

Assessment of Gaseous Criteria Pollutants in Bangkok Metropolitan Region, Thailand

Pornpan Uttamang¹, Viney P Aneja¹, Adel Hanna^{1,2}

¹Department of Marine, Earth, and Atmospheric Sciences, North Carolina State University, Raleigh, NC, 27695, USA

²Institute for the Environment, University of North Carolina at Chapel Hill, Chapel Hill, NC, 27517, USA

Correspondence to: Pornpan Uttamang (puttama@ncsu.edu)

Abstract. Analysis of gaseous criteria pollutants in Bangkok Metropolitan Region (BMR), Thailand, during 2010 to 2014 reveals that while the hourly concentrations of CO, SO₂ and NO₂ were mostly within the National Ambient Air Quality Standards (NAAQs) of Thailand; however, the hourly concentrations of O₃ frequently exceeded the standard. The results reveal that the problem of high O₃ concentration continuously persisted in this area. The O₃ photolytic rate constant (j_I) for BMR calculated based on assuming photostationary state (ranged from 0.008 to 0.013 s⁻¹) which is similar to the calculated j_I using the NCAR TUV model (0.021±0.0024 s⁻¹). Interconversion between O₃, NO and NO₂ indicates crossover points between the species occur when the concentration of NO_x (= NO + NO₂) is ~60 ppb. Under low NO_x regime ([NO_x] < 60 ppb), O₃ is the dominant species, while, under high NO_x regime ([NO_x] > 60 ppb), NO dominates. Linear regression analysis between the concentrations of O_x (= O₃ + NO₂) and NO_x provides the role of local and regional contributions to O_x. During O₃ episodes ([O₃]_{hourly} > 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 O₃ categories were observed during episode conditions in the region.

1. Introduction

Over the last three decades, Thailand's rapid industrialization and urbanization has led to an increase in global economic prowess (World Bank, 2018). A majority of the country's development has occurred within and around Bangkok (BKK) (13.7° N and 100.5° E), the capital city of Thailand and Bangkok Metropolitan Region (BMR). BMR is comprised of BKK and the five adjacent provinces of BKK (World Bank, 2018 and 2018a). The increase in emissions is due to accelerated growth in automotive and industrial activities. As a major metropolitan area, BMR

31 is dominated by mobile emissions sources, which contributes to the emissions of CO and NO_x,
32 precursors of ozone (O₃) formation. The emissions from industrial activities also contributes to
33 those emissions, and to the emissions of sulfur dioxide (SO₂) and the formation of particulate
34 matter. Since 1995, BMR has begun to experience air quality degradation and experienced
35 exceedances in Thailand NAAQs for particulate matter (PM) and ozone (O₃) (PCD, 2015) owing
36 to strong solar radiation (peak density of direct radiation ~1,350 kWh m⁻² yr⁻¹), high temperature
37 (yearly average ~29 °C), and high humidity (yearly average ~64%) (Kumar et al., 2012).

38 The relationship between air pollution and public health in BMR has been observed in
39 several published studies. Ruchirawat et al. (2007) reported that children who lived in BKK were
40 exposed to high levels of carcinogenic air pollutants which might cause an elevated cancer risk.
41 Buadong et al. (2009) reported the exposure to elevated PM and O₃ during the previous day, in
42 elderly patients (≥ 65 years), was associated with increasing the number of daily hospital visits for
43 cardiovascular diseases. Jinsart et al. (2002, 2012) reported police personnel and drivers in BKK
44 tended to be exposed to higher level of PM concentrations compared with the general environment.

45 Several studies have demonstrated the role of atmospheric processes in elevating
46 Thailand's O₃. Long-range transport from the Asian continent has enhanced O₃ concentrations in
47 Thailand compared to the lesser O₃ concentrations disbursed using long-range transports from the
48 Indian Ocean (Pochanart et al., 2001). This regional transport, moreover, played an important role
49 in seasonal fluctuations of O₃ in this area (Zhang and Oahn, 2002). Another factor that enhanced
50 O₃ concentrations was the atmospheric chemistry of volatile organic compounds
51 (VOCs). However, this process tended to be more important to enhance O₃ concentrations in
52 suburban areas than in urban areas (Suthawaree et al., 2012).

53 Therefore, the availability and analysis of multi-year measurements of such gaseous
54 criteria pollutants in the BMR will improve our understanding of how they contribute to the air
55 quality of this area. In this study, we analysed diurnal variations, seasonal variations and inter-
56 annual trends of gaseous pollutants including carbon monoxide (CO), nitric oxide (NO), nitrogen
57 dioxide (NO₂), SO₂ and O₃ during 2010 to 2014, in BMR. Chemical and physical processes
58 associated with high O₃ concentrations have been investigated. Since the concentrations of
59 nitrogen oxide (NO_x) were mostly measured at the monitoring station, therefore, O₃ precursors in
60 this study is referred to NO_x. The photochemical reaction for O₃ was investigated during the

61 photostationary state. The effects of local emission and regional contributions of O_x are presented.
62 The severity of air pollution concentrations in BMR on human health are assessed by performing
63 Air Quality Index (AQI).

64 **2. Methodology**

65 **2.1 Study Area**

66 Figure 1 shows a map of BMR, the location of monitoring stations in this study and major
67 monsoon winds over this region. BMR refers to BKK and the five adjacent provinces, including
68 Nakhon Pathom, Pathum Thani, Nonthaburi, Samut Prakan, and Samut Sakhon. These provinces
69 are linked to BKK in terms of traffic and industrial development (Zhang and Oanh, 2002). Thailand
70 has three official seasons—local summer (February to May), rainy (May to October) and local
71 winter (October to February) as per the Thai Meteorological Department (TMD) (TMD, 2015).
72 During the rainy season, this region’s weather is influenced by Southwest monsoon wind that
73 travels from the Indian Ocean to Thailand. This marine air mass contains high moisture, resulting
74 in the wet season in Thailand. During this season, Thailand is characterized by cloudy weather
75 with high precipitation and high humidity. From October to April, this region is influenced by
76 Northeast monsoon wind that travels from the north-eastern and the northern parts of Asia (China
77 and Mongolia). This monsoon wind brings a cold and dry air mass, which leads to the dry season
78 (local summer and local winter) in Thailand. The local winter in Thailand is characterized by cool
79 and dry weather, while the local summer is characterized by hot (35 to 40 °C) to extremely hot
80 weather (> 40 °C) due to strong solar radiation. During the dry season, storms may occur during
81 the seasonal transition (TMD, 2015).

82 Transportation and industrial sectors are considered to be the major sources of air pollutants
83 in the study area (Watcharaviton et al., 2013). In 2014, ~36 million new vehicles were registered
84 in Thailand and 29% of these cars were registered in BKK (DLT, 2015). About 56% and 28% of
85 the registered vehicles in BKK were gasoline and diesel engines. The remaining 16% were
86 Compressed Natural Gas (CNG) (DLT, 2017). In fact, the outskirts of BKK are populated with a
87 variety of metal, auto parts, paper, plastic, food and chemical manufacturing facilities and power
88 plants (DIW, 2016; 2016a; 2016b; 2016c and 2016d).

89 2.2 Data Collection and Data Analysis

90 Over the five-year period, January 1, 2010 to December 31, 2014, hourly observations from
91 15 Pollution Control Department (PCD) monitoring stations were analysed. The monitoring
92 stations are categorized into three categories: BKK sites, roadside sites, and BKK suburb sites.
93 BKK sites refer to the monitoring stations that are located within BKK's residential, commercial,
94 industrial and mixed areas. They are within ~50 to 100 m away from the road. Roadside sites refer
95 to the monitoring stations that are located in BKK within 2 to 5 m from the road (Zhang and Oanh,
96 2002). BKK suburb sites refer to the monitoring stations that are located in the provinces adjacent
97 to BKK (Figure 1). Quality assurance and quality control on the data set were performed by PCD
98 prior to receiving the data. Hourly observations of the gaseous pollutants and meteorological
99 parameters were automatically collected with auto calibration at the monitoring stations. Manual
100 quality control was performed when unusual observations were found. External audit of the
101 equipment and monitoring stations were done every year. Data availability and details of
102 equipment calibrations are provided in Section I, supplement material.

103 Gaseous species were measured at 3 m above ground level (AGL). CO was measured using
104 non-dispersive infrared detection (Thermo Scientific 48i). NO and NO₂ were measured using
105 chemiluminescence detection (Thermo Scientific 42i). SO₂ was measured using ultraviolet (UV)
106 fluorescence detection (Thermo Scientific 43i) and O₃ is measured by using UV absorption
107 photometry detection (Thermo Scientific 49i). The meteorological parameters including wind
108 speed (WS) and wind direction (WD) were measured at 10 m AGL by cup propeller and
109 potentiometer wind vanes. Temperature (T) and relative humidity (RH) were measured at 2 m
110 AGL by thermistor and thin film capacitor, respectively (Watchravitoon et al., 2013). All the
111 meteorological measurements were made by Met One or equivalent method.

112 Data analysis, statistical analysis and plots were developed using Excel 2016. Predominant
113 wind directions related to O₃ concentrations are obtained using Openair package (tool for the
114 analysis of air pollution data) on the RStudio program (<https://www.rstudio.com/>).

115 3. Result and Discussion

116 3.1 Status of Pollution in BMR during 2010 to 2014

117 Figure 2 a) to e) show the maximum and average concentrations of gaseous pollutants,
118 during 2010 to 2014 from the 15 monitoring stations. These concentrations are compared with the
119 hourly NAAQs of Thailand (NAAQs of Thailand for hourly CO, NO₂, SO₂ and O₃ are 30 ppm,
120 170 ppb, 300 ppb and 100 ppb, respectively (PCD, 2018). Since, NO is not a criteria pollutant,
121 only the maximum and average concentrations are presented. During the study period, the
122 maximum concentrations of CO, NO₂ and SO₂ were mostly in their hourly standards (an
123 exceedance of NO₂ was found at 52T monitoring station, during 2013). However, the maximum
124 concentrations of O₃ exceeded its standard. Elevated CO, NO, and NO₂ concentrations were
125 frequently observed at roadside sites than other sites. The average concentrations of CO, NO, and
126 NO₂, 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.
127 Elevated SO₂ were commonly observed at BKK suburb sites than other sites. The average
128 concentrations of SO₂ at BKK suburb sites were $\sim 4.0 \pm 2.3$ ppb. The average concentrations of O₃
129 during daytime (6:00 to 18:00 LT) over BKK sites, roadside sites and BKK suburb sites were
130 $\sim 24.4 \pm 13.5$ ppb, $\sim 18.2 \pm 12.3$ ppb and $\sim 27.7 \pm 14.7$ ppb, and those values during night-time (18:00
131 to 6:00 LT) were $\sim 11.3 \pm 3.3$ ppb, $\sim 9.1 \pm 4.9$ ppb and $\sim 14.2 \pm 5.4$ ppb, respectively. The 24-hour
132 average O₃ concentrations were highest at BKK suburb sites ($\sim 21.4 \pm 3.3$ ppb) and following by
133 BKK sites (18.6 ± 2.3 ppb) and roadside sites (13.9 ± 8.6 ppb). Statistical analysis of the
134 concentrations of gaseous pollutants at the three monitoring types are provided in Table I, Section
135 B, supplement material.

136 The seasonal variations of the gaseous pollutants reveal that, in general, elevated
137 concentrations were observed during dry seasons and those decreased during wet seasons (Figure
138 II, Section C, supplement material). Inter-annual variations of the gaseous pollutants reveal that
139 while the concentrations of CO, NO₂ and SO₂ decreased or remained constant, the concentration
140 of O₃ tended to increase during the study period (Figure III, Section D, supplement material).

141 An O₃ exceedance was recorded when an hourly concentration of O₃ was greater than 100
142 ppb (hourly O₃ standard). Figure 2 f) to g) illustrate the number of hourly O₃ exceedances, which
143 they are shown by locations and by seasons, respectively. The hourly O₃ exceedances at BKK

144 suburb sites were more frequently observed than those at the other sites. The average number of
145 hourly O₃ exceedances was ~16 hours year⁻¹ at BKK sites, ~9 hours year⁻¹ at roadside sites and ~43
146 hours year⁻¹ at BKK suburb sites. The hourly O₃ exceedances were commonly observed during dry
147 season, less during the transitional period between the seasons (May), and rarely observed during
148 wet season.

149 **3.2 Diurnal Variation of the Gaseous Species**

150 Diurnal variations of gaseous pollutant are shown in Figure 3 a) to c). The diurnal variations
151 of O₃ show a single-peak pattern (Aneja et al., 2001) with the concentrations increasing after
152 sunrise and reached the peak ~15:00 local time (LT). The concentrations begin to decline in the
153 evening and reach the minimum concentrations ~7:00 LT in the next morning. The concentrations
154 of O₃ at the peaks were ~40 ppb at BKK sites, ~30 ppb at roadside sites and ~45 ppb at BKK
155 suburb sites. The diurnal variations of NO show a bimodal pattern with the concentrations reach
156 the first- and the second-peak ~7:00 to 9:00 LT and ~21:00 to 22:00 LT, respectively. The
157 concentrations of NO at the first- and the second-peak were ~40 ppb and ~23 ppb at BKK sites,
158 ~110 ppb and ~73 ppb at roadside sites, and ~30 ppb and ~13 ppb at BKK suburb sites. The
159 concentrations of NO₂ at the first- and the second-peak were ~23 ppb and ~28 ppb at BKK sites,
160 ~33 ppb and ~37 ppb at roadside sites, and ~20 ppb and ~22 ppb at BKK suburb sites. Even the
161 diurnal variations of NO_x show a bimodal pattern, at roadside sites, the pattern was flatter than at
162 other sites. The flatter pattern of NO_x at roadside sites reveals that this monitoring station type was
163 affected by high concentration of NO_x all day. The diurnal variations of CO show a bimodal pattern
164 with the first- and the second-peak occurred ~8:00 LT and 21:00 LT, respectively. The
165 concentrations of CO at the first- and the second-peak were ~1 ppm (both peaks) at BKK sites, ~2
166 and ~1.5 ppm at roadside sites, and ~1 ppm (both peaks) at BKK suburb sites. The first peak of
167 the diurnal variations of NO, NO₂, and CO correspond with morning rush hour in BKK (7:00 to
168 9:00 LT). The second peak of those occurred ~3 to 5 hours after the evening traffic rush hour
169 (16:00 to 18:00 LT) (Leong et al., 2002), due to a combination of pollutants emissions and collapse
170 of the planetary boundary layer (weak turbulence and diffusion) during this time. The diurnal
171 variations of SO₂ show a bimodal pattern with the first- and the second-peak of SO₂ occurred ~8:00
172 LT and 21:00 LT, respectively. The concentrations of SO₂ at the first- and the second-peak were
173 ~4 ppb and ~3 ppb at BKK sites and roadside sites, and ~6 ppb and ~3 ppb at BKK suburb sites.

174 At the roadside sites, the peaks are more obvious than the other sites. The result indicates that at
175 this monitoring station type, SO₂ is primarily influenced by emissions from vehicle exhaust using
176 high sulfur content fuel (Henschel et al. 2013). It is noteworthy that BKK has a large diesel engine
177 fleet (an estimated 25 % of registered vehicles) (DLT, 2015). The diesel fuel contains ~0.035 %wt
178 Sulphur (DOEB, 2017). Season wise of the diurnal variations are provided in Figure IV, Section
179 E, supplement material.

180 Figure 4 a) to c) shows diurnal variations of rate of change of O₃ concentration ($\Delta[\text{O}_3]/\text{dt}$)
181 during dry seasons (local summer and local winter) and wet seasons at the three monitoring station
182 types (the data has been averaged for each monitoring station type to capture the rate of change of
183 O₃ concentration characteristics). The diurnal variations of $\Delta[\text{O}_3]/\text{dt}$ is a combination of O₃
184 chemistry and meteorology. In general, $\Delta[\text{O}_3]/\text{dt}$ during wet season were lower than those during
185 dry season. However, during local winter, the rates of change O₃ concentration were the highest.
186 The $\Delta[\text{O}_3]/\text{dt}$ at the three monitoring station types, during 10:00 to 11:00 LT, were 4.5 to 7.0 ppb
187 hr⁻¹ during wet seasons, 6.7 to 7.5 ppb hr⁻¹ during local summers, and 5.7 to 9.2 ppb hr⁻¹ during
188 local winters. The $\Delta[\text{O}_3]/\text{dt}$ became negative during 14:00 to 15:00 LT. As expected, the rate of
189 change of O₃ concentration was nearly constant during nighttime. Rapid changes in the mixing
190 height and solar insolation during morning increases $\Delta[\text{O}_3]/\text{dt}$. After sunset, the formation of O₃ is
191 inhibited and the planetary boundary layer becomes more stable resulting in O₃ reduction through
192 chemical reactions (for example, the oxidation of O₃ by NO_x) and physical processes (for example,
193 dry deposition to the earth surface) (Naja and Lal, 2002).

194 **3.3 Photochemical Reaction and Interconversion between O₃, NO and NO₂**

195 The primary precursors for tropospheric O₃, in the urban environment, are NO_x and non-
196 methane volatile organic compounds (VOCs), methane or CO (The Royal Society, 2008; Monks
197 et al., 2009; Cooper et al., 2014). While NO_x was measured continuously at all the monitoring
198 sites, VOCs were measured periodically only at one monitoring station limiting its usefulness as
199 part of this study. In this study, the photostationary state (PSS) is applied through the chemical
200 reactions of O₃ formation during 10:00 to 16:00 LT. This time window is chosen due to the fully
201 developed planetary boundary layer with well-mixed condition (Pochanart et al., 2001) to avoid

202 accumulation of air pollutants by surface inversion. Analysis and calculation are performed only
203 during dry season to eliminate effects of the removal process by wet deposition.

204 The relationship among NO, NO₂ and O₃ under PSS is presented by Eq. (1) (Seinfeld and
205 Pandis, 1998).

$$206 \quad [O_3]_{PSS} = \frac{j_I[NO_2]}{k_3[NO]} \quad \text{Eq. (1)}$$

207 Where $[O_3]_{PSS}$ is the concentration of O₃, at PSS, j_I and k_3 are reaction rate coefficient of
208 photochemical reaction of NO₂ and reaction rate coefficient of chemical reaction between NO and
209 O₃, respectively.

210 The values for k_3 (ppm⁻¹ min⁻¹) is calculated by Eq. (2) (Seinfeld and Pandis, 1998; Tiwari
211 et al., 2015).

$$212 \quad k_3 = 3.23 \times 10^3 \exp[-1430/T] \quad \text{Eq. (2)}$$

213 During dry seasons, the values of j_I ranged from 0.12 to 1.22 min⁻¹, and the average of those at
214 BKK sites, roadside sites and BKK suburb sites were 0.74±0.2, 0.64±0.3 and 0.55±0.3 min⁻¹,
215 respectively. The rate coefficients are calculated using the NCAR TUV model, during 2010 the
216 dry season, for 10:00 LT to 16:00 LT, at the latitude and longitude of 13.76 °N and 100.50 °E. The
217 average j_I value calculated from the NCAR TUV model is 0.021±0.0024 s⁻¹, which is similar to
218 the calculated j_I values from Eq. (1) (j_I ranges from 0.008 to 0.013 s⁻¹). The values of j_I from this
219 study are similar to those values at an urban background site in Delhi, India (values of j_I ranged
220 from 0.4 to 1.8 min⁻¹ and the average was 0.8 min⁻¹) (Tiwari et al., 2015) and those values collected
221 during a November daytime in the UK (values of j_I was ~0.14 min⁻¹) (Clapp and Jenkin, 2001).

222 The values of k_3 , during dry seasons, ranged from 28.3 to 30.9 ppm⁻¹ min⁻¹, and the average
223 of those at BKK sites, roadside sites and BKK suburb sites were 29.8±0.7, 29.7 and 29.8±0.7 ppm⁻¹
224 min⁻¹, respectively. The ratio of [NO₂] and [NO] was ~1.9. The statistical analysis of j_I (min⁻¹
225 and s⁻¹) and k_3 (ppm⁻¹ min⁻¹ and cm³ molecule⁻¹ s⁻¹) at the three monitoring station types using Eq.
226 (1), and the average j_I calculated from the NCAR TUV model are provided in Table II, Section F,
227 supplement material.

228 Figure 5 a) to c) shows the relationships between NO, NO₂ and O₃, their crossover points,
229 and concentration distributions. The crossover point among species occurs when the concentration

230 of NO_x is ~ 60 ppb. At this point, two regimes are identified, including low NO_x regime and high
231 NO_x regime. Under the low NO_x regime ($[\text{NO}_x] < 60$ ppb), O_3 is the dominant species among the
232 others and NO_2 concentrations are higher than NO for NO_x species. Conversely, under the high
233 NO_x regime ($[\text{NO}_x] > 60$ ppb), NO and NO_2 increase and the concentrations of O_3 rapidly decrease.
234 Under the high NO_x regime, the declination of O_3 trend-lines may describe O_3 removal process
235 through the titration of O_3 by NO .

236 **3.4 Local and Regional Contribution to O_x**

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

245 Figure 6 a) to c) shows the local and regional contributions of O_x at the three monitoring
246 station types. The effects of the local and regional contributions to O_x concentration are analysed
247 by plotting O_x concentrations against NO_x concentrations and fitting the plot with a linear
248 regression ($y = mx + c$). The concentration of NO_x and O_x are referred by x and y , respectively.
249 The slope of the linear regression (m) implies the local contribution and the intercept with the y -
250 axis (c) implies the regional (background) contribution (Aneja et al., 2000; Clapp and Jerkin, 2001;
251 Notario et al., 2012). Table 1 shows the comparison between fitted linear regressions from this
252 study with fitted linear regression lines from other studies. The average background O_x
253 concentrations over BMR during non-episodes ($[\text{O}_3]_{\text{hourly}} < 100$ ppb) and episodes ($[\text{O}_3]_{\text{hourly}} > 100$
254 ppb) were ~ 48 ppb and ~ 95 ppb, respectively. The local and regional contributions during the
255 episode days, in general, were about double of those during the non-episode days. The results
256 reveal that elevated O_3 concentrations during the episode days are influenced by both the local and
257 regional contributions of O_x . It is noteworthy that the pattern of the local and regional contributions
258 at roadside sites during non-episode period is composed of two NO_x concentration regimes. The

259 low NO_x regime ($\text{NO}_x < 60$ ppb) resembles the local and regional contributions during non-episode
260 over BKK suburb sites. The high NO_x regime ($\text{NO}_x > 60$ ppb) may represent typical characteristic
261 of air quality near roads.

262 The local contributions from the fitted linear regressions are compared with the local
263 contribution that is calculated from delta O_3 method. A delta O_3 (ΔO_3) analysis was performed to
264 reflect on the intensity of O_3 production in BMR area (Lindsay and Chameides, 1988). Lindsay et
265 al. (1989) analysed high- O_3 events in Atlanta, GA, and showed that rural background O_3 during
266 high O_3 concentrations ($[\text{O}_3] > 80$ ppb) in Atlanta Metropolitan Area were higher than its average
267 and the concentration of O_3 increased from ~ 15 to 20 ppb when the air mass travelled across the
268 city. This enhanced the total O_3 concentration to 80 to 85 ppb. In our study, during the different in
269 the concentrations of O_3 at the upwind and downwind monitoring stations (20T and 27T
270 monitoring station) are averaged. The conditions to calculate ΔO_3 in this study are 1) high O_3
271 concentrations ($[\text{O}_3] > 80$ ppb) were observed at least one of the two monitoring stations 2) the
272 calculation is performed 10:00 to 16:00 LT, during dry season, to avoid accumulation of air
273 pollutants by surface inversion and effects of the removal process by wet deposition 3) National
274 Oceanic and Atmospheric Administration (NOAA) HYSPLIT model backward trajectories
275 revealed N-NE, S-SW wind directions (Figure 7). Even the O_3 concentrations at the downwind
276 monitoring stations are expected to be greater than the O_3 concentrations at the upwind monitoring
277 stations, a negative ΔO_3 may be found. The negative ΔO_3 suggests deposition of O_3 and/or O_3 was
278 consumed as it passes over the city and/or there may have been a wind reversal so that air already
279 polluted by the metropolitan area was brought back in to the city (Lindsay et al., 1989). The ΔO_3
280 in BMR ranged from -53 to 86 ppb (average ~ 10.4 ppb.) and ranged from -66 to 96 ppb (average
281 ~ 9.4 ppb.) when the predominant wind direction advecting into the city were from NE and SW,
282 respectively. Thus, we find that there was ~ 10 ppb enhancement of the O_3 concentration during
283 the air pollution high O_3 concentration in BMR ($[\text{O}_3] > 80$ ppb), which corroborates local O_3
284 production analysis based on linear regression.

285 **3.5 Correlation of Air Pollutants**

286 **3.5.1 Local Sources Analysis**

287 Characteristic of emission sources are often determined by the ratios between CO and NO_x
288 (CO/NO_x) and SO₂ and NO_x (SO₂/NO_x). In general, the major sources of NO_x are point sources
289 and mobile sources. However, NO_x from point sources is more likely correlated with SO₂. NO_x
290 from mobile sources is more likely correlated with CO (Parrish et al., 1991). Therefore, the
291 characteristics of mobile source are high CO/NO_x ratios and low SO₂/NO_x ratios. In contrast to
292 mobile sources, the characteristic of point sources are low CO/NO_x ratios and high SO₂/NO_x ratios
293 (Parrish et al., 1991; Rasheed et al., 2014).

294 Table 2 shows the comparison between the CO/NO_x and SO₂/NO_x ratios from this study
295 and when compared with other studies. The ratio of CO/NO_x is 19.8 and the ratio of SO₂/NO_x is
296 0.1 over BMR. This suggests that the major contributors of primary pollutants over the BMR are
297 mobile sources. However, this region may be influenced by manufacturing facilities' point sources
298 (SO₂ contributor) on the outskirts of the BKK. These point sources will impact the concentrations
299 of SO₂, NO_x and CO. Correlations among species are provided in Table II, Section G, supplement
300 material.

301 **3.5.2 Effects of Pollutant Transport**

302 In general, O₃ has a short lifetime in polluted urban atmosphere (approximately hours).
303 However, O₃ has a longer lifetime of several weeks in the free troposphere. This occurrence may
304 allow O₃ to be transported over continental scales (Stevenson et al., 2006; Young et al., 2013;
305 Monks et al., 2015). Figure 8 shows O₃ concentrations, during episodes and non-episodes, with
306 predominant wind directions and wind speeds. The results show that O₃ exceedances are associated
307 with low wind speed and predominant wind directions i.e. origins of the air masses. In general,
308 elevated O₃ concentrations were observed with wind speed lower than 4 ms⁻¹ with northerly winds
309 (22T station), southerly winds (3T, 10T, 19T, 20T and 61T stations) and westerly winds (52T
310 station). It is noteworthy that the southerly winds, generally, bring cleaner marine air mass to the
311 land. However, under a stagnant condition (i.e. low wind speed), elevated O₃ concentrations were
312 observed during southerly winds (Sahu et al., 2013; 2013a).

313 **3.6 Air Quality Index for O₃ Management**

314 Air Quality Index (AQI) for air pollutants, in the US, is categorized into six categories
315 (good, moderate, unhealthy for sensitive groups, unhealthy, very unhealthy, and hazardous). These
316 categories are nonlinear and relate to human health (US.EPA, 2017, 2017a, 2017b). In Thailand,
317 the NAAQs for the air pollutant species is pegged at an AQI value of 100. In this study, the severity
318 of O₃ concentrations in BMR is evaluated by AQI for O₃. Table 3 provides the ambient air quality
319 over BMR during 2010 to 2014 based on the AQI of O₃. Based on the AQI for O₃, during the study
320 period, the majority of air quality over BMR was in the good AQI category (~97 %), followed by
321 the moderate air quality category (~2.3%). However, unhealthy for sensitive group (~0.7 %),
322 unhealthy (~0.3%) and very unhealthy (~0.04%) O₃ air quality categories were observed.
323 Generally, BKK suburb sites have higher number of hours that were found in the unhealthy for
324 sensitive group, unhealthy and very unhealthy categories than BKK and roadside sites. The
325 average number of hours that were found in unhealthy for sensitive group, unhealthy and very
326 unhealthy categories over BKK suburb sites were 425.8, 146.7 and 28.7 hours. The calculation of
327 the AQI for O₃ can be found in Section H, supplement material.

328 This study provides measurements and analysis for the gaseous criteria pollutants.
329 However, in order to provide a well-established air quality management policy, the integration of
330 multidisciplinary analysis is needed. This will include scientific, socioeconomic, and policy
331 analysis (Aneja et al, 2001). The results from this study revealed evidence of violations for O₃ for
332 air quality. This resulted in adverse health effects, human welfare, economics, and environment
333 over BMR. Ratio analysis suggests that the first priority should be controlling pollution emissions
334 from local sources that come primarily from mobile sources. The complexity between O₃ and its
335 precursors and the effects of pollution transport shows that decreasing only NO_x emissions and/or
336 local emissions may not be an effective policy to reduce O₃ since regional air pollution transport
337 (i.e. ozone and its precursors contribute to O₃ exceedances). To identify the proportional
338 contribution between local and regional sources of O₃ concentrations during selected O₃ episode
339 days, atmospheric modeling is needed to quantify various processes that contribute to the ambient
340 concentration at specific locations. This scientific analysis provides a frame work for the process
341 of establishing an air quality policy while developing socioeconomic impacts.

342 4. Conclusion

343 Among measured gaseous criteria pollutants, O₃ is the only specie whose concentrations
344 frequently exceed the NAAQs of Thailand. The O₃ exceedances occur during the dry season (local
345 summer and local winter) and most frequently occur over BKK sites and BKK suburb sites than
346 roadside sites. On average, the number of hourly O₃ exceedances at BKK sites, roadside sites and
347 BKK suburb sites were ~16 hours year⁻¹, ~9 hours year⁻¹ and ~43 hours year⁻¹, respectively. The
348 lower number of O₃ exceedances at roadside sites demonstrates the effects of the titration of O₃ by
349 NO, due to, high concentrations of NO that were generally observed at this monitoring type
350 (average [NO]_{hourly} = ~166.0±19.8 ppb). Under photostationary state assumption, during dry
351 season, the values of reaction rate coefficient of photochemical reaction of NO₂ (j_I) and reaction
352 rate coefficient of chemical reaction between NO and O₃ (k_3) range from 0.12 to 1.22 min⁻¹ and
353 range from 28.3 to 30.9 ppm⁻¹ min⁻¹, respectively. NO_x values of ~ 60 ppb, marks the threshold for
354 the interconversion between O₃, NO and NO₂. Under the low NO_x regime ([NO_x] < 60 ppb), O₃ is
355 the dominant species. On the other hand, under the high NO_x regime ([NO_x] > 60 ppb), the
356 concentrations of O₃ rapidly decrease. The decrease of O₃ under the high NO_x regime describes
357 the important role of NO in destroying O₃ in the atmosphere in polluted environments. The local
358 and regional contributions of O_x concentrations, under stagnant condition (wind speed < 4 m s⁻¹)
359 and origin of air masses containing O₃ and its precursors associate with elevated O₃ concentration
360 in this area. During O₃ episodes, the values of the local and regional contributions were about
361 double of those during non-episodes. Air Quality Index for O₃ reveals evidence of violations for
362 air quality standards, in BMR, resulting in potential adverse health effects. To achieve O₃
363 reduction, control strategies may be needed. Emissions from mobile sources may be the first
364 priority to manage O₃, since BMR is more likely affected by mobile sources than point sources
365 (CO/NO_x = 19.8 and SO₂/NO_x = 0.1). Due to the highly nonlinear physical and chemical processes
366 governing the atmosphere, control strategies need to be evaluated in a more comprehensive
367 approach. Air quality modelling of pollution episodes in the BMR would be an appropriate
368 approach to accurately quantify various atmospheric processes contributing to high O₃
369 concentrations in BMR.

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372 **Data Availability**

373 Hourly observations in this study were provided by Pollution Control Department (PCD),
374 Thailand.

375 Address: 92 Phahonyothin Rd, Khwaeng Samsen Nai, Khet Phaya Thai, Krung Thep Maha
376 Nakhon 10400, Thailand.

377 Phone: +66 2 298 2000

378 Website: <http://www.pcd.go.th/>

379 **Competing Interest**

380 The authors declare that they have no conflict of interest.

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388 Mr. Kurt Thurber for their assistance in the editorial review of the manuscript. We also thank the
389 two anonymous reviewers for their constructive comments.

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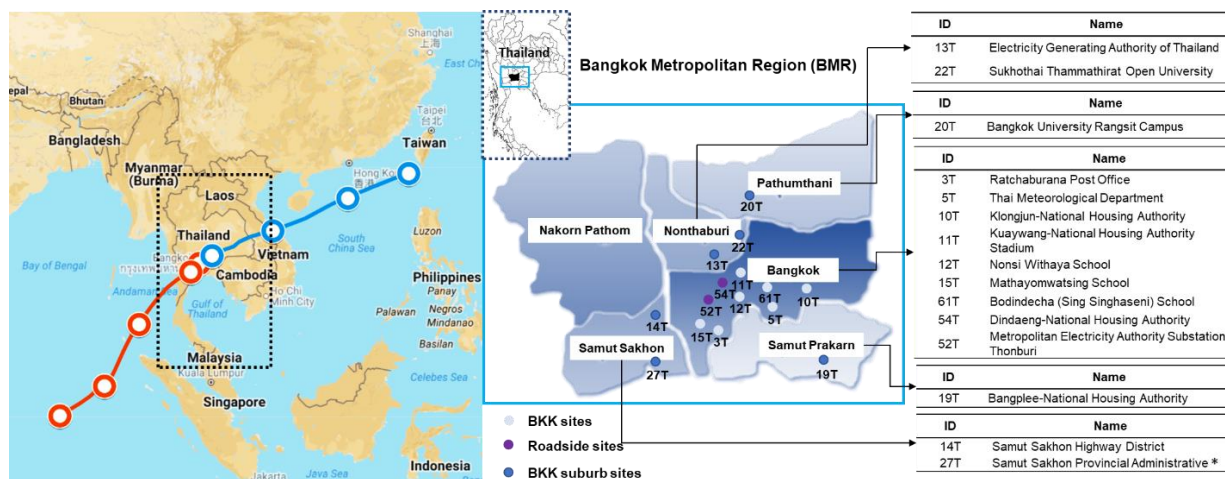
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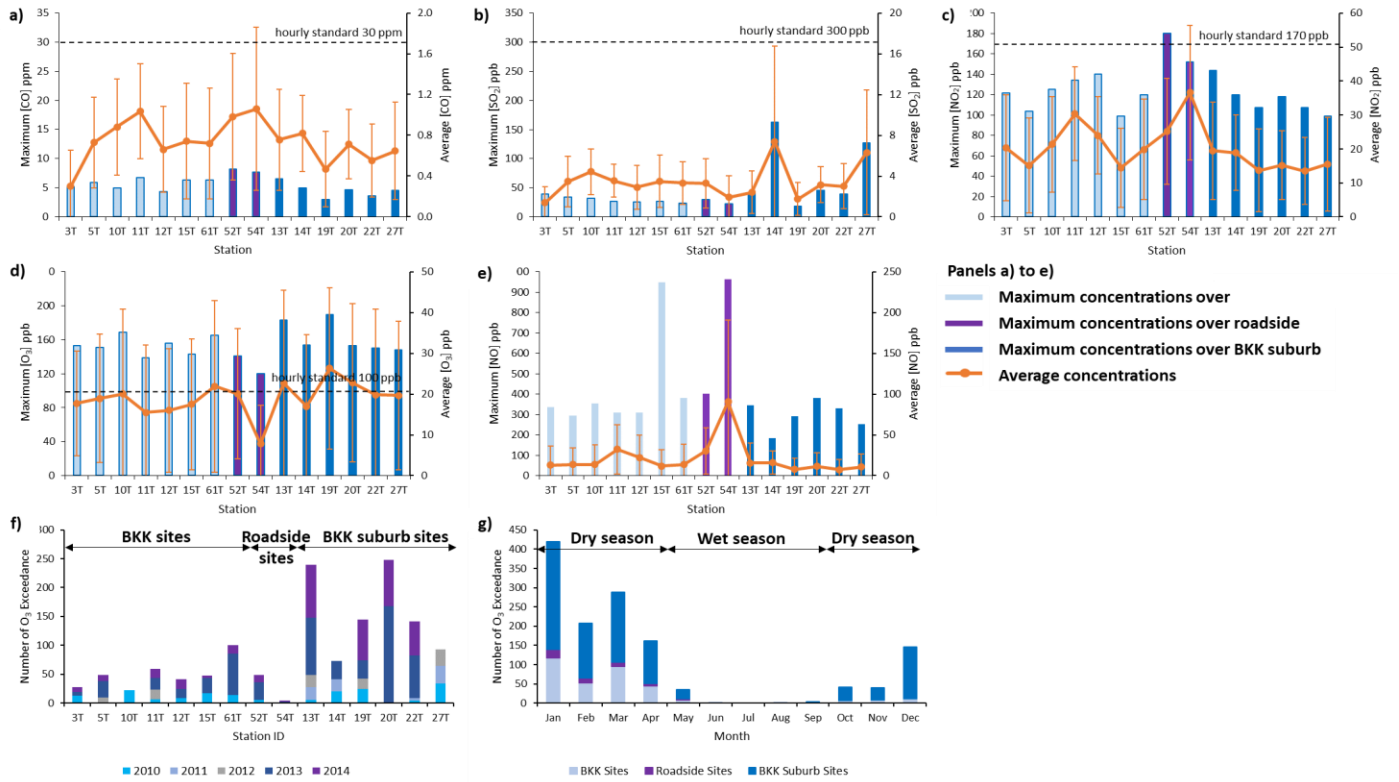
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534 **Figures:**



535
 536 **Fig. 1:** Map of BMR, the location of monitoring stations and two major monsoons winds (from NOAA HYSPLIT
 537 back trajectory model). Three monitoring station types, including BKK sites, roadside sites and BKK suburb sites are
 538 shown in light blue dots, purple dots and blue dots, respectively. (Note: * the station has been closed since 1 October
 539 2013).

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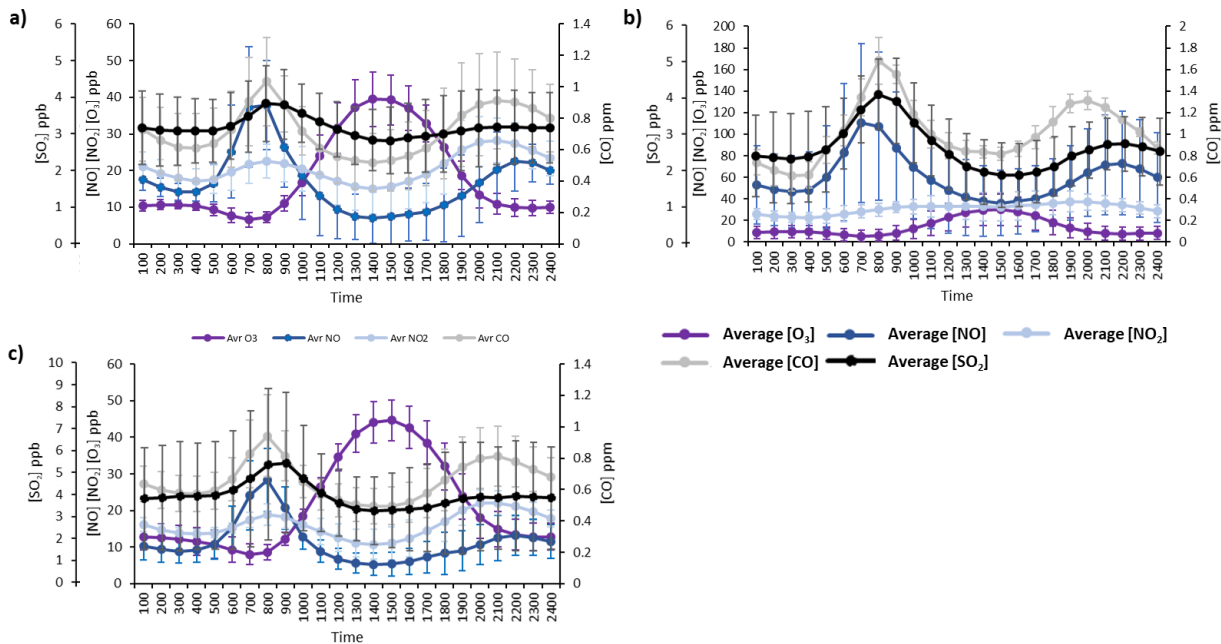


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547 **Fig 2:** Maximum (vertical bars) and average (solid line) concentrations of a) CO, b) SO₂, c) NO₂ d) O₃ and e) NO
 548 from the 15 monitoring stations, during 2010 to 2014, are compared with the hourly NAAQs (dotted line) of Thailand
 549 (except NO which is not a criteria pollutant). The number of hourly O₃ exceedances is shown by f) locations and g)
 550 seasons.

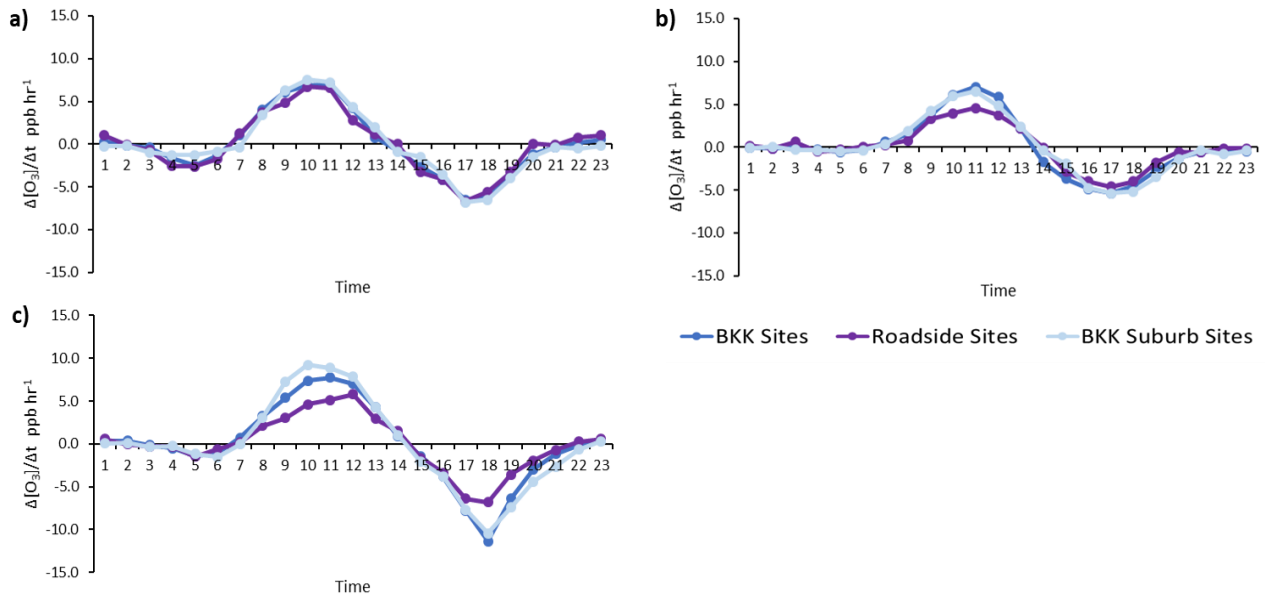
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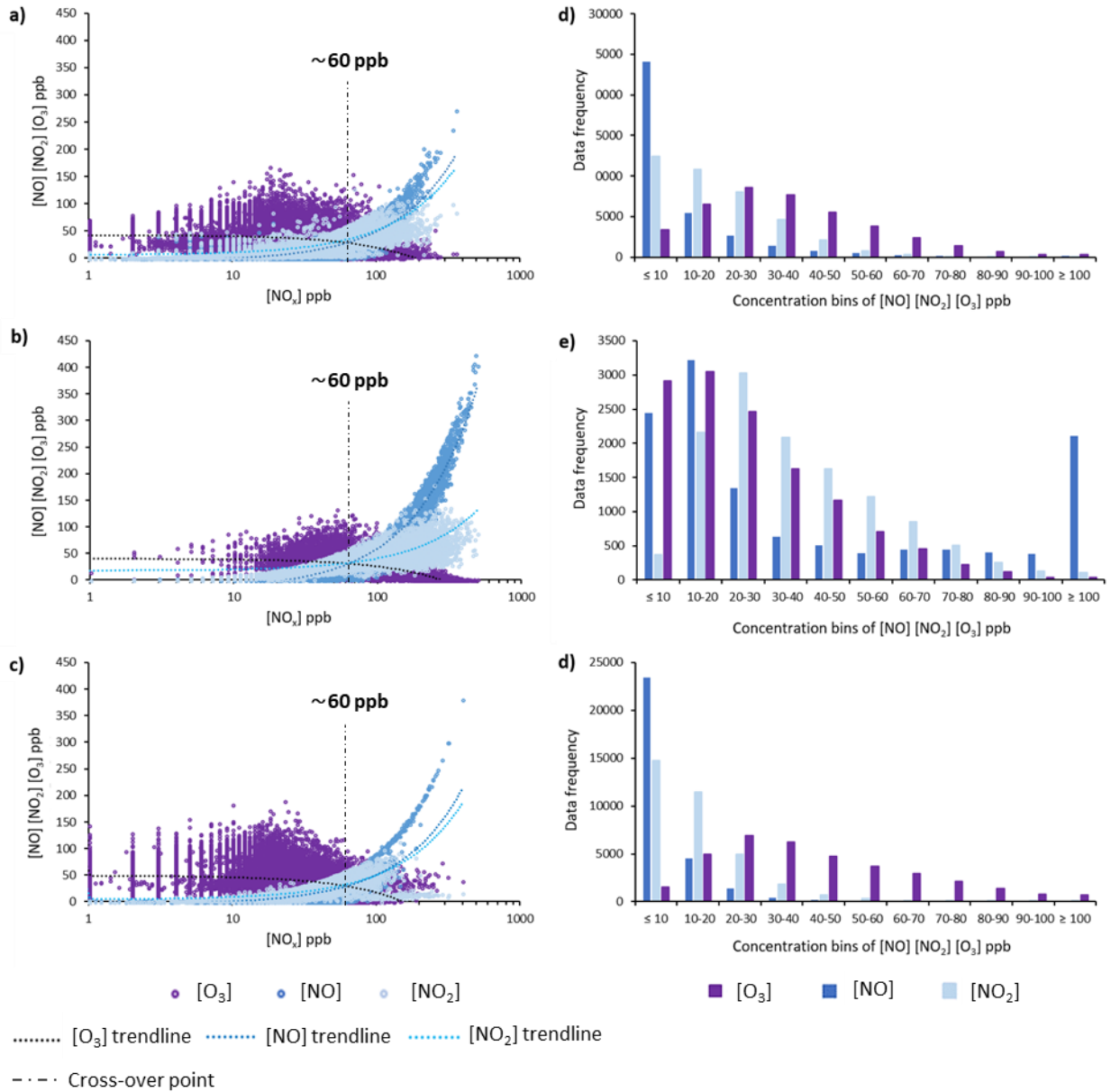
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 554 **Fig 3:** Diurnal variations of gaseous species. The plots provide the average concentrations of O₃, NO and NO₂ in ppb,
 555 the average concentrations of CO in ppm and the average concentrations of SO₂ in ppb at a) BKK site; b) roadside
 556 sites; and c) BKK suburb sites. Vertical bars provide ±1 standard deviations of the species concentrations.

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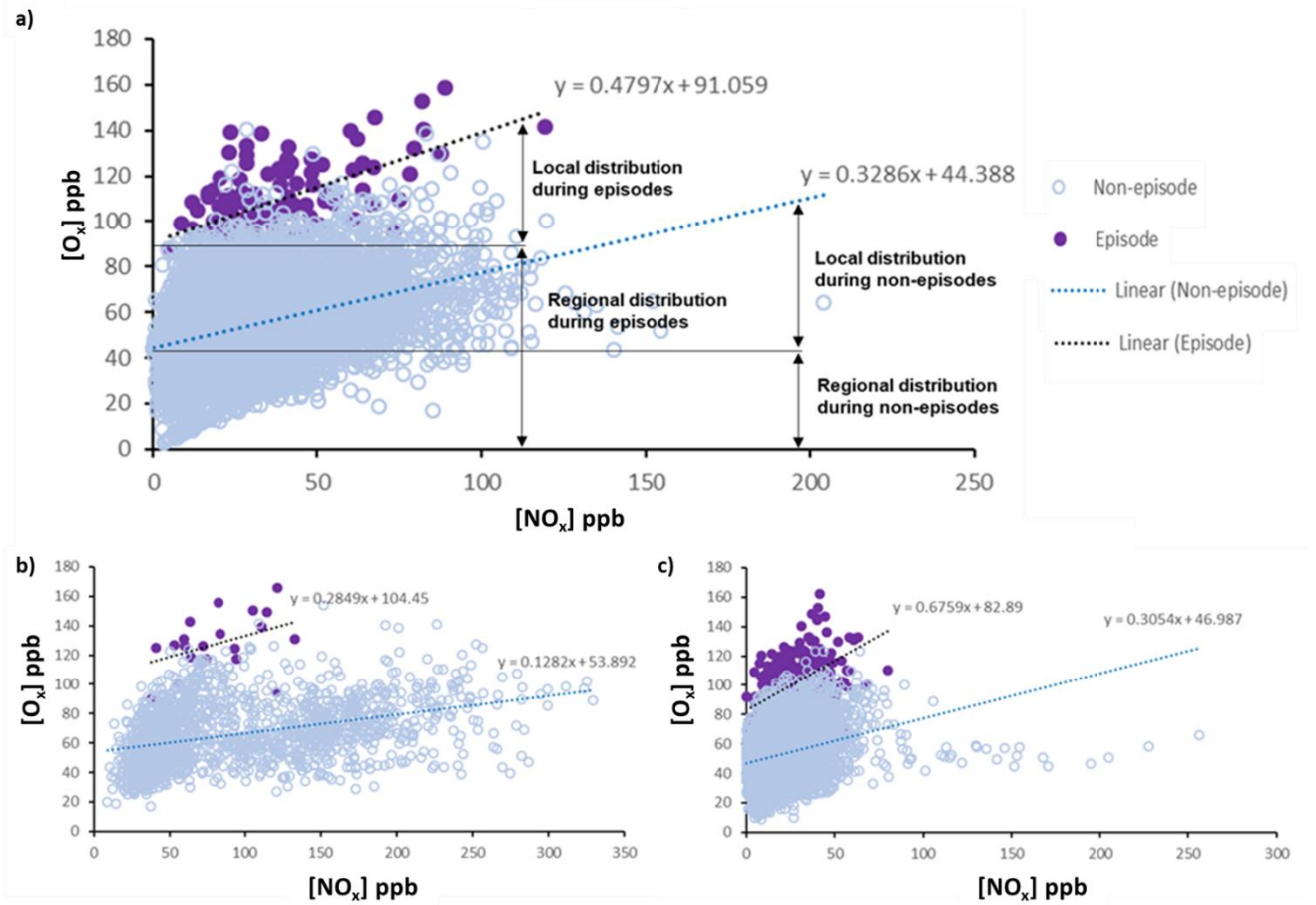
565
 566 **Fig. 4:** Diurnal variations of rate of change of O₃ concentration ($\Delta[O_3]/\Delta t$) during a) local summers b) wet seasons and
 567 c) local winters.

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 583 **Fig. 5:** Relationships and crossover points of NO, NO₂ and O₃ at a) BKK sites b) roadside sites and c) BKK suburb
 584 sites; and concentration distributions of those species at d) BKK sites e) roadside sites and f) BKK suburb sites.

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594 **Fig. 6:** Effects of local and regional contributions on O_x during non-episode and episode days at a) BKK sites, b)
 595 roadside sites and c) BKK suburb sites.

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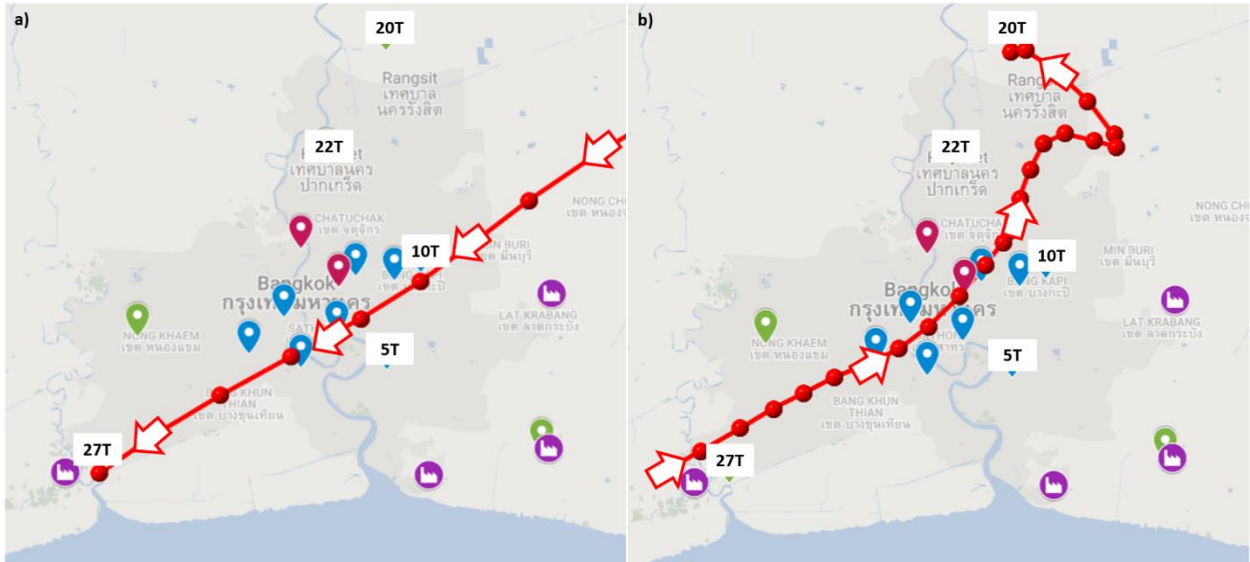
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605 **Fig. 7:** Backward trajectories from HYSPLIT model reveal a) NE wind direction (Jan 13, 2010) and b) SW wind
 606 direction (Jan 1, 2010)

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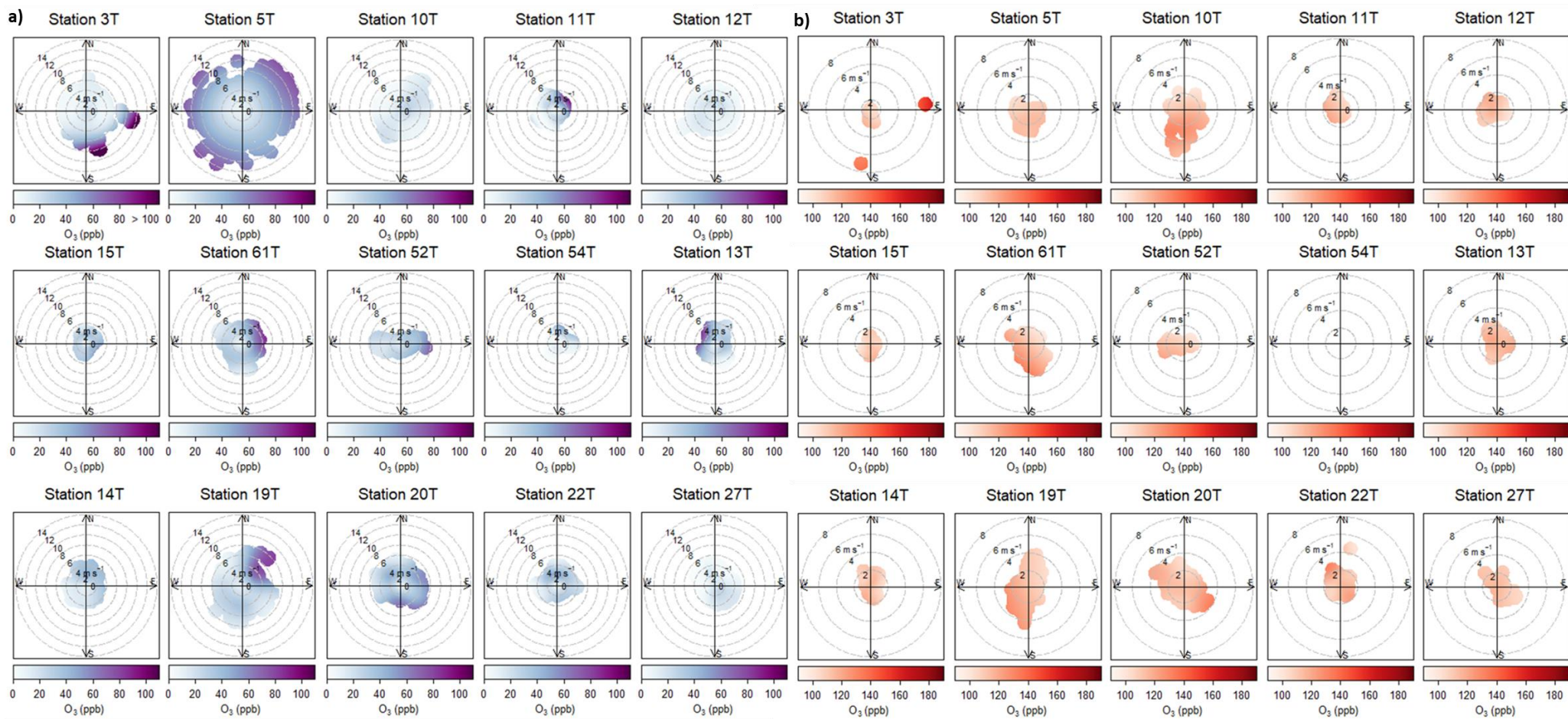
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627 **Fig. 8:** Relationship between the concentrations of O₃, wind speeds and wind directions during a) O₃ episodes ($[O_3]_{\text{hourly}} > 100$ ppb) and b) during non O₃
 628 episodes ($[O_3]_{\text{hourly}} \leq 100$ ppb), over BMR during 2010 to 2014.

629 **Tables:**

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

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

632 Note: *Clapp and Jenkin (2001), **Mazzeo et al. (2005), ***Tiwari et al. (2015)

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656 **Table 2:** The comparison of CO/NO_x and SO₂/NO_x ratios from this study with other studies (modify from Rasheed et
 657 al., 2014).

Region	Source	CO/NO _x	SO ₂ /NO _x
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		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***			> 0.3
Kolkata, and Durgapur, India***			≤ 0.13
Madrid City, Spain*		13.3	0.29
Rouen City, France**		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

658 * Fernandez-Jiménez et al., 2003

659 ** Coppalle et al., 2001

660 *** Mallik and Lal, 2014

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666 **Table 3:** Number of hours that were found in different AQI categories of O₃ over the BMR during 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	39018	32021	27959	40715	26606	33628	26442	32665	40231	31070	35429	33592	30793	34301	26873
Moderate	310	713	1023	556	367	479	1178	807	27	1620	944	1687	1340	1466	719
Unhealthy for Sensitive Group	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

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