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Cloud-to-ground lightning, meteorological conditions and surface NO_x and O_3 variation in relation to thunderstorm and lightning activities over Hong Kong

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Abstract

Cloud-to-ground (CG) lightning, meteorological conditions and corresponding surface nitrogen oxides (NO_x) and ozone (O_3) variation in relation to thunderstorm and lightning activities over Hong Kong at Kwai Chung (urban), Tung Chung (new town) and Tap Mun (background) during active lightning seasons from 2009 to 2013 were studied ~~.~~ by analyzing ~~R~~respective air quality monitoring station data along with CG lightning ~~data~~ and meteorological data~~ are analyzed~~. We observed NO_x enhancement and significant O_3 decline on lightning days. Influences of land use types, lightning activities and meteorological conditions on surface NO_x and O_3 ~~are~~ were examined. NO_x and O_3 concentration~~ distribution~~s shifted towards higher and lower ~~concentration~~ levels, respectively, for lightning days especially in the dominant wind directions. Principal component analysis/absolute principal component scores (PCA/APCS) method and stepwise multiple linear regression (MLR) analysis were employed to examine the influence of thunderstorm related lightning and meteorological parameters on surface NO_x and O_3. Wind speed was supposed to be the most important meteorological parameter affecting the concentration of NO_x, and lightning activities were observed to make a positive contribution to NO_x. Negative contribution of hot, cloudy and wet weather and positive contribution of wind speed were found to affect the concentration of O_3. Lightning parameters were also found to make a small positive contribution to O_3 concentration at Tap Mun and Tung Chung, but the net effect of lightning activities and corresponding meteorological conditions was the decrease of O_3 on lightning days. Reasonably good agreement between the predicted and observed

NO_x and O_3 values indicates that PCA/APCS-MLR is a valuable method to study the thunderstorm induced NO_x and O_3 ~~changes~~variations.

Keywords

Cloud-to-ground lightning; Nitrogen oxides; Ozone; Meteorological condition; PCA/APCS

1. Introduction

Thunderstorms which occur during the period of April to September over Hong Kong in subtropical South China are characterized by the presence of lightning activities accompanied by heavy rain, especially for those occurred in the hot and humid summer. Lightning is a major natural source of nitrogen oxides ($NO_x = NO + NO_2$) in the atmosphere. *Schumann and Huntrieser* [2007] reviewed the sources of global lightning-induced NO_x and reported a best estimate of the global lightning induced NO_x value of 5 ± 3 $Tg{\cdot}a^{-1}$, accounting for about 10% of the globle NO_x emission. NO_x in the atmosphere affects the abundance of ozone (O_3) which in turn affects the oxidizing capacity of the atmosphere [*Crutzen*, 1970; *Thompson*, 1992]. In addition, corona discharge in the vicinity of a lightning flash can also produce ozone directly [*Bozem et al.*, 2014]. *Mueller and Mallard* [2011] showed that lightning produced NO_x added as much as 25-30 ppbv (or up to about 50%) to surface 8-h average O_3 mixing ratios in southeastern U.S. in 2002 using CMAQ model analysis. *Pawar et al.* [2012] studied the effect of lightning activity on surface NO_x and O_3 over Pune, India in pre-monsoon and monsoon periods. Their result showed that NO_x increased significantly at the dissipation stage of thunderstorms, and increase in NO_x greater than the titration threshold level reduced the surface O_3 concentration.

Chinese studies on chemical effect of lightning have been mainly focused on lightning generated NO_x based on model study [*Du et al.*, 2002; *Zhou and Qie*, 2002; *Zhang et al.*, 2014; *Guo et al.*, 2006]. By in-situ measurement, *Zhou et al.* [2003] observed lightning generated NO_x in northeastern Qinghai-Tibet Plateau using a NO_x analyzer at ground level, and demonstrated that the peak values of the average volume mixing ratios of NO_x in the air were correlated with the lightning flashes during the process of thunderstorm. However, an integrated study on lightning activities, meteorological conditions and corresponding surface NO_x and O_3 variation in relation to thunderstorm has not been studied in China. In this study, NO_x and O_3 data observed at three air quality monitoring stations representing different land use types over Hong Kong are analyzed along with CG lightning data and meteorological data for lightning active seasons (from April to September) in the period of 2009 to 2013 to investigate the change of surface NO_x and O_3 between lightning and non-lightning days due to lightning activities under relevant meteorological conditions. Also, thunderstorm related meteorological and lightning parameters are examined utilizing the principal component analysis/absolute principal component scores (PCA/APCS) method and multiple linear regression (MLR) analysis to explore the influence of thunderstorm related parameters on surface NO_x and O_3 for the three land use types of interest.

2. Climatic and Land Use Characteristics of the Study Locations

Hong Kong is a coastal mega city located on the eastern side of the Pearl River Estuary, South China. The PRD area, in the immediate neighborhood of Hong Kong, exhibits the highest lightning density in China according to long-term observation by satellites [*Ma et al.*, 2005], since this region is located in the subtropical and coastal area with abundant solar heating and moisture sources. In addition, urban effect and local anthropogenic pollution also contribute to the occurrences of lightning in the PRD region [*Wang et al.*, 2011]. The climate of Hong Kong is governed by the Asian monsoon, with prevailing synoptic winds from the north and northeast in winter, east in spring and autumn, and southwest in summer [*Wang et al.*, 2001]. Most thunderstorms and lightning strokes (96.7% CG strokes) occur during the period of April to September in Hong Kong from 2009 to 2013.

Three air quality monitoring stations representing different land use types, including Tap Mun (22.4714 N, 114.3608 E), Tung Chung (22.2886 N, 113.9431 E) and Kwai Chung (22.3569 N, 114.1294 E), were selected for investigation of the effect of lightning activities and relevant meteorological conditions on surface NO_x and O_3 in this study. Tap Mun is a rural station located on Grass Island in northeastern part of Hong Kong. The highest point of Grass Island is 125 m above sea level and the sampling height of Tap Mun station is 11 m above ground and 26 m above Hong Kong Principal Datum (HKPD). Tung Chung station is located within a residential area in a new town and situated on the north-western coast of Lantau Island. Tung Chung is surrounded by mountains on three sides (west, south and east), including Lantau Peak (934 m, second highest peak in Hong Kong) and Sunset Peak (869 m, third highest peak in Hong Kong). The sampling height of Tung Chung station is 27.5 m above ground (34.5 m above HKPD). Kwai Chung is an urban station within a densely populated residential area mixed with some commercial areas, with the sampling height 13 m above ground (19 m above HKPD). Tai Mo Shan (957 m, highest peak in Hong Kong) is about 6 km to the north of Kwai Chung station and Kam Shan (369 m) is to the east of Kwai Chung. The location of the air quality monitoring stations is shown in Figure 1a.

3. Lightning, Air Quality, Meteorological Data and Relevant Information

3.1. Lightning Data

Lightning Data were obtained from the Hong Kong Observatory Lightning Location Information System. Hong Kong Lightning Location Network (HKLLN) comprises six stations, located at Chung Hom Kok (22.2202 N, 114.2035 E), Tsim Bei Tsui (22.4864 N, 114.0117 E) and Sha Tau Kok (22.5460 N, 114.2213 E) in Hong Kong, Sanshui (23.1969 N, 112.8776 E) and Huidong (22.9700 N, 114.7385 E) in Guangdong and Taipa (22.1589 N, 113.5689 E) in Macao. The location of these six stations is shown in Figure 1b. The lightning location processor used in HKLLN is Vaisala LP2000, and the sensors are Vaisala IMPACT ESP, combining Magnetic Direction Finding (MDF) and Time of Arrival (TOA) technology. Each CG lightning discharge given by the HKLLN is equivalent to the return stroke, which is different from the traditional concept of a lightning flash. The accuracy in the determination of the location of CG lightning stroke within the network is 500 m when all stations are operative, and the CG lightning stroke detection efficiency is estimated to be around 90% (http://www.weather.gov.hk/contente.htm). HKLLN can record intra-cloud (IC) lightning strokes as well which also produce NO_x. However, the detection efficiency of IC lightning is estimated to be only 10-50%. Thus IC lightning data are not taken into account in this study because of the limited detection efficiency, although IC lightning generated NO_x may also be important when there are downdrafts from the thunder clouds to surface.

To acquire comparability between each study site, we mainly focus on CG lightning strokes occurred within 10 km of the air quality monitoring station. Similar method was also used in the study by *Sonnadara et al.* [2006] and *Liu et al.* [2014]. Meanwhile, 10 km is also a representative range in the research of NO_x and tropospheric O_3 at local and urban scales [*Seinfeld and Pandis*, 2006]. It is noteworthy that a lightning day (LD) in this paper refers to a day during which at least one CG lightning stroke is detected by the HKLLN within 10 km of the study site, while a non-lightning day (NLD) refers to a day without any CG lightning stroke detected within 10 km.

3.2. Air Quality and Meteorological Data

Air quality data, including daily averaged concentrations of NO_x, O_3 and CO, were downloaded from the website of Hong Kong Environmental Protection Department (HKEPD, http://epic.epd.gov.hk/EPICDI/air/station/). NO_x were measured by chemiluminescence instruments (API 200A); O_3 was monitored with UV photometric analyzers (API 400, API 400A) and CO was measured with non-dispersive infrared absorption with gas filter correlation principle (TECO 48C, API 300). The accuracy of the air quality monitoring network was within the control limit of ±15% for gaseous pollutants and the precision of the network was within ±10%, based on the 95% probability limits.

Daily averaged meteorological data, including wind direction, wind speed, rainfall, temperature, relative humidity and cloud amount, were obtained from the weather stations which are closest to the air quality monitoring stations. Above-mentioned meteorological data were downloaded from the website of Hong Kong Observatory (HKO, http://www.weather.gov.hk/contente.htm). Convective Available Potential Energy (CAPE) value was obtained from the University of Wyoming web server (http://weather.uwyo.edu/upperair/sounding.html, station number: 45004).

4. Results and Discussion

4.1. Lightning Characteristics during Lightning Active Seasons over Three Different Land Use Observation Sites

General lightning information at each study site during lightning active seasons (April to September) from 2009 to 2013 is presented in Table 1. During the study period, there occurred 20090, 25879 and 24571 strokes within 10 km of Tap Mun, Tung Chung and Kwai Chung station, respectively. Total stroke number at rural station Tap Mun was obviously less than that at the other two stations. Studies show that lightning density tends to be higher over urban areas due to urban heat island effect and urban anthropogenic aerosol emissions [*Naccarato et al*., 2003; *Stallins et al*., 2013; *Kar and*

Liou, 2014]. Positive correlation of lightning activity with elevation has also been observed due to the enhancement of convection resulting from orographic uplifting [*Kotroni and Lagouvardos*, 2008]. Thus higher stroke number observed at Tung Chung and Kwai Chung could be a result of the relatively higher urbanization level and the surrounded mountains of these two stations.

Average peak current of the total lightning strokes (refers to the average absolute value of the peak current of both positive and negative CG lightning) was similar at each study site, ranging from 14.0 to 14.5 kA. For negative stokes, average peak current was between -14.6 and -15.9 kA. However, weaker peak current was found for positive strokes, between 5.3 and 7.6 kA, which was also much lower than the result of *Liu et al.* [2014] (21.7 to 28.1 kA) in the PRD region from 2009 to 2011. The percentage of positive lightning ranged from 8.3% to 14.6%, which was slightly higher than the finding of *Liu et al.* [2014] in the PRD region (6.1% to 10.6%). A possible explanation for the unexpected low positive peak current in this study is the inclusion of weak positive strokes. As suggested by *Cummins K.* [1998], positive flashes with peak current lower than 10 kA should be regarded as cloud discharges. However, some other researchers pointed out that "the limit of 10 kA is somewhat arbitrary, and may be related to network configuration and climate" [*Schulz et al.*, 2005]. When the weak positive strokes (< 10 kA) were removed from the dataset, the average peak current for positive strokes was from 15.2 to 20.7 kA as shown in Table S1 in the supplementary material. However, the percentage of positive lightning also sharply decreased to the incredible low range of 1.4% to 2.0%. Thus we did not exclude positive strokes less than 10 kA in the following analyses. *Zhou et al.* [2005] pointed out that higher NO_x values may be associated with higher positive CG flash proportion based on the fact that the average positive peak currents were much higher than the negative ones in their cases. However, it is not the case for us whether we remove the weak positive strokes or not in this study.

4.2. Contrast of Surface NO_x and O_3 between LDs and NLDs

Average concentrations of surface NO_x and O_3 on LDs and NLDs at each study site are presented in Table 2. Table 3 gives the mean value change (MVC) and percentage change (PC) of NO_x and O_3 concentrations on LDs and NLDs at each site. We define MVC and PC by the following two formulas.

$$\text{Mean Value Change (MVC)} = \text{Mean value on LDs} - \text{Mean value on NLDs} \quad (1)$$

$$\text{Percentage Change (PC)} = (\text{MVC} / \text{Mean value on NLDs}) \times 100\% \quad (2)$$

MVC of NO_x ranged from 2.0 $\mu g \cdot m^{-3}$ at Tap Mun to 26.4 $\mu g \cdot m^{-3}$ at Kwai Chung, with PC ranging from 4.1% to 20.5%. Positive values of NO_x MVC indicate the increase of NO_x on LDs. O_3 concentration showed a decreasing trend on LDs at all study sites. O_3 concentration decreased 18.4, 13.1 and 12.3 $\mu g \cdot m^{-3}$ on LDs at Tap Mun, Tung Chung and Kwai Chung respectively, when compared with that on NLDs. O_3 PC varied between -26.5% and -44.4%. Contrast of CO concentration between LDs and NLDs is also shown as a reference of respective anthropogenic emission intensity [*Bharali et al.*, 2015]. As can be seen in Table 2, CO concentration was lower on LDs at Tap Mun and Tung Chung (CO data were not available at Kwai Chung station), suggesting that there was no significant increase in anthropogenic emissions on LDs. In other words, NO_x enhancement and O_3 depletion on LDs over Hong Kong were mainly caused by lightning activities and associated meteorological conditions rather than the change of anthropogenic emissions.

4.3. Major Factors Affecting the Change of NO_x and O_3 between LDs and NLDs

Lightning had been observed to be efficient in producing NO_x both in labs and in direct measurements [*Wang et al.*, 1998; *Rahman et al.*, 2007]. Lightning characteristics can affect surface NO_x concentration. In turn, lightning induced surface NO_x can further affect the concentration of O_3 in the atmosphere through photochemical reactions and NO_x titration [*Pawar et al.*, 2012]. In addition, corona discharge associated with lightning can produce O_3 directly as well [*Bozem et al.*, 2014]. Land use types also have an important influence on the concentration of NO_x and O_3, as urban areas usually have more local anthropogenic emission sources compared to rural areas [*So and Wang*, 2003]. Besides, studies had shown that meteorological conditions, which affect the dispersion, transport and reaction of air pollutants [*Elminir*, 2005; *Dueñas et al.*, 2002], also play an important role in affecting the surface concentrations of NO_x and O_3. Apart from conventional meteorological parameters, CAPE value, as an indicator of atmospheric instability, has an influence on thunderstorm and lightning activities and affects the concentrations of surface pollutants as well. Here, we studied and discussed the influencing factors that possibly affect surface NO_x and O_3 which include land use types, wind direction, lightning stroke frequency (LSF, refers to the number of CG lightning strokes occurring within 10 km of each study site per day), average peak current (APC, refers to the average absolute value of the peak current of both positive and negative CG lightning strokes occurring within 10 km of the study site each day) and other meteorological parameters, including rainfall (RF), temperature (T), relative humidity (RH), wind speed (WS), cloud amount (CA) and CAPE.

4.3.1 NO_x and O_3 Concentrations for Different Land Use Types

As can be seen in Table 2, concentrations of NO_x and O_3 varied from site to site due to the differences in land use types. Kwai Chung station, which is an urban station with strong local emissions, had the highest NO_x concentrations on both LDs and NLDs, followed by new town station Tung Chung. In comparison, the rural station Tap Mun without major sources of anthropogenic emissions had the lowest concentrations of NO_x. The concentrations of O_3 were in reverse order, highest in rural area and lowest in urban area, due to the titration effect which occurred because freshly emitted NO reacted rapidly with O_3 to produce NO_2 and led to lower O_3 levels in urban and suburban areas [*Sillman*, 1999]. *Chan et al.* [1998] and *So and Wang* [2003] also found a similar surface O_3 pattern attributed to titration effect in Hong Kong. From Table 3, we find that NO_x MVC was also highest in urban area Kwai Chung (26.4 $\mu g \cdot m^{-3}$) and lowest in rural area Tap Mun (2.0 $\mu g \cdot m^{-3}$). Differences in the MVC value of NO_x among the three sites may also have an indirect relationship with land use type. As molecules diffuse faster with larger concentration gradient (according to Fick's Law), lower background NO_x concentration in rural area benefits the mixing and dilution of high-concentration lightning produced NO_x, which may result in a relatively less increase of NO_x at Tap

Mun on LDs. For O_3 MVC, it was the highest in rural area Tap Mun (-18.4 μg·m^{-3}) where showed the highest background O_3 concentration as well. O_3 MVC at Tung Chung (-13.1 μg·m^{-3}) was slightly higher than that at Kwai Chung (-12.3 μg·m^{-3}), which may be a comprehensive result of influencing factors including not only land use type but also other meteorological conditions and lightning characteristics as discussed in section 4.3.2 and 4.3.3.

4.3.2. NO_x and O_3 Concentration Distributions ~~for~~ in Different Wind Directions

Daily contributions of NO_x and O_3 from different wind directions are illustrated by the pollution roses in Figure 2. Prevailing wind direction at Tap Mun was southeast (SE) during lightning active seasons. At Tung Chung, surface wind mainly came from southwest (SW), then east (E) and SE. In terms of Kwai Chung, SE and then E wind accounted for the largest proportions. There was no significant difference in the prevailing directions between LDs and NLDs at the three stations according to the results of independent samples T test ($p > 0.05$).

Pollution roses in Figure 2 showed that the NO_x concentration distribution was different on NLDs and LDs for respective wind directions. One significant observation was that NO_x distribution shifted towards higher concentration levels during the LDs for all three stations. For example, Tap Mun exhibited a 29.9% of $NO_x > 15$ μg·m^{-3} for NLDs and a 42.6% for LDs; Tung Chung exhibited a 35.2% of $NO_x > 60$ μg·m^{-3} for NLDs and a 41.3% for LDs and Kwai Chung exhibited a 30.2% of $NO_x > 150$ μg·m^{-3} for NLDs and a 53.6% for LDs. This phenomena was more obvious for the dominant wind directions especially for Kwai Chung, which exhibited a 38.6% of $NO_x > 150$

$\mu g \cdot m^{-3}$ for NLDs when compared with a 64.1% for LDs under the prevailing SE wind direction. The O_3 concentration distribution behaved in the opposite direction and exhibited lower proportions of high O_3 concentration during LDs for the three stations. There was 51.7% of $O_3 > 60$ $\mu g \cdot m^{-3}$ for NLDs and 27.4% for LDs at Tap Mun, 28.8% of $O_3 > 60$ $\mu g \cdot m^{-3}$ for NLDs and 12.2% for LDs at Tung Chung and 37.3% of $O_3 > 30$ $\mu g \cdot m^{-3}$ for NLDs and 12.8% for LDs at Kwai Chung. We will demonstrate later that lightning related meteorological conditions played an active role in the O_3 reduction.

4.3.3. Principal Component Analysis/Absolute Principal Component Scores-Multiple Linear Regression Findings

Thunderstorm related meteorological and lightning parameters including RF, T, RH, WS, CAPE, CA, LSF and APC as well as NO_x and O_3 themselves are ~~influencing~~ factors ~~on~~ affecting surface NO_x and O_3 during lightning active periods, and their effects are examined in this section. To eliminate the problem of multicollinearity among the parameters, PCA/APCS method and stepwise MLR analysis were employed.

Principal component analysis (PCA) was first performed ~~to~~ with the input of the thunderstorm related parameters. The original inter-correlated variables were transformed into an equal number of independent uncorrelated variables (principal components) which are linear combinations of the original variables. After the transformation, a varimax rotation procedure was followed to the principal components. We extracted four factors based on the principle of eigenvalue greater than 1. Table S2 presents the varimax rotated factor loadings together with the variance contribution rates explained by each factor. The higher the loading of a variable, the more that variable contributes to the variation of the particular factor. The factor loadings that we have deemed to be relatively high were marked with boldface type in the table.

Main contributors to each factor are summarized in Table 4 according to the rotated factor loadings. Take Tap Mun as an example. F_1 was loaded heavily on -O_3, T and CAPE, which means F_1 mainly represented the variation of -O_3, T and CAPE. Negative correlation between O_3 and T and CAPE indicates the decrease of O_3 on hot days and the associated increase in CAPE ~~is~~ was the key to the possibility for the growing of convective thunderstorms [*Craven and Brooks*, 2004]. This phenomenon is closely related to the seasonal variation of O_3 concentration in Hong Kong which ~~had~~ was found to be attributed to the seasonal alternation of the prevailing oceanic and continental air masses and climate system [*Chan et al.*, 1998]. The high loading on -O_3, RF, RH and CA in F_2 implie~~s~~d the reduced formation of O_3 through photochemical reactions in cloudy and wet weather. F_3 mainly represented the variation of -NO_x and WS, suggesting the dilution effect of wind on NO_x. F_4 mostly depended on RF, LSF and APC, which indicates that intensive lightning activities were usually associated with heavy rainfall. Main contributors to the factors at Tung Chung and Kwai Chung were similar to those at Tap Mun, as shown in Table 4. The only difference was the interchange of the factors F_1 and F_2 for Tung Chung with respect to the other two sites. The inclusion of APC due to its larger loading to what is called F_2 in the other two sites promoted this factor into F_1, the first principal component for Tung Chung.

The absolute principal component scores (APCS) for each sample were subsequently calculated for the respective factors to obtain predictor variables to be included in the MLR model. Table S3 presents the equations for calculating the APCS. A detailed description of the APCS can be found in *Thurston and Spengler* [1985]. The APCS were then used as independent variables to establish predictive equations for NO_x and O_3 concentrations through stepwise MLR analysis, as follows

$$C_i = (b_0)_i + \sum \mathrm{APCS}_p \times b_{pi},\ p = 1, 2, 3, 4, \quad (3)$$

Where C_i is the concentration of pollutant i, $(b_0)_i$ is the constant term of multiple regression for pollutant i, APCS_p is the absolute principal component score for factor p, and b_{pi} is the coefficient of multiple regression of factor p for pollutant i. Table 5 presents the MLR equations for prediction of NO_x and O_3 using the APCS. The correlation coefficients for the regression analysis were from 0.786 to 0.914. NO_x and O_3 concentrations predicted by the equations were plotted against the observed values and the results are presented in Figure 3. Reasonably good agreement was found between the observed and predicted values.

As can be seen from the MLR equations in Table 5, F_3 made a significant negative contribution to NO_x concentration at all three sites, which indicates the negative contribution of wind speed to NO_x. The absolute value of linear coefficient of APCS_3 was much higher than the coefficients of other factors, indicating the great importance of wind speed on NO_x concentration. Average wind speed was relatively lower on LDs at Tap Mun (8.1 $km \cdot h^{-1}$ on LDs; 9.5 $km \cdot h^{-1}$ on NLDs) and Kwai Chung (8.8 $km \cdot h^{-1}$ on LDs; 9.7 $km \cdot h^{-1}$ on NLDs), which benefitted the increase of NO_x concentration on LDs at these two sites (Tap Mun PC=14.6%, Kwai Chung PC=20.5%). Meanwhile, average wind speed was similar on LDs and NLDs at Tung Chung (12.4 $km \cdot h^{-1}$ on LDs; 12.3

$km \cdot h^{-1}$ on NLDs), and the result was less increase of NO_x concentration on LDs (PC=4.1%). Apart from the influence of wind speed, F_4 made a positive contribution to NO_x concentration at each site, suggesting the production of NO_x by lightning activities. In addition, contributions of F_1 (representing -O_3, T and CAPE) and F_2 (representing -O_3, RF, RH and CA) to NO_x concentration were also positive at Kwai Chung. However, contribution of F_2 (F_2 represented -O_3, RF, RH and CA at Tap Mun; F_2 represented -O_3, T and CAPE at Tung Chung) to NO_x was negative at Tap Mun and Tung Chung, and F_1 (F_1 represented -O_3, T and CAPE at Tap Mun; F_1 represented -O_3, RF, RH, CA and APC at Tung Chung) made no significant contribution to NO_x at these two sites. Different contributions of F_1 and F_2 to NO_x at the three sites may possibly be due to the difference in emissions from anthropogenic sources, topographic and urban effects for the three different land use sites. Average values of relevant meteorological parameters on LDs and NLDs at each site are shown in Table S4.

For O_3 concentration, F_1 and F_2 made a negative contribution at all three sites, which was associated with the low concentration of O_3 in hot and humid summer in Hong Kong and less photochemical formation of O_3 in cloudy and wet weather. Contribution of F_3 to O_3 concentration was positive at each site, and it indicates the positive contribution of wind speed. A rise in wind speed usually implies a rise in the transport of air masses for primary pollutants, but the role played by wind speed in the change of secondary pollutants is much more complex [*Dueñas et al.*, 2002]. Positive contribution of wind speed to O_3 concentration may be due to the negative correlation between NO_x and wind speed, as there was less NO_x titration to reduce O_3 under higher wind speed. Besides, positive contribution of F_4 to O_3 concentration at Tap Mun and Tung Chung implies the production of O_3 related to rainfall and lightning activities. O_3 could be produced both directly by corona discharges [*Bozem et al.*, 2014] and indirectly through photochemical reactions related to lightning induced NO_x. However, the contribution of lightning activities to O_3 concentration was relatively small compared to the other three factors, and the net effect of lightning activities and corresponding meteorological conditions was the decrease of O_3 on LDs.

5. Conclusions

CG lightning, meteorological conditions and surface NO_x and O_3 variation in relation to thunderstorm and lightning activities over Hong Kong at Kwai Chung (urban), Tung Chung (new town) and Tap Mun (background) during April to September from 2009 to 2013 were investigated in this study. NO_x concentration increased on LDs at all three sites (increase of 4.1%-20.5%), with the greatest increase of 20.5% at urban station Kwai Chung; while O_3 concentration decreased significantly on LDs at all three sites (decrease of 26.5%-44.4%). Major influencing factors that can affect surface NO_x and O_3 have been discussed. NO_x concentration distribution was different on NLDs and LDs for respective wind directions, and it shifted towards higher NO_x concentration levels during the LDs especially in the dominant direction for all three stations. The contrary was found for O_3 concentration. The contributions of the thunderstorm related meteorological and lightning parameters to surface NO_x and O_3 were investigated for the three different land use sites based on the results of PCA/APCS method and MLR analysis. Results show that wind speed was the most important influencing factor for NO_x among these parameters, which made a significant negative contribution to the concentration of surface NO_x due to the dilution effect. Lightning activities, as a major natural source of NO_x, was found to make a positive contribution to surface NO_x at all three sites. O_3 concentration was found to be negatively related to T, CAPE, RF, RH and CA, attributed to the low concentration of O_3 in the hot, humid and thunderstorm active summer in Hong Kong and less photochemical formation of O_3 in cloudy and wet weather. Meanwhile, wind speed made a positive contribution to O_3, possibly due to the less NO_x titration to reduce O_3 under higher wind speed. Besides, the factor corresponding to lightning parameters was also found to make a small positive contribution to O_3 at Tap Mun and Tung Chung, which implies the production of O_3 related to lightning activities, but the net effect of lightning activities and corresponding meteorological conditions was the decrease of O_3 on lightning days. Reasonably good agreement was found between the model predicted values and observed values, showing that PCA/APCS-MLR can be applied for the study of thunderstorm induced NO_x and O_3 changes. Findings from this study increase our understanding of atmospheric chemistry in relation to thunderstorm and lightning activities over the subtropical climate region.

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Table 1. Lightning information at each study site during active lightning seasons (April to September) from 2009 to 2013

Station	CG Stroke Number [a]	Positive CG Stroke Number	Negative CG Stroke Number	Positive CG Stroke Proportion	Average Peak Current [b] (kA)	Average Positive Peak Current (kA)	Average Negative Peak Current (kA)
Tap Mun	20090	1662	18428	8.3%	14.0	7.6	-14.6
Tung Chung	25879	3786	22093	14.6%	14.5	6.4	-15.8
Kwai Chung	24571	3557	21014	14.5%	14.4	5.3	-15.9

[a] Here CG stroke number refers to the total number of the CG lightning strokes occurred within 10 km of the study site during the study period.

[b] Here average peak current refers to the average absolute value of the peak current of both positive and negative CG lightning strokes occurred within 10 km of the study site during the study period.

Table 2. Average and extreme concentrations of NO_x, O_3 and CO on LDs and NLDs at each site (unit: $\mu g \cdot m^{-3}$)

Station	Value Category [a]	NO_x LDs	NO_x NLDs	O_3 LDs	O_3 NLDs	CO LDs	CO NLDs
Tap Mun	AVG [b]	15.9	13.9	49.6	67.9	571	596
	SD	8.8	8.0	19.9	32.3	145	171
	MAX	54.3	50.9	117.8	175.3	1090	1120
	MIN	4.9	3.5	15.5	17.1	270	140
	N	190	605	190	605	187	604
Tung Chung	AVG [b]	54.8	52.6	36.4	49.5	500	521
	SD	27.2	30.2	17.6	26.6	170	199
	MAX	130.8	205.5	120.2	147.5	1010	1400
	MIN	8.1	6.8	9.7	6.5	90	80
	N	189	605	189	605	189	605
Kwai Chung	AVG [b]	155.0	128.7	15.4	27.7	——	——
	SD	52.3	46.5	12.8	22.6	——	——
	MAX	337.2	367.9	69.1	126.5	——	——
	MIN	21.7	41.8	1.4	3.3	——	——
	N	250	636	250	636	——	——

[a] AVG stands for average value; SD stands for standard deviation; MAX stands for maximum value; MIN stands for minimum value; N stands for number of days.
[b] Average concentration refers to the average value of daily average concentration of NO_x/O_3/CO on LDs/NLDs during the period of April to September from 2009 to 2013.

Table 3. Mean value changes and percentage changes of NO_x, O_3 and CO on LDs and NLDs at each site

Station	NO_x		O_3		CO	
	MVC [a] ($\mu g \cdot m^{-3}$)	PC [b] (%)	MVC [a] ($\mu g \cdot m^{-3}$)	PC [b] (%)	MVC [a] ($\mu g \cdot m^{-3}$)	PC [b] (%)
Tap Mun	2.0	14.6%	-18.4	-27.0%	-24	-4.1%
Tung Chung	2.1	4.1%	-13.1	-26.5%	-22	-4.2%
Kwai Chung	26.4	20.5%	-12.3	-44.4%	——	——

[a] Mean Value Change (MVC) = Mean value on LDs – Mean value on NLDs
[b] Percentage Change (PC) = (MVC / Mean value on NLDs) × 100%

Table 4. Main contributors to each factor

Station	Factor			
	F_1	F_2	F_3	F_4
Tap Mun	$-O_3$ T CAPE	$-O_3$ RF RH CA	$-NO_x$ WS	RF LSF APC
Tung Chung	$-O_3$ RF RH CA APC	$-O_3$ T CAPE	$-NO_x$ WS	RF LSF APC
Kwai Chung	$-O_3$ T CAPE	$-O_3$ RF RH CA	$-NO_x$ O_3 WS	RF LSF APC

Table 5. Multiple linear regression equations for prediction of NO_x and O_3 using APCS

Station	Regression Equation [a]	Correlation Coefficient (r)
Tap Mun	$NO_x = -1.137 \times APCS_2 - 6.267 \times APCS_3 + 1.163 \times APCS_4 + 30.544$	0.786**
	$O_3 = -18.314 \times APCS_1 - 16.614 \times APCS_2 + 9.326 \times APCS_3 + 1.629 \times APCS_4 + 214.439$	0.860**
Tung Chung	$NO_x = -10.126 \times APCS_2 - 24.979 \times APCS_3 + 1.466 \times APCS_4 + 116.507$	0.914**
	$O_3 = -16.044 \times APCS_1 - 10.359 \times APCS_2 + 9.339 \times APCS_3 + 2.879 \times APCS_4 + 138.002$	0.846**
Kwai Chung	$NO_x = 4.557 \times APCS_1 + 9.686 \times APCS_2 - 42.303 \times APCS_3 + 5.954 \times APCS_4 + 138.766$	0.887**
	$O_3 = -13.560 \times APCS_1 - 9.659 \times APCS_2 + 9.600 \times APCS_3 + 113.193$	0.912**

[a] Equations for calculating the APCS for each factor are presented in Table S3.

** Correlation is significant at the 0.01 level (bilateral).

Commented [h1]: I am not sure how you got the equations for calculating the APCS for each factor in Table S3. The values can actually be obtained directly from the SPSS running by choosing “score” function.