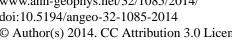
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Analysis of cloud-to-ground lightning and its relation with surface pollutants over Taipei, Taiwan

S. K. Kar^{1,*} and Y. A. Liou^{1,*}

Correspondence to: S. K. Kar (sanjib_kar@hotmail.com) and Y. A. Liou (yueian@csrsr.ncu.edu.tw)

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Abstract. Premonsoon (March–April) cloud-to-ground (CG) lightning activity over Taipei, Taiwan, is analyzed in relation to surface pollutants like particulate matter (PM₁₀), sulfur dioxide (SO₂), nitrogen oxides (NO_x) and ozone (O₃) concentration for a period of 6 years (2005–2010). Other surface parameters like aerosol optical depth and cloud top temperature are also investigated taking data from Moderate Resolution Imaging Spectroradiometer satellite products. Results reveal that SO2 is more strongly associated with CG lightning activity compared to PM₁₀ concentration. Other surface pollutants like NO_x and O₃ also show strong linear association with CG lightning flashes. Additional investigations have also been performed to extreme lightning events, particularly to a few long-lasting lightning episodes considering the concentrations of NO_x and O₃ found on days with no lightning activity as representative of the background concentration levels of the said two parameters. Results indicate that the NO_x concentration on days with lightning activity is more than 2-fold compared to the non-lightning days while the O₃ concentration is increased by 1.5-fold. Such increase in NO_x and O₃ concentration on days with lightning strongly supports the transport phenomena of NO_x and O₃ from the upper or middle troposphere to the lower troposphere by downdraft of the thunderstorm during its dissipation stage. Overall, studies suggest that enhanced surface pollution in a near-storm environment is strongly related to the increased lightning activity, which in turn increases the surface NO_x level and surface O_3 concentration over the area under study.

Keywords. Atmospheric composition and structure (pollution – urban and regional)

1 Introduction

Lightning plays a crucial role in determining the regional air chemistry. Westcott (1995) first indicated the urban effect on the enhancement of cloud-to-ground (CG) lightning over cities. Since then, considerable research work has been carried out to explore the effect of an urban area on the initiation and enhancements of CG lightning (Lyons et al., 1998; Orville et al., 2001; Soriano and Pablo, 2002; Pinto et al., 2004; Farias et al., 2009; Lal and Pawar, 2011; Pawar et al., 2012a). Several studies have indicated the direct and indirect effect of pollution on our global atmosphere. Li et al. (2011) pointed out the effect of aerosol in cloud formation process and rainfall, whereas Trenberth et al. (2000) indicated how aerosol can effectively modify our weather system. Westcott (1995) suggested the possible influence of high PM₁₀ and SO₂ concentrations on the enhancement of urban CG lightning activity. Orville et al. (2001) reported an association of urban heat island and anthropogenic pollution in the enhancement of the flash density over and downwind of the urban area after conducting a long-term analysis over the city of Houston. Soriano and Pablo (2002) reported a possible contribution of SO₂ to the increased lightning activity but did not find any significant correlation between PM₁₀ concentration and lightning flashes in central Spain. On the other hand, Naccarato et al. (2003) reported a positive correlation between PM₁₀ concentration and CG lightning over southern Brazil. Similar reports are also found in the literature indicating a possible relationship between CG lightning and both PM₁₀ and SO₂ concentration (Kar et al., 2009). Atmospheric pollutants play a crucial role in the various development stages and formation of local weather system. It is not yet well understood how the atmospheric pollutant varies

¹Center for Space and Remote Sensing Research, National Central University, Jhong-Li, Taiwan

^{*}These authors contributed equally to this work.

with different phases of prevailing weather and affects the local weather conditions. It is highly essential to know the interconnectivity between various meteorological parameters and pollutant particles to study of pollution meteorology over a particular region for a specified period of time. In general, the urban heat island circulation and the possible role for air pollution have been indicated as the prime influencing factor behind such enhancement in most of these studies. From earlier studies it has been found that urban effect on local weather is apparently due to a joint action of a thermodynamic effect due to the differential heating of the city surface and an increased aerosol concentration in the local atmosphere caused mainly by human activities (Farias et al., 2009; Lal and Pawar, 2011). However, the physical mechanisms responsible for these effects are not fully understood due to the complex correlations. In recent years, meteorologists have shown much interest in exploring the physical relationship between lightning and pollution over the globe.

The influence of atmospheric pollutants on cloud systems directly affects radiation budget and indirectly affects the cloud dynamics and microphysical properties (Tie et al., 2003; Khain et al., 2005; Wang et al., 2011). Weather and climate modification by atmospheric pollutants has been indicated by Hobbs et al. (1974). Bréon et al. (2002) reported the significant consequence of aerosols on cloud microphysics on a global scale. Positive correlations between CG lightning flash density and rainfall amount have been estimated by Petersen and Rutledge (1998). Change in cloud microphysical processes leads to a change in concentration, phase, and the size of cloud particles, which in turn affects the charge separation processes in thunderclouds. Orville et al. (2001) suggested that the enhanced pollution plays a crucial role in holding back the mean droplet size in the boundary layer; as a result more cloud water would therefore be active in differentiating the electric charge, causing more CG lightning flashes. A recent study conducted by Farias et al. (2014) suggests that urban pollution tends to saturate the intensification of storms and lightning activity at a specific level.

Moreover, the chemical reactions which determine the concentrations of ozone (O₃) and hydroxyl radical (OH) in the troposphere and stratosphere are facilitated by electric discharge-produced nitric oxide (NO) molecule. Seasonal and geographical variation of lightning-produced NO_x has been compared to other NO_x sources like anthropogenic activity, soil emissions and biomass burning by Bond et al. (2002) and indicated that the lightning-produced NO_x remains significant throughout the year. Lightning and aircraft exhaust subscribe to ambient NO levels in the troposphere. Modified tropospheric NO levels then can either enhance or reduce the O₃ levels in accordance with the local conditions and NO_x level ($NO_x = NO + NO_2$). Lightning channels primarily produce NO as a trace gas with less NO2. Once NO is produced in the atmosphere, it is always oxidized and accompanied by NO₂. NO₂ is dissociated into NO and O in the presence of sunlight. Successive chemical reaction and the photolyzation of NO_2 maintain a balance between NO_2 and NO. It has already been established that lightning-produced NO can play a crucial role in the chemistry of the earth's atmosphere, particularly, in adjusting the concentrations of O_3 and OH (Crutzen, 1973; Chameides and Walker, 1973). Noxon (1976) first reported the enhanced concentration of NO in the vicinity of lightning flashes. He also reported an increase of about 2 orders of magnitude in NO_2 concentration above ambient levels and as high as 100 ppbv (parts per billion by volume). Thereafter many reports are found in the literature advocating that lightning can produce significant increases in NO_x concentration in the troposphere and contribute notably to columnar abundance of NO_2 (Choi et al., 2005; Martin et al., 2006).

However, PM₁₀, SO₂, NO_x and O₃ production due to lightning activity over Taiwan has not yet been studied extensively. The present study has been conducted considering the data of surface PM₁₀, SO₂, NO_x and O₃ on lightning flashes during the premonsoon season. The rainfall-lightning relationship is found more consistent during premonsoon season than monsoon season as reported by Lal and Pawar (2009). A higher ratio of premonsoon lightning flash density to rain rate is reported over the Tibet Plateau than that during monsoon season by Qie et al. (2004). Higher mean cell-level flash rate during the premonsoon season than during the monsoon season has been established by Yuan and Qie (2008). Hence, the premonsoon period has been selected for the present study. Main objective of the present study is to investigate the effect of lower tropospheric air pollutants on CG lightning activity along with other meteorological parameters over Taipei. Further investigation has also been carried out to unveil the association of CG lightning flash in surface ozone production over Taipei during premonsoon (March-April) months.

2 Data and methodology

Cloud-to-ground lightning data were collected from Tai Power Company of Taiwan for the period 2005–2010. The lightning location system (LLS) of Tai Power Company was built in 1989. The LLS was upgraded to a total lightning detection system (TLDS) in 2002. The TLDS consists of seven lightning detection sensors (SAFIR 3000), which are located at the top of Ying-Tsu-Ling microwave tower, Wu-Shihpi microwave tower, Ji-Shan microwave tower, Nan-Ke extra voltage substation, Feng-Lin microwave tower, building roof of Ming-Tan power plant, and building roof of Xiao-Liou-Chiou. The location of these seven sensors, distributed throughout Taiwan, is shown in Fig. 1. Very high frequency interferometric technique is the main basis for the localization principles of SAFIR network (Richard, 1985; Richard et al., 1986). The seven lightning detection sensors, forming a lightning detection network, could detect cloud-to-ground lightning discharges, cloud lightning discharges, and breakdown events. The long-range localization of all lightning



Figure 1. Map of Taiwan showing the location of the sensors and the urban area of Taipei County.

discharges (CG and CC lightning flashes) is governed by triangulation performed on GPS time synchronized direction of arrival provided by interferometric sensor of two different detection stations in the SAFIR network. The lightning detection network average efficiency is greater than 90%, and the lightning detection localization accuracy is less than 1 km. However, especially near the edges of the network the assumption of more than 90% uniform flash detection efficiency may not be realistic.

Heights of convective condensation level (CCL) were calculated using radiosonde/rawinsonde data downloaded from the University of Wyoming website. Air pollutants data were collected from a well-organized air-quality-monitoring network operated by Taiwan Environmental Protection Administration. Taiwan's air quality monitoring network measures PM_{10} concentrations by the automatic Wedding β -gauge monitors, which is one of the US EPA-designated equivalent methods (no. EQPM-0391-081). The PM_{10} inlet is a cyclone operated at $18.9 \, \mathrm{L}\,\mathrm{min}^{-1}$. Particles are detected once every hour from its continuous collection on the filter tape, and the daily average is computed for at least 16 effective hours each

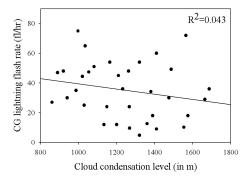


Figure 2. Scatterplot of the CG lightning flash rate and convective condensation level over Taipei during premonsoon season.

day. Seinfeld and Pandis (1998) indicated that water or moisture may be absorbed by the particles collected on the filter tape when the deliquescence point is exceeded, which may lead to an overestimation of PM_{10} concentration measurements. EPA of Taiwan performed a comparison experiment with Wedding β -gauge monitors and the manual samplers because of the frequent abundance of high humidity in the ambient air of Taiwan. Their results reveal that PM_{10} concentrations from automatic Wedding β -gauge monitor are very close to those from the manual samplers (Chang et al., 2001). For the present study PM_{10} , SO_2 , nitrogen oxides (NO_x) , and ozone (O_3) over Taipei are considered. Aerosol optical depth (AOD) data are collected from the Moderate Resolution Imaging Spectroradiometer (Aqua/Terra) satellite products.

3 Results and discussion

Convective condensation level (CCL) is assumed as the cloud base theoretically, and its variation represents various cloud dynamics (Chaudhuri and Middey, 2011; Middey and Chaudhuri, 2013). Figure 2 represents the scatterplot of the CG lightning flash rate and CCL. It shows a negative linear correlation between the plotted parameters. This result is consistent with the physical laws that when the CCL value is low, cloud formation occurs very near to the ground and the prevailing winds helps the surface pollutants to be injected into the cloud very easily. However, from Fig. 2 it is evident that CG lightning flash rate is not influenced by high or low CCL values. On the other hand, high lightning flash rates are found irrespective of CCL heights when the high concentration of surface pollutants is available over the surface. Figure 3 represents the scatterplot of the CG lightning flash rate and SPM (suspended particulate matter). A positive correlation is evident between the plotted parameters. It is seen from Fig. 3 that the values of SPM mainly concentrated around $40-120 \,\mu\mathrm{g}\,\mathrm{m}^{-3}$, but as its value increases, the CG lightning flash count also increases. This observation can be explained from cloud microphysics. The air over Taipei

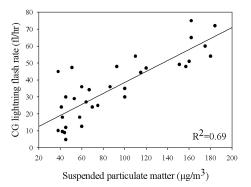


Figure 3. Scatterplot of the lightning flash rate and SPM over Taipei during premonsoon season.

is expected to be more polluted than the maritime air and hence contains a higher cloud condensation nucleus (CCN) concentration. For such differences in boundary layer aerosol concentration, the higher CCN concentrations over Taipei reduce the mean cloud droplet size which in turn decreases the process of coalescence and the droplet collision efficiency (Rogers and Yau, 1989). Therefore, more supercooled water is expected to exist at greater depths in clouds. The abundance of supercooled water may produce large graupel which in turn may enhance the lightning activity over the region concerned (Williams et al., 2005; Liou and Kar, 2010). The mean annual variation of the number of CG lightning flashes during premonsoon months are plotted with annual average particulate matter (PM₁₀), sulfur dioxide (SO₂), nitrogen oxides (NO_x), surface ozone (O₃) concentrations for the whole studied period (2005 to 2010) in Fig. 4a-d. It is worth mentioning in this context that the examination of PM_{2.5} along with PM₁₀ would have been better, since the fine particles are more likely to stabilize clouds. But PM_{2.5} is generally not measured routinely for all the stations under study, and so high concentration episodes occurring between measurements would not be reflected in the average, possibly leading to some exposure misclassification. Therefore we have restricted our analysis to only within PM₁₀. On an average almost similar trends of variations are observed for all the four air pollutant parameters with mean annual CG flash count over the years. The upward increase in the concentration of the atmospheric pollutants, reaching a relative maximum, with the maxima of CG lightning flash is clearly depicted. On and off, a fluctuating trend is also visible particularly in the PM₁₀ concentrations which may be attributed to the effect of precipitation, which occurs usually now and then with atmospheric electrical activity. Such an effect is generally connected with the general precipitation washing effect of the atmosphere.

Figure 5a–d represent the scatterplot of PM_{10} , SO_2 , NO_x and O_3 concentrations versus CG lightning flash count. The result of the scatterplot 5a and b suggests that SO_2 concentrations are better correlated with CG lightning flash count

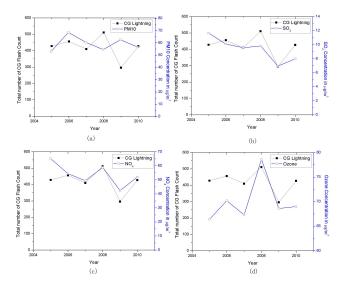


Figure 4. The mean annual variation of the number of CG lightning flashes with annual average particulate matter (PM_{10}), sulfur dioxide (SO_2), nitrogen oxides (NO_x), surface ozone (O_3) concentrations during premonsoon months for the whole studied period (2005 to 2010).

than PM₁₀ concentrations. Since the sulfate particles are generally more active than PM₁₀ in the formation of cloud droplets (Seinfeld, 1975), a comparatively higher contribution from SO₂ concentration is expected with CG lightning flashes. Hence our results corroborate well the results of Westcott (1995), who found that large annual values of SO₂ and PM₁₀ correspond generally to the large values of urban and downwind CG flashes. However, Lal and Pawar (2011) have not found any aerosol effect on lightning activity particularly over coastal cities while they have found the aerosol effect over inland stations. Very recently, Farias et al. (2014) conducted a long-term study of aerosol effect on the weekly cycles in lightning activity over São Paulo and reported that pollution tends to saturate the intensification of storm and lightning activity in a specific level. Figure 5c indicates a positive correlation between CG lightning flash count and NO_x concentration. It is also seen from Fig. 5c that the NO_x concentrations belong to the higher levels (65 μ g m⁻³) during premonsoon lightning activity over Taipei. Such high concentration of NO_x (Zhoua et al., 2005) may not be considered as a predecessor for lightning. Rather, it could be considered as the NO_x production in the lower troposphere as a result of combustion due to lightning flash. Therefore, as the lightning flash count increases, NO_x concentrations also show higher values. From Fig. 5d a positive linear correlation is again found between CG lightning flash count and nearsurface ozone concentration over Taipei. It is worth mentioning in this context that lightning-induced NO_x is not the only source of tropospheric NO_x but other sources like biomass burning, fossil fuel combustion, oxidation of atmospheric ammonia, soil release, and penetration of stratospheric NO_x

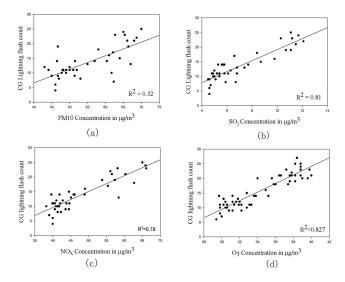


Figure 5. Scatterplot of PM_{10} , SO_2 , NO_x and O_3 concentrations versus CG lightning flash count during premonsoon months for the whole studied period (2005 to 2010).

contribute to the tropospheric NO_x as well. From the present research report it seems that premonsoon CG lightning has a significant contribution in NO_x production over the city of Taipei. It has been reported that during the warm season the southwesterly flow, enriched with abundant moisture, is potentially unstable and brings warm, moist air from the South China Sea to Taiwan (Chen and Chen, 2003). This unstable, warm, moist air from the west coast then moves towards the land and meets the central mountain ridge (CMR), where orographic lifting is expected to take place. Besides orographic lifting, orographic blocking (Li et al., 1997; Yeh and Chen, 2002) and thermally driven circulations during the diurnal heating cycle (Chen and Li, 1995) are also expected to play an important role in lifting up the atmospheric pollutants vertically. The atmospheric pollutants then remain aloft as aerosol to participate in cloud formation. Hence, the cloud microphysics is determined from the concentration and various modes of aerosol.

Figure 6 depicts the scatterplot of CG lightning flash rate with AOD. It is seen from the plot that a positive linear correlation exits between AOD and CG lightning flash rate. Such linear correlation is not unexpected, rather consistent with the physical laws. A high value of lightning flash rate means the amount of charge destroyed per unit area per unit time is very high. In an opaque atmosphere, where aerosol optical depth is higher, such charge destruction process happens faster than usual. Increased aerosol loading in cloud mixed phase region leads to an accelerated charge destruction process which in turn enhances the rate of lightning flash (Yuan et al., 2011). It is assumed that larger amount of particulate matter from the surface is expected to be migrated in clouds with low vertical extent to create excess CG lightning flash count.

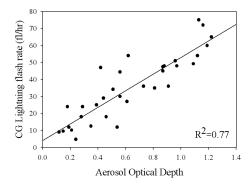


Figure 6. Scatterplot of CG lightning flash count with aerosol optical depth (AOD).

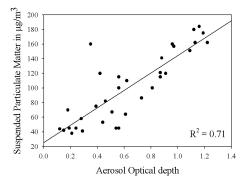


Figure 7. Scatterplot of ground-based SPM measurement and satellite-based AOD measurements.

Figure 7 represents a scatterplot of ground-based SPM measurement and satellite-based AOD measurements. High positive correlation is found between the plotted parameters over Taipei during the premonsoon season. During the premonsoon season, an innumerable amount of particulate matter from the surface goes aloft with thunderstorm updraft and participate in cloud microphysical processes (Rogers and Yau, 1989). This particulate matter is expected to be infused into a thundercloud and serve as CCN. As a result, the cloud opacity is changed and this change in cloud opacity is reflected in the AOD measurements. Anthropogenic particulate impurities in cloud water can affect the charge separation in the urban thundercloud appreciably by increasing the negative lightning activity (Steiger et al., 2002; Steiger and Orville, 2003). Higher concentration level of near-surface pollutant parameters can thus influence the CG lightning activity and flash rate. The higher number of pollutant-induced lightning flashes may then in turn influence greatly the surface NO_x production over Taipei. In later atmospheric stage this nitrogen oxide gas (NO_x) interacts with other trace gases in the atmosphere in the presence of sunlight and produces atmospheric ozone (O_3) . The chemical reaction responsible for the creation of tropospheric ozone can be written as

$$NO_2 + \text{sunlight} \rightarrow NO + O,$$
 (R1)

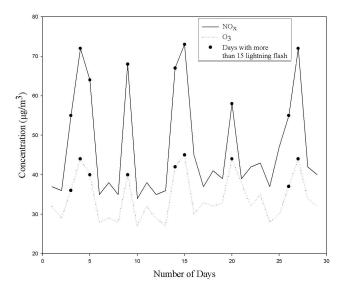


Figure 8. The variation of the daily average concentration of NO_x and O_3 with lightning days (number of lightning flashes > 15) and non-lightning days.

$$O + O_2 \rightarrow O_3. \tag{R2}$$

From the premonsoon CG lightning data we select few days with a considerable number of lightning flashes (>15) along with no lightning activity days. Then, the values are compared to the daily average concentration of NO_x and O₃ concentrations and are plotted in Fig. 8. Figure 8 reveals that the days with lightning flashes (>15) are associated with higher daily average values of NO_x and O₃ compared to the days with no lightning flash. Close examination of the results suggests that the NO_x concentration on days with lightning activity is more than 2-fold compared to the non-lightning days while the O₃ concentration is increased approximately by 1.5-fold. Such increase in NO_x and O₃ concentration on days with lightning strongly supports the transport phenomena of NO_x and O_3 from the upper or middle troposphere to the lower troposphere by downdraft of the thunderstorm during its dissipation stage. It is worth mentioning in this context that only a few studies about lower surface variation of ozone and NO_x associated with thunderstorms are found in the literature (Ott et al., 2010; Minschwaner et al., 2008; Middey and Chaudhuri, 2013). Minschwaner et al. (2008), after conducting a study in central New Mexico, indicated a large enhancement in ozone (50 % above the mean) between about 3 and 10 km altitude within an electrically active storm and inferred that the observed ozone enhancements could have resulted from a number of possible processes, including the advection of polluted air from the urban environments, photochemical production by lightning-generated NO_x from aged thunderstorm outflow, downward mixing of stratospheric air, or local production from within the thunderstorm. Several

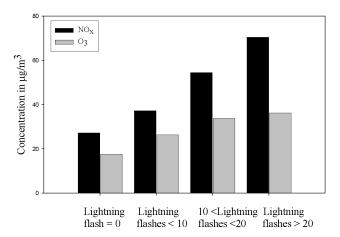


Figure 9. Variation of daily average concentrations of NO_x and O_3 with four categories of lightning days during premonsoon seasons.

studies have shown that the impacts of NO_x on ozone are complex and nonlinear (Lin et al., 1988). Recent observation also indicates that the lightning-induced NO_x production inside a thunderstorm has significant effect on surface ozone concentrations in the tropics (Pawar et al., 2012b).

Six years of CG lightning flash data are grouped into four categories during the premonsoon months. The four categories are days with no lightning flash, days fewer than 10 lightning flashes, days having 10 lightning flash per day to 20 lightning flashes per day, and days with more than 20 lightning flashes per day during premonsoon months of 2005-2010. These four categories of days are then plotted with daily average concentrations of NO_x and O₃ in Fig. 9. The variations of daily average concentrations of NO_x and O₃ with four categorically different days are prominently visible. NO_x and O₃ concentrations both are found to be increased from the days with no lightning flash to the days with lightning flashes more than 20 per day. The daily average values of NO_x and O₃ concentrations are varied from 27.27 to $76.43 \,\mu g \, m^{-3}$ for NO_x and from 17.56 to $36.28 \,\mu g \, m^{-3}$ for O₃.

4 Conclusions

The present research shows the relationship between CG lightning activity and surface pollutants over a tropical station Taipei during premonsoon seasons. Output of this study indicates that surface pollutants have a close association with CG lighting activity. This study also supports that high-frequency weather phenomena like lightning activity are not only influenced by meteorological parameters but also influenced significantly by tropospheric particulate matters. The present study strongly indicates that the SPM concentration at surface level affects the lightning flash rate during premonsoon season over Taipei which strongly corroborate the findings of Steiger et al. (2002) and Steiger and

Orville (2003), who have shown after conducting a long-term study that the CG flash density is affected by the urban area of Houston and southern Louisiana. Higher SPM concentration (> $160 \,\mu g \, m^{-3}$) corresponds to higher lightning flash rate irrespective of the cloud base formation: either it is nearer to the surface or at higher level. Near-surface SPM and PM₁₀ contribute significantly to increase AOD and hence serve as CCN and influence lightning activity. These results strongly support that pollution plays a key role in lightning enhancement (Steiger and Orville, 2003; Naccarato et al., 2003; Soriano and Pablo, 2002; Kar et al., 2009). Additional investigation suggests that the NO_x concentration on days with lightning activity is more than 2-fold compared to the nonlightning days while the O₃ concentration is increased 1-fold during premonsoon lightning activity. Such increase in NO_x and O₃ concentration on days with lightning strongly supports the transport phenomena of NO_x and O₃ from the upper or middle troposphere to the lower troposphere by downdraft of the thunderstorm during its dissipation stage. It is supposed that the increased lightning activity can enhance the NO_x production which in turn can enhance the surface O₃ formation through photochemical reaction in the presence of sunlight as is evident from the present study. A higher concentration level of 65 µg m⁻³ is found for NO_x during premonsoon lightning activity. Surface ozone is also found to be at higher concentration levels during significant lightning activity days, and it follows a similar pattern with total number of lightning flashes over the years. Overall, studies suggest that enhanced surface pollution in a near-storm environment is strongly related to the increased lightning activity, which in turn increases the surface NO_x level and surface O₃ concentration over the area under study.

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