

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

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## Abstract

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This study assesses the impact of the lightning NO (LNO) production schemes in the Community Multiscale Air Quality (CMAQ) model on ground-level air quality as well as aloft atmospheric chemistry through detailed evaluation of model predictions of nitrogen oxides ( $\text{NO}_x$ ) and ozone ( $\text{O}_3$ ) with corresponding observations for the U.S. For ground-level evaluations, hourly  $\text{O}_3$  and  $\text{NO}_x$  from the US EPA's AQS monitoring network are used to assess the impact of different LNO 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), the impact of LNO on surface  $\text{O}_3$  varies from region to region depending on the base model conditions. Overall statistics suggest that for regions where surface  $\text{O}_3$  mixing ratios are already overestimated, the incorporation of additional NO from lightning generally increased model overestimation of mean daily maximum 8-hr (DM8HR)  $\text{O}_3$  by 1-2 ppb. In regions where surface  $\text{O}_3$  is underestimated by the base model, LNO can significantly reduce the underestimation and bring model predictions close to observations. Analysis of vertical profiles reveals that LNO can significantly improve the vertical structure of modeled  $\text{O}_3$  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 leads to reduction in biases and errors and an increase in correlation coefficients at almost all the NADP/NTN sites. Among the three LNO 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 compared to the base simulation that does not include LNO.

55 **1. Introduction**

56 The potential importance of nitrogen oxides ( $\text{NO}_x$ ;  $\text{NO}_x = \text{NO} + \text{NO}_2$ ) produced by lightning  
57 (LNO<sub>x</sub>; due to the equilibrium coexistence of NO and NO<sub>2</sub> in the atmosphere, in the literature it  
58 is often collectively referred to as LNO<sub>x</sub>. However, the immediate release of lightning flashes is  
59 just NO, and the schemes in Kang et al., 2019 also generate NO emissions only, so in this paper  
60 it is primarily referred to as LNO) on regional air quality was recognized more than two decades  
61 ago (e.g. Novak and Pierce, 1993), but LNO emissions have only been added to regional  
62 chemistry and transport models during the last decade (e.g. Allen et al., 2012; Kaynak et al.,  
63 2008; Koshak et al., 2014; Smith and Mueller, 2010; Koo et al., 2010) owing in part to the  
64 limited understanding of this NO<sub>x</sub> source (Schumann and Huntrieser, 2007; Murray, 2016;  
65 Pickering et al, 2016). As a result of efforts to reduce anthropogenic NO<sub>x</sub> emissions in recent  
66 decades (Simon et al., 2015; <https://gispub.epa.gov/air/trendsreport/2018>), it is expected that the  
67 relative contribution of LNO to the tropospheric NO<sub>x</sub> burden and its subsequent impacts on  
68 atmospheric chemistry as one of the key precursors for ozone (O<sub>3</sub>), hydroxyl radical (OH),  
69 nitrate, and other species will increase in the United States and other developed countries (Kang  
70 and Pickering, 2018). The significant impact of LNO on process-based understanding of surface  
71 air quality was earlier reported by Napelenok et al. (2008), which found low-biases in upper  
72 tropospheric NO<sub>x</sub> in Community Multiscale Air Quality Model (CMAQ) (Byun and Schere,  
73 2006) simulations without LNO emissions made it difficult to constrain ground-level NO<sub>x</sub>  
74 emissions using inverse methods and Scanning Imaging Absorption Spectrometer for  
75 Atmospheric Cartography (SCIAMACHY) NO<sub>2</sub> retrievals (Bovensmann et al., 1999; Sioris et  
76 al., 2004; Richter et al., 2005). Appel et al. (2011) and Allen et al. (2012) reported that NO<sub>3</sub><sup>-</sup> wet  
77 deposition at National Atmospheric Deposition Program (NADP) sites was underestimated by a  
78 factor of two when LNO was not included.

79 LNO production and distribution were parameterized initially in global models (e.g.  
80 Stockwell et al., 1999; Labrador et al., 2005) relying on the work of Price and Rind (1992) and  
81 Price et al. (1997) in that lightning flash frequency was parameterized as a function of the  
82 maximum cloud-top-height. Other approaches for LNO parameterization include a combination  
83 of latent heat release and cloud-top-height (Flatoy and Hov, 1997), convective precipitation rate  
84 (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 (mNLDN) 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.

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## 114 2. Methodology

### 115 2.1 The LNO schemes

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

### 128 2.2 The CMAQ model and simulation configurations

129 The CMAQ model (Appel et al. 2017) version 5.2 were configured with the CB6  
130 chemical mechanism (Yarwood et al., 2010) and the AERO6 aerosol module -(Nolte et al.,  
131 2015). The meteorological inputs were provided by the Weather Research and Forecasting  
132 (WRF) model version 3.8 and the model-ready meteorological input files were created using  
133 version 4.2 of the meteorology–chemistry interface processor (MCIP; Otte and Pleim, 2010).

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

141 Emission input data were based on the 2011 National Emissions Inventory  
142 (<https://www.epa.gov/air-emissions-inventories>). The raw emission files were processed using

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).

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### 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<sub>3</sub> and NO<sub>x</sub>. 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<sub>3</sub>,  
169 NO, and NO<sub>2</sub>. 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.

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### **3. Results**

#### **3.1 Ground-level evaluation for O<sub>3</sub> and NO<sub>x</sub>**

##### **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<sub>3</sub> and daily mean NO<sub>x</sub> 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<sub>3</sub>,  
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. More importantly, all  
199 model cases with LNO exhibit slightly higher correlation coefficients than the Base simulation,  
200 suggesting the importance of including the contributions of this source for improving the spatial  
201 and temporal variability in model predictions. Additionally, the hNLDN simulation exhibited  
202 higher correlation and lower bias and error relative to the measurements indicating the value of

203 higher temporal resolution lightning activity for representing the associated NO<sub>x</sub> emissions and  
204 their impacts on tropospheric chemistry.

205 Examining the regional results for DM8HR O<sub>3</sub> 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<sub>3</sub> 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<sub>x</sub> emissions (Anderson et al., 2014). Thus,  
213 even though lightning is known to impact ambient air quality, including this additional NO<sub>x</sub>  
214 source can worsen biases in model O<sub>3</sub> 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<sub>x</sub> 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<sub>3</sub>. 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<sub>x</sub> 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<sub>x</sub> mixing ratios is small. Table 2 shows all the model cases produced similar  
234 statistics for the daily mean NO<sub>x</sub> 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<sub>3</sub> 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<sub>3</sub> 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<sub>3</sub> mixing ratios 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<sub>x</sub> 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<sub>x</sub> 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<sub>x</sub> 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<sub>x</sub>  
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<sub>3</sub> production and transport which  
257 impact radiative forcing and also downwind ground-level O<sub>3</sub> 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<sub>3</sub> and  
261 NO<sub>x</sub>. Figure 4 shows the mean diurnal profiles for hourly O<sub>3</sub> and NO<sub>x</sub> over the entire domain,

262 SE, and RM. On a domain mean basis, all model cases overestimate O<sub>3</sub> during the daytime  
263 hours, while in the SE, the overestimation spans all the hours. In RM, the model cases  
264 significantly underestimate O<sub>3</sub> 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<sub>3</sub> levels are low.  
269 This may be attributed to NO<sub>x</sub>-related nighttime chemistry in part caused by freshly released NO  
270 by cloud-to-ground lightning flashes. The diurnal variations of NO<sub>x</sub> 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<sub>x</sub> 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<sub>3</sub> at AQS sites. The spatial maps show the difference in absolute MB between the  
277 cases with lightning NO<sub>x</sub> 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.

## 292 **3.2 Vertical evaluation for O<sub>3</sub> and NO<sub>x</sub>**

### 293 **3.2.1 Ozone-sonde observations**

294 A large source of uncertainty in the specification of LNO is its vertical allocation, which can  
295 impact the model's ability to accurately represent the variability in both chemistry and transport.  
296 To further assess the impact of the vertical LNO specification on model results, we compared  
297 vertical profiles of simulated model O<sub>3</sub> with extensive ozonesonde measurements available  
298 during the study period. Figure 6 presents the vertical profiles for O<sub>3</sub> sonde measurements and  
299 paired model estimates of all model cases at Beltsville, MD and Edgewood, MD. At each  
300 location, observations from multiple days are available (one or two soundings per day) during  
301 the 2011 DISCOVER-AQ campaign in July 2011. The model evaluation was limited to days  
302 where the inclusion of LNO has an obvious impact (the mean vertical profiles of LNO cases are  
303 separable from that of the base case) on the model estimates (July 21, 22, 28 and 29 at Beltsville,  
304 and July 21, 22, 28, 29, and 30 at Edgewood). We paired the observed data with model estimates  
305 in time and space and averaged the model and observed values at each model layer. Only data  
306 below 12 km altitude are plotted in Figure 6 to exclude possible influence of stratospheric air on  
307 O<sub>3</sub>. As can be seen in Figure 6, at both locations the Base case underestimates O<sub>3</sub> mixing ratios  
308 above about 1 km, but overestimates values closer to the surface. When LNO is included in the  
309 simulations, the predicted O<sub>3</sub> mixing ratios increase relative to the Base case starting around  
310 2km, with greater divergence from the Base case at higher altitudes. The two model cases,  
311 hNLDN and mNLDN, produced similar O<sub>3</sub> levels from the surface to about 6 km, but above that  
312 altitude the mNLDN ozone mixing ratios were higher. All the model cases with LNO performed  
313 much better aloft than the Base case. Near the surface, all the model cases overestimated O<sub>3</sub>,  
314 however hNLDN had smaller bias than the other simulations. This may be attributed to the fact  
315 that only hNLDN used the observed lightning flash data directly, and as a result, LNO was  
316 estimated more accurately in time and space. This improvement in model bias at the surface is  
317 further investigated in the next section using evaluation against P-3B measurements.

### 318 **3.2.2 P-3B measurement**

319 Extensive measurements of lower tropospheric chemical composition distributions over  
320 the Northeastern U.S. are available from instruments onboard the P-3B aircraft on 14 days of the  
321 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<sub>3</sub> mixing ratios  
324 overlaid on the modeled vertical time-section for 1030 – 1730 UTC. The color-filled circles  
325 represent measured O<sub>3</sub> 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<sub>3</sub> 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<sub>3</sub> 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<sub>x</sub>, and O<sub>3</sub>  
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<sub>2</sub> in the  
340 presence of O<sub>3</sub>, and these collectively result in the NO<sub>x</sub> vertical time-section (local production  
341 plus transport) shown in the middle panel of Figure 8. NO<sub>x</sub> 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<sub>3</sub> is produced above 3 km and the maximum O<sub>3</sub> 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<sub>3</sub> is reduced consistently across the entire period, and this is the result of O<sub>3</sub> titration by NO  
346 from cloud-to-ground lightning flashes that must have been transported to this layer by storm  
347 downdrafts. Since O<sub>3</sub> 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<sub>x</sub> 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<sub>3</sub> aloft in  
354 the mNLDN case is similar to that seen in the hNLDN case. However, the near-surface reduction  
355 in O<sub>3</sub> 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<sub>x</sub> time-section is also smoothed. The pNLDN  
358 time-sections for NO, NO<sub>x</sub> and O<sub>3</sub> near the surface are similar to the mNLDN case with no  
359 change or small decreases compared to the Base case. O<sub>3</sub> 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<sub>3</sub>  
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<sub>x</sub>, and O<sub>3</sub> 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<sub>x</sub> 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<sub>3</sub> and NO,

382 respectively. The boxplots for O<sub>3</sub> 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<sub>3</sub> are noted in the hNLDN simulation.

### 386 **3.3 Deposition evaluation for nitrate**

387 In addition to contributing to tropospheric O<sub>3</sub> formation, NO<sub>x</sub> 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<sub>3</sub><sup>-</sup>). As a result, inclusion of NO<sub>x</sub> 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<sub>3</sub><sup>-</sup> 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<sub>3</sub><sup>-</sup> 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<sub>3</sub><sup>-</sup> 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<sub>3</sub><sup>-</sup> 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<sub>3</sub><sup>-</sup> when compared to the  
406 Base simulation that significantly underestimated NO<sub>3</sub><sup>-</sup> 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<sub>3</sub><sup>-</sup> wet deposition.

409  
410  
411

#### 412 4. Conclusions

413 A detailed evaluation of lightning NO<sub>x</sub> emission estimation parameterizations available in  
414 the CMAQ modeling system was performed through comparisons of model simulation  
415 results with surface and aloft air quality measurements.

416 Our analysis indicates that incorporation of LNO emissions enhanced O<sub>3</sub> production in  
417 the middle and upper troposphere, where O<sub>3</sub> mixing ratios were often significantly  
418 underestimated without the representation of LNO. Though the impact on surface O<sub>3</sub> varies  
419 from region to region and is also dependent on the accuracy of the NO<sub>x</sub> emissions from other  
420 sources, the inclusion of LNO, when it is injected at the appropriate time and location, can  
421 improve the model estimates. In regions where the base model estimates of O<sub>3</sub> were biased  
422 high, the inclusion of LNO further increased the model bias; and a systematic increase is  
423 noted in the correlation with measurements, suggesting that emissions from other sources  
424 likely drive the overestimation. Identifying how errors in emissions inputs from different  
425 sources interact with errors in meteorological modeling of mixing and transport, remains a  
426 challenging but critical task. Likewise, all the LNO schemes also enhanced the accumulated  
427 wet deposition of NO<sub>3</sub><sup>-</sup>, that was significantly underestimated by the base model without  
428 LNO throughout the modeling domain except the Pacific Coast.

429 Uncertainty remains in modeling the magnitude and spatial, temporal and vertical  
430 distribution of lightning produced NO<sub>x</sub>. LNO schemes are built on numerous assumptions  
431 and all current schemes also depend on the skill of the upstream meteorological models in  
432 describing convective activity. Nevertheless, these schemes reflect our best understanding  
433 and knowledge at the time when the schemes were implemented. The use of hourly  
434 information on lightning activity yielded LNO emissions that generally improved model  
435 performance for ambient O<sub>3</sub> and NO<sub>x</sub> as well as oxidized nitrogen wet deposition amounts.  
436 As more high-quality data from both ground and satellite measurements become available,  
437 the performance of the LNO schemes will continue to improve.

438 Since the pNLDN scheme was developed using historical data correlating lightning  
439 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

#### 443 **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-](https://www.vaisala.com/en/products/systems/lightning-detection)  
451 [detection](https://www.vaisala.com/en/products/systems/lightning-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.

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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

#### 460 **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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472



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686 **Figure Captions:**

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

690 Figure 2. Timeseries of regional-mean daily maximum 8-hr O<sub>3</sub> comparing observations (AQS)  
691 and CMAQ model predictions using the LNO<sub>x</sub> schemes to Base simulation for the  
692 domain (a), SE (b), and RM (c) in July 2011. The numbers in the parentheses following  
693 the region names are the number of AQS sites.

694

695 Figure 3. Timeseries of daily mean NO<sub>x</sub> over the domain (a), SE (b), and RM (c) in July 2011.  
696 The numbers in the parentheses following the region names are the number of AQS sites.

697

698 Figure 4. Diurnal profiles for hourly O<sub>3</sub> and NO<sub>x</sub> over the domain (a,d), SE (b,e), and RM (c,f) in  
699 July 2011.

700 Figure 5. Spatial maps of the mean bias of DM8HR O<sub>3</sub> (model – observation) differences  
701 between model case with LNO<sub>x</sub> and the Base as well as the corresponding histograms  
702 indicating the number of sites with decreased mean bias for each pair of model cases in  
703 July 2011.

704 Figure 6. Vertical profiles of O<sub>3</sub> mixing ratios from ozonesonde measurements and model  
705 simulations at Beltsville, MD (a) and Edgewood, MD (b) on the days when lightning NO  
706 produced significant impact on O<sub>3</sub> during the Discover-AQ field study in July 2011.

707 Figure 7. Overlay of P3B observed O<sub>3</sub> (1-minute mean values) over the corresponding vertical  
708 cross sections of simulated values extracted at the flying locations on July 28, 2018, (a)  
709 Base, (b) hNLDN (c) mNLDN, and (d) pNLDN. The letters marked at the bottom of the  
710 plots are P3B spiral sites, Be: Beltsville, Pa: Padonia, Fa: Fairhill, Al: Aldino, Ed:  
711 Edgewood, Es: Essex.

712 Figure 8. The vertical-time difference between hNLDN and Base during the P3B flight period on  
713 July 28, 2011 for (a) NO, (b) NO<sub>x</sub>, and (c) O<sub>3</sub>.

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<sub>x</sub>, and (c) O<sub>3</sub>.

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<sub>x</sub>, and (c) O<sub>3</sub>.

718 Figure 11. Bias (model – observation) distributions of O<sub>3</sub> (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<sub>3</sub><sup>-</sup> at  
725 NADP/NTN locations (middle), and the corresponding scatter plots (right). The bottom  
726 row shows the difference in the LNO<sub>x</sub> sensitivity simulations and the Base case estimates  
727 of wet deposition of NO<sub>3</sub><sup>-</sup>: 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.