

Response to Reviewers Comments

Assessment of Gaseous Pollutants in Bangkok Metropolitan Region, Thailand

Pornpan Uttamang, Viney P Aneja, Adel Hanna

Ref: acp-2017-1063

We wish to thank the reviewers for the careful and thoughtful review of our manuscript. We appreciate reviewer 2's comments "Overall, the article is well written and examines the interaction of Ozone with NO_x regime. The analysis was well done." All the comments and suggestions are now incorporated in the manuscript.

Reviewer #1

General Comment:

Pornpan Uttamang et al. have presented observations of CO, NO_x, SO₂ and O₃ 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 O₃ 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 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's suggestions.

Specific Comments:

Title: Authors might consider making the title of the paper more specific. Authors assess CO, NO_x, SO₂ and O₃ 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_x, SO₂ and O₃. 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, O₃ exceedances, drivers for high ozone episodes and relationship of ozone production with NO_x/NMHC ratio. These are quite relevant for the present study. Similarly, the work of Pochanart 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.

Page 2, Line 23: Authors state “possible emission sources of pollutants that associate with O₃ formation are identified”. However, such identification is not discussed in the manuscript. Authors have only used the ratio of CO/NO_x and SO₂/NO_x to identify whether the emission sources are mobile or point in nature. The method itself has an 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_x, SO₂ and O₃ 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_x, SO₂ and O₃ (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 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.

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 airmass 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 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 O₃ during daytime (6:00 AM to 6:00 PM) and during nighttime (6:00 PM to 6:00 AM) in the modified manuscript.

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 O₃ (0.10 ppm and 0.07 ppm, respectively). In this study, we compared the hourly concentrations of O₃ with the hourly O₃ standard in order to examine number of O₃ exceedances. To study the effects of O₃ on human health, we applied Air Quality Index (AQI) of O₃ instead of using the O₃ exceedance from 8-hour standard, which we believe that, using AQI of O₃ will provide more advantages than using the O₃ exceedance from 8-hour standard. Since AQI of O₃ is categorized into six categories, with four of the six categories providing the information of the severity of high O₃ concentrations on human health, from sensitive groups to healthy people; therefore, applying AQI for O₃ 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 daytime 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 (SO₂, NO_x, CO, and O₃) 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 SO₂ at all sites. In line 20 of page 7, authors speculate that SO₂ 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 SO₂ 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 NO₂ at roadside sites.

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.

We have also provided the rate of change of O₃ concentration (Naja and Lal, 2002) during the three seasons and in the three monitoring station types with explanation in the modified manuscript.

For the diurnal variations of SO₂, in the manuscript, we explained that "...The concentrations of SO₂ 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 SO₂ are nearly constant after 19:00 LT..." which the second peak of SO₂ were observed over three monitoring station types, but the magnitude of the concentrations of SO₂ over BKK sites and BKK suburb sites were small. For the diurnal variation of NO₂, 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.

Section 3.3 Interconversion between O₃, NO and NO₂ 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 O₃ formation during 10:00-16:00 LT". However, later in the section they assume photostationary state only between O₃, NO and NO₂. In polluted environments, RO₂ and HO₂ also oxidize NO to NO₂ and hence disturb the PSS of NO, NO₂ and O₃ [Mannschreck et al., 2004]. Hence the j₁ values calculated by only considering O₃, NO and NO₂ 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-NO₂-O₃ 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, j₁ 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, the photostationary state between O₃, NO and NO₂ is achieved within 60 s to 300 s during daytime [Trebs et al., 2012]. Authors should perform a calculation of j₁ 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-NO₂-O₃ 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_3 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 O₃ measurements to estimate the j_1 values and again using j_1 to explain high O₃ concentration at some sites. This is cyclic. Polynomial trend lines are used to investigate the interconversion between O₃, NO and NO₂. However, as seen from Figure 4, The fit is very poor for O₃ 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 O₃ with the gaseous criteria pollutants for the NAAQs of Thailand. The assumption of the photostationary state (PSS) ($\phi = 1$), therefore, was applied through the chemical reactions of O₃ and NO_x only.

Mannschreck et al., 2004, reviewed the PSS parameter (ϕ) as:

$$\phi = \frac{j[\text{NO}_2]}{k[\text{NO}][\text{O}_3]}$$

Where j was the photolysis rate of NO₂, and k was the rate of the chemical reaction of NO and O₃. In the Mannschreck et al., 2004, when ϕ was equal to 1, then other chemical reactions converting NO to NO₂ 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 (RO₂) played an important role to contribute to additional NO and NO₂, under clean or moderately polluted conditions. In the study of Mannschreck 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_x was 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_x concentrations the levels of peroxy radicals should approach zero, since the sink for RO₂ increases with increasing NO and since OH as a precursor for RO₂ as well as RO species are removed via reaction with NO₂”*. Therefore, we believe that the assumption of PSS holds for our study region (e.g. average of hourly concentration of NO_x at BKK sites, roadside sites, and BKK suburb sites were ~30 ppb, ~88 ppb and ~21 ppb, respectively. These NO_x values are far in excess of rural/semi-rural values in the study of Mannschreck et al., (2004)).

With regards to the calculation of j_1 , it is strongly dependent on incoming solar radiation and on other variables (i.e. the following equation):

$$j_{q,p} = \int_0^{\infty} 4\pi I_{p,\lambda} b_{a,g,q,\lambda,T} Y_{q,p,\lambda,T} d\lambda$$

Where

$4\pi I_{p,\lambda}$ = 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_3 , our intention is not to emphasize the k_3 calculation to show that our temperature measurements were reasonable, but rather to calculate j_1 . However, we have now modified our manuscript and removed using j_1 values to explain O₃ concentration.

With regards to the polynomial trend lines, we have now modified the plots by including histogram of the concentrations of O₃, NO and NO₂ to present data distribution of these species. Generally, most of the records are in low to middle concentration bins.

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 O₃ analysis, how were the back 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 O₃, the conclusion of local production is rather weak for days with O₃ 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_x and low NO_x regime should be calculated based on the ratio of NO_x OH reactivity and VOC OH reactivity or using model calculated indicators (e.g. CH₂O/NO_y, H₂O₂/HNO₃ and O₃/(NO_y-NO_x)) 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 O₃ episode was identified when hourly O₃ concentrations were greater than 100 ppb (the O₃ NAAQs for Thailand).

With regards to the linear regression, we presented “*the estimation*” of local and regional contributions of O_x. 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_x 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_x concentration regimes. The low NO_x regime (NO_x < 60 ppb) resembles the local and regional contributions during non-episode over BKK suburb sites. The high NO_x regime (NO_x > 60 ppb) may represent typical characteristic of air quality near roads”.

To estimate the local and regional contribution by plotting O_x against NO_x were reported in several published studied, 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 O₃, NO₂ and NO as a function of NO_x 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.

Mazzeoa, N. A., Venegasa, L. E. and Chorenca, H: Analysis of NO, NO₂, O₃ and NO_x 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, NO₂, NO_x, O₃ and oxidant (O_x = O₃+NO₂) 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., Aiyyer, 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, NO₂, NO_x, O₃, 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 O₃ analysis, back trajectory was determined when hourly concentration of O₃ > 80 ppb was observed either at 27T or 20T sites. By performing the backward trajectories using the NOAA HYSPLIT model, we identified the wind directions that related with high O₃ concentrations at both the monitoring stations. We calculated delta O₃ 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_x regimes in our manuscript, the low and high NO_x regimes refer to the concentrations of NO_x that are either lower or higher than the cross over point i.e. [NO_x] = 60 ppb (previous studies (Clapp and Jenkin, 2001; Notario et al., 2012; Tiwari et al., 2015) have also suggested similar NO_x regimes). Furthermore, we do not have CH₂O, NO_y, H₂O₂ and HNO₃ observations. Therefore, our analysis was limited only to NO_x species. The paper mentioned by the reviewer (Kumar et al., 2012) reported the ratio of CH₂O/NO_y, H₂O₂/HNO₃ and O₃/(NO_y-NO_x) 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 O₃, NO₂ and NO as a function of NO_x 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, NO₂, NO_x, O₃ and oxidant (O_x = O₃+NO₂) 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, NO₂, NO_x, O₃, 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_x ratio (slope of fit) because of a better correlation between the two species emitted from point sources is difficult to follow. The authors state that high CO/NO_x and low SO₂/NO_x 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_x? 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 PM₁₀ 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_x ratio. However, longer-lived NO_y should be used in place of NO_x. 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_x 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 SO₂/NO_x ratios reported in Table 3. The SO₂/NO_x 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.

With regards to the ration analysis using NO_y species, the NO_y 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 O₃ 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 O₃ concentration could also be an alternative plot. How does the wind rose look like for periods with O₃ 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 O₃ episodes ($[O_3]_{\text{hourly}} > 100$ 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 O₃, during O₃ episodes and non-episodes, in the modified manuscript. Generally, high O₃ concentrations relate with low wind speed (lower than 4 ms⁻¹) and relate with the predominant wind directions, especially, at 3T, 10T, 19T, 20T, 22T, 52T and 61T monitoring stations. Elevated O₃ concentrations associated with northerly winds were at 22T monitoring station. At 3T, 10T, 19T, 20T and 61T monitoring stations, high concentrations of O₃ associated with southerly winds. At 52T monitoring station, high concentrations of O₃ 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.

Section 3.6: Authors need to describe the calculation of air quality index. If they have used the simple hourly average O₃ concentration for calculate of AQI, then it is wrong. For calculation of hourly air quality index, O₃ 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 O₃, we calculate midpoints of 8-hour average of O₃ concentration from the average of hourly O₃ 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.

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

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) 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 NO₂ (0.17 ppm and 0.03 ppm, respectively), hourly, 24-hour and annually average standards of SO₂ (0.3 ppm, 0.12 ppm and 0.04 ppm, respectively), and hourly and 8-hour average standards of O₃ (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 CH₄ and NMVOC data from 10 out of 13 monitoring stations from BMR. Did the stations stopped monitoring CH₄ and NMVOCs?

Authors' response: Thank you. We provided the limitation in the manuscript “While NO_x 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 NO₂ 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 cm³ molecule⁻¹ s⁻¹ and s⁻¹ respectively.

Authors' response: Thank you. We calculated j_1 and k_3 in the unit of min⁻¹ and ppm⁻¹ min⁻¹, since we wanted to compare our values with other published studies that they reported their values in min⁻¹ and ppm⁻¹ min⁻¹ (Clapp and Jenkin, 2001), (Tiwari et al., 2015).

Tiwari, S., Dahiya, A., and Kumar, N.: Investigation into relationships among NO, NO₂, NO_x, O₃, 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 O₃, NO₂ and NO as a function of NO_x in the UK, Atmospheric Environment, 35(36), 6391- 6405, doi:10.1016/S1352-2310(01)00378-8, 2001.

However, we have now provided j_1 and k_3 in the unit of s⁻¹ and cm³ molecule⁻¹ s⁻¹ in the supplement material.

Page 9, line 9: The titration of O₃ with NO will not effectively reduce the O₃ concentrations.

Such a titration process with produce NO₂ which will again photolyze in the daytime and produce O₃.

Authors' response: Thank you, however, we believe that the titration of O₃ by fresh NO emitted from vehicles probably causes the lower O₃ 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 O₃ 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 O₃ 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 O₃.”* 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 O₃ precursors, especially NO, emitted from the metropolitan area of Hong Kong leads to the lower O₃ 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 O₃ by fresh NO_x 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 O₃ 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 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 O₃. It should be few days (if not few weeks) in urban atmosphere.

Authors' response: Thank you. The lifetime of O₃ that was provided in our manuscript was the lifetime in “a polluted urban atmosphere” where the lifetime of O₃ 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.

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.

Figure 2: Ambient variability should also be shown along with average values. Authors should also show the concentrations of NO, in addition to CO, SO₂, O₃, and NO₂. 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.

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.

Table 3: Authors should also include the SO₂/NO_x 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 SO₂, NO₂, 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 SO₂/NO_x ratios from the study of Mallik and Lal, 2014 in the manuscript.

Reviewer #2

We wish to thank the reviewer for the careful and thoughtful review of our manuscript. We appreciate the reviewer's comments "Overall, the article is well written and examines the interaction of Ozone with NO_x regime. The analysis was well done."

All the comments and suggestions are now incorporated in the manuscript.

Line 12: the statement is made, "On average, the number of hourly O₃ exceedences ranged from 1 - 60 hours a year." This line is confusing. The overall average should be a value, not a range. If you wish to express it as a range, then do it by year, such as 2010 that average was XX hours, 2011, the average was XX hours. This range of 1-60 hours makes no sense.

Authors' response: Thank you. We have now incorporated the change in the modified manuscript.

Section 2, Methodology line 12: When you express a range (this applies throughout, do not mix the units and values. In Section 2, Methodology line 12 you state the temperature is (~35C - 40C). This appears to read that it ranges from 35 degrees to - (minus) 40 degrees. Do this instead: (35 - 40 C).

Authors' response: Thank you. We have now incorporated the change in the modified manuscript.

Section 2, Methodology line 21: it states, " It is assumed that the monitoring sites used were representative of BMR specific patterns and trends." I think it goes without stating this that the professionals at the PCD would have done this and this does not need to be stated, but you would hope you would infer this. Remove this statement.

Authors' response: Thank you. We have now incorporated the change in the modified manuscript and the statement has been removed.

Section 2, Methodology, line 27 -29: you list the sites (19T, 20T, etc...) which mean absolutely nothing to the reader then you state in line 29 that the figure shows these. The statement that

mentions the figure should be the first line to the paragraph, not the last line. Move this line to the front so the reader can go get the figure look at it while you read the information.

Authors' response: Thank you. We have now modified the manuscript by removing site lists and referring the figure earlier in the section.

Section 2.2, line 2: you mention wind speed and direction. Is this average or vector data? Please state. This is important when calculating direction from which winds are blowing.

Authors' response: Thank you. The wind speed and wind direction are hourly averages.

Section 2.2, line 10: it is mentioned that equipment and monitoring station are calibrated every year. This is vague and could cast a shadow on validity of data. does this mean that this is done only once per year? Pollution instruments and met, or only met instruments. I am sure the PCD does calibrations more often than once annually. Please clarify this statement.

Authors' response: Thank you. As indicated in the manuscript, the data were collected, and after QA/QC, were provided to us 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 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.

Section 3.3 line 24: you use the term "atmospheric boundary layer." Is this the same as planetary boundary layer that was used previously? If it is the same term, then be consistent. If it isn't then please explain what this term means on how it differs from the PBL.

Authors' response: Thank you. We have now corrected the manuscript by using "planetary boundary layer" instead of "atmospheric boundary layer" to provide consistency in the manuscript.

Page 8, line 11: Please explain why the ratios of NO₂ and NO show significant difference. You make the statement but you don't say why. this is an important claim that you make in this paper.

Authors' response: Thank you. As suggested by the reviewer, we have now removed this from our manuscript.

Page 9, line 9: you state, "In conclusion, the titration of O₃ and NO is perhaps one of the most important processes..." Please elaborate about why this is so important.

Authors' response: Thank you. The titration of O₃ by NO is perhaps one of the most important processes to reduce O₃ concentration at roadside sites, due to this monitoring station type is more affected by fresh NO emitted from vehicles than the other monitoring station types. Several studies reported the effect of the titration of O₃ by NO, for example, Chan et al., (1998) studied surface ozone pattern in Hong Kong and reported that "*In fact, this O₃ 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 O₃ 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 O₃.” 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 O₃ precursors, especially NO, emitted from the metropolitan area of Hong Kong leads to the lower O₃ 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 O₃ by fresh NO_x 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 O₃ 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 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.

However, we have removed this from the manuscript.

Page 11, line 5: you state, "However, a negative delta O₃ may be negative. However, it appears that the data doesn't support this in the paragraph. Why is this statement made?"

Authors' response: Thank you. We put this statement to clarify that a negative delta O₃ was possibly to be observed due to O₃ deposition and/or O₃ consumption. Our analysis, negative values of delta O₃ were observed several times, however, the average of those was positive.

Assessment of ~~Air Pollution~~ Gaseous 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 below the National Ambient Air Quality Standards (NAAQs) of Thailand. However, the hourly concentrations of O₃ frequently exceeded the Thailand NAAQs standard. The maximum concentrations results reveal that the problem of high O₃ ranged from 120-190 ppb. On average, the number of hourly O₃ exceedances ranged from 1-60 hours a year depending on monitoring station locations. The exceedances occurred during the summer and winter, dry seasons, concentration continuously persisted in this area. Interconversion between O₃, NO and NO₂ indicates crossover points between the species occur when the concentration of NO_x ($[NO_x] = ([NO] + [NO_2])$) is ~60 ppb. However, when Under low NO_x regime ($[NO_x] < 60$ ppb), O₃ is the dominant species; conversely, while, under high NO_x regime ($[NO_x] > 60$ ppb), NO dominates when $[NO_x] > 60$ ppb. The calculated photochemical reaction rate (the reaction between NO₂ with sunlight), during photostationary state ranges from 0.12 to 1.22 min⁻¹. Linear regression analysis between the concentrations of O_x ($[O_x] = ([O_3] + [NO_2])$) and NO_x provides the role of local and regional contributions to O_x. Both During O₃ episodes ($[O_3]_{hourly} > 100$ ppb), the values of the local and regional O_x-contributions enhance the concentration were nearly double of O_x. Values of the local and regional O_x-contributions those during non-episode were 44-54 ppb and 0.13[NO_x] to 0.33[NO_x], respectively. Those values were about double during O₃ episodes ($[O_3] > 100$ ppb) episodes. Ratio analysis suggests that the major contributors of primary pollutants over BMR are mobile sources (CO/NO_x = 19.8). The Air Quality Index (AQI) for BMR was predominantly between good to moderate. Unhealthy, however, unhealthy O₃ categories were observed during episode conditions in the region.

Style Definition: Normal

Style Definition: Heading 2: Font: (Default) +Headings (Calibri Light), 13 pt, Not Bold, Font color: Accent 1, Space Before: 2 pt, After: 0 pt, Line spacing: 1.5 lines, Keep lines together

Style Definition: Heading 3: Font: (Default) +Headings (Calibri Light), 12 pt, Not Bold, Font color: Accent 1, Space Before: 2 pt, After: 0 pt, Line spacing: 1.5 lines, Keep lines together

Style Definition: Balloon Text: Font: (Default) Segoe UI, 9 pt

Style Definition: Bullets

Formatted: English (U.S.)

Formatted: English (U.S.)

Formatted: English (U.S.)

Formatted: English (U.S.)

31 **1. Introduction**

32 Over the last three decades, Thailand's rapid industrialization and urbanization has led to
33 an increase in global economic prowess (World Bank, 2018a). A majority of the country's
34 development has occurred within and around Bangkok (BKK) (13°45' N and 100°85' E), the
35 capital city of Thailand and Bangkok Metropolitan Region (BMR). The BKK is comprised of the
36 five adjacent provinces of BKK (World Bank, 2018a and 2018b). The increase in emissions is
37 due to accelerated growth combined with high photochemical activity, strong solar insolation,
38 high temperatures and high humidity (Kumar et al., 2012). BMR, with these conditions, has
39 begun to experience air quality degradation, in particular, enhanced secondary pollutants. Since
40 1995, BKK has experienced exceedances in Thailand NAAQs for particulate matter (PM) and
41 ozone (O₃) (PCD, 2015). The correlation between BMR air pollution and public health has been
42 observed in several published studies. Ruchirawat et al. (2007) reported the children who lived in
43 BKK are exposed to high levels of carcinogenic air pollutants which may cause an elevated
44 cancer risk. Buadong et al. (2009) reported the exposure to elevated PM and O₃ during the
45 previous day, in elderly patients (≥ 65 years), is associated with increasing the number of daily
46 hospital visits for cardiovascular diseases. Jinsart et al. (2002, 2012) reported polices and drivers
47 in BKK tended to expose higher level of PM concentrations compared with the general
48 environment, in which the concentrations of PM were already high. Role of atmospheric
49 processes in elevated O₃ in Thailand were reported in several studies. Long-range transport
50 played an important role in increasing O₃ concentration in Thailand. Generally, long-range
51 transports from the Asia continental contained higher O₃ concentrations compared with long-
52 range transports from the Indian ocean (Pochanart et al., 2001). In BMR, local emission and
53 regional transport were the major contributors to high O₃ concentrations and seasonal
54 fluctuations of O₃, respectively (Zhang and Oahn, 2002). In suburban areas of BKK, volatile
55 organic compounds (VOCs) tended to be a potential factor to enhance O₃ concentration
56 (Suthawaree et al., 2012).

57 The availability and analysis of multi-year measurements of such gaseous pollutants in
58 the BMR will improve our understanding of how they contribute to the air quality of this area.
59 As a major metropolitan area, BMR is dominated by mobile emissions sources, which
60 contributes to the emissions of CO and NO_x, precursors of ozone formation. The emissions from

61 industrial activities also dominates the BMR metropolitan area and contributes to the emissions
62 of sulfur dioxide (SO₂) and the formation of particulate matter.

63 In this study, diurnal variations, seasonal variations and inter-annual trends of gaseous
64 pollutants including carbon monoxide (CO), nitric oxide (NO), nitrogen dioxide (NO₂), SO₂ and
65 O₃ during 2010 to 2014, in BMR, have been analysed. Chemical and physical processes
66 associated with high O₃ concentrations have been investigated. Since the concentrations of
67 nitrogen oxide (NO_x) was measured at most of the monitoring station, therefore, O₃ precursors in
68 this study is referred to NO_x. The photochemical reaction was investigated during the
69 photostationary state. The effects of local emission and regional contributions of O_x are
70 presented. The severity of air pollution concentrations in BMR on human health are assessed by
71 performing Air Quality Index (AQI).

72 2. Methodology

73 ~~Bangkok (BKK), the capital city of Thailand, has the largest population and population~~
74 ~~density in Thailand. Bangkok Metropolitan Region (BMR) refers to Bangkok and~~
75 ~~2.1 Study Area~~

76 Figure 1 shows a map of BMR, the location of the monitoring stations in this study and major
77 monsoon winds over the region. BMR refers to BKK and the five adjacent provinces, including
78 Nakhon Pathom, Pathum Thani, Nonthaburi, Samut Prakan, and Samut Sakhon. These five
79 provinces are closely linked to BKK in terms of traffic and industrial development (Zhang and
80 Oanh, 2002). Since 1995, BKK has experienced exceedances in Thailand National Ambient Air
81 Quality Standard (NAAQs) for particulate matter (PM) and ozone (O₃) (PCD, 2015). The largest
82 number of O₃ exceedances ($\{O_3\}_{\text{hourly}} > 100$ ppb) occurred in the year 2000 with 174 hours of
83 exceedances (Oanh and Zhang, 2004). Furthermore, BMR is considered as a region with the
84 worst air quality in Thailand (Watcharavitoon et al., 2013). The transportation and industrial
85 sectors are considered to be the major sources of air pollutants in BKK (Watcharavitoon et al.,
86 2013). The number of vehicles in Thailand has increased since 1989. During 2014, about 36
87 million new vehicles were registered and 29 % of these cars were registered in BKK (DLT,
88 2015). About 56 % and 28 % of the registered vehicles in BKK were gasoline and diesel engines.
89 The remaining 16 % is Compressed Natural Gas (CNG). According to the database of the

Formatted: Heading 1

90 ~~Department of Industrial Work (DIW), Thailand, the number of registered manufacturing plants~~
91 ~~in Nakhon Pathom, Pathum Thani, Nonthaburi, Samut Prakan, and Samut Sakhon are 3,282~~
92 ~~(DIW, 2016), 3,756 (DIW, 2016a), 1,981 (DIW, 2016b), 7,357 (DIW, 2016c) and 6,035 (DIW,~~
93 ~~2016d). A variety of manufacturing facilities are located on the outskirts of BKK, including,~~
94 ~~metal, auto parts, paper, plastic, food, chemical manufacturing and power plants.~~

95 ~~In this study, gaseous criteria pollutants including carbon monoxide (CO), nitrogen oxide (NO),~~
96 ~~nitrogen dioxide (NO₂), sulfur dioxide (SO₂) and O₃ concentrations and trends in BMR during~~
97 ~~2010-2014 are investigated. O₃ and its precursors (only NO and NO₂) are analyzed since they are~~
98 ~~the species that were measured at a majority of the monitoring sites. Moreover, BMR~~
99 ~~experiences primarily O₃ exceedances amongst all the other gaseous criteria pollutants.~~
100 ~~Interconversion between O₃ and its precursors and photochemical reaction rate during~~
101 ~~photostationary state are examined to assess O₃ formation over BMR. Local emission, regional~~
102 ~~contribution and possible emission sources of pollutants that associate with O₃ formation are~~
103 ~~identified.~~

104 **2. Methodology**

105 **1.2 Study Area**

106 ~~BKK is located at latitude and longitude of 13°45' N and 100°85' E, over the low flat~~
107 ~~plain of Chao Praya River, elevation height 2.3 m above mean sea level. Thailand has three~~
108 ~~official seasons—local summer (around February to May), rainy (around May to October) and~~
109 ~~local winter (around October to February) as per the Thai Meteorological Department (TMD)~~
110 ~~(TMD, 2015). During the rainy season, this region's weather is influenced by Southwest~~
111 ~~monsoon wind that travels from the Indian Ocean to Thailand. This marine air mass contains~~
112 ~~high moisture, resulting in the wet season in Thailand. During this season, Thailand is~~
113 ~~characterized by cloudy weather with high precipitation and high humidity. AroundFrom~~
114 ~~October to April, this region is influenced by Northeast monsoon wind that travels from the~~
115 ~~northeasternnorth-eastern and the northern parts of Asia (China and Mongolia). This monsoon~~
116 ~~wind brings a cold and dry air mass, resulting in which leads to the dry season (local summer and~~
117 ~~local winter) in Thailand. The dry season in Thailand can be classified into two minor local~~
118 ~~seasons—winter and summer. The local winter in Thailand is characterized by cool and dry~~

Formatted: Heading 1

Formatted: Indent: First line: 1.27 cm, Space After: 0 pt, Pattern:

119 weather, while the local summer is characterized by hot (~~←~~(35 °C to 40 °C) to extremely hot
120 weather (> 40.0 °C) due to ~~the~~ strong solar radiation. During the dry season, storms may occur
121 ~~especially during the~~ seasonal ~~transition~~transition (TMD, 2015). ~~Due to its location in the~~
122 ~~coastal area of the Gulf of Thailand, land and sea breezes may play an important role on~~
123 ~~pollution dispersion over BMR. Phan and Manomaiphiboon (2012) showed that sea breezes from~~
124 ~~the Gulf of Thailand frequently occur during winter. Strong sea breezes that penetrated inland~~
125 ~~22-55 km were found during the early to mid afternoon.~~

126 Transportation and industrial sectors are considered to be the major sources of air pollutants in
127 the study area (Watcharaviton et al., 2013). For example, in 2014, ~36 million new vehicles
128 were registered in Thailand and 29 % of these cars were registered in BKK (DLT, 2015). About
129 56 % and 28 % of the registered vehicles in BKK were gasoline and diesel engines. Hourly
130 observations collected by Pollution Control Department (PCD), Thailand, from 15 monitoring
131 sites located in BMR are analyzed in this study. It is assumed that the monitoring sites used were
132 representative of BMR specific patterns and trends. The monitoring sites are categorized into
133 three categories: Bangkok (BKK) sites, Roadside sites, and BKK suburb sites). Seven Bangkok
134 sites including 3T, 5T, 10T, 11T, 12T, 15T and 61T sites, refer to the air quality monitoring sites
135 that are located within BKK's residential, commercial, industrial and mixed areas. These
136 monitoring sites are 50-100 m away from the road. Two roadside sites including 52T and 54T
137 sites, refer to the monitoring sites that are located in BKK within 2-5 m from the road (Zhang
138 and Oanh, 2002). Six BKK suburb sites including 13T, 14T, 19T, 20T, 22T and 27T sites, refer
139 to the monitoring sites that are located in provinces adjacent to BKK (Pathum Thani (site 20T),
140 Nonthaburi (sites 13T and 22T), Samut Prakan (site 19T), and Samut Sakhon (sites 14T and
141 27T)). Figure 1 shows a map of BMR with the major monsoon winds over this region and the
142 monitoring sites' location.

143 The remaining 16 % were Compressed Natural Gas (CNG) (DLT, 2017). In fact, the
144 outskirts of BKK are populated with a variety of metal, auto parts, paper, plastic, food and
145 chemical manufacturing facilities and power plants (DIW, 2016, 2016a, 2016b, 2016c, 2016d).

2.2 Data Collection and Data Analysis

The data sets in this study were provided by the PCD. Over the four-year period, January 1, 2010 to December 31, 2014, hourly observations from 15 Pollution Control Department (PCD) monitoring stations were analysed. The monitoring stations are categorized into three categories: BKK sites, roadside sites, and BKK suburb sites. BKK sites refer to the monitoring stations that are located within BKK's residential, commercial, industrial and mixed areas. They are within ~50 to 100 m away from the road. Roadside sites refer to the monitoring stations that are located in BKK within 2 to 5 m from the road (Zhang and Oanh, 2002). BKK suburb sites refer to the monitoring stations that are located in provinces adjacent to BKK (Figure 1). Quality assurance and quality control on the data set were performed by PCD prior to receiving the data. Hourly observations of the gaseous species pollutants and meteorological parameters including wind speed (WS), wind direction (WD), temperature (T) and relative humidity (RH) were automatically collected with auto calibration at the monitoring stations. Manual quality control was performed when unusual observations were found. The External audit of the equipment and monitoring stations were calibrated every year. Data availability is provided in Figure I, supplement material.

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

Data analysis, statistical data analysis (t-test) and plots are performed using Excel 2016. Predominant wind directions over BMR are illustrated by wind rose diagrams which related to O₃ concentrations are performed using WRPLOTOpenair package (tool for the analysis of air pollution data) on RStudio program (free software from Lake Environmental).

Formatted: Heading 1

Formatted: Indent: First line: 1.27 cm

Formatted: Heading 1

3. Result and Discussion

3.1 Status of Pollution in BMR during 2010- to 2014

The Figure 2 a) to e) shows the maximum and average concentration of gaseous criteria pollutants, during 2012-2010 to 2014, from the 15 monitoring sites, were analyzed and stations. These concentrations are compared with the hourly NAAQs of Thailand (NAAQs of Thailand for hourly CO, NO₂, SO₂ and O₃ are 30 ppm, 170 ppb, 300 ppb and 100 ppb, respectively) as shown in (PCD, 2018)). Since, NO is not a criteria pollutant, only the maximum and average concentrations are presented, Fig. 2(a) (d)). During the 5 years of the study period, the maximum hourly concentrations of CO, NO₂ and SO₂ were mostly lower than in their hourly standard. standards (an exceedance of NO₂ was found at 52T monitoring station, during 2013). However, the maximum concentrations of O₃ exceeded its standard. Elevated CO, NO, and NO₂ concentrations were frequently found observed at roadside sites than other sites. The average concentrations of CO, NO, and NO₂, at roadside sites, were $\sim 1.0 \pm 0.71$ ppm over roadside sites and $\sim 0.7 \pm 0.4$ ppm over BKK sites, $\sim 60.5 \pm 42.7$ ppb, and $\sim 30.9 \pm 8.1$ ppb, respectively. Elevated SO₂ were commonly observed at BKK suburb sites. The hourly maximum concentrations of CO ranged from $\sim 3-8$ ppm, than other sites. The average concentrations of NO₂SO₂ at BKK suburb sites were $\sim 32.2 \pm 17.7$ ppb, 21.1 ± 13.6 ppb and 16.3 ± 11.9 ppb over 4.0 ± 2.3 ppb. The average concentrations of O₃ during daytime (6:00 to 18:00 LT) over BKK sites, roadside sites, BKK sites and BKK suburb sites, respectively. The hourly maximum concentrations of NO₂ ranged from 62-180 ppb (an exceedance was found at 52T monitoring station, during 2013). High SO₂ concentrations were frequently found over BKK suburb sites. The average concentrations of SO₂ 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 $\sim 11.3.8 \pm 3.9$ ppb, 3.0 ± 2.1 ppb and 2.6 ± 2.3 ppb over, $\sim 9.1 \pm 4.9$ ppb and $\sim 14.2 \pm 5.4$ ppb, respectively. The 24-hour average O₃ concentrations were highest at BKK suburb sites, ($\sim 22.0 \pm 19.8$ ppb) and following by BKK sites (17.9 ± 16.9 ppb) and roadside sites. (13.3 ± 12.7 ppb). The maximum and average of gaseous pollutants the three monitoring types are provided in Table I, supplement material.

Formatted: Font: 10 pt, Bold

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

206 concentrations of CO, NO₂ and SO₂ decreased or remained constant, the concentration of O₃
207 tended to increase during the study period (Figure III, supplement material).

208 An O₃ exceedances was recorded when an hourly concentration of O₃ was greater than
209 100 ppb (hourly O₃ standard). Figure 2 f) to g) illustrate the number of hourly O₃ exceedances,
210 which they are shown by locations and by seasons, respectively. The hourly maximum
211 concentration of SO₂ ranged from 13-163 ppb. O₃ exceedances at BKK suburb sites were more
212 frequently observed than those at the other sites. The average number of hourly O₃ exceedances
213 was ~16 hours year⁻¹ at BKK sites, ~9 hours year⁻¹ at roadside sites and ~43 hours year⁻¹ at BKK
214 suburb sites. The hourly O₃ exceedances were commonly observed during dry season than during
215 the transitional period between the seasons (May) and rarely observed during wet season.

216 ~~Even though the hourly maximum concentrations of the other gaseous species were generally~~
217 ~~lower than their standards, the hourly maximum concentrations of O₃ were greater than its~~
218 ~~standard. The average concentrations of O₃ were ~22.0±19.8 ppb, 17.9±16.9 ppb and 13.3±12.7~~
219 ~~ppb over BKK suburb sites, BKK sites and roadside sites, respectively. The hourly maximum~~
220 ~~concentration of O₃ ranged from 68-190 ppb. O₃ exceedances at BKK suburb sites were more~~
221 ~~frequently occurred than those at other sites. The average number of hourly O₃ exceedances~~
222 ~~during 2010-2014 for BKK suburb sites, BKK sites and roadside sites ranged from ~43±21 hours~~
223 ~~a year, ~16±9 hours a year, and ~9 hours a year (Fig. (2(e))). Moreover, the exceedances of O₃~~
224 ~~concentration were commonly found during the dry season, especially in January (winter).~~
225 ~~During May, the transitional period between wet and dry seasons, the number of O₃ exceedances~~
226 ~~decreased and O₃ exceedance rarely occurred during wet season (Fig. (2(f))).~~

227 3.2 Diurnal Variation of the Gaseous Species

228 ~~The primary precursors for tropospheric O₃, in the urban environment, are oxide of nitrogen~~
229 ~~(NO_x; refers to NO + NO₂), and non-methane volatile organic compounds (VOCs), methane or~~
230 ~~CO (The Royal Society, 2008, Monks et al., 2009, Cooper et al., 2014). NO_x was measured~~
231 ~~continuously at all the monitoring sites. However, VOCs concentrations were measured~~
232 ~~periodically only at one monitoring station limiting its usefulness as part of this study.~~

233 ~~Diurnal variations of O₃ and its precursors over BMR during 2010-2014 gaseous pollutant are~~
234 ~~shown in Figure Fig. (3(a)-(c)). The diurnal variations of O₃ show a typical single-peak~~

Formatted: Left, Indent: First line: 1.27 cm, Space Before: 0 pt

Formatted: Heading 1

Formatted: Subscript

Formatted: Font: 10 pt, Bold

235 pattern (Aneja et al., 2001) with the concentrations ~~increase~~increased after sunrise and
236 ~~reach~~reached the peak ~~around~~~15:00 local time (LT). The concentrations begin to decline in the
237 evening and reach the minimum concentrations ~~around~~~7:00 LT ~~in~~ the next morning. The
238 concentrations ~~of O₃~~ at the peaks ~~of the diurnal variations of O₃~~ were ~40 ppb ~~over~~at BKK sites,
239 ~30 ppb ~~over~~at roadside sites and ~45 ppb ~~over~~at BKK suburb sites. The diurnal variations of
240 NO ~~and NO₂~~, show ~~double peak patterns~~a bimodal pattern with the concentrations ~~increase~~
241 ~~around 5:00 LT and~~ reach the first peak ~~around 7:00-9:00 LT before they decline. The~~
242 ~~concentrations of NO and NO₂ start rising and reach~~ and the second peak ~~around~~~7:00 to 9:00
243 ~~LT and ~21:00- to 22:00 LT, respectively.~~ The NO concentrations ~~of NO~~ at the ~~morning first-~~
244 ~~and the second-~~peak ~~over~~were ~40 ppb and ~23 ppb at BKK sites, ~110 ppb and ~73 ppb at
245 roadside sites ~~and BKK suburb sites were~~ ~40 ppb, 110 ppb and ~~,~~ and ~30 ppb. ~~In the afternoon-~~
246 ~~peak they were~~ ~23 ppb, 73 ppb ~~and~~ ~13 ppb ~~at BKK suburb sites.~~ The NO₂ concentrations ~~of~~
247 ~~NO₂ at the morning first- and the second-~~peak ~~over BKK and BKK suburb sites were~~ ~23 ppb
248 and 20 ppb and those at the afternoon peak were ~28 ppb ~~at BKK sites, ~33 ppb and~~ 22~37 ppb.
249 ~~The NO₂ concentrations over at roadside sites ranged from~~ ~22-37 ppb and were near constant
250 ~~during the day,~~ and ~20 ppb and ~22 ppb at BKK suburb sites. Even the diurnal variations of
251 ~~NO_x show a bimodal pattern, at roadside sites, the pattern was flatter than at other sites. The~~
252 ~~flatter pattern of NO_x at roadside sites reveals that this monitoring station type was affected by~~
253 ~~high concentration of NO_x all day.~~ The diurnal variations of CO show ~~double peak patterns~~a
254 ~~bimodal pattern~~ with the first- and the second-peak ~~occur around~~occurred ~8:00 LT and 21:00
255 LT. ~~The diurnal variations of CO are similar to those of NO_x, respectively.~~ The concentrations
256 ~~increase around 4:00-5:00 LT and reach~~of CO at the first ~~sharp-~~ and the second-peak were ~1
257 ~~ppm (both peaks around 8:00 LT before they decline. The CO concentrations start rising and~~
258 ~~reach the second peak at night. The CO concentrations at the morning peak were~~ ~1 ppm, 2 ppm
259 and 1 ppm and those at the night peak were ~1 ppm, 1.5 ppm and 1 ppm, ~~over~~) at BKK sites, ~2
260 and ~1.5 ppm at roadside ~~and sites,~~ and ~1 ppm (both peaks) at BKK suburb sites, ~~respectively.~~
261 ~~Diurnal patterns. The first peak of the diurnal variations~~ of NO, NO₂, and CO correspond to ~~road~~
262 ~~traffic patterns and similar to those in other big cities (Tiwari et al., 2015). The study of Leong et~~
263 ~~al. (2002) on air pollution measurement in BKK showed that, in BKK, morning rush hour~~
264 ~~occurred during in BKK (7:00- to 9:00 LT and evening rush hour occurred during 16:00-18:00~~
265 ~~LT. During traffic rush hours, traffic volume was high with low vehicle speeds. While the first~~

266 ~~peak of the diurnal pattern of pollutants occurred during the morning traffic rush hour, the). The~~
267 ~~second peak of those occurred ~3- to 5 hours after the evening traffic rush hour. This is (16:00 to~~
268 ~~18:00 LT) (Leong et al., 2002), due to a combination of pollutants emissions and collapse of the~~
269 ~~planetary boundary layer during this time. The evening planetary boundary layer is characterized~~
270 ~~by (weak turbulence and diffusion, allowing pollutants to accumulate in the layer (Arya, 1999;~~
271 ~~Jacobson, 2012).~~

272 ~~The concentrations) during this time. The diurnal variations of SO₂ show a bimodal~~
273 ~~pattern with the first- and the second-peak of SO₂ start increasing around 5:00 LT and reach~~
274 ~~maximum around 8:00 LT before the decline occurred ~8:00 LT and 21:00 LT, respectively. The~~
275 ~~concentrations of SO₂ at the morning first- and the second-peak were ~3 ppb over(both peaks) at~~
276 ~~BKK sites and, ~3 ppb (both peaks) at roadside sites, and ~6 ppb over BKK suburb sites. The~~
277 ~~concentrations of SO₂ increase again in the afternoon and reach a second peak around 21:00 LT~~
278 ~~over roadside site. Over BKK sites and BKK suburb sites, the concentrations of SO₂ are nearly~~
279 ~~constant after 19:00 LT. The concentrations of SO₂ at the second peak over roadside sites were~~
280 ~~~3 ppb and ~3-4 ppb over BKK sites and at BKK suburb sites. The double peak pattern of SO₂~~
281 ~~over At the roadside sites, the peaks are more obvious than the other sites. The result indicates~~
282 ~~that at this monitoring station type, SO₂ is primarily influenced by emission primarily emissions~~
283 ~~from vehicle exhaust using high sulfur content fuel, especially high sulfur diesel. The study of~~
284 ~~ambient air SO₂ patterns in European cities by (Henschel et al. (2013) showed that diurnal~~
285 ~~patterns of SO₂ had a double peak pattern which the morning peaks more likely related to~~
286 ~~emission during rush hour, evening peaks were possibly caused by traffic and meteorology~~
287 ~~collapse of the planetary boundary layer). It is noteworthy that BKK has a large diesel engine~~
288 ~~fleet (an estimated 25 % of registered vehicles) (DLT, 2015a2015). The diesel fuel contains~~
289 ~~~0.035 %wt Sulphur (DOEB, 2017). Given the timing of SO₂ peak (morning automotive rush~~
290 ~~hour), it is likely that SO₂ is emitted by automotive diesel engine exhaust. Season wise of the~~
291 ~~diurnal variations are provided in Figure IV, supplement material.~~

292 ~~Figure 4 a) to c) shows diurnal variations of rate of change of O₃ concentration~~
293 ~~(Δ[O₃]/dt) during dry season (local summer and local winter) and wet season at the three~~
294 ~~monitoring station types (the data has been averaged for each monitoring station type to capture~~
295 ~~the rate of change of O₃ concentration characteristics). The diurnal variations of Δ[O₃]/dt is a~~
296 ~~combination of O₃ chemistry and meteorology. In general, Δ[O₃]/dt during wet season were~~

Formatted: Indent: First line: 1.27 cm

297 lower than those during dry season. However, during local winter, the rates of change O₃
298 concentration were the highest. The Δ[O₃]/dt at the three monitoring station types, during 10:00
299 to 11:00 LT, were 4.5 to 7.0 ppb hr⁻¹ during wet season, 6.7 to 7.5 ppb hr⁻¹ during local summer,
300 and 5.7 to 9.2 ppb hr⁻¹ during local winter. The Δ[O₃]/dt became negative during 14:00 to 15:00
301 LT. As expected, the rate of change of O₃ concentration was nearly constant during nighttime.
302 Rapid changes in the mixing height and solar insolation during morning increases Δ[O₃]/dt. After
303 sunset, the formation of O₃ is inhibited and the planetary boundary layer becomes more stable
304 resulting in O₃ reduction through chemical reactions (for example, the oxidation of O₃ by NO_x)
305 and physical processes (for example, dry deposition to the earth surface) (Naja and Lal, 2002).

306 **3.3 Photochemical Reaction and Interconversion between O₃, NO and NO₂ and** 307 **Photochemical Reaction**

308 The primary precursors for tropospheric O₃, in the urban environment, are NO_x and non-
309 methane volatile organic compounds (VOCs), methane or CO (The Royal Society, 2008, Monks
310 et al., 2009; Cooper et al., 2014). While NO_x was measured continuously at all the monitoring
311 site, VOCs were measured periodically only at one monitoring station limiting its usefulness as
312 part of this study. In this study, the photostationary state (PSS) is applied through ~~at~~the chemical
313 reactions ~~for~~of O₃ formation during 10:00- to 16:00 LT. This time window is chosen due to the
314 fully developed ~~atmospheric~~planetary boundary layer with well-mixed condition (Pochanart et
315 al., 2001) ~~in order~~ to avoid accumulation ~~due to~~of air pollutants by surface inversion.
316 ~~To~~Analysis and calculation are performed only during dry season to eliminate effects of the
317 removal process by wet deposition, ~~analysis and calculation are performed only during dry~~
318 season.

319 The relationship among ~~three chemical species~~ (NO, NO₂ and O₃) under PSS is presented
320 by Eq. (1) (Seinfeld and Pandis, 1998)

$$321 \quad [O_3]_{PSS} = \frac{j_1[NO_2]}{k_3[NO]}$$

322 Eq. (1)

323 Where [O₃]_{PSS} is the concentration of O₃, at PSS, j₁ and k₃ are reaction rate coefficient of
324 photochemical reaction of NO₂ and reaction rate coefficient of chemical reaction between NO

Formatted: Heading 1

Formatted: Indent: First line: 1.27 cm

Formatted: Subscript

Formatted: Centered, Indent: Left: 3.81 cm, First line: 1.27 cm

Formatted: Font: Times New Roman

Formatted: Indent: First line: 1.27 cm

Formatted: Font: Times New Roman

325 and O₃, respectively. According to Eq. (1), the concentration of O₂ depends on the ratio of NO₂
326 and NO. Therefore, other chemical reactions or processes that affect NO₂ and NO species will
327 also affect O₃ concentrations in the atmosphere (Jacobson, 2012).

328 The ratio of NO₂ and NO are calculated only during dry season. During dry season, the values of
329 the ratios range from 0.54-4.33 in winter and from 0.87-4.33 in summer. T-test values for the
330 ratios exhibit no significant difference with season (P-value > 0.05). While there is no significant
331 difference with season, the t-test values exhibit a significant difference with locations of
332 monitoring sites. The ratios of NO₂ and NO show significantly different between roadside sites
333 and non-roadside sites (BKK sites and BKK suburb sites) with P-value < 0.05.

334 In this study, j_I is calculated based on Eq. (1), since we cannot directly measure it. The
335 values of j_I range from 0.12-1.22 min⁻¹. The values for k_3 (ppm⁻¹ min⁻¹) is calculated by Eq. (2) (Seinfeld and
336 Pandis, 1998; Tiwari et al., 2015).

337
$$k_3 = 3.23 \times 10^3 \exp[-1430/T] \quad \text{Eq. (2)}$$

Field Code Changed

338 During dry season, the values of j_I and k_3 ranged from 0.12- to 1.22 min⁻¹ in winter and
339 from 0.13-0.90 min⁻¹ in summer (Table (and 28.3 to 30.9 ppm⁻¹)). T-test values for j_I exhibit no
340 significant difference with season and location (P-value > 0.05). min⁻¹, respectively. The ratio of
341 [NO₂] and [NO] was ~1.9. The values of j_I from this study are similar to those values at an urban
342 background site in Delhi, India (values of j_I ranged from 0.4- to 1.8 min⁻¹ and the average was
343 0.8 min⁻¹) (Tiwari et al., 2015) and those values collected during a November daytime in the UK
344 (values of j_I was ~0.14 min⁻¹) (Clapp and Jenkin, 2001). The average of j_I (min⁻¹ and s⁻¹) and k_3
345 (ppm⁻¹ min⁻¹ and cm³ molecule⁻¹ s⁻¹) at the three monitoring station types are provided in Table
346 II, supplement material.

Formatted: Indent: First line: 1.27 cm

Formatted: Superscript

347 The values for k_3 (ppm⁻¹ min⁻¹) is calculated by Eq. (2) (Seinfeld and Pandis, 1998; Tiwari et al.,
348 2015).

349
$$k_3 = 3.23 \times 10^3 \exp[-1430/T] \quad (2)$$

350 During dry season, Figure 5 a) to c) shows the values of k_3 range from 28.3-29.8 ppm⁻¹ min⁻¹ in
351 winter relationships between NO, NO₂ and O₃, their crossover points, and from 30.0-30.9 ppm⁻¹
352 min⁻¹ in summer. T-test values for k_3 exhibit a significant difference with season (P-value < 0.05)
353 and no significant difference with locations of the monitoring sites (P-value > 0.05) (Table (1)).

354 Since k_3 is a function of temperature (T), therefore, the maximum values of k_3 (29.6 and 30.8
355 $\text{ppm}^{-1}\text{min}^{-1}$ in winter and summer, respectively) occur during the afternoon (around 15:00 LT)
356 when the temperature is highest. The maximum values of k_3 from this study conforms to the k_3
357 value ($29.3 \text{ ppm}^{-1}\text{min}^{-1}$) that was found at an urban background site in Delhi, India, which the
358 peak occurred at 15:00 LT (Tiwari et al., 2015).

359 Due to high value of j_1 , high O_3 concentrations are expected to be found at 11T, 20T and 52T
360 sites. However, high O_3 concentrations were found only at 20T and 52T sites, but low at 11T
361 site. The low level of O_3 concentration at 11T site has an association with the titration of O_3 by
362 NO , since high NO concentrations were observed at 11T site. In conclusion, the titration of O_3
363 by NO is perhaps one of the more important processes that control O_3 concentrations in urban
364 areas.

365 To gain a better understanding of O_3 and its precursors over BMR, the concentrations of
366 NO , NO_2 and O_3 are plotted against the concentrations of NO_x . Polynomial trend lines are added
367 in order to investigate the interconversion among these species. Figure (4(a) (c)) show
368 relationship and crossover points between the species distributions. The crossover points
369 ~~occur~~ point among species occurs when the concentration of NO_x is ~60 ppb. At this point, two
370 regimes are identified-, including low NO_x regime and high NO_x regime. Under the low NO_x
371 regime ($[\text{NO}_x] < 60 \text{ ppb}$), O_3 is the dominant species among the others, and NO_2 concentrations
372 are higher than NO for NO_x species. ~~On the other hand~~ Conversely, under the high NO_x regime
373 ($[\text{NO}_x] > 60 \text{ ppb}$), NO and NO_2 increase and, the concentrations of O_3 rapidly decrease. Under the
374 high NO_x regime, the ~~decreased~~ declination of O_3 trend-lines may describe O_3 removal process
375 through the titration of O_3 by NO .

Formatted: Indent: First line: 1.27 cm

366 3.4 Local and Regional Contribution to O_x

377 The O_x concentration is the summation of O_3 and NO_2 concentration. Under the PSS
378 condition, concentration of NO , NO_2 and O_3 approach an equilibrium and the concentration of
379 O_x may be considered constant (Keuken et al., 2009). Since the conversion between O_3 and NO_2
380 in the urban and suburban atmosphere is rapid, the use of O_x to represent production of oxidants
381 is more appropriate than only using O_3 (Lu et al, 2010). The local or NO_x -dependent contribution
382 refers to O_x concentration that is influenced by concentration of the local pollutants. The regional

Formatted: Heading 1

Formatted: Indent: First line: 1.27 cm

383 contribution or NO_x-independent refers to the background concentration of O_x that is not
384 influenced by changes of the local pollutants (Clapp and Jenkin, 2001; Tiwari et al. 2015).

385 Figure 6 a) to c) shows the local and regional contribution of O_x at the three monitoring
386 station types. The effects of the local and regional contributions to O_x concentration are
387 ~~analyzed~~analysed by plotting O_x concentrations against NO_x concentrations and fitting the plot
388 with a linear regression ($y = mx + c$). The concentration of NO_x and O_x are referred by x and y,
389 respectively. The slope of the linear regression (m) implies the local contribution, and the
390 intercept with the y-axis (c) implies the regional (background) contribution (Aneja et al., 2000;
391 Clapp and Jerkin, 2001; Notario et al., 2012). Table ~~(2)~~1 shows the comparison between the
392 fitted linear regressions from this study with other studies. The average background O_x
393 concentrations over BMR during non-episodes ([O₃]_{hourly} < 100 ppb) and episodes ~~are~~([O₃]_{hourly} >
394 100 ppb) were ~48 ppb and ~95 ppb, respectively. The local and regional contributions during
395 the episode days, in general, were about double of those during the non-episode days.
396 ~~Therefore,~~The results reveal that elevated O₃ ~~formations~~concentrations during the episode days
397 ~~were~~are influenced by both the local and regional contributions of O_x. It is noteworthy that the
398 pattern of the local and regional contributions at roadside sites during non-episode period is
399 composed of two NO_x concentration regimes. The low NO_x regime (NO_x < ~~60ppb~~60 ppb)
400 resembles the local and regional contributions during non-episode over BKK suburb sites. The
401 high NO_x regime (NO_x > ~~60ppb~~60 ppb) may represent typical characteristic of air quality near
402 roads.

403 The local contributions from the fitted linear regressions are compared with the local
404 contribution that is calculated from delta O₃ method. A delta O₃ (ΔO_3) analysis was performed to
405 reflect on the intensity of O₃ production in BMR area (Lindsay and Chameides, 1988). Lindsay
406 et al. We utilized hourly O₃ concentrations during 10:00-16:00 LT reflecting the role of
407 photochemistry in O₃ formation. Thus, the difference between O₃ concentration measured at
408 Samut Sakhon Provincial Administrative (site 27T) and Bangkok University Rangsit Campus
409 (site 20T) during the predominant wind direction should reflect the difference in the amount of
410 O₃ leaving and entering the city whenever the winds are out of the Southwest or Northeast
411 direction ((1989) analysed high-O₃ events in Atlanta, GA, and showed that rural background O₃
412 during high O₃ concentrations, Fig. (6)). This analysis provides the net increment of photochemical
413 O₃ added to an air mass over the course of the day as it advects over the city. For a more rigorous

Formatted: Indent: First line: 1.27 cm

Formatted: Font: 10 pt, Bold

414 ~~delta O₃ analysis, we need to consider the role of wind speed. Lindsay et al. (1989) analyzed~~
415 ~~high O₃ events in Atlanta, GA, and showed that rural background O₃ during episode days ([O₃] >~~
416 ~~80 ppb) in Atlanta Metropolitan Area were higher than its average and the concentration of O₃~~
417 ~~increased from ~15- to 20 ppb when the air mass travelled across the city, enhancing. This~~
418 ~~enhanced the total O₃ concentration to 80- to 85 ppb. In our study, during the different in the~~
419 ~~concentrations of O₃ overat the upwind and downwind monitoring sitesstations (20T and 27T~~
420 ~~monitoring station) are averaged-during-. The conditions to calculate ΔO₃ in this study are 1)~~
421 ~~high O₃ concentrations ([O₃] > 80 ppb) were observed at least one of the two monitoring stations~~
422 ~~2) the calculation is performed 10:00- to 16:00 LT-in, during dry season when backward~~
423 ~~trajectories from theto avoid accumulation of air pollutants by surface inversion and effects of~~
424 ~~the removal process by wet deposition 3), National Oceanic and Atmospheric Administration~~
425 ~~(NOAA) HYSPLIT model revealbackward trajectories revealed N-NE, S-SW wind directions~~
426 ~~with high O₃ concentrations ([O₃] > 80 ppb) at the monitoring sites. The(Figure 7). Even the O₃~~
427 ~~concentrations at the downwind monitoring stations are expected to be greater than the O₃~~
428 ~~concentrations at the upwind monitoring stations. However, a negative ΔO₃ may be found. The~~
429 ~~negative ΔO₃ suggests deposition of O₃ and/or O₃ was consumed as it passes over the city and/or~~
430 ~~there may have been a wind reversal so that air already polluted by the metropolitan area was~~
431 ~~brought back in to the city (Lindsay et al., 1989). The ΔO₃ in BMR ranged from -53 to 86 ppb~~
432 ~~(average about ~10.4 ppb-),) and ranged from -66 to 96 ppb (average ~9.4 ppb.) when the~~
433 ~~predominant wind direction advecting into the city were from NE and SW, respectively. Thus,~~
434 ~~we find that there was ~10 ppb enhancement of the O₃ concentration during the air pollution high~~
435 ~~O₃ concentration in BMR ([O₃] > 80 ppb), which corroborates local O₃ production analysis based~~
436 ~~on linear regression.~~

Formatted: Pattern: Clear

437 3.5 Correlation of Air Pollutants

Formatted: Heading 1

438 3.5.1 Local Sources Analysis

Formatted: Heading 1, Indent:
First line: 1.27 cm

439 Characteristic of emission sources are often determined by the ratios between CO/NO_x
440 and SO₂/NO_x. In general, the major sources of NO_x are point sources and mobile sources.
441 However, NO_x from point sources is more likely correlated with SO₂. NO_x from mobile sources
442 is more likely correlated with CO (Parrish et al., 1991). Therefore, the characteristics of mobile

443 source are high CO/NO_x ratios and low SO₂/NO_x ratios. In contrast to mobile sources, the
444 characteristic of point sources are low CO/NO_x ratios and high SO₂/NO_x ratios (Parrish et al.,
445 1991; Rasheed et al., 2014).

446 Table 32 shows the comparison between the CO/NO_x and SO₂/NO_x ratios from this
447 study and when compared with other studies. The ratio of CO/NO_x is 19.8 and the ratio of
448 SO₂/NO_x is 0.1 over BMR. This suggests that the major contributors of primary pollutants over
449 the BMR are mobile sources. However, this region may ~~also~~ be influenced by manufacturing
450 facilities' point sources (SO₂ contributor) on the outskirts of the BKK. These point sources will
451 impact the concentrations of SO₂, NO_x and CO. Correlation plots among species are provided in
452 Figure V, supplement material.

453 3.5.2 Effects of Pollutant Transport

454 In general, O₃ has a short (~~approximately hours~~) lifetime in polluted urban atmosphere-
455 (approximately hours). However, O₃ has a longer lifetime of several weeks in the free
456 troposphere. This occurrence may allow O₃ to be transported over continental scales (Stevenson
457 et al., 2006; Young et al., 2013; Monks et al., 2015). Figure 68 shows ~~high~~ O₃ concentrations
458 ({O₃}_{hourly} > 100 ppb), during episodes and non-episodes, with ~~the~~ predominant wind directions
459 over BMR during 2010 to 2014 and wind speeds. The results show that O₃ exceedances are
460 associated with ~~the local wind directions which are related to locations of the monitoring sites.~~
461 High O₃ concentrations are associated with the three low wind speed and predominant wind
462 directions; westerly, northerly and southerly winds. Elevated. In general, elevated O₃
463 concentrations associated were observed with wind speed lower than 4 ms⁻¹ with northerly winds
464 were at 11T, 13T, 14T, (22T and 27T sites. At sites station), southerly winds (3T, 5T, 10T, 12T,
465 15T, 19T, 54T 20T and 61T, high O₃ concentrations are associated with stations) and westerly
466 winds (52T station). It is noteworthy that the southerly winds. At sites 52T and 20T high,
467 generally, bring cleaner marine air mass to the land. However, under a stagnant condition (low
468 wind speed), elevated O₃ concentrations were predominantly observed with westerly wind
469 directions. The results from this study are supported by an earlier study observed (Sahu et al,
470 2013) that showed pollution concentrations over BKK related with local wind direction., 2013a,
471 2013b).

Formatted: Heading 1

3.6 Air Quality Index for O₃ Management

Enhanced ambient air pollution has an association with increased risk of adverse cardiovascular morbidity and mortality for humans. For example, increased levels of O₃ causes coughing, reduces lung function, enhances pulmonary inflammation and may increase the risk of death due to respiratory diseases (US.EPA, 2017e). While adverse health effects may occur in healthy people, enhanced ambient air pollution is a serious threat to sensitive groups (i.e. children, elders and people with respiratory system diseases). Increased lifetime exposure of tropospheric O₃ was a cause of decreased lung function in young adults (Targer et al., 2005). Air Quality Index (AQI) for air pollutants, in the US, is categorized into six. ~~Duadong et al.~~ (2009) studied the association between O₃ exposure and hospital visits for cardiovascular diseases (CVD) in the central of BKK, Thailand. The study showed a positive relationship between exposure to O₃ on the previous day with increasing number of hospital visits for CVD in elderly patients (≥ 65 years). Fann et al (2011) studied the relationship between O₃ exposure with the national public health burden in the U.S. and found O₃ associated with premature death in metropolitan areas where these numbers were greater than other habitable environs. The study of the Global Burden of Disease, Injuries, and Risk Factor study 2013 (GBD 2013) for 188 countries by Forouzanfar et al. (2015) reported the increased number of deaths during 1990 to 2013 (from 133 to 219 deaths in thousands) due to ambient O₃ pollution. World Health Organization (WHO) Regional Office for Europe, Economic Co-operation and Development (OECD) (2015) estimated the annual economic cost of premature deaths and those of morbidity from air pollution between US \$1.431 trillion and \$1.575 trillion across the countries of the WHO European Region.

For air pollutant species in the US, the AQI for each species is categorized into 6 categories (good, moderate, unhealthy for sensitive groups, unhealthy, very unhealthy, and hazardous). These categories are nonlinear and relate to human health (US.EPA, 2017, 2017a, 2017b). In Thailand, the NAAQs for the air pollutant species is pegged at an AQI value of 100. In this study, the US severity of O₃ concentrations in BMR are evaluated by AQI rating system for O₃. Table 3 provides the results were ambient air quality over BMR during 2010 to 2014 based on the following for Thailand: AQI of O₃. Based on the AQI for O₃, during the study period, the majority of air quality over BMR were was in the good AQI category (~93-99 %); 97 %, followed by the moderate air quality category (~2.3%). However, unhealthy for sensitive group (88-632 hours); (~0.7 %), unhealthy (19-209 hours) (~0.3%) and very unhealthy (2-59

Formatted: Indent: First line: 1.27 cm

503 ~~hours)(~0.04%)~~ O₃ air quality categories were ~~found during the study period. In~~
504 ~~general~~observed. Generally, BKK suburb sites have higher number of hours that were found in
505 the unhealthy for sensitive group, unhealthy and very unhealthy categories than BKK and
506 roadside sites. The average number of hours that were found in unhealthy for sensitive group,
507 unhealthy and very unhealthy categories over BKK suburb sites were 425.8, 146.7 and 28.7
508 hours. ~~Table (4) provides the ambient air quality over BMR during 2010 to 2014 based on the~~
509 ~~AQI of O₃~~The calculation of the AQI for O₃ can be found in “AQI for O₃ calculation”,
510 ~~supplement material.~~

511 This study provides measurements and analysis for the gaseous criteria pollutants.
512 However, in order to provide a well-established air quality management policy, the integration of
513 multidisciplinary analysis is needed. This will include scientific, socioeconomic and policy
514 analysis (Aneja et al, 2001). The results from this study ~~reveal~~revealed evidence of violations for
515 O₃ for air quality ~~resulting~~. This ~~resulted~~ in adverse health effects, human welfare, economics
516 and environment over BMR. Source analysis suggests ~~to control~~the first priority should be
517 ~~controlling~~ pollution ~~emission~~emissions from local sources that ~~emission~~emit primarily from
518 mobile sources ~~should be the first priority~~. The complexity between O₃ and its precursors and the
519 effects of pollution transport shows that decreasing only NO_x emissions and/or local emissions
520 may not be an effective policy to reduce O₃ since regional air pollution transport ~~contributes~~i.e.
521 ~~ozone and its precursors contribute~~ to O₃ exceedances. To identify the proportional contribution
522 between local and regional sources of O₃ concentrations during selected O₃ episode days,
523 atmospheric ~~modeling~~modelling is needed to quantify various processes that contribute to the
524 ambient concentration at specific locations. This scientific analysis provides a frame work for the
525 process of establishing an air quality policy while developing socioeconomic impacts.

526 4. Conclusion

527 _____ Among measured gaseous criteria pollutants, O₃ is the only species whose concentrations
528 frequently exceed the NAAQs of Thailand. The O₃ exceedances occur during the dry season
529 (~~local~~ summer and ~~local~~ winter) and most frequently occur over BKK sites and BKK suburb sites
530 than roadside sites; ~~which~~. On average, the number of hourly O₃ ~~exceedances at BKK sites,~~
531 ~~roadside sites and BKK suburb sites were ~16 hours year⁻¹, ~9 hours year⁻¹ and ~43 hours year⁻¹.~~

Formatted: Heading 1

532 respectively. The lower number of O₃ exceedances at roadside sites demonstrates the effects of
533 the titration of O₃ by NO played an important role, due to decrease O₃ high concentrations.
534 Interconversion between O₃ of NO and NO₂ and that were generally observed at this monitoring
535 type (average [NO]_{hourly} = ~166.0±19.8 ppb). Under photostationary state assumption, during dry
536 season, the values of reaction rate coefficient of photochemical reaction shows that of NO₂ (j_i)
537 and reaction rate coefficient of chemical reaction between NO and O₃ has a non-linear
538 relationship with its precursor with high concentrations of O₃ which occur when (k₃) range from
539 0.12 to 1.22 min⁻¹ and range from 28.3 to 30.9 ppm⁻¹ min⁻¹, respectively. NO_x concentration is
540 less than values of about 60 ppb. After this point, marks the threshold for the interconversion
541 between O₃, NO and NO₂. Under the low NO_x regime ([NO_x] < 60 ppb), O₃ concentrations is the
542 dominant species; on the other hand, under the high NO_x regime ([NO_x] > 60 ppb), the
543 concentrations of O₃ rapidly decrease, while NO_x concentrations increase. Under high NO_x
544 regime, the concentration of O₃ is influenced by NO through the titration process. The result for
545 the study shows that decreasing NO_x emission will not directly decrease O₃ concentration over
546 BMR. The regression curves reveal a background O_x concentration of ~48 ppb (non-episode) and
547 ~95 ppb (episode) over BMR. During an. The decrease of O₃ under the high NO_x regime
548 describes the important role of NO in destroying O₃ episode, both in the atmosphere in polluted
549 environments. The local and regional contributions play an important role in the increase of O_x
550 concentrations. The result reveals that, decreasing emission from only local sources may not
551 improve air quality during of O_x concentrations, under stagnant condition (wind speed < 4 m s⁻¹)
552 and predominant wind directions (northerly, southerly and westerly winds) associate with
553 elevated O₃ concentration in this area. During O₃ episodes, since the values of the local and
554 regional air pollution transport contributes to O₃ formation. Sources analysis suggests that to
555 control pollution emission from local sources, the emissions from mobile roadside sources
556 should be the first priority. contributions were about double of those during non-episodes. Air
557 Quality Index for O₃ reveal reveals evidence of violations for O₃ for air quality resulting in
558 adverse health effects, human welfare, economics and environment over BMR. air quality
559 standards, in BMR, resulting in potential adverse health effects. To achieve O₃ reduction, control
560 strategies may be needed. Emissions from mobile sources may be the first priority to manage O₃,
561 since BMR is more likely affected by mobile sources than point sources (CO/NO_x = 19.8 and
562 SO₂/NO_x = 0.1). Due to the highly nonlinear physical and chemical processes governing the

Formatted: Subscript

Formatted: Not Superscript/
Subscript

563 atmosphere, control strategies need to be evaluated in a more comprehensive approach. Air
564 quality modelling of pollution episodes in the BMR would be an appropriate approach to
565 accurately quantify various atmospheric processes contributing to high O₃ concentrations in
566 BMR.

567 **Data Availability**

568 Hourly observations in this study ~~are provided~~ were provided by Pollution Control
569 Department (PCD), Thailand.
570 Address: 92 Phahonyothin Rd, Khwaeng Samsen Nai, Khet Phaya Thai, Krung Thep
571 Maha Nakhon 10400, Thailand.
572 Phone: +66 2 298 2000
573 Website: <http://www.pcd.go.th/>

574 **Competing Interest**

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

576 **Acknowledgement**

577 We thank the Royal Thai Government for providing the Fellowship to Uttamang (ref.
578 No.1018.2/4440).

579 We thank Professor Surat Bualert, Miss Naboon Riddhiraksa, the Pollution Control
580 Department of the Ministry of Natural Resources and Environment, Bangkok, Thailand and Thai
581 Meteorological Department of the Ministry of Information and Communication Technology for
582 providing QA/QC air pollution and meteorology data. We also thank Ms. Elizabeth Adams and
583 Mr. Kurt Thurber for ~~her~~their assistance in the editorial review of the manuscript

Formatted: Heading 1

Formatted: Font: Times New Roman, Font color: Auto

Formatted: Font: Times New Roman, Font color: Auto

Formatted: Indent: First line: 1.27 cm

Formatted: Heading 1

Formatted: Indent: First line: 1.27 cm

Formatted: Heading 1

Formatted: Indent: First line: 1.27 cm

589
590
591
592
593
594
595
596
597
598
599
600

Formatted: Font: 10 pt

601 References

602 Aneja, V. P., Agarwal, A., Roelle, P. A., Phillips, S. B., Tong, Q., Watkins, N., and Yablonsky, R.: Measurements
603 and Analysis of Criteria Pollutants in New Delhi, India, Environment International, 27, 35-42, doi:10.1016/s0160-
604 4120(01)00051-4, 2001.

Formatted: Font: Times New Roman

Formatted: Normal

605 Arya, P. S.: Air pollution meteorology and dispersion, Oxford University Press, New York, USA, 1999.

606 Buadong, D., Jinsart, W., Funatagawa, I., Karita, K., and Yano. E.: Association Between PM₁₀ and O₃ Levels and
607 Hospital Visits for Cardiovascular Diseases in Bangkok, Thailand, Journal of Epidemiology, 19(4), 182-188,
608 doi:10.2188/jea.je20080047, 2009.

Formatted: Font: Times New Roman, Pattern: Clear (White)

Formatted: Normal

609 Clapp, L. J. and Jenkin, M. E.: Analysis of the relationship between ambient levels of O₃, NO₂ and NO as a function
610 of NO_x in the UK, Atmospheric Environment, 35(36), 6391- 6405, doi:10.1016/S1352-2310(01)00378-8, 2001.

Formatted: Space After: 6 pt

611 Cooper, O. R., Parrish, D. D., Ziemke, J., Balashov, N. V., Cupeiro, M., Galbally, I. E., Gilge, S., Horowitz, L.,
612 Jensen, N. R., Lamarque, J. F., Naik, V., Oltmans, S. J., Schwab, J., Shindell, D. T., Thompson, A. M., Thouret, V.,
613 Wang, Y., and Zbinden, R. M.: Global distribution and trends of tropospheric ozone: An observation-based,
614 Elementa: Science of the Anthropocene, 2(29), doi: 10.12952/journal.elementa.000029, 2019.

Formatted: Space Before: 0 pt, After: 6 pt

615 Coppalle, A., Delmas, V., and Bobbia, M.: Variability of NO_x and NO₂ concentrations observed at pedestrian level in
616 the city center of a medium size urban area, Atmospheric Environment, 35, 5361-5369, 2001.

617 Clapp, L. J. and Jenkin, M. E.: Analysis of the relationship between ambient levels of O₃, NO₂ and NO as a function
618 of NO_x in the UK, Atmospheric Environment, 35(36), 6391- 6405, doi:10.1016/S1352-2310(01)00378-8, 2001.

Formatted: Font: Times New Roman, Pattern: Clear (White)

Formatted: Space After: 6 pt

619 DIW: Manufacturing plant statistics, Nakhon Pathom, Thailand,
620 http://userdb.diw.go.th/results1.asp?pageno=1&provname=%B9%A4%C3%BB%B0%C1, last access: December
621 2016).

622 DIW: Manufacturing plant statistics, Pathum Thani, Thailand,
623 http://userdb.diw.go.th/results1.asp?pageno=1&provname=%BB%B7%D8%C1%B8%D2%B9%D5, last access:
624 December 2016a

625 DIW: Manufacturing plant statistics, Nonthaburi, Thailand,
626 <http://userdb.diw.go.th/results.1.asp?pageno=1&provname=%B%9B%9B%7BA%D%8C%3D5>, last access:
627 December 2016b.

628 DIW: Manufacturing plant statistics, Samut Prakan, Thailand,
629 <http://userdb.diw.go.th/results.1.asp?pageno=1&provname=%CA%C1%D8%B7%C3%BB%C3%D2%A1%D2%C3>,
630 last access: December 2016c.

631 DIW: Manufacturing plant statistics, Samut Sakhon, Thailand,
632 <http://userdb.diw.go.th/results.1.asp?pageno=1&provname=%CA%C1%D8%B7%C3%CA%D2%A4%C3>, last
633 access: December 2016d.

634 DLT: Transportation statistics, Department of land transport, Thailand,
635 http://apps.dlt.go.th/statistics_web/statistics.html, last access: December 2015.

636 DLT: Statistic of registered vehicle in Bangkok, categorized by fuel types, Department of land transport, Thailand, ←
637 http://apps.dlt.go.th/statistics_web/fuel.html, last access: June 2017.

638 DOEB: Notice of Department of Energy Business on determination of characteristic and quality of diesel fuel
639 (volume 5), 2011, http://elaw.doeb.go.th/document_doeb/319_0001.pdf, last access: June 2017.

640 [Fann, N., Lamson, A. D., Anenberg, S. C., Wesson, K., Risley, D., and Hubbell, B. J.: Estimating the National Public
641 Health Burden Associated with Exposure to Ambient PM_{2.5} and Ozone, *Risk Analysis*, 32\(1\), 81-95,
642 doi:10.1111/j.1529-6924.2011.01630.x, 2011.](#)

643 [Foreuzanfar, M.H. et al.: Global, regional, and national comparative risk assessment of 79 behavioral, environmental
644 and occupational, and metabolic risks or clusters of risks in 188 countries, 1990–2013: a systematic analysis for the
645 Global Burden of Disease Study 2013, *Lancet*, 386, 2287–2323, doi:10.1016/S0140-6736\(15\)00128-2, 2015.](#)

646 [Henschel, S., Querol, X., Atkinson, R., Pandolfi, M., Zeka, A., Tertre, A. L., Analistis, A., Katsouyanni, K., Chanel,
647 O., Pascal, M., Boulard, C., Haluza, D., Medina, S., and Goodman, P. G.: Ambient air SO₂ patterns in 6 European
648 cities, *Atmospheric Environment*, 79, 236-247, doi:10.1016/j.atmosenv.2013.06.008, 2013.](#)

649 [Jacobson, M. Z.: Air pollution and global warming: history, science, and solutions, Cambridge University Press,
650 Cambridge, UK, 2012.](#)

651 [Fernandez Jiménez, M. T., Climent Font, A., and Sánchez Antón, J. L.: Long term atmospheric pollution study at
652 Madrid City \(Spain\), *Water, Air, and Soil Pollution*, 142, 243–260, 2003.](#)

653 [Keuken, M., Roemer, M., and Elshout, S. V.: Trend analysis of urban NO₂ concentrations and the importance of
654 direct NO₂ emissions versus ozone/NO_x equilibrium, *Atmospheric Environment*, 43\(31\), 4780-4783,
655 doi:10.1016/j.atmosenv.2008.07.043, 2009.](#)

656 [Jinsart, W., Tamura, K., Loetkamonwit, S., Thepanondh, S., Karita, K., and Yano, E.: Roadside Particulate Air
657 Pollution in Bangkok, *Journal of the Air & Waste Management Association*, 52, 1102-1110,
658 10.1080/10473289.2002.10470845, 2002.](#)

659 [Jinsart, W., Kaewmanee, C., Inoue, M., Hara, K., Hasegawa, S., Karita, K., Tamura, K. and Yano, E.: Driver
660 exposure to particulate matter in Bangkok, *Journal of the Air & Waste Management Association*, 62:1, 64-71, doi:
661 10.1080/10473289.2011.622854, 2012.](#)

662 [Kumar, R., Naja, M., Pfister, G. G., Barth, M. C., Wiedinmyer, C. and Brasseur, G. P.: Simulations over South Asia
663 using the Weather Research and Forecasting model with Chemistry \(WRF-Chem\): chemistry evaluation and initial
664 results, *Geosci. Model Dev.*, 5, 619-648, doi:10.5194/gmd-5-619-2012, 2012.](#)

665 [Leong, S. T., Muttamara, S., and Laortanakul, P.: Air Pollution and Traffic Measurements in Bangkok Streets, *Asian
666 J. Energy Environ*, 3, 185-213, 2002.](#)

667 Lindsay, R. W., and Chameides, W. L.: High-ozone events in Atlanta, Georgia, in 1983 and 1984, *Environmental
668 Science & Technology*, 22(4), 426-431, doi:10.1021/es00169a010, 1988.

669 Lindsay, R.W., Richardson, J. L., and Chameides, W. L.: Ozone trends in Atlanta, Georgia: Have emission controls
670 been effective? *JAPCA*, 39, 40-43, 1989.

Formatted: Pattern: Clear

Formatted: Font: Times New Roman

Formatted: Pattern: Clear

Formatted: Default Paragraph Font, Font: Times New Roman, Pattern: Clear

Formatted: Font: Times New Roman

Formatted: Font: Times New Roman

Formatted: Default Paragraph Font, Font: Times New Roman, Pattern: Clear

Formatted: Font: Times New Roman

Formatted: Font: Times New Roman, Pattern: Clear (White)

Formatted: Adjust space between Latin and Asian text, Adjust space between Asian text and numbers

671 [Mazzeo, N., Venegas, L., and Choren, H.: Analysis of NO, NO_x, O₃ and NO_x concentrations measured at a green area](#)
672 [of Buenos Aires City during wintertime, Atmospheric Environment, 39\(17\), 3055-3068,](#)
673 [doi:10.1016/j.atmosenv.2005.01.029, 2005.](#)

674 [Lu, K., Y. Zhang, H. Su, T. Brauers, C. C. Chou, A. Hofzumahaus...T. Zhu \(2010\), Oxidant \(O₃+NO₂\) production](#)
675 [processes and formation regimes in Beijing, Journal of Geophysical Research, 15, doi:10.1029/2009JD012714.](#)

676 [Monks, P. S., Archibald, A. T., Colette, A., Cooper, O., Coyle, M., Derwent, R., Fowler, D., Granier, C., Law, K. S.,](#)
677 [Mills, G. E., Stevenson, D. S., Tarasova, O., Thouret, V., Schneidmesser, E., Sommariva, R., Wild, O., Williams,](#)
678 [M. L.: Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived](#)
679 [climate forcer, Atmospheric Chemistry and Physics, 15\(15\), 8889-8973, doi:10.5194/acp-15-8889-2015, 2015.](#)

680 [Naja, M. and S. Lal: Surface ozone and precursor gases at Gadanki \(13.5°N, 79.2°E\), a tropical rural site in India,](#)
681 [JOURNAL OF GEOPHYSICAL RESEARCH, 107, doi: 10.1029/2001JD000357, 2002.](#)

682 [Notario, A., Bravo, I., Adame, J. A., Díaz-de-Mera, Y., Aranda, A., Rodríguez, A., and Rodríguez, D.: Analysis of](#)
683 [NO, NO₂, NO_x, O₃ and oxidant \(Ox = O₃+NO₂\) levels measured in a metropolitan area in the southwest Iberian](#)
684 [Peninsula, Atmospheric Research, 104-105, 217-226, doi:10.1016/j.atmosres.2011.10.008, 2012.](#)

685 [Oanh, N. T., and Zhang, B.: Photochemical Smog Modeling for Assessment of Potential Impacts of Different](#)
686 [Management Strategies on Air Quality of the Bangkok Metropolitan Region, Thailand, Journal of the Air & Waste](#)
687 [Management Association, 54\(10\), 1321-1338, doi:10.1080/10473289.2004.10470996, 2004.](#)

688 [Parrish, D. D., Trainer, M., Buhr, M. P., Watkins, B. A., and Fehsenfeld, F. C.: Carbon monoxide concentrations](#)
689 [and their relation to concentrations of total reactive oxidized nitrogen at two rural U.S. sites, Journal of geophysical](#)
690 [research, 96, 9309-9320, doi:10.1029/91JD00047, 1991.](#)

691 [PCD: Thailand State of Environment 1998, Pollution Control Department, Thailand,](#)
692 [http://www.pcd.go.th/public/Publications/print_report.cfm? task=report2538, last access: November 2015,](#)

693 [Phan, T. T., and Manomphiboon, K.: Observed and simulated sea breeze characteristics over Rayong coastal area,](#)
694 [Thailand, Meteorol Atmos Phys, 116, 95-111, doi:10.1007/s00703-012-0185-9, 2012.](#)

695 [PCD: National ambient air quality and Noise standards of Thailand, Pollution Control Department, Thailand,](#)
696 [http://www.pcd.go.th/info_serv/reg_std_airsnd01.html, last access: April 2018.](#)

697 [Pochanart, P., Kreasuwun, J., Sukasem, P., Geerathadaniyom, W., Tabucanon, M. S., Hirokawa, J., Kajji, Y., and](#)
698 [Akimoto, H.: Tropical tropospheric ozone observed in Thailand. Atmospheric Environment, 35\(15\), 2657-2668,](#)
699 [doi:10.1016/s1352-2310\(00\)00441-6, 2001.](#)

700 [Rasheed, A., Aneja, V. P., Aiyyer, A., and Rafique, U.: Measurements and analysis of air quality in Islamabad,](#)
701 [Pakistan, Earth's Future, 2, 303-314, doi:10.1002/2013EF000174, 2014.](#)

702 [Ruchirawat, M., Settachan, D., Navasumrit, P., Tuntawiroon, J. and Autrup, H.: Assessment of potential cancer risk](#)
703 [in children exposed to urban air pollution in Bangkok, Thailand, Toxicology Letters, 168, 200-209,](#)
704 [doi:10.1016/j.toxlet.2006.09.013, 2007.](#)

705 [Sahu, L. K., Sheel, V., Kajino, M., and Nedelec, P.: Variability in tropospheric carbon monoxide over an urban site](#)
706 [in Southeast Asia, Atmospheric Environment, 68, 243-255, doi:10.1016/j.atmosenv.2012.11.057, 2013, 2013a,](#)

707 [Sahu, L. K., Sheel, V., Kajino, M., Gunthe, S. S., Thouret, V., Nedelec, P. and Smit, H. G.: Characteristic of](#)
708 [tropospheric ozone variability over an urban site in Southeast Asia: A study based on MOZAIC and MOZART](#)
709 [vertical profiles, Journal of Geophysical Research: Atmosphere, 118, 8729-8747, doi:10.1002/jgrd.50662, 2013b.](#)

710 [Seinfeld, J. H., and Pandis, S. N.: Atmospheric chemistry and physics: from air pollution to climate change, Wiley,](#)
711 [New York, USA, 1998.](#)

712 [Tager, I. B., Balmes, J., Lurmann, F., Ngo, L., Alcorn, S., and Künzli, N.: Chronic Exposure to Ambient Ozone and Lung](#)
713 [Function in Young Adults, Epidemiology, 16\(6\), 751-759, doi:10.1097/01.ede.0000183166.68809.b0, 2005.](#)

714 [Suthawaree, J., Tajima, Y., Khunchorn, A., Kato, S., Sharp, A., and Kajji, Y.: Identification of volatile organic](#)
715 [compounds in suburban Bangkok, Thailand and their potential for ozone formation, Atmospheric Research, 104-](#)
716 [105, 245-254, doi: 10.1016/j.atmosres.2011.10.019, 2012.](#)

Formatted: Font: Times New Roman

Formatted: Pattern: Clear

Formatted: Default Paragraph Font, Font: Times New Roman, Pattern: Clear

Formatted: Font: Times New Roman

Formatted: Font: Times New Roman

Formatted: Pattern: Clear

Formatted: Font: Times New Roman, Pattern: Clear (White)

Formatted: Font: Times New Roman

Formatted: Font: Times New Roman, Not Expanded by / Condensed by

Formatted: Font: Times New Roman

Formatted: Font: Times New Roman

Formatted: Font: Times New Roman

Formatted: Font: Times New Roman

Formatted: Font: Times New Roman

717 [Tiwari, S., Dahiya, A., and Kumar, N.: Investigation into relationships among NO, NO₂, NO_x, O₃, and CO at an](#)
718 [urban background site in Delhi, India, Atmospheric Research, 157, 119-126, doi:10.1016/j.atmosres.2015.01.008,](#)
719 [2015.](#)

720 TMD: Climate of Thailand, https://www.tmd.go.th/info/climate_of_thailand-2524-2553.pdf, last access: [November](#)
721 [2015.](#)

722 [US.EPA: Air Quality Guide for Ozone, https://airnow.gov/index.cfm?action=pubs.aqiguideozone, last access: April](#)
723 [2017.](#)

724 [US.EPA: Air Quality Index \(AQI\) Basics, https://airnow.gov/index.cfm?action=aqibasics.aqi, last access: April](#)
725 [2017a.](#)

726 [US.EPA: Daily and Hourly AQI – Ozone, https://forum.airnowtech.org/t/daily-and-hourly-aqi-ozone/170, last](#)
727 [access: April 2017b.](#)

728 [US.EPA: Health Effects of Ozone Pollution, https://www.epa.gov/ozone-pollution/health-effects-ozone-pollution,](#)
729 [last access: April 2017c.](#)

730 [Watcharaviton, P., Chio, C. P., and Chan, C. C.: Temporal and Spatial Variations in Ambient Air Quality during](#)
731 [1996–2009 in Bangkok, Thailand, Aerosol and Air Quality Research, 13, 1741-1754.](#)
732 [doi:10.4209/aaqr.2012.11.0305, 2013.](#)

733 [WHO Regional Office for Europe, OECD: Economic cost of the health impact of air pollution in Europe:](#)
734 [Clean air, health and wealth, WHO Regional Office for Europe Copenhagen, Denmark, 2015.](#)

735 [World Bank: Industrial change in the Bangkok urban region,](#)
736 <https://openknowledge.worldbank.org/handle/10986/27380?locale-attribute=en>, last access: [April, 2018a.](#)

737 [World Bank: Urbanization in Thailand is dominated by the Bangkok urban area,](#)
738 [http://www.worldbank.org/en/news/feature/2015/01/26/urbanization-in-thailand-is-dominated-by-the-bangkok-](http://www.worldbank.org/en/news/feature/2015/01/26/urbanization-in-thailand-is-dominated-by-the-bangkok-urban-area)
739 [urban-area](#), last access: [April 2018b.](#)

740 [Zhang, B., and Oanh, N. K.: Photochemical smog pollution in the Bangkok Metropolitan Region of Thailand in](#)
741 [relation to O₃ precursor concentrations and meteorological conditions. Atmospheric Environment, 36, 4211-4222,](#)
742 [doi:10.1016/S1352-2310\(02\)00348-5, 2002.](#)

Formatted: Font: Times New Roman

Formatted: Font: Times New Roman, Not Expanded by / Condensed by

Formatted: Font: Times New Roman

Formatted: Normal

Formatted: Font: Times New Roman, Not Expanded by / Condensed by

Formatted: Font: Times New Roman

Formatted: Font: Times New Roman, Not Expanded by / Condensed by

Formatted: Font: Times New Roman

Formatted: Font: Times New Roman, Not Expanded by / Condensed by

Formatted: Font: Times New Roman

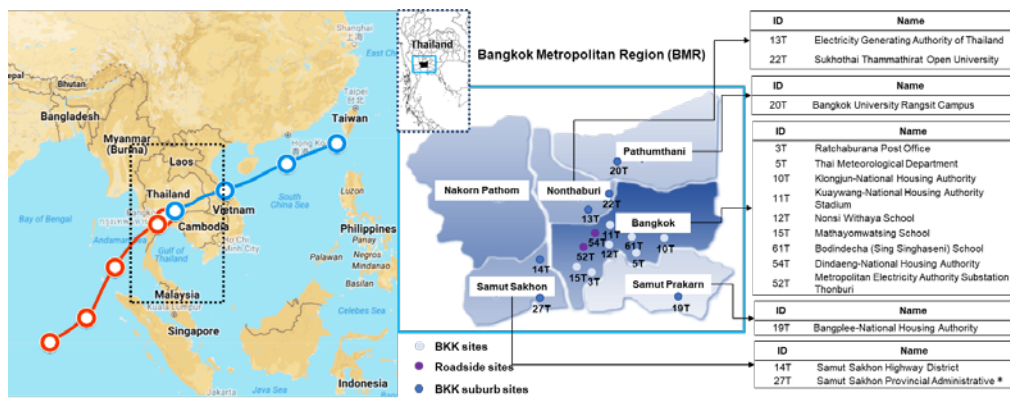
Formatted: Font: Times New Roman

Formatted: Font: Times New Roman

Formatted: Normal

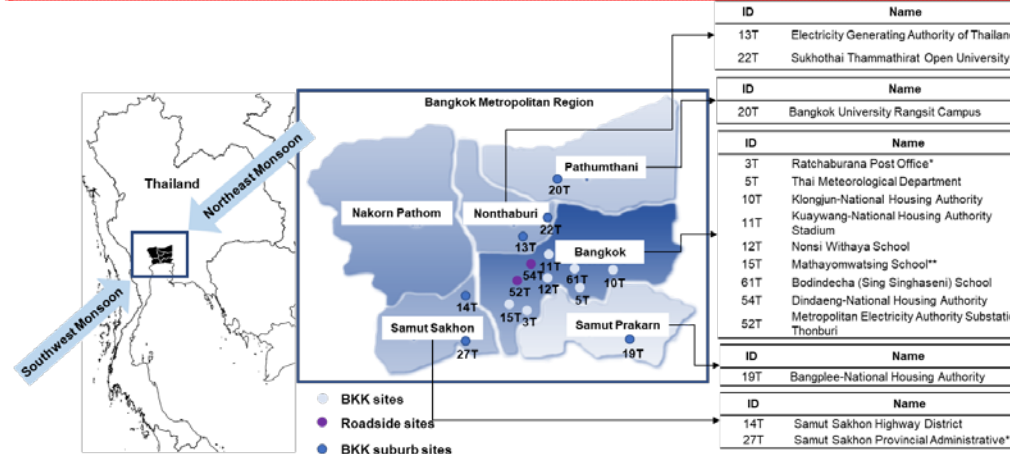
759
760
761
762
763
764
765
766
767
768
769
770
771
772
773

Figures:



Formatted: Font: 14 pt
Formatted: Font: 14 pt

Fig.



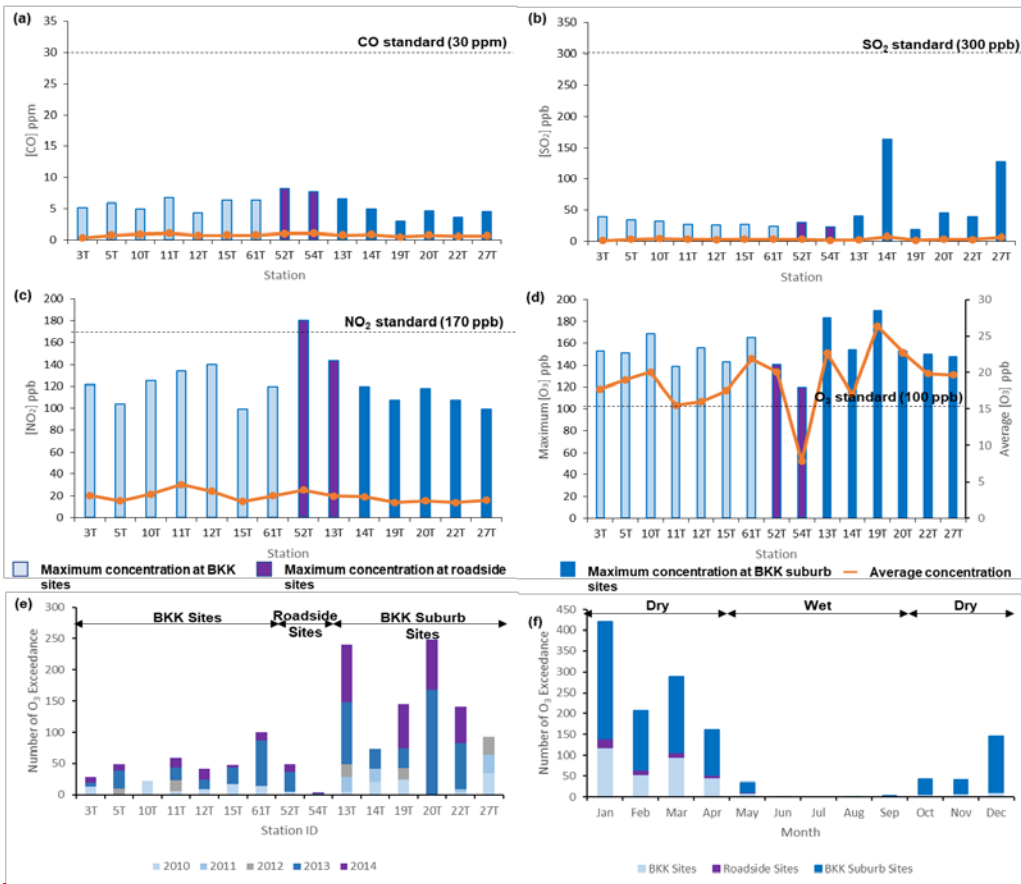
Formatted: Font: 9 pt

Figure 1: Map of BMR, including BKK monitoring station locations and five adjacent provinces, with the two major monsoons winds. The locations of three categories of (from NOAA HYSPLIT back trajectory model). Three monitoring sites station types, including BKK sites, roadside sites and BKK suburb sites are shown in light blue dots, purple dots and blue dots, respectively. (Note: * the station has been closed since 1 January 2014; ** the station has been closed since 1 August 2015; *** the station has been closed since 1 October 2013; October 2013).

Formatted: Font: Not Bold

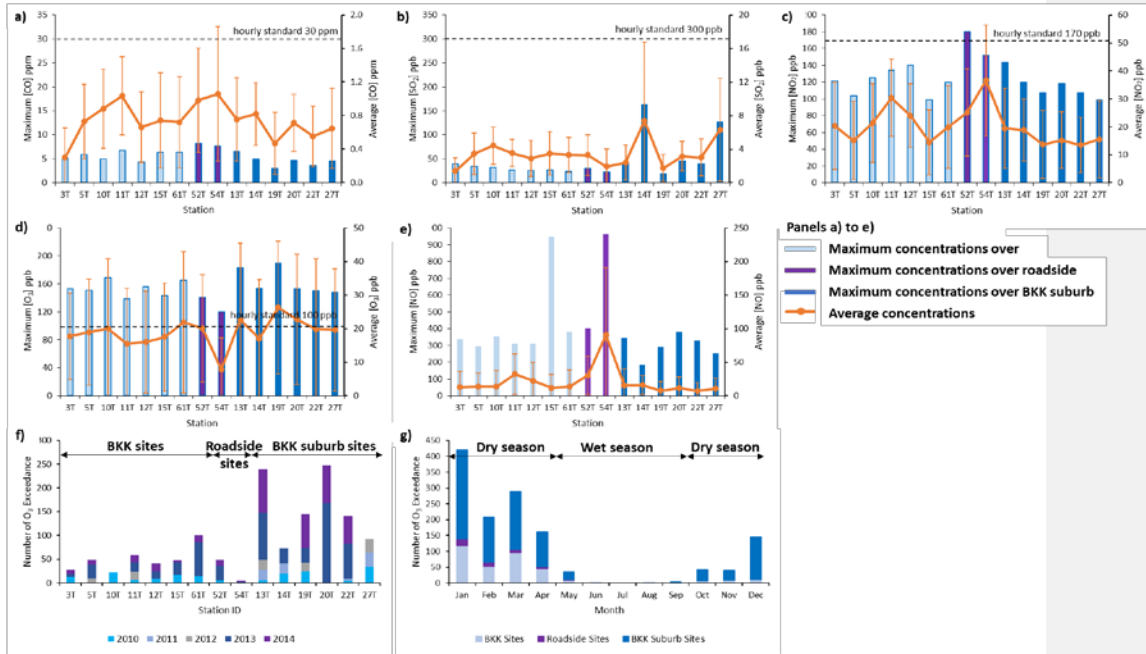
Formatted: Font: 10 pt, Not Bold, Not Italic

774
775
776
777
778
779
780
781
782
783
784
785
786



787

788 Figure



789

790 **Fig 2:** Maximum (vertical bars) and average (solid line) concentrations of (a) CO, (b) SO₂, (c) NO₂ and (d) O₃ and
 791 (e) NO from the 15 monitoring stations, during 2010- to 2014, are compared with the hourly NAAQs (dotted
 792 line) of Thailand. Three different sites, the BKK sites, roadside sites and the BKK suburb sites are represented by
 793 light blue, purple and blue colors. (except NO which is not a criteria pollutant). The number of hourly O₃
 794 exceedances of hourly O₃ concentration are shown by (e) monitoring stations, (f) locations and (g) months/seasons.

Formatted: Thai Distributed Justification

Formatted: Font: Not Bold

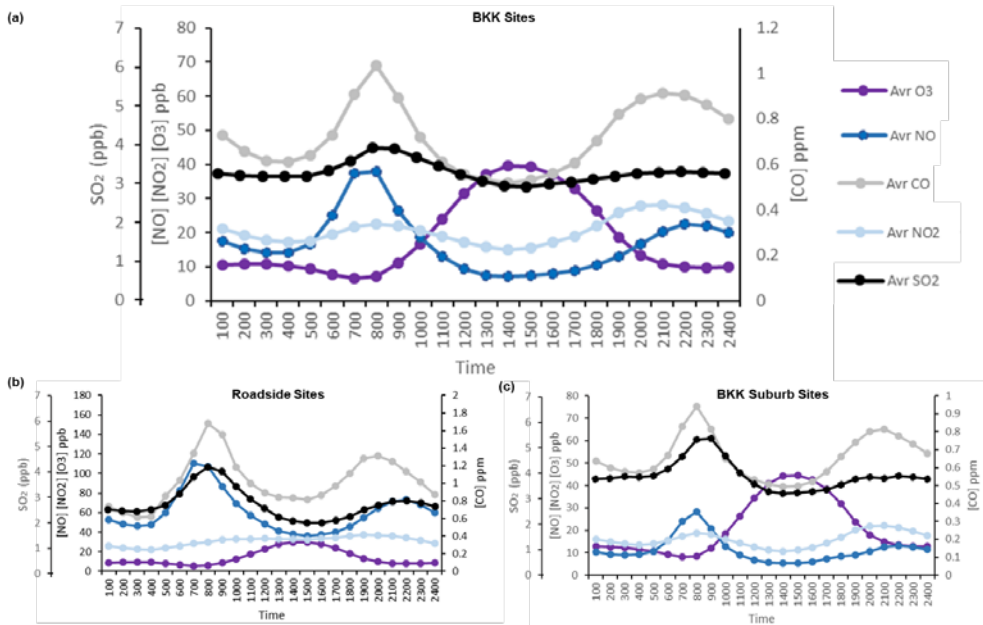
Formatted: Space Before: 6 pt, After: 6 pt

795

796

797

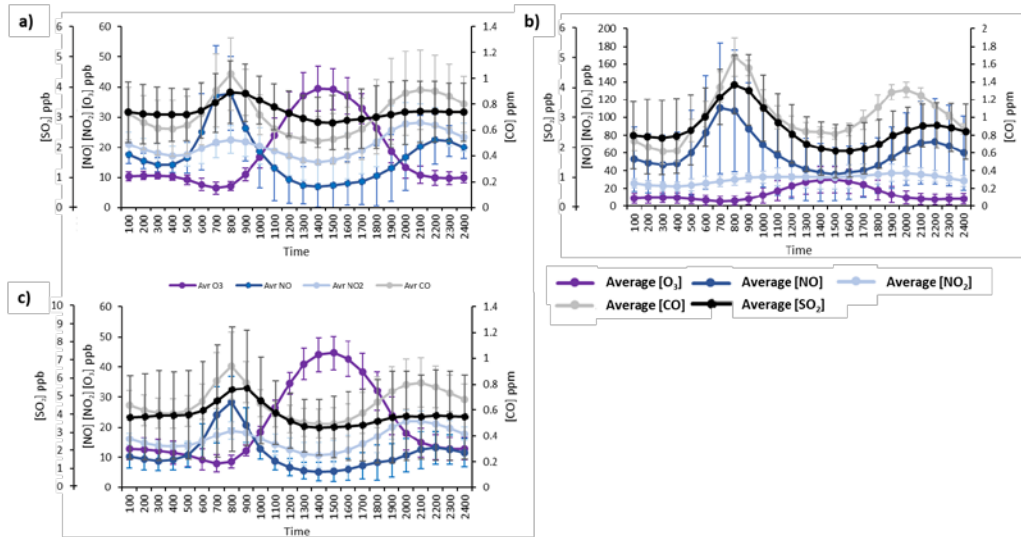
798



799

800

Figure



801

802

803

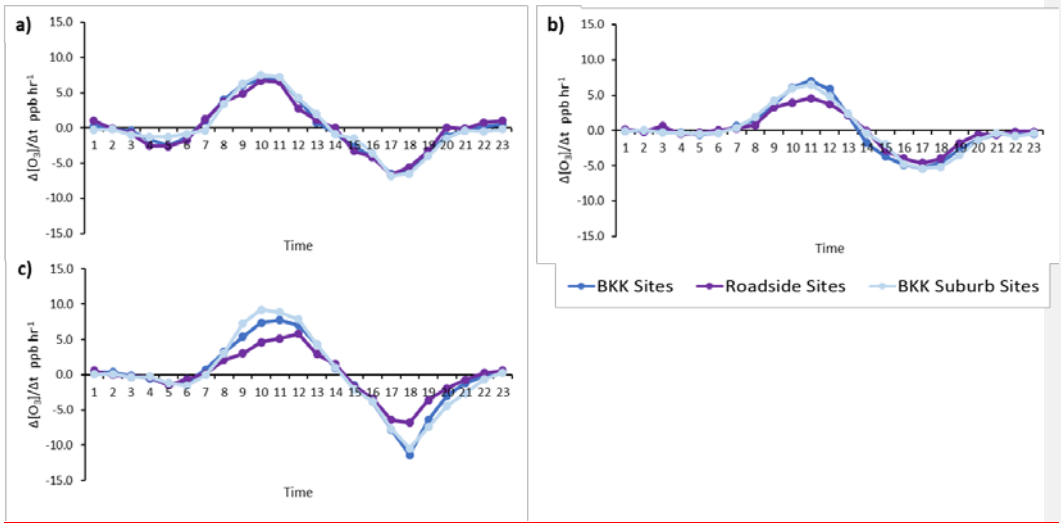
804

Fig 3: Diurnal variations of gaseous species including O₃, NO, NO₂, CO and SO₂ at (a) BKK site (b) roadside sites and (c) BKK suburb sites.

Formatted: Font: Not Bold, Not Italic

Formatted: Space Before: 6 pt, After: 6 pt

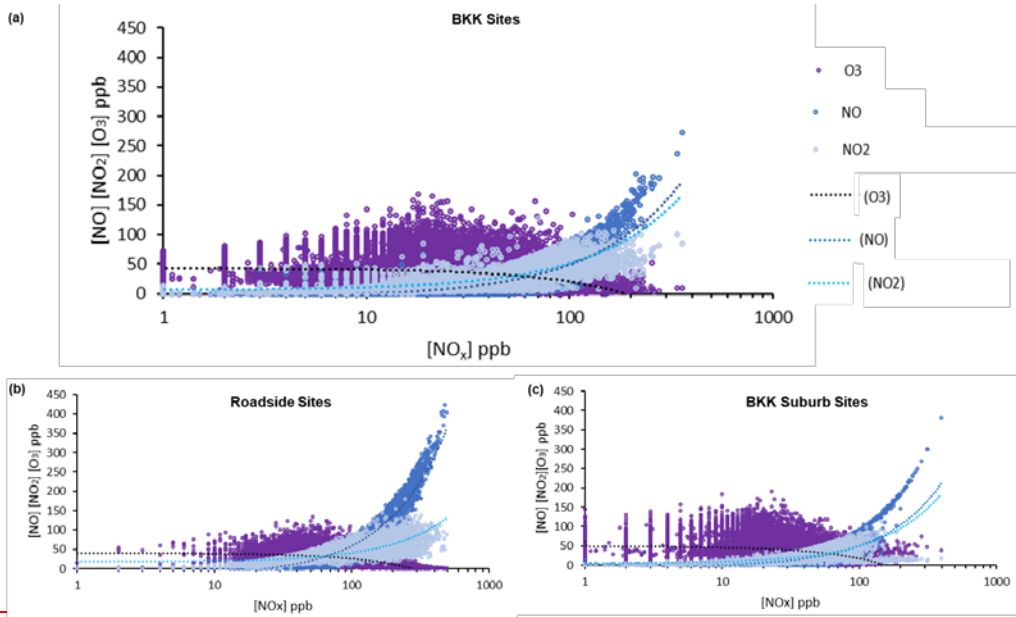
805
806
807
808
809
810



811
812
813
814
815
816
817
818

Fig.

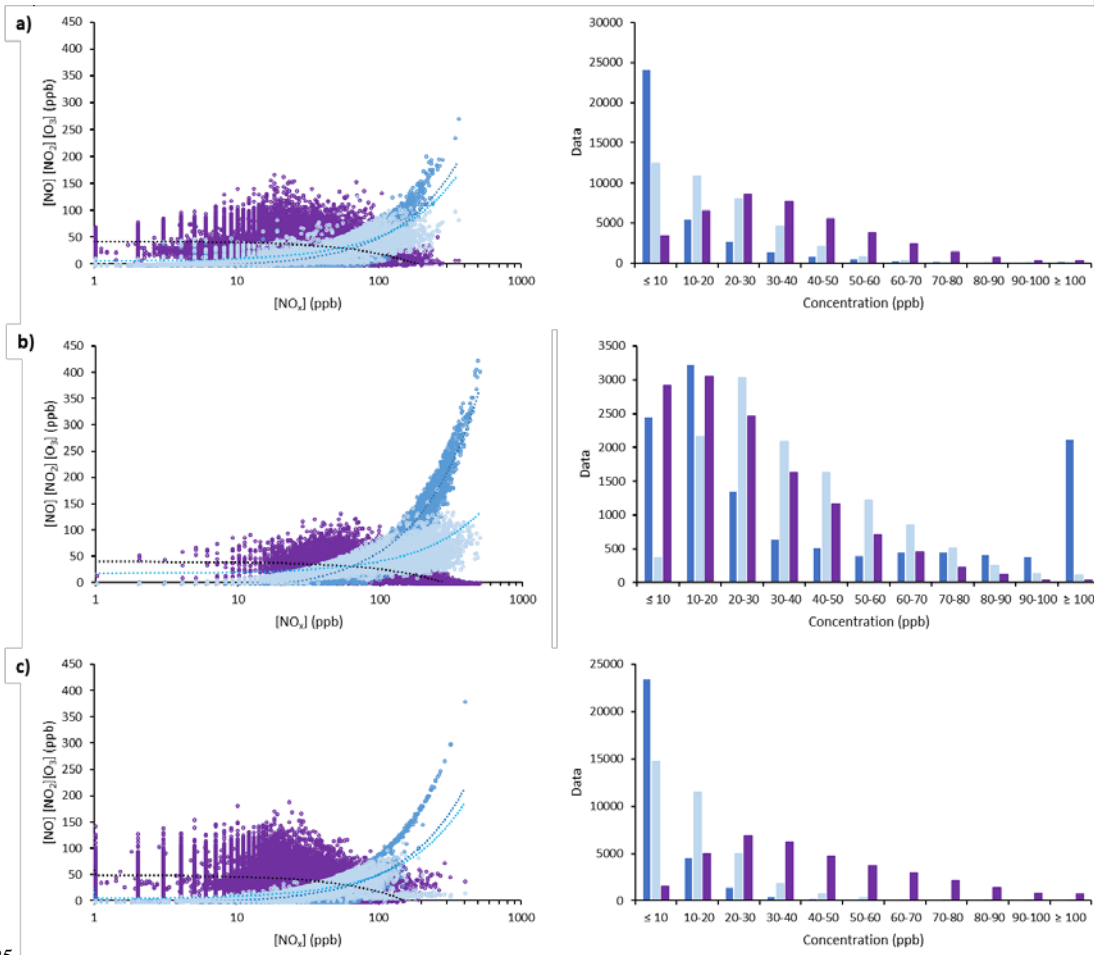
Formatted: Font: 10 pt, Bold



819
 820 **Figure 4: Interconversion between O_3 , NO and NO_2 —Diurnal variations of rate of change of O_3 concentration**
 821 **($\Delta O_3/dt$) during a) local summer b) wet season and c) local winter.**

Formatted: Font: Bold

822
 823
 824



825

826 Fig. 10:00-16:00 LT for 2010 to 2014 (a) BKK sites, (b) relationship, crossover point and concentration distribution
 827 of NO, NO₂ and O₃ at a) BKK sites b) roadside sites and (c) BKK suburb sites. The polynomial regressions provide
 828 the crossover point for NO_x (60 ppb).

Formatted: Font: 10 pt, Bold

829

Formatted: Font: 10 pt, Not Bold,
Not Italic

830

831

832

833

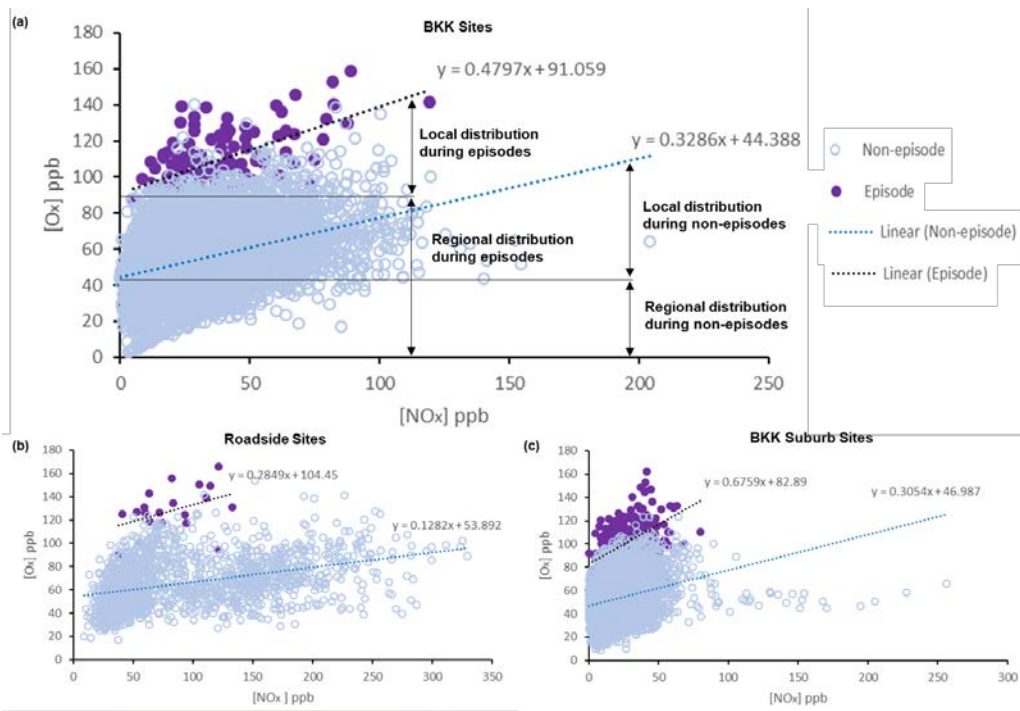
834

835

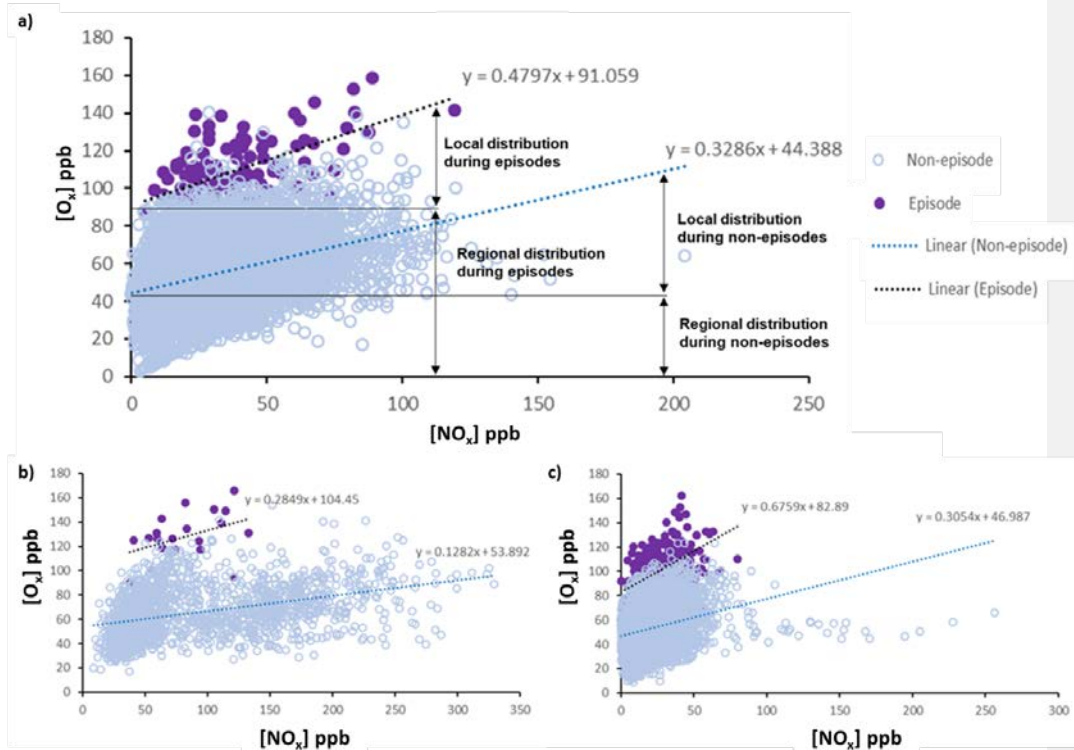
836

837

838
839
840
841
842
843

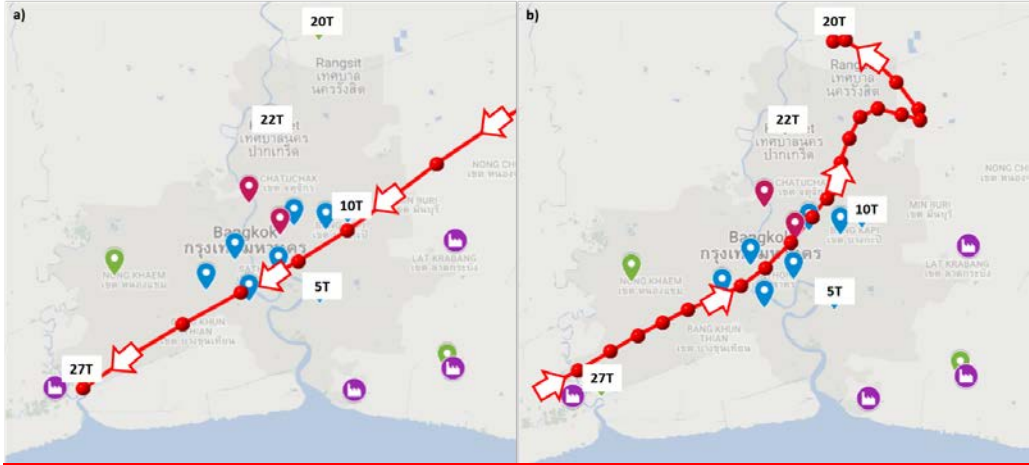


844



846
 847 **Fig. 6:** Effects of local and regional contributions on O_x during non-episode and episode days ~~over BMR~~ at (a) BKK
 848 sites at (b) roadside sites and (c) BKK suburb sites ~~during~~.

849
 850
 851
 852
 853
 854
 855
 856



857

858 **Fig. 7:** Backward trajectories from HYSPLIT model reveal a) NE wind direction (Jan 13, 2010) and b) SW wind
 859 direction (Jan 1, 2010-2014.)

Formatted: Space Before: 0 pt

860

Formatted: Indent: First line: 0 cm

861

862

863

864

865

866

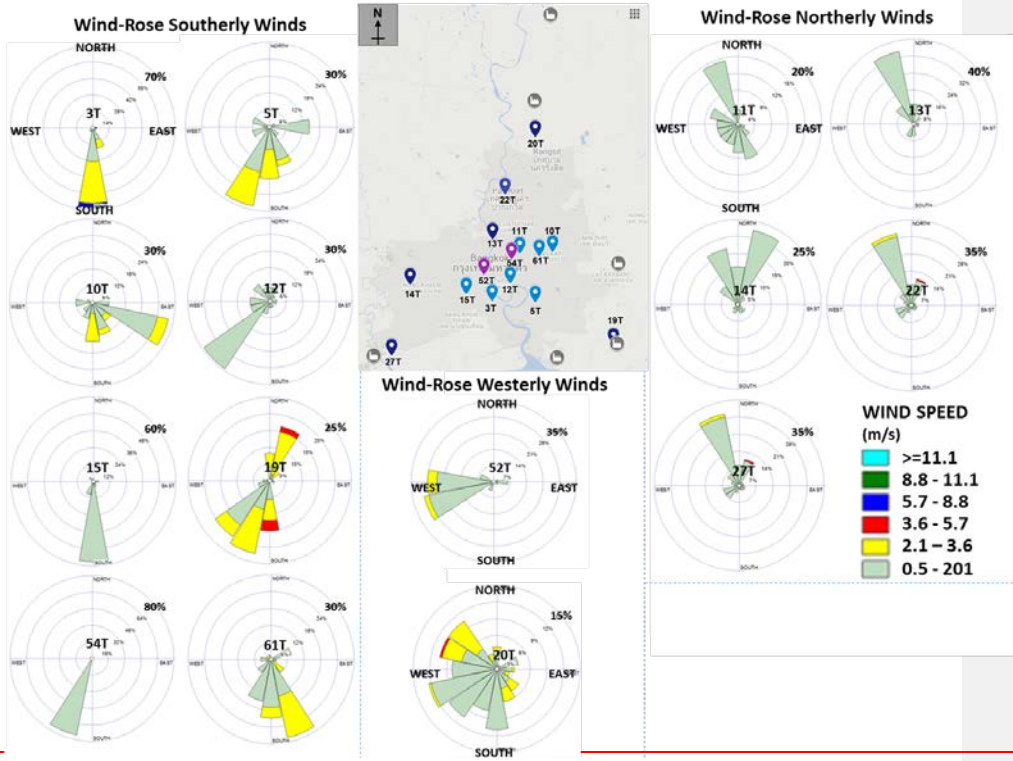
867

868

869

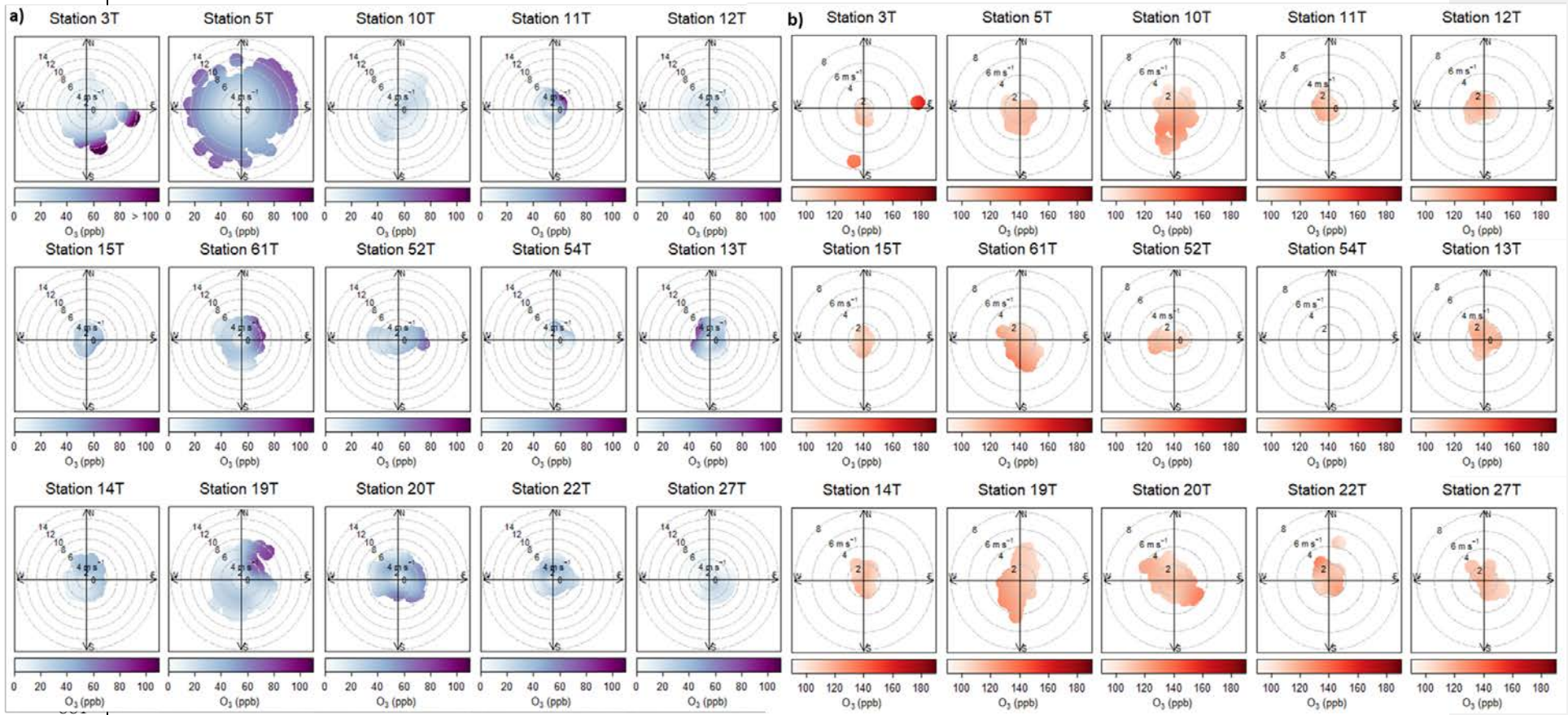
870

871



872
873
874
875
876
877
878
879
880

Figure 6



882 **Fig. 8:** Relationship between high O_3 concentration the concentrations of O_3 , wind speeds and wind directions during a) O_3 episodes ($[O_3]_{\text{hourly}} > 100$ ppb)
 883 (monitoring site ID is listed at the center of the Wind Rose Plots) and wind directions and b) during non O_3 episodes ($[O_3]_{\text{hourly}} \leq 100$ ppb), over BMR during
 884 2010 to 2014. The map illustrates several industrial areas located near the study area and the monitoring stations, blues, purples and dark blues identify BKK
 885 sites, roadside sites and BKK suburb sites; greys identify industrial areas
 886

887
888
889
890
891
892
893

894 **Tables:**

895 **Table 1:** chemical rate coefficients (k_{37}, j_i) during winter and summer from the BKK sites, roadside and BKK suburb
 896 sites, 2010-2014

Coefficient	Season	BKK sites						Roadside sites			BKK suburb sites					
		3T	5T	10T	11T	12T	15T	61T	52T	54T	13T	14T	19T	20T	22T	27T
k_3 ($\text{ppm}^{-1}\text{min}^{-1}$)	Winter	29.1	29.8	29.2	29.1	29.4	29.0	28.7	29.5	28.3	29.5	29.2	28.8	29.2	29.3	29.2
	Summer	30.1	30.9	30.4	30.2	30.5	30.2	30.1	30.4	30.5	30.5	30.7	30.0	30.8	30.9	30.0
j_i (min^{-1})	Winter	0.12	0.50	0.32	0.76	0.95	0.39	0.50	0.79	0.51	0.42	0.39	0.37	1.22	0.34	0.53
	Summer	0.13	0.51	0.47	0.72	0.43	0.57	0.23	0.90	0.36	0.50	0.69	0.49	0.86	0.36	0.46

Formatted: Space Before: 6 pt, After: 6 pt, Tab stops: Not at 1.71 cm

Formatted: Left: 2.54 cm, Right: 2.54 cm, Top: 2.54 cm, Bottom: 2.54 cm, Width: 21.59 cm, Height: 27.94 cm, Header distance from edge: 1.27 cm, Footer distance from edge: 1.27 cm, From text: 0 cm, Numbering: Continuous

897
898
899
900
901
902
903
904
905
906
907
908
909
910
911
912
913
914
915
916
917
918
919
920
921
922
923

Table 2 **Table 1:** the comparison of fitted linear regression from this study, including from BKK sites, roadside sites, and BKK suburb sites with other studies.

	Non-Episode	Episode
<u>This study</u>		
-BKK sites	$[O_x] = 0.33[NO_x]+44.39$	$[O_x] = 0.48[NO_x]+91.10$
-Roadside sites	$[O_x] = 0.13[NO_x]+53.89$	$[O_x] = 0.29[NO_x]+104.45$
-BKK suburb sites	$[O_x] = 0.31[NO_x]+47.0$	$[O_x] = 0.68[NO_x]+82.89$
UK*	$[O_x] = 0.412097[NO_x]+55.538.2$	$[O_x] = 0.097112[NO_x]+38.255.5$
Buenos Aires, Argentina**	$[O_x] = 0.099[NO_x]+22.0$	
Delhi, India***	$[O_x] = 0.54[NO_x]+28.89$	

924 *Clapp and Jenkin (2001)

925 **Mazzeo et al. (2005)

926 ***Tiwari et al. (2015)

927

928

929

930

931

932

933

934

935

936

937

938

939

940

941

942

943

944

945

946

947

948

949 **Table 32:** the comparison of CO/NO_x and SO₂/NO_x ratios from this study with other studies (modify from Rasheed
950 et al., 2014)

Region	Source	CO/NO _x	SO ₂ /NO _x
--------	--------	--------------------	----------------------------------

Formatted: Indent: First line: 1.27 cm, Tab stops: Not at 1.71 cm

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
<u>Guwahati and Nagpur, India***</u>			<u>≥ 0.3</u>
<u>Kolkata, and Durgapur, India***</u>			<u>≤ 0.13</u>
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

951 * Fernandez-Jiménez et al., 2003

952 ** Coppalle et al., 2001

953 *** Mallik and Lal, 2014

954

955

956

957

958

959

Table 43: Number of hours that were found in different AQI categories of O₃ over the BMR during 2010 to 2014.

AQI

Hour

Formatted: Indent: First line: 1.27 cm, Tab stops: Not at 1.71 cm

Formatted: Font: Times New Roman, 10 pt

	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	

960

961

962

Formatted: Tab stops: Not at 1.71 cm