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). Satellite 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 that uses the Berkeley High Resolution (BEHR) v3.0B NO_2 retrieval algorithm 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) data 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 32 ± 15 mol NO_2 flash^{-1} and 6 ± 3 mol NO_2 stroke^{-1} while LNO_x PE is 90 ± 50 mol NO_x flash^{-1} and 17 ± 10 mol NO_x stroke^{-1}. Results reveal that our method reduces the sensitivity to the background NO_2 and includes 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 are mainly produced by soil, biomass burning and fossil fuel combustion, while NO_x in the middle and upper troposphere originates 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 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 versus the commonly used cloud-top height (CTH) approach. While global lightning is predicted to increase by 5 — 16% over the next century with the CTH approach (Clark et al., 2017; Banerjee et al., 2014; Krause et al., 2014), a 15% decrease in global lightning was estimated with IFLUX in 2100 under a strong global warming scenario (Finney et al., 2018). As a result of the different effects on radiative forcing from ozone and methane, a net positive radiative forcing was found with the CTH approach while there is little net radiative forcing with the IFLUX approach (Finney et al., 2018). However, the convective available potential energy (CAPE) times the precipitation rate (P) proxy predicts a 12 $\pm$ 5% increase in the Continental US (CONUS) lightning strike rate per kelvin of global warming (Romps et al., 2014), while the IFLUX proxy predicts the lightning will only increase 3.4%/K over the CONUS. Recently, Romps (2019) compared the CAPE $\times$ P proxy and IFLUX method in cloud-resolving models. They reported that higher CAPE and updraft velocities caused by global warming could lead to the large increases in tropical lightning simulated by CAPE $\times$ P proxy, while IFLUX proxy predicts little change in tropical lightning because of the small changes in the mass flux of ice.

In the view of the regionally 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). Satellite 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 quantified 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 annual mean NO_2 column densities, more recent studies applied specific approaches to investigate LNO_x directly over active convection. 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 and did not consider 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). Their analysis was restricted to 30×60 km^2 satellite pixels where the flash rate exceeded 1 flash km^{-2} hr^{-1}. But they found LNO_x production to be highly variable and correlations between flash rate densities and LNO_x production were low in some cases. Bucsela et al. (2010) estimated 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 column 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}. 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. Since they considered NO_2 above the cloud as LNO_2 in the algorithm due to the difficulty and uncertainty in determining the background NO_2, their AMF and derived VCD of LNO_x (LNO_2) is named as $AMF_{LNO_x\mathrm{Clean}}$ ($AMF_{LNO_2\mathrm{Clean}}$) and LNO_xClean (LNO_2Clean), respectively. Note that Pickering et al. (2016) considered the two estimates of background derived from aircraft flights in the Gulf of Mexico region (3% and 33%) and subtracted the mean value (18%) from the estimated mean LNO_x production efficiency (PE) for the background bias. However, we use the original algorithm directly without correction to distinguish the effect of different AMFs on LNO_x estimation in the remainder of this paper. Unless otherwise specified, abbreviations S and V are respectively defined as the tropospheric SCD and VCD in this paper.

More recently Bucsela et al. (2019) obtained an average PE of 180 $\pm$ 100 mol NO_x/flash over East Asia, Europe and North America based on a modification of the method used in Pickering et al. (2016). A power function between LNO_x and lightning flash rate was established, while the minimum flash-rate threshold was not applied. The tropospheric NO_x background was removed by subtracting the temporal average of NO_x at each box where the value was weighted by the number of OMI pixels which meet the optical cloud pressure and CRF criteria required to be considered deep convection but have 1 flash or less instead. The lofted pollution was considered as 15% of total NO_x 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 and stratospheric NO_2 at some locations.

On the other hand, Lapierre et al. (2020) constrained LNO_2 to 1.1 $\pm$ 0.2 mol NO_2/stroke for intracloud (IC) strokes and 10.7 $\pm$ 2.5 mol NO_2/stroke for cloud-to-ground (CG) strokes over the CONUS. LNO_2 per stroke was scaled to 24.2 mol NO_x/flash using mean values of strokes per flash and the ratio of NO_x 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 that can be "seen" by the satellite. The first part is the NO_2 above clouds (pixels with CRF > 0.9) and the second part is the NO_2 detected from cloud free areas. A threshold of 3×10^{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. (2020) and Pickering et al. (2016) is the air mass factor for lightning (AMF_{LNO_x}) implemented in the basic algorithm. In Lapierre et al. (2020), 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. (2020) without geographic restrictions, the contamination by 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, Laughner and Cohen (2017) showed that the OMI AMF is increased by $\sim$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 related to these observations is reduced to $< \pm 4\%$ for OMI viewing geometries.

In this paper, we focus 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 data, lightning data, model settings and the algorithm in detail. Section 3 explores the suitable data criteria, compares different methods and evaluates the effect of background NO_2, cloud and LNO_x parameterization on LNO_x production estimation. Section 4 examines the effect of different sources of the uncertainty on 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 $\sim$ 13:45 LT (ascending node) and has a swath width of 2600 km, with a nadir field-of-view resolution of $13 \times 24\ km^2$. 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; KNMI, 2012). For the current study, we used the NASA standard product V3.0 (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 cross-track bias caused by an instrument artifact from the measured slant column;

3. The AMF for stratospheric (AMF_{strat}) or tropospheric column (AMF_{trop}) is calculated from the NO_2 profiles 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} ($V_{LNO_x} = S_{NO_2}/AMF_{LNO_x}$) by replacing the original step 5. Details of this algorithm are discussed in section 2.4.

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 combined events (IC and CG) with LIS flashes and found that the relative flash detection efficiency of ENTLN over CONUS increases from 62.4% during 2011 to 79.7% during 2013. Lapierre et al. (2020) also compared combined ENTLN and the NLDN dataset with data from the LIS during 2014 and found the detection efficiencies of IC flashes and strokes to be 88% and 45%, respectively. Since we only use the ENTLN data in 2014 as Lapierre et al. (2020) 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 divided by 0.88 and 0.45, 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 12×12 km^2 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) are 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 based on the level of neutral buoyancy parameterization (Price and Rind, 1992; Wong et al., 2013) 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 "1×200 mol NO flash^{-1}"). Simulated total flash densities are higher than ENTLN observations over the Southeast US and lower than observations in the North Central US (Fig. 2). The impact of these biases on LNO_x production is discussed and mitigated in Sect. 3.1 and 3.4. 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 outputs of 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_{NO_2} is the OMI-measured tropospheric slant column NO_2 and AMF_{LNO_x} is a customized lightning air mass factor. The concept of AMF_{LNO_x} was also used in Beirle et al. (2009) to investigate the sensitivity of satellite instruments to freshly produced lightning NO_x. In order to estimate LNO_x, we define the AMF_{LNO_x} 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 cloud radiance 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 cloud radiance fraction (CRF), 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.

Please note that the CP is a reflectance-weighted pressure retrieved by the collision-induced O_2-O_2 absorption band near 477 nm (Acarreta et al., 2004; Sneep et al., 2008; Stammes et al., 2008). For a deep convective cloud with lightning, the CP lies below the geometrical cloud top which is much closer to that detected by thermal infrared sensors, such as the CloudSat and the Aqua MODerate-resolution Imaging Spectrometer (MODIS) (Vasilkov et al., 2008; Joiner et al., 2012). Hence, much of the tropospheric NO_2 measured by OMI lies inside the cloud rather than above the cloud top. In the following, "above cloud" or "below cloud" is relative to the cloud pressure detected by OMI. The sensitivity study of Beirle et al. (2009) compared the chemical composition from the cloud bottom to the cloud top and revealed that a significant fraction of the NO_2 within the cloud originating from lightning can be detected by the satellite. This valuable cloud pressure concept has been applied not only in the LNO_x research but also in the cloud slicing method of deriving the UT O_3 and NO_x (Ziemke et al., 2009; Choi et al., 2014; Strode et al., 2017; Ziemke et al., 2017; Marais et al., 2018). As discussed in Pickering et al. (2016), the ratio of V_{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 Sect. 3.4.

To compare our results with those of Pickering et al. (2016) and Lapierre et al. (2020), we calculate their $\text{AMF}_{\text{LNO}_x\text{Clean}}$ and $\text{AMF}_{\text{NO}_2\text{Vis}}$ respectively:

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

$$AMF_{\text{NO}_2\text{Vis}} = \frac{(1-f_r)\int_{p_{\text{surf}}}^{p_{\text{tp}}} w_{\text{clear}}(p) NO_2(p)\,dp + f_r \int_{p_{\text{cloud}}}^{p_{\text{tp}}} w_{\text{cloudy}}(p) NO_2(p)\,dp}{(1-f_g)\int_{p_{\text{surf}}}^{p_{\text{tp}}} NO_2(p)\,dp + f_g \int_{p_{\text{cloud}}}^{p_{\text{tp}}} NO_2(p)\,dp} \tag{4}$$

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 $\text{AMF}_{\text{LNO}_2\text{Vis}}$ is developed for later comparison.

$$AMF_{\text{LNO}_2\text{Vis}} = \frac{(1-f_r)\int_{p_{\text{surf}}}^{p_{\text{tp}}} w_{\text{clear}}(p) NO_2(p)\,dp + f_r \int_{p_{\text{cloud}}}^{p_{\text{tp}}} w_{\text{cloudy}}(p) NO_2(p)\,dp}{(1-f_g)\int_{p_{\text{surf}}}^{p_{\text{tp}}} LNO_2(p)\,dp + f_g \int_{p_{\text{cloud}}}^{p_{\text{tp}}} LNO_2(p)\,dp} \tag{5}$$

A full definition list of the used AMFs is shown in Appendix A.

2.5 Procedures for Deriving LNO_x

V_{LNO_x} is re-gridded to 0.05° × 0.05° grids using the constant value method (Kuhlmann et al., 2014). Then, it is analyzed in 1° × 1° grid boxes with a minimum of fifty valid 0.05° × 0.05° grids to minimize the noise. The main procedures of deriving LNO_x are as follows:

CRFs (CRF ≥ 70%, CRF ≥ 90% and CRF = 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 fraction (CF) is applied to the WRF-Chem results for the successful simulation of convection. The CF is defined as the maximum cloud fraction calculated by the Xu-Randall method between 350 and 400 hPa (Xu and Randall, 1996; Strode et al., 2017). This atmospheric layer (between 350 and 400 hPa) avoids any biases in the simulation of high clouds. We choose CF ≥ 40% suggested by Strode et al. (2017) to determine cloudy or clear for each simulation grid.

Besides cloud properties, a time period and sufficient flashes (or strokes) are required for fresh LNO_x to be 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., 2020). Meanwhile, 2400 flashes box^{-1} and 8160 strokes box^{-1} per 2.4 hour time window are chosen as sufficient for detecting LNO_x (Lapierre et al., 2020). These criteria will result in a low bias in the PE results, as Bucsela et al. (2019) found that the PE is larger at small flash rates which are discarded here. Since our study focuses on developing a new AMF and compare results with other works using the similar lightning thresholds (Lapierre et al., 2020; Pickering et al., 2016), we will only discuss results based on the strict criteria in the main text. For comparisons between 2400 flashes box^{-1} criterion and 1 flash box^{-1} criterion, scatter diagrams using different lightning criteria are presented in Appendix B.

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 in addition to 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 more than half of the NO_x above the cloud must has an LNO_x source.

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 $1° \times 1°$ box. For each grid box, the mean LNO_x vertical column is obtained by averaging V_{LNO_x} values from all regridded $0.05° \times 0.05°$ 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 $1° \times 1°$ box in MJJA 2014;

(2) linear regression method: applying the linear regression to daily mean values 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 (Table 2).

A daily search of the NO_2 product for coincident ENTLN flash (stroke) data results in 99 (102) valid days under the CRF90_ENTLN condition. Taking the flash type ENTLN data as an example, the number of valid days decreases from 99 to 81 under the CRF90_ENTLN_TL1000_ratio50 condition, while LNO_x/flash increases from 52.1 $\pm$ 51.1 mol/flash to 54.5 $\pm$ 48.1 mol/flash. The result is almost the same as that under the CRF90_ENTLN_TL1000 condition which is without the condition of more than half of the above-cloud NO_x having an LNO_x source. Although this indicates the criterion of TL works well, it is better to include the ratio criterion in case of some exceptions in the different AMF methods. Since CF $\geq 40\%$ 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 $\geq$ 1000 and ratio $\geq 50\%$ are chosen as the thresholds to explore the effects of three different CRF conditions (CRF $\geq 70\%$, CRF $\geq 90\%$ and CRF = 100%) on LNO_x production (Table 3). Apart from the fewer valid days under higher CRF conditions (CRF $\geq 90\%$ and CRF = 100%), LNO_x/flash increases from 35.7 $\pm$ 36.8 mol/flash to 54.5 $\pm$ 48.1 mol/flash and decreases again to 20.8 $\pm$ 37.4 mol/flash while LNO_x/stroke enhances from 4.1 $\pm$ 3.9 mol/stroke to 7.0 $\pm$ 4.8 mol/stroke and drops again to 2.6 $\pm$ 4.0 mol/stroke (Table 3), as the CRF criterion increases from 70% to 90% and to 100%. When the CRF increases from 90% to 100%, the LNO_x PE decreases because of the higher lightning density with fewer LNO_x (not shown). The increment of LNO_x PE caused by the CRF increase from 70% to 90% is opposite to the result of Pickering et al. (2016). This is an effect of the consideration of NO_2 contamination transported from the boundary layer in our method. 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 $\geq$ 90% considering the contamination by low and mid-level NO_2 and comparisons with the results of Pickering et al. (2016) and Lapierre et al. (2020).

3.2 Comparison of LNO_x Production based on Different AMFs

Lapierre et al. (2020) 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. (2020), we choose NO_2 instead of NO_x to derive production per flash (production efficiency, PE). 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 $\geq$ 90% and a flash threshold of 2400 flashes per 2.4 h. LNO_2 PEs are mostly in the range from 20 to 80 mol/flash. LNO_2Vis PEs are smaller than LNO_2 PEs 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 56 $\pm$ 20%. The effect of cloud properties on LNO_x PE will be discussed in more detail in section 3.4. Generally, the order of estimated daily 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_2Vis 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 PE (the largest slope) is 25.2 $\pm$ 22.3 mol NO_2/flash with a correlation of 0.25 and 2.3 $\pm$ 2.1 mol NO_2/stroke with a correlation of 0.22. As shown in Fig. 3, positive percent differences between NO_2Vis PE and LNO_2Clean PE occur much fewer than negative differences. As a result, NO_2Vis PE (17.1 $\pm$ 17.2 mol NO_2/flash and 0.4 $\pm$ 1.0 mol NO_2/stroke) is smaller than LNO_2Clean PE using the linear regression method.

In order to compare our result with that of Lapierre et al. (2020), we tried to remove the CP $\leq$ 650 hPa, TL $\geq$ 1000 and ratio $\geq$ 50% conditions from criteria. But, our result based on daily summed NO_2Vis values (3.8 $\pm$ 0.5 mol/stroke) is still larger than the value of 1.6 $\pm$ 0.1 mol/stroke mentioned in Lapierre et al. (2020). This may be caused by the different version of BEHR algorithm, as Lapierre et al. (2020) 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: March 8, 2020). Note that Lapierre et al. (2020) 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 PE (18.7 $\pm$ 18.1 mol/flash and 2.1 $\pm$ 1.8 mol/stroke) is between LNO_2Clean PE and NO_2Vis PE, which coincides with the daily results in Fig. 3. Furthermore, the LNO_x PE based on the linear regression of daily summed values, the same method used in Pickering et al. (2016), is 114.8 $\pm$ 18.2 mol/flash (or 17.8 $\pm$ 2.9 mol/stroke) which is larger than 91 mol/flash in Pickering et al. (2016), possibly due to the differences in geographic location, lightning data and chemistry model.

The mean and standard deviation of LNO_2 PE under CRF $\geq$ 90% using the summation method is 46.2 $\pm$ 35.1 mol/flash and 9.9 $\pm$ 8.1 mol/stroke, while LNO_x PE is 125.6 $\pm$ 95.9 mol/flash and 26.7 $\pm$ 21.6 mol/stroke (Fig. 5). The LNO_2 PE and LNO_x PE are both higher in the Southeast U.S. (denoted by the red box in Fig. 5 panels, 75°W – 95°W, 25°N – 37°N), consistent with Lapierre et al. (2020) and Bucsela et al. (2019). Compared with Fig. 3, Figure 6a and b present some large differences between NO_2Vis PE and LNO_2Vis PE, which are consistent with what we expect for polluted regions. Meanwhile, the differences between LNO_2 PE and NO_2Vis PE 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 result in LNO_2 PE higher than NO_2Vis PE (Fig. 6c). Figure 6d 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. While peaks of the LNO_2 profile are below the CP, the ratio would also be smaller. Therefore, a better understanding of LNO_2 profile and LNO_x below clouds is required.

3.3 Effects of Tropospheric Background on LNO_x Production

With respect to the LNO_2 production, the patterns in Fig. 6 indicate the improvement of our approach is different in polluted and clean regions. To simplify the quantification, we select six grids with similar NO_2 profile ($\sim$ 100 pptv) above the cloud with CRF = 100%. These grid boxes contain the polluted and clean cities denoted by stars and triangles in Fig. 6a, respectively. Then, the differences between AMFs are dependent on fewer parameters:

$$AMF_{\mathrm{LNO_2}} = \frac{\int_{p_{cloud}}^{p_{tp}} w_{\mathrm{cloudy}}(p) NO_2(p)\, dp}{\int_{p_{surf}}^{p_{tp}} LNO_2(p)\, dp} \tag{7}$$

$$AMF_{\mathrm{NO_2Vis}} = \frac{\int_{p_{cloud}}^{p_{tp}} w_{\mathrm{cloudy}}(p) NO_2(p)\, dp}{\int_{p_{cld}}^{p_{tp}} NO_2(p)\, dp} \tag{8}$$

$$AMF_{\mathrm{LNO_2Clean}} = \frac{\int_{p_{cloud}}^{p_{tp}} w_{\mathrm{cloudy}}(p) LNO_2(p)\, dp}{\int_{p_{surf}}^{p_{tp}} LNO_2(p)\, dp} \tag{9}$$

Figure 7 compares the mean profiles of NO_2, background NO_2 and background NO_2 ratio in polluted and clean grids. Generally, the profiles of the ratio of background NO_2 over total NO_2 are C-shape because UT LNO_2 concentrations are higher than UT background NO_2 concentrations. However, the ratio profile in Fig. 7e has one peak between the cloud pressure and tropopause as background NO_2 increases and LNO_2 decreases. Besides, the percentage of UT background NO_2 in polluted regions is steady and higher than that in clean regions.

Table 4 presents the relative changes among three methods in six cities. The difference between AMF_{LNO_2} (Eq. 7) and AMF_{LNO_2Clean} (Eq. 9) is the numerator: $\int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p)NO_2(p)\,dp$ and $\int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p)LNO_2(p)\,dp$. When the ratio of LNO_2 is higher or the region is cleaner, the relative difference is smaller (e.g. 5.0% – 12.0%, Fig. 7d – f). The largest relative difference (46.3%) occurs when the ratio of background NO_2 is continuously high in the UT (Fig. 7c). As a result, our approach is less sensitive to background NO_2 and more suitable for convective cases over polluted locations. In contrast, production estimated by our method is larger than that based on NO_2Vis due to the LNO_2 below the cloud. When the cloud is higher, especially the peak of LNO profile is lower than the cloud (Fig. 7b), the relative difference is larger (121.2%) because more LNO_2 can not be included into the NO_2Vis, which has been discussed in Sect. 3.2. The relative change between AMF_{LNO_2Clean} (Eq. 9) and AMF_{NO_2Vis} (Eq. 8) depends on $\int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p)LNO_2(p)\,dp / \int_{p_{surf}}^{p_{tp}} w_{cloudy}(p)LNO_2(p)\,dp$, which is also affected by cloud not the background NO_2. The largest relative change (153.8%) occurs at New Orleans, which has the lowest cloud pressure and consequently the smallest visible column.

3.4 Effects of Cloud and LNO_x Parameterization on LNO_x Production

Figure 8a 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 as the cloud pressure decreases from 600 to 300 hPa, NO_2Vis PE is smaller than LNO_2 PE in relatively clean areas as shown in Fig. 4. Apart from LNO_2Vis, the LNO_2 PE is also affected by CP. For LNO_2 PEs larger than 30 mol/stroke, the CPs are all smaller than 550 hPa (Fig. 8b). However, smaller LNO_2 PEs (< 30 mol/stroke) occur on all levels between 650 hPa and 200 hPa. Because of the limited amount of large LNO_2 PEs and lightning data, we cannot derive the relationship between LNO_2 PE and cloud pressure or other lightning properties at this stage. Because the 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, the 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 flash: normal flashes and anomalous flashes. Because updrafts are stronger and flash rates are higher in anomalous storms, UT LNO_x concentrations are larger in anomalous 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) in which LNO_x has already been redistributed by convective transport, while the other one (preconvection) uses LNO_x production profiles made before the redistribution of convective transport (Allen et al., 2012; Luo et al., 2017). However, given the similarity of results compared to other LNO_x studies, we believe that our $1° \times 1°$ 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 330 mol NO per flash 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×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 PE and LNO_x PE. For the linear regression method (Fig. 9), LNO_2 PE is 29.8 ± 20.5 mol/flash which is 59.4% larger than the basic one (18.7 ± 18.1 mol/flash). Meanwhile, LNO_x PE (increasing from 54.5 ± 48.1 mol/flash to 88.5 ± 61.1 mol/flash) also depends on the configuration of LNO production in WRF-Chem. The comparison between Fig. 4 and Fig. 9 shows that LNO_2Clean PE and LNO_2 PE are more similar while LNO_2 PE and NO_2Vis PE present the same tendency. 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 PE. This would need detailed source analysis in WRF-Chem and is beyond the scope of this study.

Figure 10 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 regionally 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 than that of increasing numerator, resulting in a different effect on the AMF_{LNO_2} and AMF_{LNO_x}. As mentioned in Zhu et al. (2019), the lightning densities in the Southeast U.S. might be overestimated using the 2×500 mol NO $flash^{-1}$ setting and the same lightning parameterization as ours. Fortunately, the AMFs and estimated LNO_2 change little in that region. Because the Southeast U.S. has the highest flash density (Fig. 2), the NO_2 in the numerator of AMF is dominated by LNO_2. Both the SCD and VCD will increase when the model uses higher LNO_2. In other words, the sensitivity to the LNO setting decreases and the relative distribution of LNO_2 matters.

Figure 11 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 lower and higher LNO_2 PEs, respectively. The first one (Fig. 11a) is the region (36°N – 37°N, 89°W – 90°W) containing the minimal negative percent change in LNO_2 (Fig. 10c). The second one (31°N – 32°N, 97°W – 98°W), Figure 11b, has the largest positive percent change in LNO_2 (Fig. 10c). Although the relative distributions of mean LNO and LNO_2 profiles are 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 regionally dependent and an 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 into consideration. If the LNO_2/NO_2 above the cloud is large enough (Fig. 11a), 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 important (Fig. 11b).

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. 12). Note that the threshold for CRF is set to 100% to simplify Eq. (2) to Eq. (7). The overall differences of LNO_2Clean and NO_2Vis are smaller than those of LNO_2 and LNO_2Vis. Comparing the numerator and denominator in the equations, it is clear why the impact of different simulating LNO amounts is smaller in Fig. 12c 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. 12a and Fig. 12b 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. 12a are caused by the part of LNO_2 below the cloud. The uncertainty of retrieved LNO_2 and LNO_x PEs is driven by this error, and we conservatively estimate this to be $\pm$ 13% and $\pm$ 25% respectively.

4 Uncertainties Analysis

The uncertainties of the LNO_2 and LNO_x PEs are estimated following Pickering et al. (2016), Allen et al. (2019), Bucsela et al. (2019), Laughner et al. (2019) and Lapierre et al. (2020). We determine the uncertainty due to BEHR tropopause pressure, cloud radiance fraction, cloud pressure, 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-retrieving the LNO_2 and LNO_x with the perturbed values (Table 5).

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 PE and 4% for LNO_x PE) caused by the BEHR tropopause pressure. The cloud pressure bias is given as a function of cloud pressure and fraction by Acarreta et al. (2004) implying an uncertainty of 32%, the most likely uncertainty in the production analysis, for LNO_2 PE and 34% for LNO_x PE. 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.

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

$$\sigma = 0.05 \cdot \left.\frac{\partial f_r}{\partial f_g}\right|_{f_{g,pix}} \tag{10}$$

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 the LNO_2 and LNO_x PEs.

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. 3.4, 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 25% in the resulting PEs of LNO_2 and LNO_x. 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 PE 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 10% 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., 2020). 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. This is comparable with the uncertainty (25%) caused by lightning parameterization for the LNO_x type.

Recent works revealed that the modeled NO/NO_2 ratio departs from the data in the SEAC^{4}RS aircraft campaign (Travis et al., 2016; Silvern et al., 2018). Silvern et al. (2018) attributed this to the positive interference on the NO_2 measurements or errors in the cold-temperature NO-NO_2-O_3 photochemical reaction rate. We assign a 20% bias with $\pm$ 15% uncertainty to this error considering the possible positive NO_2 measurements interferences (Allen et al., 2019; Bucsela et al., 2019) and estimate the uncertainty to be 15% for LNO_x PE.

In addition, the estimation of LNO_x PE also depends on the tropospheric background NO_2. In our method, main factors affecting this factor are the emissions inventory and the amount of transported NO_2. For the emissions inventory, the sources of uncertainty are assumptions, methods, input data and calculation errors. As a result, the uncertainties for different species or pollutants related to NO_2 are different and EPA also doesn't publish the quantified uncertainty measures because the parties that submit emissions estimates to EPA are not asked to include quantitative uncertainty measurements or estimates (EPA, 2015). For the simulated convective transport, Li et al. (2018) compared the cloud-resolving simulations with these based on convective parameterization and pointed out that the convective transport was weaker in the parameterization. But, we believe that the ratio condition ($LNO_2Vis/NO_2Vis \geq 50\%$) should reduce these two kinds of uncertainty and assume an uncertainty of 10%, which is less than 20% assigned in Allen et al. (2019) and Bucsela et al. (2019).

The overall uncertainty is estimated as the square root of the sum of the squares of all individual uncertainties in Table 5. The net uncertainty is 48% and 56% for LNO_2 type and LNO_x type respectively. The mean LNO_2/flash, LNO_x/flash, LNO_2/stroke, LNO_x/stroke based on the linear regression and summation method are 32 mol/flash, 90 mol/flash, 6 mol/stroke and 17 mol/stroke. Applying the corresponding uncertainty to these mean values, we arrive at 32 ± 15 mol LNO_2/flash, 90 ± 50 mol LNO_x/flash, 6 ± 3 mol LNO_2/stroke and 17 ± 10 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 PE of 100 – 250 mol/flash which is higher than but overlaps with our estimate. Pickering et al. (2016) estimated LNO_x PE to be 80 ± 45 mol per flash for the Gulf of Mexico. This is 50% smaller than our flash-based results over the CONUS, if we use the same linear regression method which is based on the daily summed values instead of daily mean values. 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. (2020) yields lower LNO_2 PE of 1.6 ± 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 ± 110 moles (122% larger than our results) LNO_x produced per flash over the North America, this is related to the different algorithm, lightning data and lightning thresholds.

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. It works in both clean and polluted regions because of the consideration of tropospheric background pollution in the definition of AMFs. 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 production efficiencies over the CONUS. Considering all the uncertainties (Table 5) and applying the summation and regression method, the final mean production efficiencies are estimated to be 32 ± 15 mol LNO_2/flash, 90 ± 50 mol LNO_x/flash, 6 ± 3 mol LNO_2/stroke and 17 ± 10 mol LNO_x/stroke.

Compared with Lapierre et al. (2020), we find that the LNO_2 production could be larger when the below-cloud LNO_2 is taken into account, especially for the high clouds. Meanwhile, if the method of Pickering et al. (2016) is applied without the background NO_2 correction, the derived LNO_x production efficiency is similar to ours in clean regions or regions with high LNO_2 concentration above the cloud, but it could be overestimated more than 18% in polluted regions. Finally, implementing profiles generated with different model settings of lightning (1×200 mol NO flash^{-1} and 2×500 mol NO flash^{-1}), we find that the larger LNO production setting leads to 62% larger retrieval of LNO_x on average despite some regionally dependent effects caused by the nonlinear calculation of AMF. Both the ratio of the tropospheric LNO_2 above the cloud to the total tropospheric LNO_2 and the ratio of LNO_2 to NO_2 cause different comprehensive effects due to the nonlinear calculation of AMF_{LNO_2} and AMF_{LNO_x}.

Since other regions, like China and India, have much more NO_2 pollution 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 scales.

Code and data availability. The retrieval algorithm used in Sect. 2.4 is available at https://github.com/zxdawn/BEHR-LNOx (last access: March 8, 2020; 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

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

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

where f_{r} is the cloud radiance 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.

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

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

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

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

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

Appendix B: LNO_x Production based on Lower Lightning Thresholds

While we used 2400 flashes box^{-1} and 8160 strokes box^{-1} per 2.4 hour time window for detecting LNO_x, here we show results obtained when using 1 flash box^{-1} and 3.4 strokes box^{-1} in the same time window. We note that the WRF total lightning threshold is also reduced to 1 flash box^{-1}, but we keep the ratio condition unchanged. Briefly, the condition is CRF90_ENTLN1(3.4)_TL1_ratio50 as shown in Table 1.

Similarly, the order of estimated daily PEs is LNO_2Clean > LNO_2 > NO_2Vis > LNO_2Vis (Fig. B1). Compared with Fig. 4, the LNO_2 per flash and LNO_x per flash are larger while PEs based on stroke data are smaller. Considering the additional boxes of fewer lightning counts, differences in the daily mean flashes and NO_x results in different PEs and the relationship presents more like the power function as mentioned in Bucsela et al. (2019).

Instead of using the nonlinear regression of power function:

$$y = \alpha x^{\beta} \tag{B1}$$

where x is flashes or strokes and y is NO_2 or NO_x, we take the logarithm of both sides and apply the linear regression to data:

$$\log_{10} y = \log_{10}\alpha + \beta \log_{10} x \tag{B2}$$

As expected, the linear regression based on logarithmized data performs better in this situation and yields α = 38 kmol, and β = 0.3 for LNO_x per flash (Fig. B2). Since we use the unbinned data (flashes not divided into many groups), we compare our results with Bucsela et al. (2019) based on the same kind of data (α = 10.3 kmol, and β = 0.42). The large difference of α is related to the method of estimating LNO_x, different lightning data (WWLLN and ENTLN) and different regions (northern midlatitudes and CONUS). Note that the resolution (13×24 km^2) of OMI could weaken the signal of LNO_x. We believe the phenomenon of higher production efficiency as flash rate decreases (Fig. B3) could be explored in much detail with higher resolution data like the TROPOMI data.

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.

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

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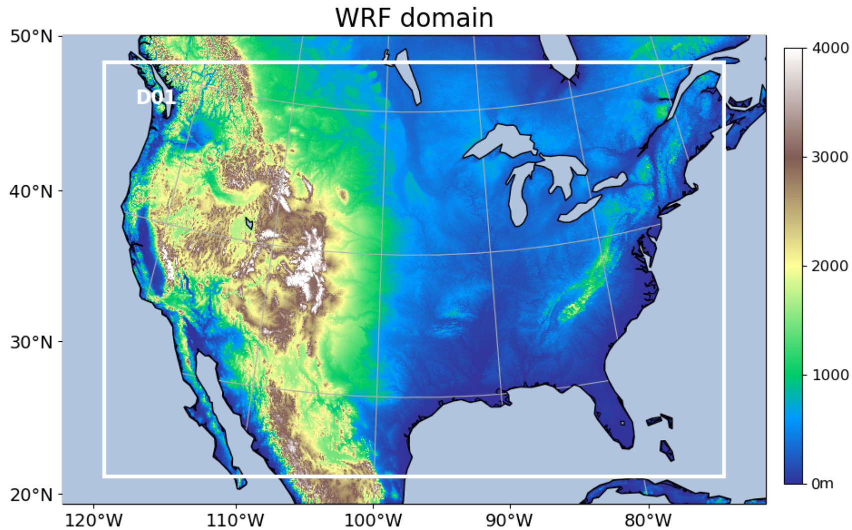

Figure 1. Domain and terrain height (m) of the WRF-Chem simulation with 350 x 290 grid cells and a horizontal resolution of 12 km.

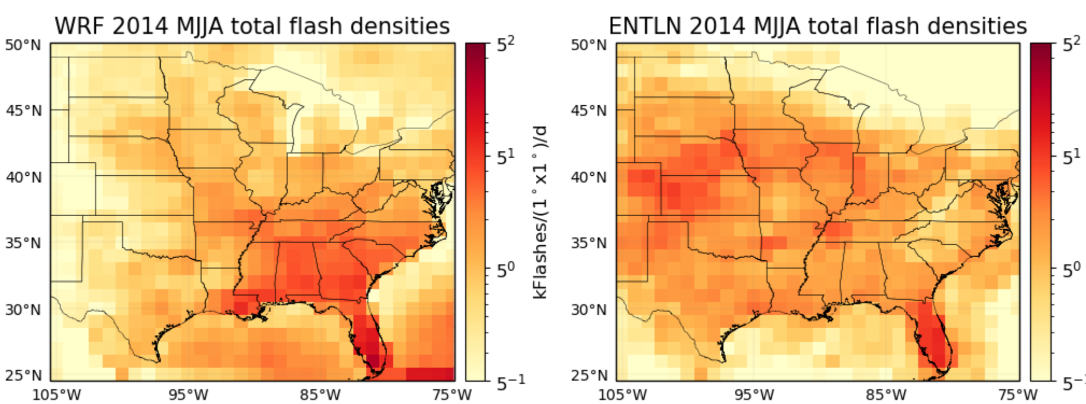

Figure 2. Comparison between total flash densities from ENTLN and WRF-Chem during MJJA 2014.

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%
CRFα_ENTLN1(3.4)_TL1_ratio50	CRF $\geq \alpha$ + ENTLN flashes(strokes) $\geq$ 1(3.4) + TL $\geq$ 1 + ratio $\geq$ 50%

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

Table 2. LNO_x production efficiencies for different combinations of criteria defined in Table 1.

Condition[1]	**ENTLN data type**[2]	**LNO_x/flash or LNO_x/stroke**	**R value**	**Intercept (10^6mol)**	**Days**[3]
CRF90_ENTLN	Flash	52.1 ± 51.1	0.20	0.21	99
CRF90_CF40_ENTLN	Flash	84.2 ± 31.5	0.54	-0.04	70
CRF90_ENTLN_TL1000	Flash	61.9 ± 49.1	0.27	0.33	83
CRF90_CF40_ENTLN_TL1000	Flash	63.4 ± 52.9	0.38	0.26	38
CRF90_ENTLN_TL1000_ratio50	Flash	54.5 ± 48.1	0.25	0.39	81
CRF90_CF40_ENTLN_TL1000_ratio50	Flash	90.0 ± 65.0	0.46	0.15	32
CRF90_ENTLN	Stroke	6.7 ± 4.1	0.31	0.23	102
CRF90_CF40_ENTLN	Stroke	10.3 ± 3.6	0.55	0.08	79
CRF90_ENTLN_TL1000	Stroke	7.5 ± 5.1	0.29	0.38	94
CRF90_CF40_ENTLN_TL1000	Stroke	8.6 ± 6.2	0.39	0.27	46
CRF90_ENTLN_TL1000_ratio50	Stroke	7.0 ± 4.8	0.29	0.42	93
CRF90_CF40_ENTLN_TL1000_ratio50	Stroke	8.9 ± 7.0	0.39	0.31	40

[1]These conditions are defined in Table 1. [2]The thresholds of ENTLN data are 2400 flashes box^{-1} and 8160 strokes box^{-1} during the period of 2.4 h before OMI overpass time. [3]The number of valid days with specific criteria in MJJA 2014.

Table 3. LNO_x production efficiencies for different thresholds of CRF with coincident ENTLN data, TL $\geq$ 1000 and ratio $\geq$ 50%.

CRF (%)	ENTLN data type[1]	LNO_x/flash or LNO_x/stroke	R value	Intercept (10^5mol)	Days[2]
70	Flash	35.7 ± 36.8	0.21	4.91	85
90	Flash	54.5 ± 48.1	0.25	3.90	81
100	Flash	20.8 ± 37.4	0.13	5.67	71
70	Stroke	4.1 ± 3.9	0.21	5.16	96
90	Stroke	7.0 ± 4.8	0.29	4.16	93
100	Stroke	2.6 ± 4.0	0.14	5.41	82

[1]The thresholds of ENTLN data are 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.

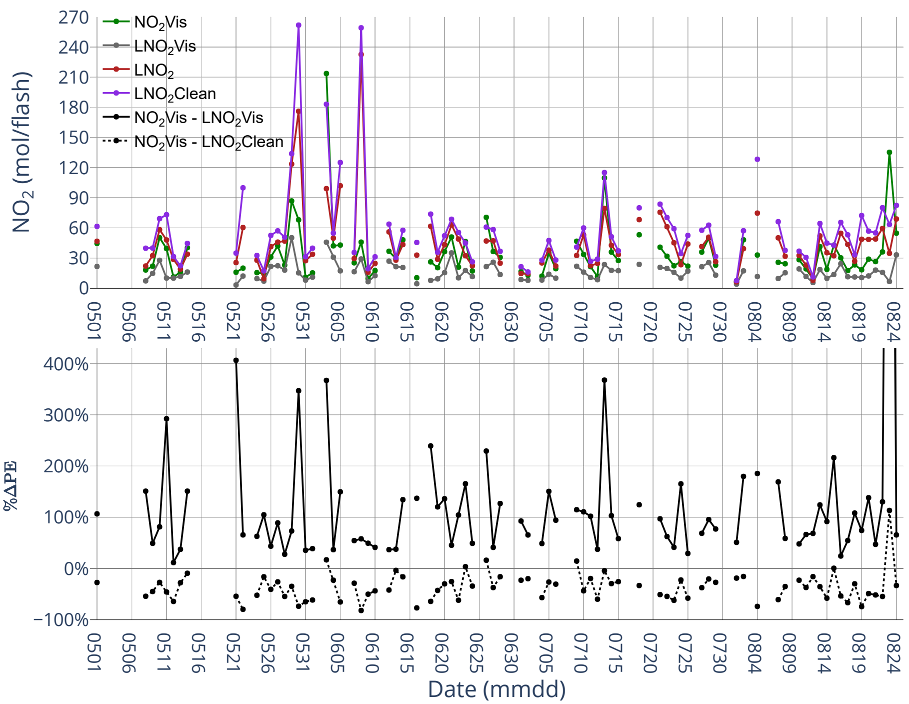

Figure 3. (top) Time series of NO_2Vis, LNO_2Vis, LNO_2 and LNO_2Clean production per day over the CONUS for MJJA 2014 with CRF $\geq$ 90% and a flash threshold of 2400 flashes per 2.4 h. (bottom) Time series of the percent differences between NO_2Vis and LNO_2Vis and the percent differences between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

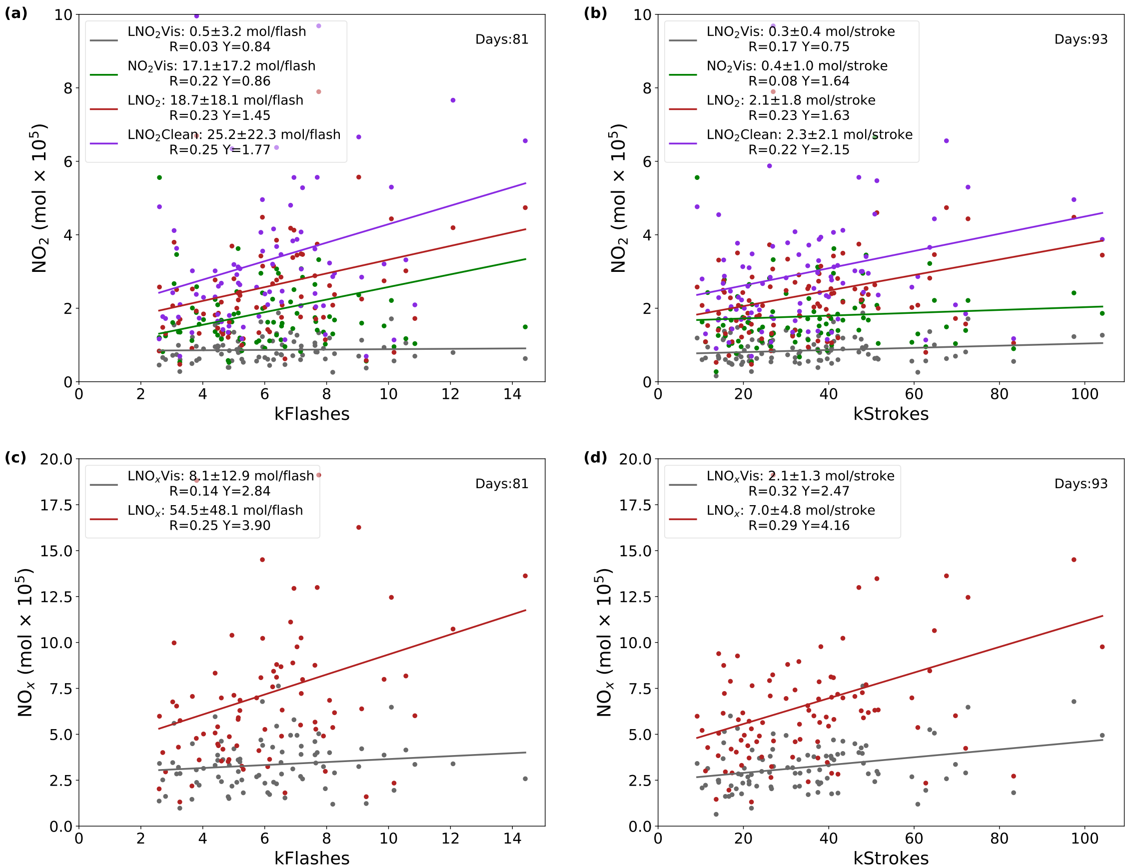

Figure 4. (a) Daily NO_2Vis, LNO_2Vis, LNO_2 and LNO_2Clean versus ENTLN total flashes data. (b) Same as (a) but for strokes. (c) Daily LNO_xVis and LNO_x versus total flashes. (d) Same as (c) but for strokes.

Table 4. The percent changes in the estimated production when using different methods based on the same a priori profiles.

	City[1]	(LNO_2Clean - LNO_2)/LNO_2	(LNO_2 - TropVis)/TropVis	(LNO_2Clean-TropVis)/TropVis
Polluted	Lansing	24.2%	49.5%	85.6%
	New Orleans	13.3%	121.2%	153.8%
	Orlando	46.3%	37.5%	101.3%
Clean	Huron	12.0%	56.4%	75.2%
	Charles Town	12.0%	82.2%	104.1%
	Tarboro	5.0%	86.0%	95.3%

[1] Locations are denoted in Fig. 6a.

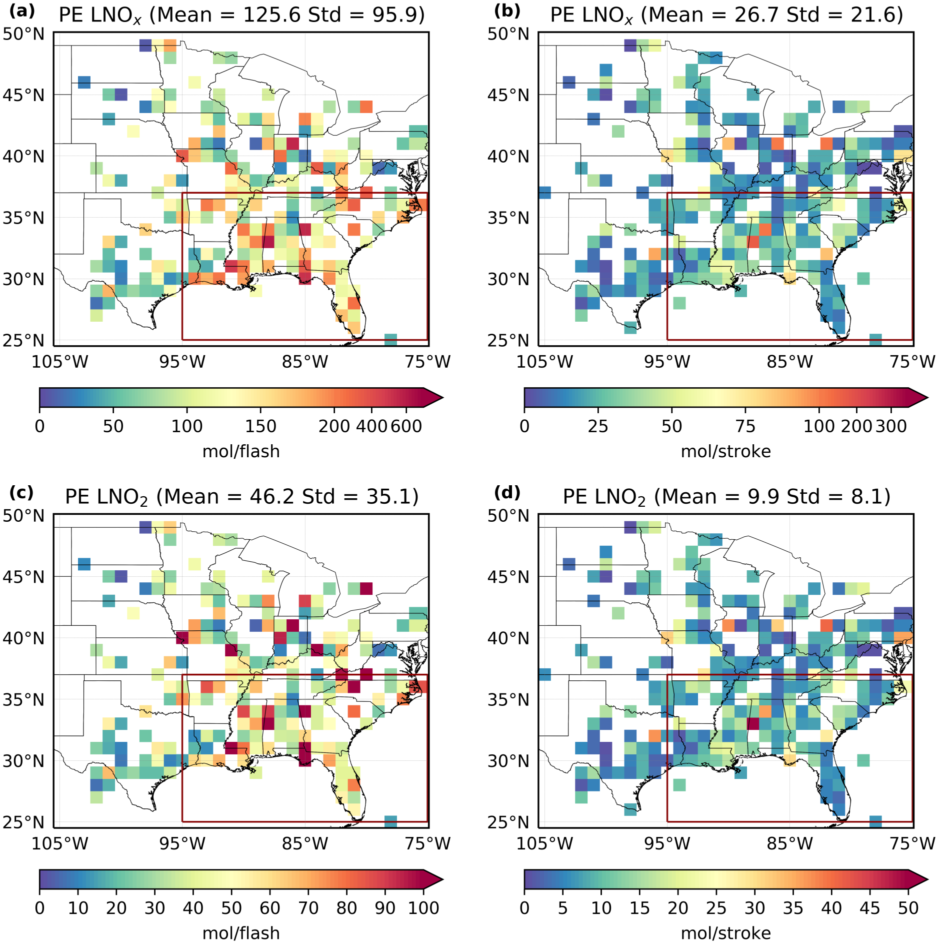

Figure 5. (a) and (c) Maps of 1°× 1°gridded values of mean LNO_x and LNO_2 production per flash with CRF ≥ 90% for MJJA 2014. (b) and (d) Same as (a) and (c) except for strokes. The southeastern US is denoted by the red box in panels a – d.

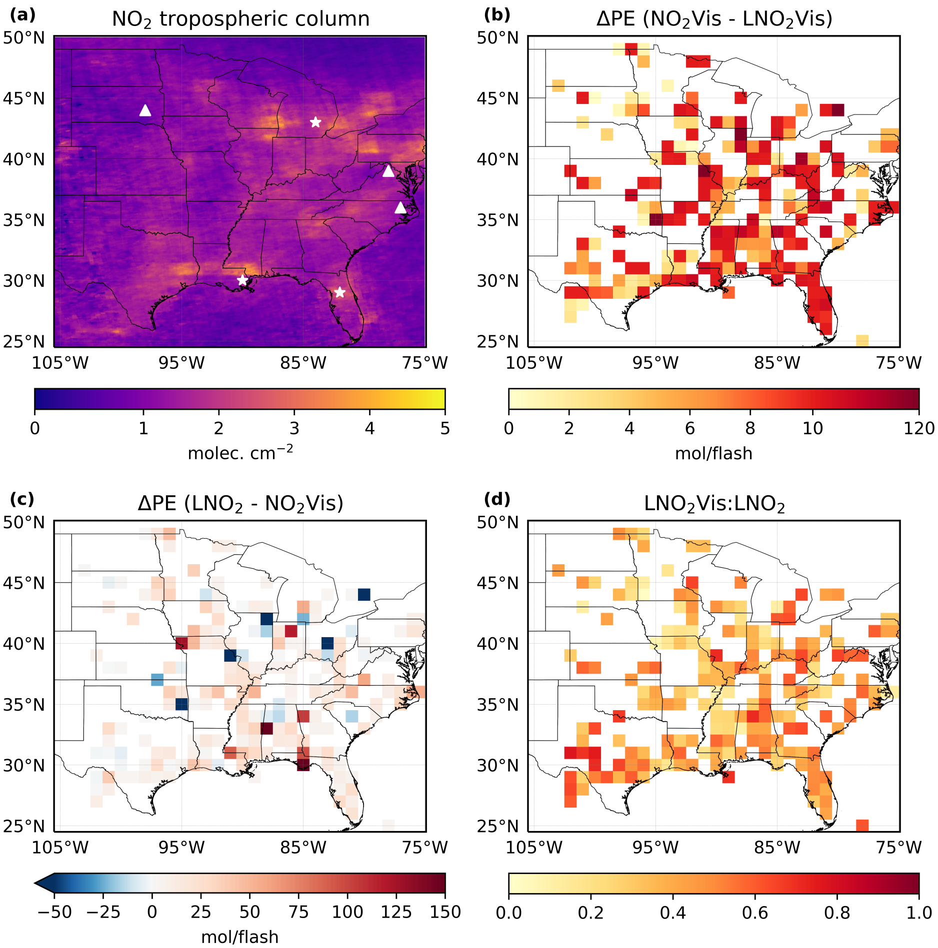

Figure 6. (a) Mean (MJJA 2014) NO_2 tropospheric column. Polluted cities are denoted by stars: Lansing, New Orleans and Orlando while clean cities are denoted by triangles: Huron, Charles Town and Tarboro. (b) The differences of the estimated mean production efficiency between NO_2Vis and LNO_2Vis with 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.

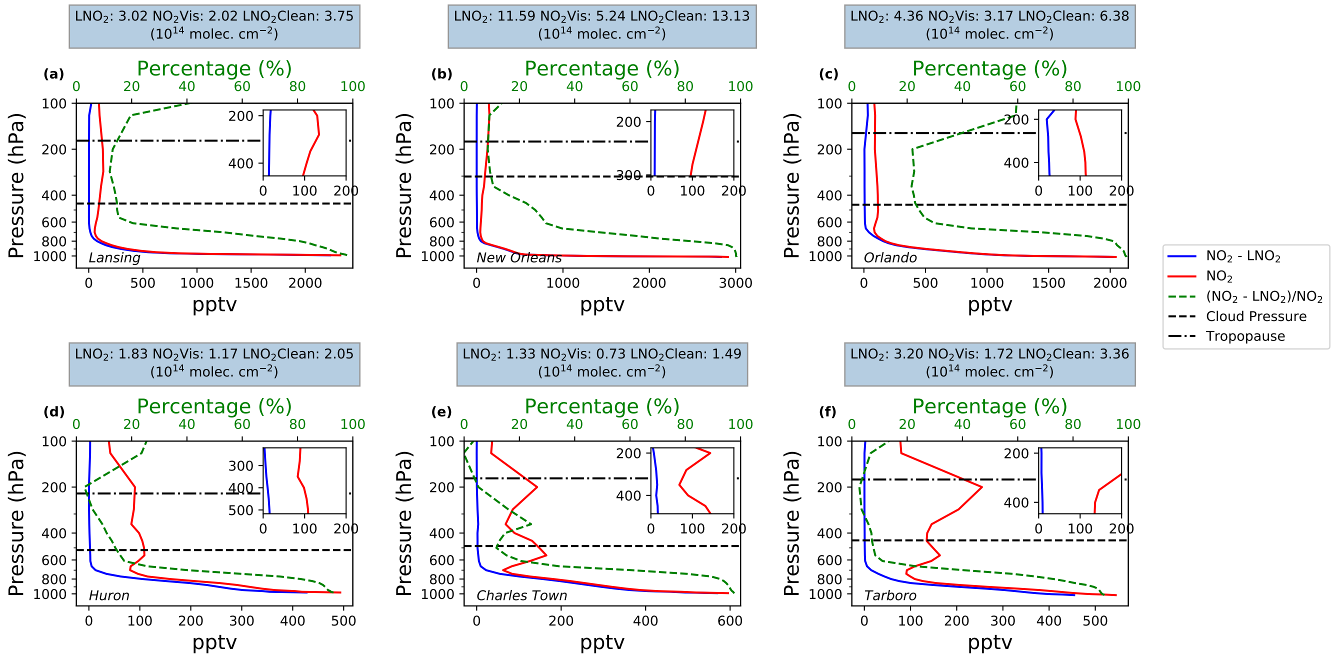

Figure 7. Comparisons of mean WRF-Chem NO_2 and background NO_2 profiles in six grids with CRF $\geq$ 100% on specific days during MJJA 2014. The top row data are selected from polluted regions (stars in Fig. 6a) while the bottom row data are from clean regions (triangles in Fig. 6a). The green dashed lines are the mean ratio profiles of background NO_2 to total NO_2. The zoomed figures show the profiles from the cloud pressure to the tropopause. The titles present the mean productions based on three different methods mentioned in Sect. 2.4.

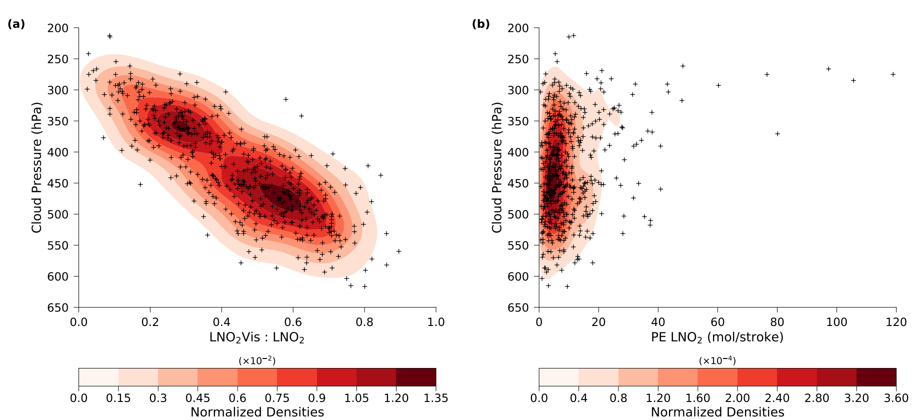

Figure 8. Kernel density estimation of the (a) daily ratio of LNO_2Vis to LNO_2 and (b) daily LNO_2 production efficiency versus the daily cloud pressure measured by OMI with CRF $\geq$ 90% for MJJA 2014. The kernel density estimation was generated by kdeplot in the Python package named seaborn.

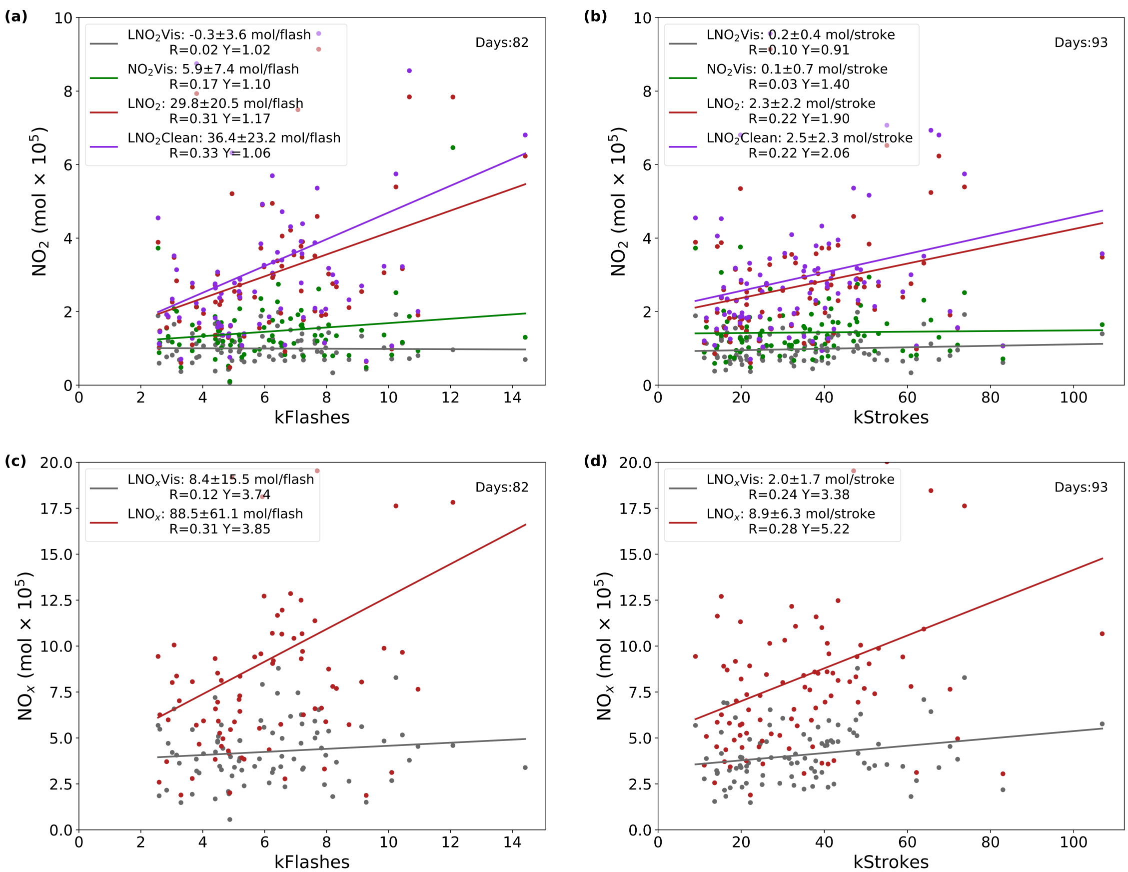

Figure 9. Same as Fig. 4 except for the 2×500 mol NO flash^{-1} configuration.

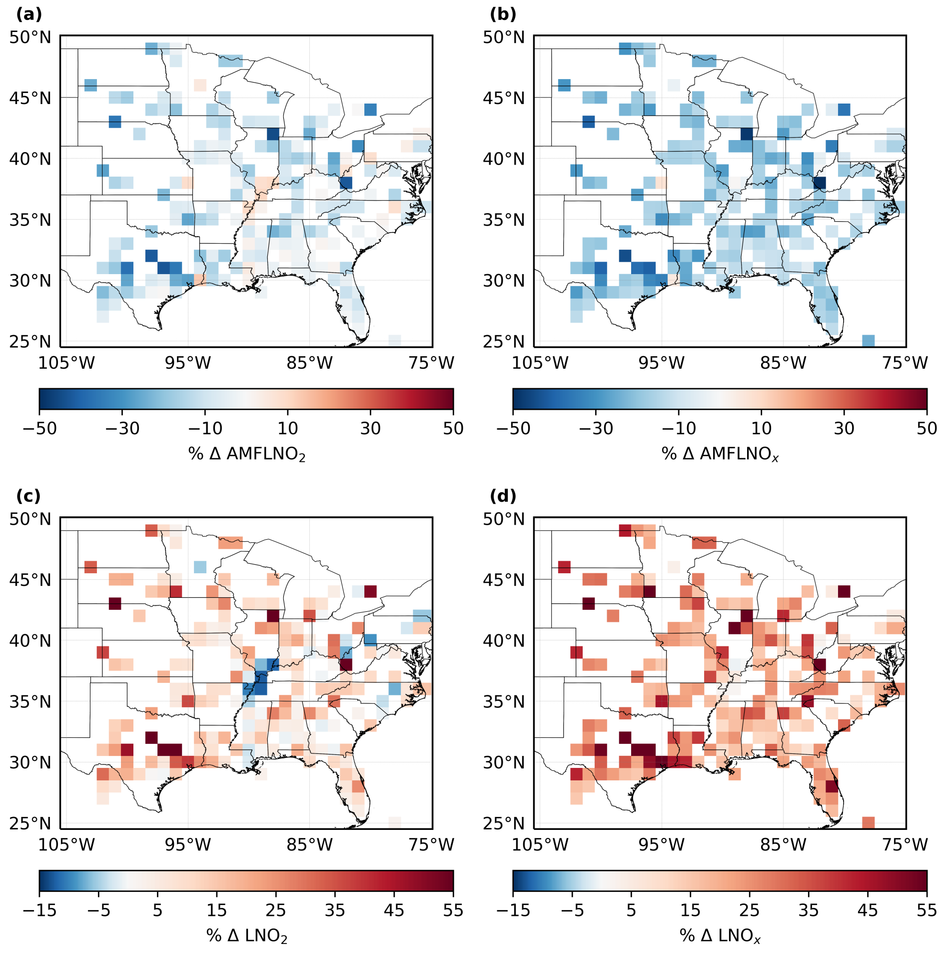

Figure 10. Average percent differences in (a) AMF_{LNO_2}, (b) AMF_{LNO_x}, (c) LNO_2 and (d) LNO_x with CRF $\geq$ 90% over MJJA 2014. Differences between profiles are generated by 2×500 mol NO $flash^{-1}$ and 1×200 mol NO $flash^{-1}$.

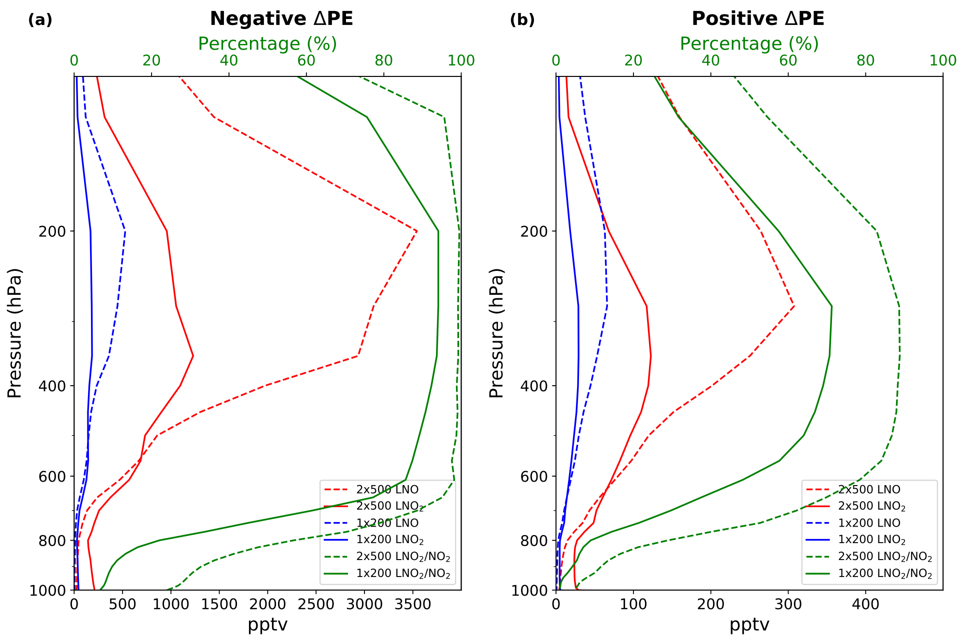

Figure 11. LNO and LNO_2 profiles with different LNO settings at (a) the region containing the minimal negative percent change in LNO_2 and (b) the region containing the largest positive percent change in LNO_2 when the LNO setting is changed from 1×200 mol NO flash^{-1} to 2×500 mol NO flash^{-1}, averaged 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}.

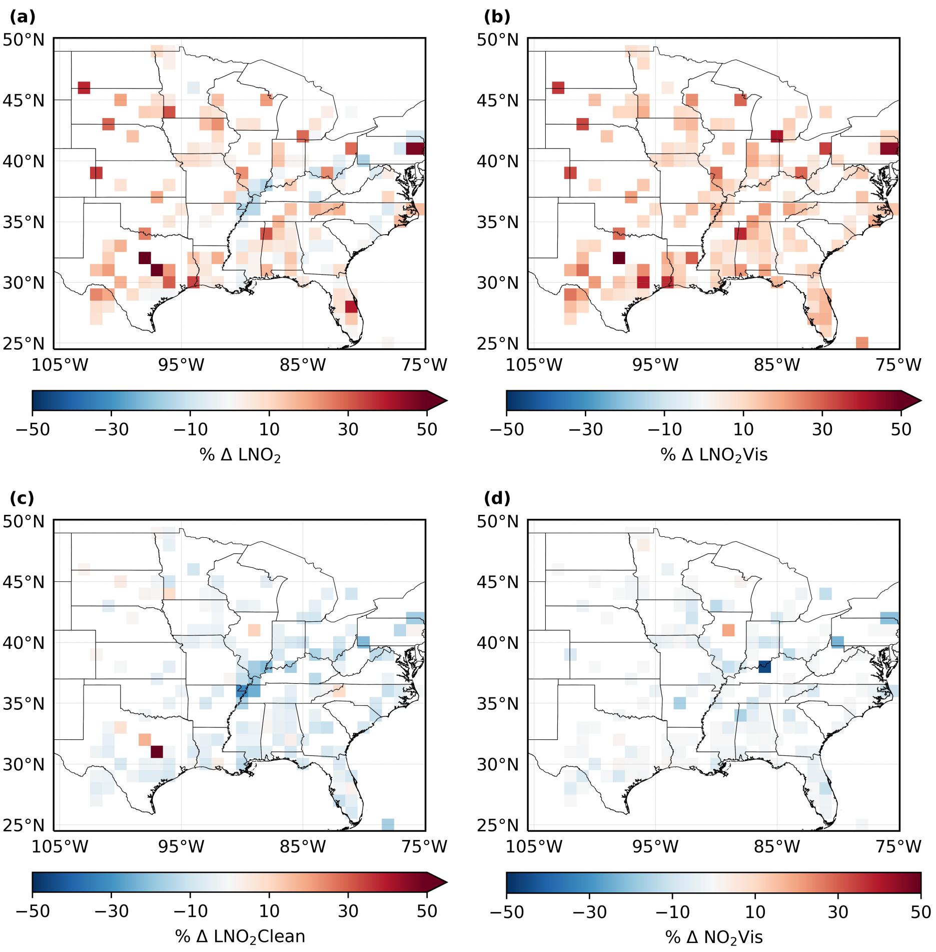

Figure 12. Average percent differences in (a) LNO_2, (b) LNO_2Vis, (c) LNO_2Clean and (d) NO_2Vis with CRF = 100% over MJJA 2014.

Table 5. Uncertainties for the estimation of LNO_2/flash, LNO_x/flash, LNO_2/stroke and LNO_x/stroke.

Type	Perturbation	LNO_2/flash[5]	LNO_x/flash[5]	LNO_2/stroke[5]	LNO_x/stroke[5]
BEHR tropopause pressure[1]	NASA product tropopause	6	4	6	4
Cloud radiance fraction[1]	$\pm$ 5%	2	2	2	2
Cloud pressure[2]	Variable	32	34	32	34
Surface pressure[1]	$\pm$ 1.5%	0	0	0	0
Surface reflectivity[1]	$\pm$ 17%	0	0	0	0
LNO_2 profile[1]	2$\times$500 mol NO $flash^{-1}$	13	25	13	25
Profile location[1]	Quasi-Monte Carlo	0	1	0	1
Lightning detection efficiency[3]	IC: $\pm$ 16%, CG: $\pm$ 5%	15	15	15	15
t_{window}[3]	2 – 4 hours	10	10	8	8
LNO_x lifetime[3]	2 – 12 hours	24	24	24	24
V_{strat}[4]	-	10	10	10	10
Systematic errors in slant column[4]	-	5	5	5	5
Tropospheric background[4]	-	10	10	10	10
NO/NO_2[4]	20% $\pm$ 15%	0	15	0	15
Net	-	49	56	48	56

$PE_{uncertainty}$ = ($Error_{rising\ perturbed\ value}$ - $Error_{lowering\ perturbed\ value}$)/2 where $Error_{perturbed\ value}$ = ($PE_{perturbed\ value}$ - $PE_{original\ value}$)/$PE_{original\ value}$.
[1] Laughner et al. (2019) [2] Acarreta et al. (2004) [3] Lapierre et al. (2020) [4] Allen et al. (2019) and Bucsela et al. (2019) [5] Uncertainty (%)

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

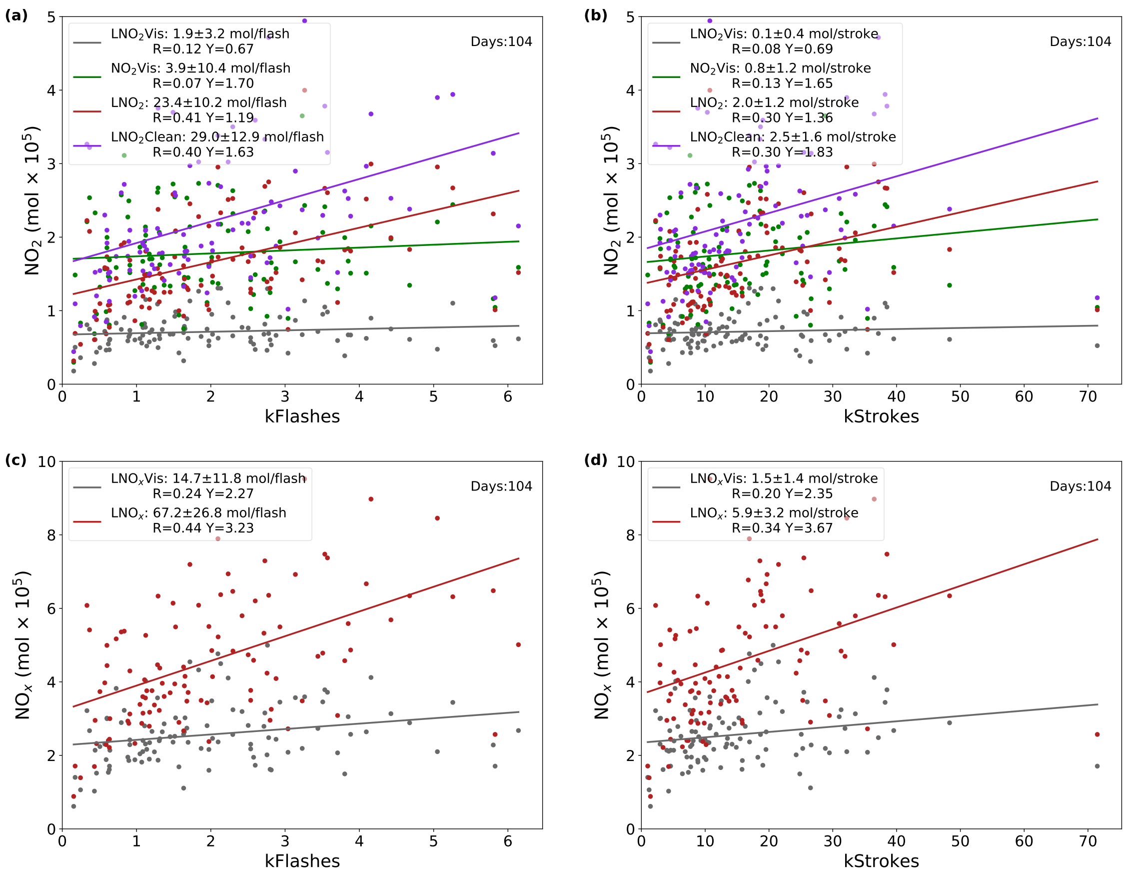

Figure B1. Linear regressions with CRF $\geq$ 90% and a flash threshold of 1 flash box^{-1} or 3.4 strokes box^{-1} per 2.4 h. (a) Daily NO_2Vis, LNO_2Vis, LNO_2 and LNO_2Clean versus ENTLN total flashes data. (b) Same as (a) but for strokes. (c) Daily LNO_xVis and LNO_x versus total flashes. (d) Same as (c) but for strokes.

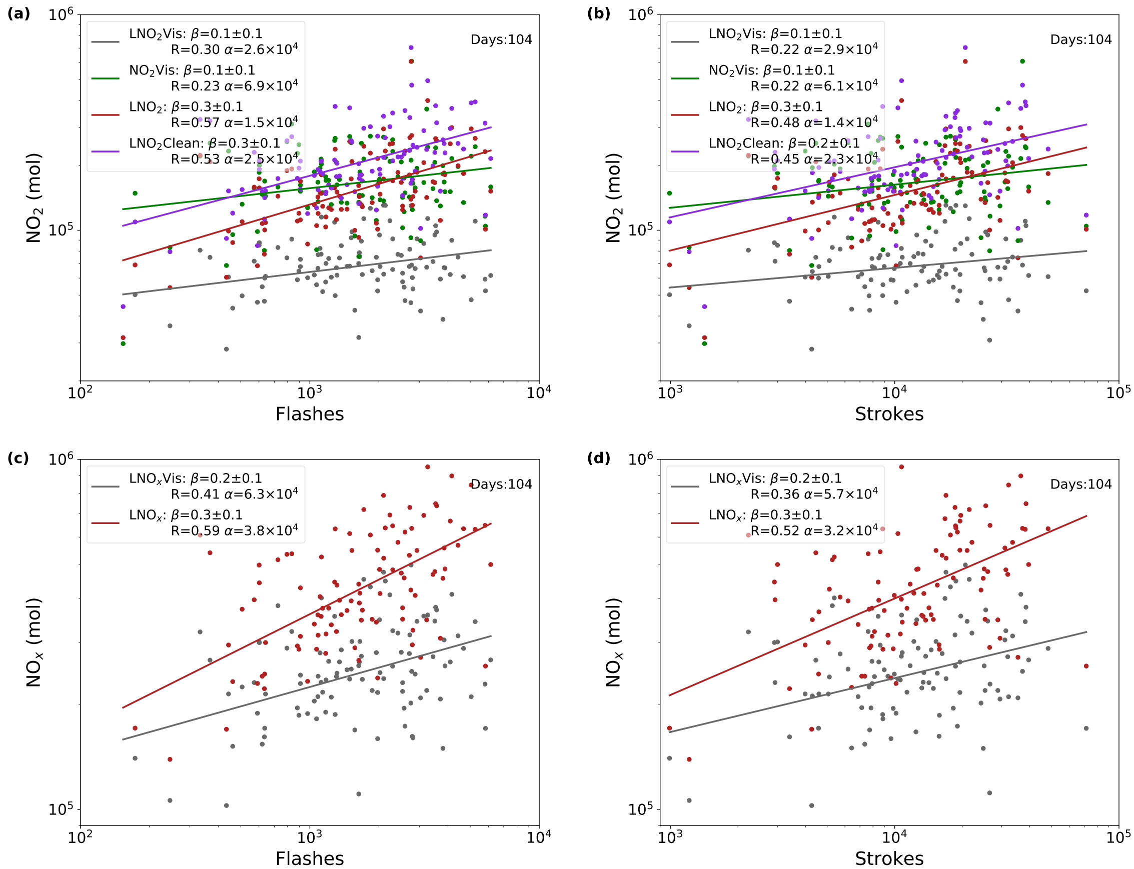

Figure B2. Same as Fig. B1 but using log-log axes.

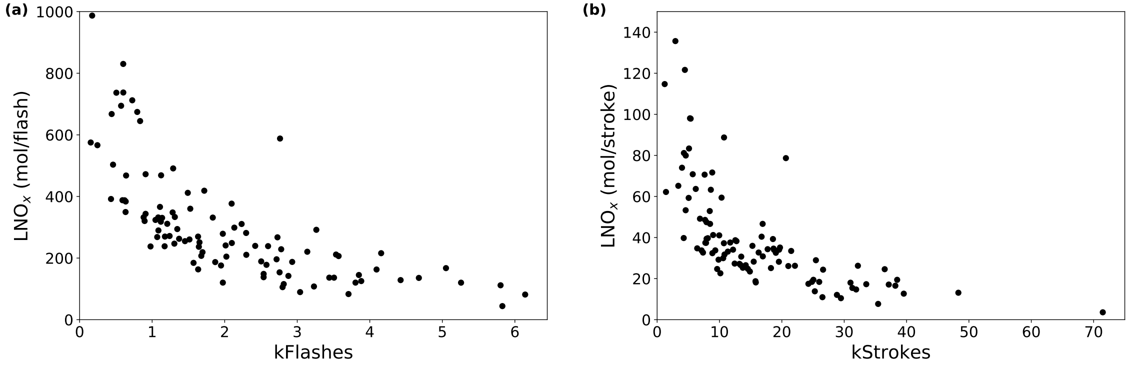

Figure B3. (a) Daily LNO_x production efficiencies versus ENTLN total flashes data, with CRF $\geq$ 90% and a flash threshold of 1 flash box^{-1}. (b) Same as (a) but for strokes.