Estimates of Lightning NO$_x$ Production based on High Resolution OMI NO$_2$ Retrievals over the Continental US

Response to Anonymous Referee #1

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We thank the reviewer for his/her positive comments and very careful reading of the main article. The individual corrections suggested are addressed below. The reviewer's comments will be shown in **red**, our response in **blue**, and changes made to the paper are shown in **black** block quotes. Unless otherwise indicated, page and line numbers correspond to the original paper. Figures, tables, or equations referenced as "Rn" are numbered within this response; if these are used in the changes to the paper, they will be replaced with the proper number in the final paper.

General Comments

1) When revising the manuscript, the authors should emphasize why they believe their approach is less sensitive to the tropospheric background than other approaches and why it may be more suitable over polluted locations.

The Section 3.2 compares the different LNO_x productions based on different AMFs. The differences are partly caused by the different consideration of tropospheric background NO_2. To make it clearer, we add another section to discuss the improved background NO_2 consideration in our approach.

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 (~ 100 pptv) above the cloud with CRF = 100%. These grid boxes contain the cities denoted by stars and triangles in Fig. 6a. Then, the differences between AMFs are dependent on fewer parameters:

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

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

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

Figure 7 (see Fig. R1) compares the mean profiles of NO_2, background NO_2 and background NO_2 ratio in polluted and clean grids. Generally, the profiles of background NO_2 ratio are C-shape because LNO_2 concentrations are higher than background NO_2 in the UT. 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 (see Table R1) presents the relative changes among three methods in six cities. The difference between $AMF_{\mathrm{LNO_2}}$ (Eq. 7) and $AMF_{\mathrm{LNO_2 Clean}}$ (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 convections over polluted locations. In contrast, production estimated by our method is larger than that based on NO_2 Vis 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_2 Vis, which has been discussed in Sect. 3.2. The relative change between $AMF_{\mathrm{LNO_2 Clean}}$ (Eq. 9) and $AMF_{\mathrm{NO_2 Vis}}$ (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 is 153.8% among the six grids where the highest clouds occur.

2) They also need a more clear rationale for how they determined what detection efficiency to use for the ENTLN flashes.

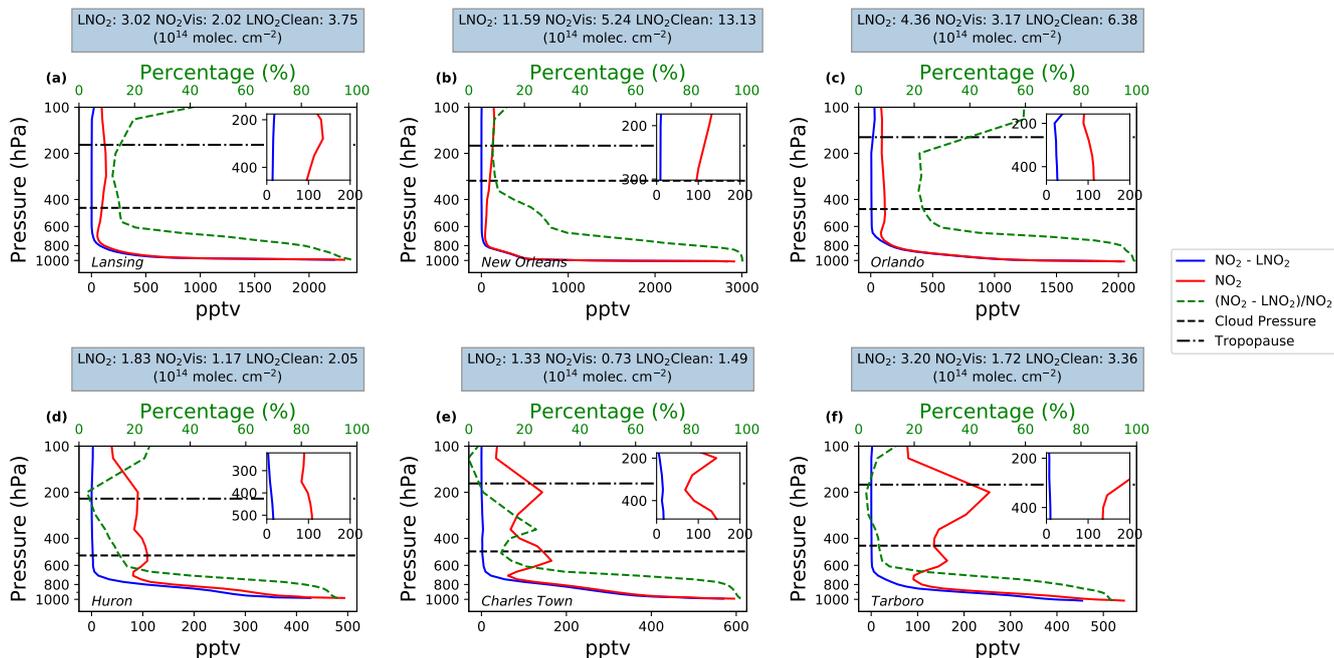Thanks. We have made it clearer (See *No. 11 – 13, Sect. Specific comments*).

Figure R1 . Comparison 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 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.

Table R1 . The percent change 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.

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Specific Comments

1) L10: Put a positive spin on your method as opposed to a negative spin on others work, i.e. mention that your method reduces sensitivity to the background and includes much of the below-cloud NO_x.

Thank you. We have reformatted the sentence:

> 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 our method reduces sensitivity to the background NO_2 and includes much of the below-cloud LNO_2.**

2) L30: Why does an increase in lightning lead to a net positive forcing but a decrease in lightning as no net effect?

Because the effect of CTH and IFLUX on radiative forcing from ozone and methane is different, the net effect should not be determined only by lightning. The detail of this is shown by Fig. 3 in Finney et al. (2018). To make this more clear, we have added the "radiative forcing" specifically.

> Finney et al. (2018) found different impacts on atmospheric composition and radiative forcing between when simulating future lightning using a new upward cloud ice flux (IFLUX) method and when the commonly used cloud-top height (CTH) approach is used. While 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 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).**

3) L63: "Since they assumed NO_2 above the clouds are all NO_x". I think this is misleading. Pickering et al. (2016) did not assume all NO_2 above the clouds was LNO_x. They assumed it included LNO_x and BL NO_x from pollution. They subtracted off a tropospheric background to account for the latter.

Although the tropospheric background NO_2 is subtracted as shown by Eq. 1 in Pickering et al. (2016), they didn't apply it to the rest of analysis. As explained by the fifth paragraph in Sect. 3.4 and the fifth paragraph in Sect. 4.4 (Pickering et al., 2016): "Given the difficulty in estimating the background, we focus on areas of active or very recently active convection where the lower tropospheric background is minimized." and "In our calculations of LNO_x in sections 4.1 through 4.3 we have assumed a zero tropospheric background due to uncertainty in how it should be determined.". To make it more clear, we have rewritten the sentence:

> 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 considered NO_2 above the cloud as LNO_2 in the algorithm due to the difficulty and uncertainty in determining of the background NO_2, their AMF and derived VCD of LNO_x (LNO_2) is named as $AMF_{LNO_x Clean}$ ($AMF_{LNO_2 Clean}$) and $LNO_x Clean$ ($LNO_2 Clean$), respectively.**

4) L68: The $3 - 30\%$ value is an estimate of $LNO_x/(LNO_x+NO_x$ from other sources). It is not an estimate of the uncertainty. Move the $3 - 30\%$ parentheses to after "background NO_x".

Moved.

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 exists in characterizing background NO$_x$ (3% $\sim$ >30%) in this region (Pickering et al., 2016).**

5) L72: What do you mean by "weighted"?

"Weighted" means that box data are weighted according to the number of OMI pixels contributing to each. We have added the explanation of "weighted":

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 subtracting 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 has 1 flash or less instead.**

6) L73: You need to specify whether the 15% adjustments were positive or negative also the "lofted pollution" was not corrected the "LNO$_x$ production" was corrected.

They considered 15% of total NO$_x$ is the transported pollution. Fixed.

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 at those locations.

7) L86: You may want to add that "Pickering et al. then subtracted off an estimate of the tropospheric background to obtain the LNO$_x$ Production.

As mentioned before (*Response 3, Sect. Specific Comments*), they did not subtract off the background NO$_2$.

8) L93: The bias of AMF relative to what?

The bias of AMF is related to aircraft observations. Fixed.

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.**

9) L106: Verify whether you used v3.0 or v3.1 of the NASA standard product

Verified.

For the current study, we used the NASA standard product **V3.0** (Krotkov et al., 2017) as input to the LNO$_x$ retrieval algorithm.

10) L110: The bias with respect to what is subtracted off?

The bias is caused by an instrument artifact. Corrected.

A corrected ("de-striped") SCD is obtained by subtracting **the cross-track bias caused by an instrument artifact** from the measured slant column;

11) L127: What region did Rudlosky analyze? Are Rudlosky's value for CG, IC, or combined flashes? Are you using ENTLN flashes or flashes from a combined ENTLN/NLDN data set? The DE values found by Rudlosky of 35% or so need to be reconciled with the 88%/45% values found by Lapierre.

Thanks for your comments on lightning data. We have changed the DE values to focus on CONUS and specified the kind of ENTLN data.

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**.

12) L121-131: Why is the NLDN DE of IC pulses relevant?

Because we only used ENTLN data instead of the combined ENTLN and the NLDN dataset, it is necessary to clarify that the NLDN DE of IC pulses is lower than ENTLN's and ENTLN DE of CG flashes or strokes is high enough to keep unchanged.

Lapierre et al. (2019) also compared combined ENTLN and the NLDN dataset with data from the LIS during 2014 and the detection efficiencies of IC flashes and strokes are 88% and 45%, respectively. **Since we only 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.

13) L121-131: You may want to include a plot showing the mean ENTLN flashes during MJJA of 2014.

Added (Fig. R2).

In addition, lightning flash rate based on the Price and Rind level of neutral buoyancy parameterization (Price and Rind, 1992) 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}"). **Although the simulated total flash densities are higher in the Southeast US and lower in the North Central US (Fig. 2), the criteria in Sect. 3.1 could limit this effect on the estimation of LNO_x production and Sect. 3.4 will use another simulation to test this problem.**
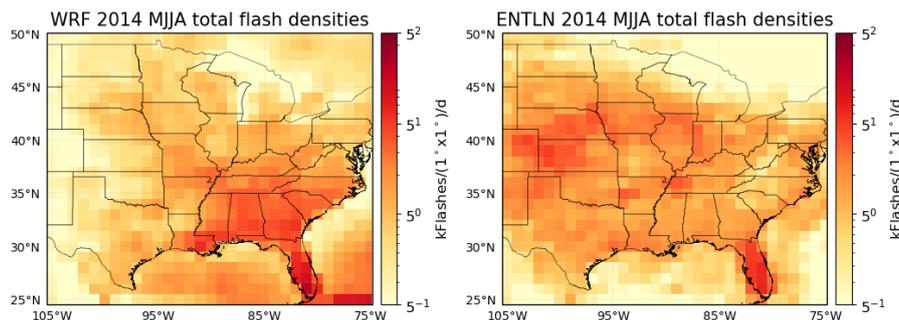

Figure R2 . Comparison between total flash densities from ENTLN and WRF-Chem during MJJA 2014.

14) L174: LNO$_x$ is regridded. At this point in the manuscript it is unclear how you generated pixel level values of LNO$_x$ that were regridded. Start paragraph by saying how you determined LNO$_x$ and then mention regridding method. Is it as simple as multiplying VLNOx by area?

Sorry for the misleading. This paragraph describes how to re-grid the V_{LNO_x} pixel values obtained in Section 2.4 to $0.05° \times 0.05°$ grids.

> V_{LNO_x} **is re-gridded to $0.05° \times 0.05°$ grids using the constant value method (Kuhlmann et al., 2014)**. Then, it is analyzed in $1° \times 1°$ grid boxes with a minimum of fifty valid $0.05° \times 0.05°$ grids which is equivalent to five satellite pixels in Pickering et al. (2016).

15) L181: Why does choosing the 350-400 hPa layer avoid biases in the simulation of high clouds?

If the model has biases in the 3D cloud fraction in the upper troposphere (higher than 350 hPa), our method is not sensitive to that bias since we don't consider the cloud fraction at levels above 350 hPa. This pressure layer was also used in Strode et al. (2017). We have added the reference for the method.

> The CFs are 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 CFs $\geq$ 40% suggested by Strode et al. (2017) to determine cloudy or clear for each simulation grid.

16) L193: I don't think "not polluted much" is appropriate for a 50% threshold. Simply state that more than half of the NO$_x$ must have an LNO$_x$ source.

Fixed.

> The ratio $\geq$ 50% indicates that more than half of the NO$_x$ above the cloud **must have a LNO$_x$ source**.

17) L216-220: Why do you think the production is relatively insensitive to the CRF threshold? Wouldn't you expect it to decrease as the CRF is increased due to less BL contamination?

Yes, it is true for the method of Pickering et al. (2016) and Lapierre et al. (2019). But, in our new method, the BL contamination is considered in the numerator of AMF$_{LNO_x}$ (Eq. 2). As a result, when the CRF is increased from 70% to 90%, the LNO$_x$ PE rises from 35.7 LNO$_x$/flash to 54.5 LNO$_x$/flash, which is opposite compared to the result of Pickering et al. (2016). On the other hand, the updraft would also affect the BL contamination detected by OMI. We have added these information to make the choice of CRF more reasonable:

> **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. (2019).

18) L228: 10-80% range is so wide as to be useless. In addition to this range include the mean and standard deviation.

Thank you for your remind. We have added the mean and standard deviation of the ratio of LNO$_x$ below the cloud.

> 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 **$56 \pm 20\%$**.

19) L266: Are the differences between NO_2Vis production and LNO_2Vis production larger over polluted grid boxes?

Yes. We mentioned that in L265.

20) L271: Not sure what you mean by "trend". Please re-write this sentence more clearly.

Clarified.

> ... 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 also be smaller** while peaks of the LNO_2 profile are below the CP.

21) L295-L300: State clearly why the difference is larger for LNO_x (25%) than for LNO_2 (11.5%)?

As suggested by Eric J. Bucsela in private, we have redone the linear regressions and stated the results instead of comparing the difference in LNO_2 PE and that in LNO_x PE.

> For the linear regression method (Fig. 9), LNO_2 production 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 production (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.

22) L300: I'd suggest deleting Figure 10. It is discussed in passing and the most important numbers from that Figure are already included in the text.

We agreed with you and have deleted that figure.

23) L334: Would the profile uncertainty be less than 13/26% for the LNO_2Clean approach, although the latter (apparently) has more uncertainty due to the tropospheric background?

We have checked the profile uncertainty of retrieved LNO_2 and LNO_x productions for the LNO_2Clean approach to be $\sim 0\%$ and 39% respectively. As a result, the LNO_2Clean approach is suitable for clean regions and the effect of profile need to be considered carefully.

24) L335-338: Why don't uncertainties in the tropospheric background contribute to uncertainties in the production?

Yes, the tropospheric background NO_2 should affect the uncertainties in the estimation of production. We have added this to the section of uncertainty.

> In addition, the estimation of LNO_x PE is also dependent 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 ($LNO_2 Vis/NO_2 Vis \geq 50\%$) 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).

25) L335-338: How would biases in the modeled NO/NO_2 ratio contribute to uncertainties in the production of LNO_x?

We have added that kind of uncertainty according to Travis et al. (2016), Silvern et al. (2018) and Bucsela et al. (2019). The uncertainty in the production of LNO_x is 20%.

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 an uncertainty of 20% to this error considering the possible positive NO_2 measurements interferences (Allen et al., 2019; Bucsela et al., 2019).

26) L392: Expand on why this approach can be used in polluted regions? Is this different than other approaches?

Added.

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.**

27) L391-409: Minimize the use to lesser known acronyms in the conclusion section. Many readers will only read the abstract and conclusions. For e.g., TL and ratio and LNO2Clean.

We reformatted the section, thanks.

Compared with former methods, our method has reduced the sensitive to background NO_2, while the method in Lapierre et al. (2019) underestimates LNO_x production efficiency because of the neglected below-cloud LNO_2 and LNO_2 production is overestimated using the method in Pickering et al. (2016) due to the over-cloud background NO_2 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 model setting leads to larger retrieval of LNO_x despite some regionally dependent effects caused by 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}.

28) L401-402: The last sentence of the 2nd paragraph of the conclusion section is poorly written. Please re-write to make its meaning clear.

Rewritten as the response above.

29) Figure 1 caption. Please specify what is plotted here. Also, did you choose this domain or is this a standard domain used by EPA or others (e.g., US1 or US2 domain)? How many east-west and north-south grid points in domain?

We chose the simulation domain and the information has been added to the caption of Figure 1 (Fig. R3).
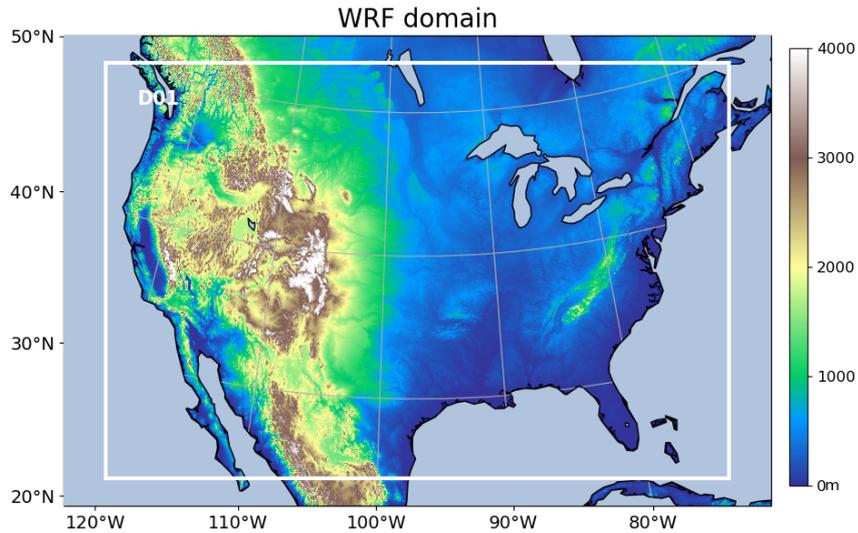
Figure R3 . Domain and terrain height (m) of the WRF-Chem simulation with 350 x 290 grid cells and a horizontal resolution of 12 km.

30) Table 1: Capitalize entln

Fixed (Table R2).

31) Table 2: under different conditions > for different thresholds

Fixed (Table R3).

32) Figure 3: May not be needed. Consider removing.

Figure 3 shows the time series of NO_2Vis, LNO_2Vis, LNO_2 and LNO_2Clean production per day and compares those with the percent differences between NO_2Vis and LNO_2Vis and the percent differences between NO_2Vis and LNO_2Clean. Readers could distinguish the difference between different methods in one figure. For example, these peaks of LNO_2Clean coincide with the NO_2 pollution above the cloud (the peaks of the percent differences between NO_2Vis and LNO_2Vis). As a result, we decide to keep this figure.

Table R2 . 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%

Table R3 . LNO_x production 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 (10^5 mol)	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 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.

Technical Corrections

1) L5: based on the program of new > that uses the

Fixed, thanks.

> 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 product and the Weather Research and Forecasting-Chemistry (WRF-Chem) model.

2) L15: surface is > surface are
L16: originate largely > originates largely

Fixed.

> 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.

3) L27: method and the commonly used the widely used cloud-top > method versus the commonly used cloud-top
L27: As lightning > While lightning
L28: have reported 5 – 16% increases > is predicted to increase by 5 – 16%

Fixed.

> 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** 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 lightning was estimated with IFLUX in 2100 under a strong global warming scenario (Finney et al., 2018).

4) L41: qualified > quantified

Fixed.

> Recent studies have determined and **quantified** LNO_x using satellite observations.

5) L50: it is > this study
L50: without the contribution > and did not consider the contribution

Fixed.

> However, **this study** assumed that all the enhanced NO_2 originated from lightning and **did not consider the contribution** of anthropogenic emissions.

6) L53: Replace sentence beginning with "The threshold of" > Their analysis was restricted to 30×60 km^2 satellite pixels where the flash rate exceeded 1 flash km^{-2} hr^{-1}.
L54: But the results of LNO_x production are highly > But they found LNO_x production to be highly

Fixed.

Their analysis was restricted to 30×60 km² 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 are low in some cases.

7) L72: removed by the weighted > removed by subtracting the weighted
L72: CRF criteria but has > CRF criteria required to be considered deep convection but has

Fixed.

The tropospheric NO$_x$ background was removed by **subtracting** 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 has 1 flash or less instead.

8) L76: at those locations > at some locations

Fixed.

However, there were negative LNO$_x$ values caused by the overestimation of the tropospheric background at **some** locations.

9) L77: On another hand > On the other hand
L79: by strokes > using mean values of strokes

Fixed.

On the other hand, Lapierre et al. (2019) constrained LNO$_2$ to 1.1 ± 0.6 mol NO$_2$/stroke for intracloud (IC) strokes and 10.0 ± 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 **using mean values of strokes** per flash and the ratio of NO to NO$_2$ in the UT.

10) L80: NO$_2$ can be "seen" > NO$_2$ that can be "seen"

Fixed.

They used the regridded Berkeley High-Resolution (BEHR) V3.0A 0.05° × 0.05° "visible only" NO$_2$ VCD (V$_{vis}$) product which includes two parts of NO$_2$ **that** can be "seen" by the satellite.

11) L94: we focused on > we focus on

Fixed.

In this paper, we **focus** on the estimation of LNO$_2$ production per flash (LNO$_2$/flash) ...

12) L99: examines different sources of the uncertainty of > examines the effect of different sources of uncertainty on the

Fixed.

Section 4 examines **the effect of** different sources of the uncertainty **on** the results.

13) L128: and the detection > and found the detection
L128: are 88% > to be 88%
L130: corrected by 88% and 45% > divided by 0.88 and 0.45

Lapierre et al. (2019) 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. (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 **divided by 0.88 and 0.45**, respectively, while CG flashes and strokes are unchanged because of the high detection efficiency.

14) L137: was used to generate > were used to generate

Outputs from the version 4 of Model for Ozone and Related chemical Tracers (MOZART-4; Emmons et al., 2010) **were** used to generate the initial and boundary conditions of chemical species.

15) L170: part of the NO_2 originated from lightning > part of the NO_2 within the cloud originating from lightning

The sensitivity study of Beirle et al. (2009) compared the chemical compositions from the cloud bottom to the cloud top and revealed that a significant fraction the NO_2 within the cloud **originating** from lightning can be detected by the satellite.

16) L180: convections > convection

Furthermore, another criterion of cloud fractions (CFs) is applied to the WRF-Chem results for the successful simulation of **convection**.

17) L184: properties of cloud > cloud properties

Besides **cloud properties**, the time period and sufficient flashes (or strokes) are required for fresh LNO_x detected by OMI.

18) L187: box^{-1} are chosen > box^{-1} per 2.4 hour time window are chosen

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., 2019).

19) L191: sources except LNO_2 > sources in addition to LNO_2

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.

20) L208: search of NO$_2$ > search of the NO$_2$
L206-210: When referring to combinations use the abbreviations given in Table 1.

Rewritten.

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 flashes 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 86.0 ± 14.0 mol/flash to 114.8 ± 18.2 mol/flash. The result is almost the same as that under **the CRF90_ENTLN_TL1000 condition which is without the condition of ratio $\geq$ 50%**.

21) L225: productions are > production values are

Fixed.

LNO$_2$ **production values** are mostly in the range from 20 to 80 mol/flash.

22) L275: 0.8 to 0.2 while the cloud is higher (smaller pressure value) > 0.8 to 0.2 as the cloud pressure decreases from 600 to 300 hPa. Note: Please correct my pressure ranges if incorrect.

Fixed, thanks. The range of cloud pressure is right.

Since the ratio of LNO$_2$Vis to LNO$_2$ decreases from 0.8 to 0.2 **as the cloud pressure decreases from 600 to 300 hPa** ...

23) L279: we can not derive that higher LNO$_2$ production relates to higher clouds > we cannot derive the relationship between LNO$_2$ production and cloud pressure

Fixed.

Because of the limited amount of large LNO$_2$ production and lightning data, we cannot derive **the relationship between LNO$_2$ production and cloud pressure** or different lightning properties at this stage.

24) L284: larger than normal > larger in anomalous than normal

Fixed.

Because updrafts are stronger and flash rates are higher in anomalous storms, UT LNO$_x$ concentrations is **larger in anomalous than normal** polarity storms.

25) L292: chose the same method > chose the same value

Fixed.

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 value** (330 mol NO per flash) that was used by Barth et al. (2012).

26) L313: relative distribution > relative distributions
L314: is similar > are similar
L315: the unrealistic > an unrealistic

Fixed.

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 region dependent and **an** unrealistic profile could appear in the UT.

27) L319: carefully in our research > carefully into consideration

Fixed.

However, the relative distribution of LNO_2 within the UT should be **taken carefully into consideration**.

28) L321: are involved > are important

Fixed.

When the condition of high LNO_2/NO_2 is not met, both relative distribution and ratio are **important**.

29) L401: find that the regionally > find a regionally

Rewritten.

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 in model leads to larger retrieval of LNO_x despite some regionally dependent effects.

30) L403: pollutions than > pollution than

Fixed.

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.

31) L409: level > scales

Fixed.

Applying current method in future studies may enhance the accuracy of LNO_x production at both local and global **scales**.

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Estimates of Lightning NO$_x$ Production based on High Resolution OMI NO$_2$ Retrievals over the Continental US

Response to Anonymous Referee #2

Xin Zhang, Yan Yin, Ronald van der A, Jeff L. Lapierre, Qian Chen,
Xiang Kuang, Shuqi Yan, Jinghua Chen, Chuan He and Rulin Shi

February 7, 2020

We thank the reviewer for his/her positive comments and very careful reading of the main article. The individual corrections suggested are addressed below. The reviewer's comments will be shown in **red**, our response in **blue**, and changes made to the paper are shown in **black** block quotes. Unless otherwise indicated, page and line numbers correspond to the original paper. Figures, tables, or equations referenced as "Rn" are numbered within this response; if these are used in the changes to the paper, they will be replaced with the proper number in the final paper.

Major Comments

The new methods that are discussed should provide better accounting for background NOx; however, it is not made clear in the manuscript how this is achieved. This point needs to be addresses in a revision.

Thank you for your advice. This is the same comment mentioned by Reviewer#1 (See *No. 1, Sect. General Comments*). We have made it clearer in a new section (Sect. 3.3).

Minor Comments

1) Line 10: previous work does not neglect below-cloud LNO$_2$. Full LNO$_2$ and LNO$_x$ profiles extending to the surface are used from the GMI model is such work as Pickering et al. (2016); Bucsela et al. (2019); Allen et al. (2019).

Thank you for your correction. Instead of the negative spin on others' work, we have reformatted the sentence to put a positive spin on our method, as mentioned in the Response to Reviewer#1 (See *No. 1, Sect. Specific Comments*). Because Lapierre et al. (2019) neglected the below-cloud LNO$_2$, it is still necessary to mention that in the summarization of our results.

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 our method reduces sensitivity to the background NO$_2$ and includes much of the below-cloud LNO$_2$.**

2) line 15: ....surface are...
line 16: originates

Corrected.

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.

3) lines 26-27: ...radiative forcing between when simulating future lightning using a new upward cloud ice flux (IFLUX) method and when the commonly used cloud-top height (CTH) approach is used.

Corrected.

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.

4) line 28-29: 5 – 16% increases in what? 15% decrease in what?

These percentage is relative to the current lightning. Corrected.

While **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 lightning** was estimated with IFLUX in 2100 under a strong global warming scenario (Finney et al., 2018).

5) line 30: ....radiative forcing due to ozone was found.... line 31: also need to compare with results of Romps et al. (2014)

We have reformatted the sentence and added the discussion of Romps et al. (2014) and Romps (2019).

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 × P proxy and IFLUX method in cloud-resolving models. They report that higher CAPE and updraft velocities caused by global warming could lead to the large increases in tropical lightning simulated by CAPE × P proxy, while IFLUX proxy predicts little change in tropical lightning because of the small changes in water mass fluxes.

6) line 39: Satellite...

Corrected.

Satellite measurements of NO_2 are a powerful tool compared to conventional platforms, because of its global coverage, constant instrument features and temporal continuity.

7) line 48: convection.

Corrected.

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 **convection**.

8) line 71: ....based on a modification of the method.... Need to describe what is different.

Thank you for your advice. We have added more descriptions about the method of Bucsela et al. (2019).

More recently Bucsela et al. (2019) obtained an average production efficiency (PE) of 180 ± 100 mol per 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.**

9) line 75: Are you sure that it is the background subtraction causing the negatives? Many of the negatives resulted from the removal of the stratospheric slant column NO_2 from the total NO_2 slant column.

Sorry for the incomplete consideration. We have added the effect of overestimated stratospheric slant column NO_2 to the sentence.

However, there were negative LNO_x values caused by the overestimation of the tropospheric background **and stratospheric NO_2** at some locations.

10) line 80: two parts of NO_2 that can be....

Fixed.

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.

11) line 81: ....above clouds (pixels with CRF > 0.9) and....

Fixed.

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.

12) line 88: ....contamination by anthropogenic....

Corrected.

> 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 **by** anthropogenic emissions must be taken into account in detail.

13) lines 129-130: not sure why this phrase about NLDN is here when you are using ENTLN and not NLDN. Remove?

Because we only used ENTLN data instead of the combined ENTLN and NLDN dataset, it is necessary to clarify that the NLDN DE of IC pulses is lower than ENTLN's and ENTLN DE of CG flashes or strokes is high enough to keep unchanged.

> Lapierre et al. (2019) 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. (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 divided by 0.88 and 0.45, respectively**, while CG flashes and strokes are unchanged because of the high detection efficiency.

14) line 142: How is flash rate parameterized?

The flash rate is parameterized by the Price and Rind level of neutral buoyancy parameterization.

> In addition, lightning flash rate **based on the level of neutral buoyancy parameterization (Price and Rind, 1992)** 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}").

15) line 175: how is this comparison of pixels computed?

The area of one satellite pixel is about 312 km^2. The minimum count of 0.5×0.5 degree grids per box is set to 50 ($\sim$ 1300 km^2) which is between 3 satellite pixels ($\sim$ 940 km^2) used in Bucsela et al. (2019) and 5 satellite pixels ($\sim$ 1560 km^2) set in Pickering et al. (2016). In other words, the number of 50 should be enough to minimize the noise. We have written the sentence to make it more clear.

> 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 minimum value is between five satellite pixels in Pickering et al. (2016) and three satellite pixels in Bucsela et al. (2019) or Allen et al. (2019).**

16) line 181: Does the Xu and Randall method consider subgrid convective clouds or only grid-scale cloud based on the microphysics parameterization that generates the gridscale clouds? If it is the latter, this method is not appropriate as a criterion to evaluate model convection.

We applied the Xu and Randall method based on the ice and cloud water fields from both Grell 3D cumulus parameterization and Lin microphysics scheme.

17) line 184: a time period......LNO_x to be detected by

Corrected.

Besides cloud properties, **a** time period and sufficient flashes (or strokes) are required for fresh LNO$_x$ **to be** detected by OMI.

18) lines 184-188: Using these criteria will result in a low bias in the PE results. Bucsela et al. (2019) found that PE is larger at small flash rates. These small flash rates are being discarded here.

Thank you for your remind about the smaller PE at high flash rates. We have added these information at the end of the paragraph.

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.

19) line 203: daily summations Is this what is done in Pickering et al. (2016)?

Yes. But, we discussed with Eric J. Bucsela recently and he suggested us to redo the linear regressions based on the daily mean LNO$_x$ and flashes. As a result, we have updated the plots of linear regression and analysis related to that.

(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).

20) line 218:....(Table 2), as the CRF criterion increases from 70% to 90% and to 100%.

Fixed.

Apart from the fewer valid days under higher CRF conditions (CRF $\geq$ 90% and CRF = 100%), LNO$_x$/flash increases from 109.0 $\pm$ 15.3 mol/flash to 114.8 $\pm$ 18.2 mol/flash and decreases again to 99.4 $\pm$ 15.3 mol/flash while LNO$_x$/stroke enhances from 16.7 $\pm$ 2.6 mol/stroke to 17.8 $\pm$ 2.9 mol/stroke and drops again to 15.6 $\pm$ 3.1 mol/stroke (Table 3)**, as the CRF criterion increases from 70% to 90% and to 100%.**

21) line 223:...to derive production per flash (production efficiency, PE).

Fixed.

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 production **per flash (production efficiency, PE)**.

22) line 228: cloud properties
line 229: PEs

Corrected.

The effect of **cloud properties** on LNO$_x$ production will be discussed in more detail in section 3.3. Generally, the order of estimated daily **PEs** is LNO$_2$Clean > LNO$_2$ > NO$_2$Vis > LNO$_2$Vis.

23) line 259: ratio of CG to IC I don't think there was any particular assumption of this ratio in Pickering et al. (2016).

... possibly due to the differences in geographic location, lightning data and chemistry model considered by Pickering et al. (2016) and this study.

24) lines 263-264: this is not obvious from the Figure 5 plots

To make it clear, we have replotted that figure with annotation (Fig. R1).

The LNO_2 and LNO_x production 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. (2019) and Bucsela et al. (2019).
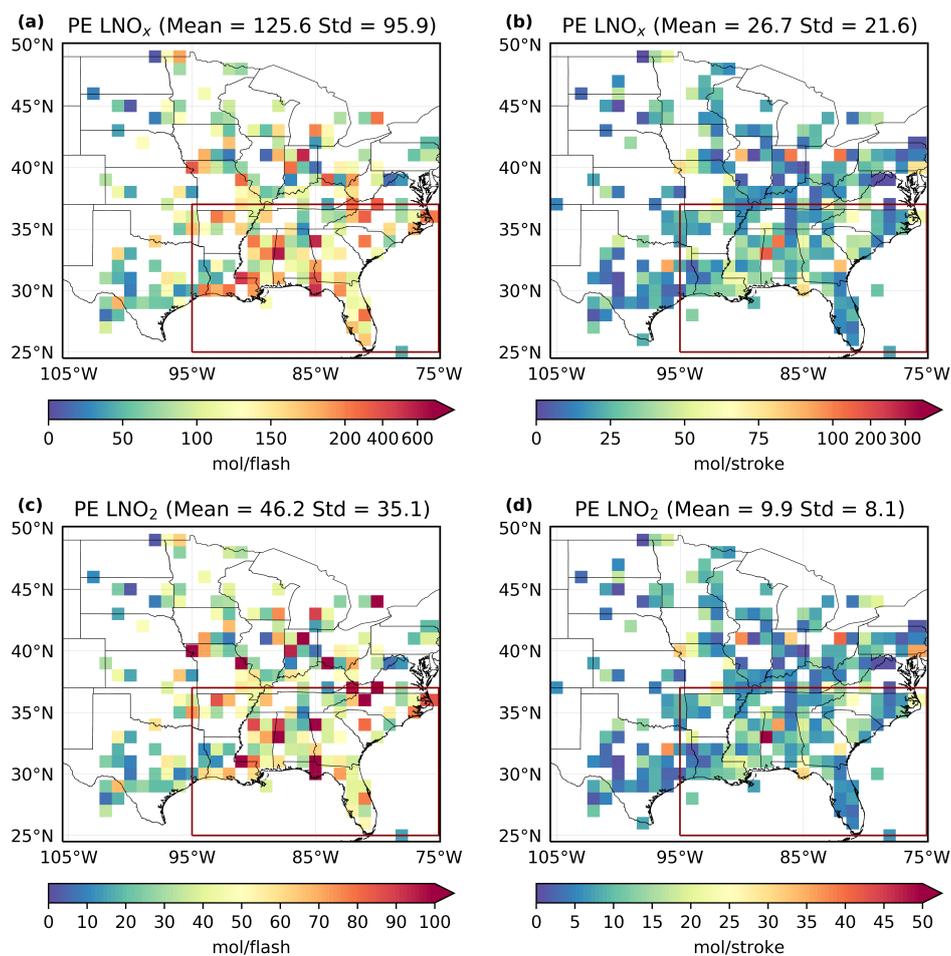
Figure R1 . (a) and (c) Maps of $1° \times 1°$ gridded values of mean LNO_x and LNO_2 production per flash with CRF $\geq 90\%$ for MJJA 2014. (b) and (d) Same as (a) and (c) except for strokes. The southeastern US is denoted by the red box in panels a – d.

25) line 271: peaks of the LNO$_2$ profile are...

Corrected.

> The ratio would also be smaller while **peaks of the LNO$_2$ profile are** below the CP.

26) line 275: from 0.8 to 0.2 as the cloud height increases ...

Thanks. We have corrected that sentence with exact interval of the cloud pressure.

> Since the ratio of LNO$_2$Vis to LNO$_2$ decreases from 0.8 to 0.2 as the cloud pressure **decreases from 600 to 300 hPa** ...

27) line 278: LNO$_2$ production (< 30 mol/stroke) occurs

Corrected.

> However, smaller LNO$_2$ **production (< 30 mol/stroke) occurs** on all levels between 650 hPa and 200 hPa.

28) line 284: Is this necessarily true? LNO$_x$ production per flash may be smaller in high flash rate storms.

Sorry for the misleading. It should be the LNO$_x$ concentrations instead of LNO$_x$ production per flash. Corrected.

> Because updrafts are stronger and flash rates are higher in anomalous storms, **UT LNO$_x$ concentrations** is larger in anomalous than normal polarity storms.

29) lines 287-288: ... (postconvection) in which LNO$_x$ has already been redistributed.........uses LNO$_x$ production profiles.......(Allen et al., 2012; Luo et al., 2017)

Corrected.

> 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**).

30) line 296: 2×500 mol NO flash^{-1} This designation can be confusing. Some readers may think you mean 1000 mol/flash

We prefer that designation, because we actually changed the adjusting factor and the LNO production at the same time instead of only changing the LNO production per flash. The definition of 1×200 mol NO flash^{-1} has been used in the model description (Section 2.3). If readers think we use 1000 mol/flash, that should be fine because the effect of that on LNO$_x$ estimation is the same.

31) line 308: different than that....

Corrected.

> 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$_{LNO_x}$.

32) line 401-402: we find that the effect is regionally dependent. Both........NO$_2$ cause different comprehensive effects due to nonlinear.....

Thanks. We have rewritten these sentences.

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 model setting leads to larger retrieval of LNO$_x$ despite some regionally dependent effects**. 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$_{LNO_x}$.

33) Table 2: How many grid boxes per day typically qualify under your criteria?

The mean number of valid grid boxes is 4.2 and 5.3 for flash and stroke data, respectively, as shown in the time series of valid grid boxes (Fig. R2).
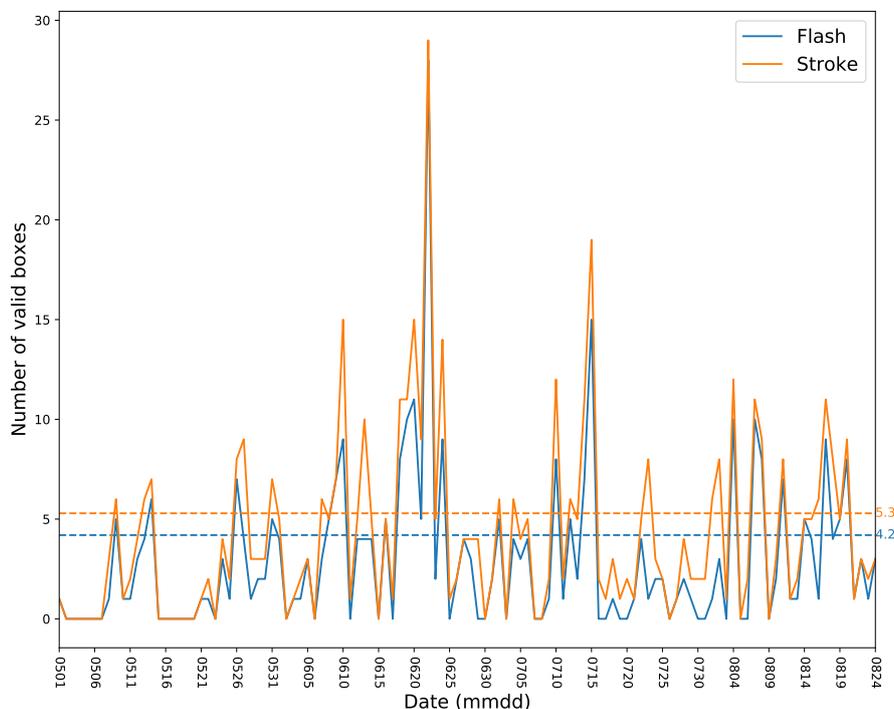

Figure R2 . Time series of valid grid boxes over the CONUS for MJJA 2014 with the criteria (CRF $\geq$ 90%, entln flashes(strokes) $\geq$ 2400(8160), TL $\geq$ 1000 and ratio $\geq$ 50%).

34) Table 3: Need to indicate how the percentage difference was calculated in the caption. Which one was in the denominator?

The percentage difference was calculated by raising and lowering the perturbed value, this will effectively calculate a mean percent difference. The equation is: $PE_{uncertainty} = (Error_{rising\ perturbed\ value} - Error_{lowering\ perturbed\ value})/2$ where $Error_{perturbed\ value} = (PE_{perturbed\ value} - PE_{original\ value})/PE_{original\ value}$. We have added these information to the caption of that table (Table R1).

Table R1 . 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]	Constant AMF: 0.46	23	23	23	23
Surface pressure[1]	$\pm$ 1.5%	0	0	0	0
Surface reflectivity[1]	$\pm$ 17%	0	0	0	0
LNO_2 profile[1]	2×500 mol NO flash^{-1}	15	29	14	29
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	20	20	20
Net	-	48	54	47	54

$PE_{uncertainty} = (Error_{rising\ perturbed\ value} - Error_{lowering\ perturbed\ value})/2$ where $Error_{perturbed\ value} = (PE_{perturbed\ value} - PE_{original\ value})/PE_{original\ value}$.
[1]Laughner et al. (2019) [2]Beirle et al. (2009) [3]Lapierre et al. (2019) [4]Allen et al. (2019) and Bucsela et al. (2019) [5]Uncertainty (%)

35) Figures 7 and 8: What do the various colors of shading indicate? What are the scales for the horizontal plot at the top and the vertical plot on the side?

The colors of shading correspond to the regions, which contain the respective normalized density estimated by kernel density estimation, using kdeplot in the Python package named seaborn (https://seaborn.pydata.org/generated/seaborn.kdeplot.html). Because the shading has shown the density, the marginal plots have been removed in case of any confusions (Fig. R3).
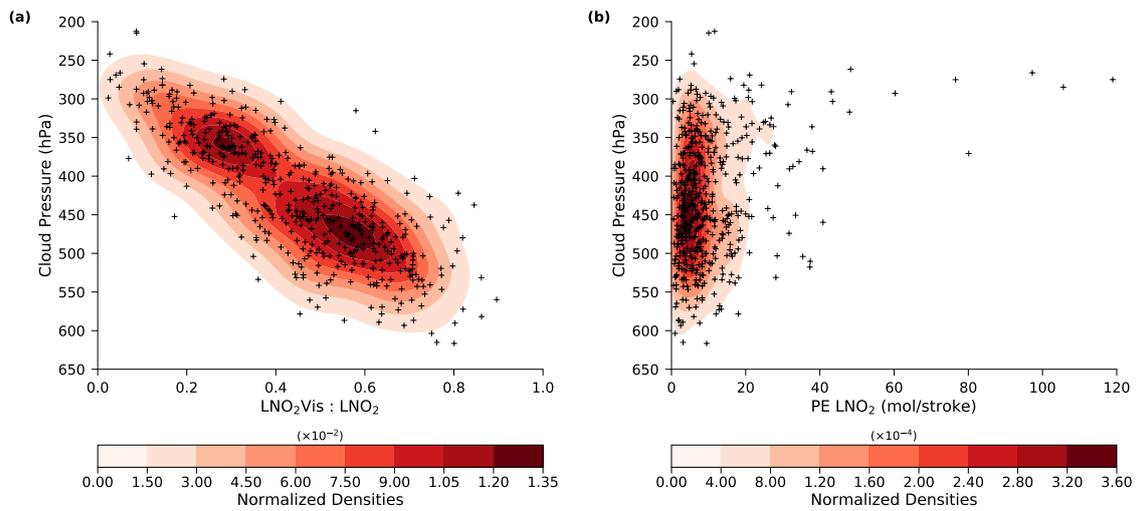
Figure R3 . Kernel density estimation of the (a) daily ratio of LNO_2Vis to LNO_2 and (b) daily LNO_2 production efficiency versus the daily cloud pressure measured by OMI with CRF $\geq$ 90% for MJJA 2014.

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Estimates of Lightning NO$_x$ Production based on High Resolution OMI NO$_2$ Retrievals over the Continental US

Response to Anonymous Referee #3

Xin Zhang, Yan Yin, Ronald van der A, Jeff L. Lapierre, Qian Chen,
Xiang Kuang, Shuqi Yan, Jinghua Chen, Chuan He and Rulin Shi

February 7, 2020

We thank the reviewer for his/her positive comments and very careful reading of the main article. The individual corrections suggested are addressed below. The reviewer's comments will be shown in **red**, our response in **blue**, and changes made to the paper are shown in **black** block quotes. Unless otherwise indicated, page and line numbers correspond to the original paper. Figures, tables, or equations referenced as "Rn" are numbered within this response; if these are used in the changes to the paper, they will be replaced with the proper number in the final paper.

Major Comments

1) The authors derive equations for LNO$_x$ by treating clouds as reflecting surfaces. This simplification is required for many radiative transfer models which can not handle multiple scattering in 3d clouds. Thus, several previous studies follow this approach. However, the authors should still be aware of this simplification and state this clearly in the manuscript. Formulations like "NO$_2$ above/below the cloud" are misleading, as for thick thunderstorm clouds, most of the LNO$_x$ is WITHIN the cloud, with a high sensitivity from OMI at the cloud top, gradually decreasing towards the cloud bottom. In this context, the authors should discuss what the "cloud top" derived from OMI O$_2$-O$_2$ measurements actually means for a thunderstorm cloud.

We appreciate the reviewer's suggestion on the "cloud top" and have added the discussion of this concept in Sect. 2.4.

The concept of AMF_{LNO_x} was also used in Beirle et al. (2009) to investigate the sensitivity of satellite instruments for freshly produced lightning NO$_x$.

....

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, 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 compositions from the cloud bottom to the cloud top and revealed that a significant fraction 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.

2) There is one crucial omission which might require major revisions: The derived AMFLNO$_x$ will strongly depend on pcloud (e.g. eq. 2). But this key input parameter is missing in the error budget in table 3. This has to be revised and the uncertainties caused by pcloud have to be discussed accordingly.

We have added the uncertainty caused by pcloud (Table R1), Thanks.

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-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. **Beirle et al. (2009) obtained a mean total sensitivity of 0.46 (σ = 0.09) for LNO$_x$ in the sensitivity study, implying an uncertainty of 23% due to cloud pressure in our study.**

Table R1 . 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]	Constant AMF: 0.46	23	23	23	23
Surface pressure[1]	$\pm$ 1.5%	0	0	0	0
Surface reflectivity[1]	$\pm$ 17%	0	0	0	0
LNO_2 profile[1]	2×500 mol NO flash^{-1}	15	29	14	29
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	20	20	20
Net	-	48	54	47	54

$PE_{uncertainty}$ = ($Error_{rising\ perturbed\ value}$ - $Error_{lowering\ perturbed\ value}$)/2 where $Error_{perturbed\ value}$ = ($PE_{perturbed\ value}$ - $PE_{original\ value}$)/$PE_{original\ value}$.
[1]Laughner et al. (2019) [2]Beirle et al. (2009) [3]Lapierre et al. (2019) [4]Allen et al. (2019) and Bucsela et al. (2019) [5]Uncertainty (%)

Minor Comments

1) 5: Skip "program of".

Fixed.

> 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 product** and the Weather Research and Forecasting-Chemistry (WRF-Chem) model.

2) 130: must be square km.

Fixed.

> 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.

3) 146: The concept of defining an "AMF" for converting SNO_2 into LNO_x was also used in Beirle et al., AMT 9, 1077-1094, 2009, (see eq. 9 therein).

Thank you for your remind of that paper we did not read before. It actually could help us explain our methods and results a lot. We have added that concept before discussing the "Cloud Top" (See *No. 1, Sect. Major comments*).

4) 268: I recommend to extend "Production" to "Production estimates"

Extended.

> 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 **estimates** than NO_2Vis production **estimates**.

5) 395: "we find that the regionally dependent effect" - unclear, please revise.

Revised.

> 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 model setting leads to larger retrieval of LNO_x despite some regionally dependent effects caused by nonlinear calculation of AMF.**

6) Fig. 1: Please make the legends clearer and remove cryptic labels ("crf90_entin_tl")

After we discussed with Eric J. Bucsela privately, we have redone the linear regressions based on the daily mean LNO_x and flashes, as mentioned in the Response to Reviewer#2 (See *No. 19*). Because the different colors and dots could mislead readers, we have decided to summarize the linear regression results in Table 2 (Table R2).

7) Figs. 2 and 9: Please use less, but larger labels on x-axis.

Table R2 . LNO$_x$ production 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 (10^6 mol)	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 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. [3] The number of valid days with specific criteria in MJJA 2014.

We have modified the former figure (Fig. 3/ Fig. R1) and deleted the later one as mentioned in the Response to Reviewer#1 (See *No. 22, Sect. Specific Comments*).
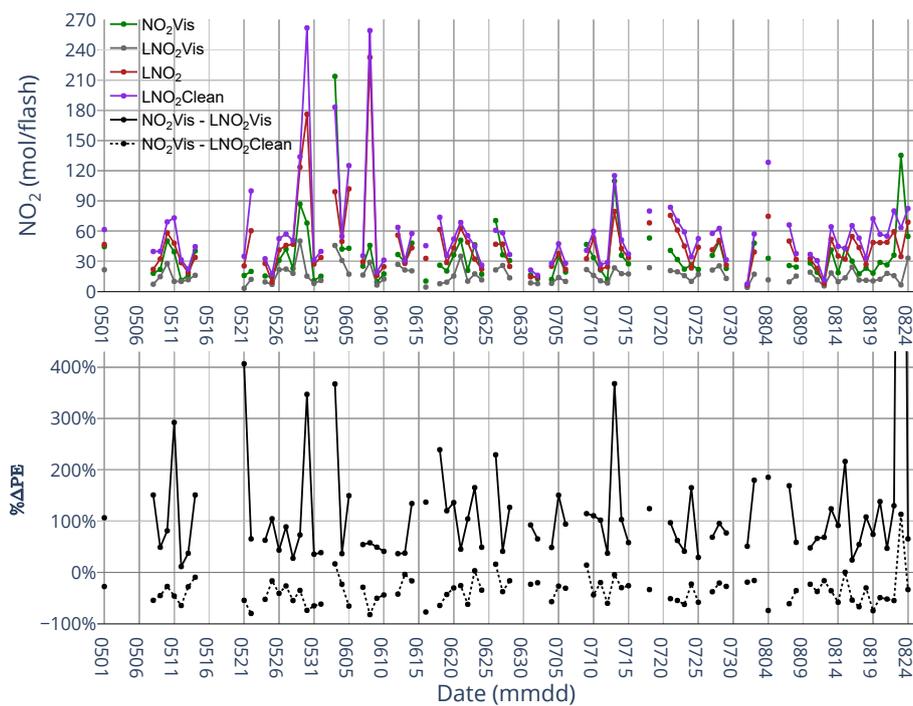
Figure R1 . (top) Time series of NO_2Vis, LNO_2Vis, LNO_2 and LNO_2Clean production per day over the CONUS for MJJA 2014 with CRF $\geq$ 90% and a flash threshold of 2400 flashes per 2.4 h. (bottom) Time series of the percent differences between NO_2Vis and LNO_2Vis and the percent differences between NO_2Vis and LNO_2Clean with CRF $\geq$ 90%. The value of black dot on August 23 (not shown) is 1958%.

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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~~ 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 ~~based on the program of new~~ that uses the 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~~ 32 $\pm$ ~~16~~ 15 mol NO$_2$ flash^{-1} and ~~8~~ 6 $\pm$ 3 mol NO$_2$ stroke^{-1} while LNO$_x$ PE is ~~120~~ 90 $\pm$ ~~52~~ 49 mol NO$_x$ flash^{-1} and ~~22~~ 17 $\pm$ 9 mol NO$_x$ stroke^{-1}. Results reveal that ~~former methods are more sensitive to~~ our method reduces sensitivity to the background NO$_2$ and ~~neglect~~ 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 ~~is~~ are mainly produced by soil, biomass burning and fossil fuel combustion, while NO$_x$ in the middle and upper troposphere ~~originate~~ 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).

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With the recent updates of UT NO_x chemistry, the day time lifetime of UT NO_x is evaluated to be ~ 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~~ versus the commonly used ~~the widely used~~ cloud-top height (CTH) approach. ~~As lightning with the CTH approach have reported~~ While lightning is predicted to increase by 5 — 16% ~~increases~~ over the next century with the CTH approach (Clark et al., 2017; Banerjee et al., 2014; Krause et al., 2014), a 15% decrease in 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 ~~compositions~~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 report 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 water mass fluxes.

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~~ 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 ~~qualified~~ 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 ± 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~~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~~ this study assumed that all the enhanced NO_2 originated from lightning ~~without 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

60 Network (WWLLN). ~~The threshold of high flash rates is that the summation of the corrected flashes within the satellite pixel~~ (Their analysis was restricted to 30×60 km^2 ~~) in the last one hour must be greater than~~ satellite pixels where the flash rate exceeded 1 ~~flashes/km^2 /h. But the results of~~ flash km^{-2} hr^{-1}. But they found LNO_x production ~~are~~ to be 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,

65 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}),

70 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~~ considered NO_2 above the cloud ~~are all~~ 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 $AMF_{LNO_x Clean}$ ($AMF_{LNO_2 Clean}$) 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

75 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 ± 45 mol NO_x per flash. Among several substantial sources of uncertainty, significant uncertainty exists in characterizing background NO_x (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 ± 100 mol per flash over East Asia,

80 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 ~~the weighted~~ subtracting temporal average of NO_x at each box ~~which meets~~ where the value was weighted by the number of OMI pixels which meet the optical cloud pressure and CRF criteria ~~but has 0 —~~ required to be considered deep convection but has 1 ~~flashes~~ flash or less instead. The lofted pollution was ~~corrected by~~ considered as 15% of

85 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 ~~at those~~ and stratospheric NO_2 at some locations.

   On ~~another~~ the other hand, Lapierre et al. (2019) constrained LNO_2 to 1.1 ± 0.6 mol NO_2/stroke for intracloud (IC) strokes and 10.0 ± 4.9 mol NO_2/stroke for cloud-to-ground (CG) strokes over the ~~continental US (CONUS)~~ CONUS. LNO_2 per stroke

90 was scaled to 54.4 mol NO_x/flash ~~by~~ using mean values of 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 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. (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~~ 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 ~~focused~~ 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, 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 the effect of different sources of the uncertainty ~~of~~ 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 13×24 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)~~(Dobber et al., 2008; KNMI, 2012). For the current study, we used the NASA standard product ~~v3~~ 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 ~~bias~~ 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 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.

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~~ combined events (IC and CG) with LIS flashes and found that the relative flash detection efficiency of ENTLN ~~increases from 21.6~~over CONUS increases from 62.4% during 2011 to ~~31.4~~79.7% during 2013. Lapierre et al. (2019) also compared combined ENTLN and the NLDN dataset with data from the LIS ~~and~~ during 2014 and found the detection efficiencies of IC flashes and strokes ~~are~~ to be 88% and 45%, respectively. Since we only 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%~~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) ~~was~~ were 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) 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}"). Although the simulated total flash densities are higher in the Southeast US and lower in the North Central US (Fig. 2), the criteria in Sect. 3.1 could limit this effect on the estimation of LNO$_x$ production and Sect. 3.4 will use another simulation to test this problem. 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$_{\text{LNO}_x}$ near convection is calculated according:

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

where S_{NO_2} is the OMI-measured tropospheric slant column NO$_2$ and AMF$_{\text{LNO}_x}$ is a customized lightning air mass factor. The concept of AMF$_{\text{LNO}_x}$ is defined was also used in Beirle et al. (2009) to investigate the sensitivity of satellite instruments for freshly produced lightning NO$_x$. In order to estimate LNO$_x$, we define the AMF$_{\text{LNO}_x}$ 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 radiance cloud fraction):

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

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.

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, 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 compositions from the cloud bottom to the cloud top and revealed that a significant fraction 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., 201 . As discussed in Pickering et al. (2016), the ratio of V$_{\text{LNO}_2}$ seen by OMI to V$_{\text{LNO}_x}$ 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. (2019), we calculate their $AMF_{\mathrm{LNO}_x\,\mathrm{Clean}}$ and $AMF_{\mathrm{NO}_2\,\mathrm{Vis}}$ respectively:

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

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

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 $AMF_{\mathrm{LNO}_2\,\mathrm{Vis}}$ is developed for comparison later.

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

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

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

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$_{\mathrm{LNO}_x}$ is partly influenced by p_{cloud}. The effects of LNO$_2$ below the cloud will be discussed in section 3.4.

2.5   Procedures for Deriving LNO$_x$

LNO$_x$ V$_{\mathrm{LNO}_x}$ is re-gridded to $0.05° \times 0.05°$ pixels like the BEHR product and grids using the constant value method (Kuhlmann et al., 2014). Then, it is analyzed in $1° \times 1°$ grid boxes with a minimum of fifty valid pixels which is equivalent to $0.05° \times 0.05°$ grids to minimize the noise. The minimum value is between five satellite pixels in Pickering et al. (2016) and three satellite pixels in Bucsela et al. (2019) or Allen et al. (2019). The main procedures used to derive LNO$_x$ are as follows:

CRFs (CRFs $\geq$ 70%, CRFs $\geq$ 90% and CRFs = 100%) and CP $\leq$ 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 convection. The CFs are defined as the maximum cloud fraction calculated by the Xu-Randall method between 350 and 400 hPa (Xu and Randall, 1996)(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 CFs $\geq$ 40% suggested by Strode et al. (2017) to determine cloudy or clear for each simulation grid.

Besides ~~properties of cloud , the~~ cloud properties, a time period and sufficient flashes (or strokes) are required for fresh

215 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 $1° \times 1°$ box over the CONUS (Lapierre et al., 2019). 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., 2019). 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.

220 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~~ 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 ~~LNO_2 is not polluted much~~ more than half of the NO_x above the cloud must have a LNO_x

225 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 ~ 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

230 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;

235 (2) linear regression method: applying the linear regression to daily ~~summations~~ 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

240 applied to the original data with a linear regression method (~~Fig. 2~~ Table 2).

A daily search of the NO_2 product for coincident ENTLN flash (stroke) data results in 99 (102) valid days under the ~~condition of CRF $\geq$ 90%~~ CRF90_ENTLN condition. 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% CRF90_ENTLN_TL1000_ratio50 condition, while LNO_x/flash increases from ~~86.0~~ 52.1 $\pm$ ~~14.0~~ 51.1 mol/flash to ~~114.8~~ 54.5 $\pm$ ~~18.2~~ 48.1 mol/flash. The result is

245 almost the same as ~~the one without~~ that under the CRF90_ENTLN_TL1000 condition which is without the condition of ratio

8

$\geq 50\%$. 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 $\geq 40\%$ 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 ≥ 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$_x$
250 production (Table 3).

Apart from the fewer valid days under higher CRF conditions (CRF $\geq 90\%$ and CRF $= 100\%$), LNO$_x$/flash increases from ~~109.0~~35.7 $\pm$ ~~15.3~~36.8 mol/flash to ~~114.8~~54.5 $\pm$ ~~18.2~~48.1 mol/flash and decreases again to ~~99.4~~20.8 $\pm$ ~~15.3~~37.4 mol/flash while LNO$_x$/stroke enhances from ~~16.7~~4.1 $\pm$ ~~2.6~~3.9 mol/stroke to ~~17.8~~7.0 $\pm$ ~~2.9~~4.8 mol/stroke and drops again to ~~15.6~~2.6 $\pm$ ~~3.1~~4.0 mol/stroke (Table 3)~~.~~, as the CRF criterion increases from 70% to 90% and to 100%. The increment of LNO$_x$ PE caused
255 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 ~~former studies~~the results of Pickering et al. (2016) and Lapierre et al. (2019).

260 ~~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 $\geq 90\%$ with flash threshold of 2400 flashes. (b)~~ ~~Same as (a) except with a stroke threshold of 8160 strokes.~~

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
265 with those of Pickering et al. (2016) and Lapierre et al. (2019), we choose NO$_2$ instead of NO$_x$ to derive ~~productions~~production per flash (production efficiency, PE). In Fig. 3, time series of NO$_2$Vis, LNO$_2$Vis, LNO$_2$ and LNO$_2$Clean 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$_2$ ~~productions~~production values are mostly in the range from 20 to 80 mol/flash. LNO$_2$Vis 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%
270 of the LNO$_x$ column lies below the CP, while the ratio in our WRF-Chem simulation is ~~10% – 80%~~56 $\pm$ 20%. The effect of ~~clouds~~cloud properties on LNO$_x$ production will be discussed in more detail in section 3.4. Generally, the order of estimated daily ~~production efficiencies (PEs)~~PEs is LNO$_2$Clean $>$ LNO$_2$ $>$ NO$_2$Vis $>$ LNO$_2$Vis. The percent difference in the estimated PE (ΔPE) between NO$_2$Vis and LNO$_2$Vis indicates a certain amount of background NO$_2$ exists above clouds. Overall, the tendency of that ΔPE is consistent with another ΔPE between NO$_2$Vis and LNO$_2$Clean. When the region is highly polluted
275 (ΔPE between NO$_2$Vis and LNO$_2$ is larger than 200%), PEs based on NO$_2$Vis and LNO$_2$Clean are significantly overestimated. In other words, NO$_2$Vis and LNO$_2$Clean are more sensitive to background NO$_2$. The extent of the overestimation of NO$_2$Vis is larger than that of LNO$_2$Clean in highly polluted regions, while it is usually opposite in most regions.

Figure 4 shows the linear regression for ENTLN data versus NO$_2$Vis, LNO$_2$Vis, LNO$_2$ and LNO$_2$Clean with the same criteria as shown in Fig. 3. LNO$_2$Clean production (the largest slope) is ~~49.1~~25.2 $\pm$ ~~8.4~~22.3 mol NO$_2$/flash with a correlation

280 of ~~0.79 and 7.5~~ 0.25 and 2.3 ± ~~1.3~~ 2.1 mol NO_2/stroke with a correlation of ~~0.77~~ 0.22. 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~~ 17.1 ± ~~2.7~~ 17.2 mol NO_2/flash and ~~3.6~~ 0.4 ± ~~0.5~~ 1.0 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 <u>daily summed</u> NO_2Vis

285 <u>values</u> (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

290 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: February 7, 2020). 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

295 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~~ 18.7 ± ~~6.9~~ 18.1 mol/flash and ~~6.3~~ 2.1 ± ~~1.1~~ 1.8 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 <u>based on daily summed values (not shown)</u> is 114.8 ± 18.2 mol/flash (or 17.8 ± 2.9 mol/stroke) which is larger than

300 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~~ <u>and chemistry model</u> considered by Pickering et al. (2016) and this study.

The mean and standard deviation of LNO_2 production under CRF ≥ 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

305 LNO_2 and LNO_x production are both higher in the ~~South Central~~ Southeast U.S. (~~88°~~ <u>denoted by the red box in Fig. 5 panels,</u> <u>75°</u>W – ~~103~~ 95°W, 28.5°N – 39°N) ~~and Southeast U.S. (79°W~~ – 85°W, 25°N – ~~35~~ 37°N), consistent with Lapierre et al. (2019) and Bucsela et al. (2019). Compared with Fig. 3, ~~Fig. 6(a) and (b)~~ <u>Figure 6a and b</u> 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

310 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 <u>estimates</u> than NO_2Vis production <u>estimates</u> (Fig. 6c). Figure 6~~(d)~~ <u>d</u> 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~~ also be smaller while peaks of the LNO$_2$ profile ~~is~~ are below the CP. 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

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 cities denoted by stars and triangles in Fig. 6a. Then, the differences between AMFs are dependent on fewer parameters:

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

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

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

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 background NO$_2$ ratio are C-shape because LNO$_2$ concentrations are higher than background NO$_2$ in the UT. 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$_{\mathrm{LNO_2}}$ (Eq. 7) and AMF$_{\mathrm{LNO_2Clean}}$ (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 convections over polluted locations. In contrast, production estimated by our method is larger than that based on NO$_2$Vis 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$_2$Vis, which has been discussed in Sect. 3.2. The relative change between AMF$_{\mathrm{LNO_2Clean}}$ (Eq. 9) and AMF$_{\mathrm{NO_2Vis}}$ (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 is 153.8% among the six grids where the highest clouds occur.

3.4 Effects of Cloud and LNO$_x$ Parameterization on LNO$_x$ Production

340 Figure ~~7~~ 8a presents the daily distribution of CP and the ratio of LNO$_2$Vis to LNO$_2$ during MJJA 2014 with the criteria defined in section 3.1 under CRF $\geq$ 90%. Since the ratio of LNO$_2$Vis to LNO$_2$ decreases from 0.8 to 0.2 ~~while the cloud is higher (smaller pressure value)~~ as the cloud pressure decreases from 600 to 300 hPa, NO$_2$Vis production is smaller than LNO$_2$ in relatively clean areas as shown in Fig. 4. Apart from LNO$_2$Vis, 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~~ 8b). However, smaller LNO$_2$ ~~productions~~

345 production (< 30 mol/stroke) ~~occur~~ occurs on all levels between 650 hPa and 200 hPa. Because of the limited amount of large LNO$_2$ production and lightning data, we ~~can not derive that higher~~ cannot derive the relationship between LNO$_2$ production ~~relates to higher clouds~~ and cloud pressure 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

350 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~~ higher in anomalous storms, UT LNO$_x$ ~~production is larger~~ concentrations is 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 dis-

355 tributing LNO$_x$ in models: LNO$_x$ profiles (postconvection) ~~are simulated after~~ in which LNO$_x$ ~~is~~ has already been redistributed by convective transport, while the other one (preconvection) uses LNO$_x$ production profiles made before the redistribution of convective transport ~~(Luo et al., 2017)~~ (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° × 1° results based on postconvective LNO$_x$ profile are sufficient for estimating average LNO$_x$ production.

360 ~~Scatter plots of the daily LNO$_2$ production efficiency versus the daily cloud pressure measured by OMI with CRF $\geq$ 90% for MJJA 2014.~~

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~~ value (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

365 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$_2$ and LNO$_x$ productions. For the linear regression method (Fig. 9), LNO$_2$ production is ~~46.4~~ 29.8 $\pm$ ~~7.8~~ 20.5 mol/flash which is ~~11.5~~ 59.4% larger than the basic one (~~41.6~~ 18.7 $\pm$ ~~6.9~~ 18.1 mol/flash). ~~However~~ Meanwhile, LNO$_x$

370 production (increasing from ~~114.8~~ 54.5 $\pm$ ~~18.2~~ 48.1 mol/flash to ~~143.4~~ 88.5 $\pm$ ~~24.0~~ 61.1 mol/flash) ~~depends to a large extent~~ also depends 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 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 ~~region~~ 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 ~~in~~ 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.

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 both lower and higher LNO_2 production. 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 ~~distribution~~ distributions of mean LNO and LNO_2 profiles ~~is~~ 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 region dependent and ~~the~~ 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 ~~in our research~~ 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 ~~involved~~ 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 ~~:~~

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

Eq. (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. 12~~(c) and (d)~~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. 12~~(a)~~a and Fig. 12~~(b)~~b is the denominator: the total tropospheric

405   LNO$_2$ vertical column and visible LNO$_2$ vertical column respectively. As a result, the negative values in Fig. 12~~(a)~~a is caused by the part of LNO$_2$ below the cloud. The comparison between ~~Figure 4 and Figure~~ Fig. 4 and Fig. 9 shows that LNO$_2$Clean and LNO$_2$ values are more similar while LNO$_2$ and NO$_2$Vis 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~~15% and ± ~~26~~29% respectively.

4   Uncertainties Analysis

410   The uncertainties of the LNO$_2$ and LNO$_x$ production are estimated following Pickering et al. (2016), Allen et al. (2019), Bucsela et al. (2019), Lapierre et al. (2019) and Laughner et al. (2019). 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-retrieval of the LNO$_2$ and LNO$_x$ with the perturbed values (Table 5).

415   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. Beirle et al. (2009) obtained a mean total sensitivity of 0.46 (σ = 0.09) for LNO$_x$ in the sensitivity study, implying an uncertainty of 23% due to cloud pressure in our study. 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

420   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

425    $\partial f_r / \partial f_g$ 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

430   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

435   to an uncertainty of ~~13% and 26~~15% and 29%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}.
440 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~~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,
445 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,
450 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.

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; Silve
455 . 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 an uncertainty of 20% to this error considering the possible positive NO$_2$ measurements interferences (Allen et al., 2019; Bucsela et al., 2019).

In addition, the estimation of LNO$_x$ PE is also dependent 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
460 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,
465 we believe that the ratio condition (LNO$_2$Vis/NO$_2$Vis $\geq$ 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 ~~37% and 43~~48% and 54% 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 ~~44~~32 mol/flash, ~~120~~90 mol/flash, ~~8~~6
470 mol/stroke and ~~22~~17 mol/stroke. Applying the corresponding uncertainty to these mean values, we arrive at ~~44~~32 $\pm$ ~~16~~15 mol LNO$_2$/flash, ~~120~~90 $\pm$ ~~52~~49 mol LNO$_x$/flash, ~~8~~6 $\pm$ 3 mol LNO$_2$/stroke and ~~22~~17 $\pm$ 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,

475 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

480 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 works in both clean and polluted regions because of the consideration of tropospheric background pollution in the definition of AMFs. It uses specific criteria combin-

485 ing 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~~ production efficiencies over the CONUS. Considering all the uncertainties (Table 5) and applying the summation and regression method, the final mean ~~PEs~~ production efficiencies are estimated to be ~~44~~ 32 $\pm$ ~~16~~ 15 mol LNO$_2$/flash, ~~120~~ 90 $\pm$ ~~52~~ 49 mol LNO$_x$/flash, ~~8~~ 6 $\pm$ 3 mol LNO$_2$/stroke

490 and ~~22~~ 17 $\pm$ 9 mol LNO$_x$/stroke.

Compared with former methods, ~~we find that NO$_2$Vis and LNO$_2$Clean are more~~ our method has reduced the sensitive to background NO$_2$, while ~~NO$_2$Vis underestimates PE~~ the method in Lapierre et al. (2019) underestimates LNO$_x$ production efficiency because of the neglected below-cloud LNO$_2$ and LNO$_2$ ~~Clean overestimates LNO$_2$ production~~ production is overestimated using the method in Pickering et al. (2016) due to the over-cloud background NO$_2$ in polluted regions. Finally, implement-

495 ing 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 ~~regionally dependent effect~~ larger LNO production model setting leads to larger retrieval of LNO$_x$ despite some regionally dependent effects caused by nonlinear calculation of AMF. Both the ~~relative distribution of~~ ratio of the tropospheric LNO$_2$ above the cloud to the total tropospheric LNO$_2$ and the ratio of LNO$_2$ to NO$_2$ ~~would take the comprehensive effect for differences by~~ cause different comprehensive effects due to the nonlinear calculation of AMF$_{LNO_2}$ and AMF$_{LNO_x}$.

500 Since other regions, like China and India, have much more NO$_2$ ~~pollutions~~ 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

505 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 ~~levels~~scales.

Code and data availability. The retrieval algorithm used in Sect. 2.4 is available at https://github.com/zxdawn/BEHR-LNOx (last access: February 7, 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

510

$$AMF_{\text{LNO}_2} = \frac{(1-f_r)\int_{p_{surf}}^{p_{tp}} w_{clear}(p)NO_2(p)\,dp + f_r \int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p)NO_2(p)\,dp}{\int_{p_{surf}}^{p_{tp}} LNO_2(p)\,dp} \tag{A1}$$

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

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

515 (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.

$$AMF_{\text{LNO}_2\text{Clean}} = \frac{(1-f_r)\int_{p_{surf}}^{p_{tp}} w_{clear}(p)LNO_2(p)\,dp + f_r \int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p)LNO_2(p)\,dp}{\int_{p_{surf}}^{p_{tp}} LNO_2(p)\,dp} \tag{A3}$$

$$AMF_{\text{NO}_2\text{Vis}} = \frac{(1-f_r)\int_{p_{surf}}^{p_{tp}} w_{clear}(p)NO_2(p)\,dp + f_r \int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p)NO_2(p)\,dp}{(1-f_g)\int_{p_{surf}}^{p_{tp}} NO_2(p)\,dp + f_g \int_{p_{cloud}}^{p_{tp}} NO_2(p)\,dp} \tag{A4}$$

520 $$AMF_{\text{NO}_x\text{Vis}} = \frac{(1-f_r)\int_{p_{surf}}^{p_{tp}} w_{clear}(p)NO_2(p)\,dp + f_r \int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p)NO_2(p)\,dp}{(1-f_g)\int_{p_{surf}}^{p_{tp}} NO_x(p)\,dp + f_g \int_{p_{cloud}}^{p_{tp}} NO_x(p)\,dp} \tag{A5}$$

$$AMF_{\text{LNO}_2\text{Vis}} = \frac{(1-f_r)\int_{p_{surf}}^{p_{tp}} w_{clear}(p)NO_2(p)\,dp + f_r \int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p)NO_2(p)\,dp}{(1-f_g)\int_{p_{surf}}^{p_{tp}} LNO_2(p)\,dp + f_g \int_{p_{cloud}}^{p_{tp}} LNO_2(p)\,dp} \tag{A6}$$

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

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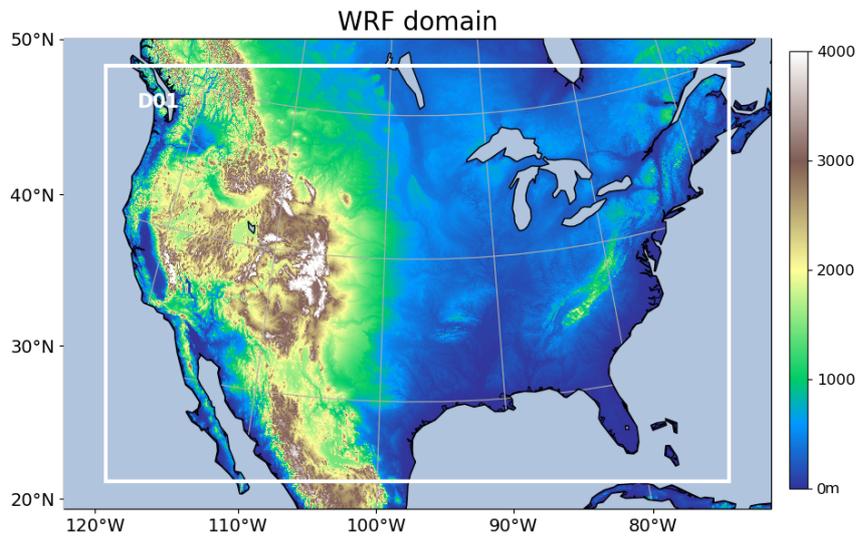
Figure 1. ~~The 12-km resolution domain for~~ Domain and terrain height (m) of the WRF-Chem ~~simulations~~simulation with 350 x 290 grid cells and a horizontal resolution of 12 km.
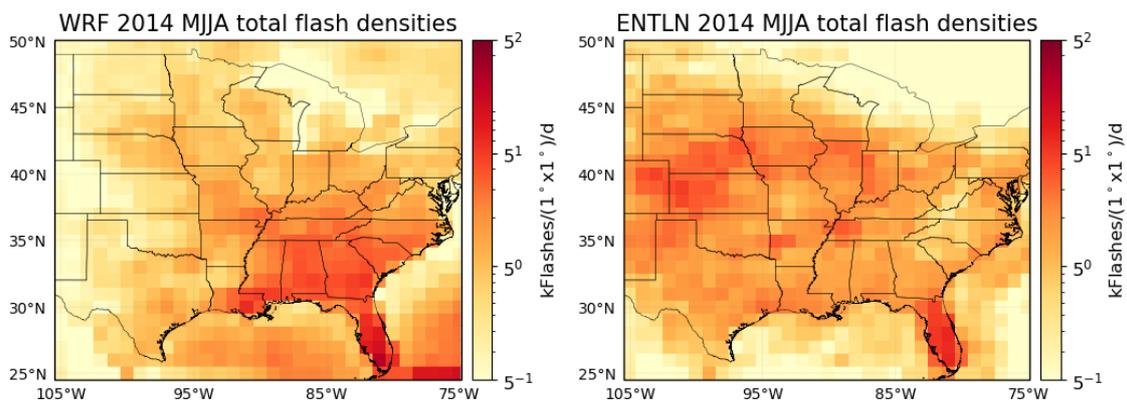
Figure 2. Comparison between total flash densities from ENTLN and WRF-Chem during MJJA 2014.

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 +

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

Table 2. LNO_x production 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 (10^6 mol)	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 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. [3] The number of valid days with specific criteria in MJJA 2014.

Table 3. LNO$_x$ production ~~under~~ for different ~~conditions~~ 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 ($10^{\sim6\sim5}$ mol)	Days[2]
70	Flash	~~109.0~~ 35.7 ± ~~15.3~~ 36.8	~~0.84~~ 0.21	~~0.23~~ 4.91	85
90	Flash	~~114.8~~ 54.5 ± ~~18.2~~ 48.1	~~0.82~~ 0.25	~~0.15~~ 3.90	81
100	Flash	~~99.4~~ 20.8 ± ~~15.3~~ 37.4	~~0.84~~ 0.13	~~0.10~~ 5.67	71
70	Stroke	~~16.7~~ 4.1 ± ~~2.6~~ 3.9	~~0.79~~ 0.21	~~0.58~~ 5.16	96
90	Stroke	~~17.8~~ 7.0 ± ~~2.9~~ 4.8	~~0.78~~ 0.29	4.16	93
100	Stroke	~~15.6~~ 2.6 ± ~~3.1~~ 4.0	~~0.75~~ 0.14	~~0.16~~ 5.41	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.
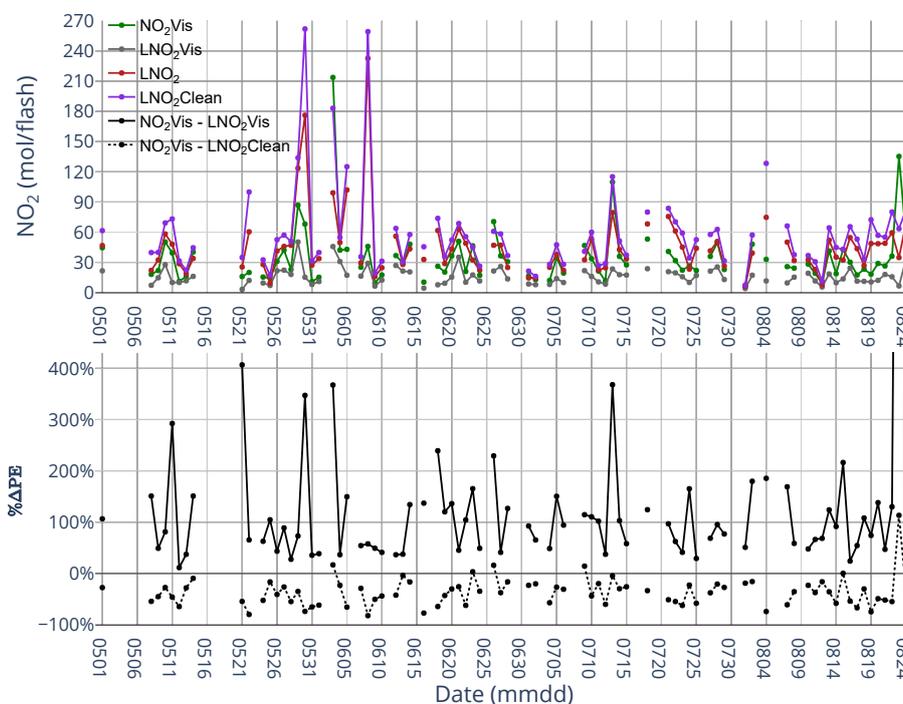
Figure 3. (top) Time series of NO$_2$Vis, LNO$_2$Vis, LNO$_2$ and LNO$_2$Clean production per day over the CONUS for MJJA 2014 with CRF $\geq$ 90% and a flash threshold of 2400 flashes per 2.4 h. (bottom) Time series of the percent differences between NO$_2$Vis and LNO$_2$Vis and the percent differences between NO$_2$Vis and LNO$_2$Clean with CRF $\geq$ 90%. The value of black dot on August 23 (not shown) is 1958%.
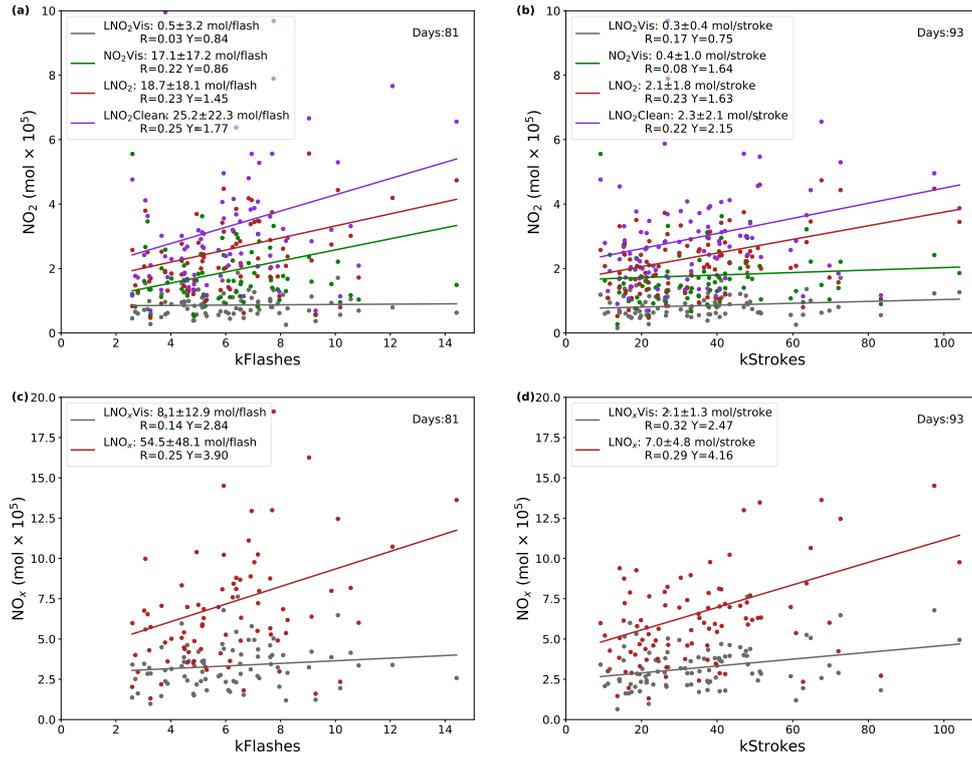
Figure 4. (a) Daily NO_2 Vis, LNO_2 Vis, LNO_2 and LNO_2 Clean versus ENTLN total flashes data. (b) Same as (a) but for strokes. (c) Daily LNO_x Vis and LNO_x versus total flashes. (d) Same as (c) but for strokes.

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

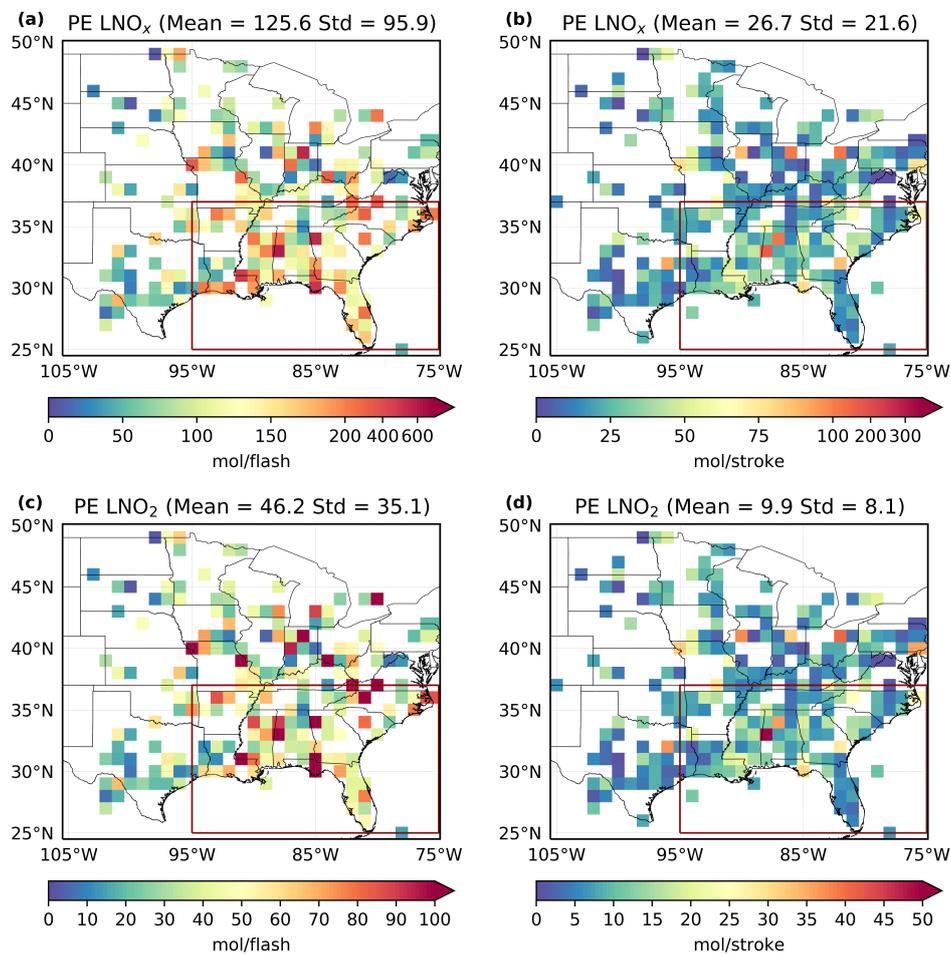|	City[1]	$(LNO_2Clean - LNO_2)/LNO_2$	$(LNO_2 - TropVis)/TropVis$	$(LNO_2Clean-TropVis)/TropVis$
Polluted	Lansing	24.2%	49.5%	85.6%
	New Orleans	13.3%	121.2%	153.8%
	Orlando	46.3%	37.5%	101.3%
Clean	Huron	12.0%	56.4%	75.2%
	Charles Town	12.0%	82.2%	104.1%
	Tarboro	5.0%	86.0%	95.3%

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

Figure 5. (a) and (c) Maps of $1° \times 1°$ gridded values of mean LNO_x and LNO_2 production per flash with CRF $\geq$ 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.
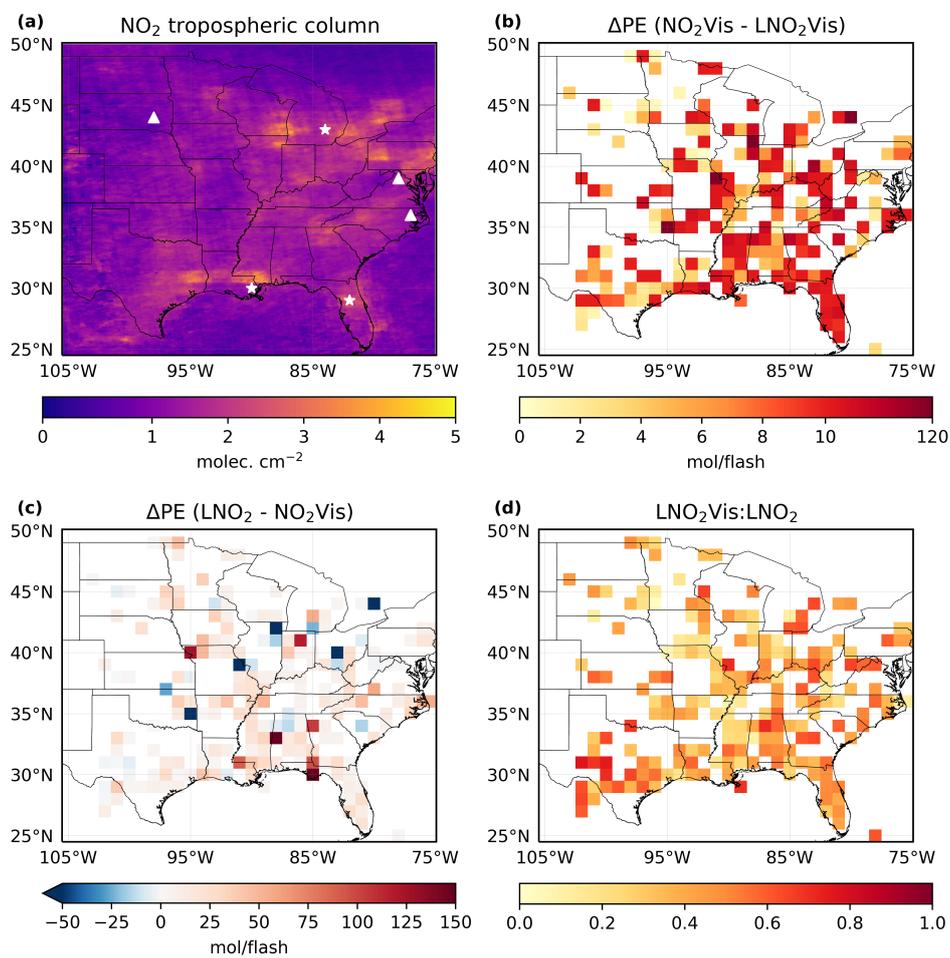
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Figure 6. (a) Mean (MJJA 2014) NO$_2$ tropospheric column. <u>Polluted cities are denoted by stars: Lansing, New Orleans and Orlando while clean cities are denoted by triangles: Huron, Charles Town and Tarboro.</u> (b) The differences of the estimated mean production efficiency between NO$_2$Vis and LNO$_2$Vis with CRF $\geq$ 90%. (c) The same differences as (b) but between LNO$_2$ and NO$_2$Vis. (d) The ratio of LNO$_2$Vis to LNO$_2$.
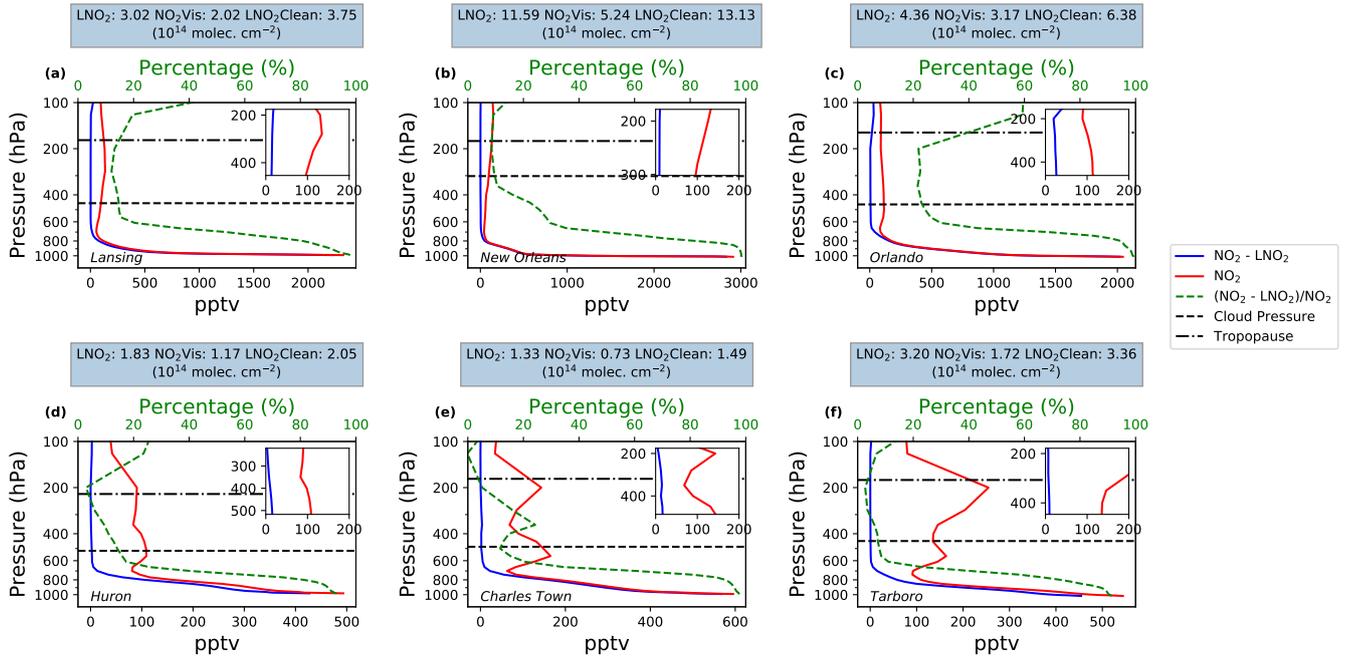
Figure 7. Comparison of mean WRF-Chem NO_2 and background NO_2 profiles in six grids with CRF $\geq$ 100% on specific days during MJJA 2014. The top row data are selected from polluted regions (stars in Fig. 6a) while the bottom row data are from clean regions (triangles in Fig. 6a). The green dashed lines are the mean ratio profiles of background NO_2 to 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.
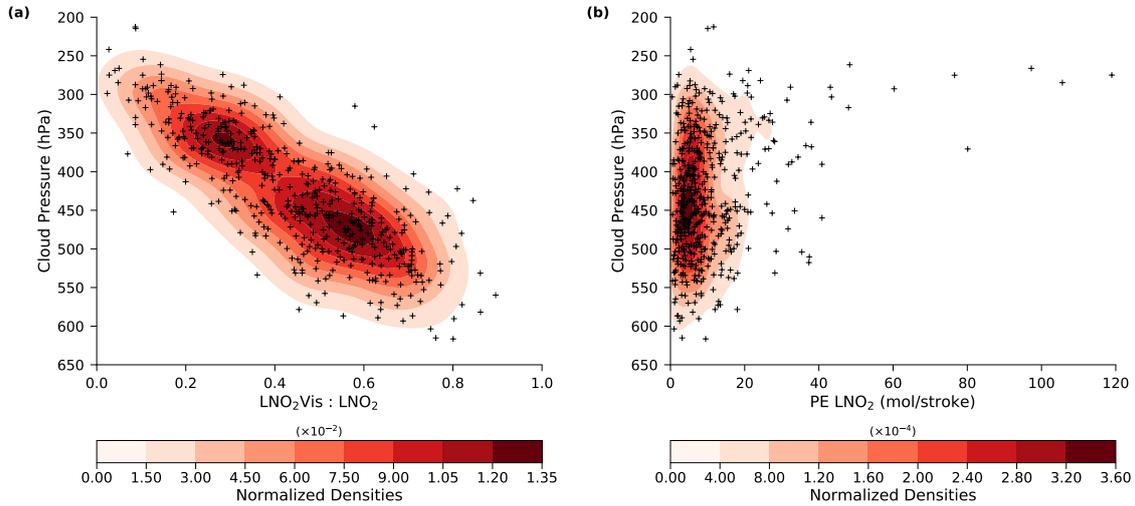
Figure 8. ~~Scatter plots~~ Kernel density estimation of the (a) daily ratio of LNO_2Vis to LNO_2 and (b) daily LNO_2 production efficiency versus the daily cloud pressure measured by OMI with CRF $\geq$ 90% for MJJA 2014.
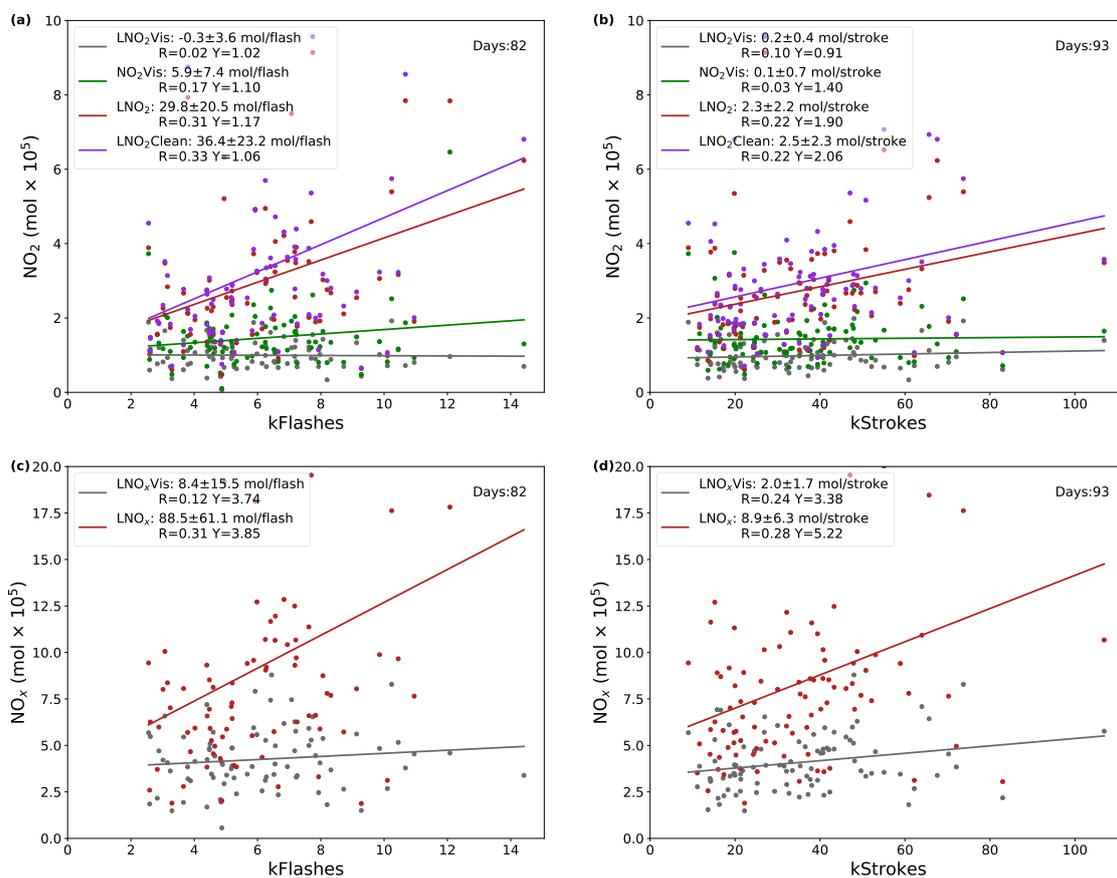
Figure 9. Same as ~~Figure~~ Fig. 4 except for 2×500 mol NO flash^{-1} configuration.
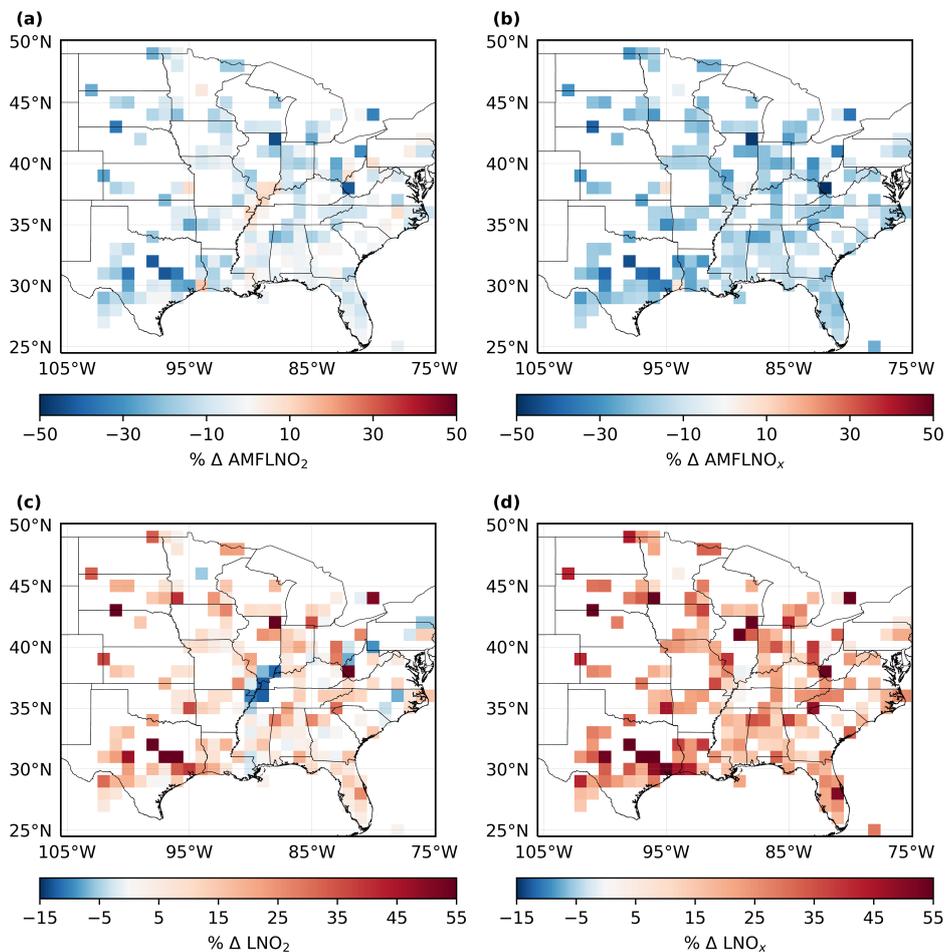
Figure 10. Average percent difference in (a) AMF$_{LNO_2}$, (b) AMF$_{LNO_x}$, (c) LNO$_2$ and (d) LNO$_x$ with CRF $\geq$ 90% over MJJA 2014. Difference between profiles are generated by 2×500 mol NO flash^{-1} and 1×200 mol NO flash^{-1}.
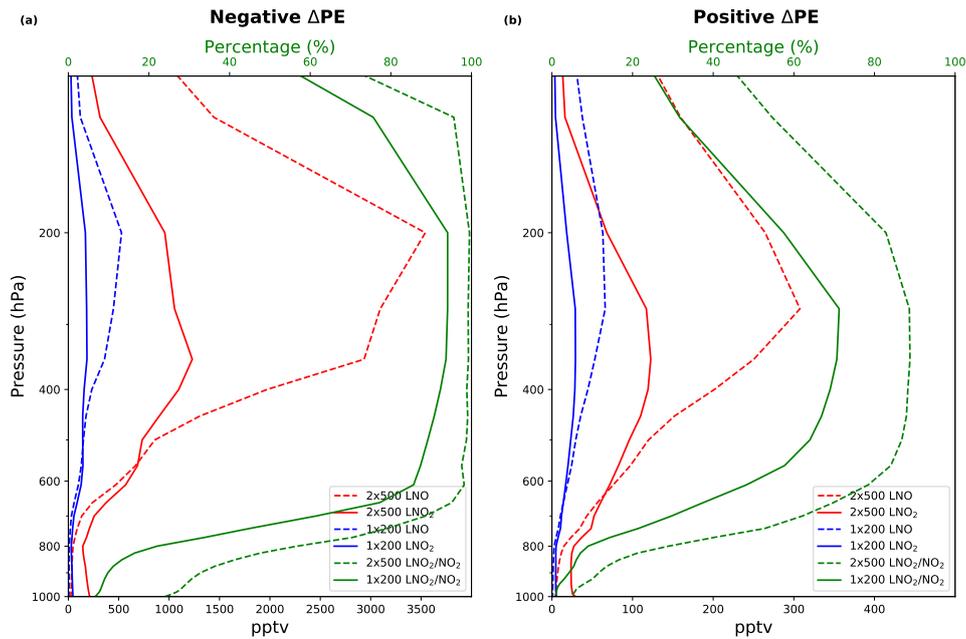
Figure 11. LNO and LNO_2 profiles with different LNO settings at (a) the region containing the minimal negative percent change in LNO_2 and (b) the region containing the largest positive percent change in LNO_2 when the LNO setting is changed from 1×200 mol NO flash^{-1} to 2×500 mol NO flash^{-1}, averaged 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}.
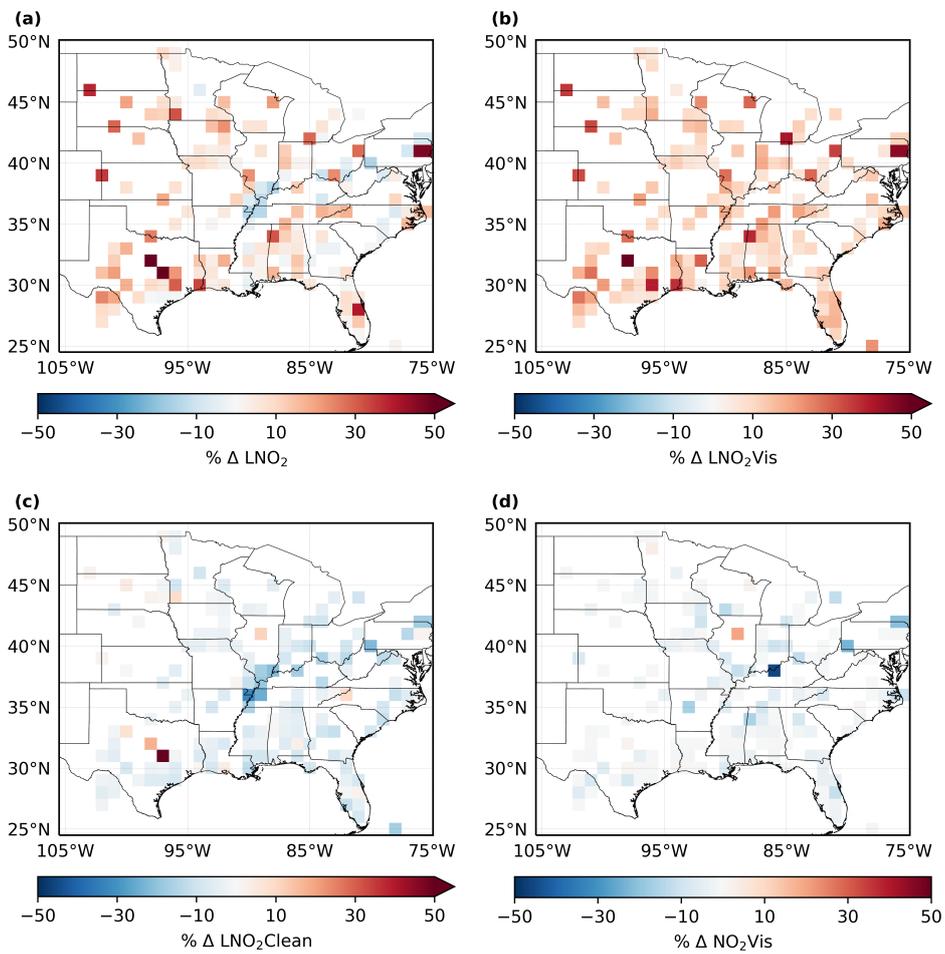
Figure 12. Average percent difference in (a) LNO_2, (b) LNO_2Vis, (c) LNO_2Clean and (d) NO_2Vis with CRF = 100% over MJJA 2014.

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[4,5]	LNO_x/flash[4,5]	LNO_2/stroke[4,5]	LNO_x/stroke[4,5]
BEHR tropopause pressure[1]	NASA product tropopause	6	4	6	4
Cloud radiance fraction[1]	$\pm$ 5%	2	2	2	2
Cloud pressure[2]	Constant AMF: 0.46	23	23	23	23
Surface pressure[1]	$\pm$ 1.5%	0	0	0	0
Surface reflectivity[1]	$\pm$ 17%	0	0	0	0
LNO_2 ~~Profile~~profile[1]	2×500 mol NO flash^{-1}	~~13~~15	~~26~~29	~~13~~14	~~26~~29
Profile location[1]	Quasi-Monte Carlo	0	1	0	1
Lightning detection efficiency[2,3]	IC: $\pm$ 16%, CG: $\pm$ 5%	15	15	15	15
t_{window}[2,3]	2 – 4 hours	10	10	8	8
LNO_x lifetime[2,3]	2 – 12 hours	24	24	24	24
V_{strat}[3,4]	-	~~15~~10	~~15~~10	~~15~~10	~~15~~10
Systematic errors in slant column[3,4]	-	5	5	5	5
Tropospheric background[4]	-	10	10	10	10
NO/NO_2[4]	-	20	20	20	20
Net	-	~~37~~48	~~43~~54	~~36~~47	~~43~~54

$PE_{uncertainty} = (Error_{rising\ perturbed\ value} - Error_{lowering\ perturbed\ value})/2$ where $Error_{perturbed\ value} = (PE_{perturbed\ value} - PE_{original\ value})/PE_{original\ value}$.

[1] Laughner et al. (2019) [2] Beirle et al. (2009) [3] Lapierre et al. (2019) [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