

Lightning NO_x and Impacts on Air Quality

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Abstract Lightning generates relatively large but uncertain quantities of nitrogen oxides (NO_x), critical precursors for ozone and hydroxyl radical (OH), the primary tropospheric oxidants. Lightning NO_x strongly influences background ozone and OH due to high ozone production efficiencies in the free troposphere, effecting small but non-negligible contributions to surface pollutant concentrations. Lightning globally contributes 3–4 ppbv of simulated annual-mean policy-relevant background (PRB) surface ozone, comprised of local, regional, and hemispheric components, and up to 18 ppbv during individual events. Feedbacks via methane may counter some of these effects on decadal time scales. Lightning contributes ~1 % to annual-mean surface particulate matter, as a direct precursor and by promoting faster oxidation of other precursors. Lightning also ignites wildfires and contributes to nitrogen deposition. Urban pollution influences lightning itself, with implications for regional lightning-NO_x production and feedbacks on downwind surface pollution. How lightning emissions will change in a warming world remains uncertain.

Keywords Lightning · Reactive nitrogen oxides · Air quality · Policy-relevant background

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Introduction

Lightning strongly impacts the composition of trace gases in the troposphere. The extreme temperatures achieved inside a lightning flash channel generate relatively large quantities of reactive nitrogen oxides (NO_x ≡ NO + NO₂), which in the presence of sunlight, water vapor, and CO or volatile organic compounds (VOCs), drive the production of ozone (O₃) and hydroxyl radical (OH), the principal tropospheric oxidants [24, 75, 90]. Ozone itself is a biological irritant and pollutant, annually causing an estimated 140–900 k premature deaths and 11–18 billion USD in crop damage worldwide [8, 147]. The tropospheric oxidants collectively guide the atmospheric evolution, distribution, and removal of many trace chemical species of human interest, including air pollutants, oxidized nutrients, and reactive greenhouse gases [2, 99].

Although the global source of NO_x produced by lightning (2–8 Tg N yr⁻¹) [141] is several times smaller than that from present-day anthropogenic and biomass burning sources (~26 Tg N yr⁻¹) [77], lightning has a disproportionately stronger influence on tropospheric burdens of ozone and OH (Fig. 1), reflecting its emission directly into the free troposphere. Here, NO_x lifetimes are a few days instead of hours as at the surface, and ozone production efficiencies (OPE) per unit NO_x are an order of magnitude higher [45, 47, 118, 138, 161, 175, 190]. Variability in global mean OH is most sensitive to lightning of any major emission source because of the location and altitude of its emission, and because lightning lacks co-emission of reduced carbon species that would counteract the influence from NO_x [2, 52, 75, 88, 102, 103].

Despite the large influence of lightning on the background composition of the troposphere, it remains one of the least understood sources of NO_x [141]. Because its impacts on the surface are small relative to the free troposphere,

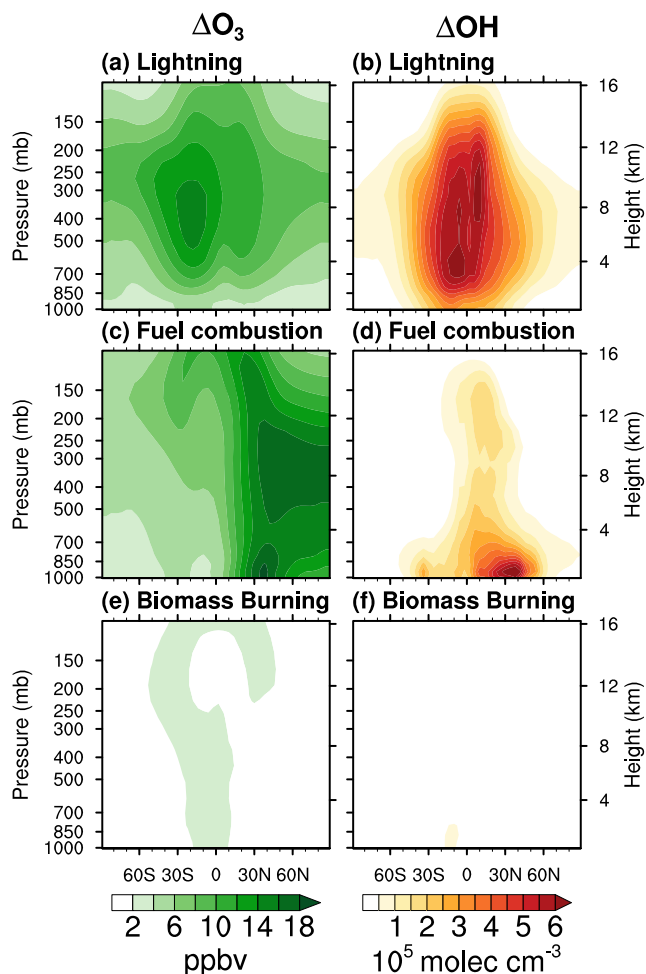


Fig. 1 Impact of different emission sources on zonal mean abundances of ozone (left column) and OH (right column) over the period 1998–2006. From top to bottom: contributions from lightning, anthropogenic fuel combustion, and biomass burning are estimated by differencing a full simulation with ones in which each source was individually removed. Simulations shown are described by Murray et al. [102]

most lightning NO_x studies have largely ignored the surface. Conversely, regional air quality models used for chemical forecasts and policy studies historically have not had any lightning NO_x emissions at all, and are only just beginning to implement the capability to do so [3, 66, 71, 168].

Focus, however, is shifting toward quantifying natural contributions to surface pollution. As air quality regulations tighten in the interest of public health, marginal improvements from anthropogenic pollution controls are becoming increasingly difficult to obtain. Furthermore, surface pollution levels are rising in some locations despite local emission reductions, such as ozone in the American West [28, 62, 112, 149]. This has shifted interest in recent years toward source apportionment of background concentrations of surface pollutants. The “policy-relevant background” (PRB) refers to baseline pollutant levels that are beyond the control

of policymakers in a given airshed. PRB may include contributions from natural processes (e.g., lightning, wildfires, volcanoes, terrestrial and marine biosphere, stratospheric intrusions), sectors unregulated for air quality (e.g., agriculture, methane), and the transport of upwind anthropogenic pollution and precursors. Estimates of North American Background (NAB) ozone in the USA range from 14 to 50 ppbv, or 20–70 % of the current standard [39, 40, 85, 94, 188]. In practice, PRB must be estimated using model simulations. A recent comparison of NAB ozone in two independent models noted that inter-model differences over the southwestern USA during summer were greatly sensitive to differential treatments of the lightning NO_x source [40].

The purpose of this review is to summarize the processes by which lightning contributes to PRB levels of major pollutants in surface air, and to provide quantitative estimates wherever possible. The intent is to be global in scope, although much of the research to date has focused on the continental USA. The remaining sections are as follows: the “Production of Trace Gases by Lightning” section overviews how lightning produces NO_x and other trace gases in the troposphere, and its main uncertainties. The “Lightning Impacts on Surface Air Quality” section describes how lightning NO_x influences PRB levels of major pollutants. The “Impact of Surface Air Pollution on Lightning” section summarizes evidence that urban air pollution influences lightning flash rates and its implications for NO_x production and regional chemistry. The “Future Trends in Lightning and Air Quality” section provides a summary of how lightning and its impacts on surface air quality are expected to change in the coming century. Finally, the “Conclusions” section ends with recommendations.

Production of Trace Gases by Lightning

Lightning-NO Production and Characterization

The rapid heating and cooling within a lightning stroke channel converts otherwise stable N_2 and O_2 into relatively significant quantities of nitric oxide (NO), by the same process as high-temperature fuel combustion [186]. Other species are also produced by this process, including NO_2 , CO, and ozone, but in negligible quantities [24, 141, 169]. Because NO is quickly oxidized to NO_2 in the atmosphere, and both rapidly cycle with one another in sunlight, the source is usually referred to as lightning NO_x . Characterizing the source of NO_x from lightning requires knowledge of the spatial and temporal occurrence of lightning flashes, the amount of NO produced per flash, and the vertical distribution of its release.

The global lightning flash distribution is climatologically well constrained and shown in Fig. 2. Satellite observations

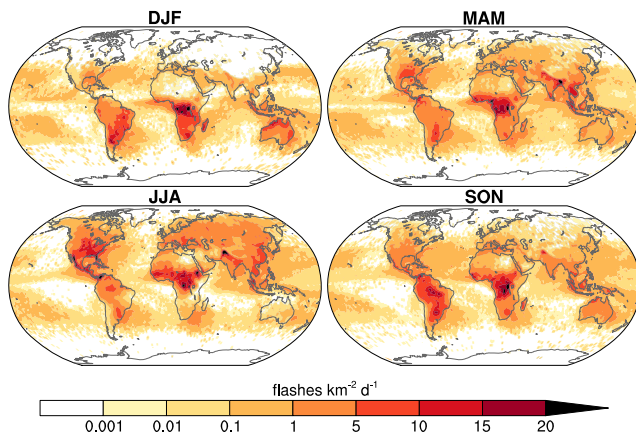


Fig. 2 Global distribution of lightning flash densities (flashes $\text{km}^{-2} \text{day}^{-1}$). Seasonal climatologies for 1995–2010 are shown from combined climatologies of the NASA OTD and LIS satellite instruments [23]

from low-earth orbit report a global mean flash rate of $46 \text{ flashes s}^{-1}$ for 1995–2010, with little interannual variability [23]. Most lightning activity occurs within deep-convective clouds in the tropics and subtropics, over land masses, during seasons of greatest solar insolation, and locally during late afternoon and early evening [26, 141]. Ground-based networks provide real-time coverage of lightning location and some physical properties (e.g., polarity, peak current) in many countries, and research arrays record comprehensive 4-D lightning physics at many sites. Most lightning flashes are inter- or intra-cloud (IC) rather than cloud-to-ground (CG) [125], although there is large spatial and temporal variability in the exact ratio [17, 74, 95, 120, 132, 171], with possible implications for lightning-NO production and chemistry in the boundary layer. However, flash rate parameterizations in global models generally show low skill in reproducing the lightning distribution

unless adjusted to the satellite observations; those parameterized upon mixed-phase particle collisions or cloud-ice flux perform the best [38, 61, 101, 162].

The amount of NO produced per flash remains a key uncertainty. NO production is sensitive to peak current, rate of energy dissipation, air density, channel length, and strokes per flash (multiplicity), all of which vary from flash to flash [29, 71, 73, 169]. An extensive literature review in 2007 reported a mean value of $250 \text{ mol NO flash}^{-1}$, and considered estimates between 33 and $660 \text{ mol NO flash}^{-1}$ to be plausible with observations of reactive nitrogen sinks [141]. Values subsequently reported in the literature are shown in Table 1 and range from 17 to $700 \text{ mol NO flash}^{-1}$ [14, 22, 29, 30, 55–58, 63, 71, 88, 93, 98, 110, 122]. The multi-model mean and standard deviation of lightning NO used by 13 global atmospheric chemistry models in a recent study is $5.3 \pm 2.1 \text{ Tg N yr}^{-1}$ [79], which corresponds to $260 \pm 100 \text{ mol NO flash}^{-1}$ using the observed global flash rate. There is disagreement as to whether CG and IC flashes produce the same amount of NO on average [29, 31, 32, 110] or whether CG flashes produce more [71]. Observations suggest that average NO-yields per flash are higher in the extratropics than tropics [55], although a recent data assimilation that simultaneously optimized the lightning-NO source to satellite observations of multiple chemical species reported large spatial heterogeneity [98]. Detailed process-based modeling of NO production on top of comprehensive observations of lightning physics find large variability in NO-yields per flash [71], highlighting the difficulty of extrapolating global mean NO yields from individual studies.

The vertical distribution of lightning NO is another key uncertainty. Care must be taken to distinguish between the vertical distribution of NO production and that following

Table 1 NO yields per flash in recent studies that have explicitly estimated lightning-NO yields using observational constraints

mol(NO) flash^{-1}	Method	Location	Reference
250 (33–600)	Literature review		[141]
67–201	Field study	Brazil	[55]
120–389	Field study	Oceania	[56]
90	Process-based model	Global	[29]
50	Satellite	Global	[14]
100–250	Satellite	Central America	[22]
500	Cloud-resolving chemistry model	North America	[110]
520	Chemical transport model	North America	[63]
71–188	Field study	West Africa	[57]
480	Chemical transport model	North America	[93]
500	Cloud-resolving chemistry model	Oceania	[30]
101 (82–171)	Process-based model	North America	[71]
310 (0–700+)	Chemical data assimilation	Global (Local)	[98]
246	Chemical transport model	Global	[88]
194 (71–533)	Field study	North America	[122]

the mixing and in-cloud chemistry of a convective event. The latter is more relevant for air quality models that are too coarse to explicitly resolve cloud physics. This distribution was originally thought to be bimodal, with maxima at both the surface and in the upper troposphere [119]. A more recent study reported unimodal distributions, with maxima at 5–7 km in midlatitude storms and 13–14 km over tropical land masses [110]. It is unknown how much geographic variability occurs in these distributions, but there is a fair amount of temporal variability over Northern Alabama [71].

Figure 3 shows the distribution of lightning NO_x emissions in global atmospheric chemistry models. Models

differ greatly in their meridional and vertical distributions. As the NO_x lifetime increases from hours in the boundary layer to days in the upper troposphere [161, 190], it is important to note that global lightning NO_x magnitudes estimated by constraining models to top-down constraints such as ozone sondes will be highly sensitive to the vertical distribution assumed.

Other Mechanisms

Hot-channel processes are not the only mechanism by which lightning produces trace gases in the troposphere. Corona

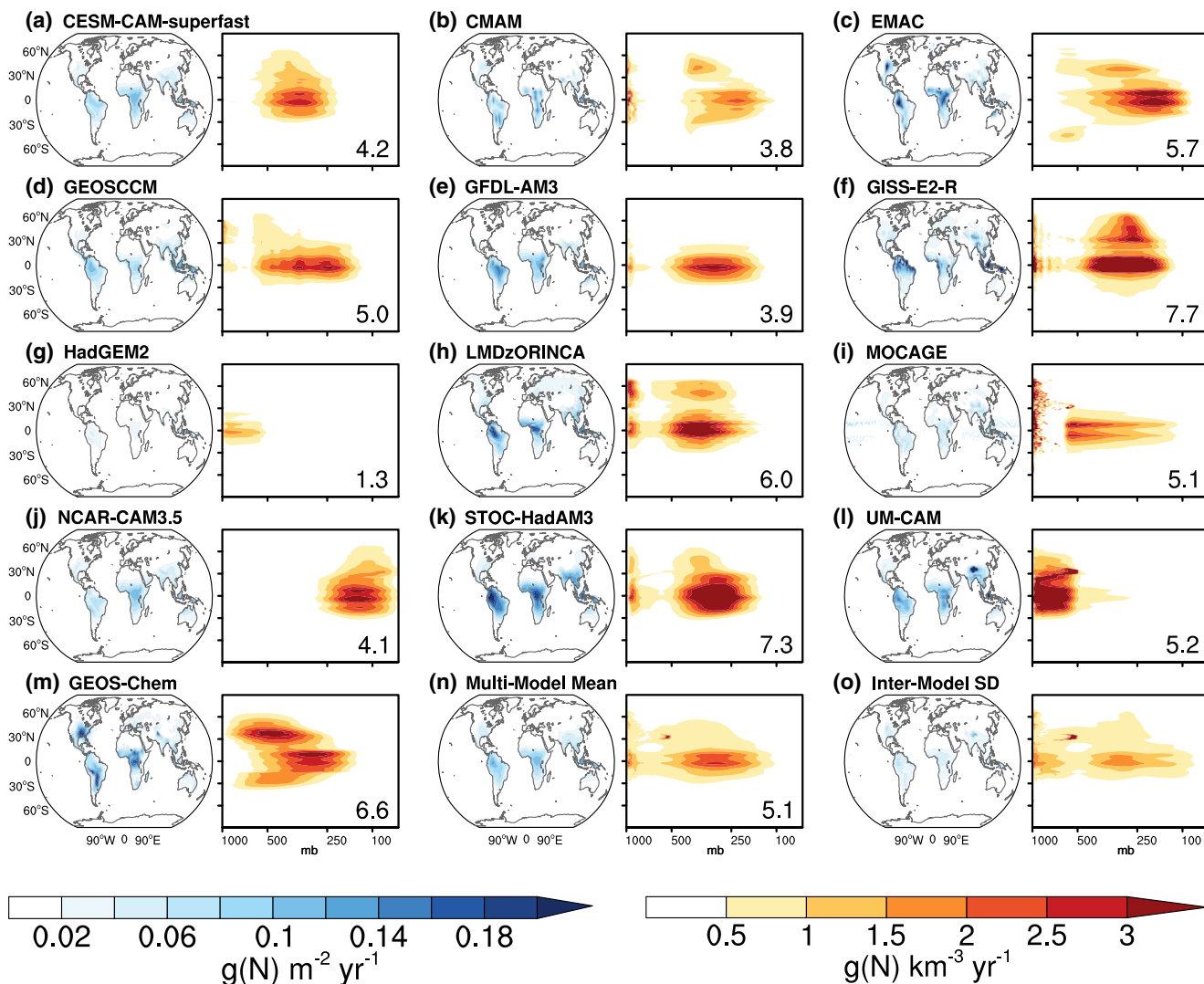


Fig. 3 Large differences in lightning NO_x emissions between global atmospheric chemistry models. Each panel shows two plots with latitude as a shared ordinate axis. The *left plot* shows the spatial distribution of the vertically integrated source in $\text{g(N) m}^{-2} \text{yr}^{-1}$. The *right plot* shows the zonal mean distribution as a function of altitude in $\text{g(N) km}^{-3} \text{yr}^{-1}$. The number reports the global mean source in Tg(N) yr^{-1} . Panels **a–f** show decadal climatologies archived by the

Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) for the 2000s [79]. Panel **m** shows emissions from GEOS-Chem using MERRA meteorology for 2004–2012 and constrained to the LIS/OTD climatology [101]. Panels **n** and **o** respectively show the multi-model mean and inter-model standard deviation of the thirteen models

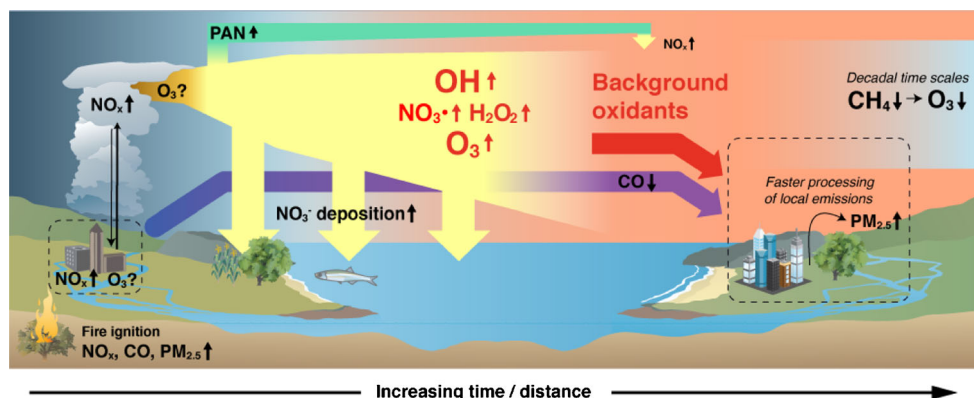
discharges in the vicinity of lightning flashes directly produce significant quantities of ozone and N_2O . Production of NO is negligible relative to that in the hot channel [50, 116, 130, 148, 184]. Field measurements indicate that the global source of ozone produced directly by lightning-induced corona discharges could range from 2.5 to 250 Tg $\text{O}_3 \text{ yr}^{-1}$, with best estimates near 100 Tg $\text{O}_3 \text{ yr}^{-1}$ [19, 97]. This is small compared to the total tropospheric chemical production term ($\sim 4900 \text{ Tg } \text{O}_3 \text{ yr}^{-1}$ [182]), but could be locally important near thunderstorms, and has been invoked to explain surface measurements of NO_x and ozone following thunderstorms [16].

Lightning is the dominant natural ignition source for wildfires, and fires generate trace gases and particles that have substantial downwind impacts on air quality, including NO_x , CO, and VOCs [82]. Lightning was responsible for 15 % of ignitions but 63 % of all area burned in the USA from 2001 to 2015 [107]; this fraction varies throughout the world and in time. Indirect impacts of lightning on air quality via fire emissions are beyond the scope of this review. However, it is noted that the relationship between lightning frequency, fire ignition, and emissions is not necessarily linear. Ignition and subsequent emissions depend on the frequency of CG flashes with continuing currents, local fuel load and moisture content, meteorology and orography, and active or passive fire suppression by humans [83, 117, 172].

Lightning Impacts on Surface Air Quality

Lightning influences air quality across a range of temporal and spatial scales, as summarized by Fig. 4. The following subsections outline the impacts by major pollutant. Figure 5 shows the mean impact of lightning on surface pollutants in the recent past using simulations of the GEOS-Chem model. Table 2 compares these results with the literature, and puts them in context with the impacts from anthropogenic and fire emissions.

Fig. 4 Schematic summarizing ways in which lightning may influence surface air quality, as described in the text. *Short vertical arrows* indicate whether the species or process increases (\uparrow), decreases (\downarrow), or could do either (?) in response to an increase in lightning activity



Nitrogen Dioxide

Trace gas measurements of surface air show relatively large enhancements of NO_x immediately following local thunderstorm activity. These may reflect direct production by lightning in the boundary layer or convective transport from within the cloud. Reported surface- NO_x enhancements associated with thunderstorms are scarce. Those that exist report peak values of about 40 ppbv over India, 25 ppbv over Taiwan, and 30 ppbv (mostly as NO_2) over northern Alabama, and persist from minutes during the day to a few hours at night [16, 64, 96, 114, 115, 165]. However, reported surface concentrations never exceed current health guidelines for short-term NO_2 exposure (1-h mean 100 ppbv [174]). Observations of NO_x in fresh convective outflows sampled from aircraft (e.g., up to ~ 6 ppbv NO_x during the Deep Convective Clouds & Chemistry (DC3) Experiment [106, 122]) are also beneath the exposure guidelines.

Ozone

Lightning NO_x has relatively small but nonetheless significant impacts on surface ozone across a wide range of spatial and temporal scales.

Local Impacts

Air masses most recently influenced by lightning are highly elevated in NO_x relative to their surroundings, which can drive either net ozone production or loss depending on the availability of CO, VOCs, and solar radiation. In air parcels with sufficiently high ratios of NO_x to CO+VOCs, NO reacts directly with and destroys ozone; this “ NO_x titration” of ozone also dominates at night when photochemical production disappears, but can photolyze back to ozone at sunrise if not lost prior. During daytime, if the ratio of NO_x to CO+VOCs is sufficiently low, net ozone production occurs. The efficiency of ozone production increases at lower NO_x concentrations [145].

A number of recent studies have reported changes in observed surface NO_x and ozone concentrations immediately

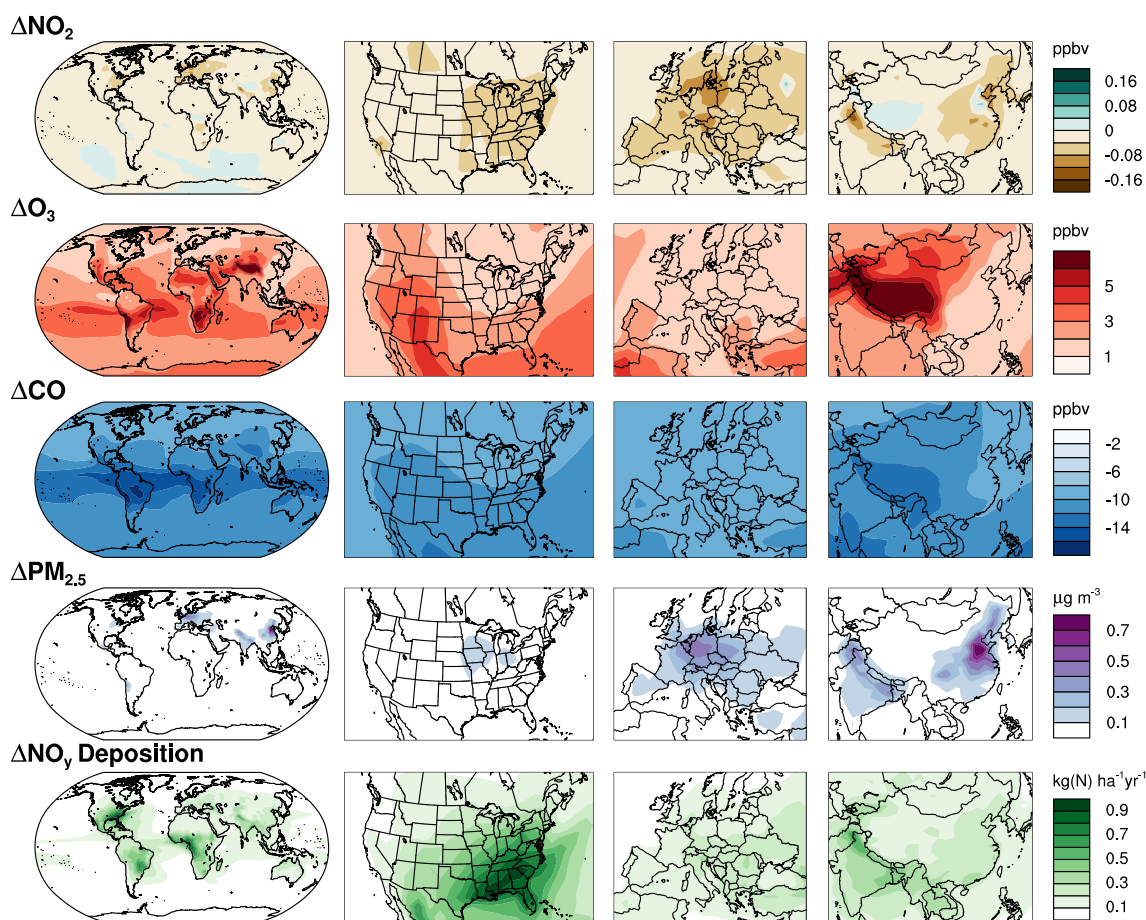


Fig. 5 Annual mean impact of lightning on surface pollution. The *left column* shows the global impact, while the *right three columns* show regional detail for North America, Europe, and South and East Asia. Simulations using version 9.02 of the GEOS-Chem model (<http://www.geos-chem.org>) are driven by meteorology from the MERRA reanalysis at 2° latitude by 2.5° longitude horizontal resolution for 2004–2012. The lightning contribution is estimated as the residual

between a full simulation and one without lightning NO_x . Lightning emissions are as described by Murray et al. [101], except over the USA where flash rates are prescribed from the National Lightning Detection Network (NLDN). Methane is prescribed from observations and not allowed to adjust, so these values reflect impacts on sub-decadal time scales (see the “Global” section)

following local thunderstorm activity. Surface NO_x was always enhanced, but the ozone response depended on the local photochemical regime. Thunderstorm- NO_x enhancements in surface air at Pune, India were sufficiently elevated above titration thresholds to drive ozone destruction in 17 % of 60 thunderstorms examined; thunderstorm- NO_x enhancements of 19 ± 10 ppbv were associated with ozone decreases of 6 ± 4 ppbv, whereas weaker NO_x enhancements of 2.2 ± 1.5 ppbv were associated with ozone increases of 10.4 ± 3.4 ppbv [114].

However, two studies report positive correlations between observed lightning flash rates and surface concentrations of NO_x and ozone over both Kolkata, India ($\Delta \text{O}_3 / \Delta \text{NO}_x = +0.6$ ppbv/ppbv) [96] and Taipei, Taiwan ($\Delta \text{O}_3 / \Delta \text{NO}_x = +1.3$ ppbv/ppbv) [64], suggesting that ozone production was always maintained over the megacities. Surface NO_x associated with thunderstorms has also

been observed to titrate ozone at nighttime, such as a 41 ppbv increase in NO_x associated with a 26 ppbv decrease in ozone during the evening of Aug 8, 2012 over Hyderabad, India [16, 165]. Marginally increasing the fraction of lightning NO_x released directly into the boundary layer in a regional air quality model of the contiguous USA led to increased NO_x titration of surface ozone. This was attributed to the propensity of lightning to occur when photolysis was limited in late afternoon, early evening, or under cloudy conditions [66].

Observations of additional surface trace gases and/or modeling will be necessary to assess the relative contributions of local boundary-layer chemistry to the observed ozone anomalies. These anomalies could also reflect convective transport of air masses that may have undergone in-cloud chemistry or are of remote origin (e.g., stratosphere,

Table 2 Contributions to surface air pollutants from lightning in regional and global models

Source/Metric	ΔO_3		$\Delta MDA8$		$\Delta PM_{2.5}$		ΔD_{NO_y}		ΔDN		Comp. ^a	Domain ^b	Averaging period	Model ^c	References
	ppbv	%	ppbv	%	$\frac{\mu g}{m^3}$	%	$\frac{kg(N)}{ha\ year}$	%	$\frac{kg(N)}{ha\ year}$	%					
Lightning															
Local Max			-6–46								R	NA	Jul.–Aug. 2004	C	[66]
Local Mean							28, 34				R	NA	JJA 2004	M	[33]
Local Mean					0–0.1						R	NA	2002	C	[70]
Local Mean	0–6		1.5–4.5				30				R	NA	JJA 2004, 2006	C	[3]
Regional Mean											R	NA	JJA 2006–2008	G	[187]
Local Mean	0–3										R	NA	Aug. 2006	C	[71]
Local Mean			0.5–10								R	NA	JJA 2006–2008	G	[188]
Hemispheric Mean	1	3.2			0.04	0.9					H	NH	2006	D	[185]
Local Mean		10–40									H	G	1990–1999	E	[45]
Global Mean	3.7	12			0.02	0.2	0.03	8.2	0.03	3.6	H	G	1998–2006	G	[102]
Pop.-Wghtd. ^d Mean	3.1	6.9			0.11	0.4	0.07	3.8	0.06	1.1	H	G	1998–2006	G	[102]
Crop-Wghtd. ^d Mean	3.3	7.8			0.08	0.4	0.08	5.0	0.07	1.8	H	G	1998–2006	G	[102]
Global Mean	2.9	10	3.2	11	0.02	0.3	0.12	12	0.13	5.2	H	G	2004–2012	G	This work
Local Mean	0.7–11	2–27	1–12	2–28	-0.03–0.88	0–6.8	0–1.01	0–55	0–1.02	0–30	H	G	2004–2012	G	This work
Pop.-Wghtd. ^d Mean	2.5	6.2	2.9	6.4	0.14	0.6	0.31	6.1	0.32	1.9	H	G	2004–2012	G	This work
Crop-Wghtd. ^d Mean	2.7	6.9	3.2	7.1	0.10	0.5	0.31	8.2	0.32	2.8	H	G	2004–2012	G	This work
Fires^e															
Hemispheric Mean	1.3	3.9			1.18	25					H	NH	2006	D	[185]
Local Mean		2–25									H	G	1990–1999	E	[45]
Local Global Mean	1.4	4.4			0.59	4.7	0.05	13	0.10	12	H	G	1998–2006	G	[102]
Pop.-Wghtd. ^d Mean	1.6	3.7			1.05	4.2	0.10	5.2	0.19	3.4	H	G	1998–2006	G	[102]
Crop-Wghtd. ^d Mean	1.9	4.4			1.32	6.4	0.12	7.6	0.24	6.0	H	G	1998–2006	G	[102]
Fuel Combustion^e															
Local Mean		25–80									H	G	1990–1999	E	[45]
Global Mean	8.1	26			1.09	8.7	0.29	70	0.57	65	H	G	1998–2006	G	[102]
Pop.-Wghtd. ^d Mean	17	40			9.08	36	1.62	84	4.90	87	H	G	1998–2006	G	[102]
Crop-Wghtd. ^d Mean	15	36			6.02	29	1.27	78	3.29	81	H	G	1998–2006	G	[102]

Absolute and percent of total contributions are shown for surface ozone (O₃), 8-h max daily ozone (MDA8), fine particulate matter (PM_{2.5}), oxidized N deposition (D_{NO_y}), and total N deposition (D_N). Where also available, fire and anthropogenic fuel contributions shown for comparison. Ranges indicate spatial spread

^aComponents of changes reflected in value: R = Local + Regional; H = Local + Regional + Hemispheric Background

^bDomain: G = Global; NH = Northern Hemisphere; NA = North America

^cModels: C = CMAQ; D = DEHM; E = E93/C; G = GEOS-Chem; M = MOZART4

^dPopulation and crop-area weights from the HYDE3.1 database for the year 2005 [69]

^eCombustion sources include impact of NO_x, CO, VOCs, SO₂, and particulates. Fires emit ~6 Tg(N) yr⁻¹ and fuel combustion ~17 Tg(N) yr⁻¹ [77]

urban), or are possibly enhanced due to direct production in corona discharges.

Regional

Lightning NO_x that detrains into the free troposphere has a lifetime of days, and is therefore able to influence regional background ozone concentrations.

The integrated impact of a lightning- NO_x plume will depend on its initial conditions. Initial titration of ozone in fresh lightning- NO_x plumes is more readily achieved in the free troposphere, as CO and VOC concentrations are typically lower than at the surface. However, the plume will eventually dilute and become ozone-producing with ever increasing OPE. A recent study that embedded a concentrated lightning-plume chemistry parameterization into a global chemical transport model found that the combined influence of NO_x titration and reduced OPE in high- NO_x plumes suppressed background ozone concentrations up to 8 % over lightning source regions [44]. However, that parameterization assumes lightning NO was always injected into clean background air. Deep convection can mix together air parcels of complex origins; the DC3 field campaign sampled convective outflow over the central USA influenced by lightning, lofted surface air, stratospheric intrusions, and smoke plumes [12, 59, 111, 140]. A case study of thunderstorms that ingested smoke during the campaign concluded that the anvil outflow, enriched in CO and VOCs from the wildfires and NO_x from lightning, was initially capable of substantial downwind ozone production, producing up to 14 ppbv of ozone over a period of 2 days in the plumes [5]. An earlier case study of a storm in the same region found ~ 10 ppbv of ozone produced in lightning-influenced plumes within 1 day [32].

Regional air quality models assess the sensitivity of downwind surface pollutants to emissions that occur within their own domain. Several studies have implemented lightning-NO production into the US EPA Community Multi-scale Air Quality Model (CMAQ), with flash rates prescribed from the continuous National Lightning Detection Network, but for different years and with different NO yields and vertical distributions [3, 66, 70, 71, 151, 167]. The inclusion of North American lightning NO_x only led to domain-wide average increases of ~ 2 ppbv in surface ozone; instantaneous contributions never exceeded 10 ppbv. Contributions to the policy-relevant maximum daily 8-h average (MDA8) surface ozone metric were usually under 4.5 ppbv, small relative to the current US standard (70 ppbv) or the total NAB (30–50 ppbv), which also includes contributions from hemispheric backgrounds, intercontinental transport, fires, and stratospheric intrusions [85, 94, 164]. However, two studies reported that the absolute lightning contribution to MDA8 decreased on days with the highest

ozone [3, 66]. Maximum lightning contributions to surface ozone were in the Southwest, followed by the Gulf Coast [3, 66, 70, 71]. Similar results were reported in a regional version of the GEOS-Chem model, but with slightly higher contributions from lightning to MDA8 surface ozone in the Southwest during summertime (6–8 ppbv mean; 17.9 ppbv max). Lightning contributed more to MDA8 surface ozone than wildfires or Californian anthropogenic emissions in much of the Southwest [188]. Summertime surface ozone in the Southwest is particularly sensitive to lightning because of high flash rates and transport pathways associated with the North American monsoon, sunny conditions conducive to photochemical production, and high boundary-layer heights that promote downward mixing of ozone to the surface [3, 188].

Intercontinental

A large fraction of NO_x exported from continental source regions is of lightning origin, even from regions heavily influenced by anthropogenic pollution [33, 54, 93]. Ozone in the free troposphere has a lifetime of months, allowing for hemispheric transport, and thereby allowing lightning NO_x to influence background ozone at the top of the boundary layer over downwind continents. Lightning NO_x also can enhance long-range transport of ozone by reinvigorating production in plumes that may have become depleted in NO_x , such as those from fires [48]. Lightning NO_x can react with lofted hydrocarbon-rich surface air to form peroxyacetylnitrate (PAN), which is extremely stable at the cold temperatures of the upper troposphere and may transported very long distances until descent and decomposition back into NO_x [100, 161]. Lightning contributes 40–80 % of PAN columns in the tropics and 20–40 % in the extra-tropics [41]. Furthermore, lightning NO_x strongly influences background abundances of the hydroxyl radical (OH) [52, 53, 75, 102, 103], the dominant tropospheric oxidant and primary sink for CO and VOCs, thereby decreasing inter-hemispheric transport of these ozone precursors.

Global or hemispheric model studies have examined the role of lightning on tropospheric ozone distributions, but do not generally report the impact on surface concentrations, which are small relative to free-tropospheric changes. The Danish Eulerian Hemispheric Model (DEHM) reports that lightning NO_x contributes up to 3 % of annual mean surface ozone concentration in the northern extratropics, and up to 20 % of the surface ozone concentrations in the tropics for the year 2006 [185]. The E39/C global chemistry-climate model reported that lightning NO_x contributes 10–15 % of zonal mean surface ozone for 1990–1999 in the northern extratropics, 15–30 % in the tropics, and 30–50 % in the southern extratropics, although that model employed a simplified tropospheric chemical mechanism

[45]. Table 2 reports global mean changes in surface ozone mixing ratios for 1998–2006 in the GEOS-Chem model [102], and for new simulations for 2004–2012 shown and described by Fig. 5. These values include both regional and hemispheric contributions. Lightning contributes 2–4 ppbv (10 %) to annual mean surface ozone in GEOS-Chem, although locally may contribute up to 11 ppbv or 27 %. The contribution decreases when weighted by population or crop area. Impacts on annual mean MDA8 values are slightly higher. The mean influence of lightning on ozone is less than anthropogenic fuel combustion but greater than biomass burning.

Global

The ultimate legacy of lightning on background ozone manifests on time scales well past the NO_x or ozone lifetimes by triggering important feedbacks in the coupled ozone-OH-CO-methane system [176]. Methane is primarily lost via reaction with OH in the tropical troposphere, with a present-day lifetime of 11.2 ± 1.3 yr against this sink [124], and therefore the methane loss rate is especially sensitive to lightning NO_x emissions [10, 52, 103]. A positive perturbation in lightning NO_x will initially drive increases in background ozone via reaction with the oxidation products of CO and non-methane VOCs, as described in the previous sections. However, the associated increase in OH will ultimately decrease methane on decadal time scales, as well as the source of CO from methane oxidation. As methane is well-mixed in the troposphere, this will drive global mean reductions in tropospheric ozone. In some places, the global methane-ozone decreases of a sustained lightning enhancement (e.g., due to climate change) could offset regional NO_x -ozone increases. Most of the model studies that examine the sensitivity of ozone to lightning NO_x have used prescribed methane concentrations, and therefore are only valid in the context of short-term impacts.

Carbon Monoxide

Lightning strongly influences the global mean OH burden, and therefore the CO lifetime. Global mean OH is particularly sensitive to lightning NO_x because of the location and altitude of its production and because lightning lacks co-emission of reduced carbon species that would counteract the NO_x influence [52, 75, 102, 103]. Therefore, lightning increases the CO loss rate, although this will be partially countered by increases in chemical production from VOC oxidation in the troposphere. No study in the literature has reported quantitative estimates of how lightning NO_x impacts the global or surface CO budget. Figure 5 shows lightning reducing CO by 9–18 ppbv (~ 10 %) in GEOS-Chem, and allowing methane to adjust would cause further

reductions. Lightning impacts on zonal mean background CO are probably larger in a local airshed than the impacts on chemical aging rates of concentrated CO plumes coming from remote fires or urban areas.

Particulate Matter

Particulate matter in the fine mode (less than $2.5 \mu\text{m}$ in diameter; $\text{PM}_{2.5}$) is associated with health risks at all levels [109]. Lightning NO_x impacts particulate pollution both directly as a nitrate aerosol precursor, and indirectly through perturbations to tropospheric oxidants.

Most lightning NO_x is eventually oxidized to nitric acid (HNO_3), which will condense into ammonium nitrate (NH_4NO_3) particles in the presence of excess ammonia (NH_3) following the neutralization of any sulfate aerosol particles [13, 49, 179]. NH_4NO_3 aerosol exists primarily in the fine mode, and is presently ~ 10 % (1.2–28 %) of observed annual mean $\text{PM}_{2.5}$ mass over northern midlatitude continents [189]. HNO_3 may also absorb onto mineral dust or sea salt particles to form fine- and coarse-mode nitrate aerosol [87, 139]. Nitrate aerosol may also be produced through heterogeneous reactions of gaseous nitrogen radicals such as NO_3 , N_2O_5 , and HNO_3 on aerosol surfaces [60], by replacing chloride in sea-salt aerosol with sufficiently low pH [67], and through nighttime oxidation of VOCs by NO_3 into low-volatility organic nitrates [42].

Changes in background oxidants driven by upwind NO_x changes can generate significant downwind increases in aerosol abundances over populated regions due to faster oxidation rates of SO_2 to sulfate and NO_x to nitrate [11, 86, 163]. Furthermore, lightning NO_x is an important precursor for gaseous nitrate radical (NO_3), the dominant nighttime oxidant [21]. Increasing NO_3 abundances increases the fraction of dimethylsulfide (DMS) from marine phytoplankton that is oxidized to SO_2 (the largest natural source of SO_2), and thereby sulfate aerosol [154]. The importance of nighttime oxidation of biogenic VOCs by NO_3 for secondary organic aerosol production is an active area of research [43].

An attribution study of annual mean surface $\text{PM}_{2.5}$ using the CMAQ regional model found North American lightning contributed up to $0.1 \mu\text{g m}^{-3}$ over the USA; greatest absolute contributions were in southern states, especially Florida [70]. A similar study of the northern hemisphere using the DEHM found lightning to contribute 1 % to hemispheric mean surface $\text{PM}_{2.5}$ ($0.04 \mu\text{g m}^{-3}$). However, lightning contributed up to 3 % over portions of North America, Southeast Asia, and West Africa, up to 10 % over Indonesia, and up to 20 % over the Amazon [185]. A study examining aircraft- NO_x impacts using the GEOS-Chem global model reported that source contributed up to $0.15 \mu\text{g m}^{-3}$ in annual mean surface $\text{PM}_{2.5}$ over China and Europe, primarily through perturbations to background oxidants

from cruise-altitude emissions; aircraft NO_x is globally 5–10 times smaller than lightning NO_x , but concentrated in the northern midlatitudes [11]. Lightning contributes less than 1 % to surface $\text{PM}_{2.5}$ in GEOS-Chem (Fig. 5 and Table 2). These contributions are small compared to current regulatory standards for annual mean $\text{PM}_{2.5}$, including those in the USA ($12 \mu\text{g m}^{-3}$) and China (non-urban: $15 \mu\text{g m}^{-3}$, urban: $35 \mu\text{g m}^{-3}$).

Nitrogen Deposition

All nitrogen fixed by lightning will ultimately deposit back to the surface via wet or dry processes, primarily in the form of nitrate [78]. Nitrogen is a critical nutrient, but excess oxidized and reduced nitrogen species contribute to ecosystem acidification, eutrophication, and biodiversity reduction [18, 27, 46]. Lightning NO_x has a more distributed impact on deposition than surface NO_x sources, due to its emission into the free troposphere where lifetimes are longer and winds are stronger [51, 143]. However, lightning has traditionally not been included in studies of adverse consequences of nitrogen deposition [159].

Attribution studies that have included and isolated lightning impacts have generally focused on the continental USA using regional models. Lightning contributed 28–34 % of oxidized nitrogen deposition in MOZART-4 in summer 2004, depending on assumptions about the lightning source magnitude [33]. This is consistent with a study using CMAQ that reports lightning contributed 30 % to oxidized nitrogen deposition and 9 % to total N deposition in summer 2006, with most of the flux occurring in eastern states. Including lightning NO_x eliminated a large bias in the simulated nitrate deposition fluxes versus observations [3, 6]. Regional simulations of GEOS-Chem found all natural sources (lightning, soil microbes, and fires) contribute 10 % to annual total N deposition in the eastern USA for 2006–2008, but up to 40 % in the southwest due to lightning [187].

Additional studies have used inverse modeling to attribute relative source contributions to N deposited at specific locations. The reported contributions from lightning are: 0–7% of total N deposited and 1–17 % of total oxidized nitrogen (NO_y) across eight biodiversity hotspots worldwide (although with only 2.3 Tg N yr^{-1} of lightning NO_x) [113], 0.5 and 4 % of total N deposited, respectively, over the Yellow and South China Seas [192], and 1–22 % of total N deposited at eight US National Parks [84].

Lightning globally contributes 12 % of NO_y deposition and 5 % total N deposition in GEOS-Chem for 2004–2012 (Table 2). Greatest absolute contributions are to the southeastern USA, the Cerrado of Brazil, and equatorial western Africa (Fig. 5). Greatest relative contributions are to the

remote tropical oceans (up to 55 % of NO_y and 30 % of total N deposition).

Impact of Surface Air Pollution on Lightning

Observational evidence suggests that aerosol pollution influences the frequency and physical properties of lightning, with implications for regional lightning NO_x production and downwind photochemistry.

Ground-based networks show enhanced lightning frequencies immediately downwind of urban areas [152]. Statistically significant increases in CG strikes have been observed downwind of large cities in the central and southeastern USA [108, 156, 173], southern Europe [7, 36, 131], southeastern Brazil [105], South Korea [65], central and northeastern India [25, 76, 96], and Taiwan [64]. In addition, the percentage of positive-polarity flashes also generally decreases. Urbanization has also been implicated in driving temporal trends and variability in lightning observations over southeastern Brazil [121] and central and southeastern China [180, 181].

Urban lightning enhancements are thought to result from a combination of urban atmospheric thermodynamics, surface properties, and aerosol pollution [173]. The presumed influence of aerosol particles is to reduce cloud droplet sizes, leading to reduced warm rain coalescence, and therefore an enhancement of cloud water reaching the mixed-phase region. Collisions between hail, graupel, and ice particles in the presence of supercooled liquid water lead to charge separation and accumulation [137, 177]. Cloud-resolving models predict enhanced electrification when aerosol loads increase [92, 144, 160, 191]. However, the relationship between aerosol loading and flash rate is observed to be nonlinear, and too much aerosol will suppress lightning by stabilizing the atmospheric column through radiative effects [4, 34, 181].

Aerosol lightning enhancements are readily observed in rural and remote regions; frequencies are enhanced downwind of large point sources and highways [155, 158], wildfires [4, 80, 81, 91, 104, 150], and volcanic eruptions [183]. Nevertheless, a main uncertainty in isolating an aerosol effect on flash rates is the complicating role that variability in atmospheric thermodynamics also plays in influencing lightning [1, 68, 157]. In urban areas, it is particularly difficult to isolate any aerosol effects from urban land cover influences. Several recent studies have reported modest but significant weekday enhancements in urban flash rates throughout the world, correlated with aerosol pollution [15, 35, 36, 146, 153]. The existence of weekly cycles are controversial [135], but could be used to isolate the impact of weekday pollution enhancements on lightning over base-

line contributions from landcover and pollution. In addition, observations indicate that the CG flash rate decreased while the fraction of positive-polarity flashes increased over the continental USA from 2003 to 2012 [72], all while aerosol concentrations decreased [123]. The lightning trends are expected if the urban-lightning effect was driven at least in part by aerosol abundances.

The NO_x-yield per flash and its vertical distribution are sensitive to channel location and current (see the “[Production of Trace Gases by Lightning](#)” section). The negative-polarity CG flashes favored downwind of urban areas tend to have lower peak currents, and only 30–50 % have continuing currents [9, 129]. Therefore, urban-influenced lightning may have lower NO_x-yields per flash. Therefore, aerosol pollution may alter the magnitude and location of the lightning NO source, with consequences on downwind surface ozone (“[Ozone](#)” section), aerosols (“[Particulate Matter](#)” section), and potentially even back onto lightning.

Future Trends in Lightning and Air Quality

How lightning may respond to a warming climate in the coming decades remains uncertain. Studies show strong sensitivity of local lightning activity to changes in surface temperature on diurnal through interannual time scales [178]. Lightning activity increases over land masses during warm phases of the El Niño-Southern Oscillation (ENSO), despite decreased convective frequency in some regions [136]. Drier tropics in a warmer future may lead to increases in extreme convection, and therefore enhanced lightning [126]. However, there is no guarantee that temperature alone will induce long-term changes because of other processes that can influence deep convection [127, 178]. For example, models expect convective available potential energy (CAPE) to increase but tropospheric wind shear to decrease, which will have opposite influences on lightning [20].

Table 3 summarizes the results of an ensemble of 12 global chemistry-climate models driven by 4 future anthropogenic emission scenarios [79]. The ensemble predicts

Table 3 Changes in lightning-NO_x emissions in 2030 and 2100 relative to 2000

Warming scenario	Δ2030 (%)	Δ2100 (%)
RCP2.6	3 ± 5	7 ± 8
RCP4.5	1 ± 8	9 ± 17
RCP6.0	3 ± 3	17 ± 5
RCP8.5	4 ± 9	27 ± 30

Multi-model mean ± standard deviation shown per future scenario in the ACCMIP study [79]. RCP8.5 has the greatest warming

lightning NO_x emissions will change +2.8 % by 2030 and +16 % by 2100, although there is very large uncertainty in these estimates. Furthermore, many of these models parameterize lightning flash rates using an empirical relationship that determines continental flash rates as a fifth-order power function of convective cloud-top depths [128]. Small changes in cloud-top depths manifest as very large changes in predicted flash rates. Since the cloud-top relationship is unable to reproduce some major features of the present-day lightning spatial and seasonal distribution [101, 162], it is unclear what predictive skill it has for future (or past) climates. Another study found a strong empirical relationship between observations of the total CG flash rate to total precipitation and CAPE over the continental USA. When applied to changes in precipitation and CAPE predicted by 11 global climate models, the relationship predicts a 50 % increase in CG lightning from 1996–2005 to 2079–2088 [133].

Even if lightning NO_x were to remain constant, its relative contribution to surface air quality will increase if anthropogenic precursor emissions decline, as predicted by some emission scenarios [166]. However, absolute contributions would rise as well; reductions in anthropogenic NO_x will increase free-tropospheric ozone production efficiencies (particularly in the northern hemisphere) and thereby increase the amount of ozone and OH generated from an identical amount of lightning NO_x. This will be especially true for ozone if actions are not taken to curb anthropogenic methane emissions. Emissions of SO₂ are also expected to decrease, but NH₃ emissions are likely to increase with expected growth in global food production [13]. Both will act to increase the fraction of lightning NO_x converted to nitrate aerosol, and therefore surface PM_{2.5}.

Conclusions

Lightning generates relatively large but uncertain quantities of NO_x, a critical precursor for the production of ozone and hydroxyl radical (OH) in the troposphere. Lightning NO_x has a disproportionately stronger influence on ozone and OH than surface-NO_x sources, due to higher OPEs above the boundary layer where most lightning NO_x is released. Direct exposure to lightning NO_x at the surface is small and short-lived relative to exposure guidelines. Few ground-based observation studies have estimated impacts of fresh lightning NO_x at the surface, and most have been in South Asia. It could be useful to perform these measurements in other regions with high lightning activity, including the USA. Nevertheless, model studies indicate that the primary influence of lightning on surface air quality is via its large-scale influence on background oxidants, rather than by local

photochemistry following release into the boundary layer.

Model sensitivity studies suggest lightning contributes 3–4 ppbv (~10%) of annual-mean PRB surface ozone, comprised of local, regional, and hemispheric components, and up to 18 ppbv during individual events. However, long-term feedbacks via methane may counter some of these effects on decadal time scales. Lightning contributes ~1 % to annual-mean surface PM_{2.5} concentrations, both as a direct precursor of nitrate aerosol and by promoting faster oxidation of PM_{2.5} precursors. Lightning is a natural source of nitrogen deposition to ecosystems, and large-scale shifts or trends in lightning could perturb ecosystems, especially those already impacted by large anthropogenic enhancements or in remote regions. Lightning indirectly impacts composition as a major source of wildfire ignition.

The magnitude and distribution of lightning NO in space and time remains poorly characterized, which limits the ability to fully quantify surface impacts. Major uncertainties include the inability of models to reproduce observed flash distributions, large uncertainties and apparent variability in NO-yields per flash, and a limited understanding of the vertical distribution despite high sensitivity of the response. Further uncertainties that limit our ability to accurately predict surface impacts involve uncertainties in the rate of mixing between the free troposphere and boundary layer [142], and in background reactive nitrogen chemistry [89]. Evidence suggests that urban pollution influences the frequency and physical properties of lightning itself, with implications for regional lightning-NO_x production and feedbacks on downwind surface pollution. How lightning will respond to a warming world remains uncertain, but models presently predict that global lightning-NO_x production will increase in the coming century, possibly imposing a climate penalty on air quality. Future studies should target reducing uncertainty in all these areas.

A first-order problem is representing lightning occurrence in air quality models. Global models show low skill in reproducing satellite lightning climatologies. Regional air quality models have the ability to prescribe CG flash rates from real-time ground detection networks, but assumptions must still be made about how to convert CG to total flash rates. Furthermore, prescribed lightning does not guarantee co-location within simulated deep convection, where fresh lightning NO_x may come in contact with lofted polluted surface air. Global models should move away from the simple cloud-top height relationship [128] in favor of parameterizations built upon cloud-microphysical parameters (e.g., cloud-ice flux [38]). It is also strongly recommended that meteorological reanalyses begin to assimilate lightning observations and provide flash rates in their standard products. In addition to improving the representation of lightning-NO production in chemistry-transport models driven by these meteorological fields, inclusion of lightning

data would also benefit the assimilation of cloud properties, precipitation, and convective mixing itself [37, 170].

Several regional air-quality studies cite Kaynak et al. [66] as justification for not including lightning in their simulations. However, as ozone regulations tighten and marginal costs on pollution controls increase, even 2–10 ppbv contributions to surface ozone become increasingly relevant for non-attainment regions. Furthermore, the regional-model simulations summarized in this review have not examined periods since 2008. There have been considerable reductions in anthropogenic NO_x and other pollutants since then in the USA [134], which will shift the relative role that lightning plays in the region, especially in areas that are becoming more NO_x-limited. Regional models should also examine health impacts of lightning-induced changes in pollutant levels in heavily populated regions of the tropics and subtropics outside the USA.

Looking forward, the Geostationary Lightning Mapper (GLM) is set to launch on the GOES-R weather satellite in late 2016, providing continuous observations of total lightning flash rates over North America for the first time. GLM will be operational during the launch (ca. 2019–2021) of the Tropospheric Emissions Monitoring of POLLution (TEMPO) satellite instrument, which will provide spatiotemporally continuous observations of key tropospheric species over North America. Collectively, and in conjunction with existing modeling and observational resources, they will offer unprecedented constraints for assessing the role of lightning on surface air quality.

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Compliance with Ethical Standards

Conflict of Interests LTM states that there is no conflict of interest.

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