As one of the largest natural sources of nitrogen oxides, it is estimated that lightning-induced NOx (LNOx) contributes up to 15 percent of the total global NOx emissions budget. Following on from the focus of last month’s topic, Advances in Air Quality Modeling, this article considers the impact of LNOx on air quality through robust LNOx production and distribution schemes in air quality models.
Ground-level ozone ($O_3$), predominantly formed from photochemical reactions in the atmosphere, responds to varying precursor emissions, meteorology, and climate. To protect human health and welfare, the National Ambient Air Quality Standards (NAAQS) for $O_3$ (https://www.epa.gov/ozone-pollution/table-historical-ozone-national-ambient-air-quality-standards-naaqs) have been tightened through the years. In response to the tightened $O_3$ standards, extensive control measures for nitrogen oxides (NOx), one of the crucial precursors in $O_3$ formation, have been implemented across the United States, and they have led to significant reduction in anthropogenic NOx emissions in the past two decades.\(^1\)

Assessments on $O_3$ trends\(^5\) have suggested varying $O_3$ responses to NOx reductions during different seasons and in different regions in United States. Though $O_3$ mixing ratios have been generally decreasing with the emission reductions, summertime $O_3$ in the Intermountain West regions during pollution episodes occasionally exceeds the current 70 parts per billion by volume (ppbv) NAAQS standard with little overall trend despite stringent precursor emission controls. The researchers have attributed these summertime anomalies to the increasing Asian emissions and more frequent occurrences of hot extremes, but one important component, lightning-induced NOx (LNOx), is missing in their assessments. As one of the largest natural NOx sources, it is estimated that LNOx contributes approximately 10 to 15 percent of the total global NOx emissions budget.\(^4\) Lightning activity and the associated distribution of LNOx exhibits strong spatial and temporal variations.\(^5\) To accurately assess the impact of LNOx on air quality, the LNOx contributions to the total NOx emissions need to be quantified in space and time, which entails robust LNOx production and distribution schemes in air quality models.

In the Community Multiscale Air Quality (CMAQ)\(^6\) model, for retrospective simulations, we have implemented a LNOx production scheme based on hourly gridded cloud-to-ground lightning flashes from the National Lightning Detection Network (NLDN)\(^7\) and satellite-based climatological intracloud to cloud-to-ground flash ratios\(^8\) to estimate gridded hourly total LNOx across the contiguous United States. However, the relative impact of LNOx on near-surface $O_3$ depends not only on lightning activity, but also on meteorology and NOx emissions from other sources, such as anthropogenic NOx and soil nitric oxide (NO) emissions.

Using the 2011 National Emissions Inventory (NEI) for anthropogenic NOx emissions and soil NO emissions estimated using CMAQ inline biogenic emission model, we quantify the relative contributions of LNOx to the total NOx emissions budget in time and space for April to September 2011 over the contiguous United States. Model simulations with and without LNOx were assessed against measurements from the U.S. Environmental Protection Agency's Air Quality System (AQS) (https://www.epa.gov/aqs) and ozone-sonde data collected from the Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality (DISCOVER-AQ) 2011 campaign (http://www.nasa.gov/).

LNOx and Its Relative Contributions to Total NOx

Figure 1 shows the spatial distribution of NOx from anthropogenic sources: (a) soil NO; (b) LNOx generated by CMAQ using NLDN lightning flash data; and (c) the relative contribution of LNOx to total NOx ($a+b+c$) during July 2011. Figure 1d indicates that the LNOx ratios are largest over the Rocky Mountains area (see Figure 2a) followed by the Southeast region; in some limited areas, the LNOx ratios can reach more than 90 percent of the total NOx emissions, suggesting that LNOx is the primary NOx emissions over these areas in that specific month.

To help identify the regional and monthly differences in LNOx contributions, Figure 2b presents the monthly LNOx ratios from April to September 2011 over the entire domain and for several sub regions. Over the contiguous United States, the contribution from LNOx emissions to the total NOx emissions ranges from 10 percent (September) to 22 percent (July).

Averaged over the six-month period, the Southeast region has the largest LNOx ratios (20%) followed by the Rocky Mountains area (16%). During July and August, the largest LNOx ratios are observed for the Rocky Mountains region (30% and 28%, respectively). LNOx ratios over Pacific Coast

One important component, lightning-induced NOx (LNOx), has been missing from most assessments of high ozone pollution episodes.
region are the smallest, contributing less than 3 percent across all months. Daily LNOx contributions for the South-east and Rocky Mountains regions over six-month period are presented in Figure 3. In the Southeast region from June through August, LNOx is frequently a significant contribution to total NOx on a daily basis; the same is true for the Rocky Mountains region (see Figure 3b), but the time shifted later from the middle of June to the middle of September.

Figure 1. Total monthly emissions (in 10^6 moles per grid cell 12 km x 12 km, over 144-km² area) and the LNOx ratios during July, 2011: (a) anthropogenic NOx, (b) soil NO, (c) LNOx, and (d) the ratio of LNOx to total NOx emissions.

Figure 2. (a) Regions and (b) the monthly ratio of LNOx to the total NOx emissions over the domain and each of the regions.
Impact on Air Quality
To evaluate the impact of LN O x on air quality, simulations with LN O x (U.S. National Lightning Detection Network [NLDN] data) and without LN O x (Base) using CMAQv5.2 over 12 km contiguous United States domain were performed. Figure 4 shows the gridded daily lightning flash rate from NLDN and the corresponding changes (NLDN–Base) of the maximum surface hourly O 3 mixing ratios on July 11, 2011. The maximum change of hourly O 3 mixing ratio ranges mostly from 6 ppbv to 30 ppbv in the regions of lightning flashes.

Mean Bias (MB = model–observed) between modeled and observed daily Maximum 8-hr O 3 (DM8HRO3) mixing ratios from Air Quality System (AQS) sites located in the Southeast and Rocky Mountains regions are calculated, and the mean differences of mean bias (MBDIFF = MBNLDN – MBBASE) between the NLDN and the Base are presented in Figure 5. Although LN O x to total NO x ratios are the largest in both the Southeast and Rocky Mountains regions among all the sub regions, the impact on surface O 3 is very different. In Southeast region, DM8HRO3 is overestimated in the Base compared to observations, which becomes worse (most often by only ~1 ppbv) with the addition of extra LN O x emissions in the NLDN, resulting in an increase in mean bias for most days. The opposite is true for the Rocky Mountains region as DM8HRO3 is underestimated in the Base, so the addition of LN O x in the NLDN results in a reduction in mean bias compared to the Base.

To examine the impact of LN O x on vertical O 3 profiles, we utilized ozone-sonde data at Beltsville and Edgewood, Maryland, measured during the 2011 DISCOVER-AQ campaign on days when significant lightning impact was observed in the model simulations at each location. We paired the observed ozone-sonde data with model predictions in time and space and take the average over all the ozone-sondes (one or two measurements per day) on the selected days at each location. As shown in Figure 6, the Base significantly underestimates O 3 aloft, but overestimates O 3 near the surface, while with the addition of LN O x in the NLDN reduces both the under-estimation aloft and the near-surface overestimation. And while the bulk statistics don’t reveal the benefit in model performance from including LN O x emissions over the regions such as Southeast, where surface O 3 mixing ratios are already over predicted by the Base model, the added value is apparent from the improved vertical structure of O 3, at least for the two locations presented here. LN O x emissions represent a potentially significant contribution to the total NO x emissions budget across much of the United States during the warmer months, and therefore it is important to include LN O x in regional model simulations to improve the accuracy of air quality predictions.

Summary
The CMAQ model (Version 5.2) contains a lightning NO x
Figure 5. Daily mean bias difference (NLDN-Base) for the Southeast and Rocky Mountains regions at the AQS monitoring sites (the number in the parentheses following the region names are the number of AQS sites).

Figure 6. Mean vertical \(O_3\) profiles from ozone-sonde observations and model simulations at Beltsville (a) and Edgewood (b) on days when significant lightning impact was observed near each location.
Lightning NOx Emissions by Daiwen Kang and Kenneth Pickering

An algorithm that uses observed lightning flashes along with an assumed NOx production per flash and a scheme to distribute the NOx in the vertical atmosphere. A CMAQ simulation for summer 2011 shows that the LNOx to total NOx emission ratio are highest in the Southeast and Rocky Mountains regions. The addition of lightning NOx emissions reduces the daily CMAQ low bias for maximum 8-hr O3 by as much as 5 ppbv in the Rocky Mountains region. The lightning source of NOx should be included in regional air quality models.

Daiwen Kang

Daiwen Kang is a Physical Scientist with the Computational Exposure Division of the U.S. Environmental Protection Agency's National Exposure Research Laboratory. E-mail: kong.daiwen@epa.gov.

Kenneth E. Pickering

Kenneth E. Pickering is a Research Professor in the Department of Atmospheric and Oceanic Science at the University of Maryland, College Park. E-mail: pickerin@umd.edu.

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References


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