Response to Reviewers Comments

Assessment of Gaseous Pollutants in Bangkok Metropolitan Region, Thailand

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Ref: acp-2017-1063

We wish to thank the reviewers for the careful and thoughtful review of our revised manuscript. All comments and suggestions now are incorporated in the manuscript. The main manuscript, supplement, figures and tables have been reviewed and modified following reviewers' comments. Furthermore, the manuscript has been reviewed by an English Editor.

Reviewer#1

General Comment:

I appreciate the major revision undertaken by the authors. They have improved the quality of analysis. Thanks for including a timeline of the measurements, inter-annual plots and seasonal variation. However, some of the important crucial concerns are still present. I had to face hard time to read the interactive discussion for my first review, where several special characters and formulae were not typeset properly. Some examples are on page C15 and Page C16. There have been several careless mistakes in the supplement. While the main text mentions values of j_1 in the range of 0.12-1.22 min^{-1}, corresponding values provided in the supplement table 1 are ~ 29 min^{-1}. The figure captions and legends are difficult to follow and sometimes even not explained properly. Examples are Main text figure 3, Main text figure 5, supplement Figure 5, An important concern I want to raise for the editor is related to the journal scope which is focused on studies with general implications for atmospheric science rather than investigations that are primarily of local or technical interest. How does this article fit in the scope of ACP considering the investigation of air pollution of a region presented in this study?

Authors' response: Thank you.

1) We have corrected and have also improved the table by including maximum, minimum, means and standard deviations of j_1 and k_3 based on observations at the three monitoring station types, and calculated j_1 based on modeling analysis. More details can be found in authors' response in comment 3.1.

Table II is changed from:

Table II: chemical rate coefficients during dry season at BKK sites, roadside and BKK suburb sites, 2010 to 2014

Rate coefficient	Unit	BKK sites	Roadside sites	BKK suburb sites
j_1	min^{-1}	29.7±0.7	29.7±1.0	29.8±0.7
	s^{-1}	0.004±0.002	0.007±0.0001	0.006±0.003
k_3	$ppm^{-1}\ min^{-1}$	0.47±0.2	0.64±0.3	0.55±0.3
	$cm^3\ molecule^{-1}\ s^{-1}$	$2.02e^{-14}±2.1e^{-16}$	$2.03e^{-14}±1.2e^{-18}$	$2.03e^{-14}±1.4e^{-16}$

Table is changed to:

Table II: Statistical analysis of the chemical rate coefficients (j_1 and k_3) based on an observational analysis during dry seasons at BKK sites, roadside and BKK suburb sites, 2010 to 2014.; and statistical analysis of j_1 based on a modeling analysis at the latitude and the longitude of 13.76 ˚N, 100.50 ˚E in a dry season, 2010.

Sites	**Rate coefficient**											
	$\boldsymbol{j_1}$						$\boldsymbol{k_3}$					
	min^{-1}			**s^{-1}**			**ppm^{-1} min^{-1}**			**cm^3 molecule^{-1} s^{-1}**		
	Max	**Min**	**Average**	**Max**	**Min**	**Average**	**Max**	**Min**	**Average**	**Max**	**Min**	**Average**
Based on observation*												
BKK	0.95	0.12	0.74±0.2	0.016	0.004	0.008±0.035	30.9	28.6	29.8±0.7	$2.06e^{-14}$	$1.99e^{-14}$	$2.02e^{-14}\pm2.01e^{-16}$
Roadside	0.90	0.36	0.64±0.3	0.015	0.011	0.013±0.002	30.6	28.3	29.7	$2.03e^{-14}$	$2.03e^{-14}$	$2.03e^{-14}$
BKK suburb	1.22	0.34	0.55±0.3	0.022	0.007	0.010±0.004	30.9	28.8	29.8±0.7	$2.04e^{-14}$	$2.01e^{-14}$	$2.03e^{-14}\pm1.34e^{-16}$
Based on modeling**												
13.7 ˚N, 100.5 ˚E						0.021±0.002						

2) As suggested by the reviewer, we have now improved Figure 3 caption in the manuscript.

Figure 3 caption is changed from:

Fig 3: Diurnal variations of gaseous species including O_3, NO, NO_2, CO and SO_2 at a) BKK site b) roadside sites and c) BKK suburb sites.

Figure 3 caption is changed to:

Fig 3: Diurnal variations of gaseous species. The plots provide the average concentrations of O_3, NO and NO_2 in ppb, the average concentrations of CO in ppm and the average concentrations of SO_2 in ppb at a) BKK site; b) roadside sites; and c) BKK suburb sites. Vertical bars provide ±1 standard deviations of the species concentrations.

3) We have improved Figure 5 and its caption in the manuscript. All the legends are now clear; and we have provided clarity on the crossover points.

Figure 5 is changed from:

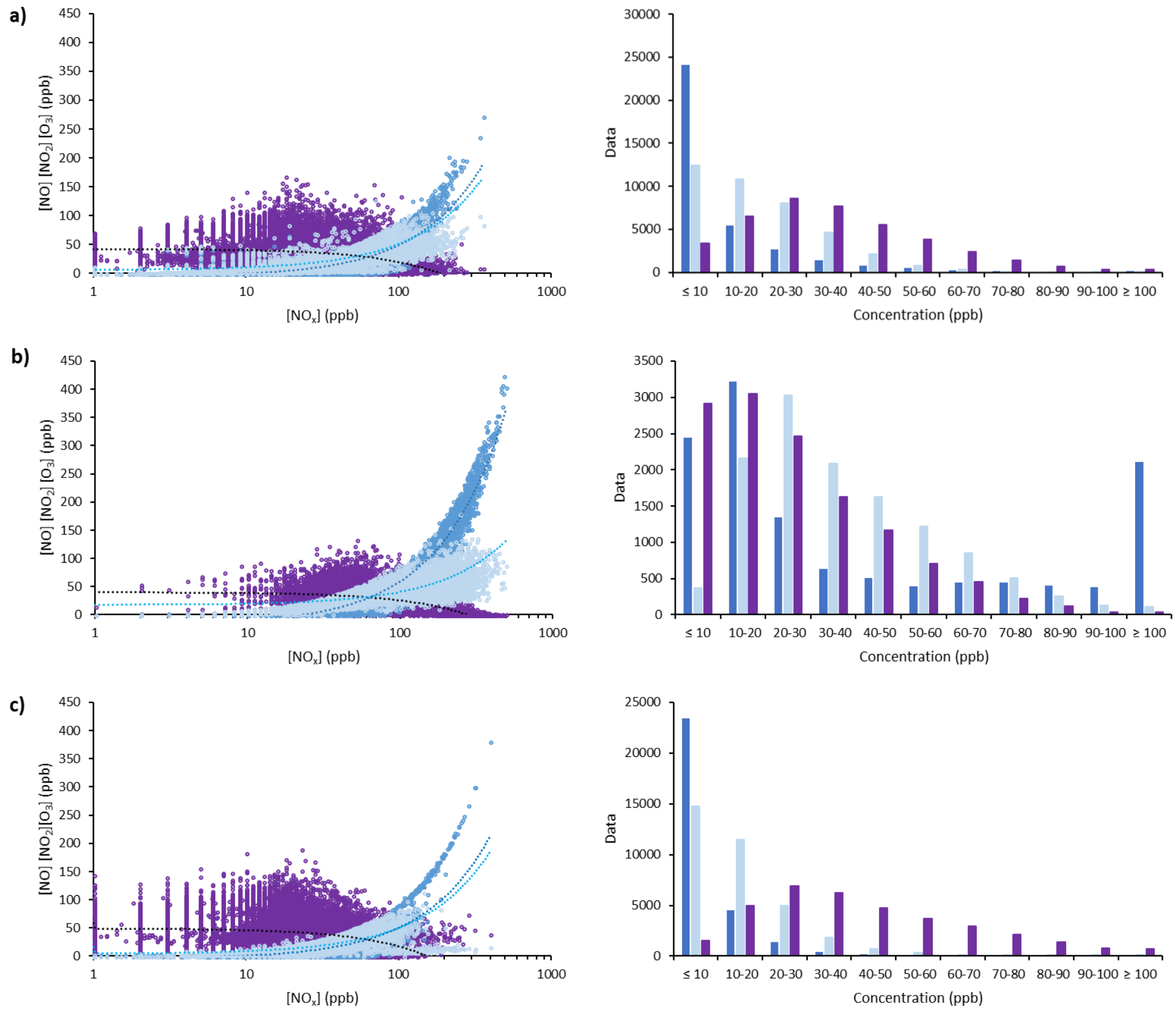

Fig. 5: relationship, crossover point and concentration distribution of NO, NO_2 and O_3 at a) BKK sites b) roadside sites and c) BKK suburb sites.

Figure 5 is changed to:

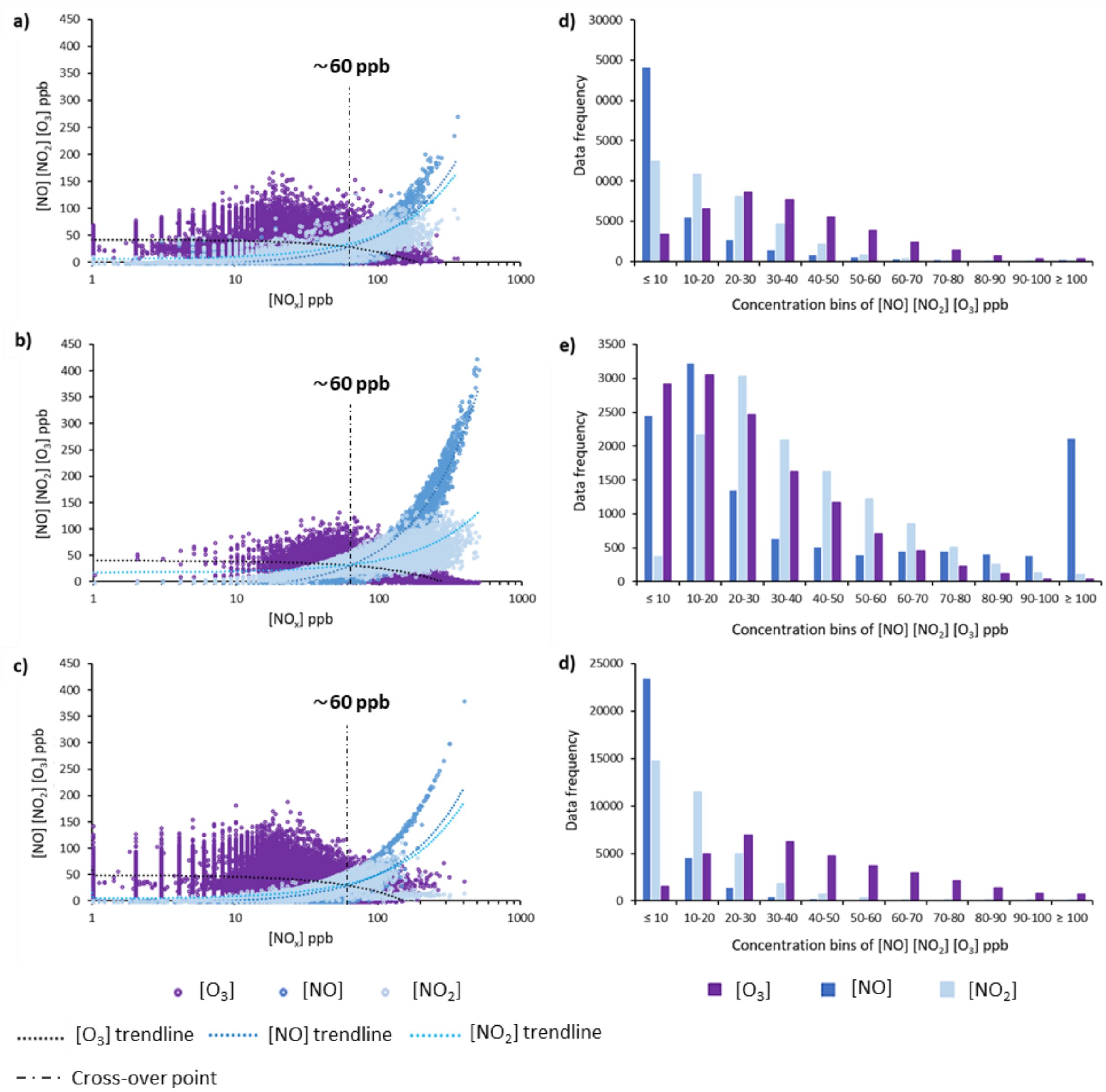

Fig. 5: Relationships and crossover points of NO, NO_2 and O_3 at a) BKK sites b) roadside sites and c) BKK suburb sites; and concentration distributions of those species at d) BKK sites e) roadside sites and f) BKK suburb sites.

4) We have now removed Figure V in the supplement and replaced it by a table that provides the correlations between CO and NO_x; and SO_2 and NO_x (Table III). We have also provided the correlation among the species at all the monitoring sites in this table. Table III provides comprehensive statistical information.

Table III: Correlation between CO and NO_x; and SO_2 and NO_x at BKK sites, roadside sites and suburb sites, during 2010 to 2014; together with ±1 standard deviation.

Station type	Station ID	Correlation	
		CO and NO_x	**SO_2 and NO_x**
BKK sites	**3T**	0.76	0.34
	5T	0.56	0.37
	10T	0.76	0.36
	11T	0.68	0.33
	12T	0.61	0.26
	15T	0.64	0.29
	61T	0.85	0.28
	Average	0.69±0.10	0.32±0.04
Roadside sites	**52T**	0.73	0.49
	54T	0.72	0.56
	Average	0.72	0.53
Suburb sites	**13T**	0.92	0.32
	14T	0.64	0.11
	19T	0.47	0.39
	20T	0.55	0.21
	22T	0.71	0.27
	27T*	0.77	0.53
	Average	0.68±0.16	0.30±0.15

Note: *the correlations are calculated based observations during 2010 to 2013

Some other concerns:

1. Gaseous pollutants in the title is still too broad a domain for a study reporting only O_3, CO, NO_x and SO_2.

Authors' response: Thank you. We have now modified the title from "Assessment of Gaseous Pollutants in Bangkok Metropolitan Region, Thailand" to "Assessment of Gaseous **Criteria** Pollutants in Bangkok Metropolitan Region, Thailand"

2. The details of calibrations are still not provided. Given the long measurement period reported in this study, it is very important to know how the instrument response drifted over time.

Authors' response: Thank you. According to a document of the Pollution Control Department, Thailand (PCD): term of reference (TOR) for air quality detectors and air quality monitoring stations in the notification of PCD, Number 17/2559, date November 17, 2016.

Detector details:

SO_2 detectors:

range: 0-500 ppb to 0-20 ppm with auto ranging or better.
lower detection limit: < 1 ppb
precision: 0.5 ppb or < 1% of reading or better
zero drift: < 1 ppb/24-hour

span drift: < 1% of reading/ 24-hour

NO_x detectors:

range: 0-500 ppb to 0-20 ppm with auto ranging or better.
lower detection limit: < 0.5 ppb
precision: 0.5 ppb or < 1% of reading or better
zero drift: < 1 ppb/24-hour
span drift: < 1% of full scale/ 24-hour

CO detectors:

range: 0-50 ppm to 0-200 ppm with auto ranging or better.
lower detection limit: < 0.05 ppm
precision: < 1% of reading or better
zero drift: < 0.1 ppm/24-hour
span drift: < 1% of reading/ 24-hour

O_3 detectors:

range: 0-500 ppb to 0-10 ppm with auto ranging or better.
lower detection limit: < 0.6 ppb
precision: 1% of reading or better
zero drift: < 1 ppb/24-hour
span drift: < 1% of reading/ 24-hour

Detector/ data loggers/ air inlets calibration/ maintenance:

single point calibration for detectors: every 15 days
multi-point calibration with 3 span levels (20 %, 40 % and 80 %): every 90 days
mass flow adjustments: every 90 days
molybdenum converter for NO_2 detectors: at least 4 times in 730 days
zero air generators: at least 4 times in 730 days
data loggers maintenance: every 15 days
air inlets maintenance: every 15 days

Acceptance data criteria:

1. Span drifts
   span drift: < ± 10 % of full scale for NO_2, SO_2, CO detectors
   span drift: < ± 7 % of full scale O_3 detectors
2. Zero checks
   zero drift: < ± 5 ppb for NO_2, SO_2 and O_3 detectors
   zero drift: < ± 0.4 ppm for CO detectors

We have now included this information in section A, the supplement material.

3. Several major conclusions are drawn from poor correlation.

3.1 Section 3.3. I am not convinced by the PSS analysis performed by the authors in the revised manuscript. Apart from the method by Trebs et al. (as suggested in the first review), authors could have used NCAR TUV model for calculation of j1. Even in the polluted environment like in Delhi, deviation from PSS was observed at NO_x values more than 10 ppb (Chate el al 2014). At such high NO_x concentration, systematic deviation from PSS with Leighton ration less than 1 was observed.

Value of Leighton ratio =1 is a very rare finding in ambient environment. Hence, I again question the validity of conclusion drawn on this assumption. I again ask the authors to calculate j1 using TUV model or using solar radiation and check the Leighton ratio. In any case, given that j1 only depends on actinic flux, quantum yield and absorption cross-section, how would the authors explain a variation of more than an order of magnitude during the daytime hours of the same season (line 210 of the revised manuscript). Chate, D. M., et al. (2014), Deviations from the O3NONO2 photo-stationary state in Delhi, India, Atmospheric Environment, 96(0), 353-358, doi:http://dx.doi.org/10.1016/j.atmosenv.2014.07.054.

Authors' response: Thank you. As suggested by the reviewer, we have calculated the j_1 values using the NCAR Tropospheric Ultraviolet and Visible (TUV) Radiation model for 2010. We have used the missing information from scientific published values for the air quality monitoring stations. Those variables are

1. Overhead O_3 column in Dobson unit. The data is retrieved from National Aeronautics and Space Administration (NASA) website (https://ozoneaq.gsfc.nasa.gov/tools/ozonemap/) at the latitude and longitude of 13.76 ˚N and 100.50 ˚E (Bangkok, Thailand location).

2. Surface albedo. The data is retrieved from Janjai, S., Wanvong, W., and Laksanaboonsong, J.:The Determination of Surface Albedo of Thailand Using Satellite Data, The 2nd Joint International Conference on Sustainable Energy and Environment (SEE 2006), 21-23 November 2006, Bangkok, Thailand.

3. Cloud optical depth. The data is retrieved from NASA Earth Observations (NEO) (https://neo.sci.gsfc.nasa.gov/view.php?datasetId=MYDAL2_M_CLD_OT).

4. Aerosol optical depth and single scattering albedo (SSA). The data is retrieved from Janjai, S., Nunez, M., Masiri1, I., Wattan, R., Buntoung, S., Jantarach, T., and Promsen, W.: Aerosol Optical Properties at Four Sites in Thailand, Atmospheric and Climate Sciences, 2, 441-453, 2012.

The rate coefficients are calculated in 2010 for the dry season (January, February, March, April, May, October, November and December), during 10:00 LT to 16:00 LT, at the latitude and longitude of 13.7 ˚N and 100.5 ˚E. The j_1 values calculated from the NCAT TUV model are now shown in section F, supplement material in Table II.

Table II: Statistical analysis of the chemical rate coefficients (j_1 and k_3) based on an observational analysis during dry seasons at BKK sites, roadside and BKK suburb sites, 2010 to 2014.; and statistical analysis of j_1 based on a modeling analysis at the latitude and the longitude of 13.7 ˚N, 100.5 ˚E in a dry season, 2010.

Sites	Rate coefficient											
	j_1						k_3					
	min^{-1}			s^{-1}			ppm^{-1} min^{-1}			cm^3 $molecule^{-1}$ s^{-1}		
	Max	Min	Average	Max	Min	Average	Max	Min	Average	Max	Min	Average
Based on observation*												
BKK	0.95	0.12	0.74±0.2	0.016	0.004	0.008±0.035	30.9	28.6	29.8±0.7	$2.06e^{-14}$	$1.99e^{-14}$	$2.02e^{-14}$±$2.01e^{-16}$
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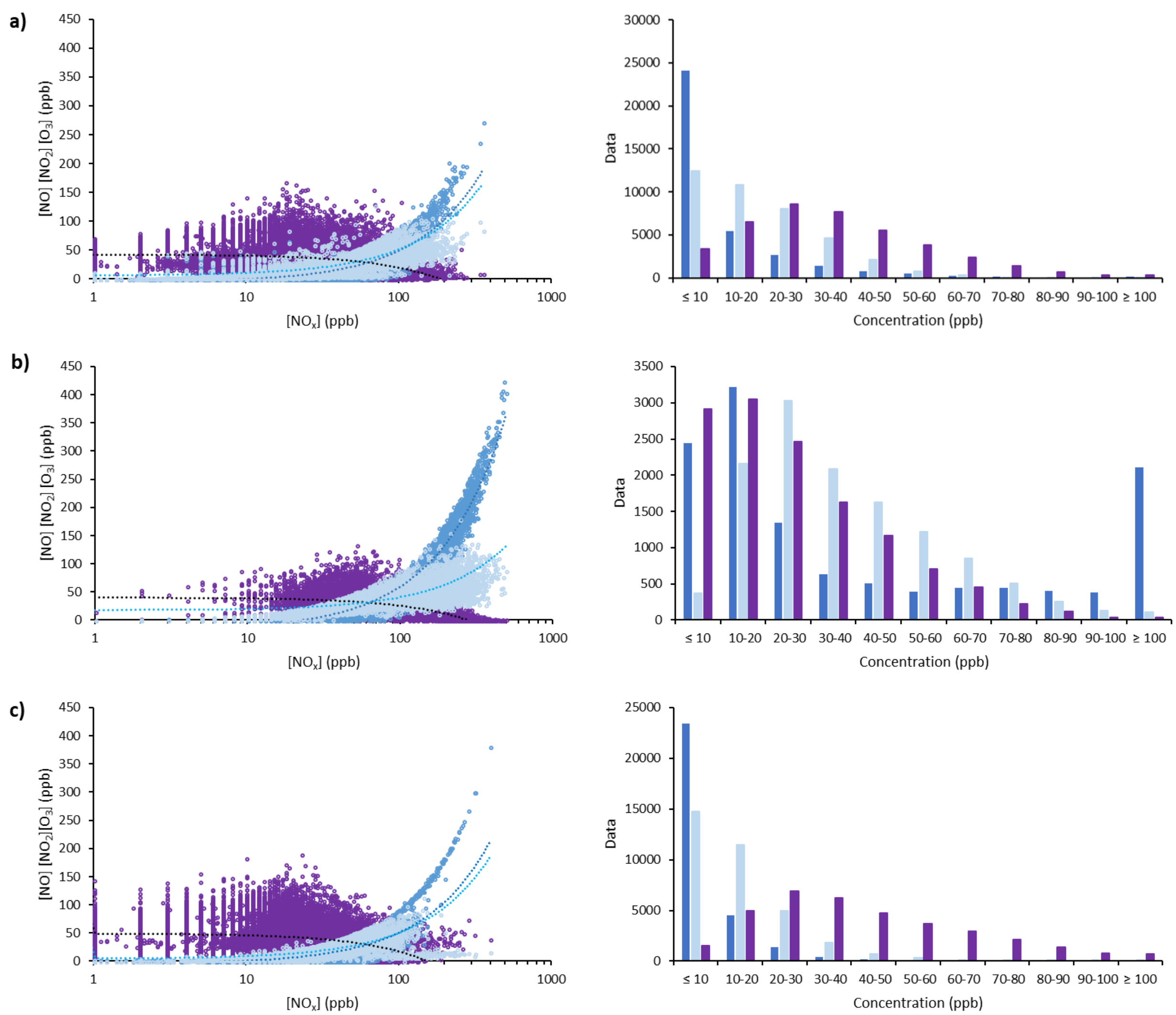
BKK suburb	1.22	0.34	0.55±0.3	0.022	0.007	0.010±0.004	30.9	28.8	29.8±0.7	$2.04e^{-14}$	$2.01e^{-14}$	$2.03e^{-14}$±$1.34e^{-16}$
Based on modeling**												
13.7 ˚N, 100.5 ˚E						0.021±0.002						

The average j_1 value calculated from the NCAR TUV model is 0.021±0.0024 s^{-1}, which is similar to our calculations based on observations in Table I of the manuscript (j_1 ranges from 0.008 to 0.013 s^{-1}).

The manuscript now includes the comparison of the j_1 result from the NCAR TUV model with our calculation. We are very encouraged by the similarity of the two results.

3.2 Cross over point and regime identification: First of all, legends are not provided in this figure 5. If I assume the purple points to be O_3, still the fit statistics (which are not even provided either in text or in figure) are very poor. So the conclusion drawn regarding cross over points are not robust. There is no clear crossover point for the BKK sites.

Authors' response: Thank you. We have modified Figure 5 by providing legends and clarity on the cross-over points.
Figure 5 is changed from:



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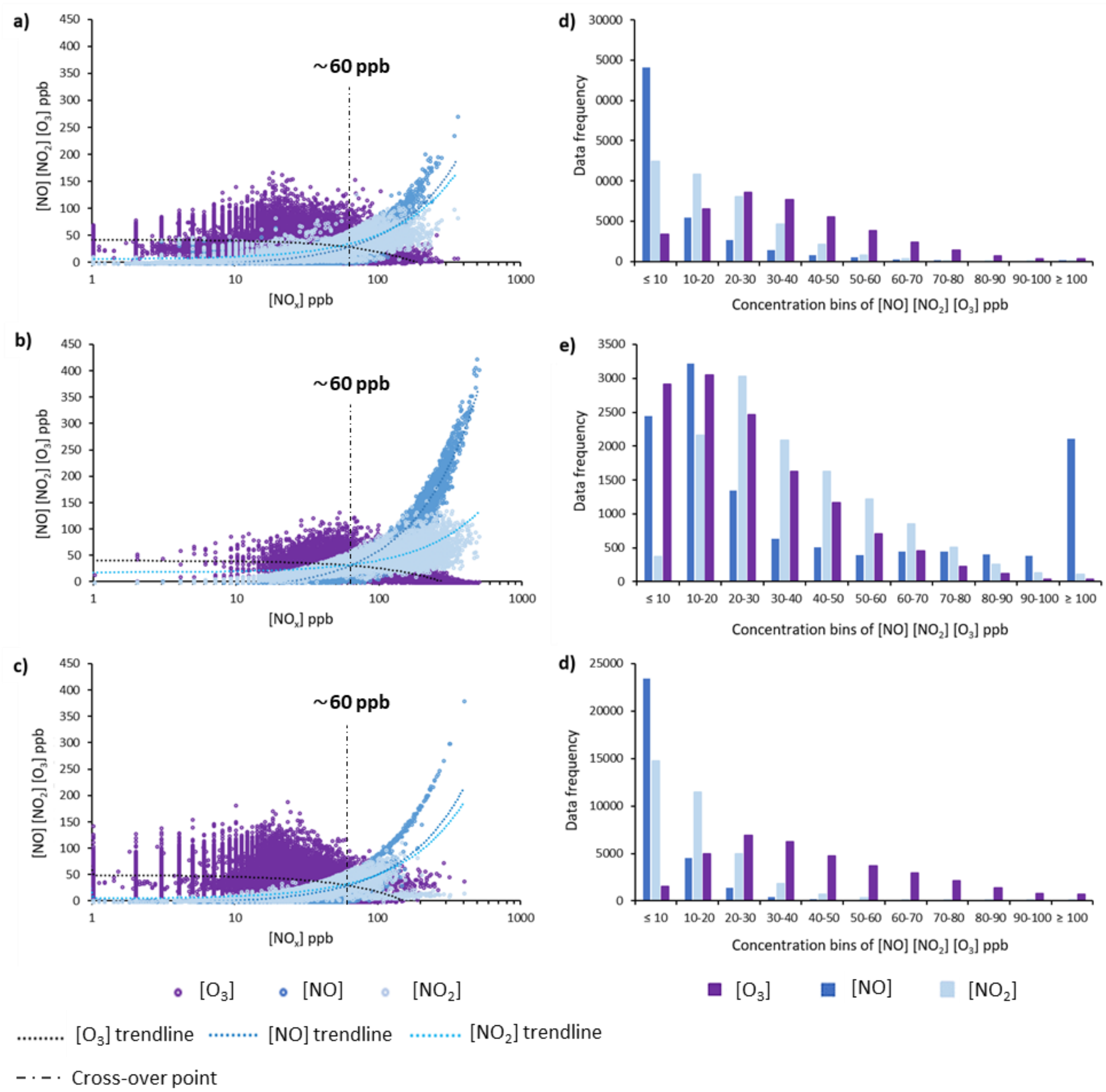

Fig. 5: Relationships and crossover points of NO, NO_2 and O_3 at a) BKK sites b) roadside sites and c) BKK suburb sites; and data distributions of those species at d) BKK sites e) roadside sites and f) BKK suburb sites.

3.3 Section 3.4: The scatter plots have very poor fit for Fig 6a and Fig 6c for the non-episode events. In addition to the slope and intercept, authors should also consider the goodness of fit before drawing any conclusion.

Authors' response: Thank you. This is a very large (2010 to 2014) and robust air quality data set. Figure 6 provides the best linear regression lines during O_3 episodes and non-episodes condition and its relationship to the O_3 precursor NO_x. This has also been articulated by reviewer#2.

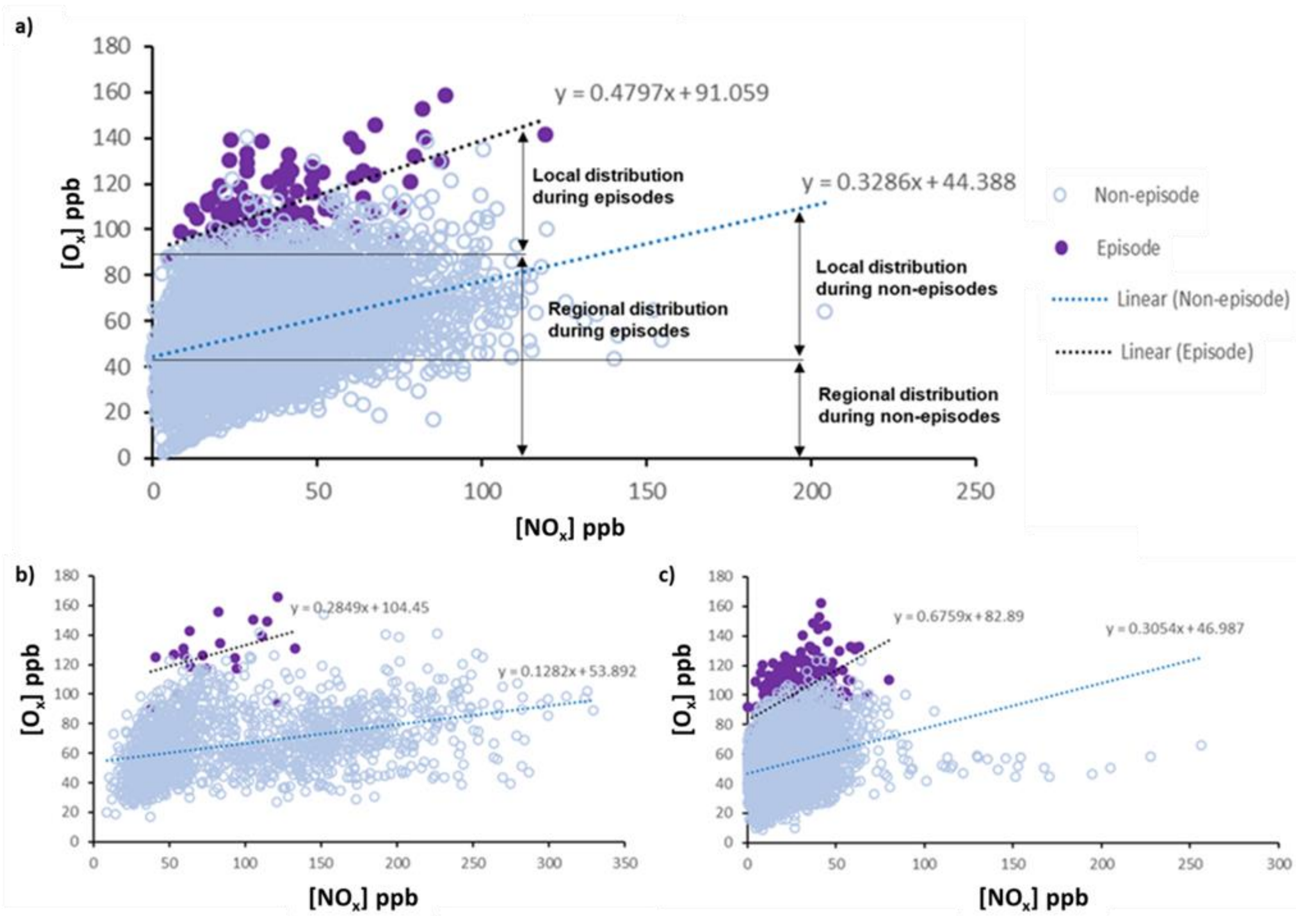

Fig. 6: Effects of local and regional contributions on O_x during non-episode and episode days at a) BKK sites, b) roadside sites and c) BKK suburb sites.

3.4 Section 3.5.1 (Figure V of the supplement): Even in the best case, the r2 is less than 0.3 in the best case. What is the significance of local source analysis based on such poor statistics? Why the frequency distribution of SO2 (I assume it is frequency distribution has no information is provided either in figure caption or text) has wiggles in between.

Authors' response: We have now removed Figure V in the supplement and replaced it by a table that provides the correlations between CO and NO_x; and SO_2 and NO_x (Table II). We have also provided the correlation among the species at all the monitoring sites in this table. Table II provides comprehensive statistical information.

Table III: Correlation between CO and NO_x; and SO_2 and NO_x at BKK sites, roadside sites and suburb sites, during 2010 to 2014; together with ±1 standard deviation.

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	11T		0.68	0.33
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	15T		0.64	0.29
	61T		0.85	0.28
		Average	0.69±0.10	0.32±0.04
Roadside sites	**52T**		0.73	0.49
	54T		0.72	0.56
		Average	0.72	0.53
Suburb sites	**13T**		0.92	0.32
	14T		0.64	0.11
	19T		0.47	0.39
	20T		0.55	0.21
	22T		0.71	0.27
	27T*		0.77	0.53
		Average	0.68±0.16	0.30±0.15

Note: *the correlations are calculated based observations during 2010 to 2013

3.5 Lines 265-272: The statistics are too poor for the conclusion of ~10 ppb enhancement in O_3. The spread in delta O_3 ranges from -66 to +96 ppb.

Authors' response: Thank you. The delta O_3 analysis for Atlanta Metropolitan Region has been published (Lindsay and Chameides, 1988; Lindsay et al., 1989). This reference is provided in the manuscript. As discussed in the manuscript, ~10 ppb enhancement in O_3 for BMR is the average for the observation. These results are similar to Lindsay and Chameides, 1988 and Lindsay et al., 1989.

Reviewer#2

We thank the reviewer for the thoughtful reviews and comments. We are please that "I am writing to you that I accept all revisions to the comments and suggestions that I made to the manuscript". Moreover, we are pleased that the reviewer rates the manuscript as "Excellent" for the three categories including Scientific significance, Scientific quality and Presentation quality.