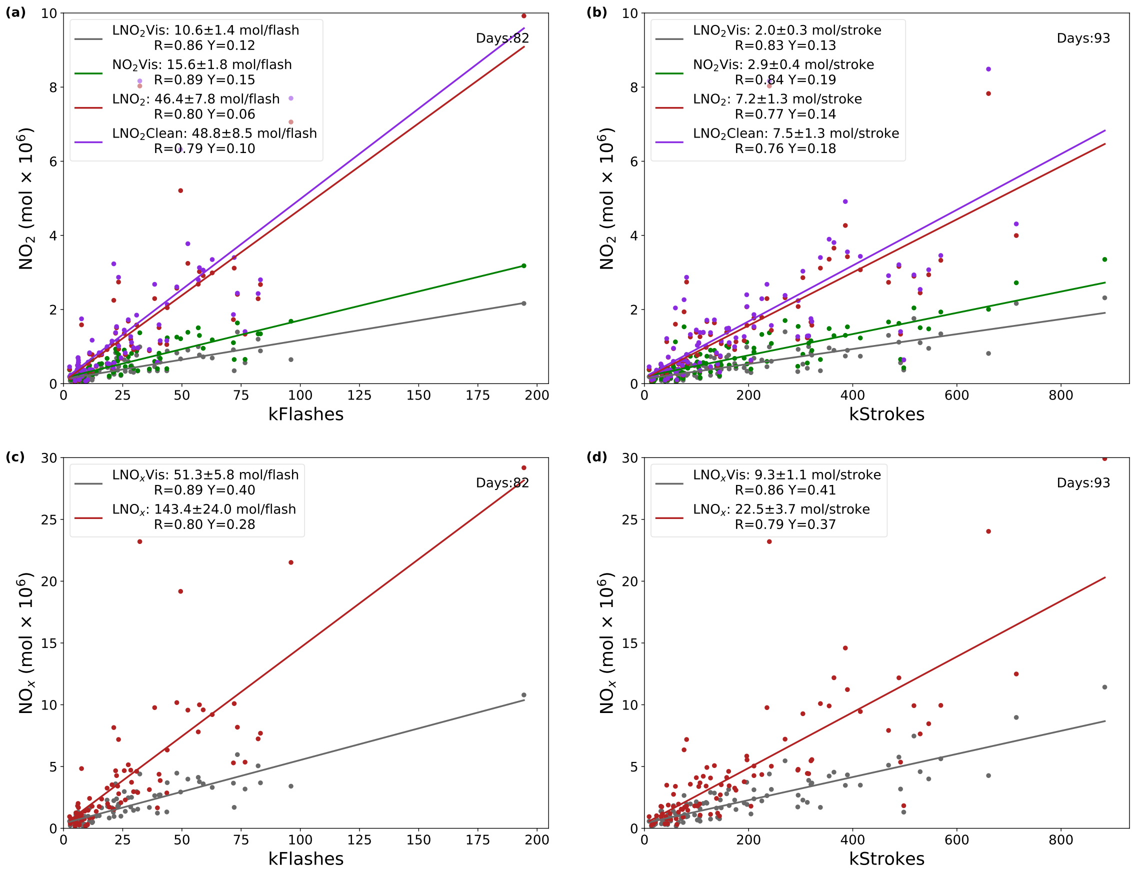

Figure 9. Same as Figure 4 except for 2×500 mol NO flash^{-1} configuration.

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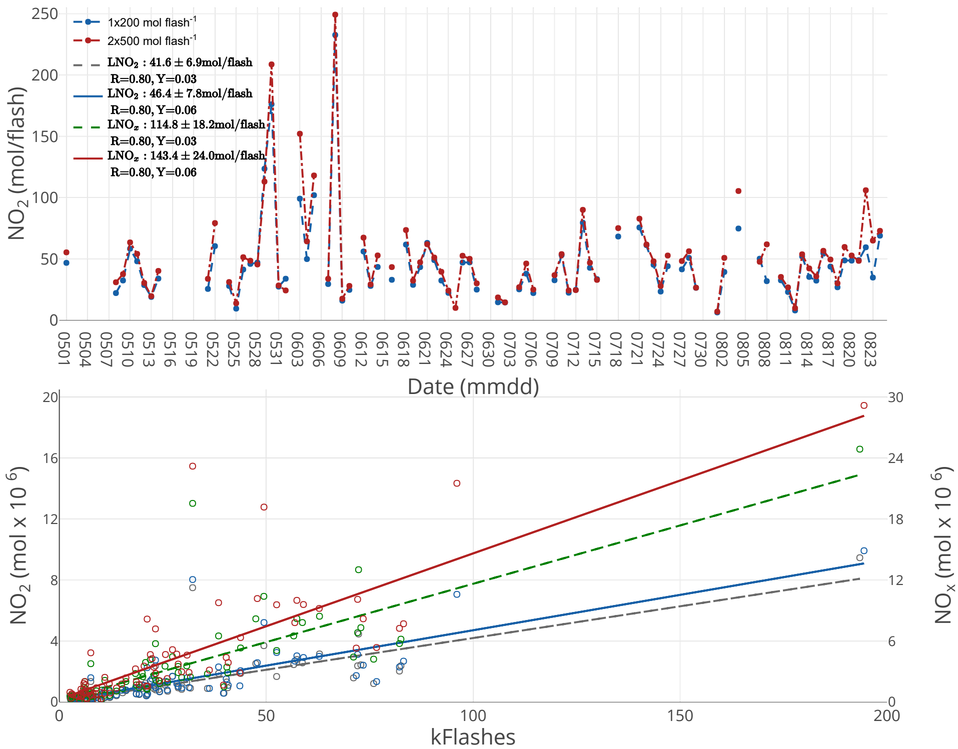

Figure 10. (top) Time series of LNO_2 production per day over the CONUS for MJJA 2014 with CRF $\geq$ 90% and a flash threshold of 2400 flashes per 2.4 h. Blue lines mark the basic LNO configuration (200 mol NO flash^{-1} and 1$\times$base flashrate) while red lines mark 500 mol NO flash^{-1} and 2$\times$base flashrate. (bottom) Daily LNO_2 and LNO_x versus ENTLN total flashes data. Dashed lines are based on basic LNO configuration while solid lines stand for the larger LNO configuration.

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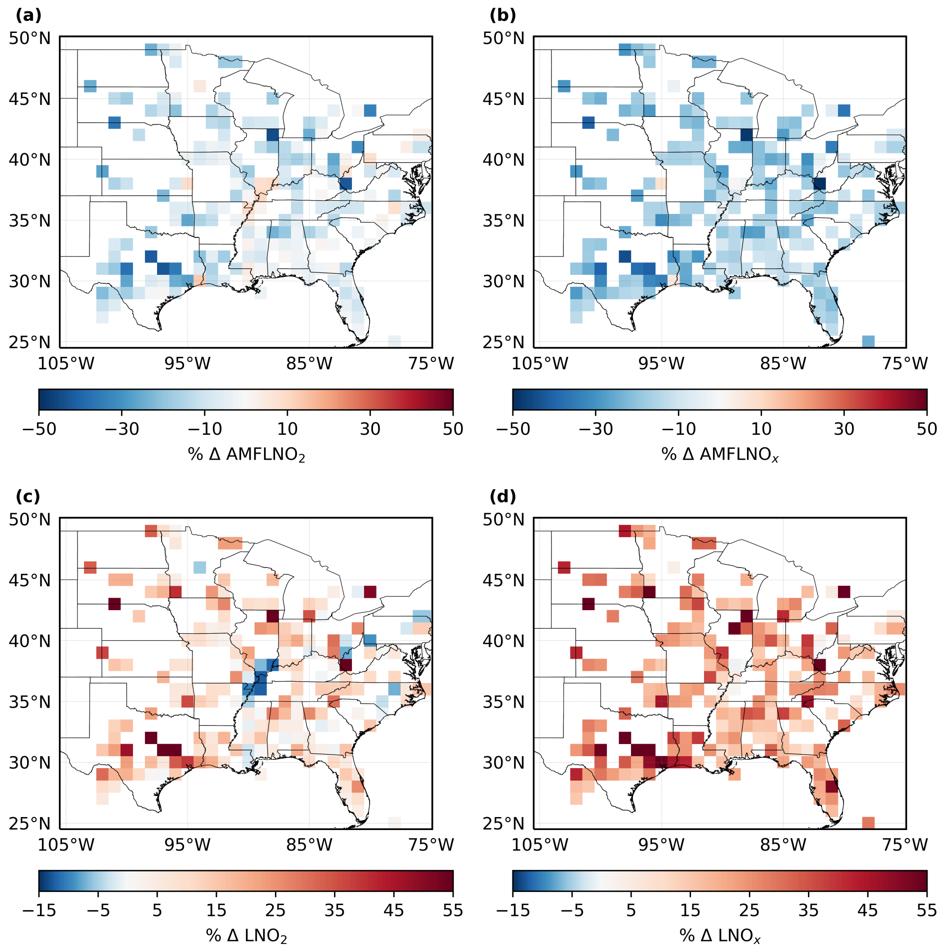

Figure 11. Average percent difference in (a) AMF_{LNO_2}, (b) AMF_{LNO_x}, (c) LNO_2 and (d) LNO_x with CRF $\geq$ 90% over MJJA 2014. Difference between profiles are generated by 2×500 mol NO flash^{-1} and 1×200 mol NO flash^{-1}.

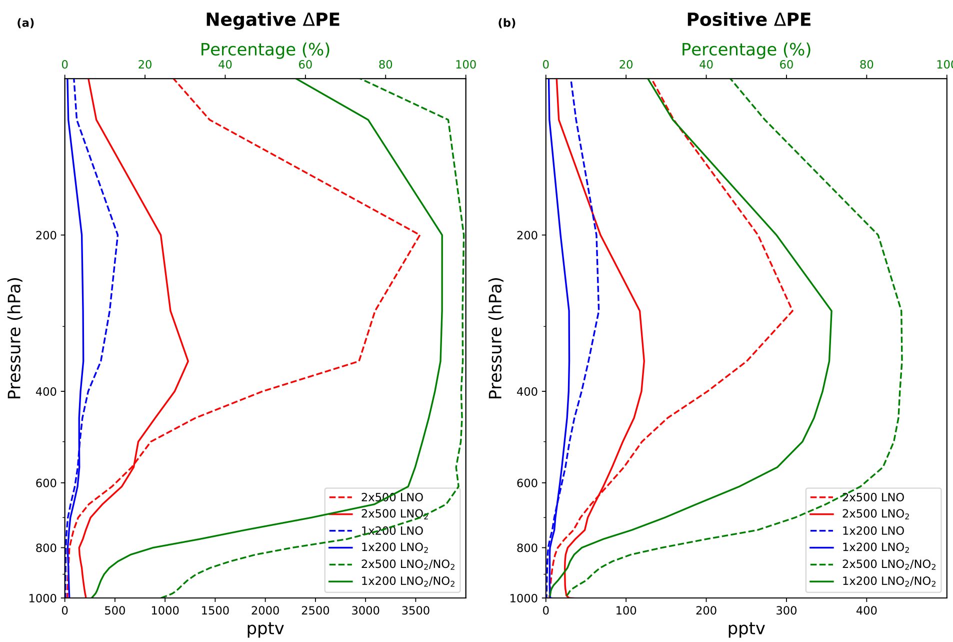

Figure 12. LNO and LNO_2 profiles with different LNO settings at (a) the region containing the minimal negative percent change in LNO_2 and (b) the region containing the largest positive percent change in LNO_2 when the LNO setting is changed from 1×200 mol NO flash^{-1} to 2×500 mol NO flash^{-1}, averaged over MJJA 2014. The profiles using 1×200 (2×500) mol NO flash^{-1} are shown in blue (red) lines. Solid (dashed) green lines are the mean ratio of LNO_2 to NO_2 with 1×200 (2×500) mol NO flash^{-1}.

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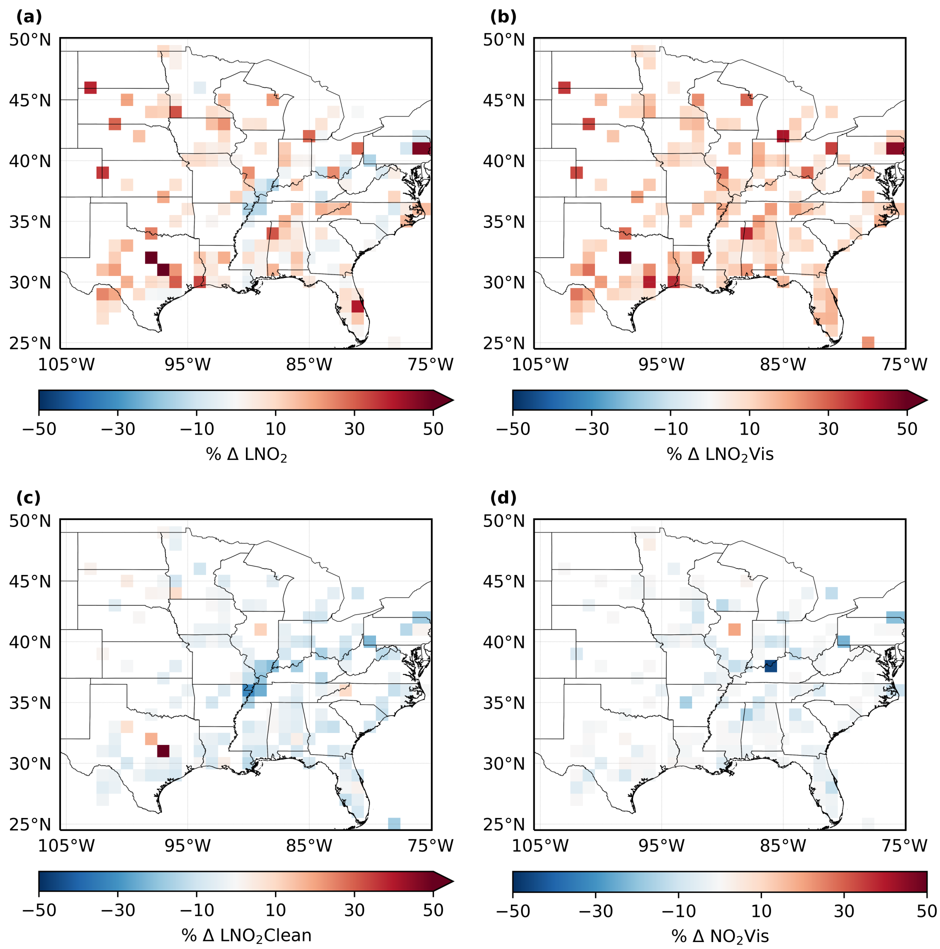

Figure 13. Average percent difference in (a) LNO_2, (b) LNO_2Vis, (c) LNO_2Clean and (d)NO_2Vis with CRF = 100% over MJJA 2014.