Midlatitude Lightning NO\textsubscript{x} Production Efficiency Inferred From OMI and WWLLLN Data

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Abstract Oxides of nitrogen are critical trace gases in the troposphere and are precursors for nitrate aerosol and ozone, which is an important pollutant and greenhouse gas. Lightning is the major source of NO\textsubscript{x} (NO + NO\textsubscript{2}) in the middle to upper troposphere. We estimate the production efficiency (PE) of lightning NO\textsubscript{x} (LNO\textsubscript{x}) using satellite data from the Ozone Monitoring Instrument and the ground-based World Wide Lightning Location Network in three northern midlatitudes, primarily continental regions that include much of North America, Europe, and East Asia. Data were obtained over five boreal summers, 2007–2011, and comprise the largest number of midlatitude convective events to date for estimating the LNO\textsubscript{x} PE with satellite NO\textsubscript{2} and ground-based lightning measurements. In contrast to some previous studies, the algorithm assumes no minimum flash-rate threshold and estimates freshly produced LNO\textsubscript{x} by subtracting a background of aged NO\textsubscript{x} estimated from the Ozone Monitoring Instrument data set itself. We infer an average value of 180 ± 100 moles LNO\textsubscript{x} produced per lightning flash. We also show evidence of a dependence of PE on lightning flash rate and find an approximate empirical power function relating moles LNO\textsubscript{x} to flashes. PE decreases by an order of magnitude for a 2 orders of magnitude increase in flash rate. This phenomenon has not been reported in previous satellite LNO\textsubscript{x} studies but is consistent with ground-based observations suggesting an inverse relationship between flash rate and size.

Plain Language Summary Oxides of nitrogen (NO\textsubscript{x} = NO + NO\textsubscript{2}) are minor gases in the atmosphere but are important in its chemistry. Their major source in the upper troposphere is lightning, which creates nitric oxide (NO) by breaking apart nitrogen and oxygen molecules. Estimating how much total NO is produced requires knowledge of the average production efficiency of individual lightning flashes. From midlatitude satellite measurements of nitrogen dioxide (NO\textsubscript{2}) and lightning detections from ground, we find a mean production efficiency of 180 moles NO per lightning flash, somewhat smaller than the ~250 mol/flash averaged globally in previous studies. We also show that production efficiency is smaller in storms with more frequent lightning.

1. Introduction

Trace gases individually represent less than 1% of all components of the Earth’s atmosphere but play significant roles in atmospheric chemistry. In particular, nitric oxide (NO) and nitrogen dioxide (NO\textsubscript{2}), collectively NO\textsubscript{x}, are critical in regulating concentrations of other trace gases. In pollution-free regions, the total NO\textsubscript{x} column is dominated by the stratospheric component. There, NO\textsubscript{x} occurs naturally as a by-product of photo-dissociation of N\textsubscript{2}O transported across the tropopause and plays a major role in the catalytic destruction of stratospheric ozone (Finlayson-Pitts & Pitts, 1999; Seinfeld & Pandis, 1998). Significant amounts of NO\textsubscript{x} also exist in the troposphere with global production estimates of ~48 Tg N/year (Miyazaki et al., 2016). Tropospheric NO\textsubscript{x} is generated by high-temperature reactions involving N\textsubscript{2} and O\textsubscript{2}. These are mainly anthropogenic and include combustion of fossil fuels and biomass burning. On average, some 90% of tropospheric NO\textsubscript{x} resides in the boundary layer, where it is a precursor to lower tropospheric ozone.

The major natural sources of tropospheric NO\textsubscript{x} are soil emissions and lightning (Finlayson-Pitts & Pitts, 1999; Seinfeld & Pandis, 1998; Vinken et al., 2014). In the upper troposphere, the lightning source dominates. It is estimated that lightning NO\textsubscript{x} (LNO\textsubscript{x}) accounts for 60–70% of free-tropospheric NO\textsubscript{x} (Allen et al., 2010). In the mid-upper troposphere, lightning dissociates N\textsubscript{2} and O\textsubscript{2}, into free N and O within the extremely hot flash channel. These in turn react with ambient N\textsubscript{2} and O\textsubscript{2} to produce NO, which remains after the
lightning channel cools. During the conversion between NO and NO$_2$, ozone is generated in the presence of H$_2$O and organic peroxy radicals, collectively called RO$_2$. Ozone is a significant greenhouse gas in the upper troposphere (e.g., Rap et al., 2015). It is sensitive to LNO$_x$ amounts, which are believed to be responsible for 35–45% of global free-tropospheric ozone (Allen et al., 2010; Dahlmann et al., 2011; Liaskos et al., 2015). Pickering et al. (1993, 1996) have shown persistent ozone enhancements downwind of convection. These long-range enhancements are consistent with NO$_x$ lifetimes of at least two to three days in the upper troposphere (Jaeglé et al., 1998; Martin et al., 2007; Schumann & Huntrieser, 2007), although initial lifetimes near the lightning source have been shown to be as short as 2–12 hr (Nault et al., 2017). In addition to its direct effect on radiative forcing, ozone can photodissociate and react with water vapor to create hydroxyl (OH), a strong oxidant. Among other roles, OH plays a large role in the destruction of methane (CH$_4$), another major greenhouse gas (DeCaria et al., 2000, 2005; Finlayson-Pitts & Pitts, 1999; Fiore et al., 2006; Labrador et al., 2004; Liaskos et al., 2015; Seinfeld & Pandis, 1998). Although its largest chemical impact is on the free troposphere, LNO$_x$ plays a small but nonnegligible role in boundary layer air quality (Allen et al., 2012; Murray, 2016).

Production rates of LNO$_x$ depend, in part, on lightning flash rates, and knowledge of these rates is critical in developing chemical transport models (CTMs). On the mesoscale, flash rates have been shown to vary as a power function of cloud top height (Price & Rind, 1992; Williams, 1985). Modeling studies have employed CTM flash parameterizations based on cloud top height, convective precipitation rate, anvil-level ice amounts, and vertical mass transport in updrafts (Allen et al., 2010; Tost et al., 2007). Murray et al. (2012) showed that flash rates in CTMs may be constrained with satellite measurements of lightning. Globally, mean annual flash rates are estimated to be approximately 46 fl/s (e.g., Cecil et al., 2014).

Lightning NO$_x$ production estimates also require knowledge of the moles of NO$_x$ produced per flash, known as the production efficiency (PE). This quantity is considerably less well known than the global flash rate and has been estimated in theoretical, laboratory, aircraft, and satellite investigations. The review studies of Schumann and Huntrieser (2007) and Murray (2016) cite PE estimates spanning 2–3 orders of magnitude, with values ranging from ~5 to >1,000 mol/fl. Schumann and Huntrieser (2007) suggest a global average PE of 250 mol/fl, which when coupled with the mean annual flash rate yields a global production of 5 ± 3 Tg N/year when uncertainties are included. Studies, including Price et al. (1997) (theory) and Koshak (2014) (measurement and theory), have indicated PEs of cloud-to-ground flashes (CG) to be an order of magnitude higher than those of intracloud (IC) flashes. However, aircraft observations in conjunction with cloud-scale models have found that IC and CG flashes are approximately equally productive (Cummings et al., 2013; DeCaria et al., 2005; Huntrieser et al., 2011; Ott et al., 2007, 2010), with mean PE values ranging from ~70 to 700 mol/fl. Regional differences have also been noted. There is some evidence that midlatitude flashes are more productive than those in tropical regions (Hudman et al., 2007; Huntrieser et al., 2006, 2008), and several CTMs including the Goddard Earth Observing System Chemistry (GEOS-Chem) (van Donkelaar et al., 2008) and Global Modeling Initiative (GMI) CTMs (Allen et al., 2010) assume a higher PE in the midlatitudes (~500 mol/fl) than tropics (~250 mol/fl). Other investigations indicate differences in continental and marine flashes. Boersma et al. (2005) found that continental flashes were ~1.6 times more productive than those over water, while Allen et al. (2019) estimated marine flashes to be twice as productive than continental flashes, consistent with earlier findings of more energetic flashes over oceans (Beirle et al., 2014; Chronis et al., 2016).

Satellite observations of LNO$_x$ combined with ground- or satellite-based lightning flash counts have become increasingly valuable in PE investigations. Satellites directly measure NO$_2$, which must be converted to NO$_x$. Methods to estimate PE from satellite measurements generally fall into two categories: those using long-term trace-gas measurements of species like O$_3$ and HNO$_3$ to constrain NO$_x$ PEs in CTM simulations and those that constrain PE using retrievals of freshly produced NO$_x$ and concurrent lightning data. Examples of the former include the model assimilations of Boersma et al. (2005), Martin et al. (2007), and Miyazaki et al. (2014), which yielded NO$_x$ production rates equivalent to mean global PEs of 55 ± 320, 300 ± 100, and 320 ± 70 mol/fl, respectively. More recently, Marais et al. (2018) combined three years of NO$_2$ data obtained from the Ozone Monitoring Instrument (OMI) retrieved by cloud slicing (Choi et al., 2014; Ziemke et al., 2001) with lightning measurements from Optical Transient Detector (OTD) and Lightning Imaging Imaging Sensor (LIS) and the GEOS-Chem model to obtain a mean PE of 280 ± 80 mol/fl.
Investigations based on freshly produced LNOx face a number of observational hurdles. Some of these are instrument-related, such as the low spatial resolutions of satellite instruments, which can complicate comparisons with lightning in small-scale convective systems, as well as saturation of instrument pixels over bright convective clouds (L. Lamsal, H. Eskes, private communications). As in assimilation studies, the dominant stratospheric part of the satellite-measured total NO\textsubscript{2} column must be removed from the smaller LNO\textsubscript{2} component. A variety of schemes are used to do this, each yielding different results on subsynoptic scales (Beirle et al., 2016; Boersma et al., 2011; Bucsela et al., 2013; Richter & Burrows, 2002; Wenig et al., 2003; Yang et al., 2014). Ambient tropospheric NO\textsubscript{x} must also be distinguished from the fresh LNO\textsubscript{x} signal. The use of cloudy scenes limits contamination of the lightning signal by NO\textsubscript{x} in the lower troposphere with a likely anthropogenic source (Allen et al., 2019; Beirle et al., 2010; Marais et al., 2018; Pickering et al., 2016). High flash-rate restrictions have also been imposed to ensure that lightning NO\textsubscript{x} is the predominant component of the satellite signal and that the background contribution is minor (Beirle et al., 2010; Pickering et al., 2016). Approaches for estimating the background have been described by Bucsela et al. (2010), Pickering et al. (2016), and Allen et al. (2019). An additional challenge is synchronization of NO\textsubscript{x} data with concurrent lightning measurements. To date, all satellite LNO\textsubscript{x} studies have been based on low Earth orbit instruments, which make, at best, only one NO\textsubscript{x} measurement per day of a given region. Their local time (LT) for overpass is also before the late-afternoon convective peak, as with the Global Ozone Monitoring Experiment (LT = 10:30), the Scanning Imaging Absorption Cartography instrument (LT = 10:00), OMI (LT = 13:45), and the Tropospheric Monitoring Instrument (LT = 13:30).

The PEs derived from fresh LNO\textsubscript{x} measurements have generally yielded smaller PEs than assimilations. Using tropical data from the Tropical Composition, Cloud and Climate Coupling Experiment, Bucsela et al. (2010) found a mean PE of 174 ± 219 mol/fL based on OMI NO\textsubscript{x} measurements and lightning data from World Wide Lightning Location Network (WWLLN) and the Costa Rica Lightning Detection Network. Pickering et al. (2016) estimated a mean PE of 80 ± 45 mol/fL over the Gulf of Mexico, using data sets from OMI and the WWLLN. Over the same region, using Global Ozone Monitoring Experiment data, Beirle et al. (2006) derived a similar value of 90 (range of 32 to 240) mol/fL. However, Beirle et al. (2010) obtained a nearly null result in their Scanning Imaging Absorption Cartography-based study of 287 convective events, with no detectable LNO\textsubscript{x} enhancement over most storms and overall negligible correlation with flash counts. Allen et al. (2019) used updated versions of the Pickering et al. (2016) data sets, similar to those of the present study to estimate a PE value of 170 ± 100 mol/fL in the tropics. They also noted a decrease in PE with increasing lightning flash rate, as well as the higher values over water relative to land.

The present study examines three midlatitude regions (eastern North America, Europe, and East Asia and adjacent waters), using 15 months of data from the five boreal summers (June-July-August) of 2007–2011 to investigate the LNO\textsubscript{x} PE. NO\textsubscript{x} observations from OMI are compared with lightning flashes detected by the WWLLN and converted to NO\textsubscript{x} using air mass factors based on LNO\textsubscript{x} and LNO\textsubscript{2} profiles from a chemical-transport model. NO\textsubscript{x} retrieved in regions of deep convection without lightning flashes are subtracted as the tropospheric NO\textsubscript{x} background. This investigation and the tropical study of Allen et al. (2019) are the largest-scale satellite studies to date (in terms of observed convective events) of the amount of NO\textsubscript{x} production from observed flash data.

2. Data Description
2.1. OMI
The Dutch-Finnish OMI spectrometer is one of four instruments on NASA’s Aura satellite, launched 15 July 2004 (Levett et al., 2006; Schoeberl et al., 2006). The satellite is in a Sun-synchronous orbit with equator and midlatitude crossing times of 13:45 and ~13:30 LT, respectively. OMI operates in push-broom configuration with a swath spanning 2,600 km. In normal mode there are 60 pixels across the swath, and the field of view of nadir pixels is 13 × 24 km\textsuperscript{2}. There are ~1,600 swaths per orbit and ~15 orbits per day. Midlatitude LTs across a swath can differ from nadir by as much as ±1 hr, and there is significant overlap between adjacent orbits. However, averaging of data in the present study minimizes the effects of LT variation. Beginning in 2007, OMI pixels were affected by the row anomaly (RA), first described by Dobber et al. (2008). The RA reduced valid data across each swath by ~3 to ~30 pixels between 2009 and 2011, and doubled the time required for
global coverage from one to two days. Bad-pixel flagging and CCD dark current also grew by a factor of ~2 during the 2007–2011 period, and signal-to-noise ratio decreased (Schenkeveld et al., 2017).

NO$_2$ slant columns densities (SCDs) used in this study are obtained from the v3.0 NASA OMI NO$_2$ standard data product (Krotkov et al., 2017; Marchenko et al., 2015). Relative to SCDs in the previous v2.1 product (Boersma et al., 2011; Bucsela et al., 2013), the v3.0 SCDs are 10–40% smaller. The v3.0 algorithm employs an iterative spectral fitting routine in the 402–464-nm range that corrects errors in wavelength registration and separately fits the Ring spectrum due to rotational Raman scattering and absorption cross sections for NO$_2$, H$_2$O, and C$_2$H$_2$O$_2$ (glyoxal). Data quality at these wavelengths has been relatively stable over the instrument’s lifetime. Radiometric degradation, stray light interference, and wavelength calibration errors have remained below 3%, 0.5%, and 0.002 nm, respectively (Dobber et al., 2008; Marchenko & DeLand, 2014; Schenkeveld et al., 2017). However, NO$_2$ SCD measurements between 2005 and 2015 over the central Pacific showed an increase in standard deviation from $0.8 \times 10^{15}$ to $1.0 \times 10^{15}$ cm$^2$ (Krotkov et al., 2017).

NO$_2$ SCDs are combined with level-2 NO$_2$ stratospheric vertical column densities (VCDs) and stratospheric air mass factors (AMFs), also from the v3.0 product. AMFs are based on a priori cloud and terrain inputs as well as chemical-transport and radiative-transfer model output. Cloud optical centroid pressure (OCP) and cloud radiance fraction (CRF) are obtained from the OMI O$_2$–O$_2$ algorithm, where OCP is the effective cloud top pressure visible from OMI (Acarreta et al., 2004; Sneep et al., 2008; Stamnes et al., 2008; Vasilkov et al., 2009). Terrain pressures and reflectivities are obtained, respectively, from a digital elevation model and OMI clear-sky measurements (Kleipool et al., 2008). The GMI chemistry-transport model (section 2.3) computes monthly NO$_2$ profile shapes and tropopause pressures for the AMFs. Atmospheric scattering weights are calculated by TOMRAD (Davé, 1965). The AMFs are also used in the stratosphere-troposphere separation (STS) algorithm (Bucsela et al., 2013) to derive the stratospheric VCDs from the total SCDs.

### 2.2. WWLLN

The World Wide Lightning Location Network is a continuously operating ground-based global array of very low frequency radio wave sensors that detect sferics from lightning (Dowden et al., 2002; Lay et al., 2005; Virts et al., 2013). The number of active sensors grew from 11 at initial global deployment in 2003 to ~30 in 2007, to ~60 in early 2012 (Hutchins et al., 2012). The increase between 2003 and 2007, alone, led to a growth in the number of lightning detections by ~165%. Relative to LIS, detection efficiencies (DEs) in 2011 were estimated to be on the order 6–15% in the Western Hemisphere tropics by Rudlosky and Shea (2013) and ~20% and ~30% by Pickering et al. (2016) in the ±60° latitude band and Gulf of Mexico regions, respectively. Detection range is ~10,000 km, allowing reasonable coverage with a sparse array (Lay et al., 2005; Rodger et al., 2006, 2009). Spatial and temporal accuracies are ~5 km and <10 μs, respectively (Abarca et al., 2010). Sensitivities are higher for CG than IC flashes (Rodger et al., 2009; Rudlosky & Shea, 2013), although we do not distinguish between the two in the present study.

The WWLLN detections used in this study are calibrated against satellite climatologies to estimate WWLLN DEs. Similar approaches were used by Pickering et al. (2016) and Allen et al. (2019). In the present study, adjustment factors were applied to the observed WWLLN strokes to ensure that the mean WWLLN flash rate over the 2007 to 2014 time period at each grid box matches that of the v2.3 OTD/LIS climatology (Cecil et al., 2014). The climatology is derived from measurements by the Optical Transient Detector (Boccippio et al., 2004, 2002), operational from 1995 to 2000, and the Lightning Imaging Sensor, with useful full-year data during the years 1997 to 2013. Comparisons were limited to the latitude ranges of the instruments, which are ±35° for LIS and ±70° for OTD. Details of the methodology are given in Appendix A. WWLLN flashes are summed over the 1-hr period before 13:30 LT, the approximate OMI overpass time.

Figure 1 shows WWLLN sensor locations and Northern Hemisphere detection efficiencies. In the geographical domain of this study, area-weighted DEs in 2007 (2011) were 3.6% (10.6%), 2.3% (4.5%), and 2.7% (8.0%), respectively. These values correspond to an average ~3–8% increase over five years in the probability that WWLLN detects a given flash. Thus, the chance of detecting a flash more than doubles. The impact of inter-annual variability is likely small, since it is only approximately ±10% during the period. Since most flashes occur over land, where DEs are smaller (see Figure 1), the actual fraction of flashes detected by WWLLN is...
10–50% less than the area-weighted DE. The small magnitude and high spatial and temporal variability of the DE make the WWLLN flash counts a major source of uncertainty in the present results.

2.3. GMI

NASA's Global Modeling Initiative model (Allen et al., 2010; Duncan et al., 2007; Strahan et al., 2007, 2013; Ziemke et al., 2006) is the CTM used in both the OMI standard NO2 product and here to estimate shapes of LNO2 and LNOx vertical profiles for AMF calculations. The GMI simulations for this study were driven by meteorological fields from GEOS-5 Modern Era Retrospective analysis for Research and Applications (MERRA) (Rienecker et al., 2011). Year-specific monthly mean GMI output was computed for 2007–2011 using Emission Database for Global Atmospheric Research 2000 fossil fuel data and biomass burning emission inventories (van der Werf et al., 2010) with annual scaling factors from the Goddard Earth Observing System-Chemical Transport Model (GEOS-Chem) (van Donkelaar et al., 2008). In this study, we compute LNO2 and LNOx profiles as the difference between model output with and without a lightning source. The lightning contribution is based on Allen et al. (2010) and assumes a PE of 500 mol/fl poleward of ±26° and 250 mol/fl equatorward of ±26°. This stepwise change in PE affects modeled amounts of LNO2 and LNOx but has relatively little impact on their on their profile shapes. Between latitudes 20° and 40°, latitudinal variation of AMF is nonsystematic with a range of ~0.3 to ~0.7. Any AMF discontinuity near the 26° latitude line is <0.05.

2.4. Data Domain

The domain for this study was selected based on the quality of available OMI and WWLLN data. Increases in noise and the effects of the row anomaly significantly compromised OMI data from 2009 onward. However, increasing WWLLN DEs reduced uncertainties in the WWLLN flash counts during the same period. The five summers of 2007–2011 were chosen as a compromise between the years of highest OMI and WWLLN data quality. Measurements during this period were taken in three midlatitude regions having high climatological lightning frequency. The regions are (1) the eastern and Central United States and southern Canada along with adjacent parts of the western Atlantic, Gulf of Mexico, and Caribbean between longitudes −115° and −55° and latitudes 20° to 60°; (2) Europe and the Mediterranean, between longitudes −10° and 60° and latitudes 30° to 60°; and (3) East Asia and the western Pacific between longitudes 90° and 150° and latitudes 20° to 60°. Further data selection criteria are given in section 3.
3. Method

Here we describe retrieval of LNOx amounts from the level-2 OMI NO2 data. Vertical-column tropospheric NOx over deep convective grid boxes ($V_{\text{LNOx}^*}$) is derived from OMI pixel data:

$$V_{\text{LNOx}^*} = \left( S - V_{\text{strat Zonal}} \cdot A_{\text{strat}} \right) / A_{\text{LNOx}}$$  \hspace{1cm} (1)

Here $S$ is the total SCD from the OMI NO2 spectral fit v3.0 algorithm (Marchenko et al., 2015). $A_{\text{strat}}$ is the stratospheric air mass factor, which depends on viewing geometry. $A_{\text{LNOx}}$ is the air mass factor that converts the tropospheric NO2 slant column to the NOx vertical column, denoted here as $V_{\text{LNOx}^*}$ (see Allen et al., 2019; Pickering et al., 2016). The asterisk indicates that the vertical column includes contributions from nonlightning NOx sources and nonrecent lightning; that is, a background correction has not been made. $V_{\text{strat Zonal}}$ is the zonally averaged v3.0 stratospheric VCD ($V_{\text{strat}}$; Bucsela et al., 2013; Krotkov et al., 2017). It is obtained by smoothing $V_{\text{strat}}$ in pixels with CRF > 0.97 and OCP < 500 hPa, using a ±180° longitude and ±3° latitude running boxcar. Zonal smoothing eliminates longitudinal variations in stratospheric NO2 concentration, tropopause height, and the a priori troposphere used in the STS algorithm. The smoothing is needed because the STS algorithm can erroneously assign small amounts of tropospheric NO2 to the stratosphere (Allen et al., 2019; Beirle et al., 2016; Bucsela et al., 2013). We multiply the smoothed stratospheric VCD by the stratospheric AMF and subtract from the total SCD to obtain the tropospheric NO2 SCD, that is, the term in parentheses in equation (1).

The tropospheric SCD is divided by an air mass factor, $A_{\text{LNOx}}$, computed from model GMI NO2 and NOx profiles and TOMRAD (Davé, 1965) scattering weights. The profiles were created using the difference between model runs with and without a lightning source of NO. Following Pickering et al. (2016), the profile at a given location is chosen from the day with the third largest LNOx column during a given month and year and is considered representative of moderate-to-active convective environments. Midlatitude profile examples for June 2007 are shown in Figure 2. Conceptually, $A_{\text{LNOx}}$ is the ratio of the modeled tropospheric LNO2 SCD to the modeled LNOx* VCD from tropopause to ground. Retrieved $V_{\text{LNOx}^*}$ therefore includes LNOx* below the OCP that cannot be directly observed by OMI. For the domain of this study, the mean OMI OCP is 483 hPa with a standard deviation of ~100 hPa. The corresponding GMI fraction of the LNOx column below the OCP is ~10–30%. This range is consistent with profiles from the cloud-resolved simulations of Ott et al. (2010) from the CRYSTAL-FACE campaign.

Only pixels with CRF > 0.97 and OCP < 500 hPa are used in the analysis. These restrictions favor data from the bright, opaque clouds associated with deep convection (Pickering et al., 2016), and minimize contamination by low- and middle-level NO2, especially over low-level stratus clouds, which enhance visibility of ambient NO2 immediately above them (Martin et al., 2002a).

LNOx* vertical columns are binned in the 1° longitude × 1° latitude grid boxes used for the WWLLN flashes, with a minimum of 3 OMI pixels per box. The number of LNOx* molecules in each box is obtained by multiplying the average of the vertical columns from the pixels by grid box area. This scheme was chosen over pixel-area weighting (e.g., Nault et al., 2017), which imparts too much weight to pixels at swath edges, where adjacent orbits overlap, and local times can differ from local time at nadir (13:30 LT) by as much as 1 hr. We count WWLLN flashes in a 1-hr window prior to the 13:30 LT OMI overpass. This window minimizes advection of fresh lightning NOx out of boxes before the OMI measurement. The choice of 1 hr is based on mean midlatitude GEOS-5 MERRA upper tropospheric (UT) wind speeds, which are estimated in the range 11 to 21 m/s. It is shorter than the 3-hr window used by Pickering et al. (2016) in their Gulf of Mexico study, where summertime UT wind speeds are 6 to 11 m/s.

Figure 2. The 2007 June profiles of LNO2 and LNOx from GMI for longitude 85°W, latitude 40°N. Profiles are the difference between NO2 (NOx) from simulations without lightning and NO2 (NOx) from simulations with a lightning source of NO.
A tropospheric NOx background is subtracted from the LNOx* in each grid box to remove ambient NOx not generated within the 1-hr flash window. The background is a weighted temporal average of boxes at each geographic location having zero to one flash during the window. Background boxes are subject to the same OCP and CRF restrictions as boxes with lightning and are weighted according to the number of OMI pixels contributing to each. The gridded background is smoothed with a $5^\circ \times 5^\circ$ boxcar to lessen noise and gaps. Subtraction of the two-dimensional background array from the three-dimensional array (longitude, latitude, day) of LNOx* yields an array of LNOx values, defined as LNOx* corrected for background. Grid boxes containing both valid LNOx and nonzero flashes will be referred to as “flashing boxes.” LNOx was also corrected for convectively lofted pollution and chemical decay. The amount of lofted pollution is assumed to be proportional to LNOx, as both are assumed to scale with convective updraft strength. Therefore, pollution is considered a fraction of the LNOx signal, rather than an absolute component of the mean background, which consists of boxes without active lightning. The magnitude is derived from DeCaria et al. (2000, 2005), who examined a midlatitude storm NNE of Denver near the Wyoming border on a day with boundary layer flow from the east. They estimated pollution to comprise <~20% of total anvil-level NOx. We choose a slightly lower 15% pollution correction to account for the mix of rural and urban regions in the data domain. The estimate of chemical decay is based on the 3-hr LNOx lifetime of Nault et al. (2017), which was obtained from measurements in near-field convective outflow. With this value, the average decay over a 1-hr period is ~15%, and we adjust measured LNOx upward accordingly. This adjustment approximately cancels the lofted-boundary layer NOx correction; however, both corrections are included in the algorithm.

4. Results

4.1. Geographic Distribution

Maps of OMI and WWLLN data are shown in Figure 3. The red boxes outline the three areas of study—eastern North America, Europe, and East Asia. The fields shown are temporal means of $V_{\text{ini}}-V_{\text{StratZonal}}$ ($\Delta V_{\text{NO2}}$), LNOx*, LNOx, and lightning in flashing boxes for the five summer (June–July–August) periods. $V_{\text{ini}}$ is the “initial” vertical column retrieved from OMI, defined as $S/A_{\text{strat}}$, or the ratio of the total OMI NO2 slant column to the (approximately geometrical) stratospheric AMF. As such, it depends only on the OMI spectral fit with no further geophysical assumptions. In the figure, the fields have been smoothed with a $3^\circ$ longitude $\times 3^\circ$ latitude boxcar for clarity. Qualitatively, Figure 3 shows that the three regions of study contain higher $\Delta V_{\text{NO2}}$, LNOx*, LNOx, and lightning relative to other northern midlatitude areas. Spatial correlations of LNOx* with lightning are highest in China and the southeastern United States. These regions also have relatively high flash rates. Pearson’s correlation coefficients, $r$, between LNOx* and lightning are 0.22, 0.12, 0.25, and 0.22 for North America, Europe, East Asia, and the combined region, respectively. Correlations between LNOx and lightning are weaker, in part due to noise in the subtracted background, but can be seen qualitatively on scales of ~2,000 km. The $r$ values for LNOx are 0.16, 0.15, 0.21, and 0.18 for North America, Europe, East Asia, and their sum, respectively. The significance of these small $r$ values will be examined in section 4.3. Regions with low WWLLN detection efficiencies may be misclassified as background, lessening the contrast between flashing and nonflashing boxes and reducing the LNOx signal. For example, the LNOx* enhancement over NW India and Pakistan is weak in the LNOx field. Lightning climatologies from OTD/LIS do show enhanced lightning in this region (Cecil et al., 2014), but detected flashes are few during the time immediately preceding OMI overflights. The disparity may indicate poor WWLLN coverage with incorrect DE estimates, an inaccurate diurnal distribution of flashes, or lingering ambient LNOx* from earlier convection. Differences in the LNOx* and LNOx fields in the southeastern United States that are less prominent in the LNOx field may also result from lightning before the 1-hr flash window, including recirculation around the Bermuda High (Cooper et al., 2006, 2007).

4.2. Mean Production Efficiency

The 15-month LNOx and 1-hr flashes were summed over all grid boxes in the data domain to estimate an average PE. This approach is comparable to the summation method of Pickering et al. (2016). Mean LNOx is 130 kmol per flashing box, which is 45% of mean LNOx*. On average, the same flashing boxes contain 740 WWLLN flashes. We compute the ratio of total LNOx to total flashes and obtain an average PE of 180
± 100 moles LNO$_x$ per flash. Bias and error estimates are discussed in section 5. We define the average PE as described to facilitate comparisons with previous studies. If this PE value is representative of all flashes globally, combining it with the estimated mean global flash rate (Cecil et al., 2014) would yield an annual global LNO$_x$ budget of 3.5 ± 2.0 Tg N/year. The possibility of regional differences in PE is examined in sections 4.4 and 5.2.

### 4.3. Data Correlations

In Figure 4, $\Delta V_{NO_2}$ and LNO$_x$ from individual flashing boxes are plotted against DE-adjusted WWLLN flash rates. The points represent the ~32,000 flashing boxes in the data set. Approximately 39% of these have negative LNO$_x$ values due, in part, to overestimation of the tropospheric background at those locations. $\Delta V_{NO_2}$ also contains negative values, indicating some overestimation of stratospheric NO$_2$. Data at higher flash rates are relatively sparse, with less than 10% of flashing boxes containing over 2 kfl/hr and ~1% having rates exceeding 6 kfl/hr (see also Figure 11). Because of these issues, any relationship between the OMI data and lightning is difficult to discern in the figures. Correlations with adjusted flash rates are $r = 0.20$ for $\Delta V_{NO_2}$ and 0.18 for LNO$_x$.

Although the correlations are not large, they are significant. The $p$ value corresponding to Figure 4b is $p < 0.0005$. The significance can be further demonstrated by comparing correlations between WWLLN flashes in a given box with OMI-derived LNO$_x$ in other boxes. The plots in Figure 5 show $r$ values

---

**Figure 3.** Mean daily data for JJA 2007–2011 per 1° longitude × 1° latitude box, averaged over flashing boxes. (a) $\Delta V_{NO_2}$ (10$^{14}$ molec/cm$^2$), (b) LNO$_x$ (10$^4$ moles), (c) LNO$_x$ (10$^4$ moles), and (d) mean daily WWLLN 1-hr flash counts (10$^2$). The red boxes outline the three geographic regions (North America, Europe, and East Asia) examined in this study.
when flash counts are compared with LNOx in surrounding boxes on the same and different days. In Figure 5b, $r = 0.18$ in the central box represents the value for the LNOx correlation with lightning in the same boxes on the same days. The correlations in surrounding boxes are lower and may be due to advection and/or lightning occurring before the 1-hr integration period. The slight enhancement northeast (to the upper right) of the central box is consistent with the typical southwesterly flow environment for northern midlatitude convection (e.g., Markowski & Richardson, 2011). Figures 5a and 5c also illustrate spatial correlations but compare LNOx on a given day with 1-hr flashes from the preceding and following days, respectively. In the central boxes, LNOx shows minimal correlation with lightning from the previous day (maximum value of $r = 0.08$ in the central box) and approximately none with lightning on the next day ($r = 0.04$ in the central box). Together these results are strong evidence that a sizable portion of the LNOx in each grid box is produced by lightning flashes in the same box during the hour before OMI overpass.

A relationship between the OMI and WWLLN data becomes clearer when the lightning-NOx metrics $V_{\text{init}}$, $\Delta V_{\text{NO2}}$, $\text{LNOx}^*$, and LNOx are binned by flash rate, as shown in Figures 6a–6d, respectively, using three OCP thresholds. The fields from Figures 6a–6d represent successively greater amounts of processing applied to the OMI data. Specifically, $V_{\text{init}}$ depends only on OMI slant columns (and a geometrical AMF), $\Delta V_{\text{NO2}}$ assumes a stratospheric estimate, $\text{LNOx}^*$ adds the assumption of a model-based AMF, and LNOx includes the additional tropospheric background estimate. All fields exhibit qualitatively similar behavior, showing...
a nonlinear dependence on flash rate, with the slopes of the curves implying a decrease in production efficiency as flash rate increases. The curves in Figures 6a–6d are also somewhat more linear for OCP < 400 hPa than for larger OCP thresholds, resulting in higher LNOx* and LNOx (Figures 6c and 6d) at the highest flash rates (see section 4.5). However, \( V_{\text{init}} \) and \( \Delta V_{\text{NO2}} \) (Figures 6a and 6b) are both uniformly lower at all flash rates for OCP < 400 hPa due to neglect of LNO2 in those fields below the OCP altitude. We quantify these effects below and discuss geophysical bases for the nonlinearity in section 5.

4.4. Quantitative Dependence of PE on Flash Rate

In Figure 7, LNOx values are averaged in 500-fl/h bins between 0 and 10,000 fl/hr, with each bin containing at least three flashing boxes. The linear correlation coefficient for the binned data is \( r = 0.87 \). Two weighted fits were performed: an ordinary least squares linear fit (blue) and a power function fit (red) given by

\[
y = a + b \times x \\
y = \alpha x^\beta
\]

respectively, where \( x \) is kfl/hr and \( y \) is kmol LNOx. Weights are inverse standard errors of the mean, shown as error bars. The LNOx PEs are the derivatives of equations (2) and (3). For the linear fit we find \( a = 79 \pm 29 \) kmol and \( b = 64 \pm 10 \) mol/fl, where \( b \) is the regression-based PE, assuming a linear relationship between flashes and LNOx. The power law coefficient is \( \alpha = 8.0 \pm 0.8 \) kmol, with exponent \( \beta = 0.45 \pm 0.01 \). Fits to the unbinned LNOx data yield \( a = 94 \) kmol, \( b = 45 \) mol/fl, \( \alpha = 10.3 \) kmol, and \( \beta = 0.42 \), with negligible standard errors. While binning clarifies the relationship between LNOx and flashes, the comparable fitted values suggest that results do not depend strongly on binning scheme (see also section 4.5). Reduced chi squares \( (\chi^2_1) \) for the linear and power-function fits to the binned data are 10.1 and 1.5, respectively. The comparison shows the power function to be a much better fit than a straight line. In Figure 7a, the data are plotted with
linear scaling on the axes, while Figure 7b is a log-log plot. The slope of the orange line in Figure 7b corresponds to a direct proportion (constant PE) between moles LNOx and flashes, while the horizontal green line represents no relation between the two (vertical placement of the orange and green lines is arbitrary). The power function with $\beta \sim 0.4$ lies between the two. The slope of power function fit corresponds to a PE that decreases by over an order of magnitude, from 290 to 20 mol/fl for flash rates between 100 and 10,000 fl/hr. The average PE of 180 mol/fl derived from all flashes and LNOx over the entire data domain is an intermediate value.

Figure 8 shows that the flash-rate dependences for the three individual midlatitude regions are similar to that of the combined region. The smaller data sets in North America and Europe yield $r$ values of 0.59 and 0.42, respectively, which are less than found in the combined region. However, the value of $r$ for East Asia is 0.86, approximately the same as the combined value. The reduced chi-squares for each of the three regions all support a nonlinear relationship between flashes and the LNOx produced. Average PEs obtained by summation in each region are comparable, with values for North America, Europe, and East Asia of 200, 150, and 160 mol/fl, respectively. Results are summarized in Table 1.

4.5. Dependence on OCP

Vertical profiles of mean flash rate and mean LNOx as a function of OCP are shown in Figure 9. Flash and LNOx values for flashing boxes have been averaged in 30-hPa-wide bins. The average flash rate increases with decreasing OCP (Figure 9a), indicating more frequent flashes in deeper convection. This result is in qualitative agreement with the findings of Williams (1985) and Price and Rind (1992), who describe a power
Table 1
Average PE and Regression Analysis of Binned Data From the Three Geographic Regions Separately and Combined

| Region     | r    | b (mol/l) | $r^2$ (linear) | $r^2$ (power) | PE$_{avg}$ (mol/l) |
|------------|------|-----------|----------------|---------------|-------------------|
| North America | 0.59 | 60 | 0.42 | 5.79 | 1.84 | 200 ± 110 |
| Europe     | 0.42 | 41 | 0.39 | 2.04 | 1.40 | 150 ± 90  |
| East Asia  | 0.86 | 67 | 0.51 | 5.77 | 1.78 | 160 ± 100 |
| Combined   | 0.87 | 64 | 0.45 | 10.1 | 1.49 | 180 ± 100 |

Note. Statistics are computed with flashes averaged in 20 bins of 0.5 kft/hr.

law dependence on altitude, with exponents of ∼5 and 1.7 for continental and marine regions, respectively. Sparse high-flash rate marine data in the present study preclude a comparison of the two regions. However, using WWLLN flash rates from all boxes, we find an overall weaker altitude dependence, corresponding to an exponent of 0.87 ± 0.04, as indicated by the dashed line. The fifth power dependence for continental regions is theoretical and based on IR cloud top heights rather than OCP-derived altitudes as used here. Significant averaging is used in the flash-count processing, which could also obscure the apparent altitude dependence. Figure 9b shows average LNO$_x$ as a function of OCP. LNO$_x$ increases with altitude at large OCPs but decreases and becomes more variable above the 450-hPa level. The cause of the decrease could be related to a bias in the model profiles below the OCP, although a somewhat weaker altitude dependence of LNO$_x$ is plausible at higher altitudes, where increases in LNO$_x$ due to more flashes may be countered by decreases in PE. This behavior contrasts with the finding of Boersma et al. (2005) that LNO$_2$ increases with the fifth power of altitude, a result similar to the continental flash-rate dependence of Price and Rind (1992).

As noted in section 4.3, Figure 6 shows that the flash-rate dependence varies with OCP range. The dependences of the fields $V_{init}$, $\Delta V_{NO2}$, LNO$_x^*$, and LNO$_x$ on flash rate become slightly less linear at larger OCPs (lower cloud tops). Exponents, $\beta$, for a power law fit to LNO$_x$ versus flash, binned as in Figures 7 and 8, are 0.55, 0.45, and 0.40 at OCP thresholds of 400, 500, and 600 hPa, respectively, indicating a progressive decrease in linearity. The $\beta$ values for the three geographic regions in Table 1 reflect a similar dependence on OCP. Mean OCPs are largest over the convectively active regions of Europe and smallest over convection in SE Asia. These two regions also have the smallest and largest $\beta$ values, respectively.

Fitting statistics are shown in Figure 10 as a function of the number of bins in the 0 to 10,000 flash-per-hour range. The correlation coefficient, $r$, decreases from roughly 0.9 to 0.5 at all OCPs (with a small overall decrease toward smaller OCPs) as the number of bins increases from 20 to 200 due to increased scatter. With OCP thresholds of 500 and 600 hPa, reduced chi-squares for the linear fits decrease from approximately 12 to 3.5 and >12 to 4.5, respectively, while those for the power-function fits are relatively constant at ~1.5. For an OCP threshold of 400 hPa, the respective reduced chi-squares for the linear and power-function fits have ~constant values of 3.5 and 2.5. At that threshold, the smaller disparities between linear and power function fits as well as the weaker dependence of the linear fit on number of bins result from both poorer statistics (due to sparser data above the 400-hPa level) and $\beta$ values closer to unity than for larger OCP thresholds. Qualitatively, it was found that this OCP dependence does not depend significantly on the form of the OCP restrictions (e.g., whether OCP thresholds or narrow ranges of OCP values are specified), the model-estimated fraction of LNO$_x$ below OCP (implicit in $A_{LNOx}$), or whether a background is subtracted. Furthermore, there is no known dependence of WWLLN DE on cloud top height. The effects could conceivably result from variations in flash properties, including duration, extent, and radiance, for high- and low-topped convection, but further exploration is needed to draw any conclusions regarding geophysical causes. The role of flash extent is examined in section 5.

Figure 9. Profiles of (a) 1-hr flashes and (b) LNO$_x$ (10$^3$ moles) versus OMI OCP (hPa) (the y axis). Data have been averaged in 30-hPa-wide bins. The dashed line in (a) is a power law fit to the lightning data.
5. Discussion

5.1. Flash-Rate Dependent Production

The present results are the first satellite-based study to suggest that LNO\textsubscript{x} production follows a power function with exponent 0 < β < 1. This relationship appears robust at rates above ~100 fl/hr in each grid box but must be modified near a zero-flash rate to avoid an infinite PE. The mean PE is a weighted average of PEs at specific flash rates with weights proportional to the number of flashes at each rate. For a unit flash integration period, let \( N \) be the total number of flashing boxes. Also, let \( x \) be the flash rate per box, \( f(x) \) be the PE, and \( p(x) \, dx \) be the probability of a flash rate between \( x \) and \( x + dx \). Integrating over flash rate, the total number of flashes is

\[
\phi = N \int x \, p(x) \, dx
\]  

(4)

The corresponding number of moles LNO\textsubscript{x} produced is

\[
\mu = N \int f(x) \, p(x) \, dx
\]

(5)

and the weighted mean production efficiency is \( \text{PE} = \mu/\phi \). Figures 11a–11c are histograms of \( p(x) \), \( x \, p(x) \), and \( f(x) \). Figure 11a is effectively a density histogram of the scatterplots in Figure 4. All three quantities decrease with flash rate, and Figure 11b indicates that total flash count is dominated by storms with the lowest flash rates. Flash rates in the 32,000 flashing boxes range from 1 to 45,000 fl/hr. Approximately 90% of boxes have rates less than 2,000 fl/hr and these account for 50% of all flashes.

The smaller PE at high flash rates may explain difficulties in detecting significant LNO\textsubscript{x} in some studies. Pickering et al. (2016) imposed a minimum threshold of 1,000 fl/hr in 1° × 1° grid boxes and obtained a relatively low PE of 80 mol/fl. The 287 cases examined by Beirle et al. (2010) were restricted to rates equivalent to a threshold of 9,000 fl/hr. They found an LNO\textsubscript{x}–flash correlation coefficient of only 0.04 and PEs less than 15 mol/fl in approximately half of their examined cases. These values are consistent with the present study at comparably high flash rates.

5.2. Geophysical Implications

Studies have suggested that PE may be directly related to flash size, with larger flashes producing more LNO\textsubscript{x} (Carey et al., 2014; Huntrieser et al., 2008; Marais et al., 2018). During the TROCCINOX campaign, Huntrieser et al. (2008) attributed lower tropical PE values to shorter stroke lengths relative to those of mid-latitude storms. The effect of flash extent and LNO\textsubscript{x} production was quantified by Carey et al. (2014). They applied the NASA Lightning Nitrogen Oxides Model (Koshak, 2014) to observations made during the Deep Convective Cloud and Chemistry campaign and computed LNO\textsubscript{x} production per meter of channel length from a Lightning Mapping Array, based on laboratory measurements and theoretical assumptions.
They found LNOx production to be highly correlated with flash extent (correlation coefficient of $r = 0.99$), indicating that the PE is controlled almost entirely by the flash size.

Evidence from field campaigns have also shown a relationship between flash rate, flash size, and updraft strength. In a study of two 2004 supercells, Bruning and MacGorman (2013) reported that increasing updraft strength is associated with smaller flash extents and higher average flash rates. Similar associations were noted by Carey et al. (2005), Kuhlman et al. (2006), and Weiss et al. (2012), as well as studies from the Deep Convective Cloud and Chemistry campaign (Barth et al., 2015; Bruning & Thomas, 2015; Carey et al., 2014; Mecikalski et al., 2015). Bruning and Thomas (2015) demonstrated that the anticorrelation between flash rate and size is strongest during the decay phase of the storm as the updraft weakens and flash rates drop, while the total energy from lightning diminishes. Marais et al. (2018) noted a stronger correlation of LNOx with flash extent than with flash duration or radiance, implying that the dependence of flash extent on flash rate may be the dominant factor driving the PE dependence on flash rate, consistent with Carey et al. (2014). Together, the above studies provide a possible geophysical basis for the dependence of PE on flash rate found in the present investigation.

Many CTMs parameterize LNOx production with the assumption that midlatitude flashes produce more NO per flash than tropical flashes based on analyses of data from field campaigns (Allen et al., 2010; Hudman et al., 2007; Murray et al., 2012; Ott et al., 2010). In general, tropical production is constrained by satellite and sonde measurements of O3, while upward adjustments in midlatitude production have been applied to match in situ aircraft data (Martin et al., 2002b; Martin et al., 2006). The contention of Huntrieser et al. (2008) that higher midlatitude PEs are related to longer midlatitude strokes was based, in part, on speculation that the stronger wind shear in that region generates longer flashes. However, this hypothesis is contradicted by the association between stronger wind shear and stronger updrafts (e.g., Markowski & Richardson, 2011). These updrafts yield more frequent, but smaller, less productive flashes, as noted above. The midlatitude PE of the present study does not differ significantly from the tropical value of 170 ± 100 mol/fl from Allen et al. (2019), given the large uncertainties in the two results. The two are comparable since both analyses were based on the relationship between $V_{LNOx}^*$ and WWLLN flashes, although the method of estimating PE from the relationship differed. Marais et al. (2018) also found no significant difference between middle and tropical latitudes and noted that the higher GEOS-Chem midlatitude PE of 500 mol/fl overestimated observed OMI NO2.

5.3. Estimate of Uncertainty in the Average PE

In general, PE estimates are strongly dependent on the methods used to process the data (see Appendix B). For this study, the sensitivity of the PE to algorithmic assumptions was used to quantify systematic errors, which are the main components of the error budget. Statistical errors in OMI and WWLLN data were found to be negligible, as in Bucsela et al. (2010). We discuss the error sources individually and combine them to obtain a net uncertainty in the mean PE.

5.3.1. Stratosphere

Stratospheric NO2 is the largest component of the total NO2 column, constituting ~95% of the NO2 vertical column for all grid boxes in the data domain and ~85% for boxes with >5,000 fl/hr. As such, it is a
significant, though not the largest, contributor to the total error budget, as described below. Zonal smoothing of the NASA OMI NO2 stratosphere mitigates aliasing of parts of the tropospheric signal into the stratosphere (Allen et al., 2019; Beirle et al., 2016; Bucsela et al., 2013). Although the smoothing does alias stratospheric features that depart from the zonal mean into the troposphere, these departures have a mean value of ~0 and do not bias the average PE. Figure 12 shows mean $V_{\text{init}}$ and mean stratospheric NO2 with and without zonal smoothing as a function of flash rate. $V_{\text{init}}$ increases by $\sim 0.5 \times 10^{15}$ molecules/cm$^2$ as the flash rate increases from ~0 to ~12,000 fl/hr. Over the same range, $V_{\text{StratZonal}}$ increases slightly by $0.04 \times 10^{15}$. However, $V_{\text{strat}}$ shows a larger increase of $0.15 \times 10^{15}$ as the flash rate increases due to stratospheric aliasing of LNO2. The difference in the mean values of $V_{\text{strat}}$ for boxes with a flash rate of zero, which are used for the background estimate, and the values in flashing boxes introduce a low bias in LNOx and PE for the unsmoothed stratosphere. The PE derived from the latter is 120 mol/ft, which is ~30% lower than that derived from a zonally smoothed stratosphere.

To test the effects of stratospheric column errors, a uniform bias of ±2 × $10^{14}$ cm$^2$ was applied to the zonally smoothed stratosphere. The magnitude of this bias is in line with previous stratospheric NO2 uncertainty estimates (Boersma et al., 2004, 2007; Bucsela et al., 2006). It is twice the error assumed by Allen et al. (2019) and Bucsela et al. (2010) in the tropics, where stratospheric columns are approximately half those at midlatitude, and the absolute uncertainty is assumed to be smaller. A ±2 × $10^{14}$ cm$^2$ error is 5–10% of the total midlatitude NO2 column, ~90% of which is stratospheric. However, its effect on PE is only ±14% since it is uniform and is partially canceled in the background subtraction. Without background subtraction, the stratospheric bias would have a >90% effect on PE. The stratospheric component of the PE error is taken to be ±15% (see also Allen et al., 2019).

5.3.2. Transport and Chemistry Effects

In this study, all LNOx from lightning in the 1-hr integration period before the 13:30 LT OMI overpass is assumed to be accounted for in the retrieval and full correction made for contamination by ambient NOx. The fact that OMI measurements are made well before the late-afternoon convective peak complicates the retrieval by decreasing the component of fresh LNOx in the total NOx signal. Derived LNOx amounts are affected by errors in estimates of lofted pollution, LNOx lifetime, advection, and background. The 3-hr lifetime for LNOx in near-field of convection (Nault et al., 2017) is shorter than previous estimates of approximately two to eight days (Jaegle et al., 1998; Martin et al., 2007; Schumann & Huntrieser, 2007), which would make chemical loss negligible. B. Nault (private communication) gives an uncertainty range in his lifetime of 2 to 12 hr depending on the proximity to deep convection. In a 1-hr window, this introduces a PE uncertainty of ±10%. For boundary layer contamination, we assume a possible range of 10–20% for lofted pollution (DeCaria et al., 2000, 2005), which brackets our 15% downward adjustment in PE by ±5%.

Advection of LNOx from flashing boxes could result in a negative PE bias. If upper tropospheric wind speeds are ~16 m/s (a reasonable value for the midlatitudes in summer), a simple calculation shows that approximately 30% of the LNOx would be advected out of the box during the hour before measurement, with a reduction in PE by the same amount. The net loss would be lower if advection into the boxes from upwind sources compensates for some of the loss. The amount of this loss may also be estimated by comparing PEs derived from flashes over 1- and 2-hr intervals. Assuming an accurate NOx, higher flash rates over a longer window should be offset by a smaller background, with minimal net effect on PE. It was found that the PE based on 2-hr flashes is 15% smaller than the PE derived from 1-hr flashes. We attribute this difference mainly to advection. Overall, we estimate advection introduces a potential negative bias in PE of ~20% and assume an uncertainty of ±10%.
The mean midlatitude background estimated from nonflashing boxes is 55 ± 10% of total LNOx*, where the uncertainty is the standard deviation of interannual variability. This is ~3 times the a priori 18 ± 15% background of Pickering et al. (2016) in the Gulf of Mexico. The ±10% background error here propagates as a PE uncertainty of ±15%. Background days not only lack flashes during the 1-hr flash-integration window but also contain relatively few flashes during preceding hours. As such, even though they meet the CRF and OCP criteria used in this study, they may represent a less convectively active environment than that of flashing boxes, which are more likely to have flashes preceding the integration window. This may introduce a bias in the background, and therefore PE, although the effect is difficult to quantify. We assign an uncertainty of ±20%, which is less than the ~30% uncertainty obtained by Allen et al. (2019), who based their value on the sensitivity of PE to uncertainties in the y intercept of their regression-based estimate of PE.

### 5.3.3. NO₂ Profiles

Retrieved LNOx* is inversely proportional to the air mass factor $A_{\text{LNOx}}$ in equation (1). Beirle et al. (2009, 2010) estimated AMFs for a variety of storm environments and found that a fixed average AMF of 0.46 was adequate for their LNOx retrievals from Scanning Imaging Absorption Cartography NO₂ data. We find this value reasonable, since substitution of the Beirle et al. AMF decreased the present PE by only 13%. The AMF is computed with GMI a priori NO₂ and NO₃ profile shapes from days with the third largest LNOx column in a given month but is relatively insensitive to the rank within the month. Using the first or tenth largest columns changed the PE by only ~5%. Midlatitude simulations put 10–15% of LNOx at altitudes below 600 hPa, ~20% below 500 hPa, and ~30% below 400 hPa. Between OCP thresholds of 400 and 600 hPa, the average PE varies by ±12%. We adopt a net PE uncertainty of ±15% due to errors in profile shape and OCP threshold.

The partitioning of NOₓ into NO and NO₂ in GMI/GEOS-Chem, which affects the AMF, has been re-examined recently by Travis et al. (2016) and Silvern et al. (2018). They found GEOS-Chem NO/NO₂ ratios near and above 10 km to be approximately a factor of 2 larger than those of in situ measurements from the SEAC4RS campaign. Travis et al. (2016) attribute this to model underestimation of NO₂ and RO₂, but Silvern et al. (2018) suggest that their required model adjustments of peroxy radicals are inconsistent with observations. Instead, Silvern et al. (2018) posit a combination of model errors in the NO₂-to-NO photolysis rate and the NO + O₃ reaction rate, $k_1$, along with possible bias in the in situ data due to a neglected labile NO₃ reservoir. They noted that a $k_1$ increase of a factor of 1.4 and a photolysis rate decrease of 20% reduced the model NO/NO₂ by ~40%. The net effect is a ~28% reduction in NOₓ/NO₂ and hence an increase in $A_{\text{LNOx}}$ with a corresponding 28% decrease in PE. However, given that their factor of 1.4 represents an ~2σ change in $k_1$, that there are also possible significant measurement interferences, and that Silvern et al. and Travis et al. are relatively recent studies, we account for the potential error as a 20% high bias with a ±15% uncertainty (see also Allen et al., 2019).

### 5.3.4. WWLLN DE

The WWLLN flash counts are the largest source of PE uncertainty. Pickering et al. (2016) estimated a ±30% uncertainty in their WWLLN counts for the Gulf of Mexico, based on two independent schemes for estimating the DE, which differed by 25–30%. Their DEs were ~10–25% in the years 2007–2011. Citing North Alabama Lightning Mapping Array (Koshak et al., 2004) data, Allen et al. (2019) questioned whether the Pickering et al. ±30% was too low, given the small size of the Gulf region and the temporal variation of their DEs. In their tropical study, Allen et al. assigned uncertainties to the WWLLN data proportional to area-size and WWLLN DE. Adopted uncertainty values were ±25% for the tropics as a whole and approximately twice that value for subregions that included the tropical Americas, Africa, and the Pacific, as well as the Gulf of Mexico. The mean area-weighted DE for the tropics was ~13%, and the entire geographic area covered was $2.6 \times 10^8$ km². In the present study, the mean area-weighted DE is ~6% over our total geographic area of $0.6 \times 10^8$ km². Based on these considerations, we assign a conservative uncertainty of ±45% to our WWLLN flash counts.

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**Table 2**

*Potential Biases and Errors in the Average PE Estimate*

| Error source                        | Bias ± error (%) | Bias ± error (mol/l) |
|-------------------------------------|------------------|----------------------|
| Stratosphere                        | 0 ± 15           | 0 ± 27               |
| NO₂ lifetime                        | 0 ± 15           | 0 ± 18               |
| Lofted pollution                    | 0 ± 5            | 0 ± 9                |
| Advection loss                      | −20 ± 10         | −36 ± 18             |
| Tropospheric background             | 0 ± 20           | 0 ± 36               |
| OCP and below-cloud LNOₓ            | 0 ± 15           | 0 ± 27               |
| [NO]/[NO₂] ratio                    | 20 ± 15          | 36 ± 27              |
| WWLLN detection efficiency          | 0 ± 45           | 0 ± 81               |
| NO₂ slant columns                   | 0 ± 5            | 0 ± 9                |
| Net PE bias and uncertainty         | 0 ± 58           | 0 ± 100              |

---
5.3.5. Net Uncertainty
The net uncertainty in average PE combines the major sources of systematic error described above and summarized in Table 2. An additional uncertainty of ±5% is included to account for errors in the NO₂ slant columns (Allen et al., 2019; Krotkov et al., 2017; Zara et al., 2017). The biases are defined as the change in PE resulting from neglect of an error source. Net positive and negative biases turned out to be equal in magnitude and so have no net effect on average PE. Following Bucsela et al. (2010), Pickering et al. (2016), and Allen et al. (2019), errors are added in quadrature. The resulting uncertainty is ±58%.

6. Conclusions
The production efficiency of LNOₓ and its apparent dependence on flash rate have been explored with OMI NO₂ and WWLLN lightning measurements. We obtain an average summertime northern midlatitude PE of 180 ± 100 mol/flash. The midlatitude data set and algorithm used here are comparable to those of the tropical study of Allen et al. (2019) and the Gulf of Mexico study of Pickering et al. (2016). Both were based on OMI and WWLLN data but employed significantly different approaches. The Allen et al. tropical results do not provide compelling evidence of a systematic difference in PE between the tropics and midlatitudes, since the error bars in theirs and the present study have significant overlap. This conclusion was also reached by Marais et al. (2018), who obtained a relatively large PE of 280 mol/flash. However, their use of climatological NO₂ data from cloud slicing and an OTD/LIS lightning climatology in constraining the GEOS-Chem model differs from the present study and complicates direct comparisons with our results. Pickering et al.’s smaller 80 mol/flash is also attributable to algorithmic differences, particularly involving tropospheric background estimation, flash-rate threshold, and NOₓ lifetime. Their high flash-rate threshold and that of Beirle et al. (2010) are consistent with the small or difficult-to-measure PEs found in those studies, given the strong inverse relationship between PE and flash rate that we have shown here.

We find an approximate power law relationship between the rates of LNOₓ production and flashes, corresponding to a PE that decreases by an order of magnitude when flash rate is increased by ~2 orders of magnitude. A slightly weaker dependence of PE on flash rate is seen over convective clouds having an OCP < 400 hPa. A possible mechanism for the decrease in PE with flash rate may be inferred from several LMA studies showing that flash size and flash rate are also inversely correlated (e.g., Bruning & Thomas, 2015). The flash-rate dependence implies a need for caution when extrapolating PE values from limited data sets to estimate global LNOₓ production. For such estimates, flash rate and size distributions must be taken into account. However, if northern midlatitude distributions in the boreal summer are representative of those globally, then our average estimate of 180 ± 100 mol/flash is equivalent to 3.5 ± 2.0 Tg N/year LNOₓ, or roughly 16% of total global NOₓ production.

Future satellite missions with improved instrumentation and temporal coverage will help verify and refine the present PE. In particular, the Tropospheric Monitoring Instrument is providing NOₓ measurements unaffected by OMI’s row anomaly and at higher spatial resolution (3.5 × 7 km²) from low Earth orbit (Veefkind et al., 2012). Geostationary instruments, with their measurements spanning the afternoon diurnal peak of convection, will likely prove revolutionary in answering questions raised in previous LNOₓ studies. The Tropospheric Emissions: Monitoring of Pollution instrument (Zoogman et al., 2017) and Geostationary Environment Monitoring Spectrometer (Kim, 2012) will be such instruments. Their data can be combined with continuous DE-adjusted flash counts from the GOES-16 and GOES-17 Geostationary Lightning Mapper instruments (Goodman et al., 2013).

Appendix A: WWLLN DE Estimation
The detection efficiency (DE) of WWLLN strokes with respect to v2.3 OTD/LIS flashes (Cecil et al., 2014) was calculated using WWLLN data for 2007–2014 and used to adjust the WWLLN strokes so that the mean monthly flash rate at each grid box over the 2007–2014 period matches the OTD/LIS climatology; however, only WWLLN data for 2007 to 2011 were used in the estimation of LNOₓ PE.
1. WWLLN stroke data for individual days are partitioned into 15-min time periods and aggregated onto a 2° latitude × 2.5° longitude grid.
2. The gridded stroke data are smoothed temporally and spatially via the applications of a running 31-day average, a 3-hr average, and a 3-point north-south and east-west boxcar smoother.
3. The smoothed WWLLN strokes are averaged over 85 12-month periods with starting dates between January 2007 and January 2014. The annually averaged flash rates from the resulting 85-member time series are then divided by the v2.3 OTD/LIS climatological annual flash rate to obtain annual adjustment factors for each grid box.
4. Initial monthly adjustment factors are obtained by averaging annual scaling factors from the 12 annual periods that contain the month of interest. For example, the July 2011 scaling factor is obtained by averaging scaling factors from 12 periods beginning with the August 2010 to July 2011 period and ending with the July 2011 to June 2012 period. Effectively, the adjustment factors are weighted averages of a 23-month period with flashes from the month of interest having a weighting of 12, flashes from the month before and after the month of interest having a weighting of 11, flashes from months two months away from the month of interest having a weighting of 10, and so forth.
5. The month-specific adjustment factors are smoothed via the 3 times application of a seven-point boxcar average and interpolated onto a 1° × 1° global grid.
6. The month-specific adjustment factors are applied to the raw WWLLN flashes and the total flash rate for 2007–2014 is determined and compared to the v2.3 OTD/LIS climatology at each grid box. The adjustment factors are then adjusted to ensure that the mean monthly average flash rate over the 2007–2014 time period at each grid box matches the OTD/LIS climatological flash rate.

The updated month-specific adjustment factors are applied to the raw WWLLN strokes and the total flash rate for 2007–2014 is determined and compared to the bihourly OTD/LIS Low Resolution Annual Diurnal Climatology (Cecil et al., 2014) after it is interpolated from its original 2.5° × 2.5° UTC grid onto a 1° × 1° hourly grid as a function of LT. The adjustment factors are then modified to ensure that the mean hourly flash rate over the 2007–2014 period at each grid box matches the diurnal climatology.

Appendix B: Algorithm Comparisons

We examined methods used in other OMI-based studies to illustrate the sensitivity of derived PEs to algorithmic assumptions, even for comparable data sets. We discuss the Gulf of Mexico investigation of Pickering et al. (2016), the tropical case study of Bucsela et al. (2010), and the climatological study of Marais et al. (2018).

Pickering et al. (2016) used both regression and summation methods to obtain the much smaller mean PE of 80 ± 45 mol/ft over the Gulf of Mexico, and 84 mol/ft estimated by summation only. We applied the present algorithm to the Gulf region and obtained 148 mol/ft. Beginning with this value, elements of the Pickering et al. algorithm that differed with those of the present study were then applied sequentially to demonstrate their relative impacts, expressed as multiplicative factors. Values greater (less) than 1 mean that the sequential change resulted in an increase (decrease) in PE. The differences are (1) NASA OMI NO2 v2.1 instead of v3.0 (1.04; see Krotkov et al., 2017), (2) WWLLN flash version of Pickering et al. instead of the Allen et al. update (1.48), (3) a 3-hr flash window instead of 1 hr (0.65), (4) a three-day LNOx, lifetime instead of 3 hr (0.62), (5) an unsmoothed stratosphere (0.80), (6) an 18% tropospheric background instead of the present OMI-data-based approach (3.00), and (7) a 1,000-fl/hr threshold (0.39). The net effect is a combined factor of 0.568. Multiplied by the initial 148 mol/ft derived with the present method, it yields a PE of 86 mol/ft, which is approximately the Pickering et al. summation value.

Bucsela et al. (2010) examined four convective systems during the Tropical Composition, Cloud and Climate Coupling Experiment. Estimated PEs from each ranged from 87 to 246 mol/ft, with a mean value and uncertainty of 174 ± 219 mol/ft. Their flash counts were obtained from WWLLN on three of the days and Costa Rica Lightning Detection Network on one, with NO2 data from the v1.0 NASA OMI NO2 standard product. The latter included tropospheric AMFs and a wave-2 stratosphere, modified for the purpose of their study. The $A_{\text{LNOx}}$ was based on a measured composite LNO2 profile and GMI photolysis ratios. As in the present study, tropospheric backgrounds were estimated from convection-free days during the experiment. However, the outflow regions analyzed were not restricted to those with high cloud fractions, and this...
potentially compromised the accuracy of their background estimate. Their average tropical PE of 174 mol/fl is similar to the present midlatitude value, but the uncertainty is large, and it is based on a limited number of events.

In their LNOx study, Marais et al. (2018) used climatological NO2 and lightning data to derive a PE of 280 ± 80 mol/fl. Seasonal mean OMI LNOx columns from the v3.0 NASA OMI standard product were obtained by cloud slicing (Choi et al., 2014; Ziemke et al., 2001), with an OCP range of 280–450 hPa and no explicit adjustment for LNO2 below OCP. Since climatological data represent ambient NOx, accumulated from multiyear lightning activity, no background subtraction is used. The 2006–2008 OMI data were divided into geographic regions 20° × 32° in longitude and latitude and compared with a lightning climatology from OTD/LIS, which was also used in the present study to calibrate the WWLLN counts. The PE was estimated by constraining the LNOx source strength in GEOS-Chem to best fit the OMI cloud-sliced NO2 observations. Unlike the present study, they made no correction for a model discrepancy in the NO/NO2 ratio relative to aircraft data (see section 5.3.3), which could partially account for their relatively high PE. Because of the above differences, comparison of our mean PE with theirs is more challenging than with Pickering et al. (2016).

The recent study of Allen et al. (2019) in the tropics is also similar to the present one. Allen et al. compared OMI LNOx data (OMI LNOx* amounts measured during five austral and boreal summers with WWLLN flashes counted 1–6 hr before OMI overpass. The flashes and LNOx* were processed with the same methods used here. However, their average PE of 170 ± 100 mol/fl was estimated by linear regression, rather than by the present summation method of dividing total LNOx by total flashes. In their approach, the line slope and intercept are the PE and tropospheric background, respectively. Analysis in separate geographic areas revealed higher PEs where flash rates were lower, as well as a larger mean PE over marine relative to continental regions. Their flash-rate dependence is consistent with the present study, and their mean tropical PE is statistically equal to our midlatitude value of 180 ± 100 mol/fl.

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