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Role of Lightning NOx in Ozone Formation: A Review

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Abstract—This paper provides an overview on the spatiotemporal distribution and evolution mechanism of lightning. The predominant mechanism of ozone formation in the upper troposphere is lightning-induced precursors such as oxides of nitrogen (NOx), carbon monoxide (CO), and hydrocarbons (HC). Lightning-induced NOx (LNOx) is one of the major ordinary sources of NOx in the upper atmosphere, particularly in the tropical region, but it is still highly uncertain as to the exact quantity. Various ground measurements, satellite observations and modelling studies on the lightning and global NOx source rate have been extensively studied and compared to find the variability in estimated global lightning-induced NOx. Lightning can influence the climate via the production of nitrogen oxides ($NO + NO_2 = NOx$) followed by the production of ozone, another efficient greenhouse gas. The global annual lightning NOx of 5 ± 3 Tg $year^{-1}$ has been estimated by modelling studies with an ozone creation efficiency of 6.5 ± 4.7 times that of surface NOx sources. Understanding and quantifying the processes and production of lightning and LNOx is important for assessment of ozone concentrations and its associated impacts on the global climate.

Keywords: Lightning, ozone, NOx, cloud, thunderstorm.

1. *Introduction*

Lightning is a natural phenomenon that occurs due to opposite charge separation and accumulation within a thunderstorm cloud. It may be cloud-to-cloud exchange of charges, i.e. intra-cloud lightning, or cloud-to-ground exchange of charges, i.e. cloud-to-ground lightning. Lightning phenomena mostly occur during volcanic eruptions, tremendously intense forest fire events, heavy snowstorms, large hurricanes, and in thunderstorms. Lightning largely occurs in atmospheric cumulonimbus cloud. Since the cumulonimbus clouds are uplifted through convection, often growing from small cumulus clouds over a hot and moist surface, there is a better opportunity for lightning. More lightning phenomena happen over the land surface in comparison to the ocean surface because of the strong opposite charge attraction (Brook and Kitagawa, 1960; Orville and Henderson 1986; Christian et al. 2003; Kandalgaonkar et al. 2005; Ranalkar and Chaudhary 2009; Tinmaker et al. 2010; Lal et al. 2014, Siingh et al. 2015). When we compare an ocean storm with a land storm, the land storm flash rate is greater than that of the ocean (Mach et al. 2011). To explain the land–ocean contrast, two mechanisms are suggested: in the first mechanism, the difference in response of the land and ocean to the solar radiation are defined. The land surface becomes hotter inducing more unstable conditions with stronger updrafts that influence the ice microphysical process leading to charge generation and lightning in clouds (Kumar and Kamra 2012; Tinmaker et al. 2019). The law of physics describes the strong rising air current called updrafts, which forms cumulonimbus clouds (Cotton et al. 2011). In the second mechanism, larger concentrations of cloud condensation nuclei over land causes more numerous and smaller cloud droplets which suppress the coalescence processes and provide more super-cooled droplets in the mixed-phase region where they participate in the charge generation processes (Joshi et al. 2008; Matsui et al. 2016; Shi et al. 2015; Williams et al. 2005).

Atmospheric electrical discharge generates various new trace molecules of gases with the interaction

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of ambient constituents of the atmosphere. Nitric oxide (NO) molecules can be produced directly or indirectly through electric discharge and play an important role in determining the chemistry of the troposphere and stratosphere (Crutzen 1970). A lightning channel is a potent source of NO as a trace gas in the middle and upper troposphere with a considerably smaller concentration of nitrogen dioxide (NO_2) (Bond et al. 2002; Tie et al. 2002; Williams and Stanfill 2002). Once NO is produced, it undergoes atmospheric oxidation and is converted to NO_2, establishing a balance between NO and NO_2 on a time scale of minutes. In most of the literature, NO and NO_2 are collectively referred to as NOx. Apart from lightning, a relatively negligible amount of NOx produced in the lower troposphere is transported upward through convection (Choi et al. 2005). Thus, lightning is a major source of NOx in the upper troposphere and lower stratosphere (Levy et al. 1996; Zhang et al. 2000). The convective available potential energy (CAPE) is a useful tool for determining severe weather potential at a given location and time. The CAPE magnitude and vertical distribution play important roles in determining the updraft velocity and vertical distribution of hydrometeors, which participate in the charge separation mechanism inside thunderstorm clouds (Williams 1985). In general, an atmosphere with larger CAPE is likely to produce a stronger updraft and more vigorous and electrically active storms (Tinmaker et al. 2015). The relationship between maximum updraft and CAPE is

$$\mathrm{Wmax} = \sqrt{2\mathrm{CAPE}}(\mathrm{Williams\ etal.}, 2005).$$

The understanding of the lightning-induced nitrogen oxides (NOx) (mol per flash) and its global budget [Teragram (Tg)] is important for the assessment and prediction of the spatial and temporal trend of the nitrogen oxide concentration, ozone concentration, oxidizing capacity of the atmosphere, and also the lifetime of the tropospheric trace gases. Further, this knowledge can be utilized for source apportionment and emission contribution to the global budget, particularly uncertain potent sources such as aviation emissions and stratospheric exchange from the surface processes. This study presents a detailed review on the lightning behavior, its classification, formation mechanism and satellite instruments for detection of lightning flash rates. The importance of NOx and lightning-induced NOx as precursor in ozone formation has also been discussed. In Sect. 2, various ground measurements and satellite observations of lightning are studied in detail. Section 3 deals with understanding the electric behavior of a thunderstorm, its formation and classification. The hotspots of lightning are discussed in Sect. 4. Finally, in Sect. 5 and 6, evolution of lightning global annual NOx source rate has been extensively studied and compared with global modelling studies to find the variability in estimated lightning-induced NOx budget.

2. *Instrumentation and Measurements*

Clouds and precipitation phenomena are more difficult to forecast in the tropics than at higher subtropical regions, due to the difference in solar insolation and earth rotation (Williams 2017, 2005; Williams, and Stanfill 2002). There is a great need to study and monitor precipitation and the associated release of energy over the tropical and subtropical regions because these phenomena relatively influence the global climate pattern and consequently result in a change to the environment. Understanding of tropical precipitation is also important for weather and climate prediction, as this event contains three fourths of the energy that drives atmospheric wind circulation (Kummerowet al. 2000).

Prior to satellite observation for larger coverage, ground instruments were in use with higher accuracy. Ground instruments such as lightning detectors and radio receivers detect lightning discharge under thunderstorm activity with limited spatial coverage. The first such device was invented in 1894 by Alexander Stepanovich Popov; it was the world's first radio receiver, an electronic device capable of receiving radio waves and converting them into the desired information. Another observation network, the World Wide Lightning Location Network (WWNNL), uses ground-based receivers in different countries of the world, capable of detecting lightning events with a current intensity greater than 5 kA (Gharaylou et al. 2020). The WWLLN has been providing data since 15 August 2004 to the present.

In 1995, the National Aeronautics and Space Administration (NASA) launched the Optical Transient Detector (OTD), as a prototype of the Lightning Imaging Sensor (LIS) of a tropical rainfall measuring mission, onboard the Microlab-1, later renamed OrbView-1, a small communication satellite. This was the first instrument specifically designed to detect lightning from space during both day and night with storm-scale resolution (Christian et al. 2003).

Advancing ahead, US's and Japan's space agencies planned for a joint earth observation mission to monitor the tropical rainfall activity, which was popularly known as the Tropical Rainfall Measuring Mission (TRMM), launched in 1997 with a working tenure of 17 years, which lasted until 2015. The TRMM term refers to both the mission itself and the satellite, with five onboard instruments, namely precipitation radar, microwave imager, visible infrared scanner, Clouds and the Earth's Radiant Energy System (CERES) and LIS. Prior to this mission, the distribution of global rainfall pattern was known with only 50% certainty (Simpson et al. 1996). A lightning imaging sensor is the main instrument designed (Fig. 1) for monitoring the total count of lightning flashes in each granule of its coverage range of ± 39.5 latitude (Christian et al. 1999). The LIS has a smaller field of view (FOV) than that of a OTD but higher detection efficiency (Williams et al. 2000). The LIS has the ability to detect and locate cloud-to-ground, intra-cloud and inter-cloud lightning in the tropical region, by the identification of the changes in cloud brightness, even with sunlit clouds in the background. It covers a storm scale resolution of 5–10 km at temporal resolution of 2 ms and a flash detection efficiency of 93 ± 4% at night and 73 ± 11% during day time. After the TRMM, an LIS was placed on the international space station (ISS) in Feb 2017 (Erdmann et al. 2020). On the space station, the LIS monitors total global lightning in both day and night and covers almost the entire globe (Liu et al. 2020). The ISS operates in low earth orbit (LEO) and traverses an area of the surface up to three times per day (two times in tropical regions). Lightning observation of a specific point lasts up to 90 s per overpass due to the ISS orbit characteristics and the LIS FOV of approximately 655 × 655 km^2 (Erdmann et al. 2020).

Additionally, a Geostationary Lightning Mapper (GLM) was launched on November 19, 2016 aboard the first Geostationary Operational Environmental

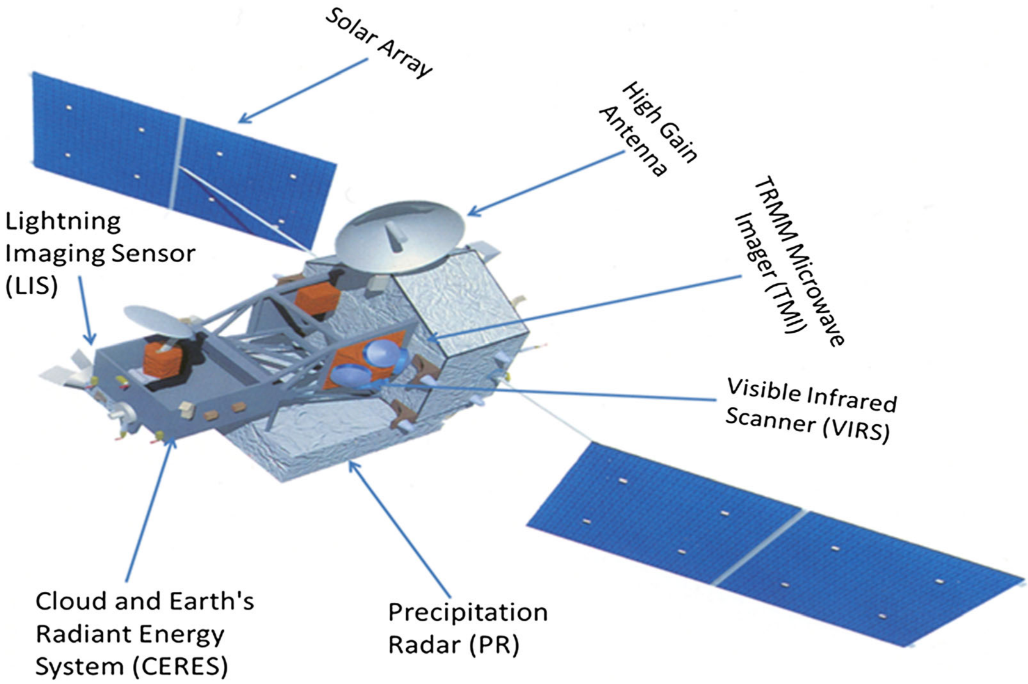

Figure 1
Tropical Rainfall Measuring Mission (TRMM) (Source: https://trmm.gsfc.nasa.gov/overview_dir/background.html)

Satellite (GEOS) R-Series spacecraft (Rudlosky et al. 2019). The satellite Geostationary Lightning Mapper (GLM) continuously monitors lightning activity extensively over a near hemispheric field of view and records the spatio-temporal contrast at an unprecedented level. The GLM provides continuous aggregate lightning measurements over America and adjacent oceans with coverage up to 54° North and South. It detects all lightning strikes be it cloud to cloud or cloud to ground. GLM is designed to detect on average $> 70\%$ within 24 hours of blackheads, better performance at night ($\sim 90\%$) than during the day ($\sim 70\%$). Another satellite, Feng-Yun-4A (FY-4A), uses a lightning mapping imager (LMI) sensor and provides lightning event and flash data by grouping events (Liu et al. 2021). FY-4A is the China Meteorological Administration (CMA) second-generation three-axis stable geostationary weather satellite series developed by the China Academy of Space Technology (CAST). FY-4A launched in December 2016 and it has been operational since May 2018. FY-4A has spatial resolution about 6.8 km at SSP (sub-satellite point) and works at 777.4 nm. Event data are the most basic unit of lightning signals obtained by FY-4A LMI (Liu et al. 2020).

3. *Electrical Behavior of Thunderstorms*

3.1. *Mechanism of Lightning and Thunderstorms*

Lightning develops when the atmosphere is unstable. This is when warm air is presents underneath much colder air. As the warm air rises, it cools and condenses forming small droplets of water. A rising air mass carries water vapor and, on meeting the cooler air, usually condenses and releases heat, giving rise to convective storm activity. The center column of cumulonimbus clouds can have an updraft exceeding 120 km h^{-1}. This same updraft gives rise to an electric charge separation within the systems, which ultimately leads to lightning activity.

Thunderstorms develop within a cloud under the influence of gravity that pulls the larger water or ice particles towards the bottom of the cloud. This separation of smaller, positively charged particles and negatively charged particles creates an electric imbalance with an enormous electric potential of millions of volts across a storm cloud. It is the natural electrostatic discharge that occurs in an electrical charge separated region of a cloud system and is usually associated with thunderstorms. These are short-lived localized storms, which have vertical air motion, humidity and instability. The lightning intensity in a thunderstorm provides useful information relating to the dynamics and microphysics of convective storms (Williams et al. 1991). Lightning intensity is related to a convective cloud development stage (Williams et al. 1989), the intensity of updraft (Lhermitte and Williams 1985), the cloud liquid water content (Saunders et al. 1991), and the cloud structure (Rust et al. 1981). Mo st of the time the higher frequency of the thunderstorm occurs because of convergence of two different types of storm fronts (dry and moist wind) (Lal et al. 2018). Also, high land features such as hills or mountain top regions are the most prone to cloud-to-ground lightning events because of the minimum distances between the earth's surface and the cloud's lower base (Boccippio et al. 2001).

3.2. *Electrification*

The primary source of lightning is the associated cloud type and developed electrical structure within it. The most common typw is the cumulonimbus cloud, sometimes called a thundercloud. However, not every cumulonimbus cloud produces lightning (Imyanitov et al. 1971). Lightning phenomena are usually associated with a convective cloud system with a vertical extent ranging from 3 to 20 km and horizontal dimensions ranging from 3 km to > 50 km, also they have a characteristic strong updraft of ≥ 10 m s^{-1}. Lightning produced by a thundercloud is usually a structured system of charge induction and distribution within it, these structures vary according to cloud development and contamination by forest fire smoke, volcanoes, sandstorms, nuclear explosions and other anthropogenic emission (vehicular emission, stack emission, etc.). The distribution and motion of hydrometeors (liquid or frozen water particles in the atmosphere) and free ions present in the complex atmospheric system produce the thunderstorm clouds. The basic structure of the cloud charge distribution includes a net positive

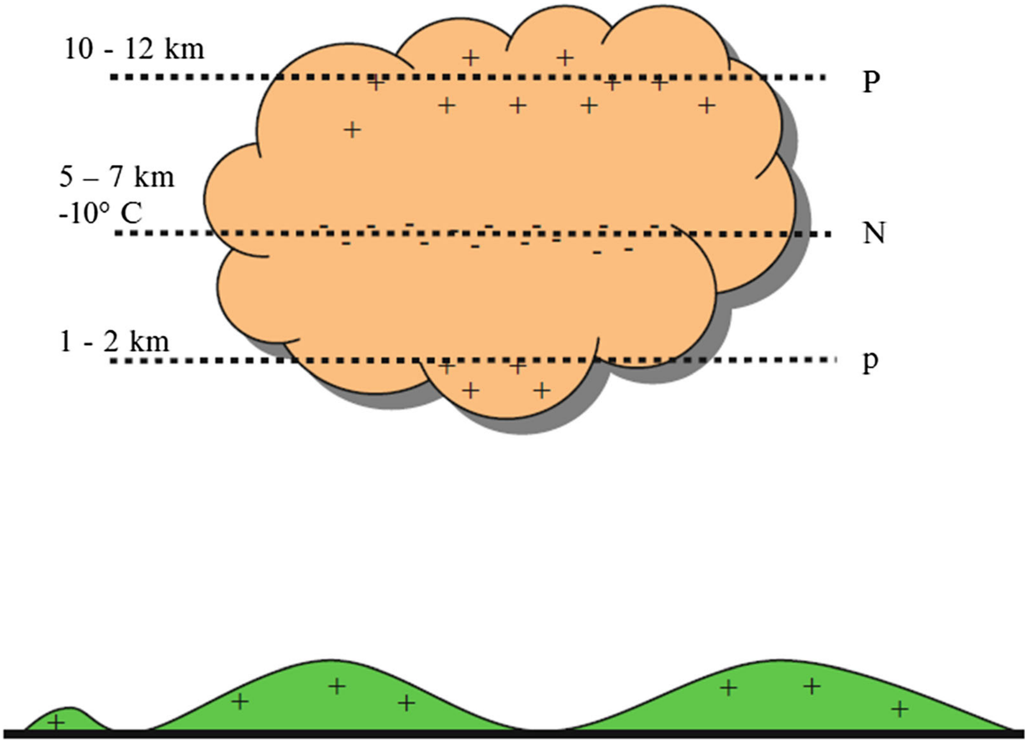

Figure 2
An idealized tri-pole charge structure of a thundercloud (Source: Cooray 2015)

charge near the top, a net negative charge below it, and an additional positive charge at the bottom of the cloud, as illustrated in Fig. 2 (Cooray 2015 and Krehbiel 1986). The positive charge at the top and the negative charge in the middle is due to the existence of polarity and does not affect the net charge in any area of the cloud (MacGorman and Rust 1998). In addition, there is a small but important positive charge buildup near the bottom of the thunderstorm cloud due to precipitation and warmer temperatures. A cloud is a relatively good electrical insulator, and leakage currents between the charged regions are spatially separated on the order of kilometers by their polarity. So far in the scientific domain there have been several reviews available on cloud electrification mechanisms (Moore and Vonnegut 1977; Phillips 1967; Stolzenburg, and Marshall 2009; Takahashi 1978, 1984; Vonnegut 1953; Williams and Heckman 1993; Wilson 1956). However, in this review only the non-inductive collisional graupel-ice mechanism and the convection mechanism have been considered. There is a wide consensus that the graupel-ice mechanism is the dominant mechanism for cloud electrification. In the convective mechanisms, the electric charges are supplied to the cloud system by external sources such as warm air currents that carry positive charge to the top of a growing system; downdrafts caused by expansion and cooling of the growing cumulus, carrying negative charge towards the cloud base, which produces positive corona at the earth's surface. In a charged ambient, the hydrometeor also appears to be polarized and is spatially separated. The illustration of the convective mechanism of cloud electrification is shown in Fig. 3.

However, in the non-inductive collisional graupel-ice mechanism, hydrometeors do not need to be polarized by the ambient electric field. In this mechanism the electric charges are produced by collisions between graupel and small ice crystals in the presence of water droplets. Graupels are precipitation particles with an high fall speed (≥ 0.3 m s^{-1}), and ice or cloud particles have a lower fall speed and smaller size in comparison. This

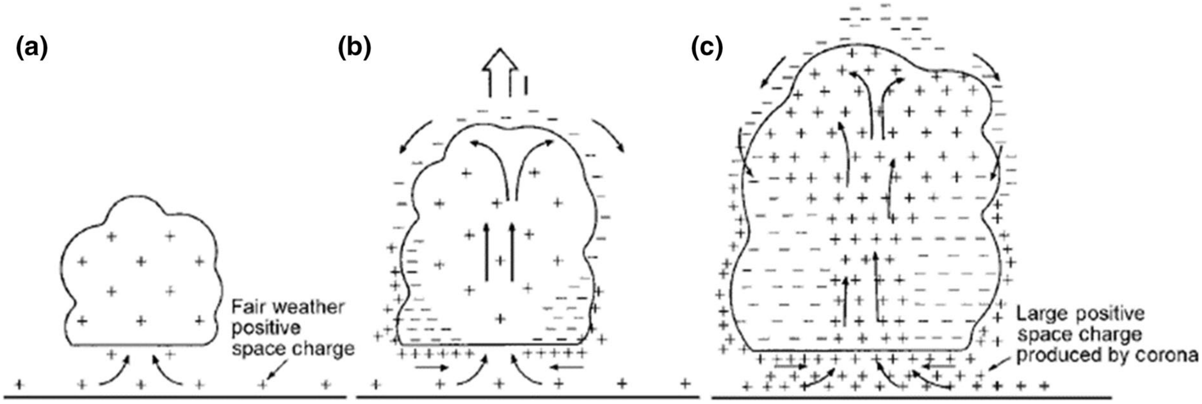

Figure 3
Schematic diagram of convection mechanism of cloud electrification (Source: MacGorman and Rust 1998)

mechanism also explains the classical tri-polar cloud charge structure; the illustration of the graupel-ice mechanism of cloud electrification is shown in Fig. 4.

3.3. Lightning Classification and Hotspot

Lightning discharges can be classified by the path of discharge into the following categories

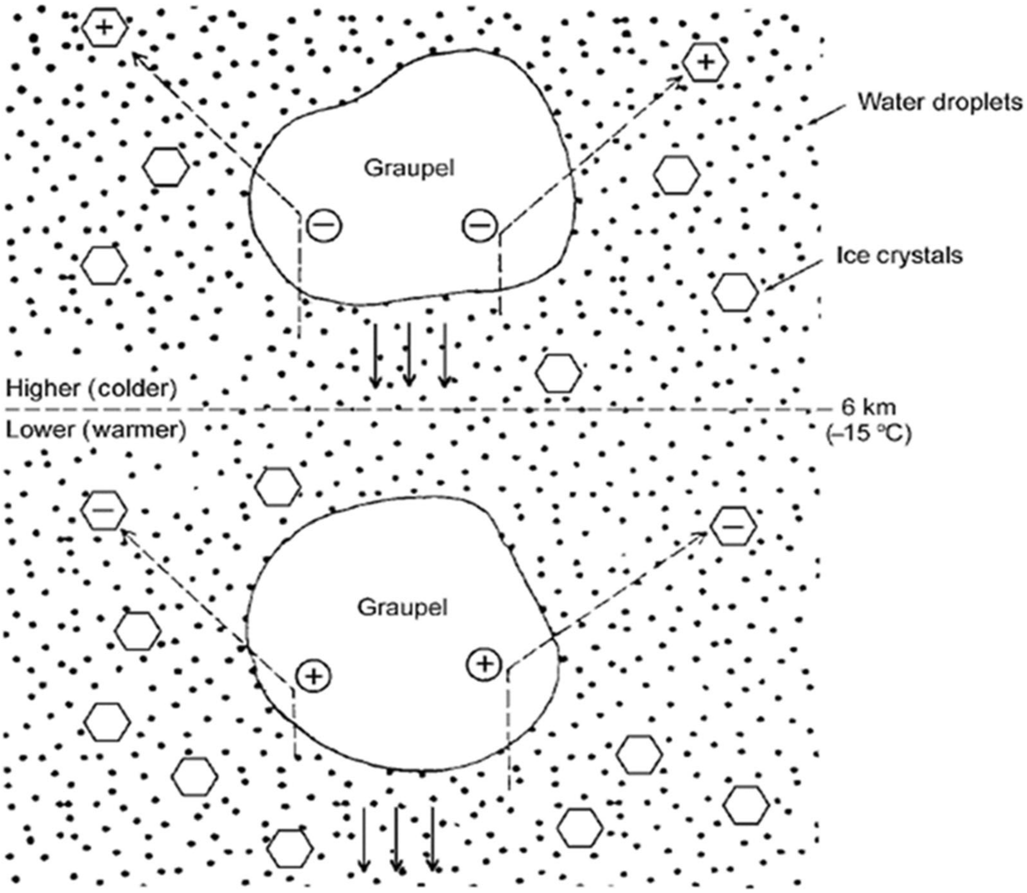

Figure 4
Illustration of the graupel-ice mechanism of cloud electrification (Source: Rakov and Uman 2005)

(MacGorman and Burgess 1994; Rust et al. 1981; Seimon 1993):

- Intra-cloud flashes which occur entirely inside a cloud (IC).
- Cloud-to-cloud (or inter-cloud) flashes (CC).
- Cloud-to-ground flashes which occur between a cloud and the ground (CG).
- Air discharge flashes which occur between a cloud and the surrounding air.

Intra-cloud flashes (IC) are known to be the most frequent type of lightning discharges that occur in the earth's atmosphere (Cooray 2003; Shao and Krehbiel 1996). Since IC lightning flashes occur inside a thundercloud, mostly remote electric field observations are used to investigate this feature (Marshall et al. 2005). It has been found that IC flashes bridge the gap between the middle negative layer of a thundercloud and its upper positive layer. Cloud-to-ground flashes can be classified into four types according to the direction of their initial lead propagation (either upward or downward) and the polarity of effective charge transfer (either positive or negative) (Uman 1987). This classification is schematically shown in Fig. 5. Apart from this conventional classification, an individual lightning flash can transfer both negative and positive charges during its course. This type of lightning discharge is called a bipolar flash, which are as frequent as positive flashes (Rakov and Uman 2005).

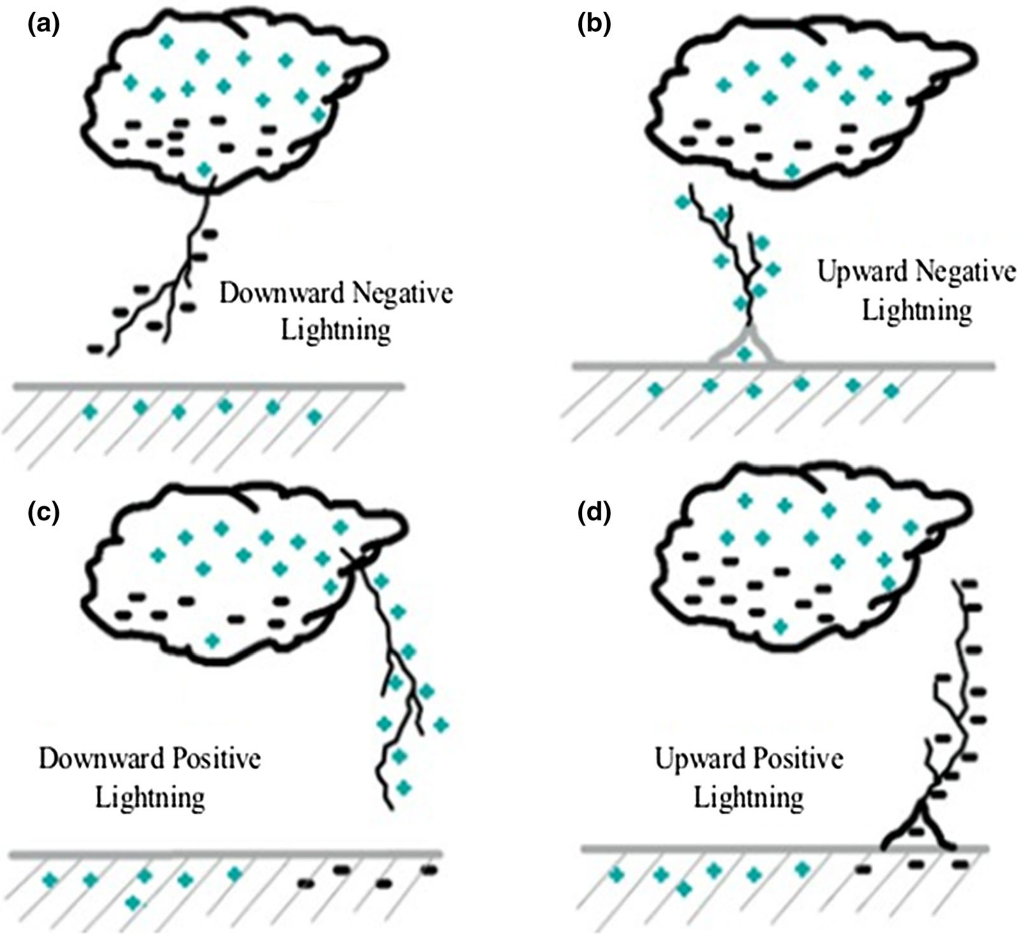

Figure 5
Four category of cloud-ground flashed based on the effective polarity of charge transfer to ground (Source: Rakov and Uman 2005)

Lake Maracaibo is a famous lightning hotspot (233 flashes km^{-2} year^{-1}) (Albrecht et al. 2016) and is known as the lightning capital of the world. Lake Maracaibo is situated in South America with very warm and humid climatic conditions near the valley of the Northern Andes Mountains in Colombia. As the wind patterns often carry warm and humid air from the Caribbean southward into the Gulf of Venezuela and Lake Maracaibo, once air enters, it gets trapped by the tall mountains across the region. The closest populated place to this hotspot over Lake Maracaibo is Lagunillas in the state of Zulia on the east side of the lake, about 60 km away (Table 1). Another lightning hotspot is the Democratic Republic of the Congo which is situated in central Africa with the second highest number of lightning flashes (greater than 205 flashes km^{-2} year^{-1}) (Cecil et al. 2015; Christian et al. 2003; Goodman et al. 2013). Table 1 shows that the greatest lightning hotspot over India is Rajouri (Jammu and Kashmir), and the second is Brahmaputra Valley of Eastern India. Lightning varies from season to season over India. The maximum lightning hot spot over India is in the northeast in pre-monsoon season, while during monsoon season it is highest in the northern area, i.e. Jammu and Kashmir. In pre-monsoon season, the NE region of India experiences high-frequency convection activity and high cloud tops; hence, the probability of a lightning event is more likely. Because of the increase in global temperature, the intensity of thunderstorms, lightning and dust storms is increasing day by day (Yadava et al. 2020).

3.4. *Evolution of Lightning Activity*

Vertical development of the updraft parcel in thunderstorm activity initiates the process of charge separation and accumulation leading to lightning strikes (Workman and Reynolds 1949; Brook and Kitagawa 1960; Williams et al. 1989, Williams and Lhermitte 1983; Carey and Rutledge 1996; Rutledge et al. 1992; Lal et al. 2018). Intra-cloud lightning flashes are usually the first strikes that appear for the first several minutes after thunderstorm development, whereas cloud-to-ground strikes increase in frequency later in the thunderstorm life cycle (Goodman et al. 1988; Reap and MacGorman 1989; Workman and Reynolds 1949). Mostly cloud-to-cloud and other discharges that do not contact the ground dominate the total lightning activity (Williams 2001). They are more frequent and reach peak activity when the vertical cloud development is greatest (Goodman et al. 1988; Williams et al. 1989), while the cloud-to-ground flashes are less frequent and erratic, and differ in terms of growth and decay (Williams 2001). In an elevated charge parcel during updraft intensification, the separation between the charge centers ideally influences both flash rate and type. This can also be explained by the theory of electric field, which is inversely proportional to the square of the distance from the charge (MacGorman and Rust 1998). In addition, the intense updraft of the parcel suspends the graupel-laden mean negative current (MNC) region closer to the mean positive current (MPC) region causing the electric field to be stronger above the MNC region, which further favors the intra-cloud flashes. Within a towering cloud (such as cumulonimbus), the MNC regions accumulate negatively charged hailstones while positively charged ice crystals constitute the region of the MPC. As the storm updraft moves through the cloud, the lighter ice crystals are carried upward leaving behind the denser hailstones to fall to the bottom. Thus, both the MNC at the bottom region and the MPC at the top develop at the same time.

With the breakdown phase, the updraft threshold decreases with decreasing pressure resulting in more lightning at higher altitudes in comparison to lower altitudes. Further weakening of updraft-downdraft and descending of precipitation particles towards the surface increases the cloud-to-ground activity. Numerous studies support the elevated charge mechanism (e.g., Ziegler and MacGorman 1994; Stolzenburg et al. 1998; Lang and Rutledge 2002).

Atmospheric temperature and moisture also play major roles in the concentration of graupel and ice accumulation. Further, a significant interaction between the developed graupel and ice at the higher altitude in the presence of super-cooled water leads to charge attraction and flash events. Within cumulus clouds, the size of hydrometeors (graupel and ice), charge separation and lightning frequency are governed by updraft speed of wind and volume (Lal et al. 2014). There is highly nonlinear relationship between

Table 1

Top 10 FRD (flashes km^{-2} year^{-1}) for each continental landmass, indicating its position in the global ranking, latitude (Lat) and longitude (Lon) position on TRMM LIS 0.1° climatology grid, as well as the name of the nearest populated place (PPL), and distance from grid point (Dist), according to the GeoNames database (www.geonames.org/)

Global rank	FRD	Lat (°)	Lon (°)	PPL	Country	PPL lat (°)	PPL lon (°)	Dist (km)
South America								
1	232.52	9.75	−71.65	Lake Maracaibo (Lagunillas)	Venezuela	10.13	−71.26	60.1
4	172.29	7.55	−75.35	Cáceres	Colombia	7.58	−75.35	3.4
7	138.61	8.85	−73.05	El Tarra	Colombia	8.58	−73.09	30.9
11	124.26	5.75	−74.95	Norcasia	Colombia	5.58	−74.89	20.4
18	114.19	8.45	−74.55	Majagual	Colombia	8.54	−74.62	12.6
25	105.73	8.15	−76.85	Turbo	Colombia	8.09	−76.73	14.8
46	95.38	11.15	−72.95	Barrancas	Colombia	10.96	−72.79	27.8
74	87.96	−17.25	−65.05	Chimoré	Bolivia	−16.95	−65.14	34.9
78	87.61	10.35	−70.95	El Corozo	Venezuela	10.12	−71.04	27.5
136	77.02	10.45	−75.35	Santa Rosa	Colombia	10.44	−75.37	2.2
Africa								
2	205.31	−1.85	27.75	Kabare	Democratic Republic of the Congo (DRC)	−2.50	28.79	136.2
3	176.71	−3.05	27.65	Kampene	DRC	−3.60	26.67	124.9
5	143.21	−0.95	27.95	Sake	DRC	−1.57	29.04	140.0
8	129.58	5.25	9.35	Nguti	Cameroon	5.33	9.42	11.7
9	129.50	0.25	28.45	Butembo	DRC	0.14	29.29	94.3
10	127.52	−1.55	20.95	Boende	DRC	−0.28	20.88	141.2
14	117.98	0.55	20.35	Boende	DRC	−0.28	20.88	109.7
15	117.19	−2.45	26.95	Kindu	DRC	−2.94	25.92	126.7
16	116.78	6.95	10.45	Baissa	Nigeria	7.23	10.63	36.6
19	112.17	0.35	26.65	Kisangani	DRC	0.52	25.19	163.3
Asia								
6	143.11	34.45	72.35	Daggar	Pakistan	34.51	72.48	14.0
12	121.41	33.35	74.55	Rajauri	India	33.38	74.31	22.6
13	118.81	33.75	70.75	Doaba	Pakistan	33.42	70.74	36.2
22	108.03	14.55	43.45	Aladi yah	Yemen	14.53	43.57	13.2
28	104.59	33.85	73.25	Murree	Pakistan	33.91	73.39	14.5
31	101.79	25.25	91.95	Cherrapunji	India	25.30	91.70	26.1
42	97.02	4.75	103.05	Paka	Malaysia	4.64	103.44	44.7
45	95.92	1.95	103.85	Kota Tinggi	Malaysia	1.74	103.90	24.2
50	94.64	3.75	98.05	Tenggulun	Indonesia	3.99	98.01	27.3
52	93.96	3.15	101.65	Kuala Lumpur	Malaysia	3.14	101.69	4.2
North America								
17	116.76	14.35	−91.15	Patulul	Guatemala	14.42	−91.17	7.6
29	103.23	14.85	−92.05	Catarina	Guatemala	14.85	−92.08	2.8
33	100.63	22.35	−83.95	San Luis	Cuba	22.29	−83.77	20.1
34	100.24	18.55	−74.35	Chambellan	Haiti	18.57	−74.32	4.0
37	99.39	13.15	−87.25	San Jerónimo	Honduras	13.18	−87.14	12.7
39	98.22	22.35	−80.65	Rodas	Cuba	22.34	−80.56	9.8
40	98.06	21.75	−78.85	Venezuela	Cuba	21.74	−78.80	5.8
47	95.32	22.85	−82.15	Mañalich	Cuba	22.81	−82.15	4.3
82	86.96	22.25	−105.2	Rosamorada	Mexico	22.12	−105.2	14.9
90	85.78	18.15	−77.65	Balaclava	Jamaica	18.17	−77.64	2.6
Oceania								
61	92.15	−15.35	125.35	Derby	Australia	−17.30	123.63	284.4
83	86.75	−14.45	126.55	Kununurra	Australia	−15.78	128.74	278.0
228	65.11	−16.65	124.75	Derby	Australia	−17.30	123.63	139.6
308	59.69	−15.65	128.45	Kununurra	Australia	−15.78	128.74	34.6
316	59.19	−4.75	142.95	Ambunti	Papua New Guinea	−4.22	142.82	61.0
327	58.57	−15.25	129.45	Kununurra	Australia	−15.78	128.74	95.8

Table 1 *continued*

Global rank	FRD	Lat (°)	Lon (°)	PPL	Country	PPL lat (°)	PPL lon (°)	Dist (km)
355	57.13	−13.15	131.05	McMinns Lagoon	Australia	−12.55	131.11	66.6
381	55.57	−13.95	129.95	Darwin	Australia	−12.46	130.84	191.6
471	51.35	−19.15	137.85	Mount Isa	Australia	−20.73	139.50	245.6
477	51.22	−17.45	126.05	Halls Creek	Australia	−18.22	127.67	191.6

A minimum distance of 100 km from a previous ranked grid point is applied

Source: Albrecht et al. (2016)

lightning rate and storm depth (Vonnegut 1953; Williams 1985).

$$f_c = L^5$$

where f_c is the lightning rate for continental thunderstorms (flashes $\min^{-1}$) and L is the storm depth or height (m). Lightning frequency is directly related to cloud parameter (cloud height), cloud radius and updraft speed (Baker et al. (1995, 1999). Thunderstorm lightning has been considered a major source of oxides of nitrogen [nitric oxide (NO) and nitrogen dioxide (NO_2)]; it is a natural mechanism for the fixation of atmospheric nitrogen (Hutchinson 1954). Lightning-induced oxides of nitrogen (LNOx) have several important implications to understand the changing atmospheric chemistry and climate change (WMO 1999; IPCC 2001).

4. *Lightning-Induced NOx*

4.1. Lightning-Induced NOx: Precursor of Ozone

Ozone (O_3) is good in the stratosphere, while considered bad in the lower atmosphere as it is an injurious air pollutant for both humans and the environment. O_3 is a greenhouse gas with a global mean radiative forcing of 0.40 (0.20–0.60) W m^{-2} since the pre-industrial period (Myhre et al. 2013; Stevenson et al. 2013). The major precursors of ozone in the lower troposphere are carbon monoxide (CO), hydrocarbons (HC) and NOx (NO + NO2). All these species are emitted to the lower atmosphere from anthropogenic sources such as thermal power plants, industry, transportation, fossil fuels and biomass burning, and from natural sources such as biogenic emissions and climate processes such as lightning (Lal et al. 2012).

One of the major natural sources of nitrogen oxide (NO) is lightning in the middle and upper troposphere while in lower troposphere it is negligible. The extreme temperature inside a lightning strike channel converts the atmospheric abundant stable N_2 and O_2 into relatively significant quantities of NO, similar to high-temperature fuel combustion; however, this process also produces other trace species such as NO_2, CO and ozone (O_3) in a smaller amount. Further, through oxidation, NO is rapidly converted to NO_2 until equilibrium is reached. In the middle and upper troposphere both (NO + NO_2) have longer residence time. During daytime, NO_2 undergoes photolysis and forms atomic oxygen, which reacts with an oxygen molecule to produce ozone (O_3). As a source of atomic oxygen, lightning NOx is thought to be the most important natural ozone precursor. As the day grows, ozone accumulates and is photolyzed in the presence of water vapor and produces hydroxyl radical (OH), the main tropospheric oxidants controlling the lifetime of many important climate gases including methane and hydrogenated halocarbons (IPCC AR4 WG1 2007). Global lightning-induced NOx is one of the largest natural sources of NOx in the upper troposphere, in particular in the tropical region (Sinha and Toumi 1997; WMO 1999). However, in the total atmospheric NOx budget, lightning-induced NOx production rate is considered least, because globally it cannot be measured directly (Logan 1983; Lawrence et al. 1995; Lee et al. 1997). For better quantification of lightning-produced NOx, knowledge of spatial and temporal occurrence of lightning flashes as well as the amount of NO produced per flash is required with its vertical distribution. As it has been observed and studied, most lightning activity occurs within deep convective clouds in the tropical and subtropical regions and is most prominent during high solar insolation (afternoon and early evening) over the land masses (Christian et al. 2003; Schumann et al. 2007). Previous studies to determine the amount of lightning-induced NOx have been based on theoretical, laboratory and field studies mainly by extrapolating measurements of emissions from individual lightning or thunderstorm events to the global scale (Chameides et al. 1977; Lee et al. 1997; Huntrieser et al. 1998; Bradshaw et al. 2000). Based on similar studies, a global estimate of lightning-induced NOx is about 5 Tg per year (nitrogen mass unit per year) with an uncertainty range of 1–20 Tg A^{-1} (Labrador et al. 2004; Cook et al. 2000; Liaw et al. 1990). Satellite observations is one of the advanced ways to globally monitor the lightning flashes and NO_2 column distribution (Christian et al. 2003; Ridley et al. 2004). The amount of the NO produced per flash is still a matter of uncertainty. Most field studies, aircraft mission, satellite observation and various cloud resolving chemistry models estimate $\sim$ 17 to 700 mol NO flash^{-1}as shown in Table 2. Schumann and Huntrieser, 2007 in an extensive literature review reported a mean value of 250 mol flash^{-1} with an estimated range of 33–600 mol NO flash^{-1}. Ground observational studies suggest that average NO yield per flash is higher in the extra tropics than tropics (Huntriser et al. 2008) and ranges from 82 to 257 mol NO flash^{-1}. The aircraft mission and cloud resolving model simulation output mean ranges from 160 to 360 mol NO flash^{-1}. However, there is an uncertainty as to whether cloud-to-ground and intra-cloud flashes produce the same amount of NO on average or whether cloud-to-ground produces more (Cooray et al. 2009; Koshak et al. 2014; Ott et al. 2010).

4.2. *Ozone Formation Mechanism*

Nitrogen oxide is a critical component of the lower atmosphere which directly alters the amount of ozone (Crutzen, 1974) and hydroxyl radical OH. Ozone is also known as a strong oxidant, a strong absorber of ultraviolet radiation and a greenhouse gas (WMO 1999). Ozone is formed and destroyed by photochemistry of NOx. (Liu 1987). In region with low NOx level (e.g. in the tropic marine boundary layer) the net effect is ozone damage. In regions with

Table 2

NO yield per flash estimated by various recent studies

Year	Method	Mol (NO)/flash	Location
Ridley et al. (2004)	Aircraft data and model	55–382	South florida
DeCaria et al. (2005)	Aircraft data and cloud model	345–460	STERAO-A
Beirle et al. (2006)	Satellite GOME	32–240	Subtropical
Schumann and Huntrieser (2007)	Literature review	250 (33–600)	Global
Huntrieser et al. (2008)	Field study	67–201	Brazil
Huntrieser et al. (2009)	Field study	120–389	Oceania
Coorey et al. (2009)	Process based model	90	Global
Beirle et al. (2010)	Satellite	50	Global
Bucsela et al. (2010)	Satellite	100–250	Central America
Huntrieser et al. (2011)	Field study	71–188	West Africa
Martini et al. (2011)	Chemistry transport model	480	North America
Cumming et al. (2013)	Cloud-resolving-chem model	500	Oceania
Koshak et al. (2014)	Process based model	100 (182–171)	North America
Miyazaki et al. (2014)	Chemical data assimilation	310 (0–700)	Global
Liaskos et al. (2015)	Chemical transport model	246	Global
Pollack et al. (2016)	Field study	194 (71–253)	North America
Laughner and Cohen (2017)	WRF-chem coupled DC3	500–665	Southeast US
Arndt et al. (2018)	Satellite	500 (320–500)	Europe
Zhu et al. (2019)	WRF-chem Simulation	500	North America

NOx concentration above a critical level (but not very high), e.g. in the upper troposphere, ozone production dominates. The critical NOx level depends on the ozone mixing ratio, may be as low. Formation of ozone involve a series of chemical reactions where precursor such as volatile organic compounds and oxides of nitrogen ($NOx = NO + NO_2$) react to presence of sun light. The emission of NOx has both natural sources (such as lightning, soil and oxidation of ammonia) and anthropogenic sources, such as fossil fuel combustion, power plants and biomass burning (Logan et al. 1981; Ghude et al. 2010). The lifetime of NOx is up to few days, and it is formed directly by lightning in the upper troposphere (Tie et al. 2002). The amount of NOx by lightning is not a linear function of energy discharge (Chameides et al. 1977). Therefore, NOx is typically the limiting precursor in ozone production (Jaffe and Wigder 2012). NOx is a key species in the photochemistry of troposphere. There are quick photochemical/chemical inter-conversions between NO and NO_2 through the following reaction (Levine et al. 1984).

$$NO + O_3 \rightarrow NO_2 + O_2, \quad (1)$$

$$NO_2 + hv \rightarrow NO + O \quad \lambda \leq 420 \text{ nm}. \quad (2)$$

The oxygen atom resulting from the photolysis of NO_2 in reaction (2) leads to formation of ozone (O_3) in the troposphere by the following reaction.

$$O + O_2 + M \rightarrow O_3 + M \quad (3)$$

Here *M* in the equation is third body to receive extra energy.

4.3. Source-Wise NOx

Lightning is a significant source of NOx particularly in the upper troposphere (Ridley et al. 1996; DeCaria et al. 2005); however, it accounts for roughly 10–15% of NOx input in the troposphere (Bradshaw et al. 2000; Schumann and Huntrieser 2007; Tuck 1976). In the upper atmosphere, NOx lasts for a longer duration and thus its ozone producing potential is greater (Pickering et al. 1998; Liu et al. 1987) than in the boundary layer, where the majority of NOx is added to the atmosphere. Lightning strikes produce between 5 and 10 Tg N year^{-1}, while nitrogen oxides induced by lightning play a small part in surface O_3 concentrations. The seasonal highest global production of lightning-induced NOx occurs during post monsoon (JJA) and the lowest production occurs during winter (DJF) period. Because of this natural imbalance, the northern hemisphere had a natural bias in tropospheric ozone, even in pre-industrial times. Future climate change may change the pattern and enhances the global lightning activity resulting in an increase in tropospheric ozone loading (Toumi et al. 1996). Biomass burning during spring (harvesting months of April–May) increases air pollution during winter months (November–January) due to lower mixing layer height, also play important role in the intra seasonal variations of NO_2 and O_3 concentration (Pawar et al. 2017). In the atmosphere, the present source rate of global NOx (Tg A^{-1}) is dominated by the anthropogenic sources (28–32 Tg A^{-1}), IPCC 2001, biomass burning (4–24 Tg A^{-1}), soil (4–16 Tg A^{-1}) (Lee et al. 1997) and LNOx (1–20 Tg A^{-1}) out of the total about 50 Tg A^{-1}shown in Table 3. So far, the globally emitted LNOx nitrogen mass per annum shown in Table 4 is mostly based on observations from ground, satellite and flash extrapolation, storm extrapolation and global lightning frequency. Estimated values shown in Table 4 are in the range of 2 to 9 Tg A^{-1}; however, in spite of considerable progress, the uncertainty remains.

5. Summary

Lightning is an activity of spatiotemporal charge accumulation and distribution between two systems, either cloud-to-cloud or cloud-to-ground, and the process has been explained by the precipitation theory and deep convection theory. However, most studies favor the concept of upper unstable atmospheric condition development due to the convergence of two different air systems, i.e. a warm vertical air column on a cooler upper air column. In addition to the convection and precipitation processes, other atmospheric constituents also affect lightning activity, such as higher values of aerosol loading, inhibiting deep convection and diminishing electrical activity in the cloud system. Some observational studies suggest that warmer surface

Table 3

Overall sources of NOx

Serial no.	Annual NOx sources	T_g N year^{-1}
1	Fossil fuel burning	28–32 (28)
2	Biomass burning	4–24 (10)
3	Lightning	1–20 (5–10)
4	Soil emission	1–16 (5)
5	Aviation	0.7–1 (0.7)
6	N_2O degradation	0.1–1 (0.4)
	Total	~ 50

Source: Schumann and Huntrieser (2007)

Table 4

Best estimate of LNOx and total NOx emissions in reviews and assessments

References	Best estimate of LNOx source rate (and range) in nitrogen mass (T_g A^{-1})
Lawrence et al. (1995)	2 (1–8)
Levy et al. (1996)	4 (2–6)
Lee et al. (1997)	5 (2–20)
Huntrieser et al. (1998)	4 (0.3–22)
WMO (1999)	5 (2–20)
Ehhalt (1999)	7 (4–10)
Bradshaw et al. (2000)	6.5 (2–10)
IPCC (2001)	5 (2–13)
Tie et al. (2002)	3.5–7
Galloway et al. (2004)	5.4
Beirle et al. (2004)	2.8 (0.8–14)
Boersma et al. (2005)	3.5 (1.1–6.4)
Law et al. (2006)	2–9
Schumann and Huntrieser (2007)	5 (2–8)
Huntrieser et al. (2009)	5.7–6.6
Jourdain et al. (2010)	6
Martini et al. (2011)	5
Murray et al. (2012)	5.5–6.5
Miyazaki and Eskes (2014)	4.9–7.7
Liaskos et al. (2015)	2.5–10
Nault et al. (2017)	9

temperatures are correlated with increased lightning across diurnal to inter-annual time scales, although whether such a relationship applies to longer term climate change remains uncertain. Lightning being one of the most energetic natural phenomena happening in the upper tropospheric region, is the most potent source of the oxidized nitrogen in the upper- and mid-tropospheric column. Since nitrogen and oxygen are the most abundant species in the atmosphere, due to extreme temperature inside a lightning strike channel, these species are converted into NO and NO_2 (i.e. collectively called NOx), similar to high-temperature fuel combustion produced NOx. However, this process also produces other trace species such as carbon monoxide (CO) and ozone (O_3) in a smaller amount. In the tropospheric region, photochemical production of ozone depends strongly on the availability of precursors, NOx, CO, CH_4, and non-methane hydrocarbons (NMHC). Since carbon monoxide and methane are more or less produced and balanced by surface processes emission and atmospheric production, the concentration of CO and CH_4 are abundant throughout the troposphere; the only ozone production rate limiting factor left is the availability of reactive oxidized nitrogen species, i.e. NOx (Fehsenfeld and Liu 1994). So far, in the concerned scientific domain it has been concluded that a major source of NOx in the troposphere is nitrogen oxides produced by lightning (e.g. IPCC 1995). The lightning source of NOx is very important for the balance of the oxidized form of nitrogen not only in the troposphere but also in the lower stratosphere. Understanding and quantifying the processes and production of LNOx is important for assessment of future ozone concentrations and its associated impacts on atmospheric chemistry.

Because of spatiotemporal variability, meteorological conditions, lightning events and resultant NOx as a precursor of tropospheric ozone production, is expected to respond to any direct changes to the atmosphere and so in the global climate. Understanding the sensitivity of this response is important for assessment of future LNOx concentrations and associated ozone formation. At the same time, it also becomes a complex task to observe minutely the lightning events due to their very short duration. Thus, an accurate estimation of the amount of NOx produced per flash is still a matter of uncertainty, so is the resultant global budget of LNOx in the atmosphere. In the atmosphere, the present source rate of global NOx (Tg A^{-1}) is dominated by the anthropogenic sources (28 to 32 Tg A^{-1}, IPCC 2001), biomass burning (4–24 Tg A^{-1}), soil (4–16 Tg A^{-1}) and LNOx (1–20 Tg A^{-1}) out of the total about 50 Tg A^{-1}shown in Table 3. So far, the globally emitted

LNOx nitrogen mass per annum shown in Table 4, is mostly based on estimation from ground-based monitoring, observation from aircraft flying near clouds, satellite-based observation on a limited spatial scale (country to continent) and output from various model simulations (chemistry transport model, cloud-resolved-chem model, WRF-chem model and process-based model) on a global scale. All these observations, simulations and estimations are sparse and heterogeneous in time and space.

In this review, a total of 19 research articles available between 2004 and 2020 have been analyzed and concluded with strong confidence of 250 mol NO/flash oscillating between the 17 and 700 mol NO/flash, while a total of 21 research articles available between 1995 and 2017 have been analyzed for the estimate of LNOx source rate in terms of nitrogen-mass per annum and concluded a global annual estimate of 5 ± 3 Tg A^{-1}, which has been considered widely in all these studies. In spite of the considerable progresses in the enhancement of ground-based monitoring network, remote observation and model advancement for the lightning processes and production the uncertainty in global annual estimation remains.

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