Effect of lightning activity on surface NO_x and O_3 over a tropical station during premonsoon and monsoon seasons

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Received 27 September 2011; revised 5 January 2012; accepted 6 January 2012; published 9 March 2012.

[1] We study the effect of lightning activity on surface NO_x and O_3 over Pune, India (18.54°N, 73.81°E) for the years 2005 to 2008 during thunderstorm events in premonsoon and monsoon periods. Surface concentration of NO_x is found to be increased significantly at the dissipation stage of thunderstorms. It is observed that increase in NO_x greater than titration threshold level reduces the surface ozone concentration. However, in some cases when NO_x increases but it does not reach the titration threshold limit, it helps in the production of ozone. Thus, results suggest that lightning production of NO_x inside a thunderstorm can lead to significant impacts on surface ozone concentrations in the tropics. Enhancement in NO_x at the surface after thunderstorm activity is much greater in premonsoon periods compared to the monsoon period.

Citation: Pawar, V., S. D. Pawar, G. Beig, and S. K. Sahu (2012), Effect of lightning activity on surface NO_x and O_3 over a tropical station during premonsoon and monsoon seasons, *J. Geophys. Res.*, *117*, D05310, doi:10.1029/2011JD016930.

1. Introduction

[2] Nitric oxide (NO) is the most important electric-discharge-produced molecule, primarily because it facilitates chemical reactions in the troposphere and stratosphere that determine the concentrations of ozone (O_3) and hydroxyl radical (OH). *Bond et al.* [2002] compared the geographical and seasonal production of NO_x by lightning to other sources like anthropogenic activity, biomass burning, and soil emissions. Their results indicate that production of NO_x by tropical lightning is significant throughout the year. In the troposphere, lightning and aircraft exhaust contribute to ambient NO levels, which can either increase or decrease O_3 levels depending on the local conditions and level of NO_x ($NO_x = NO + NO_2$). Lightning channels in air generate NO primarily as a trace gas with considerably less NO_2. NO once present, is always accompanied by NO_2 produced from oxidation of NO. In the sunlit troposphere, NO_2 is photolyzed (broken into NO and O by light photons); subsequent chemical reactions and the photolyzation of NO_2 establish a balance between NO and NO_2 on a time scale of minutes. In the 1970s, it was established that NO produced by atmospheric electric discharges can indeed play an important role in the chemistry of the Earth's atmosphere, particularly, as noted earlier, in regulating the concentrations of O_3 and OH [*Crutzen*, 1970, 1973; *Chameides and Walker*, 1973]. The first direct observations of enhanced NO concentrations in the vicinity of lightning flashes were made by NO_x *on* [1976, 1978] who used a solar absorption spectrometer to measure NO_2 concentrations below the cloud bases of lightning producing and nonlightning producing storms. He observed increases in NO_2, about 2 orders of magnitude above ambient levels and as high as 100 ppbv (parts per billion by volume). Evidence of NO_x production by lightning was given by airborne measurements in and near mature thunderstorms. Lightning can lead to significant increases in NO_x in the middle and upper troposphere and contribute significantly to columnar abundance of NO_2. [*Choi et al.*, 2005; *Zhang et al.*, 2000, 2003; *Tie et al.*, 2002; *Bond et al.*, 2001; *Pickering et al.*, 1998; *Delmas et al.*, 1997; *Ridley et al.*, 1996; *Beirle et al.*, 2006; *Martin et al.*, 2006]. *Schumann and Huntrieser* [2007] reviewed the global lightning-induced nitrogen oxides source. In the atmospheric boundary layer ozone is generated and removed by catalytic and photochemical reactions involving CO, VOCs (Volatile Organic Compounds) and NO_x. The major oxidants ozone and hydroxyl (OH) are not emitted directly into the atmosphere and their concentrations are determined by complex interplay between dynamics and chemistry which is function of NO_x, CO, H_2O and hydrocarbons. However, NO_x and ozone due to lightning activity for the Indian region has not been studied extensively. It may be noted that monthly average lightning flash counts over Gangetic West Bengal are maximum in the month of May [*Kandalgaonkar et al.*, 2006] which is normally the hottest month of the year in most parts of India. However, the spatial distribution pattern of ozone and its precursors over the Indian subcontinent during this season is mainly controlled by the dynamics and in some cases by episodic events. In this study the data of surface NO_x and ozone on thunderstorm events have been analyzed along with electric field measurements in premonsoon and monsoon seasons.

2. Methodology and Data

2.1. Observation Site Location, Wind, NO_x Emissions

[3] The observational site is situated at the northwest region of Pune city (18.54°N, 73.81°E), which is located at a

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0148-0227/12/2011JD016930

height of 559 m above sea level on the leeside of western Ghats. The site is influenced by the Indian southwest monsoon because its location is close to the Arabian Sea. The site receives southwesterly winds pattern during the southwest monsoon season (June–September), and northeasterly winds during winter (October–January) and some periods of summer (February–May). Figure 1 is a map of India showing the observation site and climatological winds at 925 hPa for a premonsoon month (May) (Figure 1a); and monsoon month (July) averaged for 30 years from 1971 to 2000 taken from NCEP wind reanalysis (Figure 1b).

[4] Figures 2a and 2b show the high-resolution gridded (0.25°) anthropogenic NO_x emissions over Indian region in 2008 from all the sources and from fossil fuel, respectively. Total NO_x emission for India is found to be around 6583 Gg/yr in 2008 as compared to 5723 Gg/yr for 2005 with an increase of around 15% (860 Gg). The spatial pattern shows that maximum NO_x emission is found to be of the order of 80–200 Gg yr^{-1} box^{-1} over western, Indo-Gangetic Plain (IGP) area, eastern, southeastern regions and eastern parts of central India as well. Fossil fuel consumption is major contributor to emissions in Indian cities. Coal used in thermal power stations, transport sector, and industrial practices are the dominant contributors to the above regions, followed by biofuel used for residential purposes in the above discussed regions. The contribution of fossil fuel is found to be around 90% with an increasing trend over many parts of the Indian geographical region. Anthropogenic NO_x emission (~54 Gg/yr) in Pune (the eight largest metropolis in India, (and the second largest city in the state of Maharashtra) is mainly from the transport sector, followed by fossil fuel used in the industrial and residential sectors. An emission of the order of ~39 Gg/yr from transport sector is the major contributor to total NO_x emission in Pune city followed by industrial (~12 Gg/yr) and residential emission (~3.5 Gg/yr). There has been a rise of 33% in NO_x emission since 2005, and future increases are anticipated. Sources of NO_x emission from the transport sector is scattered over city, accompanied by a large number of industrial units [*Ali et al.*, 2009; *Beig and Brasseur*, 2006; *Sahu*, 2010].

2.2. Measuring Instruments

[5] The electric field is measured with an AC field mill kept in a pit with its sensor flush with the ground. It can measure electric field in the range of $\pm$12.5 KV m^{-1} and has a response time of 0.1 ms. The field mill is calibrated in the laboratory by placing a flat circular plate 10 cm above and parallel to another similar grounded plate kept level with the field mill sensors and raising it to different potentials. Its response is found to be linear with the electric field of up to $\pm$12.5 KV m^{-1} applied between the sensors of the field mill and the plate placed above them. Signals from the field mill are amplified and fed through coaxial cables to a data logger which digitizes the signals using a 12-bit analog-to-digital converter for recording and storage of the data at a frequency of 10 Hz. Details of the equipment are given by *Pawar and Kamra* [2002, 2004]. Total flash rate is counted from the electric field record; a field change of at least 600 Vm^{-1} occurring in a period of 2 s being taken as a lightning-induced change [*Pawar and Kamra*, 2002, 2004].

[6] The analyzers used in the present study for the measurements of different pollutants are U.S. EPA (United States Environmental Protection Agency) approved. Ozone is measured using an analyzer (O342M, Environment S.A., France) based on the UV absorption technique, using a wavelength of UV radiation at 253.7 nm. The analyzer automatically incorporates the corrections due to changes in temperature and pressure in the absorption cell and drift in the intensity of UV Lamp. The lower detection limit of the analyzer is 1 ppbv with noise of about 0.5 ppbv, under 10 s response times. The instrument recorded the data in the range of resolution 1 min to 24 h. In these study hourly averages of concentration is used. The NO_x is measured with the Horiba, Japan made APNA 365 analyzer based on the technique of chemiluminescence. Most of the NO_x measurements depend upon some kind of surface conversion device to convert NO_2 to NO (Molybdenum in the present case), which can then be detected by chemiluminescence. It has been well established now that in the molybdenum converter instruments other gas phase nitrogen containing compounds are converted by molybdenum oxide catalysts to NO and therefore can be reported as NO_2 [*Winer et al.*, 1974]. Therefore, many studies [*Lamsal et al.*, 2008, 2010; *Dunlea et al.*, 2007] have shown that large positive bias can exist in the measurements of NO_x done by such instruments. Even though, in our instrument no attempt has done to find a bias in the measurements, however as the bias is mainly due to the concentrations of nitrogen containing compounds, which may not vary on day to day basis. Therefore, we think that bias will be same for thunderstorm and nonthunderstorm days; moreover most of the thunderstorms have occurred during same time of the day. Therefore, we feel that biases in the measurements may not affect much on the conclusions of this study. This instrument records the data from the intervals of 3 min to 3 h. In this study data is sampled at 3 min interval and averaged for 1 h. The lower detection limit of the analyzer is 100 pptv. However, most of the observed values are found to be well above this limit, which provides very large signal-to-noise ratio for corrected measurements. On thunderstorm and nonthunderstorm days we analyze surface measurements of electric field, NO_x and O_3, which show the influence of thunderstorm activity on the above parameters.

3. Results

[7] The region around Pune generally experiences thunderstorms during the premonsoon season (March–June) and on initial days of postmonsoon (September–November) months, whereas thunder is rarely observed during the monsoon period (July–August). The development of isolated thunderstorms in this area in the premonsoon season is generally due to instability of the lower troposphere created by surface heating. Base height of these thunderstorms is generally 1 to 2 km. Thunderstorms in this region are generally stationary.

3.1. Thunderstorms During the Premonsoon

[8] Over the Pune region thunderstorms are frequently observed during April to May. Table 1 shows the detail statistical information about total number of events studied during the premonsoon and monsoon in the period of 2005–2008. We analyzed a total of 60 events of thunderstorms of which, 45 cases belong to premonsoon season and 15 to monsoon season. In this study we analyzed electrical

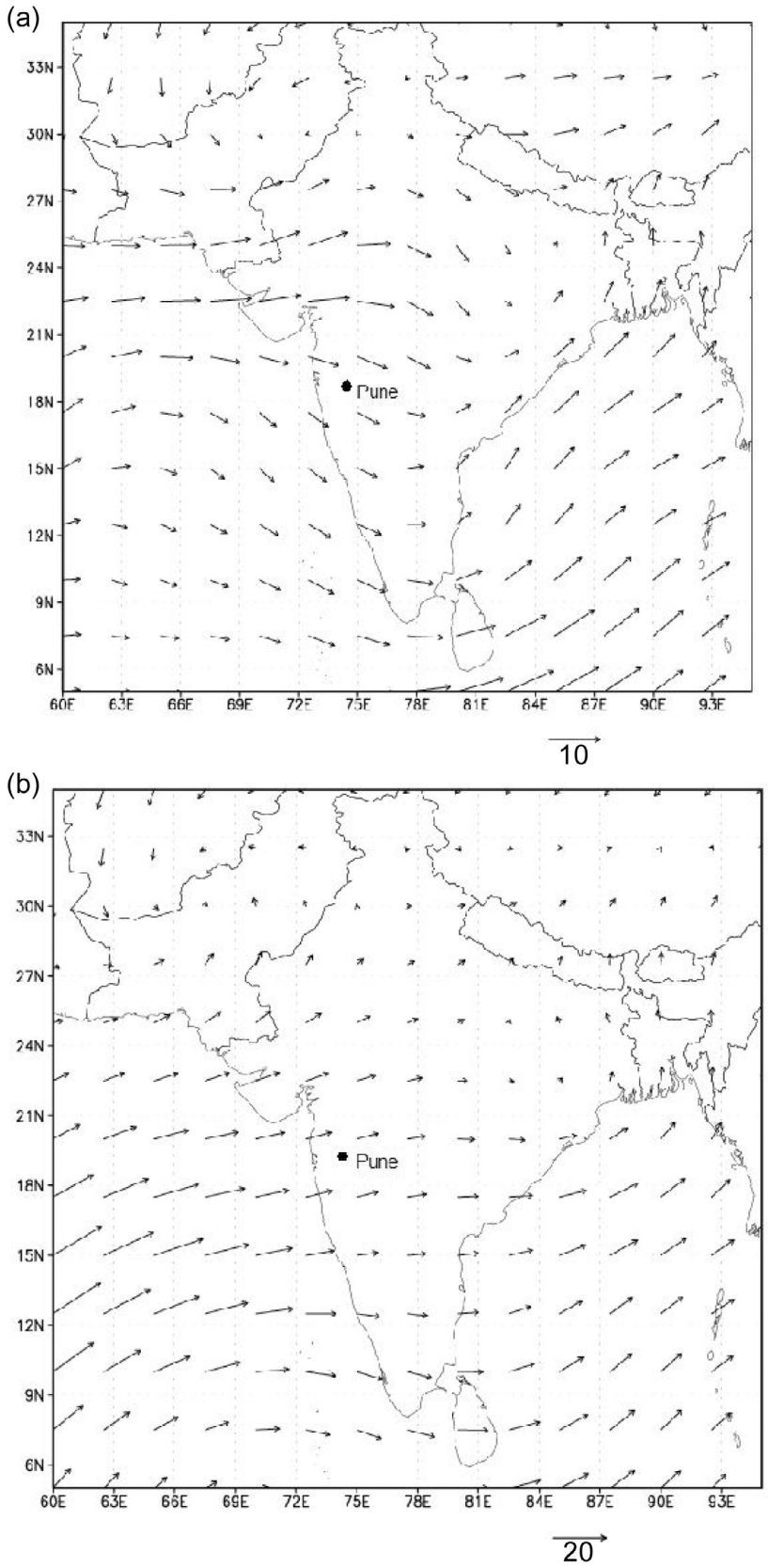

Figure 1. Map of India showing position of observation site and climatological winds over India for (a) a premonsoon month May and (b) a monsoon month July averaged for 30 years from 1971 to 2000 at 925 mb taken from NCEP wind reanalysis.

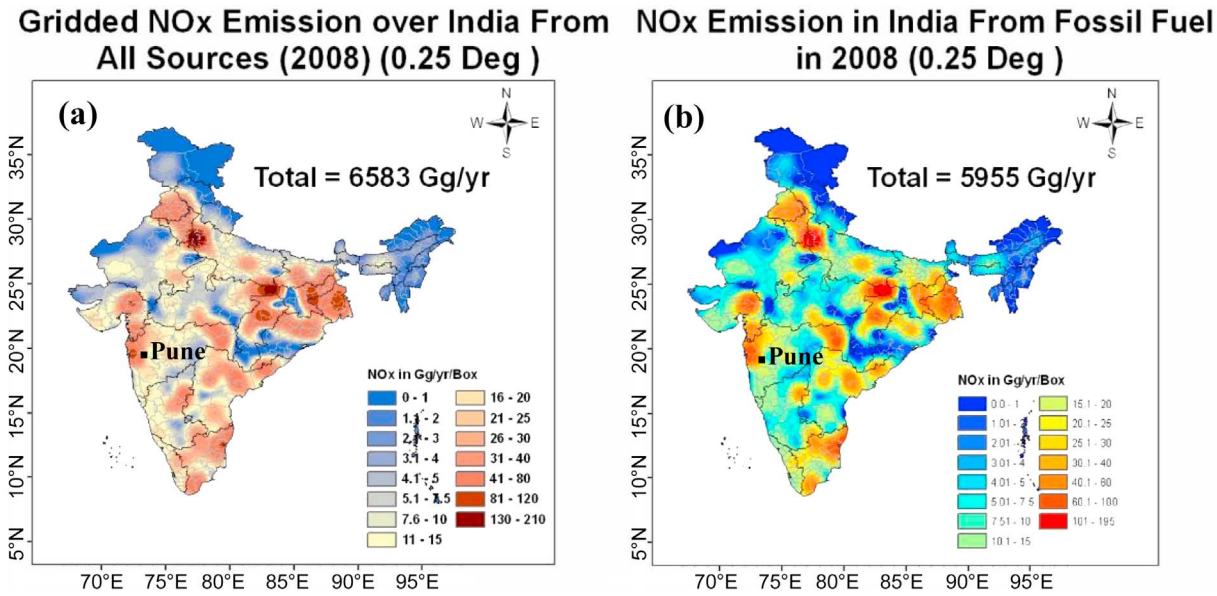

Figure 2. High-resolution gridded (0.25°) NO_x emission in India in 2008 from (a) all sources and (b) fossil fuel.

parameters (lightning flash rate, electric field) with the chemical species (NO_x, O_3) during the thunderstorm activity using the thunderstorm and nonthunderstorm day data. Figures 3a and 3b show the lightning flash rate per five minutes and electric field during thunderstorm observed on 3 May 2005, respectively. Lightning flash rate has been derived from electric field data. Figures 4a and 4b show diurnal variations of NO_x and ozone observed on 3 May 2005 as well as 5 days before and after the thunderstorm day (nonthunderstorm days). The thunderstorm developed southeast of the observatory at about 16:30 h Local Time (LT). As shown in Figure 3b the electric field became positive and increased rapidly around 17:30 and lightning activity began. Intense lightning activity was observed for 2 h from 17:10 to 19:10 with decreases in frequency thereafter. The lightning activity stopped completely by 19:15 h LT. It is well known that the lightning is a major source of NO_x in the middle and upper troposphere and contributes significantly to abundance of NO_2 [*MacGorman and Rust*, 1998; *Choi et al.*, 2005; *Beirle et al.*, 2006; *Martin et al.*, 2006; *Schumann and Huntrieser*, 2007]. However, there are only a few studies about surface variation of NO_x and ozone associated with thunderstorms [*Betts al.*, 2002; *Ott et al.*, 2010; *Minschwaner et al.*, 2008]. The lightning frequency and electric field records shown in Figures 3a and 3b clearly suggest that the dissipation stage of this thunderstorm had started at about at 19:00 h. The concentrations of NO_x and ozone observed on nonthunderstorm days are considered as representative of the background levels. As shown in Figure 4a NO_x concentration (14 ppb) after 2–3 h (21.00) of peak lightning activity is more than the twofold compared to the nonthunderstorm days. This increase in NO_x suggests the transport of NO_x from the upper or middle troposphere by downdraft of the thunderstorm during its dissipation stage. Figure 4b shows the variation of ozone on 3 May 2005 thunderstorm day with 5 days before and after the thunderstorm day (nonthunderstorm days). A study by *Minschwaner et al.* [2008] shows enhancements in ozone between about 3 and 10 km altitude within an electrically active storm in central New Mexico. However, our analysis shows a decrease in ozone following the dissipation stage of thunderstorm (Figure 4b). Several studies have shown that the impacts of NO_x on ozone are complex and nonlinear [*Lin et al.*, 1988]. The decrease of ozone following the thunderstorm on 3 May 2005 can be attributed due to the "titration reaction" which became dominant only after the NO_x titration threshold is reached.

Table 1. Present Concentrations of NO_x and O_3 for Premonsoon and Monsoon Thunderstorm Events

Season	Number of Cases Observed	Concentration Range		
			Thunderstorm Period[a]	
		NO_x Limit	NO_x (ppb)	O3 (ppb)
Premonsoon	10	NO_x above threshold	+19 (±10.35)	–5.625 (±3.92)
	35	NO_x below threshold	+2.26 (±1.6)	+10.87 (±3.93)
Monsoon	Not Found	NO_x above threshold	-	-
	15	NO_x below threshold	+1.91 (±1.11)	+9.31 (±1.25)

[a]Values outside parentheses are average values; values within parentheses are standard deviation.

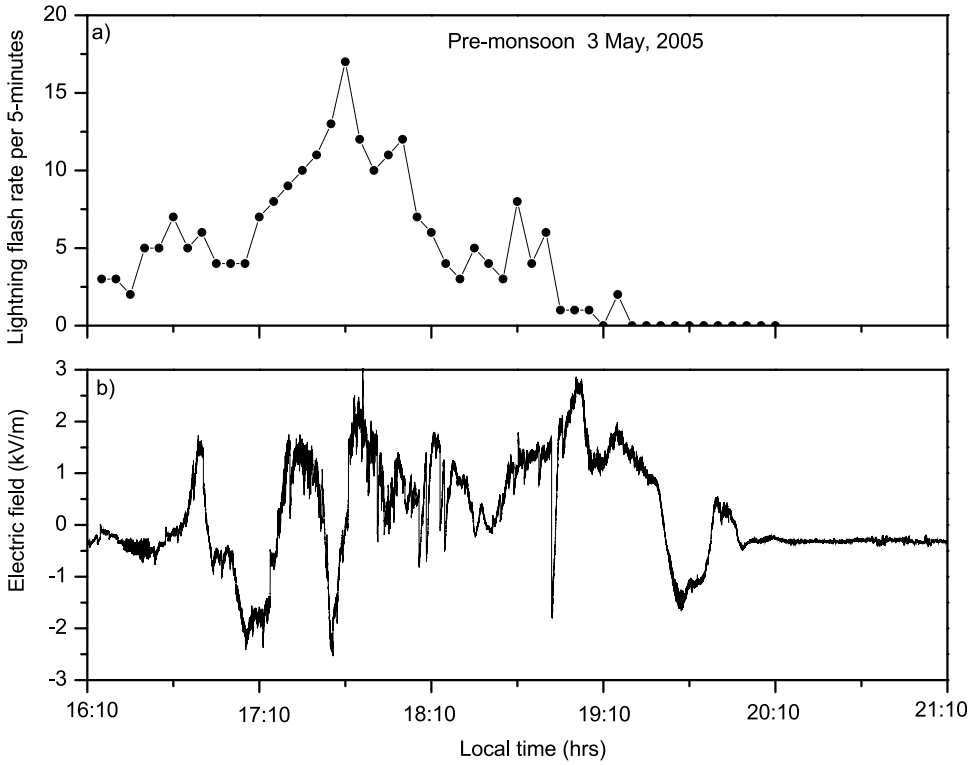

Figure 3. (a) Lightning flash rate per 5 min. (b) Nature of electric field with respect to hourly time scale during thunderstorm on 3 May 2005.

Ozone production is typically associated with hydrocarbon oxidation that produces HO_2 or peroxy radicals (RO_2) such that

(R1) $$HO_2 + NO \rightarrow OH + NO_2$$

(R2) $$RO_2 + NO \rightarrow RO + NO_2$$

Combined with reactions (R3) and (R4), these reactions convert NO to NO_2 without the consumption of ozone, resulting in net ozone production.

(R3) $$NO + O_3 \rightarrow O_2 + NO_2$$

$$NO_2 + \text{Sunlight} \rightarrow NO + O$$

(R4) $$O + O_2 \rightarrow O_3$$

O_3 production increases linearly with hydrocarbon concentrations but varies inversely with NO_x concentrations (hydrocarbon-limited regime) because the O_3 production rate is limited by the supply of hydrocarbons. The dependence of O_3 production on NO_x and hydrocarbons is very different between the two regimes. Ozone concentrations (ppbv) simulated by a regional photochemical model is a function of NO_x and hydrocarbon emissions [*Sillman et al.*, 1990]. In the NO_x-limited regime, hydrocarbon emission controls are of no benefit for decreasing O_3. In the hydrocarbon-limited regime, NO_x emission controls cause an increase in O_3. O_3 production varies linearly with the NO concentration but is independent of hydrocarbons (NO_x-limited regime) because the O_3 production rate is limited by the supply of NO_x. On a thunderstorm and nonthunderstorm day peak of O_3 occurs during afternoon hours (12:00–15.00) because of strong solar radiation (Figure 4b). After 2–3 h of thunderstorm activity (21.00 h) NO_x slowly increases and reaches concentration (14 ppb) above the titration threshold level and starts to destroys the O_3 ($\sim$15 ppb) via titration reaction. Peak of NO_x and dip of O_3 at 21:00 h on thunderstorm day is clearly seen in Figures 4a and 4b because of thunderstorm and lightning activity. However, the threshold limit is not fixed and varies from region to region. Based on the long-term data and their analysis as reported earlier from the same station [*Beig and Brasseur*, 2006; *Beig et al.*, 2007], the threshold limit is estimated to be between 12 and 20 ppb [*Beig et al.*, 2010].

[9] We discuss one more case of premonsoon thunderstorm observed on 3 June 2008. This thunderstorm developed at about 16:00 h LT local time and lasted for about 2 h; the flash rate along with 5 min average of electric field is shown in Figures 5a and 5b. Lightning flash rate increased very rapidly and reached 120 flashes per 5 min during the active stage of the thunderstorm and then decreased gradually to less than one flash per minute at about 18:00 h. As shown in Figure 5b with the onset of the thunderstorm, the electric field increased to 2 kV/m and lightning activity started at about 16:20 h. In Figures 6a and 6b we have plotted diurnal variations of NO_x and ozone observed on 3 June 2008 as well as 5 days before and after the day of the thunderstorm (nonthunderstorm days). As shown in Figure 6a, NO_x shows considerable increase at 21:00 h. This considerable increase in NO_x observed on 3 June 2008 at 21:00 h is clearly due to transport of NO_x from the upper or middle troposphere

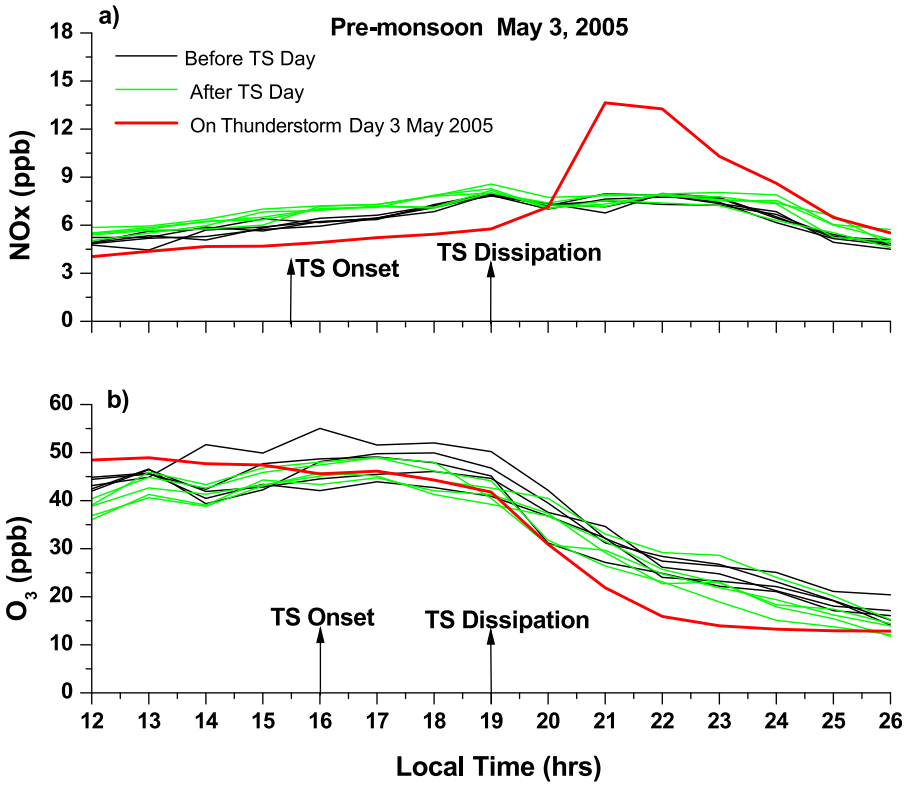

Figure 4. Comparative plot of surface (a) NO_x and (b) O_3 on thunderstorm day (3 May 2005) (red line) with 5 days (28 April to 2 May 2005) before thunderstorm, fair weather days (black line), and 5 days (4–8 May 2005) after thunderstorm fair weather days (green line).

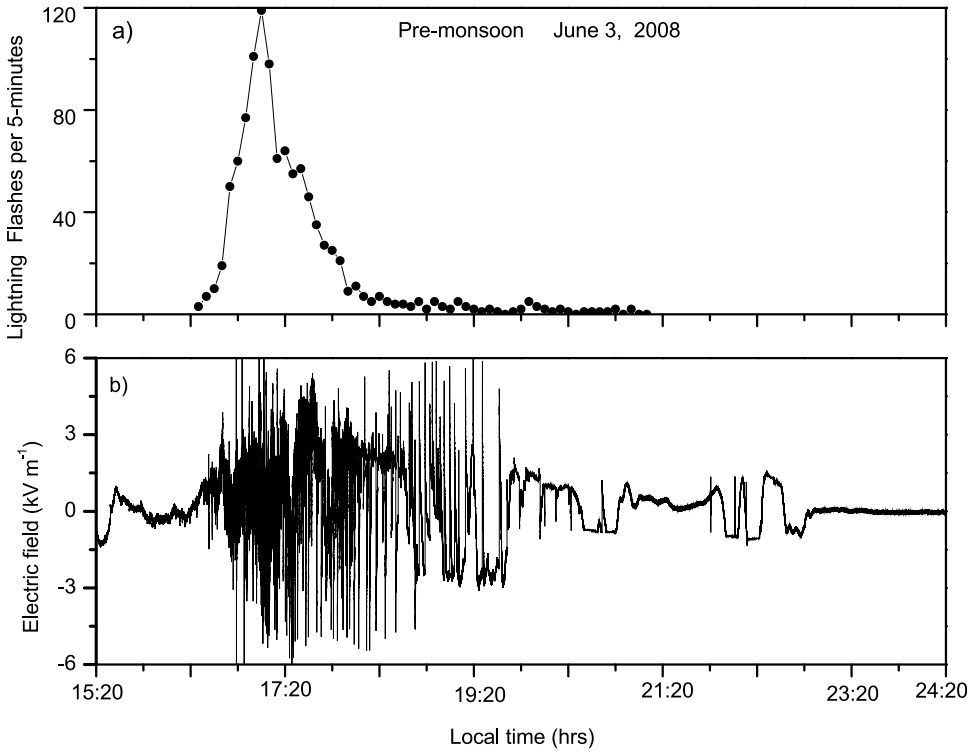

Figure 5. (a) Lightning flash rate per 5 min. (b) Nature of electric field with respect to hourly time scale during thunderstorm activity on 3 June 2008.

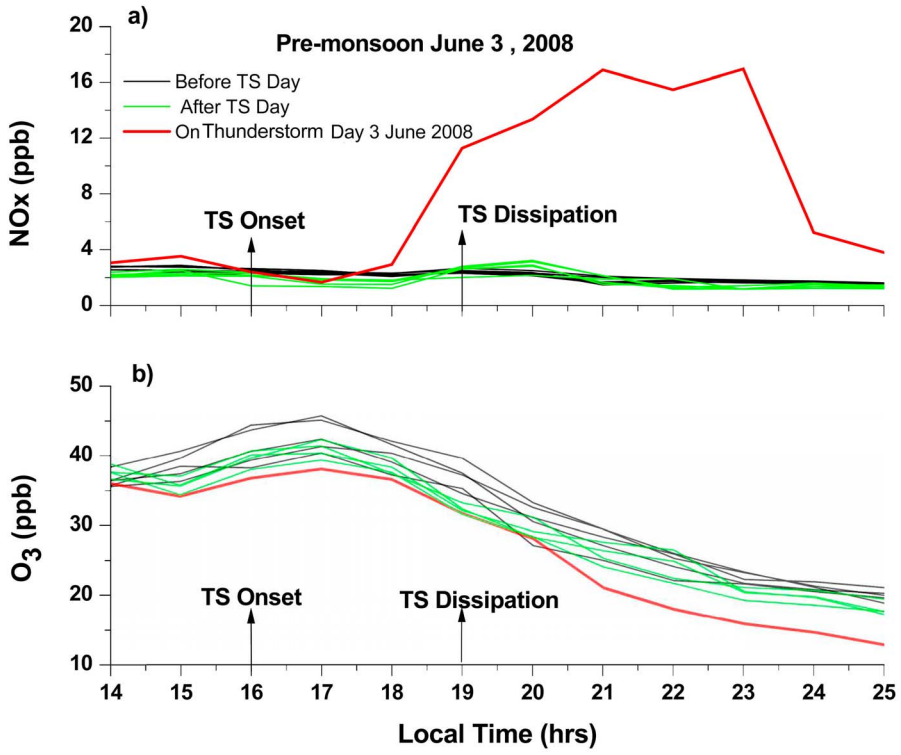

Figure 6. Comparative plot of surface (a) NO_x and (b) O_3 on thunderstorm day (3 June 2008) (red line) with 5 days (29 May to 2 June 2008) before thunderstorm, fair weather days (black line), and 5 days (4–8 June 2008) after thunderstorm fair weather days (green line).

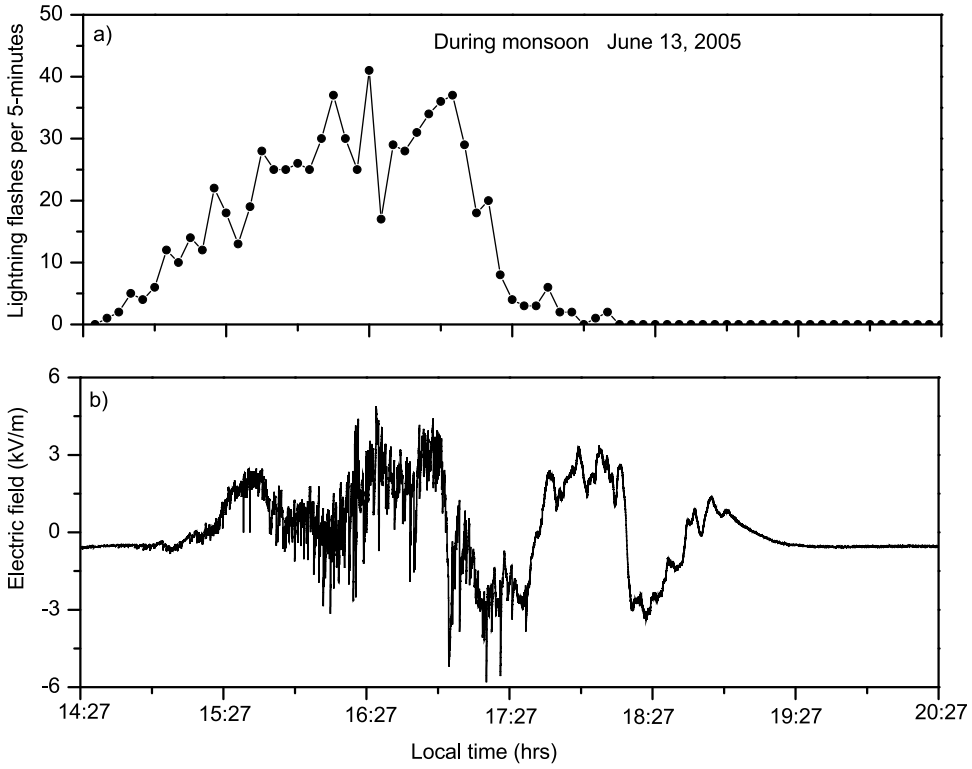

Figure 7. (a) Lightning flash rate per 5 min. (b) Nature of electric field with respect to hourly time scale during thunderstorm activity on 13 June 2005.

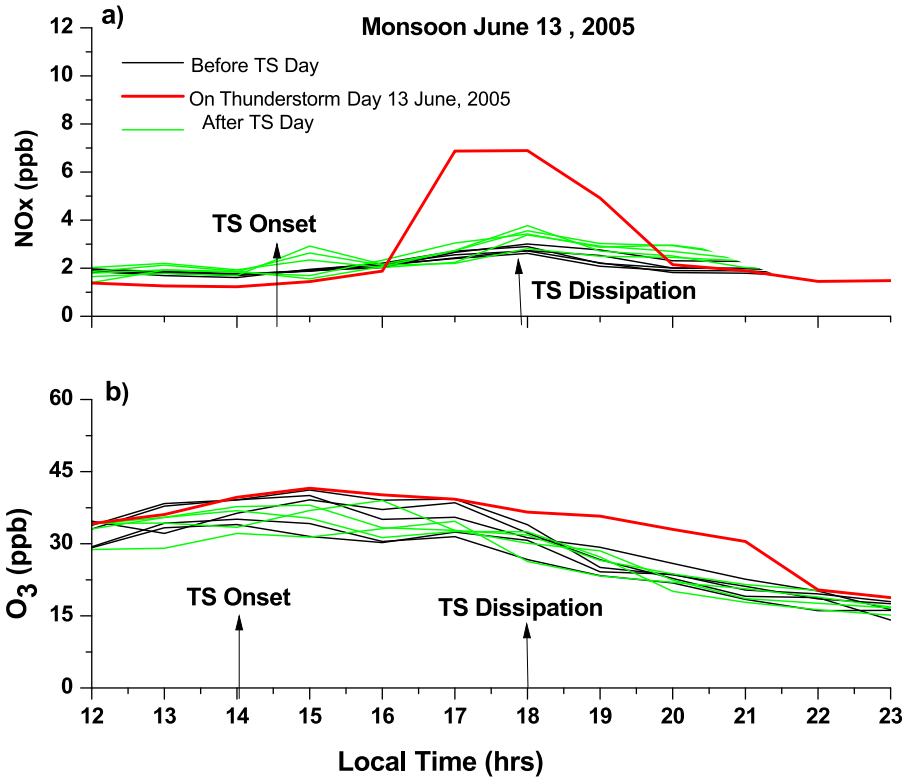

Figure 8. Comparative plot of surface (a) NO_x and (b) O_3 on thunderstorm day (13 June 2005) (red line) with 5 days (8–12 June 2005) before thunderstorm, fair weather days (black line), and 5 days (14–18 June 2005) after thunderstorm fair weather days (green line).

with the downdraft of the thunderstorm during its dissipation stage. Figure 6b shows the variation of ozone on 3 June 2008 and 5 days variations of ozone before and after thunderstorm day. During the dissipation stage (20:00–21:00 h) of the thunderstorm, the surface NO_x (17 ppb) increases sharply and at the peak level becomes four times more than the normal fair weather days (Figure 6a). The increase at peak hours is found to be from 4 ppb for nonthunderstorm days (fair weather days) to 17 ppb during thunderstorm hours. The rate of increase is high in the magnitude of NO_x concentration which is close to titration threshold limit. As a result the ozone level drops down due to the titration reaction. We did not find any enhancement in ozone during the active period of the thunderstorm (Figures 4b and 6b) as also reported earlier by *Minschwaner et al.* [2008]. In fact it is observed that in most of the cases, ozone showed low concentrations during the active period of thunderstorms. This decrease in concentration may be due to reduced solar radiation because of the presence of thunderclouds. However, it is observed that there is a reduction in ozone during the dissipation stage of the thunderstorm as seen in Figures 4b and 6b. During premonsoon season 10 out of 45 cases shows that there is large enhancement in NO_x concentration above the titration threshold during or after the dissipation of the thunderstorm and reduction in surface ozone concentration.

3.2. Thunderstorms During Monsoon

[10] Indian summer monsoon onset occurs in the first week of June over the Pune region, and thunderstorms are rarely observed during July to September. About 15 cases of thunderstorms are observed during this period. The thunderstorms formed in this period are low in intensity because of weak heat convection during the monsoon period. Figure 7a shows the lightning flash rate per five minutes observed on 13 June 2005 and Figure 7b shows the electric field recorded during a monsoon thunderstorm observed on the same day. The thunderstorm developed southeast of observatory around 14:40 h. The electric field became positive and increased rapidly at about 14:55 h and lightning activity began. Intense lightning activity was observed for 2 h from 15:30 h to 17:30 h, and then stopped completely at about 18:00 h. Diurnal variations of NO_x and O_3 observed on 13 June 2005 along with 5 days before and after the thunderstorm day (nonthunderstorm days) are shown in Figures 8a and 8b. On 13 June 2005, thunderstorm developed little early in the afternoon, i.e., at 14:40 h. On that day during dissipation stage of thunderstorm NO_x increased considerably at 17.00 h, its magnitude reached up to 7 ppb and remained less than the titration threshold limit as a result ozone increased.

4. Discussion

[11] Generally, in the tropics thunderstorms develop in barotropic environments with very small vertical wind shear and therefore, the updraft and thus the growth of the storm is nearly vertical [*Williams*, 2001]. The accumulated condensation in the upper regions of storm frequently descends in place to create the low-level downdraft. In the extratropics, thunderstorms develop in baroclinic environments with large vertical wind shear [*Williams*, 2001]. Therefore most of the

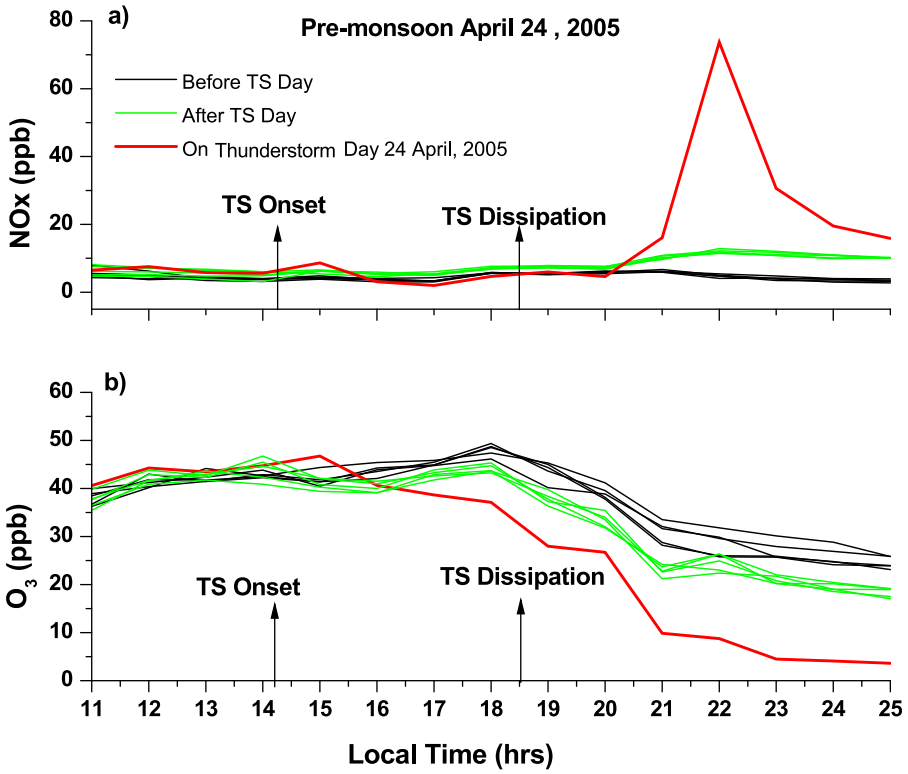

Figure 9. Comparative plot of surface (a) NO_x and (b) O_3 on thunderstorm day (24 April 2005) (red line) with 5 days (19–23 April 2005) before thunderstorm, fair weather days (black line), and 5 days (25–29 April 2005) after thunderstorm fair weather days (green line).

time dissipation stages of such thunderstorms are of shorter duration compared to the extratropical thunderstorms. *Pawar and Kamra* [2007] have shown that dissipation time for thunderstorms over Pune (India) are 24% of those for the large quasi-stationary thunderstorms occurring at Florida and 55% of the convective air mass thunderstorms at New Mexico. Therefore, it is expected that the NO_x transported to the surface, below tropical thunderstorms in the dissipation stage can be considerably higher than in extratropical thunderstorms. The observations discussed here show a many fold increase in NO_x during the dissipation stage of thunderstorms and in some cases very large increases have been observed. On 24 April 2005 thunderstorm NO_x concentration increase (70 ppb) ten times more than the background. *Noxon* [1976, 1978] observed that lightning flashes increases NO_2, about 2 orders of magnitude above ambient levels and as high as 100 ppbv (parts per billion by volume). *Bond et al.* [2002] found that the production of NO_x by tropical lightning is significant throughout the year. Lightning accounts for almost all of the NO_x emitted over the oceans and 50–90% of NO_x emitted over some continental areas on a seasonal basis [*Bond et al.*, 2002]. The large amounts of fresh NO_x emissions do not immediately contribute to photochemical ozone production and can lead to ozone decrease by both direct titration and nighttime chemistry that further reduces ozone. The observed NO_x concentration at the surface is higher than what it would be for an annual average as the summer months are the lightning-active times of the year. On 13 June 2005 (Figure 8) before and during the thunderstorm activity NO_x concentration is almost same as a nonthunderstorm day, but after 1–3 h of thunderstorm activity NO_x concentration increases more than twofold of nonthunderstorm day.

[12] Table 1 presents concentrations of NO_x and O_3 for premonsoon and monsoon thunderstorm events. For each thunderstorm event, we have calculated the difference in NO_x between the peak NO_x after the storm activity on thunderstorm day and the NO_x on nonthunderstorm day for the same hour after the storm (not for anytime during the day). The averages of these differences were then calculated for each season and NO_x regime, and the results are presented in Table 1. The above method was also applied to the ozone measurements. For each thunderstorm event, we have calculated the difference in the O_3, when the peak in NO_x was observed on thunderstorm day hours and nonthunderstorm day for the same hour as taken for the storm day. It should be noted here that in Table 1 we present the differences in values of peak NO_x and minimum of O_3 after the storm, not for anytime during the day.

[13] During premonsoon, 10 out of 45 events showed that NO_x (+19 ($\pm$10.35)) increases above titration threshold limit. This enhanced NO_x affect the O_3 and reduces O_3 ($-$5.625 ($\pm$3.92)) concentration via titration reaction. In these 10 cases fourfold increase is found in NO_x on thunderstorm days compared to that on the nonthunderstorm days NO_x. On the other hand during premonsoon 35 cases show that NO_x (+2.26 ($\pm$1.6)) increases but remains below titration threshold value. This enhanced NO_x helps to increase the O_3 (+10.87 ($\pm$3.93)) concentration. During monsoon 15 cases show that NO_x increases but remains below titration threshold value. This enhanced NO_x helps to increase the O_3

concentration. During monsoon not a single case found which show NO_x increases above titration threshold value.

[14] The ozone concentration show sharp decrease exactly when, NO_x reaches maximum and crosses a threshold limit (Figures 4, 6, and 9). However, in some cases the ozone concentration does not show much variation from its normal values. It has been observed that premonsoon thunderstorms are more efficient in lightning production than the monsoon thunderstorms [*Williams et al.*, 1992; *Lal and Pawar*, 2009]. Enhancement in NO_x after thunderstorm activity is much greater in the premonsoon period compared to that in the monsoon period (Table 1). Lightning activity in a thunderstorm is closely related to storm dynamics. In the severe thunderstorms lightning frequency is high [*Williams*, 2001] and therefore more number of electric discharges gives the large amount of NO_x [*Zhang et al.*, 2000, 2003].

5. Conclusions

[15] Present work shows the effect of lightning activity on surface NO_x and O_3 over a tropical station during premonsoon and monsoon seasons. Results indicate that if NO_x concentrations cross the titration threshold level it reduces the ozone concentration. However, if enhanced NO_x levels remain below the titration threshold level then it either contributes to elevated ozone levels or does not make any impact on surface ozone, depending on meteorological conditions.

[16] **Acknowledgments.** The author Vidya S. Pawar would like to acknowledge the motivation and support from P. N. Sen and P. Pradeep Kumar, Department of Atmospheric and Space Sciences, University of Pune, India.

References

Ali, K., G. Beig, D. M. Chate, G. A. Momin, S. K. Sahu, and P. D. Safai (2009), Sink mechanism for significantly low level of ozone over the Arabian Sea during monsoon, *J. Geophys. Res.*, *114*, D17306, doi:10.1029/2008JD011256.

Beig, G., and G. Brasseur (2006), Influence of anthropogenic emissions on tropospheric ozone and its precursors over the Indian tropical region during a monsoon, *Geophys. Res. Lett.*, *33*, L07808, doi:10.1029/2005GL024949.

Beig, G., S. Gunthe, and D. B. Jadhav (2007), Simultaneous measurements of ozone and its precursors on a diurnal scale at a semi-urban site in Pune, *J. Atmos. Chem.*, *57*, 239–253, doi:10.1007/s10874-007-9068-8.

Beig, G., H. K. Trimbake, and S. H. Kulkarni (2010), Climatology of ozone based on the base line data of ozone and its precursors at Pune, technical report, 109 pp., Indian Inst. of Technol. Madras, Madras, India.

Beirle, S., et al. (2006), Estimating the NO_x produced by lightning from GOME and NLDN data: A case study in the Gulf of Mexico, *Atmos. Chem. Phys.*, *6*, 1075–1089, doi:10.5194/acp-6-1075-2006.

Betts, A. K., J. D. Fuentes, M. Garstang, and J. H. Ball (2002), Surface diurnal cycle and boundary layer structure over Rondônia during the rainy season, *J. Geophys. Res.*, *107*(D20), 8065, doi:10.1029/2001JD000356.

Bond, D. W., R. Zhang, X. Tie, G. Brasseur, G. Huffines, R. E. Orville, and D. J. Boccippio (2001), NO_x production by lightning over the continental United States, *J. Geophys. Res.*, *106*, 27,701–27,710, doi:10.1029/2000JD000191.

Bond, D. W., S. Steigera, R. Zhang, X. Tie, and R. E. Orville (2002), The importance of NO_x production by lightning in the tropics, *Atmos. Environ.*, *36*, 1509–1519, doi:10.1016/S1352-2310(01)00553-2.

Chameides, W. L., and J. C. G. Walker (1973), A photochemical theory of tropospheric ozone, *J. Geophys. Res.*, *78*, 8751–8760, doi:10.1029/JC078i036p08751.

Choi, Y., Y. Wang, T. Zeng, R. V. Martin, T. P. Kurosu, and K. Chance (2005), Evidence of lightning NO_x and convective transport of pollutants in satellite observations over North America, *Geophys. Res. Lett.*, *32*, L02805, doi:10.1029/2004GL021436.

Crutzen, P. J. (1970), The influence of nitrogen oxides on the atmospheric ozone content, *Q. J. R. Meteorol. Soc.*, *96*, 320–325, doi:10.1002/qj.49709640815.

Crutzen, P. J. (1973), A discussion of the chemistry of some minor constituents in the stratosphere and troposphere, *Pure Appl. Geophys.*, *106*, 1385–1399, doi:10.1007/BF00881092.

Delmas, R., D. Serca, and C. Jambert (1997), Global inventory of NO_x sources, *Nutr. Cycl. Agroecosyst.*, *48*, 51–60, doi:10.1023/A:1009793806086.

Dunlea, E. J., et al. (2007), Evaluation of nitrogen dioxide chemiluminescence monitors in a polluted urban environment, *Atmos. Chem. Phys.*, *7*, 2691–2704, doi:10.5194/acp-7-2691-2007.

Kandalgaonkar, S. S., M. I. R. Tinmaker, M. K. Kulkarni, and A. Nath (2006), Lightning and rainfall activity over Gangetic West Bengal, *J. Atmos. Electr.*, *26*, 37–50.

Lal, D. M., and S. D. Pawar (2009), Relationship between rainfall and lightning over central Indian region in monsoon and pre-monsoon seasons, *Atmos. Res.*, *92*, 402–410, doi:10.1016/j.atmosres.2008.12.009.

Lamsal, L. N., R. V. Martin, A. van Donkelaar, M. Steinbacher, E. A. Celarier, E. Bucsela, E. J. Dunlea, and J. P. Pinto (2008), Ground-level nitrogen dioxide concentrations inferred from the satellite-borne Ozone Monitoring Instrument, *J. Geophys. Res.*, *113*, D16308, doi:10.1029/2007JD009235.

Lamsal, L. N., R. V. Martin, A. van Donkelaar, E. A. Celarier, E. J. Bucsela, K. F. Boersma, R. Dirksen, C. Luo, and Y. Wang (2010), Indirect validation of tropospheric nitrogen dioxide retrieved from the OMI satellite instrument: Insight into the seasonal variation of nitrogen oxides at northern mid-latitudes, *J. Geophys. Res.*, *115*, D05302, doi:10.1029/2009JD013351.

Lin, X., M. Trainer, and S. C. Liu (1988), On the nonlinearity of the tropospheric ozone production. *J. Geophys. Res.*, *93*(D12), 15,879–15,888, doi:10.1029/JD093iD12p15879.

MacGorman, D. R., and W. D. Rust (1998), *The Electrical Nature of Storms*, 422 pp., Oxford Univ. Press, Oxford, U. K.

Martin, R. V., C. E. Sioris, K. Chance, T. B. Ryerson, T. H. Bertram, P. J. Wooldridge, R. C. Cohen, J. A. Neuman, A. Swanson, and F. M. Flocke (2006), Evaluation of space-based constraints on global nitrogen oxide emissions with regional aircraft measurements over and downwind of eastern North America, *J. Geophys. Res.*, *111*, D15308, doi:10.1029/2005JD006680.

Minschwaner, K., L. E. Kalnajs, M. K. Dubey, L. M. Avallone, P. C. Sawaengphokai, H. E. Edens, and W. P. Winn (2008), Observation of enhanced ozone in an electrically active storm over Socorro, NM: Implications for ozone production from corona discharges, *J. Geophys. Res.*, *113*, D17208, doi:10.1029/2007JD009500.

Noxon, J. F. (1976), Atmospheric nitrogen fixation by lightning, *Geophys. Res. Lett.*, *3*, 463–465, doi:10.1029/GL003i008p00463.

Noxon, J. F. (1978), Tropospheric NO_2, *J. Geophys. Res.*, *83*, 3051–3057, doi:10.1029/JC083iC06p03051.

Ott, L. E., K. E. Pickering, G. L. Stenchikov, D. J. Allen, A. J. DeCaria, B. Ridley, R. F. Lin, S. Lang, and W. Tao (2010), Production of lightning NO_x and its vertical distribution calculated from three-dimensional cloud-scale chemical transport model simulations, *J. Geophys. Res.*, *115*, D04301, doi:10.1029/2009JD011880.

Pawar, S. D., and A. K. Kamra (2002), Recovery curves of the surface electric field after lightning discharges occurring between the positive charge pocket and negative charge center in a thundercloud, *Geophys. Res. Lett.*, *29*(23), 2108, doi:10.1029/2002GL015675.

Pawar, S. D., and A. K. Kamra (2004), Evolution of lightning and the possible initiation/triggering of lightning discharge by the lower positive charge centre in an isolated thundercloud in tropics, *J. Geophys. Res.*, *109*, D02205, doi:10.1029/2003JD003735.

Pawar, S. D., and A. K. Kamra (2007), The end-of-storm oscillation in tropical air mass thunderstorms, *J. Geophys. Res.*, *112*, D03204, doi:10.1029/2005JD006997.

Pickering, K. E., Y. Wang, W.-K. Tao, C. Price, and J.-F. Müller (1998), Vertical distribution of lightning NO_x for use in regional and global chemical transport models, *J. Geophys. Res.*, *103*, 31,203–31,216.

Ridley, B. A., J. E. Dye, J. G. Walega, J. Zheng, F. E. Grahek, and W. Rison (1996), On the production of active nitrogen by thunderstorms over New Mexico, *J. Geophys. Res.*, *101*, 20,985–21,005, doi:10.1029/96JD01706.

Sahu, S. K. (2010), Emission inventories of major atmospheric pollutants for India using GIS technique and their impact on the distribution of tropospheric ozone, PhD thesis, Univ. of Pune, Pune, India.

Schumann, U., and H. Huntrieser (2007), The global lightning-induced nitrogen oxides source, *Atmos. Chem. Phys.*, *7*, 3823–3907, doi:10.5194/acp-7-3823-2007.

Sillman, S., J. A. Logan, and S. C. Wofsy (1990), The sensitivity of ozone to nitrogen oxides and hydrocarbons in regional ozone episodes, *J. Geophys. Res.*, *95*, 1837–1851, doi:10.1029/JD095iD02p01837.

Tie, X. X., R. Y. Zhang, G. Brasseur, and W. F. Lei (2002), Global NO_x production by lightning, *J. Atmos. Chem.*, *43*, 61–74, doi:10.1023/A:1016145719608.

Williams, E. R. (2001), The electrification of severe storms, *Meteorol. Monogr.*, *28*(50), 527–528, doi:10.1175/0065-9401-28.50.527.

Williams, E. R., S. A. Rutledge, S. C. Geotis, N. Renno, E. Rasmussen, and T. Rickenbach (1992), A radar and electrical study of tropical hot tower, *J. Atmos. Sci.*, *49*, 1386–1395, doi:10.1175/1520-0469(1992)049<1386:ARAESO>2.0.CO;2.

Winer, A. M., J. W. Peters, J. P. Smith, and J. N. Pitts Jr. (1974), Response of commercial chemiluminescent NO-NO_2 analyzers to other nitrogen containing compounds, *Environ. Sci. Technol.*, *8*, 1118–1121, doi:10.1021/es60098a004.

Zhang, R., N. T. Sanger, R. E. Orville, X. Tie, W. Randel, and E. R. Williams (2000), Enhanced NO_x by lightning in the upper troposphere and lower stratosphere inferred from the UARS global NO_2 measurements, *Geophys. Res. Lett.*, *27*, 685–688, doi:10.1029/1999GL010903.

Zhang, R., X. X. Tie, and D. W. Bond (2003), Impacts of anthropogenic and natural NO_x sources over the U.S. on tropospheric chemistry, *Proc. Natl. Acad. Sci. U. S. A.*, *100*, 1505–1509, doi:10.1073/pnas.252763799.

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