

## ***Interactive comment on “Assessment of Air Pollution in Bangkok Metropolitan Region, Thailand” by Pornpan Uttamang et al.***

**Pornpan Uttamang et al.**

puttama@ncsu.edu

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General Comment: Pornpan Uttamang et al. have presented observations of CO, NO<sub>x</sub>, SO<sub>2</sub> and O<sub>3</sub> from 15 monitoring sites at understudied Bangkok Metropolitan Region (BMR) for a fiveyear- long period from 2010-2014. Background pertaining to the air-quality in terms of PM and O<sub>3</sub> exceedance events in the BMR is provided. However, the authors do not mention the knowledge gap or scientific question that they want to address from this study. I have major concerns with the paper which include description of analytical methods and discussion about quality control (calibration and sampling protocols, filter criteria) of dataset used. The statistical analysis is also weak which mostly covers average/maximum over the entire study periods, without going into details of specific seasons, inter-annual trends and pin-pointing the season-specific

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emission sources /formation processes and removal processes of the pollutants. The conclusions are drawn either from the regression lines having poor fit parameters or oversimplification of methods for source identification available in the peer-reviewed literature. The manuscript needs to address the major concerns (highlighted in specific comments) before it can be considered further. After performing the analysis suggested in the specific comments, corrections and restructuring the paper, the scientific outcome might be significantly different from the present version and should be considered as a new publication.

Authors' response: Thank you. The last paragraph of the Introduction (line 17 – 24) succinctly provides both the knowledge gap and scientific question being addressed. We have now made the statistical analysis more robust based on reviewer' suggestions.

Specific Comments: Title: Authors might consider making the title of the paper more specific. Authors assess CO, NO<sub>x</sub>, SO<sub>2</sub> and O<sub>3</sub> air pollution and not overall air pollution in general. Authors' response: Thank you. We have now modified the title to “Assessment of Gaseous Pollutants in Bangkok Metropolitan Region, Thailand”

Introduction: The authors have included a description of auto-mobile fleet and manufacturing industries in the introduction which should rather be a part of the site description. The introduction is poorly structured. Authors should include a brief literature review of the previous works from BMR, outlook from these studies and what are the knowledge gaps they want to address from this paper. The authors should also mention, why they have chosen to study CO, NO<sub>x</sub>, SO<sub>2</sub> and O<sub>3</sub>. At-least a line each about their importance regarding atmospheric chemistry and air quality should be present. The authors have referred to Zhang and Oanh, [2002] for the site description. However, the findings there should also be mentioned in the introduction, as Zhang and Oanh, [2002] have analyzed monthly and diel variation, O<sub>3</sub> exceedances, drivers for high ozone episodes and relationship of ozone production with NO<sub>x</sub>/NMHC ratio. These are quite relevant for the present study. Similarly, the work of Pocha-

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nant et al., [2001] should be highlighted in the introduction. I found few other studies (mentioned below) which are relevant to the present work and should be highlighted in the introduction. There might be several more! Jinsart, W., Tamura, K., Loetkamonwit, S., Thepanondh, S., Karita, K., and Yano, E.: Roadside Particulate Air Pollution in Bangkok, *Journal of the Air & Waste Management Association*, 52, 1102-1110, 10.1080/10473289.2002.10470845, 2002. Suthawaree, J., Tajima, Y., Khunchornyakong, A., Kato, S., Sharp, A., and Kajii, Y.: Identification of volatile organic compounds in suburban Bangkok, Thailand and their potential for ozone formation, *Atmospheric Research*, 104-105, 245-254, 10.1016/j.atmosres.2011.10.019, 2012

Authors' response: Thank you. We have now modified the Introduction to include the discussion in the references provided by the reviewer.

"1. Introduction Over the last three decades, Thailand's rapid industrialization and urbanization has led to an increase in global economic prowess (World Bank, 2018a). A majority of the country's development has occurred within and around Bangkok (BKK) (13°45' N and 100°85' E), the capital city of Thailand and Bangkok Metropolitan Region (BMR). The BKK is comprised of the five adjacent provinces of BKK (World Bank, 2018a and 2018b). The increase in emissions is due to accelerated growth combined with high photochemical activity, strong solar insolation, high temperatures and high humidity (Kumar et al., 2012). BMR, with these conditions, has begun to experience air quality degradation, in particular, enhanced secondary pollutants. Since 1995, BKK has experienced exceedances in Thailand NAAQs for particulate matter (PM) and ozone (O<sub>3</sub>) (PCD, 2015). The correlation between BMR air pollution and public health has been observed in several published studies. Ruchirawat et al. (2007) reported the children who lived in BKK are exposed to high levels of carcinogenic air pollutants which may cause an elevated cancer risk. Buadong et al. (2009) reported the exposure to elevated PM and O<sub>3</sub> during the previous day, in elderly patients ( $\geq 65$  years), is associated with increasing the number of daily hospital visits for cardiovascular diseases. Jinsart et al. (2002, 2012) reported policemen and drivers in BKK tended to expose higher

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level of PM concentrations compared with the general environment, in which the concentrations of PM were already high. Role of atmospheric processes in elevated O<sub>3</sub> in Thailand were reported in several studies. Long-range transport played an important role in increasing O<sub>3</sub> concentration in Thailand. Generally, long-range transports from the Asia continental contained higher O<sub>3</sub> concentrations compared with long-range transports from the Indian ocean (Pochanart et al., 2001). In BMR, local emission and regional transport were the major contributors to high O<sub>3</sub> concentrations and seasonal fluctuations of O<sub>3</sub>, respectively (Zhang and Oahn, 2002). In suburban areas of BKK, volatile organic compounds (VOCs) tended to be a potential factor to enhance O<sub>3</sub> concentration (Suthawaree et al., 2012). The availability and analysis of multi-year measurements of such gaseous pollutants in the BMR will improve our understanding of how they contribute to the air quality of this area. 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 formation. The emissions from industrial activities also dominates the BMR metropolitan area and contributes to the emissions of sulfur dioxide (SO<sub>2</sub>) and the formation of particulate matter. In this study, diurnal variations, seasonal variations and inter-annual 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> during 2010 to 2014, in BMR, have been analysed. Chemical and physical processes associated with high O<sub>3</sub> concentrations have been investigated. Since the concentrations of nitrogen oxide (NO<sub>x</sub>) was measured at most of the monitoring station, therefore, O<sub>3</sub> precursors in this study is referred to NO<sub>x</sub>. The photochemical reaction was investigated during the photostationary state. The effects of local emission and regional contributions of Ox are presented. The severity of air pollution concentrations in BMR on human health are assessed by performing Air Quality Index (AQI)."

Page 2, Line 23: Authors state "possible emission sources of pollutants that associate with O<sub>3</sub> formation are identified". However, such identification is not discussed in the manuscript. Authors have only used the ratio of CO/NO<sub>x</sub> and SO<sub>2</sub>/NO<sub>x</sub> to identify whether the emission sources are mobile or point in nature. The method itself has an

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inherent limitation which is mentioned later in the specific comment for the section.

Authors' response: Thank you. We have now modified the Introduction. Please refer to our discussion in the comment "Section 3.5 Page 11, Line 16" below.

Methodology: The exact measurement period should be mentioned in this section. This paper discusses a five-year-long measurement period and shows data over 15 different measurement stations and authors should provide a time-line for data availability for each station. Authors' response: Thank you. We have now provided the measurement period in the modified manuscript and provided a time-line for data availability for each station as part of the supplement material.

Page 3, line 22: What is the basis of the assumption that monitoring sites used were representative of BMR specific patterns and trends?

Authors' response: Thank you. Based on the reviewer, we have modified the manuscript by removing the sentence.

Data Collection and Data Analysis: I have major concerns with this section. Authors did not provide any sampling details. The trace gas analysers for CO, NO<sub>x</sub>, SO<sub>2</sub> and O<sub>3</sub> are known to have drifts with time. Authors mention that equipment and monitoring stations are calibrated every year. This is not enough. There should be frequent zero drift check for CO (at-least daily) and for NO<sub>x</sub>, SO<sub>2</sub> and O<sub>3</sub> (at-least once a week). The linearity of the detection should also be checked with calibration experiments performed at-least once a month. The authors did not provide any information about the drift in the sensitivity of instruments over the period of 5 years. Detection limits of the trace gas analysers and uncertainties of the measurements should also be provided.

Authors' response: Thank you. As indicated in the manuscript, the data were collected, and after QA/QC, were provided by the Pollution Control Department (PCD), Thailand. Data loggers are calibrated/ checked at least every 15 days. Air inlets are cleaned at least every 15 days. Equipment is single-point calibrated and multi-point calibrated at

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least every 15 days and at least every 3 months. Monitoring stations and equipment are audited by external auditors every year. We have modified our manuscript to make a clarification.

"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 auto calibration at the monitoring stations. Manual quality control was performed when unusual observations were found. External audit of the equipment and monitoring stations were done every year. "

Page 4, line 6: Authors mention that quality assurance and quality control on the dataset were performed by PCD prior to receiving the data. What are these quality controls? Authors' response: Thank you. QA/AC protocols are published in the PCD, Thailand government document.

Page 4, line 9: What are the manual quality controls? What are the criteria for choosing unusual observations? Authors' response: Thank you. QA/AC protocols are published in the PCD, Thailand government document. However, we did not provide any additional guidelines for data collection.

Result and Discussion Section 3.1: Authors have only provided maximum and average over the entire five-year period. Since they have continuous one hour time resolution dataset from 15 monitoring stations for a five year long period, authors should also include inter-annual variability and seasonal statistics at-least for different monitoring station types. Given the advantage of also having wind speed/ wind direction data, authors should consider comparing various air mass fetch regions for some monitoring sites. For ozone, it makes more sense to separate daytime and night-time before reporting the average concentrations. The authors discuss extensively about 1-hour exceedance of ozone concentrations, but there is no description of how are these exceedance events calculated. One cannot compare the hourly average concentrations directly with the NAAQS. What about the ozone exceedance from 8-h standard? Bangkok

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air quality standard provides criteria for both 1-hour and 8-hour average ozone. 8-h average is intended to provide a better protection from long term ozone exposure.

Authors' response: Thank you. We have now provided inter-annual plots and seasonal variation plots for gaseous criteria pollutants from the monitoring stations. In the plots, we have averaged the data for each monitoring type station (from BKK sites, roadside sites and BKK suburb sites). This figure is now included in the supplementary material. However, the discussion associated with this information is now included in the manuscript. Moreover, we have now included the average concentrations of O<sub>3</sub> during daytime (6:00 AM to 6:00 PM) and during nighttime (6:00 PM to 6:00 AM) in the modified manuscript.

"3.1 Status of Pollution in BMR during 2010 to 2014 Figure 2 a) to e) shows the maximum and average concentration of gaseous pollutants, during 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<sub>2</sub>, SO<sub>2</sub> and O<sub>3</sub> 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<sub>2</sub> and SO<sub>2</sub> were mostly in their hourly standards (an exceedance of NO<sub>2</sub> was found at 52T monitoring station, during 2013). However, the maximum concentrations of O<sub>3</sub> exceeded its standard. Elevated CO, NO, and NO<sub>2</sub> concentrations were frequently observed at roadside sites than other sites. The average concentrations of CO, NO, and NO<sub>2</sub>, 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<sub>2</sub> were commonly observed at BKK suburb sites than other sites. The average concentrations of SO<sub>2</sub> at BKK suburb sites were  $\sim 4.0 \pm 2.3$  ppb. The average concentrations of O<sub>3</sub> during daytime (6:00 to 18:00 LT) over BKK sites, roadside sites and BKK suburb sites were  $\sim 24.4 \pm 13.5$  ppb,  $\sim 18.2 \pm 12.3$  ppb and  $\sim 27.7 \pm 14.7$  ppb, and those values during night-time (18:00 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 average O<sub>3</sub> concentrations were highest at BKK sub-

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urb sites ( $\sim 22.0 \pm 19.8$  ppb) and following by BKK sites ( $17.9 \pm 16.9$  ppb) and roadside sites ( $13.3 \pm 12.7$  ppb). The maximum and average of gaseous pollutants the three monitoring types are provided in Table I, supplement material. The seasonal variations of the gaseous pollutants reveal that, in general, elevated concentrations were observed during dry season and those decreased during wet season (Figure II, supplement material). Inter-annual variations of the gaseous pollutants reveal that, while the concentrations of CO, NO<sub>2</sub> and SO<sub>2</sub> decreased or remained constant, the concentration of O<sub>3</sub> tended to increase during the study period (Figure III, supplement material). An O<sub>3</sub> exceedances was recorded when an hourly concentration of O<sub>3</sub> was greater than 100 ppb (hourly O<sub>3</sub> standard). Figure 2 f) to g) illustrate the number of hourly O<sub>3</sub> exceedances, which they are shown by locations and by seasons, respectively. The hourly O<sub>3</sub> exceedances at BKK suburb sites were more frequently observed than those at the other sites. The average number of hourly O<sub>3</sub> exceedances was  $\sim 16$  hours year<sup>-1</sup> at BKK sites,  $\sim 9$  hours year<sup>-1</sup> at roadside sites and  $\sim 43$  hours year<sup>-1</sup> at BKK suburb sites. The hourly O<sub>3</sub> exceedances were commonly observed during dry season than during the transitional period between the seasons (May) and rarely observed during wet season. "

With regards to segregating wind direction data we performed a more robust back trajectory analysis. Moreover, we provided wind-rose plots for each of the monitoring stations and discussed it in the manuscript.

The National Ambient Air Quality Standards of Thailand provides hourly and 8-hour average standards of O<sub>3</sub> (0.10 ppm and 0.07 ppm, respectively). In this study, we compared the hourly concentrations of O<sub>3</sub> with the hourly O<sub>3</sub> standard in order to examine number of O<sub>3</sub> exceedances. To study the effects of O<sub>3</sub> on human health, we applied Air Quality Index (AQI) of O<sub>3</sub> instead of using the O<sub>3</sub> exceedance from 8-hour standard, which we believe that, using AQI of O<sub>3</sub> will provide more advantages than using the O<sub>3</sub> exceedance from 8-hour standard. Since AQI of O<sub>3</sub> is categorized into six categories, with four of the six categories providing the information of the severity

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of high O<sub>3</sub> concentrations on human health, from sensitive groups to healthy people; therefore, applying AQI for O<sub>3</sub> will provide better information for air quality management.

Section 3.2 Diurnal Variation of the Gaseous Species: Regional meteorology has strong influence on primary emission processes, production of secondary pollutant e.g. ozone and ambient concentrations of pollutants. I would recommend season wise analysis of diel variation of gaseous species. For example, the authors can refer to the work of Gaur et al. [2014] and Kumar et al. [2016]. This would also enable to identify the periods when ozone production is maximum during the year. Authors should also analyse, how does rate of formation of ozone from sunrise until it attains the peak day-time values changes at different sites and in different seasons. Authors could refer to the work of Naja and Lal [2002]. Gaur, A., Tripathi, S. N., Kanawade, V. P., Tare, V., and Shukla, S. P.: Four-year measurements of trace gases (SO<sub>2</sub>, NO<sub>x</sub>, CO, and O<sub>3</sub>) at an urban location, Kanpur, in Northern India, *Journal of Atmospheric Chemistry*, 1-19, 10.1007/s10874-014-9295-8, 2014. Kumar, V., Sarkar, C., and Sinha, V.: Influence of post-harvest crop residue fires on surface ozone mixing ratios in the N.W. IGP analyzed using 2 years of continuous in situ trace gas measurements, *J. Geophys. Res.*, 121, 3619–3633 10.1002/2015JD024308, 2016. Naja, M., and Lal, S.: Surface ozone and precursor gases at Gadanki (13.5\_N, 79.2\_E), a tropical rural site in India, *Journal of Geophysical Research: Atmospheres*, 107, 10.1029/2001jd000357, 2002. Authors should provide an explanation for why a second peak is not observed in the diel profiles of SO<sub>2</sub> at all sites. In line 20 of page 7, authors speculate that SO<sub>2</sub> is emitted by automotive diesel engine exhaust. If we observe the diel profile of NO from the BKK sites, a bimodal profile is observed which is attributed to traffic emissions. Moreover, even if we assume that manufacturing facilities point sources are the SO<sub>2</sub> contributors as mentioned in line 23 of page 11, their emission strength would not vary over the time scale of a day and a bimodal profile driven by boundary layer meteorology should be observed. Similarly, authors should also provide an explanation for the relatively flatter diel profile of NO<sub>2</sub> at roadside sites.

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Authors' response: Thank you. We have now provided season wise analysis of diel variation of gaseous species for the three monitoring station types in the supplementary material and a discussion is provided in the manuscript.

"3.1 Status of Pollution in BMR during 2010 to 2014 Figure 2 a) to e) shows the maximum and average concentration of gaseous pollutants, during 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<sub>2</sub>, SO<sub>2</sub> and O<sub>3</sub> 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<sub>2</sub> and SO<sub>2</sub> were mostly in their hourly standards (an exceedance of NO<sub>2</sub> was found at 52T monitoring station, during 2013). However, the maximum concentrations of O<sub>3</sub> exceeded its standard. Elevated CO, NO, and NO<sub>2</sub> concentrations were frequently observed at roadside sites than other sites. The average concentrations of CO, NO, and NO<sub>2</sub>, 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<sub>2</sub> were commonly observed at BKK suburb sites than other sites. The average concentrations of SO<sub>2</sub> at BKK suburb sites were  $\sim 4.0 \pm 2.3$  ppb. The average concentrations of O<sub>3</sub> during daytime (6:00 to 18:00 LT) over BKK sites, roadside sites and BKK suburb sites were  $\sim 24.4 \pm 13.5$  ppb,  $\sim 18.2 \pm 12.3$  ppb and  $\sim 27.7 \pm 14.7$  ppb, and those values during night-time (18:00 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 average O<sub>3</sub> concentrations were highest at BKK suburb sites ( $\sim 22.0 \pm 19.8$  ppb) and following by BKK sites ( $17.9 \pm 16.9$  ppb) and roadside sites ( $13.3 \pm 12.7$  ppb). The maximum and average of gaseous pollutants the three monitoring types are provided in Table I, supplement material. The seasonal variations of the gaseous pollutants reveal that, in general, elevated concentrations were observed during dry season and those decreased during wet season (Figure II, supplement material). Inter-annual variations of the gaseous pollutants reveal that, while the concentrations of CO, NO<sub>2</sub> and SO<sub>2</sub> decreased or remained constant, the concentration of O<sub>3</sub> tended to increase during the study period (Figure III, supplement

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material). An O<sub>3</sub> exceedance was recorded when an hourly concentration of O<sub>3</sub> was greater than 100 ppb (hourly O<sub>3</sub> standard). Figure 2 f) to g) illustrate the number of hourly O<sub>3</sub> exceedances, which they are shown by locations and by seasons, respectively. The hourly O<sub>3</sub> exceedances at BKK suburb sites were more frequently observed than those at the other sites. The average number of hourly O<sub>3</sub> exceedances was ~16 hours year<sup>-1</sup> at BKK sites, ~9 hours year<sup>-1</sup> at roadside sites and ~43 hours year<sup>-1</sup> at BKK suburb sites. The hourly O<sub>3</sub> exceedances were commonly observed during dry season than during the transitional period between the seasons (May) and rarely observed during wet season. " 3.2 Diurnal Variation of the Gaseous Species Diurnal variations of gaseous pollutant are shown in Figure 3 a) to c). The diurnal variations of O<sub>3</sub> show a single-peak pattern (Aneja et al., 2001) with the concentrations increased after sunrise and reached the peak ~15:00 local time (LT). The concentrations begin to decline in the evening and reach the minimum concentrations ~7:00 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 of NO show a bimodal pattern with the concentrations reach the first- and the second-peak ~7:00 to 9:00 LT and ~21:00 to 22:00 LT, respectively. The concentrations of NO at the first- and the second-peak were ~40 ppb and ~23 ppb at BKK sites, ~110 ppb and ~73 ppb at roadside sites, and ~30 ppb and ~13 ppb at BKK suburb sites. The concentrations of NO<sub>2</sub> at the first- and the second-peak were ~23 ppb and ~28 ppb at BKK sites, ~33 ppb and ~37 ppb at roadside sites, and ~20 ppb and ~22 ppb at BKK suburb sites. Even the diurnal variations of 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 high concentration of NO<sub>x</sub> all day. The diurnal variations of CO show a bimodal pattern with the first- and the second-peak occurred ~8:00 LT 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 of NO, NO<sub>2</sub>, and CO correspond to morning rush hour in BKK (7:00

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to 9:00 LT). The second peak of those occurred ~3 to 5 hours after the evening traffic rush hour (16:00 to 18:00 LT) (Leong et al., 2002), due to a combination of pollutants emissions and collapse of the planetary boundary layer (weak turbulence and diffusion) during this time. The diurnal variations of SO<sub>2</sub> show a bimodal pattern with the first- and the second-peak of SO<sub>2</sub> occurred ~8:00 LT and 21:00 LT, respectively. The concentrations of SO<sub>2</sub> at the first- and the second-peak were ~3 ppb (both peaks) at BKK sites, ~3 ppb (both peaks) at roadside sites, and ~6 ppb ~3 ppb at BKK suburb sites. At the roadside sites, the peaks are more obvious than 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 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). The diesel fuel contains ~0.035 %wt Sulphur (DOEB, 2017). Season wise of the diurnal variations are provided in Figure IV, supplement material. Figure 4 a) to c) shows diurnal variations of rate of change of O<sub>3</sub> concentration ( $\Delta[\text{O}_3]/\text{dt}$ ) during dry season (local summer and local winter) and wet season at the three monitoring station types (the data has been averaged for each monitoring station type to capture the rate of change of O<sub>3</sub> concentration characteristics). The diurnal variations of  $\Delta[\text{O}_3]/\text{dt}$  is a combination of O<sub>3</sub> chemistry and meteorology. In general,  $\Delta[\text{O}_3]/\text{dt}$  during wet season were lower than those during dry season. However, during local winter, the rates of change O<sub>3</sub> concentration were the highest. 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 hr<sup>-1</sup> during wet season, 6.7 to 7.5 ppb hr<sup>-1</sup> during local summer, and 5.7 to 9.2 ppb hr<sup>-1</sup> during local winter. The  $\Delta[\text{O}_3]/\text{dt}$  became negative during 14:00 to 15:00 LT. As expected, the rate of change of O<sub>3</sub> concentration was nearly constant during nighttime. Rapid changes in the mixing height and solar insolation during morning increases  $\Delta[\text{O}_3]/\text{dt}$ . 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). " We have also provided the rate of change of O<sub>3</sub> concentration (Naja

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and Lal, 2002) during the three seasons and in the three monitoring station types with explanation in the modified manuscript.

"Figure 4 a) to c) shows diurnal variations of rate of change of O<sub>3</sub> concentration ( $\Delta[\text{O}_3]/\text{dt}$ ) during dry season (local summer and local winter) and wet season at the three monitoring station types (the data has been averaged for each monitoring station type to capture the rate of change of O<sub>3</sub> concentration characteristics). The diurnal variations of  $\Delta[\text{O}_3]/\text{dt}$  is a combination of O<sub>3</sub> chemistry and meteorology. In general,  $\Delta[\text{O}_3]/\text{dt}$  during wet season were lower than those during dry season. However, during local winter, the rates of change O<sub>3</sub> concentration were the highest. 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 hr<sup>-1</sup> during wet season, 6.7 to 7.5 ppb hr<sup>-1</sup> during local summer, and 5.7 to 9.2 ppb hr<sup>-1</sup> during local winter. The  $\Delta[\text{O}_3]/\text{dt}$  became negative during 14:00 to 15:00 LT. As expected, the rate of change of O<sub>3</sub> concentration was nearly constant during nighttime. Rapid changes in the mixing height and solar insolation during morning increases  $\Delta[\text{O}_3]/\text{dt}$ . 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)"

For the diurnal variations of SO<sub>2</sub>, in the manuscript, we explained that "...The concentrations of SO<sub>2</sub> increase again in the afternoon and reach a second-peak around 21:00 LT over roadside sites. Over BKK sites and BKK suburb sites, the concentrations of SO<sub>2</sub> are nearly constant after 19:00 LT..." which the second peak of SO<sub>2</sub> were observed over three monitoring station types, but the magnitude of the concentrations of SO<sub>2</sub> over BKK sites and BKK suburb sites were small. For the diurnal variation of NO<sub>2</sub>, at the roadside sites also showed a bimodal distribution, but flatter than those at other sites. However, we have now included the clarification and explanation in the manuscript.

"The diurnal variations of SO<sub>2</sub> show a bimodal pattern with the first- and the second-

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peak of SO<sub>2</sub> occurred ~8:00 LT and 21:00 LT, respectively. The concentrations of SO<sub>2</sub> at the first- and the second-peak were ~3 ppb (both peaks) at BKK sites, ~3 ppb (both peaks) at roadside sites, and ~6 ppb ~3 ppb at BKK suburb sites. At the roadside sites, the peaks are more obvious than 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 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). The diesel fuel contains ~0.035 %wt Sulphur (DOEB, 2017). Season wise of the diurnal variations are provided in Figure IV, supplement material. "

"The concentrations of NO<sub>2</sub> at the first- and the second-peak were ~23 ppb and ~28 ppb at BKK sites, ~33 ppb and ~37 ppb at roadside sites, and ~20 ppb and ~22 ppb at BKK suburb sites. Even the diurnal variations of 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 high concentration of NO<sub>x</sub> all day. "

Section 3.3 Interconversion between O<sub>3</sub>, NO and NO<sub>2</sub> and Photochemical Reaction: I have major concerns again with this section. In line 23, authors mention "the photostationary state (PSS) is applied through all chemical reactions for O<sub>3</sub> formation during 10:00-16:00 LT". However, later in the section they assume photostationary state only between O<sub>3</sub>, NO and NO<sub>2</sub>. In polluted environments, RO<sub>2</sub> and HO<sub>2</sub> also oxidize NO to NO<sub>2</sub> and hence disturb the PSS of NO, NO<sub>2</sub> and O<sub>3</sub> [Mannschreck et al., 2004]. Hence the  $j_1$  values calculated by only considering O<sub>3</sub>, NO and NO<sub>2</sub> in the PSS would not be accurate. Mannschreck, K., Gilge, S., Plass-Duelmer, C., Fricke, W., and Berresheim, H.: Assessment of the applicability of NO-NO<sub>2</sub>-O<sub>3</sub> photostationary state to long-term measurements at the Hohenpeissenberg GAW Station, Germany, Atmos. Chem. Phys., 4, 1265-1277, 10.5194/acp-4 1265-2004, 2004. Moreover,  $j_1$  values are strongly dependent on incoming solar radiation and mentioning an average over 10:00 L.T. until 16:00 L.T. will be oversimplification. In the moderately polluted environment,

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the photostationary state between O<sub>3</sub>, NO and NO<sub>2</sub> is achieved within 60 s to 300 s during daytime [Trebs et al., 2012]. Authors should perform a calculation of j<sub>1</sub> at similar timescales. Trebs, I., Mayol-Bracero, O. L., Pauliquevis, T., Kuhn, U., Sander, R., Ganzeveld, L., Meixner, F. X., Kesselmeier, J., Artaxo, P., and Andreae, M. O.: Impact of the Manaus urban plume on trace gas mixing ratios near the surface in the Amazon Basin: Implications for the NO-NO<sub>2</sub>-O<sub>3</sub> photostationary state and peroxy radical levels, *Journal of Geophysical Research: Atmospheres*, 117, 10.1029/2011JD016386, 2012. I cannot understand, why the authors emphasize the calculated k<sub>3</sub> values. It depends on a single parameter which is temperature! Do the authors want to show that their temperature measurements are reasonable or their calculation is accurate? Next, the authors are using O<sub>3</sub> measurements to estimate the j<sub>1</sub> values and again using j<sub>1</sub> to explain high O<sub>3</sub> concentration at some sites. This is cyclic. Polynomial trend lines are used to investigate the interconversion between O<sub>3</sub>, NO and NO<sub>2</sub>. However, as seen from Figure 4, The fit is very poor for O<sub>3</sub> in all the three cases. So inference drawn using these fits would not be conclusive.

Authors' response: Thank you. In our study, we evaluated the relationship between O<sub>3</sub> with the gaseous criteria pollutants for the NAAQs of Thailand. The assumption of the photostationary state (PSS) ( $\bar{I}_T = 1$ ), therefore, was applied through the chemical reactions of O<sub>3</sub> and NO<sub>x</sub> only.

Mannscheck et al., 2004, reviewed the PSS parameter ( $\bar{I}_T$ ) as:

Where j was the photolysis rate of NO<sub>2</sub>, and k was the rate of the chemical reaction of NO and O<sub>3</sub>. In the Mannscheck et al., 2004, when  $\bar{I}_T$  was equal to 1, then other chemical reactions converting NO to NO<sub>2</sub> and local emissions of either compound were negligible. However, these cases were rare and were limited to very polluted conditions. On the other hand, peroxy radicals (RO<sub>2</sub>) played an important role to contribute to additional NO and NO<sub>2</sub>, under clean or moderately polluted conditions. In the study of Mannscheck et al., (2004), the measurement was performed in a rural site, generally, the site was affected by relatively clean air masses (yearly average of NO<sub>x</sub> was

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below 3.5 ppb). The site was surrounded by forests (70%, mostly coniferous) and agricultural pastures (30%). The distance to the nearest urban and major industrial areas was about 80 km. Furthermore, the study mentioned that "for high NO<sub>x</sub> concentrations the levels of peroxy radicals should approach zero, since the sink for RO<sub>2</sub> increases with increasing NO and since OH as a precursor for RO<sub>2</sub> as well as RO species are removed via reaction with NO<sub>2</sub>". Therefore, we believe that the assumption of PSS holds for our study region (e.g. average of hourly concentration of NO<sub>x</sub> at BKK sites, roadside sites, and BKK suburb sites were ~30 ppb, ~88 ppb and ~21 ppb, respectively. These NO<sub>x</sub> values are far in excess of rural/semi-rural values in the study of Mannscheck et al., (2004)). With regards to the calculation of j<sub>1</sub>, it is strongly dependent on incoming solar radiation and on other variables (i.e. the following equation):

$$j_1(q,p) = \int_0^\infty \bar{I}_T \bar{I}_0 \bar{a}(p,\lambda) b_-(a,g,q,\lambda,T) Y_-(q,p,\lambda,T) d\lambda$$

Where  $\bar{I}_T$  = Actinic flux  $b_-(a,g,q,\lambda,T)$  = Average absorption cross section  $Y_-(q,p,\lambda,T)$  = Average quantum yield

However, these variables were not measured in our study at the monitoring stations. With regards to the calculation of k<sub>3</sub>, our intention is not to emphasize the k<sub>3</sub> calculation to show that our temperature measurements were reasonable, but rather to calculate j<sub>1</sub>. However, we have now modified our manuscript and removed using j<sub>1</sub> values to explain O<sub>3</sub> concentration. With regards to the polynomial trend lines, we have now modified the plots by including histogram of the concentrations of O<sub>3</sub>, NO and NO<sub>2</sub> to present data distribution of these species. Generally, most of the records are in low to middle concentration bins. (Fig. 5)

Section 3.4: What are the criteria for differentiation between episodes and non-episodes? For the linear regression presented in this section, one can observe significant scatter around the fitted line. In some cases, (for example roadside sites, non-episode), one can clearly observe two different regions in the plots and a single linear fit over entire dataset cannot be justified. For the delta O<sub>3</sub> analysis, how were the back

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trajectories calculated? How many trajectories per day and how many days backward trajectories at what height were calculated? Authors should also provide the number of days/hours when N-NE and S-SE wind directions respectively were observed. How was the agreement between local wind directions and the wind directions derived from NOAA HYSPLIT model? Given the large scatter around average of 10 ppb delta O<sub>3</sub>, the conclusion of local production is rather weak for days with O<sub>3</sub> concentrations > 80 ppb. The sentence structuring is poor and was difficult to follow. This also needs improvement. The conclusion regarding crossover points is drawn from polynomial regressions which have very poor fit parameters (and not even mentioned in the paper). The high NO<sub>x</sub> and low NO<sub>x</sub> regime should be calculated based on the ratio of NO<sub>x</sub> OH reactivity and VOC OH reactivity or using model calculated indicators (e.g. CH<sub>2</sub>O/NO<sub>y</sub>, H<sub>2</sub>O<sub>2</sub>/HNO<sub>3</sub> and O<sub>3</sub>/(NO<sub>y</sub>-NO<sub>x</sub>)) as described by Kumar et al., [2011]. Classification based on cross over points are an oversimplification of the polynomial fits. Kumar, R., Naja, M., Pfister, G. G., Barth, M. C., Wiedinmyer, C., and Brasseur, G. P.: Simulations over South Asia using the Weather Research and Forecasting model with Chemistry (WRF-Chem): chemistry evaluation and initial results, *Geosci. Model Dev.*, 5, 619-648, 10.5194/gmd-5-619-2012, 2012.

Authors' response: Thank you. We had explained that an O<sub>3</sub> episode was identified when hourly O<sub>3</sub> concentrations were greater than 100 ppb (the O<sub>3</sub> NAAQs for Thailand).

With regards to the linear regression, we presented "the estimation" of local and regional contributions of Ox. Furthermore, we also compared the result from our study to results from other studies (using similar linear regression method).

For the two different observed NO<sub>x</sub> regions at roadside sites, we provided in our discussion the following "It is noteworthy that the pattern of the local and regional contributions at roadside sites during non-episode period 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-episode over BKK suburb sites. The high NO<sub>x</sub> regime (NO<sub>x</sub>

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> 60 ppb) may represent typical characteristic of air quality near roads".

To estimate the local and regional contribution by plotting Ox against NO<sub>x</sub> were reported in several published studies, for example Clapp and Jenkin (2001), Aneja et al., (2001), Mazzeo et al., (2005), Tang et al., (2009), Notario et al., (2012), Rasheed et al., (2014), Tiwari et al., (2015). These studies provide similar plots to our study. All these references are cited in the manuscript.

Clapp, L. J. and Jenkin, M. E.: Analysis of the relationship between ambient levels of O<sub>3</sub>, NO<sub>2</sub> and NO as a function of NO<sub>x</sub> in the UK, *Atmospheric Environment*, 35(36), 6391- 6405, doi:10.1016/S1352-2310(01)00378-8, 2001. Aneja, V. P., Agarwal, A., Roelle, P. A., Phillips, S. B., Tong, Q., Watkins, N., and Yablonsky, R.: Measurements and Analysis of Criteria Pollutants in New Delhi, India, *Environment International*, 27, 35-42, doi:10.1016/S0160-4120(01)00051-4, 2001. Mazzeo, N. A., Venegasa, L. E. and Chorenco, H.: Analysis of NO, NO<sub>2</sub>, O<sub>3</sub> and NO<sub>x</sub> concentrations measured at a green area of Buenos Aires City during wintertime, *Atmospheric Environment*, 39, 3055-3068, doi:10.1016/j.atmosenv.2005.01.029, 2005. Tang, G., Li, X., Wang, Y., Xin, J., and Ren, X.: Surface ozone trend details and interpretations in Beijing, 2001-2006, *Atmos. Chem. Phys.*, 9, 8813-8823, 2009. Notario, A., Bravo, I., Adame, J. A., Díaz-de-Mera, Y., Aranda, A., Rodríguez, A., and Rodríguez, D.: Analysis of NO, NO<sub>2</sub>, NO<sub>x</sub>, O<sub>3</sub> and oxidant (Ox = O<sub>3</sub>+NO<sub>2</sub>) levels measured in a metropolitan area in the southwest Iberian Peninsula, *Atmospheric Research*, 104-105, 217-226, doi:10.1016/j.atmosres.2011.10.008, 2012. Rasheed, A., Aneja, V. P., Aiyer, A., and Rafique, U.: Measurements and analysis of air quality in Islamabad, Pakistan, *Earth's Future*, 2, 303-314, doi:10.1002/2013EF000174, 2014. Tiwari, S., Dahiya, A., and Kumar, N.: Investigation into relationships among NO, NO<sub>2</sub>, NO<sub>x</sub>, O<sub>3</sub>, and CO at an urban background site in Delhi, India, *Atmospheric Research*, 157, 119-126, doi:10.1016/j.atmosres.2015.01.008, 2015. With regards to the delta O<sub>3</sub> analysis, back trajectory was determined when hourly concentration of O<sub>3</sub> > 80 ppb was observed either at 27T or 20T sites. By performing the backward trajectories using the NOAA

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HYSPLIT model, we identified the wind directions that related with high O<sub>3</sub> concentrations at both the monitoring stations. We calculated delta O<sub>3</sub> when air masses were observed from NE to SW or vice versa direction (about 200 records during the study period). In general, it should be noted that there is good agreement between the local station wind direction measurement (wind-roses analysis) and the back-trajectory analysis.

With regards to the low and high NO<sub>x</sub> regimes in our manuscript, the low and high NO<sub>x</sub> regimes refer to the concentrations of NO<sub>x</sub> that are either lower or higher than the cross over point i.e. [NO<sub>x</sub>] = 60 ppb (previous studies (Clapp and Jenkin, 2001; Notario et al., 2012; Tiwari et al., 2015) have also suggested similar NO<sub>x</sub> regimes). Furthermore, we do not have CH<sub>2</sub>O, NO<sub>y</sub>, H<sub>2</sub>O<sub>2</sub> and HNO<sub>3</sub> observations. Therefore, our analysis was limited only to NO<sub>x</sub> species. The paper mentioned by the reviewer (Kumar et al., 2012) reported the ratio of CH<sub>2</sub>O/NO<sub>y</sub>, H<sub>2</sub>O<sub>2</sub>/HNO<sub>3</sub> and O<sub>3</sub>/(NO<sub>y</sub>-NO<sub>x</sub>) based on the modeling analysis performed by WRF-Chem model. This modeling analysis is not within the scope of our study. Clapp, L. J. and Jenkin, M. E.: Analysis of the relationship between ambient levels of O<sub>3</sub>, NO<sub>2</sub> and NO as a function of NO<sub>x</sub> in the UK, *Atmospheric Environment*, 35(36), 6391- 6405, doi:10.1016/S1352-2310(01)00378-8, 2001. Notario, A., Bravo, I., Adame, J. A., Díaz-de-Mera, Y., Aranda, A., Rodríguez, A., and Rodríguez, D.: Analysis of NO, NO<sub>2</sub>, NO<sub>x</sub>, O<sub>3</sub> and oxidant (O<sub>x</sub> = O<sub>3</sub>+NO<sub>2</sub>) levels measured in a metropolitan area in the southwest Iberian Peninsula, *Atmospheric Research*, 104-105, 217-226, doi:10.1016/j.atmosres.2011.10.008, 2012. Tiwari, S., Dahiya, A., and Kumar, N.: Investigation into relationships among NO, NO<sub>2</sub>, NO<sub>x</sub>, O<sub>3</sub>, and CO at an urban background site in Delhi, India, *Atmospheric Research*, 157, 119-126, doi:10.1016/j.atmosres.2015.01.008, 2015.

Section 3.5 Page 11, Line 16: A good correlation implies good correlation coefficient (r) for a linear regression and not necessarily a large value of slope. Authors' logic of having a high CO/NO<sub>x</sub> ratio (slope of fit) because of a better correlation between the two species emitted from point sources is difficult to follow. The authors state

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that high CO/NO<sub>x</sub> and low SO<sub>2</sub>/NO<sub>x</sub> ratio is characteristic of mobile sources. What are the values they referring to? Is there a threshold? What is the correlation coefficient of the liner regression between CO and NO<sub>x</sub>? Such correlation plots should at-least be provided in supplement. Since the authors have a great advantage of having the data from multiple receptor locations, they should use some statistical source apportionment models (for example, Positive Matrix Factorization (PMF) or the work by Garg and Sinha [2017]) Garg, S., and Sinha, B.: Determining the contribution of long-range transport, regional and local source areas, to PM<sub>10</sub> mass loading in Hessen, Germany using a novel multi-receptor based statistical approach, *Atmospheric Environment*, 167, 566-575, 10.1016/j.atmosenv.2017.08.029, 2017. The authors have referred to the work of Parrish et al. [1991] for local source identification using CO/NO<sub>x</sub> ratio. However, longer-lived NO<sub>y</sub> should be used in place of NO<sub>x</sub>. This method can be used for estimating the background concentration of a short-lived species by performing a lognormal regression with a long-lived species. Simply using the ratio of CO and NO<sub>x</sub> to conclude the dominance of mobile source over point sources or vice versa by performing a linear regression over entire dataset of a group of specific monitoring station type will be a wrong over-interpretation of these ratios. This is also evident from the SO<sub>2</sub>/NO<sub>x</sub> ratios reported in Table 3. The SO<sub>2</sub>/NO<sub>x</sub> values are very similar for all the types of sites and even higher for roadside sites as compared for suburban and BKK sites. Based on authors assertion, it should be minimum for roadside sites among the three categories.

Authors' response: Thank you. The threshold or value to classify the difference between emissions from mobile sources and point sources has not been quantified definitively, however, the relative magnitude of the ratios provides an insight on source characteristics. We have compared our results with other published studies from different urban locations in US., Europe, and Asia before making our conclusion. It should also be noted that Positive Matrix Factorization (PMF) or the work by Garg and Sinha [2017] is in general applied to particulate matter. However, we have now provided correlation plots in the supplementary material.

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With regards to the ration analysis using NO<sub>y</sub> species, the NO<sub>y</sub> data was not corrected as part of this study.

Section 3.5.2: Why are wind rose plotted for separate wind directions? It is very confusing. Authors should show a wind rose showing the fraction of wind coming from all the directions for O<sub>3</sub> concentrations higher than 100 ppb. Higher fraction of wind from a particular direction would automatically point out major contribution from a particular wind direction. A polar plot with wind speed as radius axis, wind direction as angle and markers coloured according to observed O<sub>3</sub> concentration could also be an alternative plot. How does the wind rose look like for periods with O<sub>3</sub> concentration less than 100 ppb? If it is different from the ones for higher concentration, this would make the conclusion of higher ozone production from a particular wind direction stronger.

Authors' response: Thank you. The wind rose plots were created from wind speeds and wind directions (blowing from), during O<sub>3</sub> episodes ([O<sub>3</sub>]hourly > 100 ppb). The wind rose plots were analyzed and, then, classified the into 3 groups, according to the predominant local wind directions, including northerly, westerly and southerly winds. These wind roses were not plotted based on wind directions alone. However, we have now provided new plots of wind speed, wind direction (blowing from) versus the concentrations of O<sub>3</sub>, during O<sub>3</sub> episodes and non-episodes, in the modified manuscript. Generally, high O<sub>3</sub> concentrations relate with low wind speed (lower than 4 ms<sup>-1</sup>) and relate with the predominant wind directions, especially, at 3T, 10T, 19T, 20T, 22T, 52T and 61T monitoring stations. Elevated O<sub>3</sub> concentrations associated with northerly winds were at 22T monitoring station. At 3T, 10T, 19T, 20T and 61T monitoring stations, high concentrations of O<sub>3</sub> associated with southerly winds. At 52T monitoring station, high concentrations of O<sub>3</sub> associated with westerly winds. Moreover, the limited back trajectory analysis (based on NOAA HYSPLIT model) corroborates these findings and are now discussed in the manuscript.

"3.5.2 Effects of Pollutant Transport In general, O<sub>3</sub> has a short lifetime in polluted urban atmosphere (approximately hours). However, O<sub>3</sub> has a longer lifetime of several

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weeks in the free troposphere. This occurrence may allow O<sub>3</sub> to be transported over continental scales (Stevenson et al., 2006; Young et al., 2013; Monks et al., 2015). Figure 8 shows O<sub>3</sub> concentrations, during episodes and non-episodes, with predominant wind directions and wind speeds. The results show that O<sub>3</sub> exceedances are associated with low wind speed and predominant wind directions. In general, elevated O<sub>3</sub> concentrations were observed with wind speed lower than 4 ms<sup>-1</sup> with northerly winds (22T station), southerly winds (3T, 10T, 19T, 20T and 61T stations) and westerly winds (52T station). It is noteworthy that the southerly winds, generally, bring cleaner marine air mass to the land. However, under a stagnant condition (low wind speed), elevated O<sub>3</sub> concentrations were observed (Sahu et al., 2013a, 2013b)."

Section 3.6: Authors need to describe the calculation of air quality index. If they have used the simple hourly average O<sub>3</sub> concentration for calculate of AQI, then it is wrong. For calculation of hourly air quality index, O<sub>3</sub> concentration for a give hour should be taken as the average for the previous 4 hours, current hour and next 3 hours. However, it is recommended to consider 8-hour AQI as mentioned previously in the review.

Authors' response: Thank you. To calculate AQI for O<sub>3</sub>, we calculate midpoints of 8-hour average of O<sub>3</sub> concentration from the average of hourly O<sub>3</sub> concentration of the previous four hours, at the given hour and the following three hours (this analysis is similar to the reviewer's suggestion). To get a valid calculation, at least 6 of 8 records (75%), are needed. Then we compared the calculated midpoints with the AQI table. However, we have now included this information in the supplement material.

US.EPA (2017), Air Quality Index (AQI) Basics, Available from: <https://airnow.gov/index.cfm?action=aqibasics.aqi>, (Accessed April 2017). US.EPA (2017), Daily and Hourly AQI – Ozone, Available from: <https://forum.airnowtech.org/t/daily-and-hourly-aqi-ozone/170>, (Accessed April 2017).

Technical comments: Page 1 Line 27 – page 2 line 2; Page 2, lines 11-16: These are better suited for site description.

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Authors' response: Thank you. We have moved the information to "Section 1.2 Study Area".

"2.1 Study Area Figure 1 shows a map of BMR, the location of the monitoring stations in this study and major monsoon winds over the region. BMR refers to BKK and the five adjacent provinces, including Nakhon Pathom, Pathum Thani, Nonthaburi, Samut Prakan, and Samut Sakhon. These provinces are closely 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 (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 high 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 north-eastern 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 °C) to extremely hot weather (> 40 °C) 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). For example, 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. The remaining 16 % were Compressed Natural Gas (CNG) (DLT, 2017). In fact, the outskirts of BKK are populated with a variety of metal, auto parts, paper, plastic, food and chemical manufacturing facilities and power plants (DIW, 2016, 2016a, 2016b, 2016c, 2016d). "

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Page 2, line 17: NO is Nitric oxide and not nitrogen oxide. Nitrogen oxides refer to the family of oxides of nitrogen.

Authors' response: Thank you. This typographical error is now corrected.

Page 2, Line 20: What is the basis of the statement "Moreover, BMR experiences primarily O3 exceedances amongst all the other gaseous criteria pollutants."

Authors' response: Thank you. The current study provides the basis for this statement.

Page 4, Line 4: Figure 1 should be mentioned earlier in the section.

Authors' response: Thank you. We have now mentioned the figure earlier in the section.

Page 4, Line 20: What are the "equivalent instruments"?

Authors' response: Thank you. The US EPA provides on its website ([https://archive.epa.gov/emap/archive-emap/web/html/qa\\_terms.html](https://archive.epa.gov/emap/archive-emap/web/html/qa_terms.html)) clarity to equivalent method. Often it may also be referred to as Alternate method which is any body of procedures and techniques of sample collection and/or analysis for a characteristic of interest which is not a reference or approved equivalent method but which has been demonstrated in specific cases to produce results comparable to those obtained from a reference method.

Page 5, Line 3: Is the measurement period 2012-2014 or 2010-2014?

Authors' response: Thank you. This was a typographical error, which has now corrected.

Page 5, Line 7: What is the hourly "standard"?

Authors' response: Thank you. The National Ambient Air Quality Standards of Thailand provides hourly and 8-hour average standards of CO (30 ppm and 9 ppm, respectively), hourly and annually average standards of NO<sub>2</sub> (0.17 ppm and 0.03 ppm, respectively),

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hourly, 24-hour and annually average standards of SO<sub>2</sub> (0.3 ppm, 0.12 ppm and 0.04 ppm, respectively), and hourly and 8-hour average standards of O<sub>3</sub> (0.10 ppm and 0.07 ppm, respectively).

Page 6, line 5: Authors mention “VOCs concentrations were measured periodically only at one monitoring station limiting its usefulness as part of this study”. However, Zhang et al. [2002] have reported CH<sub>4</sub> and NMVOC data from 10 out of 13 monitoring stations from BMR. Did the stations stopped monitoring CH<sub>4</sub> and NMVOCs?

Authors’ response: Thank you. We provided the limitation in the manuscript “While NO<sub>x</sub> was measured continuously at all the monitoring site, VOCs were measured periodically only at one monitoring station limiting its usefulness as part of this study”.

Page 6, Line 12 and Figure 3: What is the explanation for a rather flat diel profile of NO<sub>2</sub> at roadside sites. Roadside sites are influenced maximum by traffic emissions, and one would expect a bimodal shape of diel profile.

Authors’ response: Thank you. We have now included the explanation in the manuscript.

Page 8: The rate constants and photolysis frequencies should be expressed in cm<sup>3</sup> molecule<sup>-1</sup> s<sup>-1</sup> and s<sup>-1</sup> respectively.

Authors’ response: Thank you. We calculated j<sub>1</sub> and k<sub>3</sub> in the unit of min<sup>-1</sup> and ppm<sup>-1</sup> min<sup>-1</sup>, since we wanted to compare our values with other published studies that they reported their values in min<sup>-1</sup> and ppm<sup>-1</sup> min<sup>-1</sup> (Clapp and Jenkin, 2001), (Tiwari et al., 2015).

Tiwari, S., Dahiya, A., and Kumar, N.: Investigation into relationships among NO, NO<sub>2</sub>, NO<sub>x</sub>, O<sub>3</sub>, and CO at an urban background site in Delhi, India, *Atmospheric Research*, 157, 119-126, doi:10.1016/j.atmosres.2015.01.008, 2015. Clapp, L. J. and Jenkin, M. E.: Analysis of the relationship between ambient levels of O<sub>3</sub>, NO<sub>2</sub> and NO as a function of NO<sub>x</sub> in the UK, *Atmospheric Environment*, 35(36), 6391- 6405,

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doi:10.1016/S1352-2310(01)00378-8, 2001. However, we have now provided j<sub>1</sub> and k<sub>3</sub> in the unit of s<sup>-1</sup> and cm<sup>3</sup> molecule<sup>-1</sup> s<sup>-1</sup> in the supplement material.

Page 9, line 9: The titration of O<sub>3</sub> with NO will not effectively reduce the O<sub>3</sub> concentrations. Such a titration process will produce NO<sub>2</sub> which will again photolyze in the daytime and produce O<sub>3</sub>.

Authors’ response: Thank you, however, we believe that the titration of O<sub>3</sub> by fresh NO emitted from vehicles probably causes the lower O<sub>3</sub> concentration observed at the roadside sites. Several studies reported the similar results, for example, Chan et al., (1998) studied surface ozone pattern in Hong Kong and reported that “In fact, this O<sub>3</sub> sink is a common feature observed in many countries in the Northern Hemisphere, such as in Great Britain and Canada. In these two countries, the urban stations in central London (Bower et al. 1989; UKPORG 1990) and Alberta (Angle and Sandhu 1988) show lower O<sub>3</sub> concentrations than their counterparts in the rural areas. This can be explained by the fact that the fresh precursor emissions from traffic and other sources cause direct chemical scavenging of O<sub>3</sub>.” And “Indeed, Bell et al. (1970, 1977) has shown that even under light wind conditions, pollutants generated from local sources will be dispersed within 2–3 h. Thus, the titration effect of the fresh O<sub>3</sub> precursors, especially NO, emitted from the metropolitan area of Hong Kong leads to the lower O<sub>3</sub> levels in the urban stations in our study.” Ghim and Chang (2002) studied ground-level ozone distribution in Korea and reported that “many studies reveal that background ozone concentrations in the Northern Hemisphere are around 35-40 ppb [Akimoto et al., 1996; Husar, 1998]. However, even in summer, monthly mean ozone levels in Korea are lower than this background level. . . . This could be primarily due to local effects of titration of O<sub>3</sub> by fresh NO<sub>x</sub> emissions, since most ozone monitoring stations are located in or near major cities [Fuentes and Dann, 1994]”. Munir et al., (2014) studied the diurnal variations of O<sub>3</sub> in the UK and reported that “the lowest ozone concentrations are exhibited by Marylebone monitoring site which is located approximately 1 m from the edge of Marylebone road. This road has six lanes and has a flow of 80,000

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vehicles per day. Most probably titration of ozone by fresh NO emitted by road transport keeps ozone concentrations low at this site.” Chan, L. Y., Chan, C. Y. and Qin, Y.: Surface Ozone Pattern in Hong Kong, *Journal of Applied Meteorology*, 37, 1153-1165, 1998. Ghim, Y. S., and Chang, Y. S.: Ground-level ozone distribution in Korea, *Journal of Geographical Research*, 105(7), 8877-8890, 2000. Munir, S., Chen, H., and Ropkins, K: Characterising the temporal variations of ground-level ozone and its relationship with traffic-related air pollutants in the United Kingdom: a quantile regression approach, *Int. J. Sus. Dev. Plann*, 9(1), 29-41, 2014.

Page 12, line 2: Please check the lifetime of O<sub>3</sub>. It should be few days (if not few weeks) in urban atmosphere.

Authors' response: Thank you. The lifetime of O<sub>3</sub> that was provided in our manuscript was the lifetime in “a polluted urban atmosphere” where the lifetime of O<sub>3</sub> is relatively short in this atmospheric condition. Monks et al., (2015) that reported “. . .ozone has a relatively short atmospheric lifetime, typically hours, in polluted urban regions where concentrations of its precursors are high, its lifetime in the free troposphere is of the order of several weeks (Stevenson et al., 2006; Young et al., 2013). . .” Monks, P. S., Archibald, A. T., Colette, A., Cooper, O., Coyle, M., Derwent, R., Fowler, D., Granier, C., Law, K. S., Mills, G. E., Stevenson, D. S., Tarasova, O., Thouret, V., Schneidemesser, E., Sommariva, R., Wild, O., Williams, M. L.: Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer, *Atmospheric Chemistry and Physics*, 15(15), 8889-8973, doi:10.5194/acp-15-8889-2015, 2015.

Page 12, Line 15- Page 12, Line 5: Such description is better suited for introduction.

Authors' response: Thank you. We have now modified the manuscript by moving this description to the introduction section

Page 13 Line 18 to page 19 line 2: Such discussion is well suited for outlook after proper restructuring.

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Authors' response: Thank you. Based on the reviewer, we have now modified our manuscript.

Figure 1: I would recommend showing airmass back trajectories rather than showing wind directions with two indicator arrows.

Authors' response: Thank you. Based on the reviewer, we have now modified the figure and included the airmass back trajectories based on NOAA HYSPLIT model. (Fig.1)

Figure 2: Ambient variability should also be shown along with average values. Authors should also show the concentrations of NO, in addition to CO, SO<sub>2</sub>, O<sub>3</sub>, and NO<sub>2</sub>. The colour for year 2010 and 2011 look same in panel “e”.

Authors' response: Thank you. We did not provide a plot of NO in the manuscript, since this species is not a criteria pollutant. However, we have added a plot of the concentrations of NO and modified the figure in the modified manuscript. (Fig. 2)

Figure 3: Quality of figure should be improved (overall presentation, axis labels and legends). Ambient variability (as interquartile range or 1 \_ standard deviation) should also be shown in addition to the average values. This should be done for other figures also in the paper.

Authors' response: Thank you. We have now modified the figure by adding standard deviations, and improved axis labels, and legends.

Figure 6: The minimum wind speed bin should be 0.5 – 2.0 (not 201). Please use the same radius scale for the wind rose plots.

Authors' response: Thank you. We have provided a new figure in the modified manuscript.

Table 1: Please refer to the comment for section 3.3.

Authors' response: Thank you. We have now modified our manuscript based on the reviewer's suggestion.

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Table 3: Authors should also include the SO<sub>2</sub>/NO<sub>x</sub> ratio reported from various cities in India for mobile sources as reported by Mallik and Lal, 2014. Mallik, C., and Lal, S.: Seasonal characteristics of SO<sub>2</sub>, NO<sub>2</sub>, and CO emissions in and around the Indo-Gangetic Plain, Environmental Monitoring and Assessment, 186, 1295-1310, 10.1007/s10661-013-3458-y, 2014.

Authors' response: Thank you. We have now included the SO<sub>2</sub>/NO<sub>x</sub> ratios from the study of Mallik and Lal, 2014 in the manuscript.

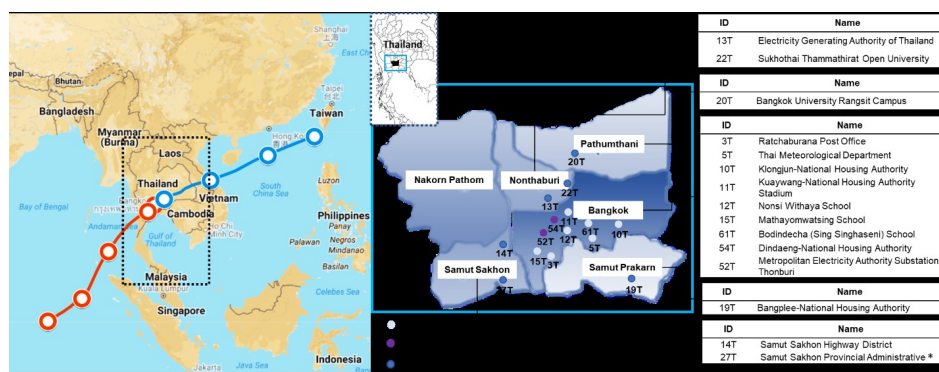
"Guwahati and Nagpur, India\*\*\* SO<sub>2</sub>/NO<sub>x</sub> > 0.3 Kolkata, and Durgapur, India\*\*\* SO<sub>2</sub>/NO<sub>x</sub> ≤ 0.13"

Please also note the supplement to this comment:

<https://www.atmos-chem-phys-discuss.net/acp-2017-1063/acp-2017-1063-AC1-supplement.pdf>

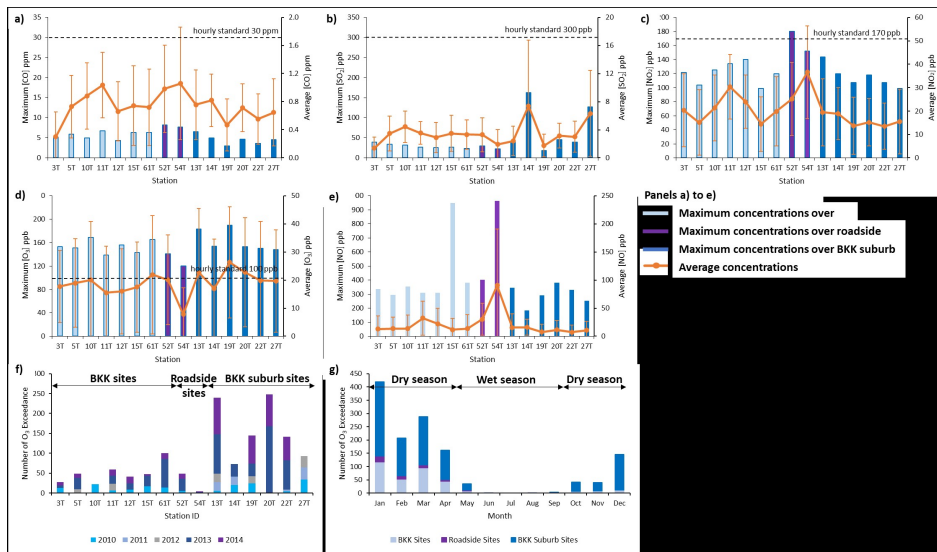
Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2017-1063>, 2017.

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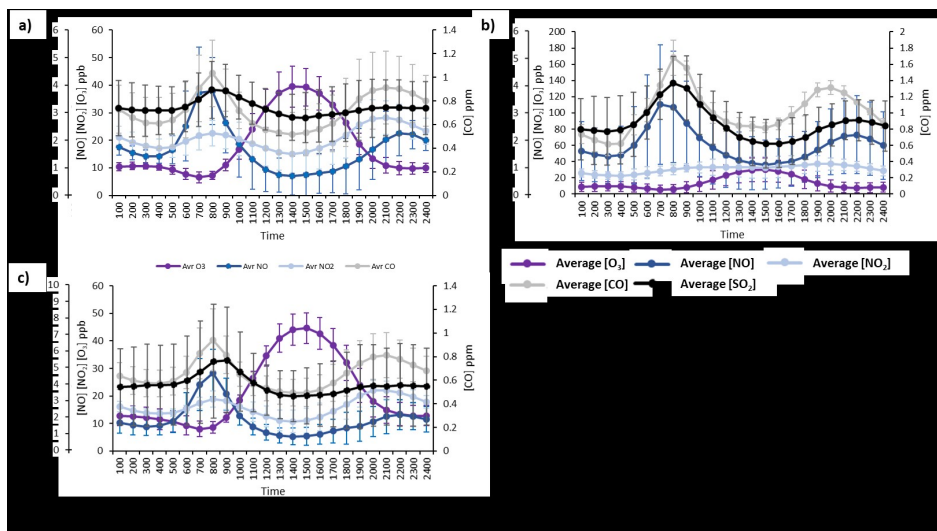
**Fig. 1.** Map of BMR, monitoring station locations and two major monsoons winds (from NOAA HYSPLIT back trajectory model). Three monitoring station types, including BKK sites, roadside sites and BKK suburb site

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**Fig. 2.** Maximum (vertical bars) and average (solid line) concentrations of a) CO, b) SO<sub>2</sub>, c) NO<sub>2</sub> d) O<sub>3</sub> and e) NO from the 15 monitoring stations, during 2010 to 2014, are compared with the hourly NAAQs (dotted)

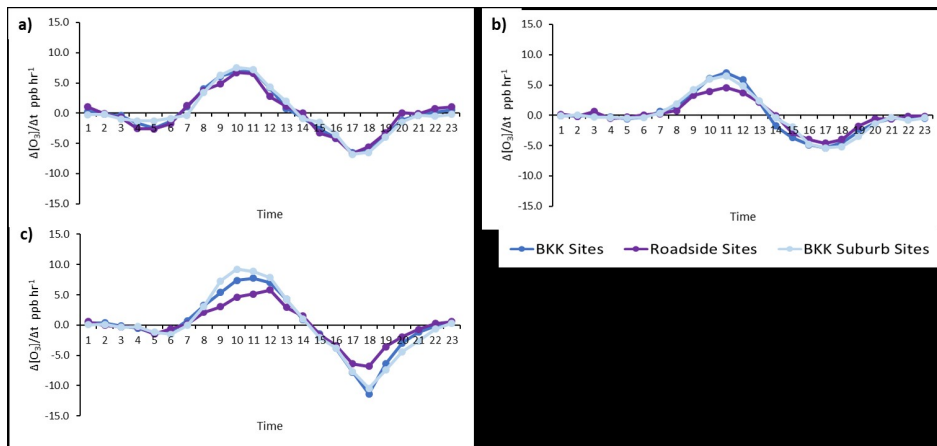
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**Fig. 3.** Diurnal variations of gaseous species including O<sub>3</sub>, NO, NO<sub>2</sub>, CO and SO<sub>2</sub> at a) BKK site b) roadside sites and c) BKK suburb sites.

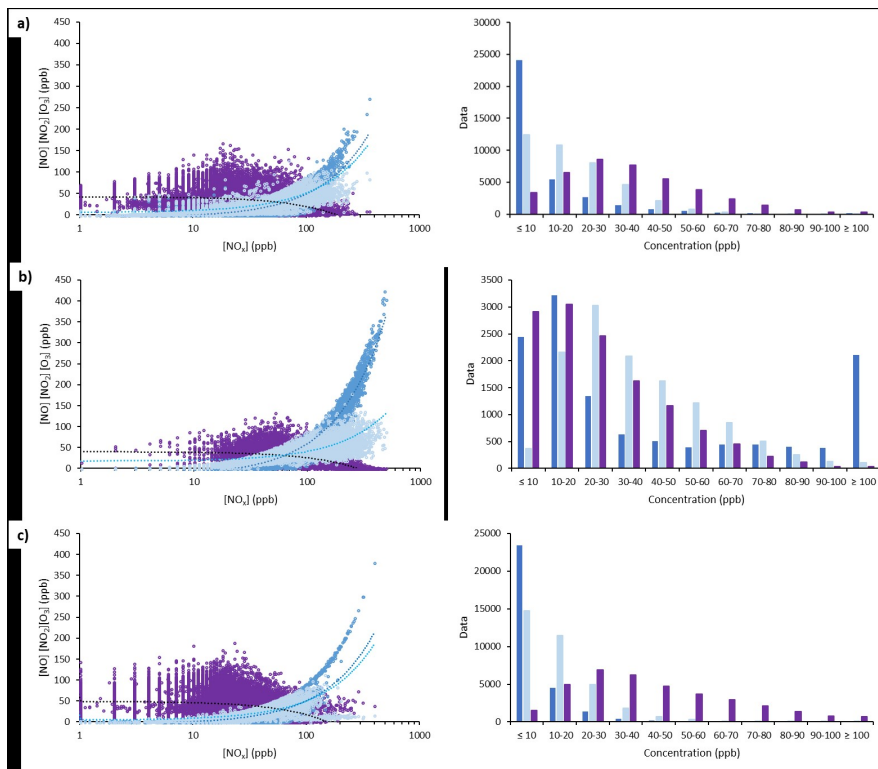
C32





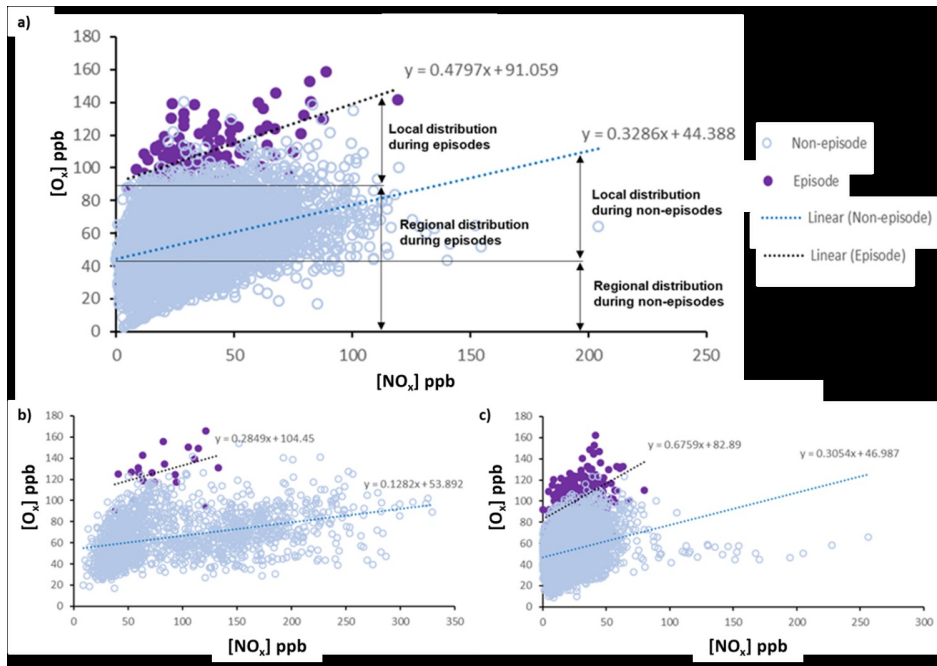
**Fig. 4.** Diurnal variations of rate of change of O<sub>3</sub> concentration ( $\Delta[O_3]/dt$ ) during a) local summer b) wet season and c) local winter.

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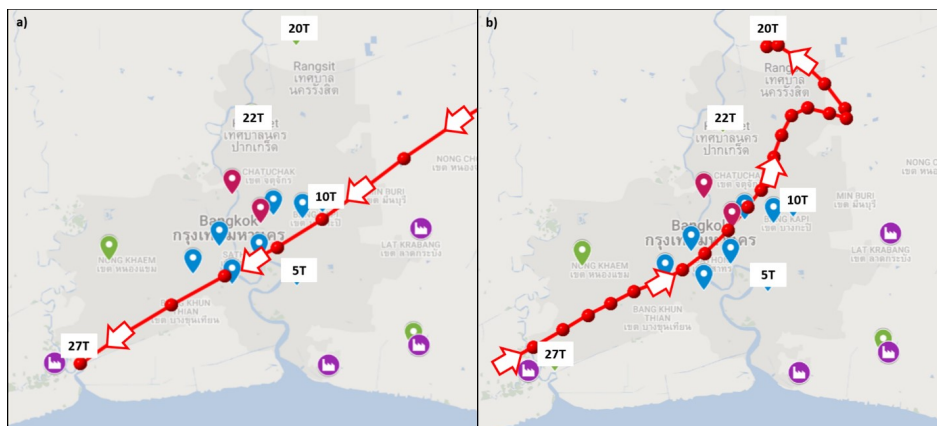
**Fig. 5.** relationship, crossover point and concentration distribution of NO, NO<sub>2</sub> and O<sub>3</sub> at a) BKK sites b) roadside sites and c) BKK suburb sites.

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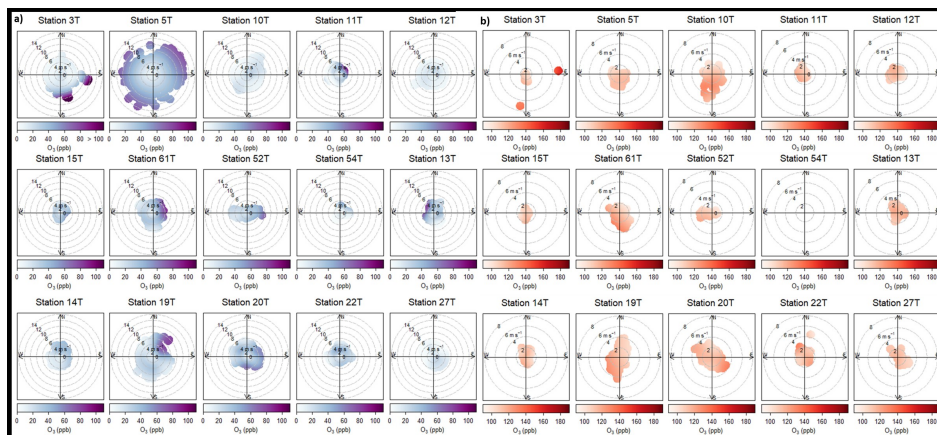
**Fig. 6.** Effects of local and regional contributions on Ox during non-episode and episode days at a) BKK sites, b) roadside sites and c) BKK suburb sites

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

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**Fig. 8.** Relationship between the concentrations of O<sub>3</sub>, wind speeds and wind directions during a) O<sub>3</sub> episodes ([O<sub>3</sub>]hourly > 100 ppb) and b) during non O<sub>3</sub> episodes ([O<sub>3</sub>]hourly ≤ 100 ppb), over BMR during 2010 t