Lightning NO_2 simulation over the Contiguous US and its effects on satellite NO_2 retrievals

Response to Anonymous Referee #1

Qindan Zhu, Joshua L. Laughner and Ronald C. Cohen

July 12, 2019

We thank the reviewers for their positive response and very careful reading of both the main article and the supplement. Our responses to the reviewer's comments and detailed changes made to the manuscript 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 "R*n*" 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. Figures, tables, and equations numbered normally refer to the numbers in the original discussion paper.

Here is my major issue with this manuscript: Throughout the manuscript the authors state that the lightning prediction is improved with use of CAPE-PR compared with CTH. However, the lightning schemes are run with different convective parameterizations which are going to produce different convective characteristics (locations, timing, frequency, amounts of precipitation, etc.). Therefore, they would need to run CAPE-PR with Kain Fritsch to truly be able to say that CAPE-PR is better. If making this additional model run is not possible, I would then suggest that throughout the paper the authors refer to CAPE-PR as Grell3D/CAPE-PR and refer to CTH as KF/CTH to reflect the fact that it is a combination of convection and lightning schemes that are producing the difference they see in lightning flash rates.

We appreciate the reviewer pointing out the inconsistency in the convective schemes. To be clarified, we used two model runs in the manuscript, one uses Grell 3D convective scheme with CTH lightning parameterization ("G3/CTH"), and another one uses Kain Fritsch convective scheme with CAPE-PR lightning parameterization ("KF/CAPE-PR"). The first one is the only option provided by WRF to parameterize lightning at convective parameterized scale.

We agree that the switch of convective scheme will affect the parameterized lightning flashes and as a result, we can't conclude the improvement in lightning flash representation in WRF-Chem is solely due to the new implemented CAPE-PR lightning parameterization. Therefore, we made another WRF-Chem run with Kain Frisch convective scheme and CTH lighting parameterization, referred as "KF/CTH". We add the description of this model run in Section 2.1.

"We analyze WRF-Chem outputs from three model runs. The first run, **referred as "G3/CTH"**, is consistent with Laughner and Cohen (2017); it selects the Grell 3D ensemble cumulus convective scheme (Grell, 1993; Grell and Dvnyi, 2002) and the CTH lightning parameterization. The Grell 3D convective scheme readily computes the neutral buoyancy level which serves as the optimal proxy for cloud top height (Wong et al., 2013). **The "G3/CTH" is the only option for the coupled convective-lighting parameterization used in WRF-Chem at a non-cloud resolving resolution (12 km). In addition, we run WRF-Chem with the CTH lightning parameterization coupled with the Kain-Fritsch cumulus convective scheme (Kain and Fritsch, 1990; Kain, 2004) ("KF/CTH") to test the effect of switching convective schemes. In the "KF/CTH" parameterization, the cloud top height is the level where the updraft vertical velocity equals to zero. Another run, referred as "KF/CAPE-PR",** selects the Kain-Fritsch cumulus convective scheme and the CAPE-PR lightning parameterization described above..."

In the context, we change CTH to either "G3/CTH" or "KF/CTH" and change CAPE-PR to "KF/CAPE-PR" accordingly. In Section 3.1, we added comparison of "KF/CTH" parameterized lightning flashes against ENTLN both in the text, Fig 1 and Table 1 (also labeled as Fig. R1 and Table R1 in this response).

"The G3/CTH parameterization fails to reproduce the spatial pattern of flashes observed by ENTLN over the CONUS. **Compared to the G3/CTH, the KF/CTH parameterization improves the spatial correlation in the southeast region of US.** The KF/CAPE-PR parameterization better captures the spatial distribution of flash densities both in the southeast region and elsewhere in CONUS."

"The model using the KF/CAPE-PR lightning parameterization yields a tight correlation and slope close to the unity over the US domain. In the southeastern US, the R^2 increases from 0.3 to 0.7 and slope is reduced from 2.08 to 0.96 with the KF/CAPE-PR parameterization compared to the G3/CTH. **The slope for KF/CTH is comparable to KF/CAPE-PR while the R^2 for KF/CAPE-PR is slightly higher.**"

"Elsewhere in CONUS, **the R^2 for KF/CAPE-PR improves significantly to 0.6 compared to both G3/CTH and KF/CTH**. The slope for KF/CAPE-PR is 1.19, which is within the uncertainty of the detection efficiency of ENTLN. In general the KF/CAPE-PR lightning parameterization captures the day-to-day variation in flash densities better than the G3/CTH **and KF/CTH parameterizations as shown by the improved R^2 values.**"

The abstract is modified to:

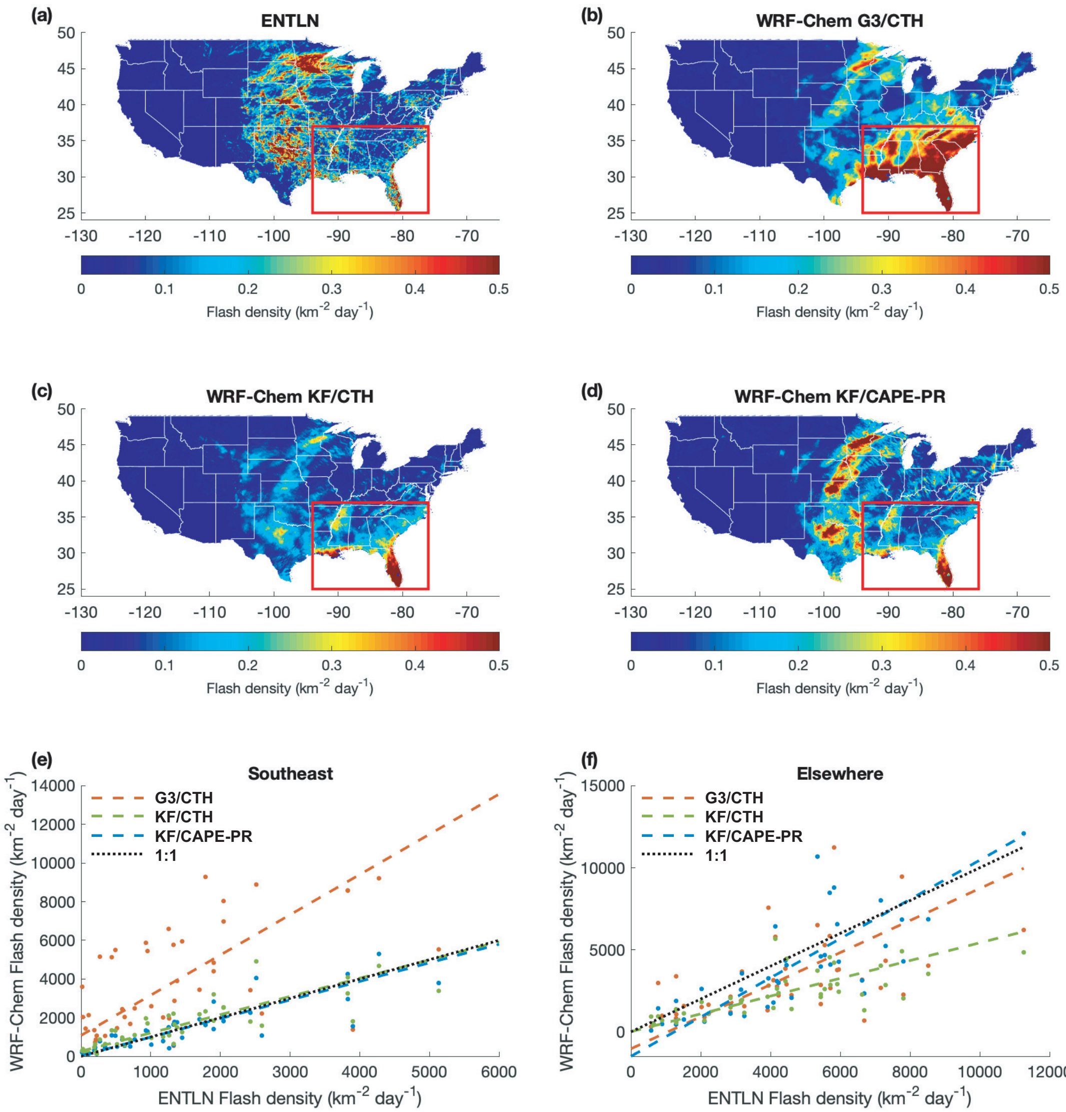

Figure R1: Observed flash densities from the ENTLN dataset **(a)** and WRF-Chem using three coupled convective-lightning parameterizations, the G3/CTH parameterization **(b)**, the KF/CTH parameterization **(c)** and the KF/CAPE-PR parameterization **(d)**, respectively. The correlation of total flash density per day between WRF-Chem outputs and ENTLN for the southeastern US (denoted by the red box in **a**-**d**) is shown in panel **(e)** and the correlation for elsewhere in CONUS is shown in **(f)**. The model using G3/CTH is in red, KF/CTH is in green, and KF/CAPE-PR is in blue. Dash lines are corresponding fits. For slope and R^2, see Table 1.

		G3/CTH	KF/CTH	KF/CAPE-PR
Southeastern	Slope	2.08	0.94	0.96
	R^2	0.30	0.67	0.72
Elsewhere	Slope	0.98	0.54	1.19
	R^2	0.27	0.48	0.62

Table R1: Correlation statistics between observed and modeled (G3/CTH, KF/CTH, KF/CAPE-PR) flash density per day averaged by regions

> "**The CAPE-PR parameterization with a regional scaling factor of 0.5 in the southeastern US, is coupled with Kain Fritsch convective scheme (KF/CAPE-PR) to generate lightning for the continental US.** We show that the **KF/CAPE-PR** scheme yields an improved representation of lightning flashes in WRF when comparing against flash density from the Earth Networks Total Lightning Network. Compared to the cloud top height (CTH) lightning parameterization **coupled with Grell 3D convective scheme (G3/CTH)** used in WRF-Chem, simulated NO_2 profiles using the **KF/CAPE-PR** parameterization exhibit better agreement with aircraft observations in the middle and upper troposphere..."

The conclusion is also modified accordingly:

> "We implement an alternative lightning parameterization based on convective available potential energy and precipitation rate into WRF-Chem **and couple it with Kain Frisch convective scheme**. We evaluate its performance in simulating lightning NO_x. We first validate it by comparing against lightning observations and conclude that the **KF/CAPE-PR** parameterization with a regional scaling factor of 0.5 in the southeastern US improves...."

Specific comments: p. 2, line 11: give examples of near-filed analyses (e.g., Huntrieser et al. (several papers); Pollack et al., 2016). add another sentence: Near-field estimates of LNOx per flash have also been make through use of cloud-resolved models with LNOx production constrained by observed flashes and aircraft data from storm anvils (e.g., DeCaria et al., 2005; Ott et al., 2010; Cummings et al., 2013).

We added the text:

> "In near-field approaches the total NO_x from direct observation close to the lightning flashes is divided by the number of flashes from a lightning observation network to yield the NO_x per flash **(e.g. Schumann and Huntrieser, 2007; Huntrieser et al., 2009; Pollack et al., 2016). Near-field estimates of LNO_x per flash have also been made through use of cloud-resolved models with LNO_x constrained by observed flashes and aircraft data from storm anvils (e.g. DeCaria et al., 2005; Ott et al., 2010; Cummings et al., 2013)**. In contrast..."

p. 3, line 4: flash count frequency distribution over time,....
Added.

> "Wong et al. (2013) showed that a model using the CTH lightning parameterization simulates erroneous flash count frequency distribution **over time** while the integrated lightning flash count is consistent with the observation. "

p. 3, ine 9: need a reference for CAPE-PR here
Added

> "we implemented the CAPE-PR lightning parameterization **(Romps et al., 2014)** into WRF-Chem"

p. 3, line 15: Provide the time periods that are being simulated here. The reader needs to know if 2012 emissions are appropriate. Otherwise, the reader doesn?t learn the simulating periods until Section 2.3.
We added the study time period at the beginning of Section 2.1

> "This study applies the Weather Research and Forecast Model coupled with Chemistry (WRF-Chem) version 3.5.1 **to the time periods May to June, 2012 and August to September, 2013.**"

p. 4, line 16: the neutral buoyancy level
Corrected, thanks.

p. 4, lines 14-22: Won't the difference in flash rate between the two model runs be partially due to different convective parameterizations and partially due to different flash rates schemes? Here is where the authors either need to add another model run (KF with CAPE-PR) or start calling the two runs Grell3D/CAPE-PR and KF/CTH.
We added another model run referred as KF/CTH. The revisions are summarized above.

p. 4, line 29: was this detection efficiency value applied to the ENTLN data? Flash counts should be divided by 0.7.
We did a more thorough survey on the detection efficiency of ENTLN and decided to correct it to 88%. As the detection efficiency of ENTLN varies by time, region, lightning type and the reference datasets, the choice of 88% is based on following elements:

1. Local studies comparing ENTLN to rocket-triggered lightning data at Florida report detection efficiency of ENTLN to be 89% during 2009-2012 (Mallick et al., 2015) and 99% during 2014-2015 (Zhu et al., 2017).

2. Lapierre et al. (submitted) found out the average detection efficiency for flashes observed by ENTLN+NLDN was 88% over CONUS relative to space-based TRMM-LIS during May-August, 2014.

In the context, we decide not to correct the ENTLN flash counts using the detection efficiency (88%) referring to Fig S1. In Fig S1, we matched the ENTLN data to LIS flashes both in time and space after the correction of LIS data based on its detection efficiency (Cecil et al., 2014) during the study period May 13-June 23, 2012. Both two datasets are then summed on 0.5°x0.5° grid cells and they show median correlation with slope of 1.0. While it indicates uncorrected ENTLN during study time period shows the best agreement with LIS observation, the detection efficiency of ENTLN is only considered as a source of uncertainty when comparing modeled lightning flashes against ENTLN. We added the following text in Section 2.2:

> "Compared to National Lightning Detection Network (NLDN), ENTLN is selected for high detection efficiencies of both CG and IC flashes. The average detection efficiency for total flashes observed by ENTLN was 88% over CONUS relative to the space-based Tropical Rainfall Measurement Mission (TRMM) Lightning Imaging Sensor (LIS) (Lapierre et al. (submitted), private communication). Shown in Fig. S1, we matched the ENTLN data to LIS flashes both in time and space after the correction of LIS data based on its detection efficiency (Cecil et al., 2014) during May 13-June 23, 2012. It shows a median correlation ($R^2 = 0.51$) with the slope of 1.0, indicating the ENTLN data during the study time period is in agreement with the LIS observation. We use the ENTLN for analysis as reported and consider the detection efficiency of ENTLN as a source of uncertainty when comparing the modeled lightning flashes."

p. 5, line 12: should the reference by 2019 instead of 2018? 2019 is the one with v3.0B in the title.

Laughner et al. (2018) is the first paper describing the upgrade to BEHR v3.0B and Laughner et al. (2019) is the followup paper evaluating BEHR 3.0B. In the context Laughner et al. (2018) should be the one for citation

p. 5, line 15: what is "NASA tropopause temperature"? Is it from the MERRA-2 product? We reformatted the sentence for clarification:

> "First, Instead of calculation based on temperature profiles from WRF-Chem (Mak et al., 2018), the tropopause pressure is switched to **GEOS-5 monthly tropopause pressure which is consistent with NASA Standard Product (SP2)**."

p. 6, lines 14-15: But, how much of this improvement might be due to use of KF convection rather than Grell 3D convection? I don?t think you can conclude that one lightning scheme is better than the other with these two simulations that use different convection schemes.

We addressed this question in the response above. Please refer to our response to the first comment.

p. 8, lines 4-5: Need to point out that this is really only true for the CTH model runs for SEAC4RS. Both model and observations are very small in this layer for DC3.

We found using the percentage change in NO_2 profiles is misleading. The large bias in NO_2 comparing against DC3 observation between 400 to 700 hpa is due to the small values both in observation and simulation. To avoid this, we replace the percentage change with absolute difference in Fig. 2 (also labeled as Fig. R2 in this response).

In the context, we corrected the sentence to:

> "NO_x from both the observations and the models are very small in the middle troposphere between 400 hPa to 700 hPa."

p. 11, lines 4-5: Here again, this conclusion needs to be modified. See above.
We modified the conclusion accordingly, please refer to the response to the first comment.

p. 15, lines 1-2: Update Laughner and Cohen reference
Corrected, thanks!

Figure S1: Since LIS only observes a swath across this region for a few minutes each day, I assume that the ENTLN data are subsetted in time to match the LIS overpass times. Is this correct? If so, you need to say that in the caption. Or, if the ENTLN data are really for the entire day/night for all days, then the comparison is not valid. The c) panel of the figure does not look to be correct. Is it really ENTLN - LIS rather than LIS - ENTLN? I see some pixels where neither appears to be correct.

We add a more detailed description of Fig. S1 in Section 2.2:

> "Shown in Fig. S1, we matched the ENTLN data to LIS flashes both in time and space after the correction of LIS data based on its detection efficiency (Cecil et al., 2014) during May 13-June 23, 2012. It shows a median correlation ($R^2 = 0.51$) with the slope of 1.0, indicating the ENTLN data during the study time period is in agreement with the LIS observation...."

It's ENTLN-LIS. Thanks for pointing out the error, we update Fig. S1 (labeled as Fig. R3 in this response) and also modified the caption:

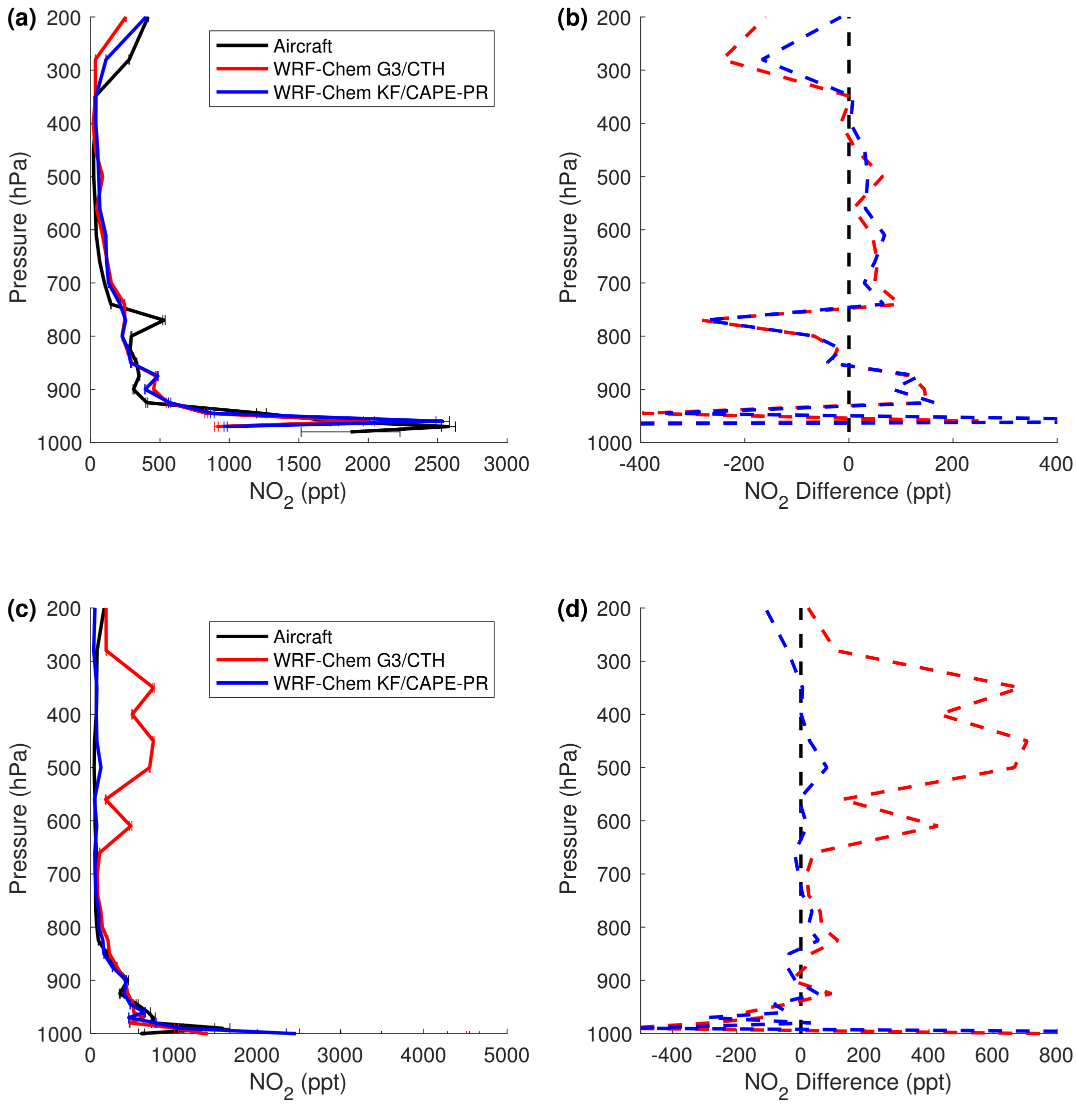

Figure R2: Comparison of WRF-Chem and aircraft NO_2 profiles from the **(a,b)** DC3, **(c,d)** SEAC4RS campaigns. Vertical NO_2 profiles are shown in **(a,c)**, the solid line is the mean of all profiles and the bars are 1 standard deviation for each binned level. The corresponding absolute difference compared to observations are shown in **(b,d)**. Aircraft measurements are shown in black, WRF-Chem using G3/CTH parameterization in red and WRF-Chem using KF/CAPE-PR parameterization in blue.

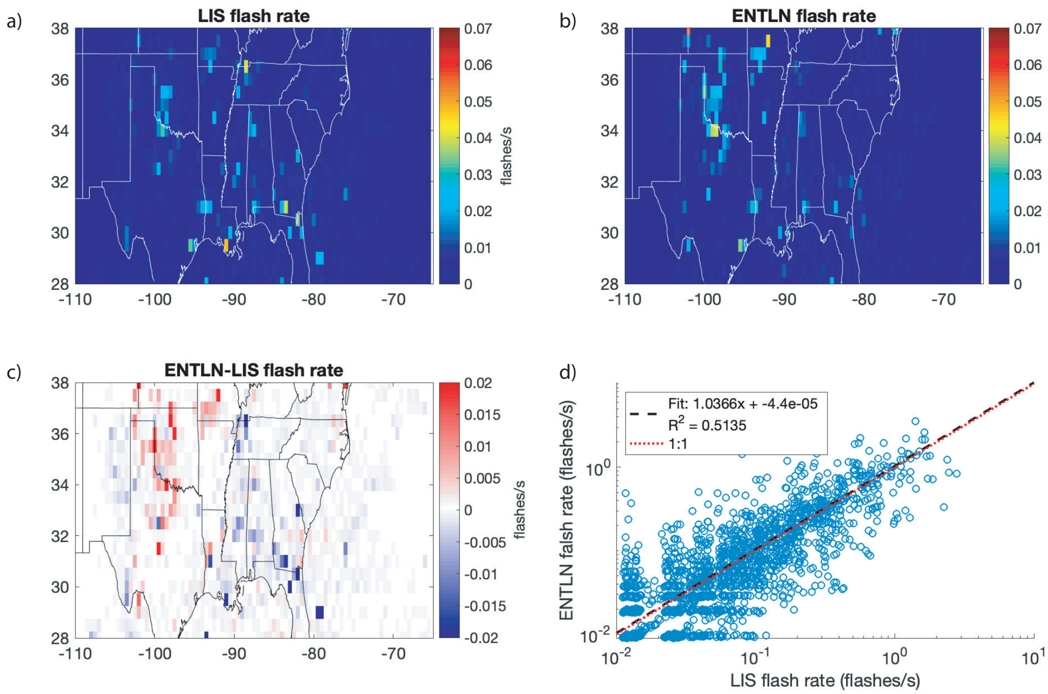

Figure R3: Comparison between flash rates observed by ENTLN and Lightning Imaging Sensor (LIS). **ENTLN data is matched to corrected LIS flashes both in time and space during May 13-June 23, 2012, and both datasets are summed onto 0.5°x 0.5°grid spacing.** **(a,b)** shows the spatial pattern of lightning flash rates measured by LIS **(a)** and ENTLN **(b)**. The plot region covers 20°N - 38°N and 130°W - 65°W. **(c,d)** are corresponding absolute difference and scatter plots between LIS and ENTLN. **LIS data is corrected using the detection efficiency from Cecil et al. (2014).**

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Response to Anonymous Referee #2

Qindan Zhu, Joshua L. Laughner and Ronald C. Cohen

July 12, 2019

We thank the reviewers for very careful reading of both the main article and the supplement. Our responses to the reviewer's comments and detailed changes made to the manuscript 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 "R*n*" 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. Figures, tables, and equations numbered normally refer to the numbers in the original discussion paper.

While I think the paper covers interesting areas of research, the results from this paper are in line with previous studies. It would be a stronger paper if more significant improvements in either modeling or analysis results were made.

We appreciate the reviewer's positive comments on the overview of the paper. The significance of this study is to improve model performance in representing lightning and lightning NO_x in WRF-Chem and to directly couple those components to a high spatial resolution OMI NO_2 retrieval.

Currently, except for convective resolved runs, lightning NO_x scheme is disable by default in WRF-Chem, and the only option provided is combing CTH lightning parameterization with Grell 3D convective scheme (G3/CTH). Our work implements CAPE-PR lightning parameterization coupled with Kain Fritsch convective scheme into WRF-Chem, and suggests it being a better proxy for lightning in WRF-Chem. To our knowledge the CAPE-PR parameterization has not previously been coupled with chemistry.

On top of the improvement in representing lightning and lightning NO_x, we further investigate its effect on satellite NO_x retrievals, and estimate the lightning NO_x production rate by comparing modeled NO_2 VCD against satellite observations. Our study strongly suggests that accurately the retrieving NO_x VCD requires a priori profiles produced from model simulation with reliable lightning NO_x. While this is not surprising our quantitive results are a useful point of reference.

Some of the discussion is rather odd. A few of the issues were raised in my initial review but no modifications were made to the paper. These issues are not complex but do require more in-depth thinking than what was given in the manuscript. I think that the uncertainties in lightning measurements and modeling, satellite measurements and retrievals, and in situ measurements should be clearly acknowledged.

We made no scientific changes after the access review as is consistent with ACP policy. This was not meant to ignore the referee comments but rather to respond here in the open discussion.

(1) Lightning modeling uncertainty. I pointed out in the initial review that the abstract statement in line 11-12 on page 1 should be deleted. Comparing NO2 VCDs between two parameterizations is not meaningful because the amount of lightning NOx (LNOx) is a function of specified IC/CG ratio, NOx yield per flash, and the vertical distribution of LNOx. Values can be easily modified to produce similar LNOx values between the two parameterizations. For the same reason, line 12-13 on page 11 in the conclusion section should be deleted. The NOx production rate per (IC or CG) flash cannot be determined with available observations.

We agree that changing some lightning characteristics will affect NO_2 VCD. Our point here is emphasizing the non-negligible effect of lightning parameterizations on NO_2 VCD retrievals. Except for giving the exact value of changes in VCD, we modify the text in abstract to:

> "Using a lightning NO_x production rate of 500 mol NO flash^{-1}, the a priori NO_2 profile generated by the simulation with the KF/CAPE-PR parameterization reduces the air mass factor for NO_2 retrievals by 16% on average in the southeastern US on the late spring and early summer compared to simulations using the G3/CTH parameterization. **This causes an average change in NO_2 vertical column density four times higher than the average uncertainty.**"

In Section 3.3, we added the following text to compare our results to the uncertainty in BEHR VCD retrievals:

> "We follow the same algorithm used in Laughner and Cohen (2017) to determine if the result is significant. The overall uncertainty due to AMF calculation for BEHR v3.0B is smaller than 30% during the study period (Laughner et al., 2019). As each grid in Fig. 3(a) is the average of 45±9 pixels, the reduced uncertainty is less than 4.5%. The overall change in VCD is four times larger than the reduced uncertainty. The switch of lightning parameterization leads to changes in VCD exceeding the averaged uncertainty in ˜94% of pixels in the southeast region of US."

We respectfully disagree with the reviewer to delete the line 12-13 on page 11. While the elements listed above affect the lightning NO_x estimate, numerous studies have used the far-field approach to constrain the lightning NO_x production rate with uncertainty considerably accounted for (Hudman et al., 2007; Martin et al., 2007; Jourdain et al., 2010; Miyazaki et al., 2014; Liaskos et al., 2015; Laughner and Cohen, 2017; Nault et al., 2017). To be more specific, as we assume CG and IC produce the same amount of lightning NO_x per flash, and we validate the KF/CAPE-PR parameterization by comparing the total flash rate against

ENTLN, IC/CG ratio does not affect our estimate of lightning NO_x production rate. For the vertical distribution of LNO_x, we use the modified version of profiles from Ott et al. (2010) based on the results from cloud-resolving model, which is consistent with Laughner and Cohen (2017). Overall we have optimized the affecting factors to reduce the uncertainty of lightning NO_x production rate estimate using the far-field approach.

The abstract statement in line 5-6 on page 1 is based on model comparison with SEAC4RS data, when the lightning activity in the Southeast is relatively low. Unless it is a science objective, aircraft in missions like SEAC4RS usually flies in sunny days and steers away from thunderstorms. The 50% reduction is also only specific to the CAPE-PR parameterization in this paper and it offers little value to other lightning parameterizations.

The abstract statement is based on model comparison with DC3 data, referring to Section 3.1. Over the time period, the lightning activity in relatively high as DC3 is designed to observe deep convection. To avoid the confusion, we add the text in Section 3.1:

> "The lightning parameterizations are compared against observations from ENTLN in Fig. 1. Each of the datasets is averaged from May 13 to June 23, 2012, **covering DC3 field campaign.**"

The 50% reduction in the southeast region of US is applied to improve the model performance in representing lightning when it uses CAPE-PR as the lightning parameterization. We are not intended to apply this value to other lightning parameterizations. We made the following changes in the abstract for clarification:

> "We implement a lightning parameterization using the product of convective available potential energy (CAPE) and convective precipitation rate (PR) into Weather Research and Forecasting-Chemistry (WRF-Chem) model. **The CAPE-PR parameterization with a regional scaling factor of 0.5 in the southeastern US, is coupled with Kain Fritsch convective scheme (KF/CAPE-PR) to generate lightning for the continental US.** We show that.... "

It is not surprising that the CTH based parameterization doesn't work well. It's a poor choice in WRF-Chem, but I presume that it's a good reason to compare it to a new scheme. It would be better that other parameters, say those used by Allen et al. (2002) and more recently Luo et al. (2017), were used.

We agree with the reviewer that CTH based parameterization fails to reproduce good representation of lightning flashes in models. Even it is a poor choice, the CTH parameterization coupled with Grell 3D convective scheme is the only option provided by WRF-Chem to include lightning and lightning NO_x into model simulation at cloud parameterized scale. Better lightning parameterizations have been discussed and evaluated in other models, for instance, WRF-CMAQ (Luo et al., 2017) and GEOS-START (Allen and Pickering, 2002). To our knowledge there is no other lightning parameterization has been implemented into WRF-Chem at convective parameterized scale. In our study, we implement CAPE-PR lightning parameterization and find it is an extraordinarily good proxy for lightning flash. While it is generally hard to compare the results across the literatures as different models, time windows and regions are chosen, we can roughly compare our results with Luo et al. (2017), which successfully implemented most common lightning parameterizations into WRF-CMAQ. Our correlation results are better than the optimal case analyzed in Luo et al. (2017) ($R^2 = 0.56$, Slope = 0.87). Therefore in our study, we only implements CAPE-PR into WRF-Chem. However, in the future, we will intend to implement more lighting parameterizations studied in Luo et al. (2017) into WRF-Chem and test their performances in representing lightning and lightning NO_x.

A third issue raised in the initial review is line 17-19 on page 4. Which convection scheme was used for the results presented later in the paper? Are there differences? I did not find the results comparing the two convective schemes. Zhao et al. (2009) compared MM5 Grell and WRF KF schemes and found large differences. My understanding is that the Grell scheme in WRF does not have the large bias in MM5. It should be discussed.

We recognize the inconsistency in convective scheme will affect the robustness of our conclusion. We agree that the switch of convective scheme will affect the parameterized lightning flashes and as a result, we can't conclude the improvement in lightning flash representation in WRF-Chem is solely due to the new implemented CAPE-PR lightning parameterization. Therefore, we made another WRF-Chem run with Kain Frisch convective scheme and CTH lighting parameterization, referred as "KF/CTH". We add the description of this model run in Section 2.1.

> "We analyze WRF-Chem outputs from three model runs. The first run, **referred as "G3/CTH"**, is consistent with Laughner and Cohen (2017); it selects the Grell 3D ensemble cumulus convective scheme (Grell, 1993; Grell and Dvnyi, 2002) and the CTH lightning parameterization. The Grell 3D convective scheme readily computes the neutral buoyancy level which serves as the optimal proxy for cloud top height (Wong et al., 2013). **The "G3/CTH" is the only option for the coupled convective-lighting parameterization used in WRF-Chem at a non-cloud resolving resolution (12 km). In addition, we run WRF-Chem with the CTH lightning parameterization coupled with the Kain-Fritsch cumulus convective scheme (Kain and Fritsch, 1990; Kain, 2004) ("KF/CTH") to test the effect of switching convective schemes. In the "KF/CTH" parameterization, the cloud top height is the level where the updraft vertical velocity equals to zero. Another run, referred as "KF/CAPE-PR",** selects the Kain-Fritsch cumulus convective scheme and the CAPE-PR lightning parameterization described above..."

In the context, we change CTH to either "G3/CTH" or "KF/CTH" and change CAPE-PR to "KF/CAPE-PR" accordingly. In Section 3.1, we added comparison of "KF/CTH" parameterized lightning flashes against ENTLN both in the text, Fig 1 and Table 1 (also labeled as Fig. R1 and Table R1 in this response).

> "The G3/CTH parameterization fails to reproduce the spatial pattern of flashes observed by ENTLN over the CONUS. **Compared to the G3/CTH, the**

		G3/CTH	KF/CTH	KF/CAPE-PR
Southeastern	Slope	2.08	0.94	0.96
	R^2	0.30	0.67	0.72
Elsewhere	Slope	0.98	0.54	1.19
	R^2	0.27	0.48	0.62

Table R1: Correlation statistics between observed and modeled (G3/CTH, KF/CTH, KF/CAPE-PR) flash density per day averaged by regions

> **KF/CTH parameterization improves the spatial correlation in the southeast region of US.** The KF/CAPE-PR parameterization better captures the spatial distribution of flash densities both in the southeast region and elsewhere in CONUS."
>
> "The model using the KF/CAPE-PR lightning parameterization yields a tight correlation and slope close to the unity over the US domain. In the southeastern US, the R^2 increases from 0.3 to 0.7 and slope is reduced from 2.08 to 0.96 with the KF/CAPE-PR parameterization compared to the G3/CTH. **The slope for KF/CTH is comparable to KF/CAPE-PR while the R^2 for KF/CAPE-PR is slightly higher.**"
>
> "Elsewhere in CONUS, **the R^2 for KF/CAPE-PR improves significantly to 0.6 compared to both G3/CTH and KF/CTH**. The slope for KF/CAPE-PR is 1.19, which is within the uncertainty of the detection efficiency of ENTLN. In general the KF/CAPE-PR lightning parameterization captures the day-to-day variation in flash densities better than the G3/CTH **and KF/CTH parameterizations as shown by the improved R^2 values.**"

The abstract is modified to:d

> "**The CAPE-PR parameterization with a regional scaling factor of 0.5 in the southeastern US, is coupled with Kain Fritsch convective scheme (KF/CAPE-PR) to generate lightning for the continental US.** We show that the **KF/CAPE-PR** scheme yields an improved representation of lightning flashes in WRF when comparing against flash density from the Earth Networks Total Lightning Network. Compared to the cloud top height (CTH) lightning parameterization **coupled with Grell 3D convective scheme (G3/CTH)** used in WRF-Chem, simulated NO_2 profiles using the **KF/CAPE-PR** parameterization exhibit better agreement with aircraft observations in the middle and upper troposphere..."

The conclusion is also modified accordingly:

> "We implement an alternative lightning parameterization based on convective available potential energy and precipitation rate into WRF-Chem **and couple**

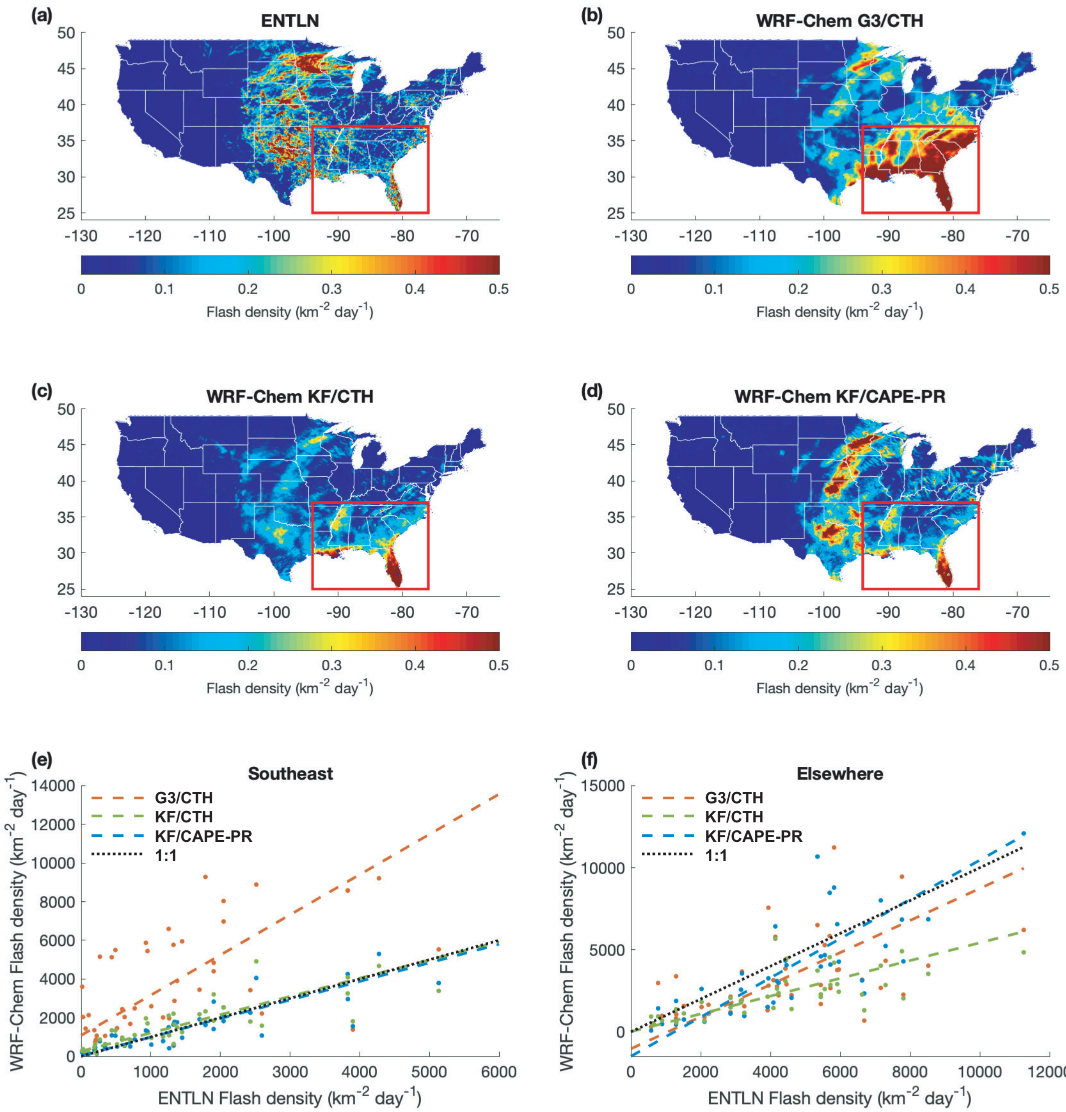

Figure R1: Observed flash densities from the ENTLN dataset **(a)** and WRF-Chem using three coupled convective-lightning parameterizations, the G3/CTH parameterization **(b)**, the KF/CTH parameterization **(c)** and the KF/CAPE-PR parameterization **(d)**, respectively. The correlation of total flash density per day between WRF-Chem outputs and ENTLN for the southeastern US (denoted by the red box in **a**-**d**) is shown in panel **(e)** and the correlation for elsewhere in CONUS is shown in **(f)**. The model using G3/CTH is in red, KF/CTH is in green, and KF/CAPE-PR is in blue. Dash lines are corresponding fits. For slope and R^2, see Table 1.

it with Kain Frisch convective scheme. We evaluate its performance in simulating lightning NO_x. We first validate it by comparing against lightning observations and conclude that the **KF/CAPE-PR** parameterization with a regional scaling factor of 0.5 in the southeastern US improves...."

(2) Lighting measurement uncertainty. Section 2.2 described the ENTLN lightning data and its use in this paper. Figure 1 in the paper by Luo et al. (2017) compared the lightning distributions observed by ENTLN to NLDN. For the data they used, ENTLN had more IC and CG lightning flash rates than NLDN. The lower IC flash rates in NLDN are likely due to a lower detection efficiency. However, the CG flash rates in NLDN are also lower. As implied by Luo et al. (2017), the NLDN CG flash rate data have been the ?gold standard? in previous LNOx studies over the US. The distributions of NLDN flash rates were also different from ENTLN data in that work. The uncertainties of ENTLN data should be acknowledged. It is another reason the two statements on lightning parametrization in the abstract (discussed above) are not robust science results and should be taken out. Some explanation is due for the reasons of not using NLDN data.

While both NLDN and ENTLN have high detection efficiency (>90%) for CG flashes, we also recognize that ENTLN observes more CG flashes than NLDN. Shown in Fig. R2, we average the flashes density over CONUS both from ENTLN and NLDN between May 13 to June 23 2012. The daily averaged CG flash density from ENTLN is tightly correlated with those from NLDN with slope of 1.5. It can be explained by discrepancy in the grouping criterions applied to produce flash counts between NLDN and ENTLN. ENTLN groups all pulses within 10 km and 700 ms of each other as a single flash, and NLDN uses 10 km and 1000 ms as the threshold. In consequence, for the same amount of CG pulses measured by both lightning observation network, ENTLN produces more flashes than NLDN according to the grouping algorithm.

In our study, we use the total lightning flashes, including cloud-to-ground (CG) and Intra-cloud (IC) flashes, to validate the lightning parameterization in WRF-Chem. Compared to NLDN, ENTLN is selected for high detection efficiencies in both CG and IC flashes.The average detection efficiency for total flashes observed by ENTLN was 88% over CONUS relative to space-based TRMM-LIS (Lapierre et al., submitted) . We also evaluate ENTLN by comparing against LIS data during our study period, and the result indicates ENTLN represent total flash rates very well. We add the following text into Section 2.2:

"Compared to National Lightning Detection Network (NLDN), ENTLN is selected for high detection efficiencies of both CG and IC flashes. The average detection efficiency for total flashes observed by ENTLN was 88% over CONUS relative to the space-based Tropical Rainfall Measurement Mission (TRMM) Lightning Imaging Sensor (LIS) (Lapierre et al. (submitted), private communication). Shown in Fig. S1, we matched the ENTLN data to LIS flashes both in time and space after the correction of LIS data based on its detection efficiency (Cecil et al., 2014) during May 13-June 23, 2012. It shows a median correlation ($R^2 = 0.51$) with the slope of 1.0, indicating the ENTLN data during the study time period is in agreement with the LIS observation. We use the ENTLN for

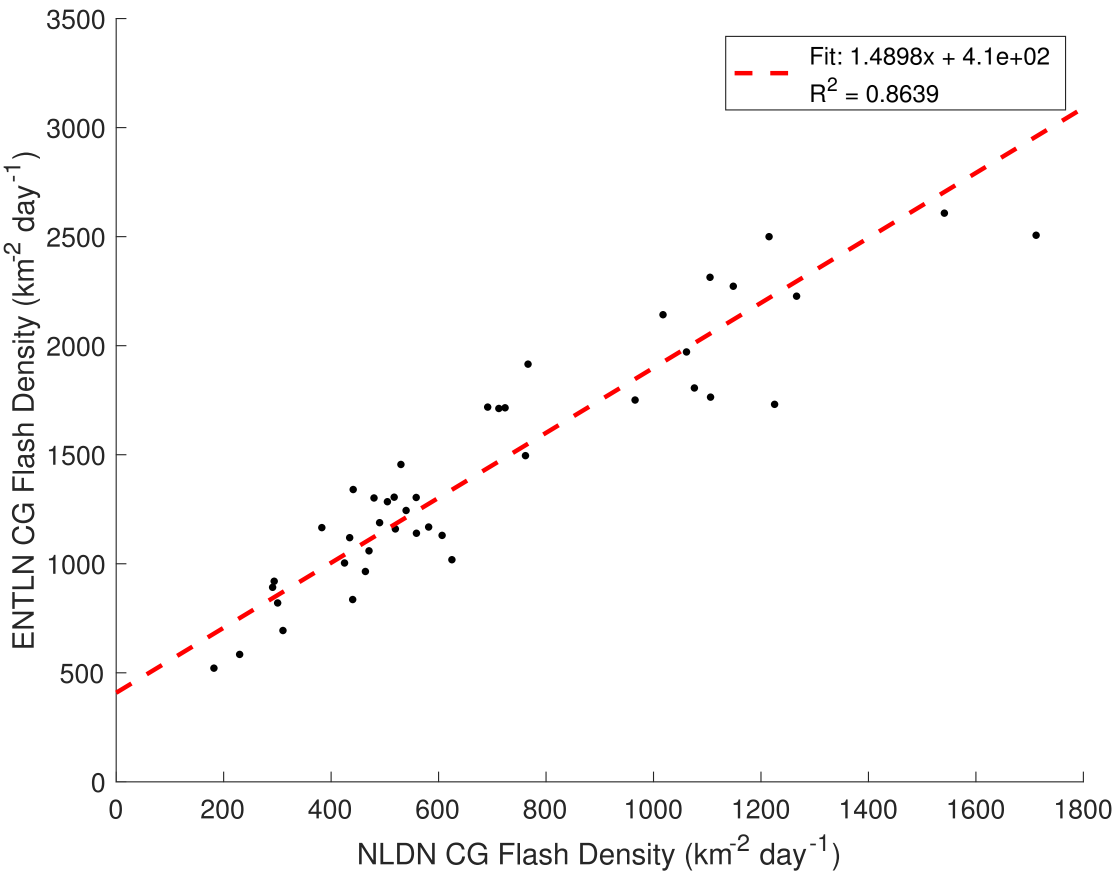

Figure R2: Comparison between CG flash density per day observed by NLDN and ENTLN. The data spans May 13 to June 23, 2012.

> analysis as reported and consider the detection efficiency of ENTLN as a source of uncertainty when comparing the modeled lightning flashes."

The comparison in Fig. S1 is inadequate since it is only for one day only. LIS observations are mostly for IC flash rates and there are good reasons to believe that a CG flash produces more NOx than an IC flash (see the relevant discussion in Luo et al. (2017)).

The comparison in Fig.S1 covers May 13 to June 23, 2012, which is consistent with the study period for comparing WRF-Chem lightning parameterizations against ENTLN. TRMM-LIS has been shown to observe CG and IC flashes equally well and the detection efficiency varies between 0.69 to 0.88 by hour of the day. To better explain Fig. S1, we add the following text in Section 2.2.

> "Shown in Fig.S1, we matched the ENTLN data to LIS flashes both in time and space after the correction of LIS data based on its detection efficiency (Cecil et al., 2014) during May 13-June 23, 2012. It shows a median correlation ($R^2 =$ 0.51) with the slope of 1.0, indicating the uncorrected ENTLN data during the study time period shows the best agreement with LIS observation."

We also add more detailed description in the caption of Fig.S1 (also labeled as Fig.R3 in this response):

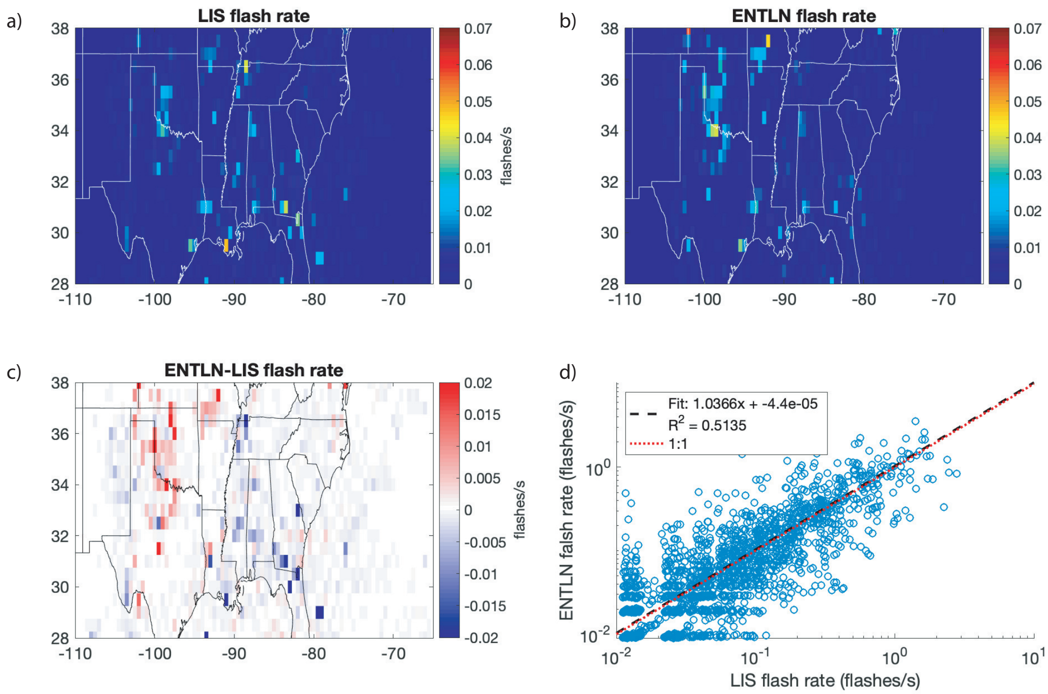

Figure R3: Comparison between flash rates observed by ENTLN and Lightning Imaging Sensor (LIS). **ENTLN data is matched to corrected LIS flashes both in time and space during May 13-June 23, 2012, and both datasets are summed onto 0.5°x 0.5°grid spacing.** **(a,b)** shows the spatial pattern of lightning flash rates measured by LIS **(a)** and ENTLN **(b)**. The plot region covers 20°N - 38°N and 130°W - 65°W. **(c,d)** are corresponding absolute difference and scatter plots between LIS and ENTLN. **LIS data is corrected using the detection efficiency from Cecil et al. (2014).**

The difference between CG and IC flashes relative to NO_x production rate is disputable. We assume CG and IC flashes produce the same amount of NO_x per flash in this study primarily due to two reasons:

1. Among literatures suggesting CG flash produces more NO_x than an IC flash, there is no agreement on quantifying the difference (Koshak et al., 2014; Carey et al., 2016; Luo et al., 2017; Lapierre et al., submitted).
2. The disputation is partially due to the ambiguity in the concept of lightning flash. By definition, a flash contains multiple strokes, and strokes are impulsive current pulses measured directly by the sensors configured in the lightning observation network. A lightning flash is classified as CG when it contains a return stroke, otherwise it is classified as IC. Lapierre et al. (submitted) indicates that a CG stroke produces much more (10 times) LNO_x than a IC stroke, of which the conclusion is consistent with (Koshak et al., 2014). However, LNO_x derived from flash rather than stroke will obscure the IG and CG variability as a IC flash contains more strokes than a CG flash.

(3) OMI retrieval uncertainties. The comparisons of Figures 4 and S2 can only be used as a qualitative not quantitative measure of lightning NOx in the model. For clean regions of the SE, the difference is on the order of 1x1015 molec cm-2. Considering that the OMI uncertainty is larger than 1.5x1015 molec cm-2, it is difficult to say which sensitivity simulation is quantitatively better. There are additional uncertainties in OMI retrievals including surface albedo, cloud, and background noise. With the uncertainties in mind, the difference among the lightning sensitivity simulations in Figure S3 is therefore insignificant. Appropriate discussion on the uncertainties of OMI retrievals and the implications for this paper should be included.

We appreciate reviewer's suggestion on discussing the uncertainty of OMI retrievals. However, we believe the results we shown in the manuscript is significant. The results represent the average of NO_2 VCD between May 13 to June 23, 2012, and 32 $\pm$ 6 pixels contribute to each value. While the global mean uncertainty for tropospheric NO_2 VCD retrievals is 1x10^{15} mole cm^{-2} (Bucsela et al., 2013), the reduced uncertainty in our analysis is $\sim 0.2 \times 10^{15}$ mole cm^{-2}, which is less than half of the RMSEs we calculated between BEHR retrievals and model simulations. We expand the description of the figures in the discussion:

> "Figure 4 shows the difference between satellite retrieved NO_2 VCD and model simulated NO_2 VCD without lightning NO_x **(a)** and with lightning NO_x production rate of 500 mol NO flash^{-1} **(b)** averaged between May 13 to June 23, 2012. Figure S2 shows difference plots with varied lightning NO_x production rates (400 and 665 mol NO flash^{-1}). The corresponding root-mean-square errors (RMSE) are included in Table S1. LNO_x production rate of 500 mol NO flash^{-1} yields the lowest RMSE of 0.41$\times10^{15}$ mole cm^{-2} between modeled and observed NO_2 VCD over CONUS. This is at the high end of previous estimates of the lightning NO_x production rate (16-700 mol NO flash^{-1}). "

> "The RMSE for urban areas (top 5% of NO_2 VCD simulated by WRF-Chem without LNO_x) remains at high value ($\sim$0.9-1.3$\times10^{15}$ mole cm^{-2}) when switching

the LNO_x production rate. It indicates that the bias in the modeled VCD over urban areas is more likely due to surface NO_2. The RMSE for non-urban areas shows pronounced change with varied LNO_x production rate. Excluding urban areas lowers the RMSE to 0.37×10^{15} mole cm^{-2} for LNO_x production rate of 500 mol NO $flash^{-1}$. The RMSEs are significant considering the uncertainty for retrievals. During the average time period, 32 ± 6 pixels contribute to each value in the plots. While the global mean uncertainty for tropospheric NO_2 VCD retrievals is 1×10^{15} mole cm^{-2} (Bucsela et al., 2013), the reduced uncertainty in our analysis is $\sim0.2\times10^{15}$ mole cm^{-2}. The calculated RMSEs are twice of the uncertainty. "

There is no description on how OMI data under high cloud-fraction conditions are treated. Those data cannot be used in the comparisons of Figures 4 and S2.

The pixels with cloud fraction larger than 0.2 are filtered out in our analysis. We add the text in the caption of Fig.4 for clarification.

> "**Figure 4.** Difference in NO_2 VCD between BEHR retrievals and WRF-Chem. **(a)** excludes LNO_x in model simulation, **(b)** adds LNO_x emission with production rate of 500 mol NO $flash^{-1}$. **(c)** includes the same LNO_x emission as **(b)** but uses NO_2 profiles scaled upward by 60% at pressure lower than 400 hPa. **The average time covers May 13 to June 23, 2012. Pixels with cloud fraction larger than 0.2 are filtered out in the analysis.**"

(4) Uncertainty of the upper tropospheric NO2/NOx ratio. In sections 4, the uncertainty of the upper tropospheric NO2/NOx ratio was discussed. This issue doesn?t affect model comparison with in situ observations when NO measurements are available. Comparisons with in situ NO, O3, and JNO2 should be included with the discussion of Figure 2. To evaluate model lightning NOx simulations, using NO measurements will get around the uncertainty of NO2/NOx ratio. Therefore, the last statement of the conclusion section (line 13-14 on page 11) is inappropriate and should be removed. This uncertainty affects the retrieval of OMI data only if the unknown interferences by other nitrates are insignificant. Furthermore, the uncertainties oof OMI data may mask out the effects. More detailed discussion should be included.

While NO measurements are available during DC3 and SEAC4RS field campaigns, we disagree that the uncertainty of NO_2/NO_x ratio will not affect the model results. Our results are consistent with Silvern et al. (2018). In Figure 1 from Silvern et al. (2018), they compared the profiles of NO/NO_2 and relative quantities on SEAC4RS flights and concluded there is no systematic model bias in ozone, temperature, or JNO_2 that would explain the error in NO/NO_2.

The incorrect nitrate chemistry forming PNs/ANs/HNO_3 will affect the result, however, we argue the error from the underestimated NO_2/NO_x ratio is still significant:

1. We use a customized version of the Regional Atmospheric Chemistry Mechanism version 2 (RACM2), the details are described by Zare et al. (2018), which has a very

detailed nitrate mechanism, so any errors should be smaller than in most model comparisons.

2. Nault et al. (2017) found a 33% error in upper tropospheric NO_2 caused by the incorrect nitrate chemistry before the modified mechanism is implemented. In this study, if we assume that most of the LNO_x falls in pressure lower than 400 hPa, then given that +60% NO_2 is too much NO_2 in the column and the +0% was too little, a ballpark estimate of ∼30% seems reasonable. That's the same order as Nault et al. (2017) saw for nitrate chemistry, so the NO_2/NO_x ratio can be significant even if nitrate chemistry is poorly constrained.

(1) P. 4, Line 26-27, Eq. (2), Luo et al. (2017) used a formula of $f = a_0 \times x^{a_1} + a_2 \times x \times y + a_3 \times y^{a_4}$, what is the reason for not including the other terms? What are the reasons for not using UMF or CPR?

The formula used in Luo et al. (2017) is based on the power law relationship. The CAPE-PR lightning parameterization used in the paper is modified based on Romps et al. (2014), from which they found tight relationship between observed lightning flash rate and CAPE times precipitation rates both from measurements.

As we mentioned above, except for CTH, neither UMF or CPR has been implemented into WRF-Chem. Our study implements CAPE-PR lightning parameterization into WRF-Chem and find out that it reproduce lightning flashes well comparing against lightning observation. In the future, more lightning parameterizations will be implemented to WRF-Chem in order to further improve its performance in representing lightning and lightning NO_x.

(2) P. 8, Figure 3, Is the large urban VCD change mostly due to Orlando? Figure 1 shows very little lightning activity in the other SE region.

Figure 1 and Figure 3 correspond to different time periods. Figure 1 shows the spatial distribution of lightning flash density averaged from May 13 to Jun 23, 2012 covering DC3; Figure 3 shows the change of VCD averaged from Aug 01 to Sep 23, 2013 as Figure 2 shows large difference in NO_2 profiles during SEAC4RS. As ENTLN is only available upon request, we currently have no ENTLN data covering the same time period as Figure 3 . However, we find out the Figure 3 **(b)** remain unaffected if we mask out the Florida area, thus the large urban VCD is not due to Orlando.

(3) P. 9, Line 6-7, the 19% value should be compared to Zhao et al. (2009).

Zhao et al. (2009) and this study are looking at different quantities. Zhao et al. (2009) discussed about how much lightning contributes to a modeled NO_2 column, while this study is looking at how the change in a priori profiles from different lightning parameterizations affects a retrieved VCD.

(4) Figures are hard to read in general.

We enlarge all figures within the space constraints of the ACP template.

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Lightning NO_2 simulation over the Contiguous US and its effects on satellite NO_2 retrievals

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Abstract. Lightning is an important NO_x source representing ~10% of the global source of odd N and a much larger percentage in the upper troposphere. The poor understanding of spatial and temporal patterns of lightning contributes to a large uncertainty in understanding upper tropospheric chemistry. We implement a lightning parameterization using the product of convective available potential energy (CAPE) and convective precipitation rate (PR) into Weather Research and Forecasting-Chemistry (WRF-Chem) modelfor North America. We show that the . The CAPE-PR parameterization with a regional scaling factor of 0.5 in the southeastern US, is coupled with Kain Fritsch convective scheme (KF/CAPE-PR) to generate lightning for the continental US. We show that the KF/CAPE-PR scheme yields an improved representation of lightning flashes in WRF when comparing against flash density from the Earth Networks Total Lightning Network. Compared to the cloud top height (CTH) lightning parameterization coupled with Grell 3D convective scheme (G3/CTH) used in WRF-Chem, simulated NO_2 profiles using the KF/CAPE-PR parameterization exhibit better agreement with aircraft observations in the middle and upper troposphere. While the Using a lightning NO_x production rate is of 500 mol NO flash^{-1}, using the a priori NO_2 profile generated by the simulation with the KF/CAPE-PR parameterization reduces the air mass factor for NO_2 retrievals by 16% on average in the southeastern US on the late spring and early summer ; yielding an overall 20% enhancement of the vertical column density compared to simulations using the CTH lightning parameterization. G3/CTH parameterization. This causes an average change in NO_2 vertical column density four times higher than the average uncertainty.

1 Introduction

Nitrogen oxides ($NO_x \equiv NO + NO_2$) are key species in the atmosphere atmospheric chemistry, affecting the oxidative capacity in the troposphere by regulating the ozone and hydroxyl radical concentrations (Crutzen, 1979). Anthropogenic sources (mainly fossil fuel combustion) are the largest contributor to the NO_x budget on a global scale. Natural sources of NO_x are also nonnegligible (Denman et al., 2007). While anthropogenic emissions of NO_x are intensively studied, natural sources are less understood (e.g. Delmas et al., 1997; Lamsal et al., 2011; Miyazaki et al., 2012). Lightning contributes to ~10% of NO_x budget on a global scale and represents over 80% of NO_x in the upper troposphere (UT) (Schumann and Huntrieser, 2007; Nault et al., 2017). Over the US, the anthropogenic NO_x emissions have been decreasing rapidly (Russell et al., 2012; Lu et al., 2015), making lightning an increasingly important source of NO_x and an increasingly large fraction of the source of column NO_2.

Ozone (O_3) in UT has long lifetime and leads to a more pronounced radiative effect than ozone elsewhere in the troposphere. Varying lightning NO_x emission (LNO_x) by a factor of four (123 to 492 mol NO flash^{-1}) yields up to 60% enhancement of UT O_3 and increases the mean net radiative flux by a factor of three (Liaskos et al., 2015). This range in the lightning NO_x production rate is similar to the current uncertainty of estimated lightning emission rates. FurtherFurthurmore, incorrect representation of LNO_x in a priori profiles for satellite NO_2 retrievals leads to biases in the retrieved NO_2 columns. This is exacerbated by the greater sensitivity of UV/Vis NO_2 retrievals to the UT NO_2(e.g. Laughner and Cohen, 2017; Travis et al., 2016).

When lightning occurs, NO is emitted as a result of high temperatures and NO_2 forms through rapid photochemistry. Numerous studies Studies report the estimated LNO_x production rate with ranges ranges widely from 16 to 700 mol NO flash^{-1} (DeCaria et al., 2005; Hudman et al., 2007; Martin et al., 2007; Schumann and Huntrieser, 2007; Huntrieser et al., 2009; Beirle et al., 2010; Bucsela et al., 2010; Jourdain et al., 2010; Ott et al., 2010; Miyazaki et al., 2014; Liaskos et al., 2015; Pickering et al., 2016; Pollack et al., 2016; Laughner and Cohen, 2017; Nault et al., 2017).

Two categories of methods, one emphasizing the near-field of lightning NO_x and the other the far-field, have previously been applied to estimate LNO_x. In near-field approaches the total NO_x from direct observation close to the lightning flashes is divided by the number of flashes from a lightning observation network to yield the NO_x per flash . (e.g. Schumann and Huntrieser, 2007; Hunt Near-field estimates of LNO_x per flash have also been made through use of cloud-resolved models with LNO_x constrained by observed flashes and aircraft data from storm anvils (e.g. DeCaria et al., 2005; Ott et al., 2010; Cummings et al., 2013). In contrast, the far-field approach uses downwind observations to constrain a regional or global chemical transport model. The emission rate of lightning NO_x is varied in the model (either ad hoc or through formal assimilation methods) until the modeled NO_x agrees with the measurements of total NO_x at the far field location (Hudman et al., 2007; Martin et al., 2007; Jourdain et al., 2010; Miyazaki et al., 2014; Liaskos et al., 2015; Laughner and Cohen, 2017; Nault et al., 2017). In general, the far-field approaches yield estimates of LNO_x at the upper end of reported range, while estimates from the near-field studies are typically at the lower end of the range. Nault et al. (2017) showed that a large part of this discrepancy is because prior near-field studies assume a long NO_x lifetime in the UT, while active peroxy radical chemistry in the near field leads to a short NO_x lifetime (~3 h). Without accounting for this chemical loss, the near-field and far-field estimates are biased low compared to each other. However, this effect cannot completely reconcile the discrepancy between LNO_x reported from near- and far- field studies.

In chemical transport models, LNO_x production is modeled by assuming a fixed number of moles of NO are produced per lightning flash, typically 250 or 500 mol NO flash^{-1} (Zhao et al., 2009; Allen et al., 2010; Ott et al., 2010). This presents an additional challenge to the far-field approaches to constrain LNO_x, as errors in the simulation of lightning flashrate will propagate into errors in the LNO_x production per flash. However, explicitly simulating the cloud scale processes that produce lightning is generally too computationally expensive to be applied in a regional or global model as it requires spatial resolution at the same scale of cloud processes. Instead, the convection is parameterized using simplified convection schemes. Lightning is then parameterized by a suite of convection parameters. The most prevalent lightning parameterization relates lightning to the cloud top height (CTH) (Price and Rind, 1992; Price et al., 1997). Price and Rind found a consistent proportionality between cloud-to-ground (CG) lightning flashes and the fifth power of cloud top height. Other meteorological variables, including upward cloud mass flux (UMF), convective precipitation rate (CPR), convective available potential energy (CAPE), cloud ice flux (ICEFLUX) have been suggested as alternative lightning proxies for CG flashes or in some cases total flashes (Allen and Pickering, 2002; Choi et al., 2005; Wong et al., 2013; Romps et al., 2014; Finney et al., 2014). When CG flashes are predicted, the total lightning rate, including CG and Intra-Cloud (IC) flashes, is derived by defining a regional dependent CG:IC ratio (Boccippio et al., 2002).

Several previous studies have evaluated the performance of these lightning parameterizations in regional and global models. Tost et al. (2007) concluded none of them accurately reproduce the observed lightning observations even though some are inter-comparable. Wong et al. (2013) showed that a model using the Grell-Devenyi ensemble convective parameterization and the CTH lightning parameterization simulates erroneous flash count frequency distribution over time while the integrated lightning flash count is consistent with the observation. Luo et al. (2017) tested the single-variable parameterizations (CTH, CAPE, UMF, CPR) and the paired parameterizations based on power law relationship (CAPE-CTH, CAPE-UMF, UMF-CTH), each of which was coupled with Kain Frisch convective scheme, and demonstrated that the two-variable parameterization using CAPE-CTH improves upon the previous single-variable parameterizations; it captures temporal change of flash rates but the simulated spatial distribution is still not satisfactory.

In this study, we implemented the CAPE-PR lightning parameterization (Romps et al., 2014) into WRF-Chem and assess the performance in reproducing lightning flash density. Our motivation is to produce a better representation of a proxy-based lightning parameterization in the regional chemistry transport model. We also evaluate the effect of modeled lightning NO_x on both the a priori profiles used in satellite NO_2 retrievals and the retrievals themselves.

2 Methods: models and observations

2.1 WRF-Chem

This study ~~employs~~ applies the Weather Research and Forecast Model coupled with Chemistry (WRF-Chem) version 3.5.1 ~~.~~ to the time periods May to June, 2012 and August to September, 2013. The model domain covers North America from 20 °N to 50 °N with 12 $\text{km}\times 12\ \text{km}$ horizontal resolution and 29 vertical layers. The North American Regional Reanalysis (NARR) provides initial and boundary conditions. Temperature, wind direction, wind speed and water vapor are nudged every 3h towards to NARR product. Chemistry initial and boundary conditions are provided by the Model for Ozone and Related Chemistry Tracers (MOZART, https://www.acom.ucar.edu/wrf-chem/mozart.shtml). Anthropogenic emissions are driven by the National Emissions Inventory 2011 (NEI 11), with a scaling factor to match the total emissions to 2012 emission from the Environmental Protection Agency (EPA, 2016). Biogenic ~~emission~~ emissions are driven by the Model of Emissions of Gases and Aerosol from Nature (MEGAN; (Guenther et al., 2006)). We use a customized version of the Regional Atmospheric Chemistry Mechanism version 2 (RACM2), the details are described by Zare et al. (2018).

The default lightning parameterization used in WRF-Chem is based on cloud top height (CTH). The parameterized lightning flash rates are proportional to a power of cloud top height with linear scaling varied by region:

$$f = \begin{cases} 3.44 \times 10^{-5} H^{4.9} & \text{Continental} \\ 6.20 \times 10^{-4} H^{1.73} & \text{Marine} \end{cases} \tag{1}$$

where f is the CG flash rate in each grid and H is the colocated cloud top height in units of kilometers.

We also implement an alternative lightning parameterization where lightning flash rates are defined to be proportional to the product of the convective available potential energy (CAPE) and precipitation rate (PR).

$$f = \begin{cases} 0.9 \times 10^{-4} \times E \times PR & \text{Southeastern CONUS} \\ 1.8 \times 10^{-4} \times E \times PR & \text{Elsewhere CONUS} \end{cases} \tag{2}$$

where f the CG flash rate in each grid cell, E the convective available potential energy and PR the convective precipitation rate. Southeastern CONUS in the context is the region between 94 °W to 76 °W and 25 °N to 37 °N. This parameterization was proposed by Romps et al. (2014). Romps et al. (2014) used a year-round observation of lightning and meteorological parameters and found a good correlation between observed lightning flash densities and observed CAPE times PR over the CONUS. CAPE-PR was further examined in Tippett and Koshak (2018) who computed the proxy in a numerical forecast model and found a fairly good agreement between the spatial pattern of the daily CG flash rate and the forecast proxy over 2003-2016. To our knowledge CAPE-PR parameterization has not previously been coupled with chemistry. Note that we compute these two meteorological variables every 72 seconds in our model setup and produce lightning flash rates in a much shorter time step compared to Romps et al. (2014) and Tippett and Koshak (2018). We also apply a regional scaling factor of 0.5 to the southeastern US (See Sec 3.1).

We analyze WRF-Chem outputs from ~~two~~ three model runs. The first run, referred as “G3/CTH”, is consistent with Laughner and Cohen (2017); it selects the Grell 3D ensemble cumulus convective scheme (Grell, 1993; Grell and Dévényi, 2002) and the CTH lightning parameterization. The Grell 3D convective scheme readily computes ~~natural~~ the neutral buoyancy level which serves as the optimal proxy for cloud top height (Wong et al., 2013). The ~~second run selects the~~ “G3/CTH” is the only option for the coupled convective-lighting parameterization used in WRF-Chem at a non-cloud resolving resolution (12 km). In addition, we run WRF-Chem with the CTH lightning parameterization coupled with the Kain-Fritsch cumulus convective scheme (Kain and Fritsch, 1990; Kain, 2004) (“KF/CTH”) to test the effect of switching convective schemes. In the “KF/CTH” parameterization, the cloud top height is the level where the updraft vertical velocity equals to zero. Another run, referred as “KF/CAPE-PR”, selects the Kain-Fritsch cumulus convective scheme and the CAPE-PR lightning parameterization described above. Compared to the Grell 3D convective scheme, the Kain-Fritsch uses the depletion of at least 90% CAPE as the closure assumption and calculates CAPE on the basis of entraining parcels instead of undiluted parcels, which also improves the calculation of precipitation rate (Kain, 2004). ~~Both runs define the~~ The lightning NO_x production rate is defined to be 500 mol NO flash^{-1}. The CG:IC ratio and the LNO_x post-convection vertical distribution are the same as used by Laughner and Cohen (2017).

2.2 ENTLN lightning observation network

To assess the performance of the lightning parameterizations we compare to lightning flashes from Earth Networks Total Lightning Network (ENTLN). ENTLN employs over 100 sensors across the United States and observes both CG and IC pulses (https://www.earthnetworks.com/why-us/networks/lightning/). All lightning pulses within 10 km and 700 ms of each other are grouped as a single flash. The IC and CG flashes are summed over the grid spacing defined in WRF-Chem. Among multiple lightning observation networks

Compared to National Lightning Detection Network (NLDN), ENTLN is selected for better coverage over the CONUS domain and for its high detection efficiency(~70%) (Rudlosky, 2015). Comparison between the ENTLN and the high detection efficiencies of both CG and IC flashes. The average detection efficiency for total flashes observed by ENTLN was 88% over CONUS relative to the space-based Tropical Rainfall Measurement Mission (TRMM) Lightning Imaging Sensor (LIS) yields a broad agreement during the time period covered in this study, as shown in Fig. S1. (Lapierre et al. (2018), private communication). Shown in Fig. S1, we matched the ENTLN data to LIS flashes both in time and space after the correction of LIS data based on its detection efficiency (Cecil et al., 2014) during May 13-June 23, 2012. It shows a median correlation ($R^2 = 0.51$) with the slope of 1.0, indicating the ENTLN data during the study time period is in agreement with the LIS observation. We use the ENTLN for analysis as reported and consider the detection efficiency of ENTLN as a source of uncertainty when comparing the modeled lightning flashes.

2.3 In Situ Aircraft Measurements

We compare our simulations to observations from aircraft campaigns that focus on deep convection. The Deep Convective Clouds and Chemistry (DC3) campaign (Barth et al., 2015) took place during May and June of 2012 over Colorado, Oklahoma, Texas and Alabama. The Studies of Emissions and Atmospheric Composition, Clouds, and Climate Coupling by Regional Surveys (SEAC4RS) (Toon et al., 2016) took place during August and September of 2013; most of the flight tracks occurred over the southeastern US. Both aircraft campaigns flew into and out of storms and sampled deep convection. The combination of these two aircraft campaigns cover the regions with the most active lightning in the domain.

2.4 Satellite Measurements

The Ozone Monitoring Instrument is an ultraviolet/visible (UV/Vis) nadir solar backscatter spectrometer launched in July 2004 on board the Aura satellite. It detects backscattered radiance in the range of 270-500 nm and the spectra are used to derive column NO_2 at a spatial resolution of 13 km×24 km at nadir (Levelt et al., 2006).

We use the Berkeley High Resolution (BEHR) v3.0B OMI NO_2 retrieval (Laughner et al., 2018). The AMF air mass factor (AMF) is calculated based on the high spatial resolution a priori input data including surface reflectance, surface elevation and NO_2 vertical profiles. In this study we apply an experimental branch of the BEHR product which differs from v3.0B in several ways. First, the tropopause pressure is switched to NASA tropopause pressure instead of calculation based on temperature profiles from WRF-Chem(Mak et al., 2018), the tropopause pressure is switched to GEOS-5 monthly tropopause pressure

		G3/CTH	KF/CTH	KF/CAPE-PR
Southeastern	Slope	2.08	~~0.98~~0.94	0.96
	R^2	0.30	0.67	0.72
Elsewhere	Slope	0.98	0.54	1.19
	R^2	0.27	0.48	0.62

Table 1. Correlation statistics between observed and modeled (G3/CTH, KF/CTH, KF/CAPE-PR) flash density per day averaged by regions

which is consistent with NASA Standard Product (SP2). Analysis shows the algorithm used in BEHR v3.0B to calculate the WRF-derived tropopause pressure is very much dependent on the vertical spacing predefined in WRF-Chem setup, which causes biases when the vertical layers are at a coarse resolution (Mak et al., 2018). Second, the NO_2 vertical profiles are outputs using the modified lightning parameterization described in Eq. 2.

3 Results

3.1 Comparison with observed lightning flash density

The lightning parameterizations are compared against observations from ENTLN in Fig. 1. Each of the datasets is averaged from May 13 to June 23, ~~2012.~~ 2012, covering DC3 field campaign. The ENTLN data is summed to the 12 km×12 km WRF grid. The G3/CTH parameterization fails to reproduce the spatial pattern of flashes observed by ENTLN over the CONUS. ~~In contrast, the~~ Compared to the G3/CTH, the KF/CTH parameterization improves the spatial correlation in the southeast region of US. The KF/CAPE-PR parameterization better captures the spatial distribution of flash densities both in the southeast region and elsewhere in CONUS. However the KF/CAPE-PR parameterization still fails to capture the gradients in flash occurrence within smaller regions. For instance, ENTLN shows that more lightning occurs along the east coast than west coast in Florida, however, WRF-Chem generates a lightning flash density of the same magnitude over both areas. Nevertheless, the KF/CAPE-PR substantially improves the model performance in reproducing lightning spatial patterns.

To evaluate the agreement quantitatively, we regress the WRF daily regional average flash densities against those measured by ENTLN. The daily regional averaged flash density is calculated by summing the total flash rates and dividing them by the corresponding regional size. The regressions are shown in Fig~~1 (d~~. 1 (e) and (~~e~~f); the correlation statistics are shown in Table 1. We also regress the data by forcing intercept equals to zero, and the results remain unaffected.

The model using the KF/CAPE-PR lightning parameterization yields a tight correlation and slope close to the unity over the US domain. In the southeastern US, the R^2 increases from 0.3 to 0.7 and slope is reduced from 2.08 to 0.96 with the KF/CAPE-PR parameterization compared to ~~CTH.~~ the G3/CTH. The slope for KF/CTH is comparable to KF/CAPE-PR while the R^2 for KF/CAPE-PR is slightly higher. Note that the improved scaling of the slope in KF/CAPE-PR is mainly caused by the scaling factor of 0.5 applied to the southeast region. In this simulation, a constant linear coefficient for KF/CAPE-PR is not adequate to represent the observed lightning over CONUS, in contrast to the finding of Romps et al. (2014). Elsewhere in

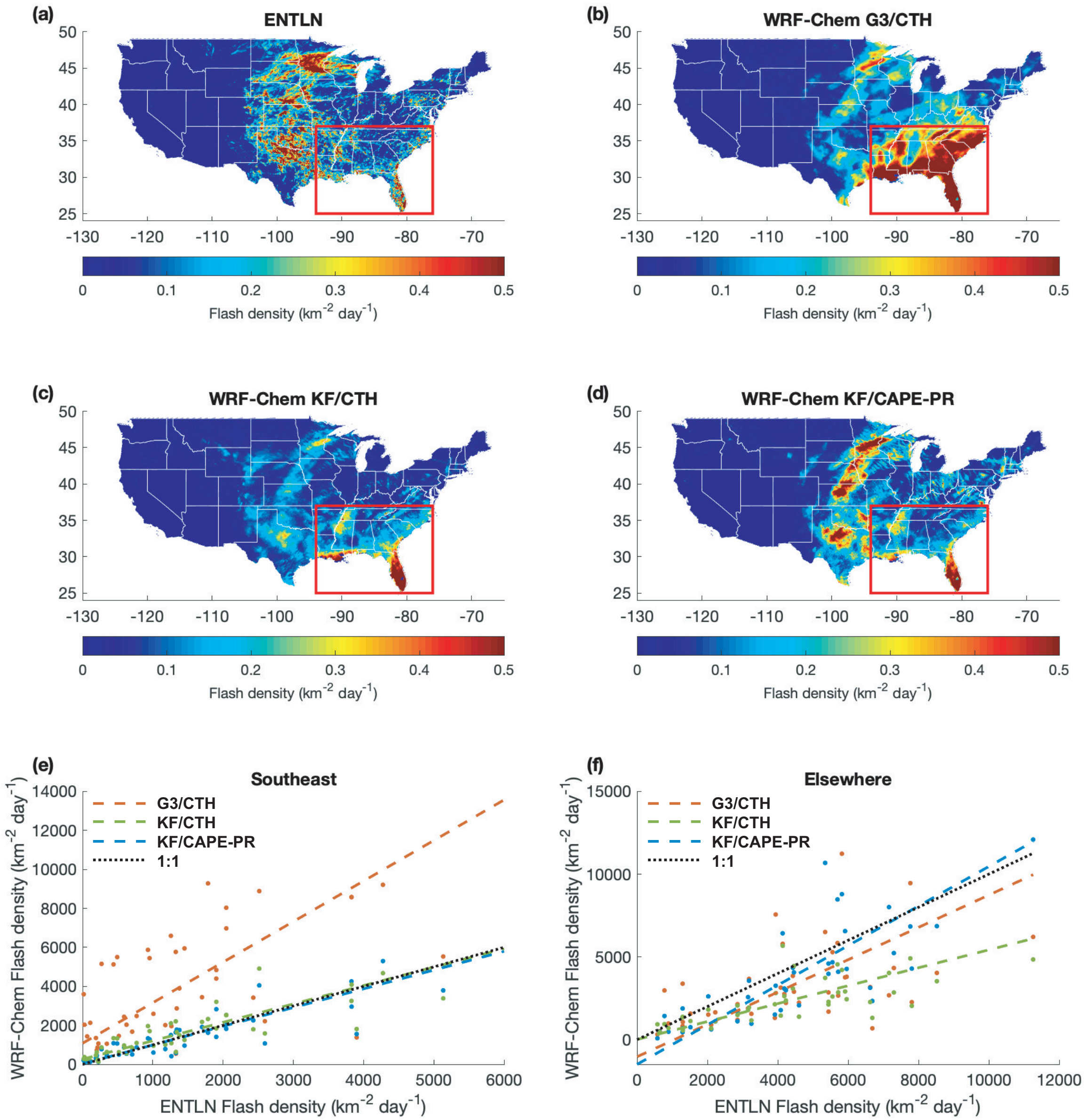

Figure 1. Observed flash densities from the ENTLN dataset **(a)** and WRF-Chem using ~~two lightning parameterization~~three coupled convective-lightning parameterizations, the G3/CTH ~~lightning~~ parameterization **(b)**, the KF/CTH parameterization **(c)** and the KF/CAPE-PR ~~lightning~~ parameterization ~~**(c)**~~**(d)**, respectively. The correlation of total flash density per day between WRF-Chem outputs and ENTLN for the southeastern US (denoted by the red box in **a**~~and **b**~~-**d**) is shown in panel ~~**(d)**~~ **(e)** and the correlation for elsewhere in CONUS is shown in ~~**(e)**~~**(f)**. The model using G3/CTH is in red, KF/CTH is in green, and ~~the model using~~ KF/CAPE-PR is in blue. Dash lines are corresponding fits. For slope and R^2, see Table 1.

		AMF G3/CTH	AMF KF/CAPE-PR	%ΔAMF	VCD G3/CTH	VCD KF/CAPE-PR	%ΔVCD
Sep 10	Urban	~~1.71~~ 1.64	~~0.55~~ 0.72	~~-67.9~~ -56.0	~~2.01~~2.19 $\times 10^{15}$	~~6.48~~5.16 $\times 10^{15}$	~~222.2~~134.9
	Rural	~~2.88~~ 1.96	~~1.89~~ 1.33	~~-34.4~~ -32.0	~~1.05~~1.11 $\times 10^{15}$	~~1.38~~1.63 $\times 10^{15}$	~~34.4~~44.9
Aug 24	Urban	~~0.74~~ 1.07	~~0.75~~ 0.95	~~1.5~~ -11.3	~~5.59~~2.56 $\times 10^{15}$	~~3.92~~2.64 $\times 10^{15}$	~~-1.8~~3.1
	Rural	~~0.82~~ 1.23	~~0.86~~ 1.25	~~4.8~~ 1.60	~~2.24~~1.91 $\times 10^{15}$	~~2.13~~1.82 $\times 10^{15}$	~~-4.8~~-4.6

Table 2. Differences for BEHR AMFs and tropospheric VCDs when using the a priori NO_2 profiles from models with G3/CTH vs KF/CAPE-PR parameterizations in the AMF calculation. For definitions of "urban" and "rural", see the text.

CONUS, the R^2 ~~improves from 0.3~~ for KF/CAPE-PR improves significantly to 0.6 ~~with slopes increasing by 20%.~~ compared to both G3/CTH and KF/CTH. The slope for KF/CAPE-PR is 1.19, which is within the uncertainty of the detection efficiency of ENTLN. In general the KF/CAPE-PR lightning parameterization captures the day-to-day variation in flash densities better than the ~~CTH parameterization~~G3/CTH and KF/CTH parameterizations as shown by the improved R^2 values.

3.2 Comparison with observed vertical profiles

We compare the WRF NO_2 profile to the average vertical profile of NO_2 measured during DC3 and SEAC4RS in Fig. 2. Data points are matched in time and space by finding the WRF-Chem output nearest in time and closest in space to a given observation. We only compare NO_2 profiles from WRF-Chem using KF/CAPE-PR against the one using G3/CTH.

The effect of lightning NO_x on the profiles is indistinguishable close to the surface. In the upper and middle troposphere, both model simulations yields similar NO_2 vertical profiles compared to the measurements from DC3. WRF-Chem using KF/CAPE-PR performs ~~better comparing profile~~ slightly better between 200 hPa to 400 hPa but the negative bias still exists. ~~The largest percentage difference occurs~~ NO_x from both the observations and the models are very small in the middle troposphere between 400 hPa to 700 hPa~~where observations cannot capture but the model predicts an appreciable amount of compared to observations.~~.

Laughner et al. (2019) previously identified a high bias of WRF-Chem UT NO_2 versus SEAC4RS in the southeast US when using the G3/CTH parameterization. The model using the KF/CAPE-PR parameterization reduces this high bias of NO_2 in the middle and upper troposphere. The KF/CAPE-PR parameterization slightly overestimates ~~it~~ NO_2 in the middle troposphere (~~400 - 530~~ 400-530 hPa) and underestimates it in the upper troposphere ($<$280 hPa), which is consistent with the comparison to observations from DC3 campaign.

3.3 Impact on BEHR NO_2 retrievals

In space-based retrievals of NO_2, ~~an air mass factor (AMF)~~ the AMF is required to convert the slant column density (SCD) obtained by fitting the observed radiances into a vertical column density (VCD). The AMF depends on scattering weights (which describe the sensitivity of the measurement to different levels of the atmosphere) and an NO_2 profile which is either measured or simulated by a chemical transport model, such as WRF-chem. Over a dark surface, the scattering weights in the

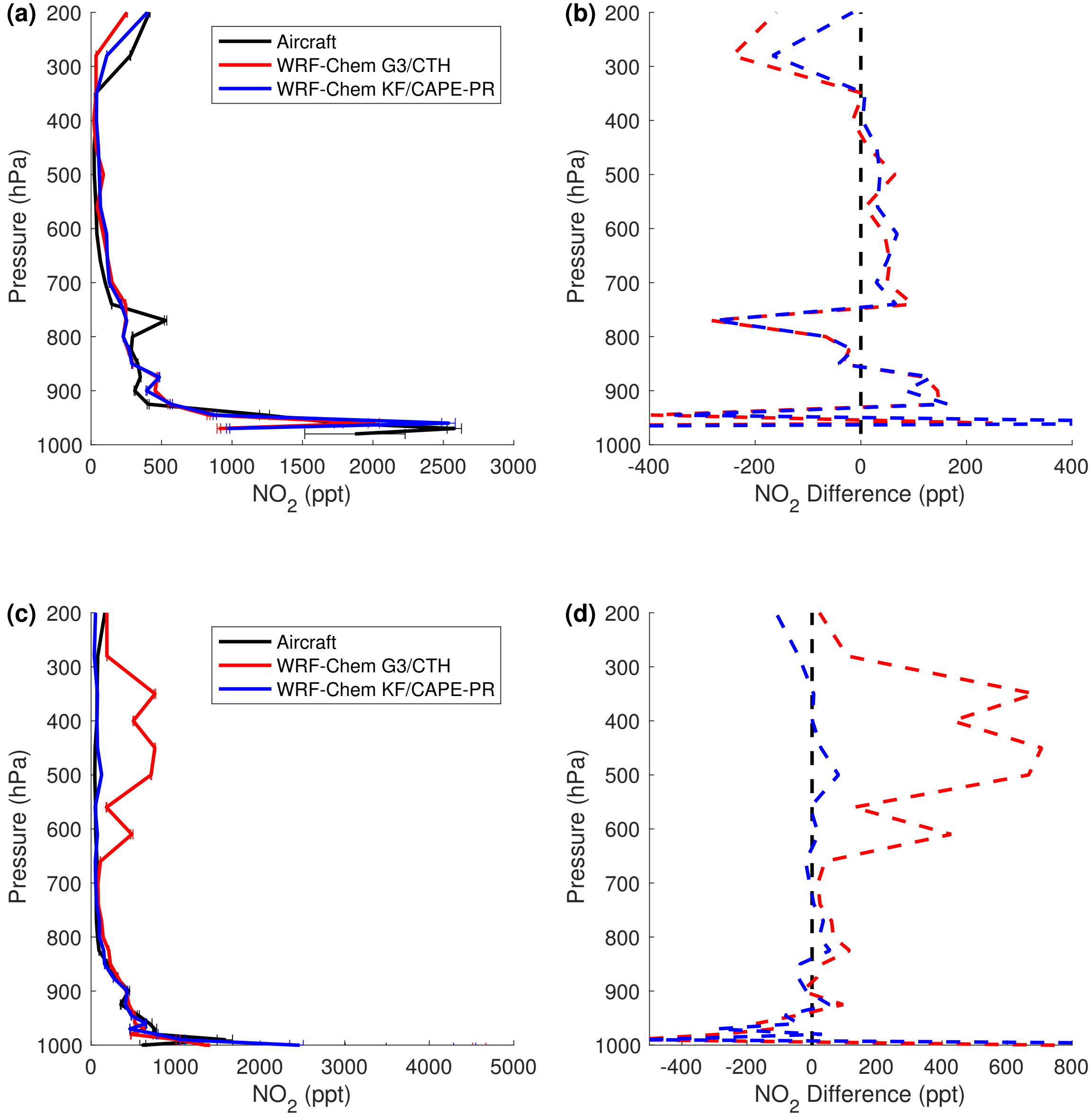

Figure 2. Comparison of WRF-Chem and aircraft NO_2 profiles from the **(a,b)** DC3, **(c,d)** SEAC4RS campaigns. Vertical NO_2 profiles are shown in **(a,c)**, the solid line is the mean of all profiles and the bars are 1 standard deviation for each binned level. The corresponding ~~relative~~ absolute difference compared to observations are shown in **(b,d)**. Aircraft measurements are shown in black, WRF-Chem using G3/CTH ~~lightning~~ parameterization in red and WRF-Chem using KF/CAPE-PR ~~lightning~~ parameterization in blue.

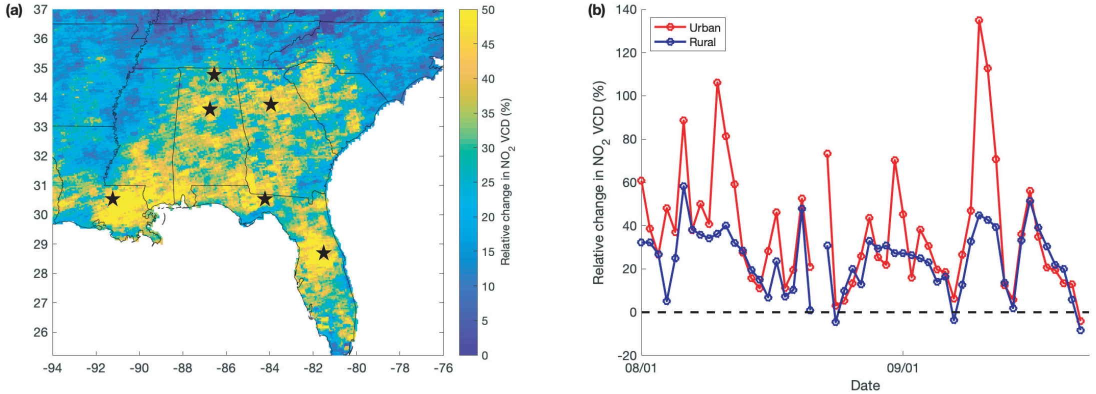

Figure 3. Relative change in BEHR NO_2 VCD over the southeastern US switching the source of a prior ~~NO2~~ NO_2 profiles from WRF-chem outputs using G3/CTH to one using KF/CAPE-PR lightning parameterization. **(a)** shows the mean spatial distribution of the changes from Aug 01 to Sep 23, 2013 and **(b)** shows the temporal variation over urban and rural areas. Medium to large cities, including Atlanta, GA; Huntsville, AL; Birmingham, AL; Tallahassee, FL; Orlando, FL; and ~~Boton~~ Baton Rouge, LA, are marked by stars in panel **(a)**.

UT are up to 10x greater than near the surface, due to the greater probability that a photon that reaches the lower troposphere will be absorbed by the surface. Therefore, errors in the UT NO_2 profile can have large effects on the AMF (e.g. Laughner and Cohen, 2017). Here, we investigate how the NO_2 profiles simulated by the KF/CAPE-PR parameterization affect the BEHR NO_2 retrievals.

~~Fig.~~ Figure 3(a) shows the relative change in tropospheric VCD averaged between Aug 01 to Sep 23, 2013 induced by changing the a priori profiles from the model using G3/CTH to the one using the KF/CAPE-PR lightning parameterization. The relative enhancement of VCD is 19% on average over southeast US but it varies significantly.

We follow the same algorithm used in Laughner and Cohen (2017) to determine if the result is significant. The overall uncertainty due to AMF calculation for BEHR v3.0B is smaller than 30% during the study period (Laughner et al., 2019). As each grid in Fig. 3(a) is the average of 45±9 pixels, the reduced uncertainty is less than 4.5%. The overall change in VCD is four times larger than the reduced uncertainty. The switch of lightning parameterization leads to changes in VCD exceeding the averaged uncertainty in ~94% of pixels in the southeast region of US.

The spatial pattern in Fig. 3(a) suggests that the magnitude of the improved representation of lightning is quite different in urban and rural areas. The cities indicated by stars and their vicinity regions are associated with substantial increase in NO_2 VCD. To quantify this, we define urban and rural areas by difference in column NO_2 ~~. We calculate VCDs using AMFs computed with a priori profiles from a simulation~~ calculated from WRF-Chem without LNO_x ~~and select the 5% and 95% percentiles of VCD as thresholds. Urban ares~~ . Urban areas are the top 5% of columns ~~and rural areas the bottom 5%, respectively. Fig~~ with the average VCD of 2.2 $\times 10^{15}$ mole cm^{-2}. The selected rural areas have the same size as urban areas and the average VCD is 0.72×10^{15} mole cm^{-2}. Figure 3(b) shows the relative change in VCD over the urban and rural areas as a function of time. The increase in VCD due to the change in profiles is more pronounced over urban areas with averaged relative change of ~~~60~~38%

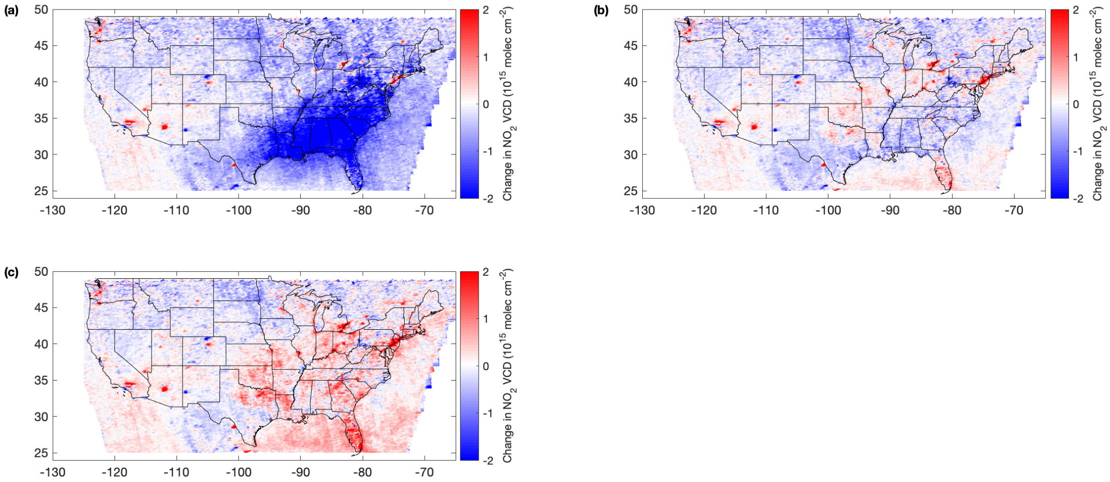

Figure 4. Difference in NO_2 VCD between BEHR retrievals and WRF-Chem. **(a)** excludes LNO_x in model simulation, **(b)** adds LNO_x emission with production rate of 500 mol NO flash^{-1}. **(c)** includes the same LNO_x emission as **(b)** but uses NO_2 profiles scaled upward by 60% at pressure lower than 400 hPa. The average time covers May 13 to June 23, 2012. Pixels with cloud fraction larger than 0.2 are filtered out in the analysis.

compared to the average change of ∼~~25~~24% in rural areas. Changes in urban VCDs span ~~0 to 220~~-10% to 135%. In contrast, using the NO_2 profiles produced by the KF/CAPE-PR simulation leads to only maximum ~~51.2~~58.3% increase in VCD over rural areas.

Table 2 presents the AMF and VCD obtained from using a priori profiles with ~~CTH or~~ G3/CTH or KF/CAPE-PR lightning parameterizations as well as the relative changes on ~~Aug 31 and Sep 14~~Sep 10 and Aug 24, 2013. ~~Aug 31~~ Sep 10 is an example of one day when the change in NO_2 profiles has a very large impact on the NO_2 VCDs. The VCD increases by ~~222~~134.9% over urban areas and ~~34~~44.9% over rural areas; the corresponding change in AMF is ~~68% and 34~~-56.0% and -32.0%, respectively. In contrast, ~~Sep 14~~ Aug 24 is an example where the lightning parameterization has very little effect. The relative change in VCD is ~~-1.8~~3.1% over urban areas and ~~-4.8~~-4.6% over rural areas.

4 Discussion

Here, we apply the improved KF/CAPE-PR simulation to the problem of constraining LNO_x production over CONUS. To do so, we vary the lightning NO_x production rate prescribed in WRF-Chem to produce the simulated map of NO_2 VCD, and compare against OMI NO_2 retrievals using a priori profiles from model simulations with the same LNO_x production rate. In our model-satellite comparisons the averaging kernel is applied to remove the representative errors introduced by a priori knowledges of NO_2 vertical profiles (Boersma et al., 2016). Figure 4 ~~(a, b), Fig. S2 and Fig. S3 show that the~~ shows the difference between satellite retrieved NO_2 VCD and model simulated NO_2 VCD without lightning NO_x **(a)** and with lightning NO_x production rate of 500 mol NO flash^{-1} ~~yields the lowest~~ **(b)** averaged between May 13 to June 23, 2012. Figure S2 shows difference plots with varied lightning NO_x production rates (400 and 665 mol NO flash^{-1}). The corresponding root-mean-square ~~error~~ errors (RMSE) are included in Table S1. LNO_x production rate of 500 mol NO flash^{-1} yields the lowest RMSE of 0.41×10^{15} mole cm^{-2} between modeled and observed NO_2 VCD ~~, which~~ over CONUS. This is at the high end of previous estimates of the lightning NO_x production rate (16-700 mol NO flash^{-1}).

The RMSE for urban areas (top 5% of NO_2 VCD simulated by WRF-Chem without LNO_x) remains at high value (~0.9-1.3×10^{15} mole cm^{-2}) when switching the LNO_x production rate. It indicates that the bias in the modeled VCD over urban areas is more likely due to surface NO_2. The RMSE for non-urban areas shows pronounced change with varied LNO_x production rate. Excluding urban areas lowers the RMSE to 0.37×10^{15} mole cm^{-2} for LNO_x production rate of 500 mol NO flash^{-1}. The RMSEs are significant considering the uncertainty for retrievals. During the average time period, 32 ± 6 pixels contribute to each value in the plots. While the global mean uncertainty for tropospheric NO_2 VCD retrievals is 1×10^{15} mole cm^{-2} (Bucsela et al., 2013), the reduced uncertainty in our analysis is ~0.2×10^{15} mole cm^{-2}. The calculated RMSEs are twice of the uncertainty.

However, we note that this lightning NO_x estimate is systematically biased high due to the negative bias in $[NO_2]/[NO_x]$ ratio in the middle and upper troposphere. The satellite observed NO_2 column serves as a proxy for total NO_x emitted by lightning. The rapid interconversion between NO and NO_2 reaches the photochemical steady state in a short time (~120s). Consequently, if the model kinetics result in an incorrect NO-NO_2 photochemical steady state ratio, this error will propagate into the LNO_x production estimate. Comparisons against aircraft measurements show $[NO_2]/[NO_x]$ ratio in the WRF-Chem simulations is around 40% smaller than observations in upper troposphere (Fig. ~~S4~~S3). Given that the simulated $[NO_2]/[NO_x]$ is too small, the model will simulate smaller NO_2 VCDs per unit of LNO_x emitted, requiring a greater LNO_x production efficiency to match satellite NO_2 VCD observations. ~~Modeled~~ Comparison of modeled NO_2 columns ~~are~~ recalculated with NO_2 profiles scaled up by 60% (the ratio of observed and modeled $[NO_2]/[NO_x]$) at pressure levels where $p < 400$ hPa ~~, and the comparison between revised model and observation~~ and observations is shown in Fig. 4 **(c)**. This suggests that the 500 mol NO flash^{-1} is greater than the actual LNO_x production rate when the bias caused by $[NO_2]/[NO_x]$ ratio is accounted for.

Several recent studies also report an underestimate in modeled $[NO_2]/[NO_x]$ ratios in ~~SE~~ southeastern US(Travis et al., 2016; Silvern et al., 2018); both feature observations from SEAC4RS field campaign to validate model simulations. Silvern et al. (2018) suggests the underestimate is either caused by an unknown labile NO_x reservoir species or error in reaction rate constant for the NO+O_3 reaction and NO_2 photolysis reaction. In contrast, Nault et al. (2017) utilizes measurements from DC3 field campaign and demonstrates a positive bias in modeled $[NO_2]/[NO_x]$ ratio compared against observations. Understanding the difference in $[NO_2]/[NO_x]$ between model and observations requires additional study, but is crucial to reducing the uncertainty in LNO_x estimates.

5 Conclusions

We implement an alternative lightning parameterization based on convective available potential energy and precipitation rate into WRF-Chem and couple it with Kain Frisch convective scheme. We evaluate its performance in simulating lightning NO_x. We first validate it by comparing against lightning observations and conclude that the KF/CAPE-PR parameterization with a regional scaling factor of 0.5 in the southeastern US improves the model representation of spatial pattern and day-to-day variation of lightning flashes. We also compare the simulated NO_2 profiles against aircraft measurements and find that the simulated NO_2 is more consistent with observations in the mid and upper troposphere.

The improved lightning NO_2 simulation has significant impact on AMFs and VCD of NO_2. Over the southeastern US the AMF is reduced by 16% on average leading to a 19% increase in the NO_2 VCD. The effects on AMF and on VCD are very locally dependent. The VCD increase over urban areas is more pronounced and can be up to over 100%. This study emphasizes the importance of including reliable lightning NO_2 in the a priori profiles for satellite retrievals.

The model-satellite NO_2 column comparison suggests 500 mol NO flash^{-1} is too high for the estimate of lightning NO_x production rate, but demonstrate demonstrates that the uncertainty in the modeled UT [NO_2]/[NO_x] ratio is a key limiting factor in constraining production efficiency over CONUS in the far-field approaches.

Data availability. The experimental branch of BEHR v3.0B product used in this study is hosted by UC Dash (Zhu et al., 2019a, b) as well as on behr.cchem.berkeley.edu. The BEHR algorithm is available at https://github.com/CohenBerkeleyLab/BEHR-core/ (Laughner and Zhu, 2018). The revised WRF-Chem code is available at https://github.com/CohenBerkeleyLab/WRF-Chem-R2SMH/tree/lightning (Zhu and Laughner, 2019).

Author contributions. RCC directed the research and QZ, JLL and RCC designed this study; JLL and QZ developed BEHR products; QZ performed the analysis and prepared the manuscript with contributions from JLL and RCC. All authors have reviewed and edited the paper.

Competing interests. The authors declare no competing interests.

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Supplement to "Lightning NO_2 simulation over the Contiguous US and its effects on satellite NO_2 retrievals"

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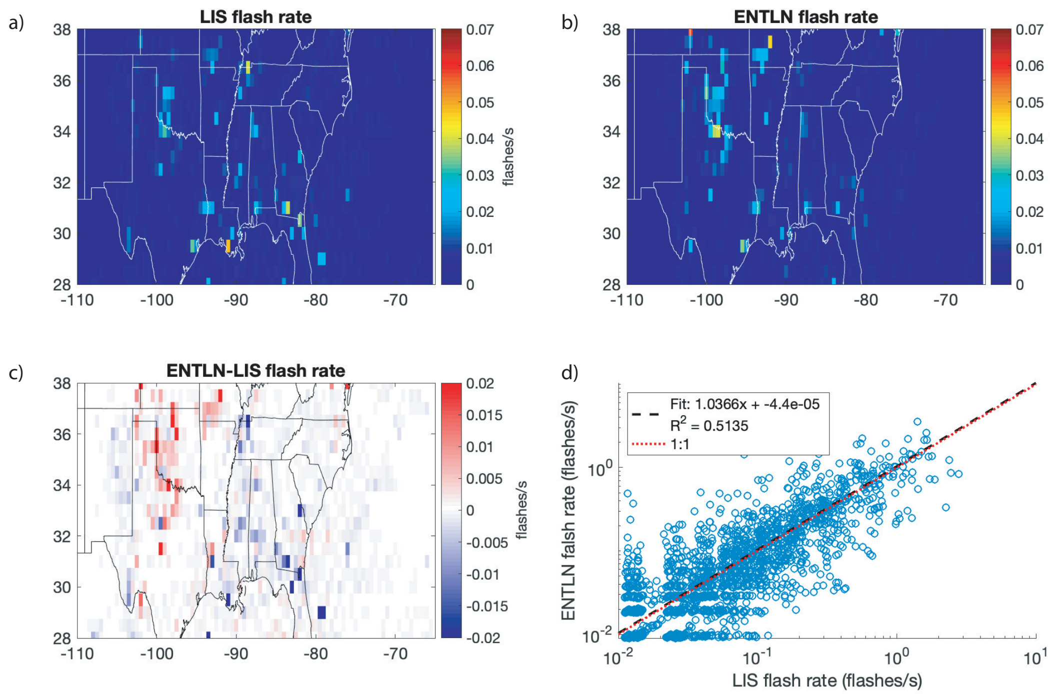

Figure S1: Comparison between flash rates observed by ENTLN and Lightning Imaging Sensor (LIS). ENTLN data is matched to corrected LIS flashes both in time and space during May 13-June 23, 2012, and both datasets are summed onto 0.5°x 0.5°grid spacing. **(a,b)** shows the spatial pattern of lightning flash rates measured by LIS **(a)** and ENTLN **(b)**. The plot region covers 20°N - 38°N and 130°W - 65°W. **(c,d)** are corresponding absolute difference and scatter plots between LIS and ENTLN. LIS data is corrected using the detection efficiency from citetcecil14.

Comparison between flash rates observed by ENTLN and Lightning Imaging Sensor (LIS). **(a,b)** shows the spatial pattern of lightning flash rates averaged from May 13 to Jun 23 2012 measured by LIS **(a)** and ENTLN **(b)**. The plot region covers 20°N - 38°N and 110°W - 65°W. **(c,d)** are corresponding absolute difference and scatter plots between LIS and ENTLN.

	No lightning	400 mol NO $flash^{-1}$	500 mol NO $flash^{-1}$	665 mol NO $flash^{-1}$
CONUS	0.92×10^{15}	0.44×10^{15}	0.41×10^{15}	0.44×10^{15}
Urban	1.30×10^{15}	0.89×10^{15}	0.91×10^{15}	1.10×10^{15}
Non-Urban	0.90×10^{15}	0.41×10^{15}	0.37×10^{15}	0.39×10^{15}

Table S1: The root-mean-square errors (RMSE) in unit of mole cm^{-2} between observed and modeled NO_2 VCD using WRF-Chem with varied LNO_x production rates (0, 400, 500, 665 mol NO $flash^{-1}$). Urban areas are selected where NO_2 columns are at top 5% calculated from WRF-Chem without lightning. Non-urban areas are CONUS excluding urban areas.

Box plot of difference in NO_2 VCD between BEHR retrievals and WRF-Chem with varied LNO_x production rate of 0, 400, 500 and 665 mol NO $flash^{-1}$. The corresponding root-mean-square errors (RMSE) are shown above.

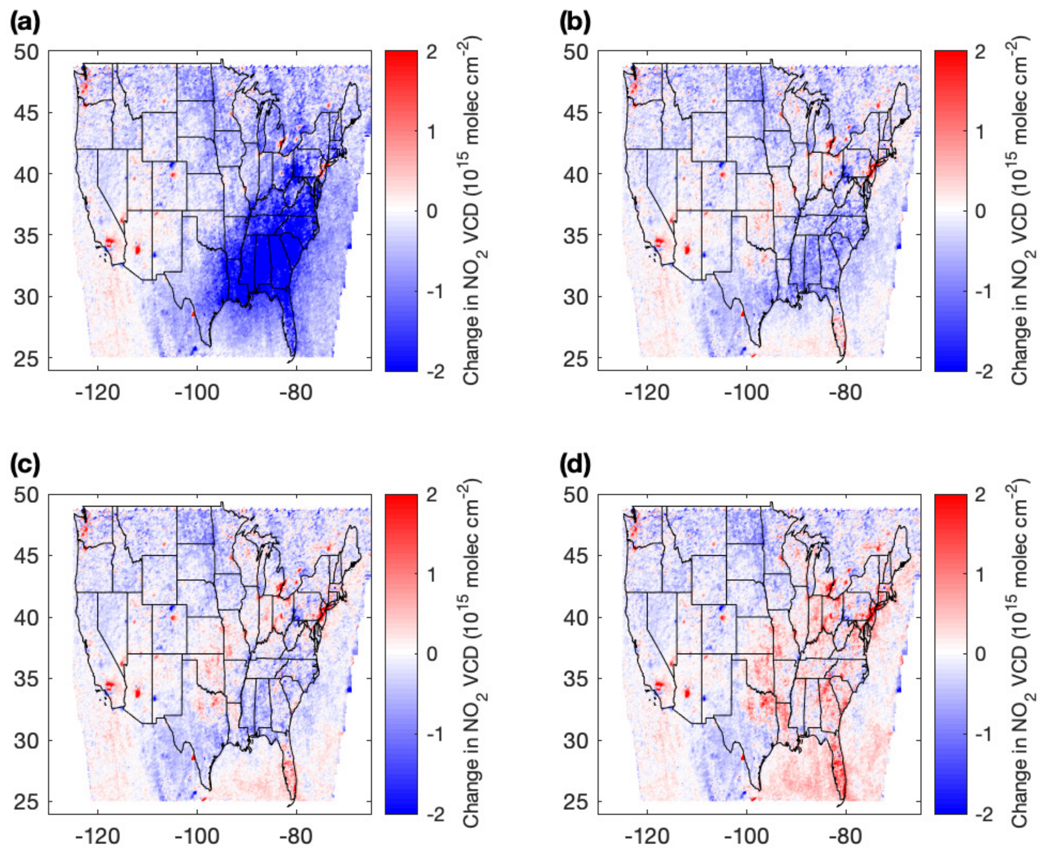

Figure S2: Difference in NO_2 VCD between BEHR retrievals and WRF-Chem **(a)** without LNO_x and with LNO_x production rate of **(b)** 400 mol NO flash^{-1}, **(c)** 500 mol NO flash^{-1} and **(d)** 665 mol NO flash^{-1}.

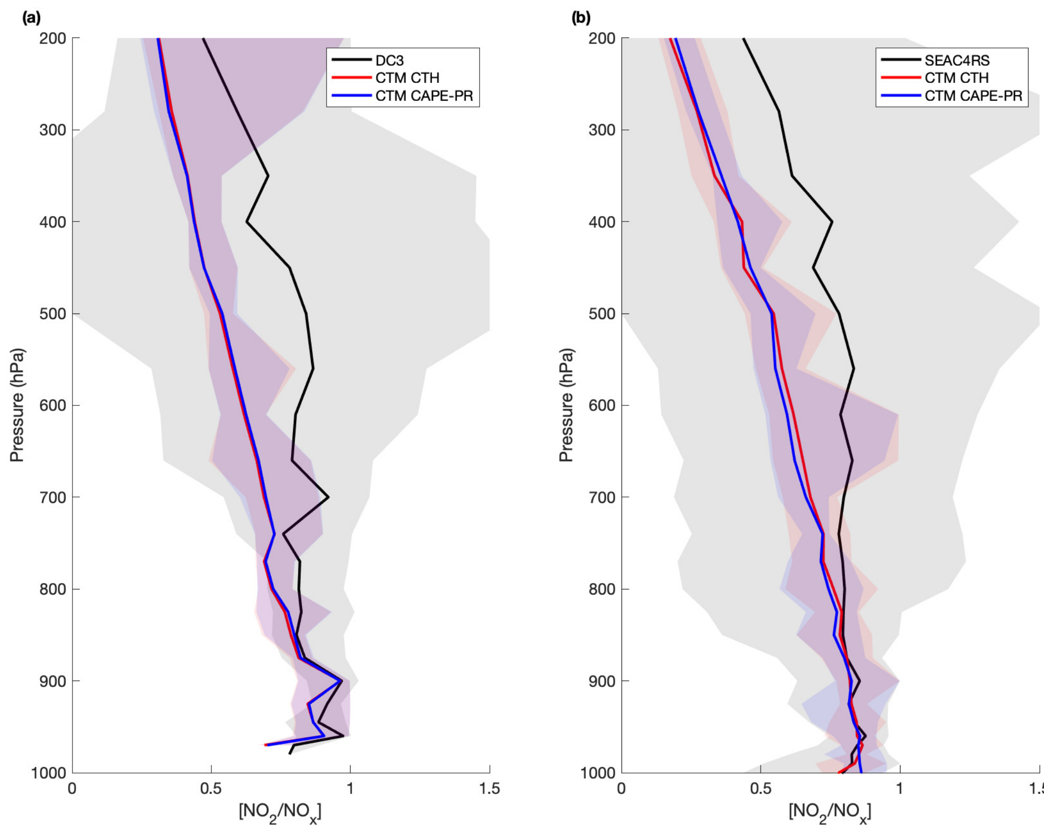

Figure S3: Comparison of WRF-Chem and aircraft $[NO_2/NO_x]$ profiles from the **(a)** DC3, **(b)** SEAC4RS campaigns.The solid line is the median of all profiles and the shaded areas are between 10th and 90th percentiles for each binned level. Aircraft measurements are shown in black, WRF-Chem using CTH lightning parameterization in red and WRF-Chem using CAPE-PR lightning parameterization in blue.