



# Assessment of gaseous criteria pollutants in the Bangkok Metropolitan Region, Thailand

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**Abstract.** The analysis of gaseous criteria pollutants in the Bangkok Metropolitan Region (BMR), Thailand, from 2010 to 2014 reveals that while the hourly concentrations of CO, SO<sub>2</sub> and NO<sub>2</sub> were mostly within the National Ambient Air Quality Standards (NAAQs) of Thailand, the hourly concentrations of O<sub>3</sub> frequently exceeded the standard. The results reveal that the problem of high O<sub>3</sub> concentration continuously persisted in this area. The O<sub>3</sub> photolytic rate constant ( $j_1$ ) for BMR calculated based on assuming a photo-stationary state ranged from 0.008 to 0.013 s<sup>-1</sup>, which is similar to the calculated  $j_1$  using the NCAR TUV model (0.021 ± 0.0024 s<sup>-1</sup>). Interconversion between O<sub>3</sub>, NO and NO<sub>2</sub> indicates that crossover points between the species occur when the concentration of NO<sub>x</sub> (= NO + NO<sub>2</sub>) is ~ 60 ppb. Under a low-NO<sub>x</sub> regime ([NO<sub>x</sub>] < 60 ppb), O<sub>3</sub> is the dominant species, while, under a high-NO<sub>x</sub> regime ([NO<sub>x</sub>] > 60 ppb), NO dominates. Linear regression analysis between the concentrations of O<sub>x</sub> (= O<sub>3</sub> + NO<sub>2</sub>) and NO<sub>x</sub> provides the role of local and regional contributions to O<sub>x</sub>. During O<sub>3</sub> episodes ([O<sub>3</sub>]<sub>hourly</sub> > 100 ppb), the values of the local and regional contributions were nearly double of those during non-episodes. Ratio analysis suggests that the major contributors of primary pollutants over BMR are mobile sources. The air quality index (AQI) for BMR was predominantly good to moderate; however, unhealthy O<sub>3</sub> categories were observed during episode conditions in the region.

## 1 Introduction

Over the last 3 decades, Thailand has experienced rapid industrialization, urbanization and economic growth (World Bank, 2018a). A majority of the country's development has occurred within and around Bangkok (BKK) (13.7° N, 100.5° E), the capital city of Thailand, and in the Bangkok Metropolitan Region (BMR). BMR is comprised of BKK and the five adjacent provinces of BKK (World Bank, 2018a, b). The increase in emissions is due to accelerated growth in automotive and industrial activities. As a major metropolitan area, BMR is dominated by mobile emissions sources, which contributes to the emissions of CO and NO<sub>x</sub>, precursors of ozone (O<sub>3</sub>) formation. The emissions from industrial activities also contribute to those emissions and to the emissions of sulfur dioxide (SO<sub>2</sub>) and the formation of particulate matter. Since 1995, BMR has begun to experience air quality degradation and experienced exceedances in Thailand National Ambient Air Quality Standards (NAAQs) for particulate matter (PM) and ozone (O<sub>3</sub>) (PCD, 2015) owing to strong solar radiation (peak density of direct radiation ~ 1350 kWh m<sup>-2</sup> yr<sup>-1</sup>), high temperature (yearly average ~ 29 °C) and high humidity (yearly average ~ 64 %) (Kumar et al., 2012).

The relationship between air pollution and public health in BMR has been examined in several published studies. Ruchirawat et al. (2007) reported that children who lived in BKK were exposed to high levels of carcinogenic air pollutants which might cause an elevated cancer risk. Buadong et al. (2009) reported that the exposure to elevated PM and O<sub>3</sub> in elderly patients (≥ 65 years) was associated with an increasing in the number of hospital visits for cardiovascular diseases on the following day. Jinsart et al. (2002, 2012)

reported the police personnel and drivers in BKK tended to be exposed to higher levels of PM concentrations compared with the general environment.

Several studies have demonstrated the role of atmospheric processes in elevating Thailand's O<sub>3</sub>. Long-range transport from the Asian continent has enhanced O<sub>3</sub> concentrations in Thailand compared to the lesser O<sub>3</sub> concentrations disbursed via long-range transports from the Indian Ocean (Pochanart et al., 2001). This regional transport, moreover, played an important role in seasonal fluctuations of O<sub>3</sub> in this area (Zhang and Oanh, 2002). Another factor that enhanced O<sub>3</sub> concentrations was the atmospheric chemistry of volatile organic compounds (VOCs). However, this process tended to be more important in enhancing O<sub>3</sub> concentrations in suburban areas than in urban areas (Suthawaree et al., 2012).

Therefore, the availability and analysis of multiyear measurements of such gaseous criteria pollutants in the BMR will improve our understanding of how they contribute to the air quality of this area. In this study, we analyzed diurnal variations, seasonal variations and interannual trends of gaseous pollutants including carbon monoxide (CO), nitric oxide (NO), nitrogen dioxide (NO<sub>2</sub>), SO<sub>2</sub> and O<sub>3</sub> in BMR from 2010 to 2014. Chemical and physical processes associated with high O<sub>3</sub> concentrations have been investigated. Since the monitoring station mostly measured concentrations of nitrogen oxide (NO<sub>x</sub>), O<sub>3</sub> precursors in this study are referred to as NO<sub>x</sub>. The photochemical reaction for O<sub>3</sub> was investigated during the photostationary state. The effects of local emission and regional contributions of O<sub>x</sub> are presented. The severity of air pollution concentrations in BMR in relation to human health is assessed by using the air quality index (AQI).

## 2 Methodology

### 2.1 Study area

Figure 1 shows a map of BMR, the location of monitoring stations in this study and major monsoon winds over this region. BMR refers to BKK and the five adjacent provinces, i.e., Nakhon Pathom, Pathum Thani, Nonthaburi, Samut Prakan and Samut Sakhon. These provinces are linked to BKK in terms of traffic and industrial development (Zhang and Oanh, 2002). Thailand has three official seasons – local summer (February to May), rainy seasons (May to October) and local winter (October to February) as per the Thai Meteorological Department (TMD) (TMD, 2015). During the rainy season, this region's weather is influenced by southwest monsoon wind that travels from the Indian Ocean to Thailand. This marine air mass contains a large amount of moisture, resulting in the wet season in Thailand. During this season, Thailand is characterized by cloudy weather with high precipitation and high humidity. From October to April, this region is influenced by northeast monsoon wind that travels

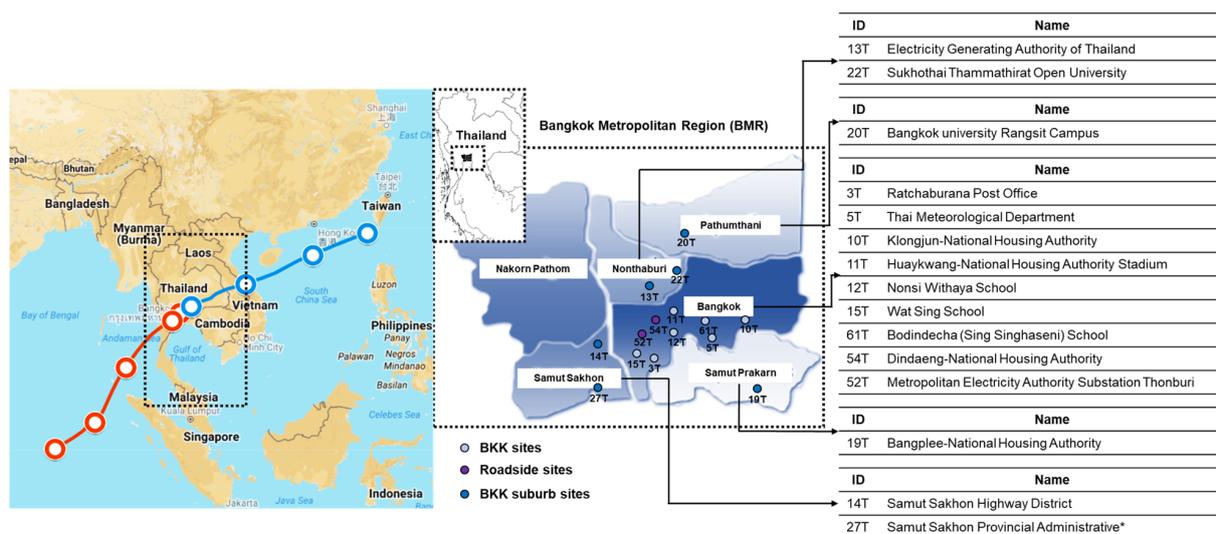
from the northeastern and the northern parts of Asia (China and Mongolia). This monsoon wind brings a cold and dry air mass, which leads to the dry season (local summer and local winter) in Thailand. The local winter in Thailand is characterized by cool and dry weather, while the local summer is characterized by hot (35 to 40°) to extremely hot weather (>40°) due to strong solar radiation. During the dry season, storms may occur during the seasonal transition (TMD, 2015).

Transportation and industrial sectors are considered to be the major sources of air pollutants in the study area (Watcharaviton et al., 2013). In 2014, ~ 36 million new vehicles were registered in Thailand, and 29 % of these cars were registered in BKK (DLT, 2015). About 56 % and 28 % of the registered vehicles in BKK were gasoline and diesel engines, respectively. The remaining 16 % were run on compressed natural gas (CNG) (DLT, 2017). There are a variety of metal, auto parts, paper, plastic, food and chemical manufacturing facilities and power plants in the outskirts of BKK (DIW, 2016a, b, c, d, e).

### 2.2 Data collection and data analysis

Over the 5-year period (1 January 2010 to 31 December 2014), hourly observations from 15 Pollution Control Department (PCD) monitoring stations were analyzed. The monitoring stations are assigned to three categories: BKK sites, roadside sites and BKK suburb sites. BKK sites refer to the monitoring stations that are located within BKK's residential, commercial, industrial and mixed areas. They are within ~ 50 to 100 m of the road. Roadside sites refer to the monitoring stations that are located in BKK within 2 to 5 m of the road (Zhang and Oanh, 2002). BKK suburb sites refer to the monitoring stations that are located in the provinces adjacent to BKK (Fig. 1). Quality assurance and quality control on the data set were performed by PCD prior to receiving the data. Hourly observations of the gaseous pollutants and meteorological parameters were automatically collected with autocalibration at the monitoring stations. Manual quality control was performed when unusual observations were found. An external audit of the equipment and monitoring stations was done every year. The data availability and details of equipment calibrations are provided in Fig. S1, Sect. S1, Supplement.

Gaseous species were measured at 3 m above ground level (a.g.l.). CO was measured using nondispersive infrared detection (Thermo Scientific 48i). NO and NO<sub>2</sub> were measured using chemiluminescence detection (Thermo Scientific 42i). SO<sub>2</sub> was measured using ultraviolet (UV) fluorescence detection (Thermo Scientific 43i), and O<sub>3</sub> is measured by using UV absorption photometry detection (Thermo Scientific 49i). The meteorological parameters including wind speed (WS) and wind direction (WD) were measured at 10 m a.g.l. by a cup anemometer and potentiometer wind vanes. Temperature (*T*) and relative humidity (RH) were measured at



**Figure 1.** Map of BMR, the location of monitoring stations and two major monsoons winds (from NOAA HYSPLIT back trajectory model). Three monitoring station types (BKK sites, roadside sites and BKK suburb sites) are shown as light blue dots, purple dots and blue dots, respectively. (Note: \* the station has been closed since 1 October 2013.)

2 m a.g.l. by a thermistor and thin film capacitor, respectively (Watchravitoon et al., 2013). All the meteorological measurements were made by Met One or an equivalent method.

Data analysis, statistical analysis and plots were developed using Excel 2016. Predominant wind directions related to  $O_3$  concentrations are obtained using the Openair package (tool for the analysis of air pollution data) on the RStudio program (<https://www.rstudio.com/>, last access: 6 February 2018).

### 3 Result and discussion

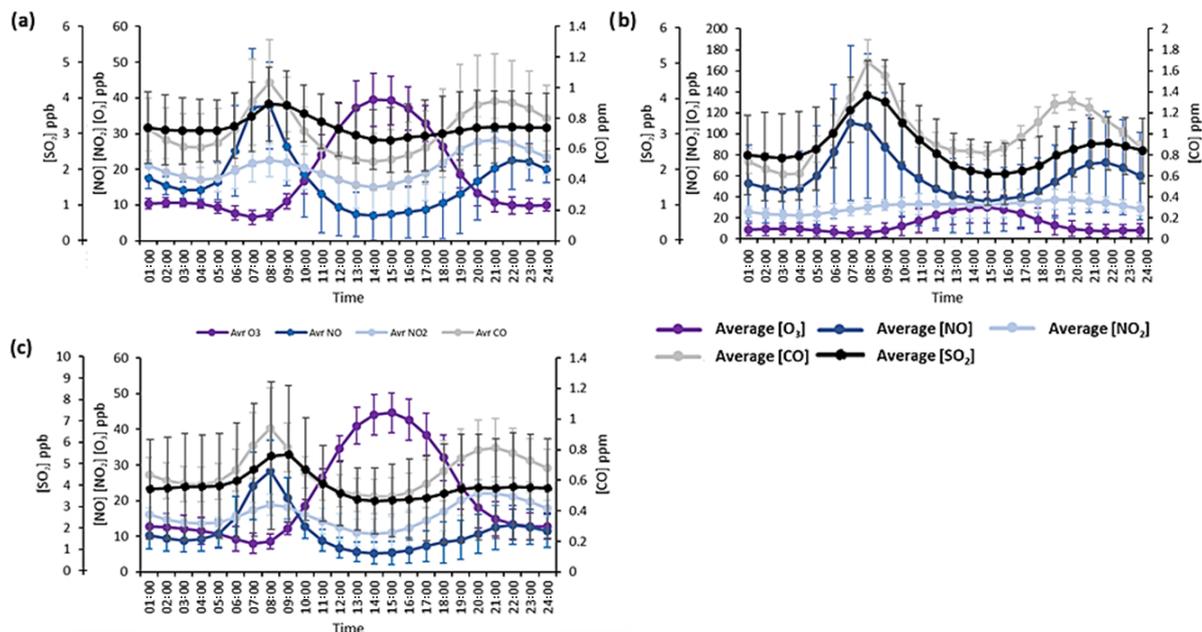
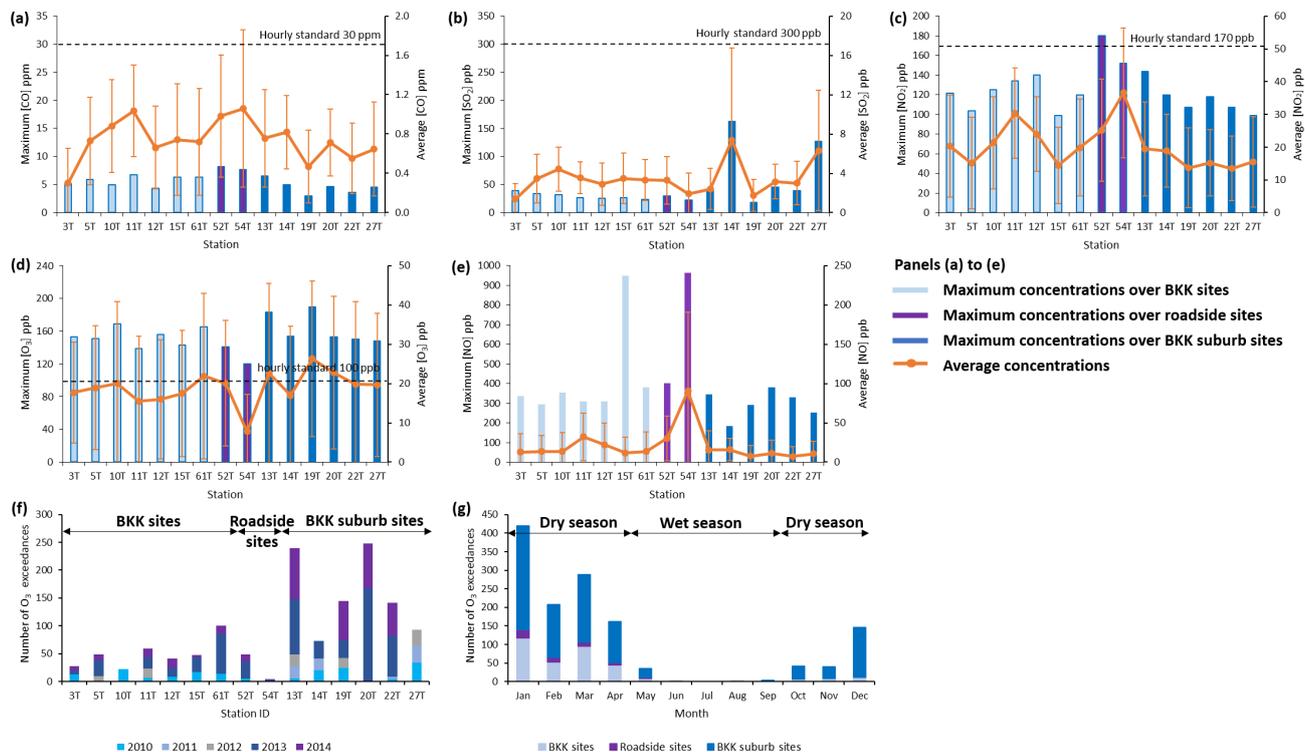
#### 3.1 Status of pollution in BMR from 2010 to 2014

Figure 2a to e show the maximum and average concentrations of gaseous pollutants, from 2010 to 2014 from the 15 monitoring stations. These concentrations are compared with the hourly NAAQs of Thailand (NAAQs of Thailand for hourly  $CO$ ,  $NO_2$ ,  $SO_2$  and  $O_3$  are 30 ppm, 170 ppb, 300 ppb and 100 ppb, respectively (PCD, 2018)). Since  $NO$  is not a criteria pollutant, only the maximum and average concentrations are presented. During the study period, the maximum concentrations of  $CO$ ,  $NO_2$  and  $SO_2$  were mostly at their hourly standards (an exceedance of  $NO_2$  was found at monitoring station 52T during 2013). However, the maximum concentrations of  $O_3$  exceeded its standard. Elevated  $CO$ ,  $NO$  and  $NO_2$  concentrations were observed more frequently at roadside sites than other sites. The average concentrations of  $CO$ ,  $NO$  and  $NO_2$  at roadside sites were  $\sim 1.0 \pm 0.1$  ppm,  $\sim 60.5 \pm 42.7$  ppb, and  $\sim 30.9 \pm 8.1$  ppb, respectively. Elevated  $SO_2$  was more commonly observed at BKK suburb sites than other sites. The average concentrations of  $SO_2$  at BKK suburb sites were  $\sim 4.0 \pm 2.3$  ppb. The average con-

centrations of  $O_3$  during the daytime (06:00 to 18:00 LT) over BKK sites, roadside sites and BKK suburb sites were  $\sim 24.4 \pm 13.5$ ,  $\sim 18.2 \pm 12.3$  and  $\sim 27.7 \pm 14.7$  ppb, and their values during the nighttime (18:00 to 6:00 LT) were  $\sim 11.3 \pm 3.3$ ,  $\sim 9.1 \pm 4.9$  and  $\sim 14.2 \pm 5.4$  ppb, respectively. The 24 h average  $O_3$  concentrations were highest at BKK suburb sites ( $\sim 21.4 \pm 3.3$  ppb), followed by BKK sites ( $18.6 \pm 2.3$  ppb) and roadside sites ( $13.9 \pm 8.6$  ppb). Statistical analyses of the concentrations of gaseous pollutants from the three monitoring station types are provided in Table S1, Sect. B, Supplement.

The seasonal variations in the gaseous pollutants reveal that, in general, elevated concentrations were observed during dry seasons and they decreased during wet seasons (Fig. S2, Sect. S3). Interannual variations in the gaseous pollutants reveal that while the concentrations of  $CO$ ,  $NO_2$  and  $SO_2$  decreased or remained constant, the concentration of  $O_3$  tended to increase during the study period (Fig. S3, Sect. S4).

An  $O_3$  exceedance was recorded when an hourly concentration of  $O_3$  was greater than 100 ppb (hourly  $O_3$  standard). Figure 2f and g illustrate the number of hourly  $O_3$  exceedances, which are shown by location and by seasons. The hourly  $O_3$  exceedances at BKK suburb sites were more frequently observed than at the other sites. The average number of hourly  $O_3$  exceedances was  $\sim 16 \text{ h yr}^{-1}$  at BKK sites,  $\sim 9 \text{ h yr}^{-1}$  at roadside sites and  $\sim 43 \text{ h yr}^{-1}$  at BKK suburb sites. The hourly  $O_3$  exceedances were commonly observed during the dry season, less so during the transitional period between the seasons (May) and rarely during the wet season.



**Figure 3.** Diurnal variations in gaseous species. The plots provide the average concentrations of O<sub>3</sub>, NO and NO<sub>2</sub> in ppb, the average concentrations of CO in ppm, and the average concentrations of SO<sub>2</sub> in ppb at (a) BKK sites, (b) roadside sites and (c) BKK suburb sites. Vertical bars provide  $\pm 1$  standard deviation of the species concentrations.

### 3.2 Diurnal variation in the gaseous species

Diurnal variations in gaseous pollutant are shown in Fig. 3a to c. The diurnal variations in O<sub>3</sub> show a single-peak pattern (Aneja et al., 2001) with the concentrations increasing after sunrise and reaching the peak ~ 15:00 local time (LT). The concentrations begin to decline in the evening and reach the minimum concentrations ~ 07:00 LT the next morning. The concentrations of O<sub>3</sub> at the peaks were ~ 40 ppb at BKK sites, ~ 30 ppb at roadside sites and ~ 45 ppb at BKK suburb sites. The diurnal variations in NO show a bimodal pattern with the concentrations reaching the first and the second peak ~ 07:00 to 09:00 and ~ 21:00 to 22:00 LT, respectively. The concentrations of NO at the first and the second peak were ~ 40 and ~ 23 ppb at BKK sites, ~ 110 and ~ 73 ppb at roadside sites, and ~ 30 and ~ 13 ppb at BKK suburb sites. The concentrations of NO<sub>2</sub> at the first and the second peak were ~ 23 and ~ 28 ppb at BKK sites, ~ 33 and ~ 37 ppb at roadside sites, and ~ 20 and ~ 22 ppb at BKK suburb sites. Even the diurnal variations in NO<sub>x</sub> show a bimodal pattern; at roadside sites the pattern was flatter than at other sites. The flatter pattern of NO<sub>x</sub> at roadside sites reveals that this monitoring station type was affected by a high concentration of NO<sub>x</sub> all day. The diurnal variations in CO show a bimodal pattern with the first and the second peak occurring ~ 08:00 and 21:00 LT, respectively. The concentrations of CO at the first and the second peak were ~ 1 ppm (both peaks) at BKK sites, ~ 2 and ~ 1.5 ppm at roadside sites, and ~ 1 ppm (both peaks) at BKK suburb sites. The first peak of the diurnal variations in NO, NO<sub>2</sub> and CO corresponds with the morning rush hour in BKK (07:00 to 09:00 LT). The second peak occurred ~ 3 to 5 h after the evening traffic rush hour (16:00 to 18:00 LT) (Leong et al., 2002), due to a combination of pollutant emissions and the collapse of the planetary boundary layer (weak turbulence and diffusion) during this time. The diurnal variations in SO<sub>2</sub> show a bimodal pattern with the first and the second peak of SO<sub>2</sub> occurring ~ 08:00 and 21:00 LT, respectively. The concentrations of SO<sub>2</sub> at the first and the second peak were ~ 4 and ~ 3 ppb at BKK sites and roadside sites and ~ 6 and ~ 3 ppb at BKK suburb sites. At the roadside sites, the peaks are more obvious than at the other sites. The result indicates that at this monitoring station type, SO<sub>2</sub> is primarily influenced by emissions from vehicle exhaust using a high sulfur content fuel (Henschel et al., 2013). It is noteworthy that BKK has a large diesel engine fleet (an estimated 25 % of registered vehicles) (DLT, 2015). Diesel fuel contains ~ 0.035 % wt sulfur (DOEB, 2017). The season-wise distributions of the diurnal variations are provided in Fig. S4, Sect. S5.

Figure 4a to c shows diurnal variations in the rate of change in O<sub>3</sub> concentration ( $\Delta[\text{O}_3]/dt$ ) during dry seasons (local summer and local winter) and wet seasons at the three monitoring station types (the data have been averaged for each monitoring station type to capture the rate of change in O<sub>3</sub> concentration characteristics). The diurnal variations in

$\Delta[\text{O}_3]/dt$  are a combination of O<sub>3</sub> chemistry and meteorology. In general,  $\Delta[\text{O}_3]/dt$  during the wet season were lower than those during dry season. However, during local winter, the rates of change in O<sub>3</sub> concentration were the highest. The  $\Delta[\text{O}_3]/dt$  at the three monitoring station types, from 10:00 to 11:00 LT, were 4.5 to 7.0 ppb h<sup>-1</sup> during wet seasons, 6.7 to 7.5 ppb h<sup>-1</sup> during local summers and 5.7 to 9.2 ppb h<sup>-1</sup> during local winters. The  $\Delta[\text{O}_3]/dt$  became negative from 14:00 to 15:00 LT. As expected, the rate of change in O<sub>3</sub> concentration was nearly constant during the nighttime. Rapid changes in the mixing height and solar insolation during the morning increases  $\Delta[\text{O}_3]/dt$ . After sunset, the formation of O<sub>3</sub> is inhibited and the planetary boundary layer becomes more stable resulting in O<sub>3</sub> reduction through chemical reactions (for example, the oxidation of O<sub>3</sub> by NO<sub>x</sub>) and physical processes (for example, dry deposition to the earth surface) (Naja and Lal, 2002).

### 3.3 Photochemical reaction and interconversion between O<sub>3</sub>, NO and NO<sub>2</sub>

The primary precursors for tropospheric O<sub>3</sub>, in the urban environment, are NO<sub>x</sub> and non-methane volatile organic compounds (VOCs), methane or CO (The Royal Society, 2008; Monks et al., 2009; Cooper et al., 2014). While NO<sub>x</sub> was measured continuously at all the monitoring sites, VOCs were measured periodically only at one monitoring station, limiting its usefulness as part of this study. In this study, the photostationary state (PSS) is applied through the chemical reactions of O<sub>3</sub> formation from 10:00 to 16:00 LT. This time window is chosen due to the fully developed planetary boundary layer with well-mixed condition (Pochanart et al., 2001) to avoid the accumulation of air pollutants by surface inversion. Analysis and calculation are performed only during the dry season to eliminate the effects of the removal process by wet deposition.

The relationship among NO, NO<sub>2</sub> and O<sub>3</sub> under PSS is presented by Eq. (1) (Seinfeld and Pandis, 1998).

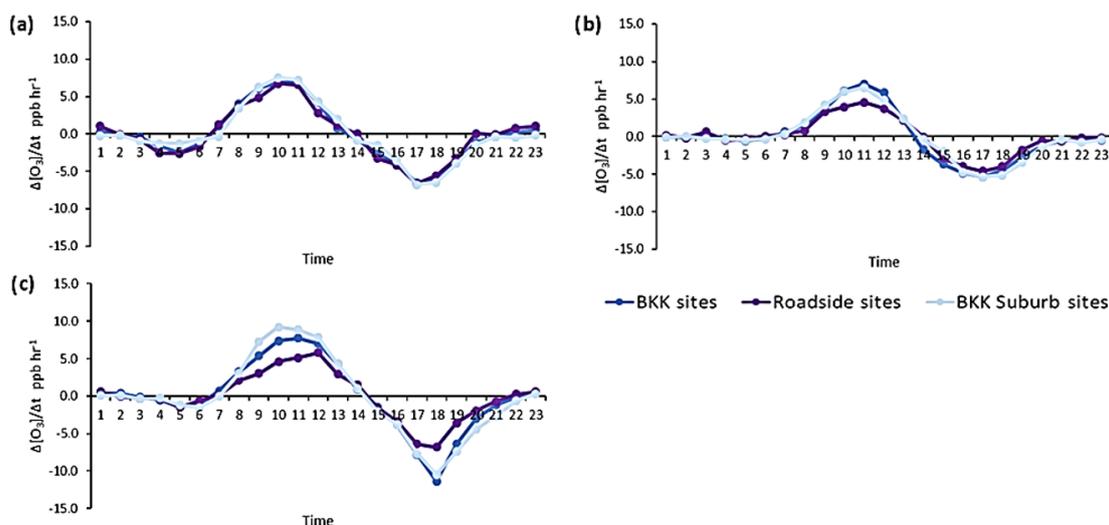
$$[\text{O}_3]_{\text{PSS}} = \frac{j_1[\text{NO}_2]}{k_3[\text{NO}]}, \quad (1)$$

where  $[\text{O}_3]_{\text{PSS}}$  is the concentration of O<sub>3</sub> at PSS and  $j_1$  and  $k_3$  are the reaction rate coefficient of the photochemical reaction of NO<sub>2</sub> and the reaction rate coefficient of the chemical reaction between NO and O<sub>3</sub>, respectively.

The values for  $k_3$  (ppm<sup>-1</sup> min<sup>-1</sup>) are calculated by Eq. (2) (Seinfeld and Pandis, 1998; Tiwari et al., 2015).

$$k_3 = 3.23 \times 10^3 \exp[-1430/T] \quad (2)$$

During dry seasons, the values of  $j_1$  ranged from 0.12 to 1.22 min<sup>-1</sup>, and the average of those at BKK sites, roadside sites and BKK suburb sites were  $0.74 \pm 0.2$ ,  $0.64 \pm 0.3$  and  $0.55 \pm 0.3$  min<sup>-1</sup>, respectively. The rate coefficients are calculated using the NCAR TUV model for 10:00 to 16:00 LT



**Figure 4.** Diurnal variations in the rate of change in  $O_3$  concentration ( $\Delta[O_3]/dt$ ) during (a) local summers, (b) wet seasons and (c) local winters.

during the 2010 dry season at the latitude and longitude of  $13.76^\circ N$  and  $100.50^\circ E$ . The average  $j_1$  value calculated from the NCAR TUV model is  $0.021 \pm 0.0024 s^{-1}$ , which is similar to the calculated  $j_1$  values from Eq. (1) ( $j_1$  ranges from  $0.008$  to  $0.013 s^{-1}$ ). The values of  $j_1$  from this study are similar to those values at an urban background site in Delhi, India (values of  $j_1$  ranged from  $0.4$  to  $1.8 min^{-1}$  and the average was  $0.8 min^{-1}$ ) (Tiwari et al., 2015) and those values collected during daytime in November in the UK (value of  $j_1$  was  $\sim 0.14 min^{-1}$ ) (Clapp and Jenkin, 2001).

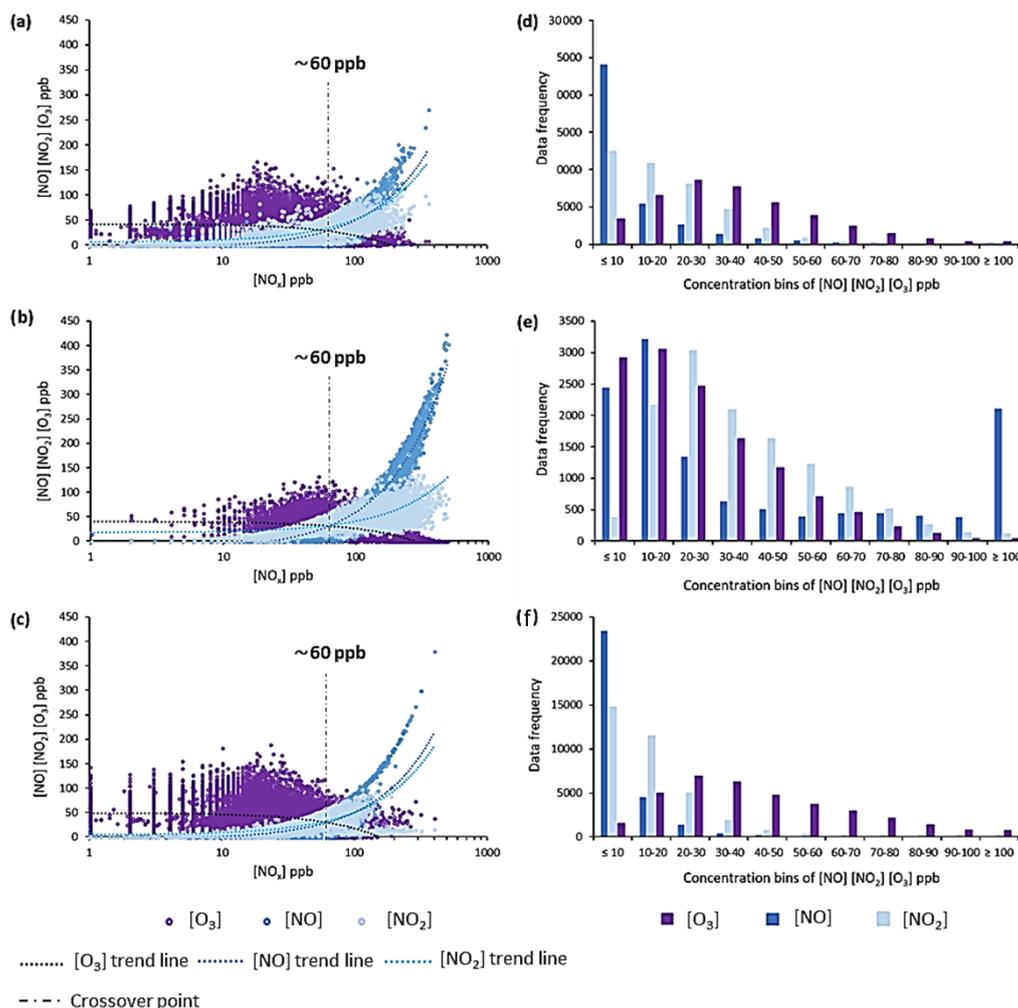
The values of  $k_3$ , during dry seasons, ranged from  $28.3$  to  $30.9 ppm^{-1} min^{-1}$ , and the average of those at BKK sites, roadside sites and BKK suburb sites were  $29.8 \pm 0.7$ ,  $29.7$  and  $29.8 \pm 0.7 ppm^{-1} min^{-1}$ , respectively. The ratio of  $[NO_2]$  and  $[NO]$  was  $\sim 1.9$ . The statistical analysis of  $j_1$  ( $min^{-1}$  and  $s^{-1}$ ) and  $k_3$  ( $ppm^{-1} min^{-1}$  and  $cm^3 molecule^{-1} s^{-1}$ ) at the three monitoring station types using Eq. (1) and the average  $j_1$  calculated from the NCAR TUV model are provided in Table S2, Sect. F.

Figure 5a to c shows the relationships between  $NO$ ,  $NO_2$  and  $O_3$ , their crossover points, and concentration distributions. The crossover point among species occurs when the concentration of  $NO_x$  is  $\sim 60$  ppb. At this point, two regimes are identified, including a low- $NO_x$  regime and a high- $NO_x$  regime. Under the low- $NO_x$  regime ( $[NO_x] < 60$  ppb),  $O_3$  is the dominant species and  $NO_2$  concentrations are higher than  $NO$  for  $NO_x$  species. Conversely, under the high- $NO_x$  regime ( $[NO_x] > 60$  ppb),  $NO$  and  $NO_2$  increase and the concentrations of  $O_3$  rapidly decrease. Under the high- $NO_x$  regime, the decline in  $O_3$  trend lines may describe the  $O_3$  removal process through the titration of  $O_3$  by  $NO$ .

### 3.4 Local and regional contribution to $O_x$

The  $O_x$  concentration is the summation of  $O_3$  and  $NO_2$  concentration. Under the PSS condition, the concentration of  $NO$ ,  $NO_2$  and  $O_3$  approaches an equilibrium and the concentration of  $O_x$  may be considered constant (Keuken et al., 2009). Since the conversion between  $O_3$  and  $NO_2$  in the urban and suburban atmosphere is rapid, the use of  $O_x$  to represent the production of oxidants is more appropriate than only using  $O_3$  (Lu et al., 2010). The local or  $NO_x$ -dependent contribution refers to  $O_x$  concentration that are influenced by a concentration of the local pollutants. The regional or  $NO_x$ -independent contribution refers to the background concentration of  $O_x$  that is not influenced by changes in the local pollutants (Clapp and Jenkin, 2001; Tiwari et al., 2015).

Figure 6a to c show the local and regional contributions of  $O_x$  at the three monitoring station types. The effects of the local and regional contributions to  $O_x$  concentration are analyzed by plotting  $O_x$  concentrations against  $NO_x$  concentrations and fitting the plot with a linear regression ( $y = mx + c$ ). The concentrations of  $NO_x$  and  $O_x$  are referred to as  $x$  and  $y$ , respectively. The slope of the linear regression ( $m$ ) implies the local contribution, and the intercept with the  $y$  axis ( $c$ ) implies the regional (background) contribution (Aneja et al., 2000; Clapp and Jerkin, 2001; Notario et al., 2012). Table 1 shows the comparison between fitted linear regressions from this study and fitted linear regression lines from other studies. The average background  $O_x$  concentrations over BMR during non-episodes ( $[O_3]_{hourly} < 100$  ppb) and episodes ( $[O_3]_{hourly} > 100$  ppb) were  $\sim 48$  and  $\sim 95$  ppb, respectively. The local and regional contributions during the episode days, in general, were about double of those during the non-episode days. The results reveal that elevated  $O_3$  concentrations during the episode days are influenced by



**Figure 5.** Relationships and crossover points of NO, NO<sub>2</sub> and O<sub>3</sub> at (a) BKK sites, (b) roadside sites and (c) BKK suburb sites and concentration distributions of those species at (d) BKK sites, (e) roadside sites and (f) BKK suburb sites.

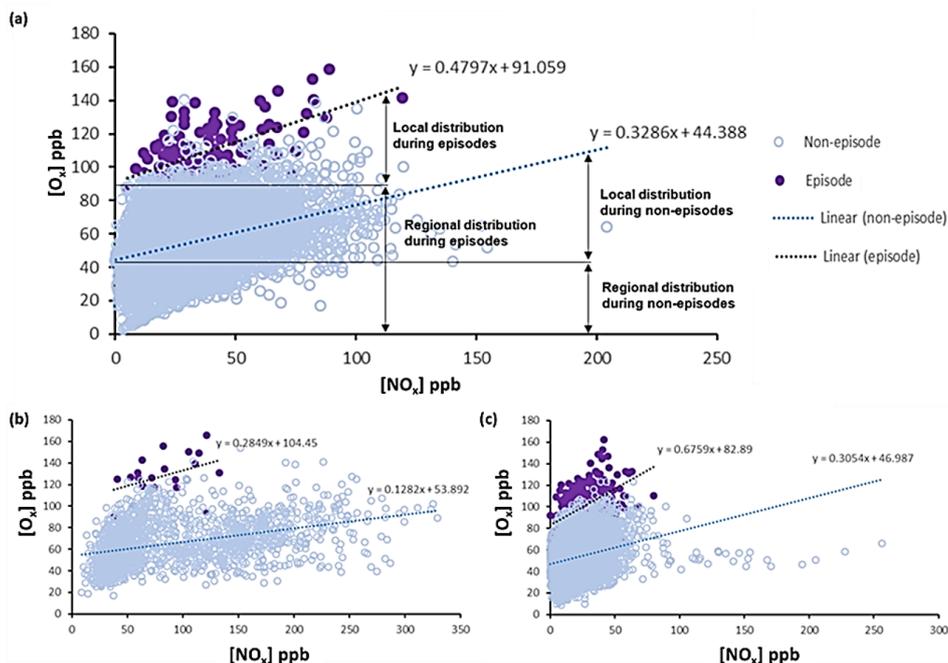
**Table 1.** The comparison of fitted linear regression lines from this study, including at BKK sites, roadside sites and BKK suburb sites with fitted linear regression lines from other studies.

	Non-episode	Episode
This study		
– BKK sites	$[O_x] = 0.33[NO_x] + 44.39$	$[O_x] = 0.48[NO_x] + 91.10$
– roadside sites	$[O_x] = 0.13[NO_x] + 53.89$	$[O_x] = 0.29[NO_x] + 104.45$
– BKK suburb sites	$[O_x] = 0.31[NO_x] + 47.0$	$[O_x] = 0.68[NO_x] + 82.89$
UK <sup>a</sup>	$[O_x] = 0.097[NO_x] + 38.2$	$[O_x] = 0.112[NO_x] + 55.5$
Buenos Aires, Argentina <sup>b</sup>	$[O_x] = 0.099[NO_x] + 22.0$	
Delhi, India <sup>c</sup>	$[O_x] = 0.54[NO_x] + 28.89$	

<sup>a</sup> Clapp and Jenkin (2001). <sup>b</sup> Mazzeo et al. (2005). <sup>c</sup> Tiwari et al. (2015).

both the local and regional contributions of O<sub>x</sub>. It is noteworthy that the pattern of the local and regional contributions at roadside sites during non-episode periods is composed

of two NO<sub>x</sub> concentration regimes. The low-NO<sub>x</sub> regime (NO<sub>x</sub> < 60 ppb) resembles the local and regional contributions during non-episodes over BKK suburb sites. The high-



**Figure 6.** The effects of local and regional contributions on  $O_3$  during non-episode and episode days at (a) BKK sites, (b) roadside sites and (c) BKK suburb sites.

$NO_x$  regime ( $NO_x > 60$  ppb) may represent the typical characteristics of air quality near roads.

The local contributions from the fitted linear regressions are compared with the local contribution that is calculated from the delta  $O_3$  method. A delta  $O_3$  ( $\Delta O_3$ ) analysis was performed to reflect on the intensity of  $O_3$  production in the BMR area (Lindsay and Chameides, 1988). Lindsay et al. (1989) analyzed high- $O_3$  events in Atlanta, GA, USA, and showed that rural background  $O_3$  during high  $O_3$  concentrations ( $[O_3] > 80$  ppb) in the Atlanta metropolitan area was higher than its average and the concentration of  $O_3$  increased from  $\sim 15$  to 20 ppb when the air mass traveled across the city. This enhanced the total  $O_3$  concentration from 80 to 85 ppb. In our study, the differences in the concentrations of  $O_3$  at the upwind and downwind monitoring stations (monitoring stations 20T and 27T) are averaged. The conditions to calculate  $\Delta O_3$  in this study are as follows (1) high  $O_3$  concentrations ( $[O_3] > 80$  ppb) were observed at at least one of the two monitoring stations; (2) the calculation is performed from 10:00 to 16:00 LT during the dry season to avoid the accumulation of air pollutants by surface inversion and the effects of the removal process by wet deposition; (3) National Oceanic and Atmospheric Administration (NOAA) HYSPLIT model backward trajectories revealed N–NE, S–SW wind directions (Fig. 7). Even the  $O_3$  concentrations at the downwind monitoring stations are expected to be greater than the  $O_3$  concentrations at the upwind monitoring stations, a negative  $\Delta O_3$  may be found. The negative  $\Delta O_3$  suggests the deposition of  $O_3$  and/or that  $O_3$  was

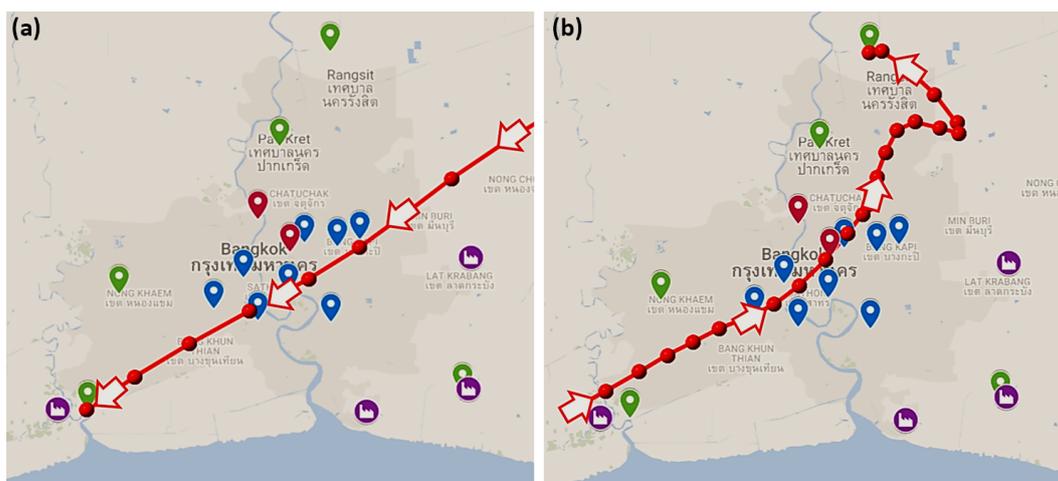
consumed as it passes over the city and/or that there may have been a wind reversal so that air already polluted by the metropolitan area was brought back in to the city (Lindsay et al., 1989). The  $\Delta O_3$  in BMR ranged from  $-53$  to 86 ppb (average  $\sim 10.4$  ppb) and ranged from  $-66$  to 96 ppb (average  $\sim 9.4$  ppb) when the predominant wind directions advecting into the city were from NE and SW, respectively. Thus, we find that there was a  $\sim 10$  ppb enhancement of the  $O_3$  concentration during the air pollution high  $O_3$  concentration in BMR ( $[O_3] > 80$  ppb), which corroborates local  $O_3$  production analysis based on linear regression.

### 3.5 Correlation of air pollutants

#### 3.5.1 Local sources analysis

The characteristics of emission sources are often determined by the ratios between CO and  $NO_x$  ( $CO/NO_x$ ) and  $SO_2$  and  $NO_x$  ( $SO_2/NO_x$ ). In general, the major sources of  $NO_x$  are point sources and mobile sources. However,  $NO_x$  from point sources is more likely correlated with  $SO_2$ .  $NO_x$  from mobile sources is more likely correlated with CO (Parrish et al., 1991). Therefore, the characteristics of mobile sources are high  $CO/NO_x$  ratios and low  $SO_2/NO_x$  ratios. In contrast to mobile sources, the characteristics of point sources are low  $CO/NO_x$  ratios and high  $SO_2/NO_x$  ratios (Parrish et al., 1991; Rasheed et al., 2014).

Table 2 shows the comparison between the  $CO/NO_x$  and  $SO_2/NO_x$  ratios from this study when compared with other studies. The ratio of  $CO/NO_x$  is 19.8, and the ratio of



**Figure 7.** Backward trajectories from the HYSPLIT model reveal a (a) NE wind direction (13 January 2010) and (b) SW wind direction (1 January 2010).

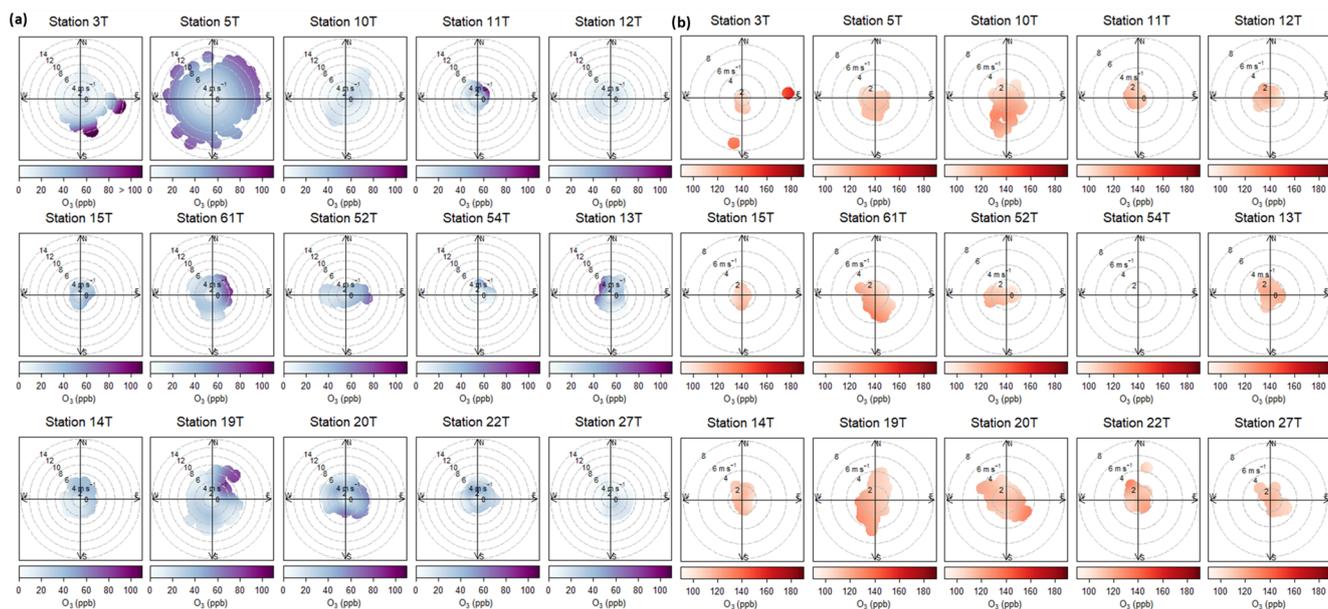
**Table 2.** The comparison of CO / NO<sub>x</sub> and SO<sub>2</sub> / NO<sub>x</sub> ratios from this study with other studies (modified from Rasheed et al., 2014).

Region	Source	CO / NO <sub>x</sub>	SO <sub>2</sub> / NO <sub>x</sub>
This study		19.8	0.1
– BKK sites		18.25	0.09
– roadside sites		21.15	0.11
– BKK suburb sites		19.20	0.09
Eastern US		4.3	0.94
	Mobile	8.4	0.05
	Point	0.95	1.8
Pennsylvania		2.6	1.7
	Mobile	7.8	0.05
	Point	0.8	2.3
Western US		6.7	0.41
	Mobile	10.2	0.05
	Point	1.2	1.1
Denver metropolitan area		7.3	0.19
	Mobile	10.5	0.05
	Point	0.18	0.44
Raleigh, NC		16.3	0.73
New Delhi, India		50	0.58
Guwahati and Nagpur, India <sup>c</sup>			> 0.3
Kolkata and Durgapur, India <sup>c</sup>			≤ 0.13
Madrid, Spain <sup>a</sup>		13.3	0.29
Rouen, France <sup>b</sup>		12–18	
Islamabad, Pakistan			
– based on Emission Inventory (2010)	Mobile	4.94	0.34
	Point	0.63	7.0
– based on ambient data		10	0.01

<sup>a</sup> Fernandez-Jiménez et al. (2003). <sup>b</sup> Coppalle et al. (2001). <sup>c</sup> Mallik and Lal (2014).

SO<sub>2</sub> / NO<sub>x</sub> is 0.1 over BMR. This suggests that the major contributors of primary pollutants over the BMR are mobile sources. However, this region may be influenced by manufacturing facilities' point sources (SO<sub>2</sub> contributor) on the

outskirts of the BKK. These point sources will impact the concentrations of SO<sub>2</sub>, NO<sub>x</sub> and CO. Correlations among species are provided in Table S3, Sect. G.



**Figure 8.** Relationship between the concentrations of  $O_3$ , wind speeds and wind directions during (a)  $O_3$  episodes ( $[O_3]_{\text{hourly}} > 100$  ppb) and (b) during non- $O_3$ -episodes ( $[O_3]_{\text{hourly}} \leq 100$  ppb) over BMR from 2010 to 2014.

### 3.5.2 Effects of pollutant transport

In general,  $O_3$  has a short lifetime in the polluted urban atmosphere (approximately hours). However,  $O_3$  has a longer lifetime of several weeks in the free troposphere. This occurrence may allow  $O_3$  to be transported over continental scales (Stevenson et al., 2006: CO /  $NO_x$ ; Young et al., 2013: CO /  $NO_x$ ; Monks et al., 2015). Figure 8 shows  $O_3$  concentrations, during episodes and non-episodes, with predominant wind directions and wind speeds. The results show that  $O_3$  exceedances are associated with low wind speed and predominant wind directions, i.e., the origins of the air masses. In general, elevated  $O_3$  concentrations were observed with a wind speed lower than  $4 \text{ m s}^{-1}$  with northerly winds (station 22T), southerly winds (stations 3T, 10T, 19T, 20T and 61T) and westerly winds (station 52T). It is noteworthy that the southerly winds, generally, bring cleaner marine air mass to the land. However, under a stagnant condition (i.e., low wind speed), elevated  $O_3$  concentrations were observed during southerly winds (Sahu et al., 2013a, b).

### 3.6 Air quality index for $O_3$ management

In the US, the AQI for air pollutants is divided into six categories (good, moderate, unhealthy for sensitive groups, unhealthy, very unhealthy and hazardous). These categories are nonlinear and relate to human health (U.S. EPA, 2017a, b, c). In Thailand, the NAAQs for the air pollutant species are pegged at an AQI value of 100. In this study, the severity of  $O_3$  concentrations in BMR is evaluated by AQI for  $O_3$ . Table 3 provides the ambient air quality over BMR from

2010 to 2014 based on the AQI of  $O_3$ . Based on the AQI for  $O_3$ , during the study period, the majority of air quality over BMR was in the good AQI category ( $\sim 97\%$ ) followed by the moderate air quality category ( $\sim 2.3\%$ ). However, the unhealthy for sensitive groups ( $\sim 0.7\%$ ), unhealthy ( $\sim 0.3\%$ ) and very unhealthy ( $\sim 0.04\%$ )  $O_3$  air quality categories were observed. Generally, BKK suburb sites have a higher number of hours that were categorized as unhealthy for sensitive groups, unhealthy and very unhealthy than BKK and roadside sites. The average number of hours that were categorized as unhealthy for sensitive groups, unhealthy and very unhealthy over BKK suburb sites were 425.8, 146.7 and 28.7 h. The calculation of the AQI for  $O_3$  can be found in Figs. S5 and S6, Sect. S8.

This study provides measurements and analysis for the gaseous criteria pollutants. However, in order to provide a well-established air quality management policy, the integration of multidisciplinary analysis is needed. This will include scientific, socioeconomic and policy analysis (Aneja et al, 2001). The results from this study revealed evidence of  $O_3$  air quality standards being breached. This resulted in adverse health effects, human welfare, economics, and environment over BMR. Ratio analysis suggests that the first priority should be controlling pollution emissions from local sources that are primarily mobile. The complex relationship between  $O_3$  and its precursors and the effects of pollution transport show that decreasing only  $NO_x$  emissions and/or local emissions may not be an effective policy to reduce  $O_3$  because of regional air pollution transport (i.e., ozone and its precursors contribute to  $O_3$  exceedances). To identify the proportional contribution between local and regional sources

**Table 3.** Number of hours for the different AQI categories of O<sub>3</sub> over the BMR from 2010 to 2014.

AQI	Hour														
	BKK sites							Roadside sites		BKK suburb sites					
	3T	5T	10T	11T	12T	15T	61T	52T	54T	13T	14T	19T	20T	22T	27T
Good	39 018	32 021	27 959	40 715	26 606	33 628	26 442	32 665	40 231	31 070	35 429	33 592	30 793	34 301	26 873
Moderate	310	713	1023	556	367	479	1178	807	27	1620	944	1687	1340	1466	719
Unhealthy for sensitive groups	88	139	225	109	82	108	295	151	0	454	288	515	632	448	218
Unhealthy	19	40	61	30	29	38	85	36	0	195	87	184	209	109	96
Very unhealthy	0	6	12	0	0	10	26	0	0	59	2	51	28	23	9
Hazardous	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0

of O<sub>3</sub> concentrations during selected O<sub>3</sub> episode days, atmospheric modeling is needed to quantify various processes that contribute to the ambient concentration at specific locations. This scientific analysis provides a framework for the process of establishing an air quality policy while analyzing socioeconomic impacts.

#### 4 Conclusions

Among measured gaseous criteria pollutants, O<sub>3</sub> is the only species whose concentrations frequently exceed the NAAQs of Thailand. The O<sub>3</sub> exceedances occur during the dry season (local summer and local winter) and more frequently occur over BKK sites and BKK suburb sites than roadside sites. On average, the number of hourly O<sub>3</sub> exceedances at BKK sites, roadside sites and BKK suburb sites was  $\sim 16$ ,  $\sim 9$  and  $\sim 43 \text{ h yr}^{-1}$ , respectively. The lower number of O<sub>3</sub> exceedances at roadside sites demonstrates the effects of the titration of O<sub>3</sub> by NO due to high concentrations of NO that were generally observed at this monitoring station type (average  $[\text{NO}]_{\text{hourly}} = \sim 166.0 \pm 19.8 \text{ ppb}$ ). Under the photostationary state assumption, during the dry season, the values of the reaction rate coefficient of the photochemical reaction of NO<sub>2</sub> ( $j_1$ ) and the reaction rate coefficient of the chemical reaction between NO and O<sub>3</sub> ( $k_3$ ) from 0.12 to 1.22  $\text{min}^{-1}$  and range from 28.3 to 30.9  $\text{ppm}^{-1} \text{ min}^{-1}$ , respectively. NO<sub>x</sub> values of  $\sim 60 \text{ ppb}$  mark the threshold for the interconversion between O<sub>3</sub>, NO and NO<sub>2</sub>. Under the low-NO<sub>x</sub> regime ( $[\text{NO}_x] < 60 \text{ ppb}$ ), O<sub>3</sub> is the dominant species. On the other hand, under the high-NO<sub>x</sub> regime ( $[\text{NO}_x] > 60 \text{ ppb}$ ), the concentrations of O<sub>3</sub> rapidly decrease. The decrease in O<sub>3</sub> under the high-NO<sub>x</sub> regime describes the important role of NO in destroying O<sub>3</sub> in the atmosphere in polluted environments. The local and regional contributions of O<sub>x</sub> concentrations under stagnant conditions (wind speed  $< 4 \text{ m s}^{-1}$ ) and the origin of air masses containing O<sub>3</sub> and its precursors are associated with elevated O<sub>3</sub> concentrations in this area. During O<sub>3</sub> episodes, the values of the local and regional contributions were about double those during non-episodes. The air quality index for O<sub>3</sub> reveals evidence of air quality standards being breached in BMR, resulting in poten-

tially adverse health effects. To achieve O<sub>3</sub> reduction, control strategies may be needed. Emissions from mobile sources may be the first priority to manage O<sub>3</sub>, since BMR is more likely to be affected by mobile sources than point sources ( $\text{CO} / \text{NO}_x = 19.8$  and  $\text{SO}_2 / \text{NO}_x = 0.1$ ). Due to the highly nonlinear physical and chemical processes governing the atmosphere, control strategies need to be evaluated in a more comprehensive approach. Air quality modeling of pollution episodes in the BMR would be an appropriate approach to accurately quantify various atmospheric processes contributing to high O<sub>3</sub> concentrations in BMR.

**Data availability.** The data may be obtained upon request from the Director of the Air Quality and Noise (AQNIS) Management Bureau, Pollution Control Department, Ministry of Natural Resources and Environment, Phahonyothin Rd, Samsen Nai, Phaya Thai, Bangkok, Thailand, 10400 (aqnis.web@gmail.com); tel: +66 2 298 2318; fax: +66 2 298 5389.

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**Author contributions.** PU is a PhD graduate student who developed the idea, analyzed the data, and performed the computations. VPA is a Professor, and AFH, Research Professor are advising the student. All the co-authors contributed in the development of the manuscript.

**Competing interests.** The authors declare that they have no conflict of interest.

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