Responses to Reviewer 1:

This article appears to be the evaluation side of GMD-2019-33 “Simulating Lightning NOX Production in CMAQv5.2: 2 Evolution of Scientific Updates”, which is cited numerous times in the manuscript, and with similar authors (although the order is not exactly the same). I would suggest to make the link more specific and make these two papers companion papers, possibly entitled “Simulating Lightning NOX Production in CMAQv5.2: part 1, new parameterizations”, and part 2: evaluation for example.

We agree with the reviewer, and we have now changed the title to “Simulating Lightning NO Production in CMAQv5.2: Performance Evaluations” to match the companion paper that has been published.

The paper is well written, concise, and of good scientific quality, with a thorough evaluation of the impact of the three new schemes that have been implemented into CMAQ.

We thank the reviewer for the overall positive assessment.

I have a few remarks that should in my opinion be addressed before final publication:

âA˘ c Please add a short descriptive summary of the three lightning schemes that are ´ evaluated in the paper

The other reviewer also made the same suggestion, so we have now added this information in the Methodology section as “2.1 The LNO schemes” in the revised manuscript on Page 4.

âA˘ c It would be desirable to remind the reader of the different chemical links between ´ NOx, O3 and nitrate precursors; this is partially done at the beginning of section 3.3 for nitrate.

We thank the reviewer for this suggestion. Even though the role of NOx in the atmospheric chemistry is well known, we agree that for completeness it would be useful to briefly summarize the role for broader readership. We have now incorporated the information in the introduction section by stating that “it is expected that the relative contribution of LNO to the tropospheric NO_x burden and its subsequent impacts on atmospheric chemistry as one of the key precursors for ozone (O_3), hydroxyl radical (OH), nitrate, and other species will increase in the United States and other developed countries”.

âA˘ c Perhaps a discussion on the skill of the forecasts of convective precipitations in ´ the WRF forecast (and possibly of its diurnal cycle) should be discussed or at least mentioned since this is a critical input of the three schemes,

The reviewer makes a good point. However, there are no observations to distinguish convective precipitation from non-convective precipitation, and usually for precipitation measurements, only aggregated daily or even longer-period products such as PRISM (a combination of rain gauge and modeling results) and STAGE (a combination of radar and

rain gauge observations) are available. Therefore, it is not readily possible to assess the forecast skills for convective precipitation, and even more so for the diurnal cycle. For this reason, we provided the monthly accumulated precipitation assessment in Figure 12 for WRF precipitation and computed the statistics over the NTN sites to form the basis for our nitrate wet deposition evaluation.

âA˘ c For nitrate, perhaps it would have been simpler to evaluate the nitrate concentrations against observations from the CASTNET network, rather than nitrate wet deposition, which depends again on modelled precipitation: this adds another layer of error/uncertainty.

We agree that the CASTNET network offers another source of evaluation for model estimated nitrate. The advantage of using the NADP/NTN network is that it is larger with more spatial coverage (196 NTN sites compared to about 75 CASTNET sites). The top row in Figure 12 provides the model bias in annual total precipitation compared to NTN observations so that it can be compared against the wet deposition bias. These plots indicate that, while there is some underestimation of precipitation in the eastern half of the US, errors in modeled precipitation do not account for 35% normalized mean bias in modeled wet deposition of nitrate across the country. This consistent bias suggests missing regional-scale emissions sources such as NO from lightning. Additionally, direct evaluation of modeled wet deposition estimates is used to inform national scale assessments of nitrogen budgets and comparison of deposition loads with critical loads. The evaluation again the NTN network is used to demonstrate how the addition of NO from lightning can help reduce bias in modeled nitrate deposition levels, increasing the credibility of using model output for critical loads analyses. It should also be noted that comparisons of aerosol NO3 predictions with ambient observations (from CASTNET and other networks) can be influenced by errors in modeled gas/aerosol partitioning influenced by uncertainties in NH3 emissions. Comparisons of total NO3 wet deposition also helps circumvent those other model error influences and better isolate the impacts of LNO emissions on total nitrate atmospheric budget.

âA˘ c Tables 1 and 2 are very big; the bold parts are not always easy to spot. Is there a ´ way to present this key information in graphics?

We agree that the tables contain a lot of information, but as a supplementary to the information that has presented in other graphics and plots, interested readers can get more detailed information from the tables for model performance over different geographical regions.

Responses to Reviewer 2

General Comments

The scope of this paper is to evaluate the impact of three Lightning NOx parameterisation schemes in WRF-CMAQ on ozone, NOx and nitrate deposition compared with a base case without such parameterisation. The use of a variety of observations at different heights is commendable and it is clearly presented. The paper is well written and easy to follow. Although differences between the three parameterisation schemes and the base case are generally large, the three schemes perform fairly similarly to each other in a number of cases presented. This is not surprising, given that the three parameterisation schemes used are different versions of the same scheme. However, the authors use all the observations in their toolbox to provide a clear explanation of where the schemes show the largest difference and try to identify the best performing scheme.

We thank the reviewer for the overall positive assessment of the manuscript.

Specific Comments

There is not enough information about the three parameterisation schemes. It would be useful to add at least a very short description here (including the vertical distribution algorithm) and then refer the reader to the relevant paper for further details.

Both reviewers have suggested including additional details on the LNO schemes. In the revised manuscript, we have now added this information to the Methodology Section as "2.1 The LNO schemes" on Page 4 of the revised manuscript.

Given that the model uses hourly or monthly observed lightning flashes information from the NLDN network, I expect this parameterisation schemes are only available for simulations of the past, e.g. hindcasts and case studies, but not for air quality forecasts (for which the observed lightning flashes are not available). Can the authors add a comments in the text to address the relevance of this work for air quality forecast or specify its intended areas of application?

In this study, three lightning NO schemes are involved. It is correct, all the schemes are related to the observed lightning flashes from NLDN network, but the formulations are different. The hourly (hNLDN) or monthly (mNLDN) schemes do depend on the availability of the observed NLDN data for their applications, but the third one, the parameterized scheme (pNLDN), was derived using historical data from the observed NLDN data and model predicated convective precipitation, and its application doesn't require the actual observed data. Instead, the lightning flashes are derived from the linear and log-linear relationship that is parameterized in the scheme. And it is specifically tailored for applications such as air quality forecast when the observed lightning flashes are not available. We have now incorporated this point in Conclusions on Page 15 of this revised manuscript.

l.184-185 "...all model cases with LNOx exhibit slightly higher correlation coefficients than the base simulation, suggesting the importance..." Looking at table 1 and 2 I see identical values for most locations and tiny differences (0.69 vs 0.70; 0.73 vs 0.74; 0.52 vs 0.53) for other cases. I would rather say that the correlation coefficients between simulations with and without LNOx are not significantly different!

Though the difference between correlation coefficients are small, but the increase is persistent through the domain and all subregions that indicates the general trend. Therefore, we describe it as slightly higher.

l.257-259 can the authors comment on why NOx is over/under-estimated during night/day-time?

The question of why NOx is over/under-estimated during night/day-time is rather a complicated issue that is currently under active investigation in many research groups with coordinated efforts. There are several hypotheses including (1) issues related to representation of vertical mixing, (2) issues related to magnitude of anthropogenic emissions, especially from the mobile sector, and (3) spatial and temporal allocation of emissions.

In Figure 4, the legend for AQS is wrong (no star symbol used in the plots)

We thank the reviewer for catching this error. It has now been corrected.

Figure 6. It would be interesting to add 2 further panels to show equivalent results for NOx profiles in the different model simulations. Can this help explain the lower surface ozone in hNLDN? If not, can the author suggest what processes are responsible for it?

We thank the reviewer for the suggestion. The impact of lightning NO_x on O_3 production generally occurs downwind of the location of lightning flashes as revealed in our later analysis related to Figures 7-10. Often it is the case that when the ozonesonde measurements indicated difference on O_3 mixing ratios, the difference of NO_x mixing ratios from the different model cases is insignificant at the same location. We however examine the issue raised by the reviewer using the aircraft measurements in detail in later section in the manuscript.

Technical Comments

l.53 "pNLDN, provides an improved estimate for LNOx compared to the base simulation that does not include LNOx." LNOx is of course improved if it is included in the simulation! I think this should be: "...provides an improvement for ozone and NOx compared to the base simulation..."

Thanks. It was a typo. It has now been revised to “an improved estimate of nitrate wet deposition”

l.65-66 "The significant impact of LNOx on surface air quality was earlier..." Given the explanation given by the authors I think this should be: "The significant impact of LNOx on process-based understanding of surface air quality was earlier..."

Thanks. The sentence has been modified as suggested by the reviewer.

l.66 replace "in that" with "which found"

Thanks, the change has been made.

l.288-289 "the vertical profile lines can be separated" this is confusing, replace with same text used later (l.308) which is much clearer.

Thanks, we have revised the manuscript as suggested.

Responses to Executive Editor

Thank you for pointing out the loose points for the code and data accessibility. We have now updated the manuscript with more accurate links.

1. The CMAQ and WRF code references point to project websites. This is insufficiently persistent and precise for GMD purposes. Please also cite a persistent public archive of the exact version of the source used.

We have now updated the links for both CMAQ and WRF to point to the specific versions:
WRF: http://www2.mmm.ucar.edu/wrf/users/wrfv3.8/updates-3.8.html
CMAQ: https://github.com/USEPA/CMAQ/tree/5.2

2. Data processing and analysis scripts are available "on request". This does not meet GMD requirements. Please provide a citation of a persistent public archive of the scripts (e.g. Zenodo).

We have now updated the dataset with all the scripts used to create the tables and plots in the manuscript as the dataset version 2.0:
https://zenodo.org/record/3360744

3. The lightning dataset is proprietary, which is acceptable. However please identify exactly the data set and version used so that a reader who wished to reproduce the work would know exactly what they needed to purchase and use.

Unfortunately, we don't have the information about the data set and its version. To obtain this dataset, one needs to contact Vaisala Inc. directly, and they would prepare the data with the region and time period from their database and that data is the original lightning flash data collected and managed by Vaisala Inc. For clarity, we added the sentence "The lightning data obtained from Vaisala Inc. is the cloud-to-ground lightning flashes over the contiguous United States".

4. The data citation to Zenodo is excellent. Please ensure that the additional data which is only "on request" is not actually required to reproduce the results.

In the data citation to Zenodo, we have provided the immediate data tables to produce the tables and plots without the "on request" data. However, the scripts provided could directly use the data tables or start from the original "on request" data to understand how the data tables are generated.

Simulating Lightning $NO_{~~x~~}$ Production in CMAQv5.2~~ Using mNLDN, hNLDN, and pNLDN Schemes~~: Performance Evaluations

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Abstract

This study assesses the impact of the lightning NO~~$_x$~~ (LNO~~$_x$~~) production schemes in the Community Multiscale Air Quality (CMAQ) model ~~(Kang et al., 2019)~~ on ground-level air quality as well as aloft atmospheric chemistry through detailed evaluation of model predictions of nitrogen oxides (NO$_x$~~$_x$~~) and ozone (O_3) with corresponding observations for the U.S. For ground-level evaluations, hourly O_3 and NO$_x$ from the US EPA's AQS monitoring network are used to assess the impact of different LNO~~x~~ schemes on model prediction of these species in time and space. Vertical evaluations are performed using ozonesonde and P-3B aircraft measurements during the DISCOVER-AQ campaign conducted in the Baltimore/Washington region during July 2011. The impact on wet deposition of nitrate is assessed using measurements from the National Atmospheric Deposition Program's National Trends Network (NADP/NTN). Compared with the base model (without LNO~~x~~), the impact of LNO~~x~~ on surface O_3 varies from region to region depending on the base model conditions. Overall statistics suggest that for regions where surface O_3 mixing ratios are already overestimated, the incorporation of additional NO~~$_x$~~ from lightning generally increased model overestimation of mean daily maximum 8-hr (DM8HR) O_3 by 1-2 ppb. In regions where surface O_3 is underestimated by the base model, LNO~~x~~ can significantly reduce the underestimation and bring model predictions close to observations. Analysis of vertical profiles reveals that LNO~~$_x$~~ can significantly improve the vertical structure of modeled O_3 distributions by reducing underestimation aloft, and to a lesser degree decreasing overestimation near the surface. Since the base model underestimates the wet deposition of nitrate in most regions across the modeling domain except the Pacific Coast, the inclusion of LNO~~x~~ leads to reduction in biases and errors and an increase in correlation coefficients at almost all the NADP/NTN sites. Among the three LNO~~x~~ schemes described in Kang et al. (2019), the hNLDN scheme, which is implemented using hourly observed lightning flash data from National Lightning Detection Network (NLDN), performs best for the ground-level, vertical profiles, and wet deposition comparisons except that for the accumulated wet deposition of nitrate, the mNLDN scheme (the monthly NLDN-based scheme) performed slightly better. However, when observed lightning flash data are not available, the linear regression-based parameterization scheme, pNLDN, provides an improved estimate for nitrate wet deposition ~~LNOx~~ compared to the base simulation that does not include LNO~~x~~.

1. Introduction

The potential importance of nitrogen oxides (NO$_x$~~x~~; NO$_x$ = NO + NO$_2$) produced by lightning (LNOx; due to the equilibrium coexistence of NO and NO$_2$ in the atmosphere, in the literature it is often collectively referred to as LNOx. However, the immediate release of lightning flashes is just NO, and the schemes in Kang et al., 2019 also generate NO emissions only, so in this paper it is primarily referred to as LNO) on regional air quality was recognized more than two decades ago (e.g. Novak and Pierce, 1993), but LNO~~x~~ emissions have only been added to regional chemistry and transport models during the last decade (e.g. Allen et al., 2012; Kaynak et al., 2008; Koshak et al., 2014; Smith and Mueller, 2010; Koo et al., 2010) owing in part to the limited understanding of this NO$_x$~~x~~ source (Schumann and Huntrieser, 2007; Murray, 2016; Pickering et al, 2016). As a result of efforts to reduce anthropogenic NOx emissions in recent decades (Simon et al., 2015; https://gispub.epa.gov/air/trendsreport/2018), it is expected that the relative contribution of LNO~~x~~ to the tropospheric NO$_x$~~x~~ burden and its subsequent impacts on atmospheric chemistry as one of the key precursors for ozone (O$_3$), hydroxyl radical (OH), nitrate, and other species will increase in the United States and other developed countries (Kang and Pickering, 2018). The significant impact of LNO~~x~~ on process-based understanding of surface air quality was earlier reported by Napelenok et al. (2008), which found ~~in that~~ low-biases in upper tropospheric NO$_x$~~x~~ in Community Multiscale Air Quality Model (CMAQ) (Byun and Schere, 2006) simulations without LNO~~x~~ emissions made it difficult to constrain ground-level ~~NOx~~ NO$_x$ emissions using inverse methods and Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) NO$_2$ retrievals (Bovensmann et al., 1999; Sioris et al., 2004; Richter et al., 2005). Appel et al. (2011) and Allen et al. (2012) reported that NO$_3^-$ wet deposition at National Atmospheric Deposition Program (NADP) sites was underestimated by a factor of two when LNO~~x~~ was not included.

LNO~~x~~ production and distribution were parameterized initially in global models (e.g. Stockwell et al., 1999; Labrador et al., 2005) relying on the work of Price and Rind (1992) and Price et al. (1997) in that lightning flash frequency was parameterized as a function of the maximum cloud-top-height. Other approaches for LNO~~x~~ parameterization include a combination of latent heat release and cloud-top-height (Flatoy and Hov, 1997), convective precipitation rate (e.g. Allen and Pickering, 2002), convective available potential energy (Choi et al., 2005), or convectively induced updraft velocity (Allen et al., 2000; Allen and Pickering, 2002). More recently, Finney et al. (2014, 2016) adopted a lightning parameterization using upward cloud ice flux at 440hPa (based upon definitions of deep convective clouds in the International Satellite Cloud Climatology Project (Rossow et al., 1996)) and implemented it in the United Kingdom Chemistry and Aerosol model (UKCA). With the availability of lightning flash data from the National Lightning Detection Network (NLDN) (Orville et al., 2002), recent LNO~~$_x$~~ parameterization schemes started to include the observed lightning flash information to constrain LNO~~$_x$~~ in regional Chemical Transport Models (CTMs) (Allen et al., 2012). In Kang et al. (2019), we described the existing LNO~~$_x$~~ parameterization scheme that is based on the monthly NLDN (mNLND) lightning flash data, and an updated scheme using hourly NLDN (hNLDN) lightning flash data in the CMAQ lightning module. In addition, we also developed a scheme based on linear and log-linear regression parameters using multiyear NLDN observed lightning flashes and model predicted convective precipitation rate (pNLDN). The preliminary assessment of these schemes based on total column LNO~~$_x$~~ suggests that all the schemes provide reasonable LNO~~$_x$~~ estimates in time and space, but during summer months, the mNLDN scheme tends to produce the most and the pNLDN scheme the least LNO~~$_x$~~.

The first study on the impact of LNO~~$_x$~~ on surface air quality using CMAQ was conducted by Allen et al. (2012) and followed by Wang et al. (2013) with different ways for parameterizing LNO~~$_x$~~ production and different model configurations. In this study, we present performance evaluations using each of the LNO~~$_x$~~ production schemes (mNLDN, hNLDN, pNLDN) described by Kang et al. (2019) to provide estimates of LNO~~$_x$~~ in CMAQ. In addition to examination of differences in air quality estimates between these schemes, we compare the model predictions to base model estimates without LNO~~$_x$~~ and evaluate the estimates from all of the simulations against surface and airborne observations.

Section 2 describes the model configuration, simulation scenarios, analysis methodology, and observational data. Section 3 presents the analysis results and Section 4 presents the conclusions.

2. Methodology

2.1 The LNO schemes

In air quality models, three steps are ~~generally~~ involved in~~to~~ generating~~e~~ LNO emissions: (1) identify lightning flashes, (2) produce the total column NO at model grid cells, and (3) distribute the column NO into model layers vertically. Three schemes to produce total column LNO emissions are examined in this study: mNLDN – based on monthly mean NLDN lightning flashes and convective precipitation predicted by the upstream meteorological model, hNLDN – directly use the observed NLDN lightning flashes that are aggregated into hourly values and gridded onto model grid cells, and pNLDN – a linear and log-linear regression parameterization scheme derived using multiyear observed lightning flash rate and model predicted convective precipitation. After total column LNO is produced at model grid cells, it is distributed onto vertical model layers using the~~the~~ double-peak vertical distribution algorithm described in ~~. Refer to~~ Kang et al. (2019), which also provides a ~~for the~~ detailed description and formulation of all the LNO~~-~~ schemes~~schemes including the vertical distribution algorithm~~.

2.2 The CMAQ model and simulation configurations

~~The three LNO$_x$ production schemes described in Kang et al (2019) were incorporated into CMAQ v5.2 (Appel et al. 2017; doi:10.5281/zenodo.1167892).~~ The CMAQ model (Appel et al. 2017) version 5.2 were configured with t~~T~~he CB6 chemical mechanism (Yarwood et al., 2010) ~~used was~~ ~~CB6 (Yarwood et al., 2010)~~ and the AERO6 aerosol module ~~was A-ERO6~~ (Nolte et al., 2015). The meteorological inputs were provided by the Weather Research and Forecasting (WRF) model version 3.8 and the model-ready meteorological input files were created using version 4.2 of the meteorology–chemistry interface processor (MCIP; Otte and Pleim, 2010).

The modeling domain covers the entire contiguous United States (CONUS) and surrounding portions of northern Mexico and southern Canada, as well as the eastern Pacific and western Atlantic oceans. The model domain consists of 299 north–south grid cells by 459 east–west grid cells utilizing 12 km x 12 km horizontal grid spacing, 35 vertical layers with varying thickness extending from the surface to 50 hPa and an approximately 10 m midpoint for the lowest (surface) model layer. The simulation time period covers the months from April to September 2011 with a 10-day spin-up period in March.

Emission input data were based on the 2011 National Emissions Inventory (https://www.epa.gov/air-emissions-inventories). The raw emission files were processed using version 3.6.5 of the Sparse Matrix Operator Kernel Emissions (SMOKE; https://www.cmascenter.org/smoke/) processor to create gridded speciated hourly model-ready input emission fields for input to CMAQ. Electric generating unit (EGU) emissions were obtained using data from EGUs equipped with a continuous emission monitoring system (CEMS). Plume rise for point and fire sources were calculated in-line for all simulations (Foley et al., 2010). Biogenic emissions were generated in-line in CMAQ using BEIS versions 3.61 (Bash et al., 2016). All the simulations employed the bidirectional (bi-di) ammonia flux option for estimating the air–surface exchange of ammonia.

There are four CMAQ simulation scenarios for this study: 1) simulation without LNO~~$_x$~~ (Base), 2) simulation with LNO~~$_x$~~ generated by the scheme based on monthly information from the NLDN (mNLDN), 3) simulation with LNO~~$_x$~~ generated by scheme based on hourly information from the NLDN (hNLDN), and 4) simulation with LNO~~$_x$~~ generated by the scheme parameterizing lightning emissions based on modeled convective activity (pNLDN) as described in detail in Kang et al. (2019). All other model inputs, parameters and settings were the same across the four simulations. The vertical distribution algorithm is the same for all the LNO~~$_x$~~ schemes as also described in Kang et al. (2019).

2.3 Observations and analysis techniques

To assess the impact of LNO~~x~~ on ground-level air quality, output from the various CMAQ simulations were paired in space and time with observed data from the EPA's Air Quality System (AQS; https://www.epa.gov/aqs) for hourly O_3 and ~~NO_x~~NO_x. To evaluate the vertical distribution, measurements of trace species from the Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality (DISCOVER-AQ; http://www.nasa.gov/missions/discover-aq) campaign conducted in the Baltimore/Washington region (e.g., Crawford and Pickering, 2014; Anderson et al., 2014; Follette-Cook et al., 2015) were used. During this campaign, the NASA P-3B aircraft measured trace gases including O_3, NO, and NO_2. Vertical profiles were obtained over seven locations – Beltsville (Be), Padonia (Pa), Fairhill (Fa), Aldino (Al), Edgewood (Ed), Essex (Es), and Chesapeake Bay (Cb) from approximately 0.3 to 5 km above ground level during P-3B flights over 14 days in July 2011. During this same period, ozonesonde measurements were taken that extended from ground level through the entire model column at two locations (Beltsville, MD and Edgewood, MD shown in Figure 1). Inclusion of LNO~~x~~ estimates in the CTM simulations also has an important impact on model estimated wet deposition of nitrate. Therefore, assessment was also performed using data from the National Atmospheric Deposition Program's National Trends Network (NADP/NTN, http://ndp.slh.wisc.edu/ntn).

Since lightning activity as well as LNO~~x~~ exhibit distinct spatial variations (Kang and Pickering, 2018), analysis was conducted for the model domain over the contiguous United States, and then for each region as shown in Figure 1. Emphasis is placed on two regions, Southeast (SE) and Rocky Mountains (RM), where lightning activity is more prevalent and LNO~~x~~ has the greatest impact on model predictions as shown in Results - increasing model bias in the SE and decreasing bias in the RM. The commonly used statistical metrics, Root Mean Square Error (RMSE), Normalized Mean Error (NME), Mean Bias (MB), Normalized Mean Bias (NMB), and Correlation Coefficient (R), in the model evaluation field as defined in Kang et al. (2005) and Eder et al. (2006) were calculated to assess the basic performance differences among all the model cases for their ground-level air quality predictions.

3. Results

3.1 Ground-level evaluation for O_3 and ~~NO_x~~NO_x

3.1.1 Statistical performance metrics

Tables 1 and 2 display the statistical model performance metrics for daily maximum 8-hr (DM8HR) O_3 and daily mean NO_x mixing ratios over the domain and each analysis region for all four model cases in July 2011 (Base, mNLDN, hNLDN, and pNLDN). The best performance metrics among the model cases are highlighted in bold. As shown in Table 1, for DM8HR O_3, the Base simulation has the lowest MB and NMB values over the Domain, while hNLDN produced the smallest RMSE and NME values. mNLDN generated the largest values for both error (RMSE and NME) and biases (MB and NMB), followed by pNLDN. More importantly, all model cases with LNO~~x~~ exhibit slightly higher correlation coefficients than the Base simulation, suggesting the importance of including the contributions of this source for improving the spatial and temporal variability in model predictions. Additionally, the hNLDN simulation exhibited higher correlation and lower bias and error relative to the measurements indicating the value of higher temporal resolution lightning activity for representing the associated NO_x emissions and their impacts on tropospheric chemistry.

Examining the regional results for DM8HR O_3 in Table 1, the statistical measures indicate that in the Northeast (NE), hNLDN outperformed all other model cases with the lowest errors and biases and highest correlation coefficient. In Southeast (SE), the Base performed better with the lowest errors and mean biases, but the correlation coefficient (R) value for hNLDN is slightly higher. Among all the LNO~~x~~ cases, mNLDN produced the worst statistics in this region. Historically, CTMs tend to significantly overestimate surface O_3 in the Southeast US (Lin et al., 2008;~~,~~ Fiore et al., 2009;~~,~~ Brown-Steiner et al., 2015; Canty et al., 2015), and this is partially ~~speculated to be~~ driven ~~in part~~ by a likely~~n~~ overestimation of anthropogenic NO_x emissions (Anderson et al., 2014)~~n estimates~~. Thus, even though lightning is known to impact ambient air quality, including this additional NO_x source can worsen biases in model O_3 ~~model performance~~ in some locations and time periods due to other errors in the modeling system. As noted in Table 1, for the SE, the MB values increased by about 1.6 ppb in mNLDN and less than 1 ppb in hNLDN and pNLDN. Nevertheless, the correlation coefficients for mNLDN and pNLDN are almost the same with the Base, and hNLDN was slightly higher (0.77 compared to 0.76). These correlations indicate that even though additional NO_x increases the mean bias, when it is added correctly in time and space, as with the case of hNLDN, the spatial and temporal correlation are slightly improved. In the Upper Midwest (UM), the lowest errors and biases among the model cases are associated with hNLDN, while the worst performance is with mNLDN. In the Lower Midwest (LM), hNLDN performed comparable with the Base, with hNLDN having the highest correlation and lowest mean errors, while the Base has the lowest mean biases. Rocky Mountain (RM) is the only region that shows an underestimation of DM8HR O_3. In this region all the model cases with LNO~~x~~ outperformed the Base case in all the metrics. Among the three model cases with LNO~~x~~, mNLDN produced the lowest MB and NMB values, while hNLDN had the lowest RMSE and NME, and the highest correlation. In the Pacific Coast (PC) region, lightning activity is generally very low compared to other regions (Kang and Pickering, 2018). All model cases with LNO~~x~~ outperformed the Base case, especially hNLDN which had the lowest mean error and bias and highest correlation among all the cases.

Most of the NO$_x$ produced by lightning is distributed in the middle and upper troposphere with only a small portion being distributed close to the surface. As a result, the impact on ground-level NO$_x$ mixing ratios is small. Table 2 shows all the model cases produced similar statistics for the daily mean NO$_x$ mixing ratios at AQS sites across the domain and within all the subregions. Although the changes in model performance are small, the model cases with LNO~~x~~ exhibit similar or slightly better performance than the Base case.

3.1.2 Time series

Figure 2 presents ~~the~~ timeseries of regional-mean observed and modeled DM8HR O$_3$ for the entire domain and the SE and RM regions during July 2011. Over the domain and in SE, all the model cases overestimate the mean DM8HR O$_3$ mixing ratios on all days with the Base being the closest to the observations. hNLDN is almost the same as the Base with slightly higher values on some days. Among all the cases, mNLDN produced the highest values on almost all days through the month, on the order of 1-2 ppb higher than the Base. In contrast, in the RM region, the Base significantly underestimates DM8HR O$_3$ mixing rations on all the days during the month, while all model cases with LNO~~x~~ improved model predictions relative to observations in the region. Among the three model cases with LNO~~x~~, mNLDN produced the lowest bias for all the days, closely followed by hNLDN.

Figure 3 displays the average daily mean NO$_x$ mixing ratios at AQS sites over the same regions as in Figure 2. On most of the days in July 2011, over the domain and in the SE, the model ~~cases~~ overestimate NO$_x$ values, and on almost half of the days, the overestimation is significant (up to 100 %). As noted in Table 2, on average, the overestimation is ~17 % over the domain and ~43 % in SE. However, in RM, the predicted NO$_x$ mixing ratios closely follow the daily observations and on average the modeled and observed magnitude is almost identical (~3 % difference). All the model cases, with or without LNO~~x~~, produced almost the same mean NO$_x$

mixing ratios at the surface. However, the different cases produce different levels of LNO~~x~~ in the middle and upper troposphere, resulting in differences in O_3 production and transport which impact radiative forcing and also downwind ground-level O_3 levels. We further explore these features in Section 3.2 which presents evaluation of modeled vertical pollutant distributions.

3.1.3 Diurnal variations

Diurnal plots are used to further examine differences in model evaluation for O_3 and NO_x. Figure 4 shows the mean diurnal profiles for hourly O_3 and NO_x over the entire domain, SE, and RM. On a domain mean basis, all model cases overestimate O_3 during the daytime hours, while in the SE, the overestimation spans all the hours. In RM, the model cases significantly underestimate O_3 across all the hours except for a few early morning hours, when the model predicted values are very close to the observations. Among all the model cases, as expected, the most prominent differences occurred during the midday hours when the photochemistry is most active. However, the difference between hNLDN (and mNLDN) and the Base is also significant during the night in the RM region, even though the O_3 levels are low. This may be attributed to NO_x-related nighttime chemistry in part caused by freshly released NO by cloud-to-ground lightning flashes. The diurnal variations of ~~NOx~~ NO_x are similar over the domain and in the regions for all model cases. Appel et al. (2017) reported a significant overestimation of ~~NOx~~ NO_x mixing ratios at AQS sites during nighttime hours and underestimation during daytime hours. The bias pattern is identical for all of the LNO~~x~~ model cases evaluated here (Figure 4).

3.1.4 Spatial variations

Figure 5 shows the impact of the different LNO~~x~~ schemes on model performance for DM8HR O_3 at AQS sites. The spatial maps show the difference in absolute MB between the cases with lightning NO_x emissions and the Base and is calculated as follows. First, the absolute MB was calculated at each site for each case, e.g. $|MB_{[Base - Obs]}|$, then the difference in absolute MB was calculated between model cases, e.g. $|MB_{[hNLDN-Obs]}| - |MB_{[Base - Obs]}|$. The histograms of the differences in absolute MB between model cases in Figure 5 are provided to show the distribution of the change in model performance across space, i.e. the frequency of an improvement in model performance versus a degradation in model performance between cases.

As shown in Figure 5, the mNLDN shows increased model bias in the east US and along the California coast, but reduced model bias in the RM. At a majority of the AQS sites, it increases the model bias (only decreases at 26.8 % (346) of the sites). The hNLDN also significantly reduces model bias in the RM with a moderate increase in the SE. Overall, in the hNLDN, the mean bias decreased at 61.2 % (791) of AQS sites. Similar to mNLDN, increases in mean bias are noted at 29.3 % (378) of the AQS sites in the pNLDN simulation. As noted in the histograms, the distribution of the model bias in the pNLDN is much narrower than both mNLDN and hNLDN, eliminating the large bias increases in mNLDN and the significant bias decreases in hNLDN.

3.2 Vertical evaluation for O_3 and ~~NO_x~~NO_x

3.2.1 Ozone-sonde observations

A large source of uncertainty in the specification of LNO~~x~~ is its vertical allocation, which can impact the model's ability to accurately represent the variability in both chemistry and transport. To further assess the impact of the vertical LNO~~x~~ specification on model results, we compared vertical profiles of simulated model O_3 with extensive ozonesonde measurements available during the study period. Figure 6 presents the vertical profiles for O_3 sonde measurements and paired model estimates of all model cases at Beltsville, MD and Edgewood, MD. At each location, observations from multiple days are available (one or two soundings per day) during the 2011 DISCOVER-AQ campaign in July 2011. The model evaluation was limited to days where the inclusion of LNO~~x~~ has an obvious impact (the mean vertical profiles of LNO cases are separable from that of the base case~~the vertical profile lines can be separated~~) on the model estimates (July 21, 22, 28 and 29 at Beltsville, and July 21, 22, 28, 29, and 30 at Edgewood). We paired the observed data with model estimates in time and space and averaged the model and observed values at each model layer. Only data below 12 km altitude are plotted in Figure 6 to exclude possible influence of stratospheric air on O_3. As can be seen in Figure 6, at both locations the Base case underestimates O_3 mixing ratios above about ~~from around~~ 1 km ~~upwards,~~ but overestimates values closer to the surface. When LNO~~x~~ is included in the simulations, the predicted O_3 mixing ratios increase relative to the Base case starting around 2km, with greater divergence from the Base case at higher altitudes. The two model cases, hNLDN and mNLDN, produced similar O_3 levels from the surface to ~~until~~ about 6 km, but above that altitude the mNLDN ozone mixing ratios were higher. All the model cases with LNO~~x~~ performed much better aloft than the Base case. Near the surface, all the model cases overestimated O_3, however hNLDN had smaller bias than the other simulations. This may be attributed to the fact that only hNLDN used the observed lightning flash data directly, and as a result, LNO~~x~~ was estimated more accurately in time and space. This improvement in model bias at the surface is further investigated in the next section using evaluation against P-3B measurements.

3.2.2 P-3B measurement

Extensive measurements of lower tropospheric chemical composition distributions over the Northeastern U.S. are available from instruments onboard the P-3B aircraft on 14 days of the DISCOVER-AQ campaign. We utilize measurements from one of the days (28 July 2011) with noticeable (the mean vertical profiles of LNO~~$_x$~~ cases are separable from that of the base case) lightning impacts, to evaluate the model simulations. Figure 7 shows measured O_3 mixing ratios overlaid on the modeled vertical time-section for 1030 – 1730 UTC. The color-filled circles represent measured O_3 mixing ratios averaged over 60 seconds and the background is the model estimated vertical profiles from the grid cells containing the P-3B flight path for that hour and location. As indicated in the Base case (Figure 7a), the model tends to overestimate O_3 mixing ratios from the surface to about 2 km, but underestimate at altitudes above 2 km. The hNLDN reduced the overestimation below 2km, e.g. fewer grid cells with mixing ratios above 90ppb (shown in red). The other two cases (mNLDN, pNLDN) did not produce the same improvement near the surface. The hNLDN also decreases the underestimation aloft compared to the Base case with O_3 mixing ratios in the 55-65 ppb range (light blue colors), better matching the measured values. This decrease in underestimation aloft is also seen in the mNLDN case, but to a lesser degree while the pNLDN case shows only slight improvement aloft over the Base simulation.

To further differentiate the three LNO~~x~~ model cases, Figures 8-10 show the difference in the time-sections between each of the model cases with LNO~~x~~ and the Base for NO, NO_x, and O_3 from all the model layers along the P-3B flight path on July 28. As seen in Figure 8, the hNLDN scheme injected most NO above 5 km with a peak between 13-14 km and only a small amount near the surface~~, with the maximum amount injected between 13-14 km~~. After release into the atmosphere, NO is quickly converted into NO_2 in the presence of O_3, and these collectively result in the NO_x ~~($NO+NO_2$)~~ vertical time-section (local production plus transport) shown in the middle panel of Figure 8. NO_x is further mixed down through the time-section and more persistent along the flight path near the surface than is NO. As a result, significant O_3 is produced above 3 km and the maximum O_3 difference appears between 9 and 14 km during the early afternoon hours (from 13:30 to 17:30). However, from surface to about 2 km, O_3 is reduced consistently across the entire period, and this is the result of O_3 titration by NO from cloud-to-ground lightning flashes that must have been transported to this layer by storm downdrafts. Since O_3 is significantly underestimated above 3 km and overestimated near the surface by the Base model, the inclusion of LNO~~x~~ greatly improved the model's performance under both conditions.

Comparison of Figure 9 (mNLDN) with Figure 8 (hNLDN) reveals that the time-sections of NO and NO_x ~~above 5 km~~ are similar above 5 km ~~for these two cases,~~ but ~~they are~~ dramatically different near the surface. The near-surface increase in ambient NO noted in the hNLDN is absent in mNLDN, and in fact there are some small decreases in NO, although the reason for this is unclear. The increase in O_3 aloft in the mNLDN case is similar to that seen in the hNLDN case. However, the near-surface reduction in O_3 is almost absent. In the pNLDN case (Figure 10), NO mixing ratios are much less than those in hNLDN and mNLDN in the upper layers as a result of less column NO being generated by the linear parameterization. The resulting NO_x time-section is also smoothed. The pNLDN time-sections for NO, NO_x and O_3 near the surface are similar to the mNLDN case with no change or small decreases compared to the Base case. O_3 mixing ratios increase by more than 30 ppb during the afternoon hours between 10 – 13 km in the pNLDN case, however the increase is not as intense and widespread as the other cases. In summary, the hNLDN scheme produces estimates that are more consistent with measurements at the surface and aloft, compared to the other simulations, reflecting the advantage of using the spatially and temporally-resolved observed lightning flash data. The model performance improvement for simulated O_3 distributions also suggests robustness in the vertical distribution scheme when LNO~~x~~ is generated at the right time and location.

To corroborate the above time-section distributions of NO, ~~NO_x~~NO_x, and O_3 in the lightning cases, the lightning NO emissions are traced back on July 28 for each case. It is found that in all cases, the lightning NO was injected approximately~~bout~~ 200 km upwind (north-west) of the flight path. The hNLDN case captured two injections: one occurred during the morning hours (5:00 to 7:00 am) and the other happened during the afternoon hours (after 2:30 pm). Both mNLDN and pNLND captured the afternoon lightning event at the later time (after 3:30 pm for mNLDN and after 4:30 pm for pNLDN) with varying intensity, but neither captured the morning lightning event, which explains why the increase of NO and ~~NO_x~~ NO_x in the hNLDN case (Figure 8) did not occur in the mNLDN and pNLDN cases (Figures 9 and 10). Also note that the significant increase of NO during the time period from 11:00 to 13:00 occurred about 5 hours after the lightning NO was injected at about 200 km upwind in the hNLDN case.

To expand on the evaluation in Figures 7-10 which focused on measurements from July 28, 2011, we retrieved all the P-3B measurements on days with noticeable lightning impact (July 21, 22, 28, and 29). The 3-D paired observation-model data were grouped together by spiral site and the mean biases (model – observation) were plotted in Figure 11 (a and b) for O_3 and NO, respectively. The boxplots for O_3 in Figure 11a suggests that the Base exhibited larger bias with greater spread (i.e. larger interquartile range) than other model cases incorporating LNO~~x~~ at most of the locations where aircraft spirals were conducted. At all locations except Aldino, the lowest mean biases in simulated NO and O_3 are noted in the hNLDN simulation.

3.3 Deposition evaluation for nitrate

In addition to contributing to tropospheric O_3 formation, NO_x oxidation also leads to gaseous nitric acid and particulate-nitrate which are eventually removed from the atmosphere by dry and wet deposition of nitrate (NO_3^-). As a result, inclusion of NO_x from lightning also plays an important role in nitrogen deposition modeling. To assess the impacts of incorporating LNO~~x~~ emissions on simulated oxidized nitrogen deposition, we compared model estimated amounts of precipitation from NTN network (http://nadp.slh.wisc.edu/ntn/) and wet deposition of NO_3^- with measurements from the NADP network (http://nadp.slh.wisc.edu/). During summer months in 2011 (June -August) the WRF model generally reproduces the observed precipitation with a slight underestimate in the east, but the Base model simulation tends to underestimate wet deposition of NO_3^- across the domain, with the greatest underestimation in the SE and UM (See Table 3 and Figure 12). All three LNO~~x~~ simulations increase wet deposition amounts of NO_3^- and decrease model bias in all regions. The bottom panel of Figure 12 shows that the mNLDN simulation resulted in the largest increase over the base model estimates. The NMB is reduced from -35 % in the Base to -15 % in mNLDN across the domain and from -32 % to -2 % in the SE. The hNLDN shows very similar model performance to the mNLDN case. In contrast, the wet deposition NO_3^- estimates from the pNLDN case are only slightly higher than the Base case, and as a result the evaluation statistics for pNLND are very similar to the Base statistics. As discussed earlier, the mNLDN tends to produce the most LNO~~x~~ among the three LNO~~x~~ schemes, thus it results in the smallest errors in terms of wet deposition of NO_3^- when compared to the Base simulation that significantly underestimated NO_3^- wet deposition. It should be noted that in addition to the LNO~~x~~ contributions, errors in modeled precipitation amounts and patterns also likely influence the underestimation of NO_3^- wet deposition.

4. Conclusions

A detailed evaluation of lightning NO_x emission estimation parameterizations available in the CMAQ modeling system was performed through comparisons of model simulation results with surface and aloft air quality measurements.

Our analysis indicates that incorporation of LNO~~x~~ emissions enhanced O_3 production in the middle and upper troposphere, where O_3 mixing ratios were often significantly underestimated without the representation of LNO~~x~~. Though the impact on surface O_3 varies from region to region and is also dependent on the accuracy of the NO_x emissions from other sources, the inclusion of LNO~~x~~, when it is injected at the appropriate time and location, can improve the model estimates. In regions where the base model estimates of O_3 were biased high, the inclusion of LNO~~x~~ further increased the model bias; and a systematic increase is noted in the correlation with measurements, suggesting that emissions from other sources likely drive the overestimation. Identifying how errors in emissions inputs from different sources interact with errors in meteorological modeling of mixing and transport, remains a challenging but critical task. Likewise, all the LNO~~x~~ schemes also enhanced the accumulated wet deposition of NO_3^-, that was significantly underestimated by the base model without LNO~~x~~ throughout the modeling domain except the Pacific Coast.

Uncertainty remains in modeling the magnitude and spatial, temporal and vertical distribution of lightning produced NO_x. LNO~~x~~ schemes are built on numerous assumptions and all current schemes also depend on the skill of the upstream meteorological models in describing convective activity. Nevertheless, these schemes reflect our best understanding and knowledge at the time when the schemes were implemented. The use of hourly information on lightning activity yielded LNO~~$_x$~~ emissions that generally improved model performance for ambient O_3 and NO_x as well as oxidized nitrogen wet deposition amounts. As more high-quality data from both ground and satellite measurements become available, the performance of the LNO~~$_x$~~ schemes will continue to improve.

Since the pNLDN scheme was developed using historical data corelating lightning activity with convective precipitation, the scheme could be employed for applications involving air quality forecasting and future projections when observed lightning information is not available.

Code and data availability

CMAQ model documentation and released versions of the source code, including all model code used in his study, are available at https://github.com/USEPA/CMAQ/tree/5.2 ~~https://www.epa.gov/cmaq~~. The data processing and analysis scripts are available upon request. The WRF model is available for download through the WRF website (http://www2.mmm.ucar.edu/wrf/users/wrfv3.8/updates-3.8.html~~http://www.wrf-model.org/index.php~~).
The raw lightning flash observation data used are not available to the public but can be purchased through Vaisala Inc. (https:// www.vaisala.com/en/products/systems/lightning-detection). The lightning data obtained from Vaisala Inc. is the cloud-to-ground lightning flashes over the contiguous United States. The immediate data behind the tables and figures are available from https://zenodo.org/record/3360744~~262~~1096 (Kang and Foley, 2019). Additional input/output data for CMAQ model utilized for this analysis are available upon request as well.

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Author Contribution

Daiwen Kang: data collection, algorithm design, model simulation, analysis, and manuscript writing.

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