Comments to the Author:

Minor comment: I do not think that the surface statistics presented in Table 1 and 2 suggest "the importance of including the contributions of this source for improving the spatial and temporal variability in model predictions". They show a very marginal gain. It is not surprising because the base model is "tuned" (input parameters and schemes adjusted) to match as well as possible existing observations. Therefore, even by making the model more realistic in terms of emissions or physics, one may degrade initially the model performances. Overall, regarding the importance of LNO, results are much more convincing for the free troposphere. I do not think you need to add "suggesting…predictions" here. The point is made more naturally in the conclusion.

The topical editor has made a very good point and we completely agree. We have now revised the manuscript by removing the sentence "suggesting … predictions" on Page 7 Lines 200 – 201. And made minor changes to make the sentence flow smoothly.

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 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) 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 LNOx 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 LNOx), the impact of LNOx 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, LNOx 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 LNOx leads to reduction in biases and errors and an increase in correlation coefficients at almost all the NADP/NTN sites. Among the three LNOx 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 LNOx compared to the base simulation that does not include LNOx.

1. Introduction

The potential importance of NO_X produced by lightning (LNO_X) 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 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 burden and its subsequent impacts on atmospheric chemistry will increase in the United States and other developed countries (Kang and Pickering, 2018). The significant impact of LNO_X on surface air quality was earlier reported by Napelenok et al. (2008), in that low-biases in upper tropospheric NOx in Community Multiscale Air Quality Model (CMAQ) (Byun and Schere, 2006) simulations without LNO_X emissions made it difficult to constrain ground-level NOx 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 LNOx 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

85   National Lightning Detection Network (NLDN) (Orville et al., 2002), recent LNO_X

86   parameterization schemes started to include the observed lightning flash information to constrain

87   LNO_X in regional Chemical Transport Models (CTMs) (Allen et al., 2012). In Kang et al. (2019),

88   we described the existing LNO_X parameterization scheme that is based on the monthly NLDN

89   (mNLND) lightning flash data, and an updated scheme using hourly NLDN (hNLDN) lightning

90   flash data in the CMAQ lightning module. In addition, we also developed a scheme based on

91   linear and log-linear regression parameters using multiyear NLDN observed lightning flashes

92   and model predicted convective precipitation rate (pNLDN). The preliminary assessment of

93   these schemes based on total column LNO_X suggests that all the schemes provide reasonable

94   LNO_X estimates in time and space, but during summer months, the mNLDN scheme tends to

95   produce the most and the pNLDN scheme the least LNO_X.

96   The first study on the impact of LNO_X on surface air quality using CMAQ was conducted

97   by Allen et al. (2012) and followed by Wang et al. (2013) with different ways for parameterizing

98   LNO_X production and different model configurations. In this study, we present performance

99   evaluations using each of the LNO_X production schemes (mNLDN, hNLDN, pNLDN) described

100   by Kang et al. (2019) to provide estimates of LNO_X in CMAQ. In addition to examination of

101   differences in air quality estimates between these schemes, we compare the model predictions to

102   base model estimates without LNOx and evaluate the estimates from all of the simulations

103   against surface and airborne observations.

104   Section 2 describes the model configuration, simulation scenarios, analysis methodology,

105   and observational data. Section 3 presents the analysis results and Section 4 presents the

106   conclusions.

107

108   **2. Methodology**

109   **2.1 The CMAQ model and simulation configurations**

110   The three LNO_x production schemes described in Kang et al (2019) were incorporated

111   into CMAQ v5.2 (Appel et al. 2017; doi:10.5281/zenodo.1167892). The chemical mechanism

112   used was CB6 (Yarwood et al., 2010) and the aerosol module was AERO6 (Nolte et al., 2015).

113 The meteorological inputs were provided by the Weather Research and Forecasting (WRF)

114 model version 3.8 and the model-ready meteorological input files were created using version 4.2

115 of the meteorology–chemistry interface processor (MCIP; Otte and Pleim, 2010).

116 The modeling domain covers the entire contiguous United States (CONUS) and

117 surrounding portions of northern Mexico and southern Canada, as well as the eastern Pacific and

118 western Atlantic oceans. The model domain consists of 299 north–south grid cells by 459 east–

119 west grid cells utilizing 12 km x 12 km horizontal grid spacing, 35 vertical layers with varying

120 thickness extending from the surface to 50 hPa and an approximately 10m midpoint for the

121 lowest (surface) model layer. The simulation time period covers the months from April to

122 September 2011 with a 10-day spin-up period in March.

123 Emission input data were based on the 2011 National Emissions Inventory

124 (https://www.epa.gov/air-emissions-inventories). The raw emission files were processed using

125 version 3.6.5 of the Sparse Matrix Operator Kernel Emissions (SMOKE;

126 https://www.cmascenter.org/smoke/) processor to create gridded speciated hourly model-ready

127 input emission fields for input to CMAQ. Electric generating unit (EGU) emissions were

128 obtained using data from EGUs equipped with a continuous emission monitoring system

129 (CEMS). Plume rise for point and fire sources were calculated in-line for all simulations (Foley

130 et al., 2010). Biogenic emissions were generated in-line in CMAQ using BEIS versions 3.61

131 (Bash et al., 2016). All the simulations employed the bidirectional (bi-di) ammonia flux option

132 for estimating the air–surface exchange of ammonia.

133 There are four CMAQ simulation scenarios for this study: 1) simulation without LNO_X

134 (Base), 2) simulation with LNO_X generated by the scheme based on monthly information from

135 the NLDN (mNLDN), 3) simulation with LNO_X generated by scheme based on hourly

136 information from the NLDN (hNLDN), and 4) simulation with LNO_X generated by the scheme

137 parameterizing lightning emissions based on modeled convective activity (pNLDN) as described

138 in detail in Kang et al. (2019). All other model inputs, parameters and settings were the same

139 across the four simulations. The vertical distribution algorithm is the same for all the LNO_X

140 schemes as also described in Kang et al. (2019).

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2.2 Observations and analysis techniques

To assess the impact of LNOx 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. 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 LNOx 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 LNOx 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 LNOx 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.

174
175 **3. Results**

176 **3.1 Ground-level evaluation for O$_3$ and NO$_X$**

177 **3.1.1 Statistical performance metrics**

178   Tables 1 and 2 display the statistical model performance metrics for daily maximum 8-hr
179 (DM8HR) O$_3$ and daily mean NOx mixing ratios over the domain and each analysis region for
180 all four model cases in July 2011 (Base, mNLDN, hNLDN, and pNLDN). The best performance
181 metrics among the model cases are highlighted in bold. As shown in Table 1, for DM8HR O$_3$,
182 the Base simulation has the lowest MB and NMB values over the Domain, while hNLDN
183 produced the smallest RMSE and NME values. mNLDN generated the largest values for both
184 error (RMSE and NME) and biases (MB and NMB), followed by pNLDN. More importantly, all
185 model cases with LNOx exhibit slightly higher correlation coefficients than the Base simulation,
186 suggesting the importance of including the contributions of this source for improving the spatial
187 and temporal variability in model predictions. Additionally, the hNLDN simulation exhibited
188 higher correlation and lower bias and error relative to the measurements indicating the value of
189 higher temporal resolution lightning activity for representing the associated NO$_x$ emissions and
190 their impacts on tropospheric chemistry.

191   Examining the regional results for DM8HR O$_3$ in Table 1, the statistical measures indicate
192 that in the Northeast (NE), hNLDN outperformed all other model cases with the lowest errors
193 and biases and highest correlation coefficient. In Southeast (SE), the Base performed better with
194 the lowest errors and mean biases, but the correlation coefficient (R) value for hNLDN is slightly
195 higher. Among all the LNOx cases, mNLDN produced the worst statistics in this region.
196 Historically, CTMs tend to significantly overestimate surface O$_3$ in the Southeast US (Lin et al.,
197 2008l Fiore et al., 2009l Brown-Steiner et al., 2015; Canty et al., 2015), and this is speculated to
198 be driven in part by an overestimation of anthropogenic NOx emission estimates. Thus, even
199 though lightning is known to impact ambient air quality, including this additional NOx source
200 can worsen model performance in some locations and time periods due to other errors in the
201 modeling system. As noted in Table 1, for SE, the MB values increased by about 1.6 ppb in
202 mNLDN and less than 1 ppb in hNLDN and pNLDN. Nevertheless, the correlation coefficients
203 for mNLDN and pNLDN are almost the same with the Base, and hNLDN was slightly higher

204 (0.77 compared to 0.76). These correlations indicate that even though additional NOx increases
205 the mean bias, when it is added correctly in time and space, as with the case of hNLDN, the
206 spatial and temporal correlation are improved. In Upper Midwest (UM), the lowest errors and
207 biases among the model cases are associated with hNLDN, while the worst performance is with
208 mNLDN. In the Lower Midwest (LM), hNLDN performed comparable with the Base, with
209 hNLDN having the highest correlation and lowest mean errors, while the Base has the lowest
210 mean biases. Rocky Mountain (RM) is the only region that shows an underestimation of
211 DM8HR O_3. In this region all the model cases with LNOx outperformed the Base case in all the
212 metrics. Among the three model cases with LNOx, mNLDN produced the lowest MB and NMB
213 values, while hNLDN had the lowest RMSE and NME, and the highest correlation. In the Pacific
214 Coast (PC) region, lightning activity is generally very low compared to other regions (Kang and
215 Pickering, 2018). All model cases with LNOx outperformed the Base case, especially hNLDN
216 which had the lowest mean error and bias and highest correlation among all the cases.

217   Most of the NO_x produced by lightning is distributed in the middle and upper troposphere
218 with only a small portion being distributed close to the surface. As a result, the impact on
219 ground-level NO_x mixing ratios is small. Table 2 shows all the model cases produced similar
220 statistics for the daily mean NO_x mixing ratios at AQS sites across the domain and within all the
221 subregions. Although the changes in model performance are small, the model cases with LNOx
222 exhibit similar or slightly better performance than the Base case.

223 **3.1.2 Time series**

224   Figure 2 presents the timeseries of regional-mean observed and modeled DM8HR O_3 for
225 the entire domain and the SE and RM regions during July 2011. Over the domain and in SE, all
226 the model cases overestimate the mean DM8HR O_3 mixing ratios on all days with the Base being
227 the closest to the observations. hNLDN is almost the same as the Base with slightly higher
228 values on some days. Among all the cases, mNLDN produced the highest values on almost all
229 days through the month, on the order of 1-2 ppb higher than the Base. In contrast, in the RM
230 region, the Base significantly underestimates DM8HR O_3 mixing rations on all the days during
231 the month, while all model cases with LNOx improved model predictions relative to
232 observations in the region. Among the three model cases with LNOx, mNLDN produced the
233 lowest bias for all the days, closely followed by hNLDN.

234   Figure 3 displays the average daily mean NOx mixing ratios at AQS sites over the same

235   regions as in Figure 2. On most of the days in July 2011, over the domain and in SE, the model

236   cases overestimate NOx values, and on almost half of the days, the overestimation is significant

237   (up to 100%). As noted in Table 2, on average, the overestimation is ~17% over the domain and

238   ~43% in SE. However, in RM, the predicted NOx mixing ratios closely follow the daily

239   observations and on average the modeled and observed magnitude is almost identical (~3%

240   difference). All the model cases, with or without LNOx, produced almost the same mean NOx

241   mixing ratios at the surface. However, the different cases produce different levels of LNOx in the

242   middle and upper troposphere, resulting in differences in O_3 production and transport which

243   impact ground-level O_3 levels. We further explore these features in Section 3.2 which presents

244   evaluation of modeled vertical pollutant distributions.

245   **3.1.3 Diurnal variations**

246   Diurnal plots are used to further examine differences in model evaluation for O_3 and

247   NO_x. Figure 4 shows the mean diurnal profiles for hourly O_3 and NO_x over the entire domain,

248   SE, and RM. On a domain mean basis, all model cases overestimate O_3 during the daytime

249   hours, while in the SE, the overestimation spans all the hours. In RM, the model cases

250   significantly underestimate O_3 across all the hours except for a few early morning hours, when

251   the model predicted values are very close to the observations. Among all the model cases, as

252   expected, the most prominent differences occurred during the midday hours when the

253   photochemistry is most active. However, the difference between hNLDN (and mNLDN) and the

254   Base is also significant during the night in the RM region, even though the O_3 levels are low.

255   This may be attributed to NOx-related nighttime chemistry in part caused by freshly released NO

256   by cloud-to-ground lightning flashes. The diurnal variations of NOx are similar over the domain

257   and in the regions for all model cases. Appel et al. (2017) reported a significant overestimation of

258   NOx mixing ratios at AQS sites during nighttime hours and underestimation during daytime

259   hours. The bias pattern is identical for all of the LNOx model cases evaluated here (Figure 4).

260   **3.1.4 Spatial variations**

261   Figure 5 shows the impact of the different LNO_x schemes on model performance for

262   DM8HR O_3 at AQS sites. The spatial maps show the difference in absolute MB between the

263   cases with lightning NO_x emissions and the Base and is calculated as follows. First, the absolute

264   MB was calculated at each site for each case, e.g. $|MB_{[Base - Obs]}|$, then the difference in absolute

265   MB was calculated between model cases, e.g. $|MB_{[hNLDN-Obs]}| - |MB_{[Base - Obs]}|$. The histograms of

266   the differences in absolute MB between model cases in Figure 5 are provided to show the

267   distribution of the change in model performance across space, i.e. the frequency of an

268   improvement in model performance versus a degradation in model performance between cases.

269   As shown in Figure 5, the mNLDN shows increased model bias in the east US and along the

270   California coast, but reduced model bias in the RM. At a majority of the AQS sites, it increases

271   the model bias (only decreases at 26.8% (346) of the sites). The hNLDN also significantly

272   reduces model bias in the RM with a moderate increase in the SE. Overall, in the hNLDN, the

273   mean bias decreased at 61.2% (791) of AQS sites. Similar to mNLDN, increases in mean bias

274   are noted at 29.3% (378) of the AQS sites in the pNLDN simulation. As noted in the histograms,

275   the distribution of the model bias in the pNLDN is much narrower than both mNLDN and

276   hNLDN, eliminating the large bias increases in mNLDN and the significant bias decreases in

277   hNLDN.

278   **3.2 Vertical evaluation for O_3 and NO_X**

279   **3.2.1   Ozone-sonde observations**

280   A large source of uncertainty in the specification of LNOx is its vertical allocation, which

281   can impact the model's ability to accurately represent the variability in both chemistry and

282   transport. To further assess the impact of the vertical LNOx specification on model results, we

283   compared vertical profiles of simulated model O_3 with extensive ozonesonde measurements

284   available during the study period. Figure 6 presents the vertical profiles for O_3 sonde

285   measurements and paired model estimates of all model cases at Beltsville, MD and Edgewood,

286   MD. At each location, observations from multiple days are available (one or two soundings per

287   day) during the 2011 DISCOVER-AQ campaign in July 2011. The model evaluation was limited

288   to days where the inclusion of LNOx has an obvious impact (the vertical profile lines can be

289   separated) on the model estimates (July 21, 22, 28 and 29 at Beltsville, and July 21, 22, 28, 29,

290   and 30 at Edgewood). We paired the observed data with model estimates in time and space and

291   averaged the model and observed values at each model layer. Only data below 12 km altitude are

292   plotted in Figure 6 to exclude possible influence of stratospheric air on O_3. As can be seen in

293 Figure 6, at both locations the Base case underestimates O_3 mixing ratios from around 1 km

294 upwards, but overestimates closer to the surface. When LNOx is included in the simulations, the

295 predicted O_3 mixing ratios increase relative to the Base case starting around 2km, with greater

296 divergence from the Base case at higher altitudes. The two model cases, hNLDN and mNLDN,

297 produced similar O_3 levels until about 6 km, but above that altitude the mNLDN ozone mixing

298 ratios were higher. All the model cases with LNOx performed much better aloft than the Base

299 case. Near the surface, all the model cases overestimated O_3, however hNLDN had smaller bias

300 than the other simulations. This may be attributed to the fact that only hNLDN used the observed

301 lightning flash data directly, and as a result, LNOx was estimated more accurately in time and

302 space. This improvement in model bias at the surface is further investigated in the next section

303 using evaluation against P-3B measurements.

304 **3.2.2 P-3B measurement**

305 Extensive measurements of lower tropospheric chemical composition distributions over

306 the Northeastern U.S. are available from instruments onboard the P-3B aircraft on 14 days of the

307 DISCOVER-AQ campaign. We utilize measurements from one of the days (28 July 2011) with

308 noticeable (the mean vertical profiles of LNO_X cases are separable from that of the base case)

309 lightning impacts, to evaluate the model simulations. Figure 7 shows measured O_3 mixing ratios

310 overlaid on the modeled vertical time-section for 1030 – 1730 UTC. The color-filled circles

311 represent measured O_3 mixing ratios averaged over 60 seconds and the background is the model

312 estimated vertical profiles from the grid cells containing the P-3B flight path for that hour and

313 location. As indicated in the Base case (Figure 7a), the model tends to overestimate O_3 mixing

314 ratios from the surface to about 2 km, but underestimate at altitudes above 2 km. The hNLDN

315 reduced the overestimation below 2km, e.g. fewer grid cells with mixing ratios above 90ppb

316 (shown in red). The other two cases (mNLDN, pNLDN) did not produce the same improvement

317 near the surface. The hNLDN also decreases the underestimation aloft compared to the Base case

318 with O_3 mixing ratios in the 55-65 ppb range (light blue colors), better matching the measured

319 values. This decrease in underestimation aloft is also seen in the mNLDN case, but to a lesser

320 degree while the pNLDN case shows only slight improvement aloft over the Base simulation.

321 To further differentiate the three LNOx model cases, Figures 8-10 show the difference in

322 the time-sections between each of the model cases with LNOx and the Base for NO, NOx, and

11

323   O_3 from all the model layers along the P-3B flight path on July 28. As seen in Figure 8, the

324   hNLDN scheme injected most NO above 5 km and small amount near the surface, with the

325   maximum amount injected between 13-14 km. After release into the atmosphere, NO is quickly

326   converted into NO_2 in the presence of O_3, and these collectively result in the NO_x ($NO+NO_2$)

327   vertical time-section (local production plus transport) shown in the middle panel of Figure 8.

328   NO_x is further mixed down through the time-section and more persistent along the flight path

329   near the surface than is NO. As a result, significant O_3 is produced above 3 km and the maximum

330   O_3 difference appears between 9 and 14 km during the early afternoon hours (from 13:30 to

331   17:30). However, from surface to about 2 km, O_3 is reduced consistently across the entire period,

332   and this is the result of O_3 titration by NO from cloud-to-ground lightning flashes that must have

333   been transported to this layer by storm downdrafts. Since O_3 is significantly underestimated

334   above 3 km and overestimated near the surface by the Base model, the inclusion of LNOx

335   greatly improved the model's performance under both conditions.

336   Comparison of Figure 9 (mNLDN) with Figure 8 (hNLDN) reveals that the time-sections

337   of NO and NOx above 5 km are similar for these two cases, but they are dramatically different

338   near the surface. The near-surface increase in ambient NO noted in the hNLDN is absent in

339   mNLDN, and in fact there are some small decreases in NO, although the reason for this is

340   unclear. The increase in O_3 aloft in the mNLDN case is similar to that seen in the hNLDN case.

341   However, the near-surface reduction in O_3 is almost absent. In the pNLDN case (Figure 10), NO

342   mixing ratios are much less than those in hNLDN and mNLDN in the upper layers as a result of

343   less column NO being generated by the linear parameterization. The resulting NOx time-section

344   is also smoothed. The pNLDN time-sections for NO, NOx and O_3 near the surface are similar to

345   the mNLDN case with no change or small decreases compared to the Base case. O_3 mixing ratios

346   increase by more than 30 ppb during the afternoon hours between $10 - 13$ km in the pNLDN

347   case, however the increase is not as intense and widespread as the other cases. In summary, the

348   hNLDN scheme produces estimates that are more consistent with measurements at the surface

349   and aloft, compared to the other simulations, reflecting the advantage of using the spatially and

350   temporally-resolved observed lightning flash data. The model performance improvement for

351   simulated O_3 distributions also suggests robustness in the vertical distribution scheme when

352   LNOx is generated at the right time and location.

353   To corroborate the above time-section distributions of NO, NO_X, and O_3 in the lightning

354   cases, the lightning NO emissions are traced back on July 28 for each case. It is found that in all

355   cases, the lightning NO was injected about 200 km upwind (north-west) of the flight path. The

356   hNLDN case captured two injections: one occurred during the morning hours (5:00 to 7:00 am)

357   and the other happened during the afternoon hours (after 2:30 pm). Both mNLDN and pNLND

358   captured the afternoon lightning event at the later time (after 3:30 pm for mNLDN and after 4:30

359   pm for pNLDN) with varying intensity, but neither captured the morning lightning event, which

360   explains why the increase of NO and NO_X in the hNLDN case (Figure 8) did not occur in the

361   mNLDN and pNLDN cases (Figures 9 and 10). Also note that the significant increase of NO

362   during the time period from 11:00 to 13:00 occurred about 5 hours after the lightning NO was

363   injected at about 200 km upwind in the hNLDN case.

364   To expand on the evaluation in Figures 7-10 which focused on measurements from July

365   28, 2011, we retrieved all the P-3B measurements on days with noticeable lightning impact (July

366   21, 22, 28, and 29). The 3-D paired observation-model data were grouped together by spiral site

367   and the mean biases (model – observation) were plotted in Figure 11 (a and b) for O_3 and NO,

368   respectively. The boxplots for O_3 in Figure 11a suggests that the Base exhibited larger bias with

369   greater spread (i.e. larger interquartile range) than other model cases incorporating LNOx at most

370   of the locations where aircraft spirals were conducted. At all locations except Aldino, the lowest

371   mean biases in simulated NO and O_3 are noted in the hNLDN simulation.

372

3.3 Deposition evaluation for nitrate

373

374   In addition to contributing to tropospheric O_3 formation, NOx oxidation also leads to gaseous

375   nitric acid and particulate-nitrate which are eventually removed from the atmosphere by dry and

376   wet deposition of nitrate (NO_3^-). As a result, inclusion of NOx from lightning also plays an

377   important role in nitrogen deposition modeling. To assess the impacts of incorporating LNOx

378   emissions on simulated oxidized nitrogen deposition, we compared model estimated amounts of

379   precipitation from NTN network (http://nadp.slh.wisc.edu/ntn/) and wet deposition of NO_3^- with

380   measurements from the NADP network (http://nadp.slh.wisc.edu/). During summer months in

381   2011 (June -August) the WRF model generally reproduces the observed precipitation with a

382   slight underestimate in the east, but the Base model simulation tends to underestimate wet

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383   deposition of NO_3^- across the domain, with the greatest underestimation in the SE and UM (See

384   Table 3 and Figure 12). All three LNOx simulations increase wet deposition amounts of NO_3^-

385   and decrease model bias in all regions. The bottom panel of Figure 12 shows that the mNLDN

386   simulation resulted in the largest increase over the base model estimates. The NMB is reduced

387   from -35% in the Base to -15% in mNLDN across the domain and from -32% to -2% in the SE.

388   The hNLDN shows very similar model performance to the mNLDN case. In contrast, the wet

389   deposition NO_3^- estimates from the pNLDN case are only slightly higher than the Base case, and

390   as a result the evaluation statistics for pNLND are very similar to the Base statistics. As

391   discussed earlier, the mNLDN tends to produce the most LNOx among the three LNOx schemes,

392   thus it results in the smallest errors in terms of wet deposition of NO_3^- when compared to the

393   Base simulation that significantly underestimated NO_3^- wet deposition. It should be noted that in

394   addition to the LNOx contributions, errors in modeled precipitation amounts and patterns also

395   likely influence the underestimation of NO_3^- wet deposition.

396

397   **4. Conclusions**

398   A detailed evaluation of lightning NOx emission estimation parameterizations available

399   in the CMAQ modeling system was performed through comparisons of model simulation

400   results with surface and aloft air quality measurements.

401   Our analysis indicates that incorporation of LNOx emissions enhanced O_3 production in

402   the middle and upper troposphere, where O_3 mixing ratios were often significantly

403   underestimated without the representation of LNOx. Though the impact on surface O_3 varies

404   from region to region and is also dependent on the accuracy of the NOx emissions from other

405   sources, the inclusion of LNOx, when it is injected at the appropriate time and location, can

406   improve the model estimates. In regions where the base model estimates of O_3 were biased

407   high, the inclusion of LNOx further increased the model bias; and a systematic increase is

408   noted in the correlation with measurements, suggesting that emissions from other sources

409   likely drive the overestimation. Identifying how errors in emissions inputs from different

410   sources interact with errors in meteorological modeling of mixing and transport, remains a

411   challenging but critical task. Likewise, all the LNOx schemes also enhanced the accumulated

412   wet deposition of NO_3^-, that was significantly underestimated by the base model without

413   LNOx throughout the modeling domain except the Pacific Coast.

414   Uncertainty remains in modeling the magnitude and spatial, temporal and vertical

415   distribution of lightning produced NOx. LNOx schemes are built on numerous assumptions

416   and all current schemes also depend on the skill of the upstream meteorological models in

417   describing convective activity. Nevertheless, these schemes reflect our best understanding

418   and knowledge at the time when the schemes were implemented. The use of hourly

419   information on lightning activity yielded LNOx emissions that generally improved model

420   performance for ambient O_3 and NO_x as well as oxidized nitrogen wet deposition amounts.

421   As more high-quality data from both ground and satellite measurements become available,

422   the performance of the LNOx schemes will continue to improve.

423

Code and data availability

425   CMAQ model documentation and released versions of the source code, including all model
426   code used in his study, are available at https://www.epa.gov/cmaq. The data processing and
427   analysis scripts are available upon request. The WRF model is available for download through
428   the WRF website (http://www.wrf-model.org/index.php).
429   The raw lightning flash observation data used are not available to the public but can be
430   purchased through Vaisala Inc. (https:// www.vaisala.com/en/products/systems/lightning-
431   detection). The immediate data behind the tables and figures are available from
432   https://zenodo.org/record/2621096 (Kang and Foley, 2019). Additional input/output data for
433   CMAQ model utilized for this analysis are available upon request as well.

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435

436   **Disclaimer:** The views expressed in this paper are those of the authors and do not necessarily
437   represent the views or policies of the U.S. EPA.

438

Author Contribution

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

442   **Kristen Foley**: data analysis and manuscript writing.

443   **Rohit Mathur**: manuscript editing.

444   **Shawn Roselle**: manuscript editing.

445 **Kenneth Pickering:** manuscript editing.

446 **Dale Allen:** manuscript editing.

447

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1   **Simulating Lightning NO Production in CMAQv5.2: Performance**

2   **Evaluations**

3

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24

Abstract

26   This study assesses the impact of the lightning NO (LNO) production schemes in the

27   Community Multiscale Air Quality (CMAQ) model on ground-level air quality as well as aloft

28   atmospheric chemistry through detailed evaluation of model predictions of nitrogen oxides

29   (NO_x) and ozone (O_3) with corresponding observations for the U.S. For ground-level evaluations,

30   hourly O_3 and NO_x from the US EPA's AQS monitoring network are used to assess the impact of

31   different LNO schemes on model prediction of these species in time and space. Vertical

32   evaluations are performed using ozonesonde and P-3B aircraft measurements during the

33   DISCOVER-AQ campaign conducted in the Baltimore/Washington region during July 2011.

34   The impact on wet deposition of nitrate is assessed using measurements from the National

35   Atmospheric Deposition Program's National Trends Network (NADP/NTN). Compared with the

36   base model (without LNO), the impact of LNO on surface O_3 varies from region to region

37   depending on the base model conditions. Overall statistics suggest that for regions where surface

38   O_3 mixing ratios are already overestimated, the incorporation of additional NO from lightning

39   generally increased model overestimation of mean daily maximum 8-hr (DM8HR) O_3 by 1-2

40   ppb. In regions where surface O_3 is underestimated by the base model, LNO can significantly

41   reduce the underestimation and bring model predictions close to observations. Analysis of

42   vertical profiles reveals that LNO can significantly improve the vertical structure of modeled O_3

43   distributions by reducing underestimation aloft, and to a lesser degree decreasing overestimation

44   near the surface. Since the base model underestimates the wet deposition of nitrate in most

45   regions across the modeling domain except the Pacific Coast, the inclusion of LNO leads to

46   reduction in biases and errors and an increase in correlation coefficients at almost all the

47   NADP/NTN sites. Among the three LNO schemes described in Kang et al. (2019), the hNLDN

48   scheme, which is implemented using hourly observed lightning flash data from National

49   Lightning Detection Network (NLDN), performs best for the ground-level, vertical profiles, and

50   wet deposition comparisons except that for the accumulated wet deposition of nitrate, the

51   mNLDN scheme (the monthly NLDN-based scheme) performed slightly better. However, when

52   observed lightning flash data are not available, the linear regression-based parameterization

53   scheme, pNLDN, provides an improved estimate for nitrate wet deposition compared to the base

54   simulation that does not include LNO.

1. Introduction

The potential importance of nitrogen oxides (NO_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 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 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 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 (Kang and Pickering, 2018). The significant impact of LNO on process-based understanding of surface air quality was earlier reported by Napelenok et al. (2008), which found low-biases in upper tropospheric NO_x in Community Multiscale Air Quality Model (CMAQ) (Byun and Schere, 2006) simulations without LNO emissions made it difficult to constrain ground-level 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 was not included.

LNO 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 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

85   convectively induced updraft velocity (Allen et al., 2000; Allen and Pickering, 2002). More

86   recently, Finney et al. (2014, 2016) adopted a lightning parameterization using upward cloud ice

87   flux at 440hPa (based upon definitions of deep convective clouds in the International Satellite

88   Cloud Climatology Project (Rossow et al., 1996)) and implemented it in the United Kingdom

89   Chemistry and Aerosol model (UKCA). With the availability of lightning flash data from the

90   National Lightning Detection Network (NLDN) (Orville et al., 2002), recent LNO

91   parameterization schemes started to include the observed lightning flash information to constrain

92   LNO in regional Chemical Transport Models (CTMs) (Allen et al., 2012). In Kang et al. (2019),

93   we described the existing LNO parameterization scheme that is based on the monthly NLDN

94   (mNLND) lightning flash data, and an updated scheme using hourly NLDN (hNLDN) lightning

95   flash data in the CMAQ lightning module. In addition, we also developed a scheme based on

96   linear and log-linear regression parameters using multiyear NLDN observed lightning flashes

97   and model predicted convective precipitation rate (pNLDN). The preliminary assessment of

98   these schemes based on total column LNO suggests that all the schemes provide reasonable LNO

99   estimates in time and space, but during summer months, the mNLDN scheme tends to produce

100   the most and the pNLDN scheme the least LNO.

101   The first study on the impact of LNO on surface air quality using CMAQ was conducted

102   by Allen et al. (2012) and followed by Wang et al. (2013) with different ways for parameterizing

103   LNO production and different model configurations. In this study, we present performance

104   evaluations using each of the LNO production schemes (mNLDN, hNLDN, pNLDN) described

105   by Kang et al. (2019) to provide estimates of LNO in CMAQ. In addition to examination of

106   differences in air quality estimates between these schemes, we compare the model predictions to

107   base model estimates without LNO and evaluate the estimates from all of the simulations against

108   surface and airborne observations.

109   Section 2 describes the model configuration, simulation scenarios, analysis methodology,

110   and observational data. Section 3 presents the analysis results and Section 4 presents the

111   conclusions.

112

113

2. Methodology

2.1 The LNO schemes

In air quality models, three steps are involved in generating 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 double-peak vertical distribution algorithm described in Kang et al. (2019), which also provides detailed description and formulation of all the LNO schemes.

2.2 The CMAQ model and simulation configurations

The CMAQ model (Appel et al. 2017) version 5.2 were configured with the CB6 chemical mechanism (Yarwood et al., 2010) and the AERO6 aerosol module (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

5

143   version 3.6.5 of the Sparse Matrix Operator Kernel Emissions (SMOKE;

144   https://www.cmascenter.org/smoke/) processor to create gridded speciated hourly model-ready

145   input emission fields for input to CMAQ. Electric generating unit (EGU) emissions were

146   obtained using data from EGUs equipped with a continuous emission monitoring system

147   (CEMS). Plume rise for point and fire sources were calculated in-line for all simulations (Foley

148   et al., 2010). Biogenic emissions were generated in-line in CMAQ using BEIS versions 3.61

149   (Bash et al., 2016). All the simulations employed the bidirectional (bi-di) ammonia flux option

150   for estimating the air-surface exchange of ammonia.

151   There are four CMAQ simulation scenarios for this study: 1) simulation without LNO

152   (Base), 2) simulation with LNO generated by the scheme based on monthly information from the

153   NLDN (mNLDN), 3) simulation with LNO generated by scheme based on hourly information

154   from the NLDN (hNLDN), and 4) simulation with LNO generated by the scheme parameterizing

155   lightning emissions based on modeled convective activity (pNLDN) as described in detail in

156   Kang et al. (2019). All other model inputs, parameters and settings were the same across the four

157   simulations. The vertical distribution algorithm is the same for all the LNO schemes as also

158   described in Kang et al. (2019).

159

160   **2.3 Observations and analysis techniques**

161   To assess the impact of LNO on ground-level air quality, output from the various CMAQ

162   simulations were paired in space and time with observed data from the EPA's Air Quality

163   System (AQS; https://www.epa.gov/aqs) for hourly O_3 and NO_x. To evaluate the vertical

164   distribution, measurements of trace species from the Deriving Information on Surface Conditions

165   from Column and Vertically Resolved Observations Relevant to Air Quality (DISCOVER-AQ;

166   http://www.nasa.gov/missions/discover-aq) campaign conducted in the Baltimore/Washington

167   region (e.g., Crawford and Pickering, 2014; Anderson et al., 2014; Follette-Cook et al., 2015)

168   were used. During this campaign, the NASA P-3B aircraft measured trace gases including O_3,

169   NO, and NO_2. Vertical profiles were obtained over seven locations – Beltsville (Be), Padonia

170   (Pa), Fairhill (Fa), Aldino (Al), Edgewood (Ed), Essex (Es), and Chesapeake Bay (Cb) from

171   approximately 0.3 to 5 km above ground level during P-3B flights over 14 days in July 2011.

172   During this same period, ozonesonde measurements were taken that extended from ground level

173   through the entire model column at two locations (Beltsville, MD and Edgewood, MD shown in

174 Figure 1). Inclusion of LNO estimates in the CTM simulations also has an important impact on

175 model estimated wet deposition of nitrate. Therefore, assessment was also performed using data

176 from the National Atmospheric Deposition Program's National Trends Network (NADP/NTN,

177 http://ndp.slh.wisc.edu/ntn).

178 Since lightning activity as well as LNO exhibit distinct spatial variations (Kang and

179 Pickering, 2018), analysis was conducted for the model domain over the contiguous United

180 States, and then for each region as shown in Figure 1. Emphasis is placed on two regions,

181 Southeast (SE) and Rocky Mountains (RM), where lightning activity is more prevalent and LNO

182 has the greatest impact on model predictions as shown in Results - increasing model bias in the

183 SE and decreasing bias in the RM. The commonly used statistical metrics, Root Mean Square

184 Error (RMSE), Normalized Mean Error (NME), Mean Bias (MB), Normalized Mean Bias

185 (NMB), and Correlation Coefficient (R), in the model evaluation field as defined in Kang et al.

186 (2005) and Eder et al. (2006) were calculated to assess the basic performance differences among

187 all the model cases for their ground-level air quality predictions.

188
189 **3. Results**

190 **3.1 Ground-level evaluation for O_3 and NO_x**

191 **3.1.1 Statistical performance metrics**

192 Tables 1 and 2 display the statistical model performance metrics for daily maximum 8-hr

193 (DM8HR) O_3 and daily mean NO_x mixing ratios over the domain and each analysis region for all

194 four model cases in July 2011 (Base, mNLDN, hNLDN, and pNLDN). The best performance

195 metrics among the model cases are highlighted in bold. As shown in Table 1, for DM8HR O_3,

196 the Base simulation has the lowest MB and NMB values over the Domain, while hNLDN

197 produced the smallest RMSE and NME values. mNLDN generated the largest values for both

198 error (RMSE and NME) and biases (MB and NMB), followed by pNLDN~~,~~. ~~and~~More

199 ~~importantly,~~ all model cases with LNO exhibit slightly higher correlation coefficients than the

200 Base simulation~~, suggesting the importance of including the contributions of this source for~~

201 ~~improving the spatial and temporal variability in model predictions~~. Additionally, the hNLDN

202 simulation exhibited higher correlation and lower bias and error relative to the measurements

203 indicating the value of higher temporal resolution lightning activity for representing the

204 associated NO_x emissions and their impacts on tropospheric chemistry.

205 Examining the regional results for DM8HR O_3 in Table 1, the statistical measures indicate

206 that in the Northeast (NE), hNLDN outperformed all other model cases with the lowest errors

207 and biases and highest correlation coefficient. In Southeast (SE), the Base performed better with

208 the lowest errors and mean biases, but the correlation coefficient (R) value for hNLDN is slightly

209 higher. Among all the LNO cases, mNLDN produced the worst statistics in this region.

210 Historically, CTMs tend to significantly overestimate surface O_3 in the Southeast US (Lin et al.,

211 2008; Fiore et al., 2009; Brown-Steiner et al., 2015; Canty et al., 2015), and this is partially

212 driven by a likely overestimation of anthropogenic NO_x emissions (Anderson et al., 2014). Thus,

213 even though lightning is known to impact ambient air quality, including this additional NO_x

214 source can worsen biases in model O_3 in some locations and time periods due to other errors in

215 the modeling system. As noted in Table 1, for the SE, the MB values increased by about 1.6 ppb

216 in mNLDN and less than 1 ppb in hNLDN and pNLDN. Nevertheless, the correlation

217 coefficients for mNLDN and pNLDN are almost the same with the Base, and hNLDN was

218 slightly higher (0.77 compared to 0.76). These correlations indicate that even though additional

219 NO_x increases the mean bias, when it is added correctly in time and space, as with the case of

220 hNLDN, the spatial and temporal correlation are slightly improved. In the Upper Midwest (UM),

221 the lowest errors and biases among the model cases are associated with hNLDN, while the worst

222 performance is with mNLDN. In the Lower Midwest (LM), hNLDN performed comparable with

223 the Base, with hNLDN having the highest correlation and lowest mean errors, while the Base has

224 the lowest mean biases. Rocky Mountain (RM) is the only region that shows an underestimation

225 of DM8HR O_3. In this region all the model cases with LNO outperformed the Base case in all the

226 metrics. Among the three model cases with LNO, mNLDN produced the lowest MB and NMB

227 values, while hNLDN had the lowest RMSE and NME, and the highest correlation. In the Pacific

228 Coast (PC) region, lightning activity is generally very low compared to other regions (Kang and

229 Pickering, 2018). All model cases with LNO outperformed the Base case, especially hNLDN

230 which had the lowest mean error and bias and highest correlation among all the cases.

231 Most of the NO_x produced by lightning is distributed in the middle and upper troposphere

232 with only a small portion being distributed close to the surface. As a result, the impact on

233   ground-level NO_x mixing ratios is small. Table 2 shows all the model cases produced similar

234   statistics for the daily mean NO_x mixing ratios at AQS sites across the domain and within all the

235   subregions. Although the changes in model performance are small, the model cases with LNO

236   exhibit similar or slightly better performance than the Base case.

237   **3.1.2 Time series**

238   Figure 2 presents timeseries of regional-mean observed and modeled DM8HR O_3 for the

239   entire domain and the SE and RM regions during July 2011. Over the domain and in SE, all the

240   model cases overestimate the mean DM8HR O_3 mixing ratios on all days with the Base being the

241   closest to the observations. hNLDN is almost the same as the Base with slightly higher values on

242   some days. Among all the cases, mNLDN produced the highest values on almost all days

243   through the month, on the order of 1-2 ppb higher than the Base. In contrast, in the RM region,

244   the Base significantly underestimates DM8HR O_3 mixing rations on all the days during the

245   month, while all model cases with LNO improved model predictions relative to observations in

246   the region. Among the three model cases with LNO, mNLDN produced the lowest bias for all

247   the days, closely followed by hNLDN.

248   Figure 3 displays the average daily mean NO_x mixing ratios at AQS sites over the same

249   regions as in Figure 2. On most of the days in July 2011, over the domain and in the SE, the

250   model overestimate NO_x values, and on almost half of the days, the overestimation is significant

251   (up to 100 %). As noted in Table 2, on average, the overestimation is ~17 % over the domain and

252   ~43 % in SE. However, in RM, the predicted NO_x mixing ratios closely follow the daily

253   observations and on average the modeled and observed magnitude is almost identical (~3 %

254   difference). All the model cases, with or without LNO, produced almost the same mean NO_x

255   mixing ratios at the surface. However, the different cases produce different levels of LNO in the

256   middle and upper troposphere, resulting in differences in O_3 production and transport which

257   impact radiative forcing and also downwind ground-level O_3 levels. We further explore these

258   features in Section 3.2 which presents evaluation of modeled vertical pollutant distributions.

259   **3.1.3 Diurnal variations**

260   Diurnal plots are used to further examine differences in model evaluation for O_3 and

261   NO_x. Figure 4 shows the mean diurnal profiles for hourly O_3 and NO_x over the entire domain,

262   SE, and RM. On a domain mean basis, all model cases overestimate O_3 during the daytime

263   hours, while in the SE, the overestimation spans all the hours. In RM, the model cases

264   significantly underestimate O_3 across all the hours except for a few early morning hours, when

265   the model predicted values are very close to the observations. Among all the model cases, as

266   expected, the most prominent differences occurred during the midday hours when the

267   photochemistry is most active. However, the difference between hNLDN (and mNLDN) and the

268   Base is also significant during the night in the RM region, even though the O_3 levels are low.

269   This may be attributed to NO_x-related nighttime chemistry in part caused by freshly released NO

270   by cloud-to-ground lightning flashes. The diurnal variations of NO_x are similar over the domain

271   and in the regions for all model cases. Appel et al. (2017) reported a significant overestimation of

272   NO_x mixing ratios at AQS sites during nighttime hours and underestimation during daytime

273   hours. The bias pattern is identical for all of the LNO model cases evaluated here (Figure 4).

274   **3.1.4 Spatial variations**

275   Figure 5 shows the impact of the different LNO schemes on model performance for

276   DM8HR O_3 at AQS sites. The spatial maps show the difference in absolute MB between the

277   cases with lightning NO_x emissions and the Base and is calculated as follows. First, the absolute

278   MB was calculated at each site for each case, e.g. $|MB_{[Base-Obs]}|$, then the difference in absolute

279   MB was calculated between model cases, e.g. $|MB_{[hNLDN-Obs]}| - |MB_{[Base-Obs]}|$. The histograms of

280   the differences in absolute MB between model cases in Figure 5 are provided to show the

281   distribution of the change in model performance across space, i.e. the frequency of an

282   improvement in model performance versus a degradation in model performance between cases.

283   As shown in Figure 5, the mNLDN shows increased model bias in the east US and along the

284   California coast, but reduced model bias in the RM. At a majority of the AQS sites, it increases

285   the model bias (only decreases at 26.8 % (346) of the sites). The hNLDN also significantly

286   reduces model bias in the RM with a moderate increase in the SE. Overall, in the hNLDN, the

287   mean bias decreased at 61.2 % (791) of AQS sites. Similar to mNLDN, increases in mean bias

288   are noted at 29.3 % (378) of the AQS sites in the pNLDN simulation. As noted in the histograms,

289   the distribution of the model bias in the pNLDN is much narrower than both mNLDN and

290   hNLDN, eliminating the large bias increases in mNLDN and the significant bias decreases in

291   hNLDN.

3.2 Vertical evaluation for O_3 and NO_x

3.2.1 Ozone-sonde observations

A large source of uncertainty in the specification of LNO 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 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 has an obvious impact (the mean vertical profiles of LNO cases are separable from that of the base case) 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 1 km, but overestimates values closer to the surface. When LNO 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 about 6 km, but above that altitude the mNLDN ozone mixing ratios were higher. All the model cases with LNO 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 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

322 noticeable (the mean vertical profiles of LNO cases are separable from that of the base case)

323 lightning impacts, to evaluate the model simulations. Figure 7 shows measured O_3 mixing ratios

324 overlaid on the modeled vertical time-section for 1030 – 1730 UTC. The color-filled circles

325 represent measured O_3 mixing ratios averaged over 60 seconds and the background is the model

326 estimated vertical profiles from the grid cells containing the P-3B flight path for that hour and

327 location. As indicated in the Base case (Figure 7a), the model tends to overestimate O_3 mixing

328 ratios from the surface to about 2 km, but underestimate at altitudes above 2 km. The hNLDN

329 reduced the overestimation below 2km, e.g. fewer grid cells with mixing ratios above 90ppb

330 (shown in red). The other two cases (mNLDN, pNLDN) did not produce the same improvement

331 near the surface. The hNLDN also decreases the underestimation aloft compared to the Base case

332 with O_3 mixing ratios in the 55-65 ppb range (light blue colors), better matching the measured

333 values. This decrease in underestimation aloft is also seen in the mNLDN case, but to a lesser

334 degree while the pNLDN case shows only slight improvement aloft over the Base simulation.

335 To further differentiate the three LNO model cases, Figures 8-10 show the difference in

336 the time-sections between each of the model cases with LNO and the Base for NO, NO_x, and O_3

337 from all the model layers along the P-3B flight path on July 28. As seen in Figure 8, the hNLDN

338 scheme injected most NO above 5 km with a peak between 13-14 km and only a small amount

339 near the surface. After release into the atmosphere, NO is quickly converted into NO_2 in the

340 presence of O_3, and these collectively result in the NO_x vertical time-section (local production

341 plus transport) shown in the middle panel of Figure 8. NO_x is further mixed down through the

342 time-section and more persistent along the flight path near the surface than is NO. As a result,

343 significant O_3 is produced above 3 km and the maximum O_3 difference appears between 9 and 14

344 km during the early afternoon hours (from 13:30 to 17:30). However, from surface to about 2

345 km, O_3 is reduced consistently across the entire period, and this is the result of O_3 titration by NO

346 from cloud-to-ground lightning flashes that must have been transported to this layer by storm

347 downdrafts. Since O_3 is significantly underestimated above 3 km and overestimated near the

348 surface by the Base model, the inclusion of LNO greatly improved the model's performance

349 under both conditions.

350 Comparison of Figure 9 (mNLDN) with Figure 8 (hNLDN) reveals that the time-sections

351 of NO and NO_x are similar above 5 km but dramatically different near the surface. The near-

352 surface increase in ambient NO noted in the hNLDN is absent in mNLDN, and in fact there are

353 some small decreases in NO, although the reason for this is unclear. The increase in O_3 aloft in

354 the mNLDN case is similar to that seen in the hNLDN case. However, the near-surface reduction

355 in O_3 is almost absent. In the pNLDN case (Figure 10), NO mixing ratios are much less than

356 those in hNLDN and mNLDN in the upper layers as a result of less column NO being generated

357 by the linear parameterization. The resulting NO_x time-section is also smoothed. The pNLDN

358 time-sections for NO, NO_x and O_3 near the surface are similar to the mNLDN case with no

359 change or small decreases compared to the Base case. O_3 mixing ratios increase by more than 30

360 ppb during the afternoon hours between $10 - 13$ km in the pNLDN case, however the increase is

361 not as intense and widespread as the other cases. In summary, the hNLDN scheme produces

362 estimates that are more consistent with measurements at the surface and aloft, compared to the

363 other simulations, reflecting the advantage of using the spatially and temporally-resolved

364 observed lightning flash data. The model performance improvement for simulated O_3

365 distributions also suggests robustness in the vertical distribution scheme when LNO is generated

366 at the right time and location.

367   To corroborate the above time-section distributions of NO, NO_x, and O_3 in the lightning

368 cases, the lightning NO emissions are traced back on July 28 for each case. It is found that in all

369 cases, the lightning NO was injected approximately 200 km upwind (north-west) of the flight

370 path. The hNLDN case captured two injections: one occurred during the morning hours (5:00 to

371 7:00 am) and the other happened during the afternoon hours (after 2:30 pm). Both mNLDN and

372 pNLND captured the afternoon lightning event at the later time (after 3:30 pm for mNLDN and

373 after 4:30 pm for pNLDN) with varying intensity, but neither captured the morning lightning

374 event, which explains why the increase of NO and NO_x in the hNLDN case (Figure 8) did not

375 occur in the mNLDN and pNLDN cases (Figures 9 and 10). Also note that the significant

376 increase of NO during the time period from 11:00 to 13:00 occurred about 5 hours after the

377 lightning NO was injected at about 200 km upwind in the hNLDN case.

378   To expand on the evaluation in Figures 7-10 which focused on measurements from July

379 28, 2011, we retrieved all the P-3B measurements on days with noticeable lightning impact (July

380 21, 22, 28, and 29). The 3-D paired observation-model data were grouped together by spiral site

381 and the mean biases (model – observation) were plotted in Figure 11 (a and b) for O_3 and NO,

382   respectively. The boxplots for O_3 in Figure 11a suggests that the Base exhibited larger bias with

383   greater spread (i.e. larger interquartile range) than other model cases incorporating LNO at most

384   of the locations where aircraft spirals were conducted. At all locations except Aldino, the lowest

385   mean biases in simulated NO and O_3 are noted in the hNLDN simulation.

386   **3.3 Deposition evaluation for nitrate**

387   In addition to contributing to tropospheric O_3 formation, NO_x oxidation also leads to gaseous

388   nitric acid and particulate-nitrate which are eventually removed from the atmosphere by dry and

389   wet deposition of nitrate (NO_3^-). As a result, inclusion of NO_x from lightning also plays an

390   important role in nitrogen deposition modeling. To assess the impacts of incorporating LNO

391   emissions on simulated oxidized nitrogen deposition, we compared model estimated amounts of

392   precipitation from NTN network (http://nadp.slh.wisc.edu/ntn/) and wet deposition of NO_3^- with

393   measurements from the NADP network (http://nadp.slh.wisc.edu/). During summer months in

394   2011 (June -August) the WRF model generally reproduces the observed precipitation with a

395   slight underestimate in the east, but the Base model simulation tends to underestimate wet

396   deposition of NO_3^- across the domain, with the greatest underestimation in the SE and UM (See

397   Table 3 and Figure 12). All three LNO simulations increase wet deposition amounts of NO_3^- and

398   decrease model bias in all regions. The bottom panel of Figure 12 shows that the mNLDN

399   simulation resulted in the largest increase over the base model estimates. The NMB is reduced

400   from -35 % in the Base to -15 % in mNLDN across the domain and from -32 % to -2 % in the

401   SE. The hNLDN shows very similar model performance to the mNLDN case. In contrast, the

402   wet deposition NO_3^- estimates from the pNLDN case are only slightly higher than the Base case,

403   and as a result the evaluation statistics for pNLND are very similar to the Base statistics. As

404   discussed earlier, the mNLDN tends to produce the most LNO among the three LNO schemes,

405   thus it results in the smallest errors in terms of wet deposition of NO_3^- when compared to the

406   Base simulation that significantly underestimated NO_3^- wet deposition. It should be noted that in

407   addition to the LNO contributions, errors in modeled precipitation amounts and patterns also

408   likely influence the underestimation of NO_3^- wet deposition.

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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 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. 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, 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 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 schemes also enhanced the accumulated wet deposition of NO_3^-, that was significantly underestimated by the base model without LNO 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 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 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 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

440   involving air quality forecasting and future projections when observed lightning information
441   is not available.

442

Code and data availability

444   CMAQ model documentation and released versions of the source code, including all model code
445   used in his study, are available at https://github.com/USEPA/CMAQ/tree/5.2
446   . The data processing and analysis scripts are available upon request. The WRF model is
447   available for download through the WRF website
448   (http://www2.mmm.ucar.edu/wrf/users/wrfv3.8/updates-3.8.html).
449   The raw lightning flash observation data used are not available to the public but can be
450   purchased through Vaisala Inc. (https:// www.vaisala.com/en/products/systems/lightning-
451   detection). The lightning data obtained from Vaisala Inc. is the cloud-to-ground lightning flashes over
452   the contiguous United States. The immediate data behind the tables and figures are available from
453   https://zenodo.org/record/3360744 (Kang and Foley, 2019). Additional input/output data for
454   CMAQ model utilized for this analysis are available upon request as well.
455
456
457   **Disclaimer:** The views expressed in this paper are those of the authors and do not necessarily
458   represent the views or policies of the U.S. EPA.

459

Author Contribution

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

463   **Kristen Foley**: data analysis and manuscript writing.

464   **Rohit Mathur**: manuscript editing.

465   **Shawn Roselle**: manuscript editing.

466   **Kenneth Pickering:** manuscript editing.

467   **Dale Allen:** manuscript editing.

468

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Figure Captions:

Figure 1. Analysis regions and ozonesonde locations during the 2011 DISCOVER-AQ field study.

Figure 2. Timeseries of regional-mean daily maximum 8-hr O_3 comparing observations (AQS) and CMAQ model predictions using the LNO_x schemes to Base simulation for the domain (a), SE (b), and RM (c) in July 2011. The numbers in the parentheses following the region names are the number of AQS sites.

Figure 3. Timeseries of daily mean NO_X over the domain (a), SE (b), and RM (c) in July 2011. The numbers in the parentheses following the region names are the number of AQS sites.

Figure 4. Diurnal profiles for hourly O_3 and NO_x over the domain (a,d), SE (b,e), and RM (c,f) in July 2011.

Figure 5. Spatial maps of the mean bias of DM8HR O_3 (model – observation) differences between model case with LNO_X and the Base as well as the corresponding histograms indicating the number of sites with decreased mean bias for each pair of model cases in July 2011.

Figure 6. Vertical profiles of O_3 mixing ratios from ozonesonde measurements and model simulations at Beltsville, MD (a) and Edgewood, MD (b) on the days when lightning NO produced significant impact on O_3 during the Discover-AQ field study in July 2011.

Figure 7. Overlay of P3B observed O_3 (1-minute mean values) over the corresponding vertical cross sections of simulated values extracted at the flying locations on July 28, 2018, (a) Base, (b) hNLDN (c) mNLDN, and (d) pNLDN. The letters marked at the bottom of the plots are P3B spiral sites, Be: Beltsville, Pa: Padonia, Fa: Fairhill, Al: Aldino, Ed: Edgewood, Es: Essex.

Figure 8. The vertical-time difference between hNLDN and Base during the P3B flight period on July 28, 2011 for (a) NO, (b) NO_X, and (c) O_3.

714   Figure 9. The vertical-time difference between mNLDN and Base during the P3B flight period

715   on July 28, 2011 for (a) NO, (b) NO_X, and (c) O_3.

716   Figure 10. The vertical-time difference between pNLDN and Base during the P3B flight period

717   on July 28, 2011 for (a) NO, (b) NO_X, and (c) O_3.

718   Figure 11. Bias (model – observation) distributions of O_3 (a) and NO (b) at each P3B spiral site

719   on July 21, 22, 28, and 29, 2011. Be: Beltsville, Pa: Padonia, Fa: Fairhill, Al: Aldino, Ed:

720   Edgewood, Es: Essex, Cb: Chesapeake Bay

721   Figure 12. The top row shows precipitation estimates from WRF (left), the bias in the WRF

722   predicted precipitation at NTN locations (middle), and the corresponding scatter plots

723   (right). The middle row shows wet deposition (Dep) of nitrate estimates from the Base

724   simulation (left), the bias in the Base model estimates of wet deposition of NO_3^- at

725   NADP/NTN locations (middle), and the corresponding scatter plots (right). The bottom

726   row shows the difference in the LNO_X sensitivity simulations and the Base case estimates

727   of wet deposition of NO_3^-: mNLDN – Base (left); hNLDN – Base (middle), and pNLDN

728   – Base (right). All maps are based on accumulated values (precipitation or wet

729   deposition) during June – August 2011. Precipitation totals are in cm and wet deposition

730   totals are in kg/ha.