#### **Response to Referee #1**

# We truly appreciate the comments and suggestions from the referee. Incorporation of the referee's comments has greatly improved our paper. Below is a detailed response to the comments. The referee's comments are in regular font and our responses are in bold font.

#### **General Comments**

This manuscript applies five different anthropogenic emission inventories with WRF/Chem to examine surface CO and O3 for Southeast Asia. The authors compared the simulations with observation data, and concluded that none of the emission inventories are better than the others. While most current studies have focused on East and South Asia, this manuscript provides some insights about Southeast Asia. The underlying work in this manuscript will be a useful contribution to the literature. While this manuscript provides detailed comparisons and long discussion, I suggest the authors summarize the major innovations besides the study region in the introduction sections to emphasize its significance and give the readers a sense of "big picture".

We have added a few sentences summarizing the major innovations besides the study region. These sentences have been added to the Introduction in the fourth paragraph, which now reads as the following.

"While previous studies (e.g. Ohara et al., 2007) have compared different emission inventories, a comparison of simulated surface CO and O<sub>3</sub> mixing ratios resulting from different emission inventories, yet using the same model framework, has not been done. Here, the Weather and Forecasting Model coupled with Chemistry (WRF-Chem) is used to examine the variability of predicted O<sub>3</sub> and CO surface mixing ratios when five different anthropogenic emission inventories (RETRO, INTEX-B, MACCity, SEAC4RS and a modified SEAC4RS) are used as inputs. By conducting this comparison using the same meteorology and chemical mechanism, differences in results due to model meteorology and chemical mechanism are mitigated. We focus this study on Southeast Asia, an area that has received little attention, yet has substantial anthropogenic and biomass burning emissions. As part of our study, we examine the effect of biomass burning emissions on surface O<sub>3</sub> and CO by contrasting results from a low biomass burning period (December) with a high biomass burning period (March)."

#### Specific Comments

Section 3.1, for each emission inventory, it is better to state whether it includes shipping emissions, whether it includes non-combustion sources, and whether it includes four sectors (residential, industry, power and transport). I feel that such kind of clarification will make the comparison in section 3.2 more meaningful, and show the readers whether these emission inventories have consistent emission sources

This is a very good suggestion. We have added a table (below) to clarify what activities are part of each emissions inventory. Further, we have added text to the manuscript (Section 3) describing which emission sectors are missing from specific emissions inventories.

						/	5
	RETRO		INTEX-B		MACCity		SEAC4RS
1.	Power	1.	Power	1.	Energy production	1.	
	Generation		Generation		and distribution		Generation
2.	Residential	2.	Industry	2.	Industry (combustion	2.	Industry
3.	Industrial		(combustion		and non-combustion)		(combustion
	combustion		and non-	3.	Land transport		and non-
4.	Industrial		combustion)	4.	Maritime transport		combustion)
	processes	3.	Residential	5.	Aviation	3.	Residential
5.	Extraction distribution	4.	Transportation <sup>a</sup>	6.	Residential and commercial	4.	Transportation <sup>b</sup>
	of fossil fuels			7.	Solvents		
6.	Solvent use			8.	Agriculture		
7.	Road			9.	Agricultural waster		
0	transport			10	burning on fields		
8.	Other mobile sources <sup>c</sup>			10	Waste		
9.	Waste						
	treatment and						
	disposal						
10	Agriculture and Landuse						
	change						

Table 1. Emission sectors used in the model simulations from each emission inventory.

<sup>a</sup>Transportation for INTEX-B includes road, railways, aviation, and maritime transportation.

<sup>b</sup>Transportation for SEAC4RS includes road, railways, and aviation transportation.

<sup>c</sup>Other mobile sources for RETRO include aviation and maritime transportation.

Line 15 on page 9352: can the authors give more details about "what was developed for Europe"? It is not quite clear how the RETRO seasonal cycle is developed.

The final report on the RETRO emissions (Schultz et al., 2005) states that the European monthly emissions used what was developed in the LOTUS-EUROS model. The LOTUS-EUROS documentation (Schaap et al., 2005) states that the monthly emission factors are derived from a critical review of what these factors should be for each emission sector and give a table listing these factors. We have added this information to Section 3.1.

Lines 23-26 on page 9353: be specific about "other emission inventories". How can the emission inventory in 2000 be used to make the conclusion about inventory in 2010? It is better to provide more references.

In the noted sentence, the "other emission inventories" are RETRO, INTEX-B and SEAC4RS. We have clarified this in the text. We have also added more information regarding the significant biases of MACCity emissions that was discussed in Lamarque et al., 2010. The new text is as follows.

"Lamarque et al. (2010) did not find significant biases in their comparison of 2000 MACCity emissions with published emission estimates (e.g. RETRO and EDGAR). However, they estimate that these emissions have an uncertainty of about a factor of 2 based on Bond et al. (2004, 2007) and Smith et al. (2010). The uncertainty of the 2010 emissions was not reported."

Line 18 on page 9354: be specific about ship emission. Does it include both international and domestic shipping?

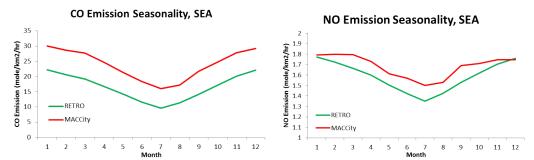
For RETRO and INTEX-B emissions, only international shipping emissions are included. MACCity emissions include international shipping, domestic shipping and fishing, but exclude military vessels. We have clarified this in the text.

"In addition to the four inventories described above, we have conducted a simulation with a combined MACCity/SEAC4RS emissions inventory, which replaces MACCity with the SEAC4RS emissions over Asia yet includes the MACCity ship emissions, which include international shipping, domestic shipping and fishing. Note, that in the RETRO and INTEX-B inventories, ship emissions represents only international shipping."

Lines 24-25 on page 9354: as the authors introduced, "both the RETRO and MACCity emission inventories have monthly temporal variability", why only RETRO is chosen to estimate monthly emissions in INTEX-B and SEAC4RS. How much uncertainty would be introduced to the final simulations with the assumption that these three emission inventories follow the same monthly allocation? Besides, will this assumption affect the following discussion that different emission inventories make little variation in modeled surface mixing ratios?

When comparing the seasonality of RETRO CO emissions with MACCity emissions (Figure below), it is evident that they have very similar temporal trends. Thus, we do not expect the choice of RETRO for CO emission seasonality to contribute to the uncertainties in the results. When comparing the seasonality of RETRO NO emission with MACCity emissions, the seasonality is not as similar as it was for CO emissions. There is a bigger difference in March NO emissions compared to February, and a bigger difference in November compared to December. These differences could introduce uncertainties in the  $O_3$  results for the region (which is mostly  $NO_x$ -limited). Therefore we have added the following sentences to the discussion on seasonality of emissions (Section 3.1).

"The MACCity seasonal variation is very similar to RETRO for CO emissions, but does differ somewhat for NO emissions. The change of NO emissions from February (when we start the simulation) to March differs between these two inventories with little change in high NO emissions for the MACCity inventory and a 5% decrease in NO emissions for the RETRO inventory. While this is a small difference, the change in NO emissions could affect O<sub>3</sub> production downwind of NO sources."



**Figure.** Seasonality of CO (left panel) and NO emissions (right panel) for Southeast Asia from the RETRO (green) and MACCity (red) emission inventories.

Equations (1)-(3): is the monthly estimate based on each grid or total emissions of over the entire model domain? What is the difference between "monthly emissions" defined here and "monthly-average emissions" defined in line 10, page 9366?

The monthly estimate was based on each grid. Monthly emissions are the emission data for each month for entire year. We apologize for the poor wording. We have replaced "The monthly-average" with "The monthly emission".

Lines 13-14 on page 9355, how to make the conclusion that biomass burning sources dominate the emissions of NOx in March? Based on Table 1, NO emission from biomass burning is less than that from anthropogenic sources.

Thank you for pointing this out. We have now changed the sentence to "In March, the biomass burning sources dominate the emissions of CO.".

Lines 1-3 on page 9356, from RETRO in 2000 to SEAC4RS in 2012, there should be emission reductions.

Thank you for pointing this out. We have now revised the sentence to contrast RETRO emissions to MACCity/SEAC4RS emissions because of the lack of ship emissions in the SEAC4RS-only emissions. The sentence has been changed to "By comparing RETRO emissions to MACCity/SEAC4RS emissions, the total anthropogenic emissions in Southeast Asia decreased by ~30% for CO and ~13% for NO<sub>x</sub> between 2000 and 2012 with 2010 ship emissions.

Lines 4-13 on page 9356, authors made comparison between different emission inventories, and mentioned that less NO emissions in SEAC4RS are due to lack of ship emissions. How much contribution do ship emissions make to the total emissions? Is it possible that some of the emissions have been reduced by applying emission control technologies?

On page 9354 lines 16-20, we said that the contribution from ship emission accounts for 15% of the NO emission and 0.1% of the CO emissions. Yes, it is possible that applying

emission control technologies has reduced some of the emissions. However, the SEAC4RS emission inventory does not include any ship emissions, which is the point of the sentence.

Lines 14-29 on page 9356, suggest authors show emissions in Ohara et al. (2007) and REAS v2.1 inventory in Table 1. It is hard to follow this paragraph without any numbers.

Thank you for the suggestion. We now include the emission estimates from REAS v1 in Table 1 (now Table 2). The estimate in the table comes from the Emissions of atmospheric Compounds & Compilation of Ancillary Data (ECCAD) web site (<u>http://eccad.sedoo.fr</u>) in order to obtain emission estimates for the same region as our model domain. We then relate those numbers to those reported in Table 6 of Ohara et al. (2007) so that we can compare the WRF-Chem emissions with the TRACE-P and EDGAR3.2 emission estimates for the Southeast Asia region. The text has been modified to include emission rates in the discussion to the following.

"The CO and NO emissions used in our study are larger than the REAS v1 emissions (Ohara et al., 2007) for our modeling domain (Table 2). The REAS v1 estimate in Table 2 comes from the Emissions of atmospheric Compounds & Compilation of Ancillary Data (ECCAD) web site (http://eccad.sedoo.fr) to obtain emission estimates for the same region as our model domain, which encompasses small regions of India and China that are not included in the Southeast Asia region denoted by Ohara et al. (2007). For our model domain the REAS v1 annual emissions are 91.4 Tg year<sup>-1</sup> for CO and 4.81 Tg year<sup>-1</sup> for NO<sub>x</sub>. For the Southeast Asia region, Ohara et al. (2007) report in their Table 6 annual CO and NO<sub>x</sub> emissions of 54.5 and 3.77 Tg year<sup>-1</sup>, respectively, but these exclude international aviation, international shipping and open biomass burning. The REAS v1 emissions are even greater than the TRACE-P, EDGAR 3.2, and IIASA CO emissions (34.0, 42.6, 39.8 Tg CO year<sup>-1</sup>, respectively) but are more similar to TRACE-P, EDGAR 3.2, and IIASA NOx emissions (3.06, 3.91, 3.94 Tg NO<sub>x</sub> year<sup>-1</sup>, respectively) for Southeast Asia (Ohara et al., 2007) as well as REAS v2.1 (Kurakawa et al., 2013), which were 36.2 Tg CO year<sup>-1</sup> and 3.00 Tg NO<sub>x</sub> year<sup>-1</sup>. Thus, the emissions used here are larger than the REAS emissions inventories as well as other previous inventories."

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	E_CO (mo	ole km <sup>-2</sup> hr <sup>-1</sup> )	E_NO (mole km <sup>-2</sup> hr <sup>-1</sup> )		
<b>Emission Inventory</b>	March	December	March	December	
<b>RETRO – 2000</b>	410,840	496,860	30,590	39,320	
INTEX-B – 2006	396,170	406,240	27,410	29,640	
MACCity – 2010	436,750	454,250	27,440	28,280	
MACCity/SEAC4RS	319,420	320,310	29,810	30,910	

Table 2. Summation of CO emissions and NO emissions (mole km<sup>-2</sup> hr<sup>-1</sup>) from all grids in the model domain for each month.

SEAC4RS – 2012	305,542	300,369	16,610	17,290
<b>Biomass Burning – 2008</b>	717,940	58,780	10,220	700
<b>REAS v1<sup>a</sup> – 2000</b>	282	,120	13,8	328

<sup>a</sup>REAS v1 emissions are from the ECCAD web site (<u>http://eccad.sedoo.fr</u>) and are the annual emissions converted to hourly emissions assuming constant emissions for the year over the WRF-Chem model domain.

Section 5.1, Figure 5 and 6, it is difficult to make comparison with current figures, I suggest the authors to add figures about the differences between modeling and observation.

Following the reviewer's suggestion, we added panels to Figure 5 and 6 to show the differences between model and observation (see Figures below). However, we are concerned that when we do this the panels get smaller and are harder to read. Further, we think that the qualitative differences between WRF and observations are pretty clear in the original Figures 5 and 6. Therefore, we have decided to use the figures that we originally submitted.

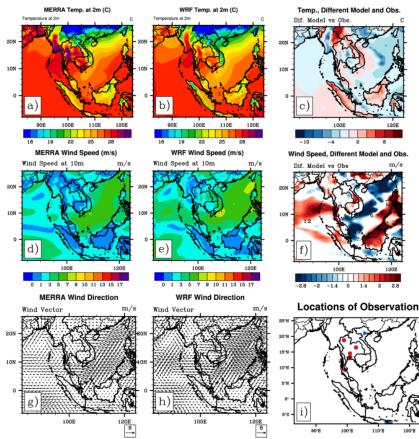
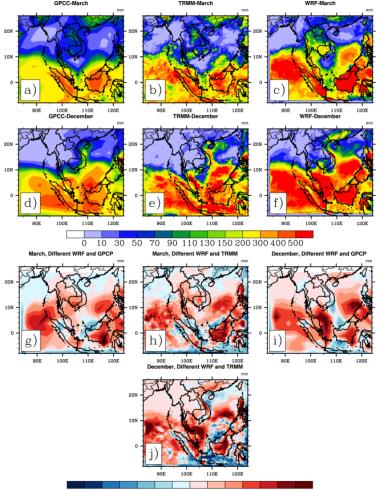


Figure 5. March 2008 monthly-averaged (a) 2 m temperature from MERRA, (b) 2 m temperature from WRF, (c) difference between (a) and (b), (d) 10 m wind speed from MERRA, (e) 10 m wind speed from WRF, (f) difference between (d) and (e), (g) 10 m wind direction from MERRA, and (h) 10 m wind direction from WRF. (i) Locations of ground-based CO and O3 measurements and ozonesonde sites.



-500400300200100-50 0 50 100 200 300 400 500

Figure 6. Accumulated precipitation (a) GPCC, March, (b) TRMM, March, (c) WRF, March, (d) GPCC, December, (e) TRMM, December, (f) WRF, December. Differences between (g) WRF and GPCC for March, (h) WRF and TRMM for March, (i) WRF and TRMM for December, and (j) WRF and TRMM for December.

Lines 3-5 on page 9362, how to separate the influence of biomass burning and anthropogenic emissions?

We determine the influence of biomass burning versus anthropogenic emissions by comparing model results from March (high biomass burning emissions) to those from December (low biomass burning emissions). We have reworded the text at these lines to clarify this approach to the following.

"By comparing the model results from March (high biomass burning emissions) to those from December (low biomass burning emissions), the influence of biomass burning emissions can be seen for all three species. CO mixing ratios are > 500 ppbv over Burma and northern Thailand during March compared to 200-500 ppbv during December." Lines 11-19 on page 9362, what is the definition of "variation" in the predicted monthly-average surface mixing ratios across the five simulations? The authors listed ranges of variation for difference pollutants, but what kind of information we are supposed get from the values of variation? What cause the variations? While the high variations of NOx were explained by differences in ship emissions, what are the reasons for high variations over land?

We apologize for not defining "variation" in the manuscript. It is the standard deviation of the 5 simulations. This is now defined in the text when we introduce the variation discussion. Because the meteorology and biomass burning emissions are the same among the 5 simulations, the primary cause for the variations are the differences in the anthropogenic emissions. As part of this, missing sectors in the emission inventories or different emissions for different years contribute to the variations. We have modified the text to bring out these causes to the following.

"The variation, which is defined as the standard deviation of the five simulations, in the predicted monthly-averaged surface mixing ratios of CO, O<sub>3</sub>, and NO<sub>x</sub> across the five simulations is highlighted in Figure 8. Because we conducted each simulation with the same meteorology and biomass burning emissions, the primary cause for the variations are the differences in the anthropogenic emissions. CO mixing ratios vary across simulations by <20%, but variations of ~30-60% are found near Bangladesh and Indonesia for both March and December. O<sub>3</sub> mixing ratios have up to 30% variation near the tip of the Malaysian peninsula and near Indonesia, but have much smaller variability elsewhere. Mixing ratios of NO<sub>x</sub> have the most variation among the simulations. The 70-100% variations for NO<sub>x</sub>, especially over the South China Sea, are from the differences in ship emissions from each inventory. There are also high NO<sub>x</sub> variations in several cities as seen by the locally high values in Fig. 8e,f due to different emission strengths in each inventory and to missing emission sectors in some inventories (e.g. shipping emissions in the SEAC4RS inventory)"

#### Lines 23-24 on page 9362, Saraburi is missing in this sentence.

Thank you for pointing this out. We have now modified the sentence to "The 6-hourly daytime (00:00, 06:00, 12:00 UTC) CO mixing ratios from WRF-Chem with each of the five inventories are compared to observations from the six ground-site measurements: Chiang Mai (CM) in northwest Thailand, Khonkaen (KK) in eastern Thailand, Nonthaburi (NTB) in the Bangkok metropolitan region, Sarabuti (SRB) just north of Bangkok, Chonburi (CB) southeast of Bangkok, and Suratthani (SRT) in the southern peninsula (Fig. 5g)."

Fig. 9 (g)-(l) and Table 3, if I understand it correctly, figures show that the model underpredicts most of December CO except some points at Chiang Mai and Saraburi. But Table 3 only shows positive bias at Chonburi, and the authors stated that higher emissions are modeled at Chonburi and Suratthani (line 8 page 9363). The authors are better to clarify these statements and explain the underestimates of CO (in both March and December) by model simulations.

We have now changed the sentence and elaborated on explaining the underprediction of CO to "In December, the predicted 6-hourly daytime surface CO for all simulations is

much less than the observations, with the exception of the Chonburi site. The large underprediction is reflected by the bias calculation (Table 4). Part of the underprediction is a result of the coarse model resolution (36 km), which cannot capture the highly variable emissions and high CO concentrations in an urban setting where the measurement site is located. However, the underprediction of CO could also be due to low anthropogenic emissions (discussed further in Section 5), a high planetary boundary layer height, which would cause dilution of surface mixing ratios, and/or missing chemistry in the model such as heterogeneous chemistry (Mao et al., 2013)."

Fig. 10 and 11 (also Fig. 14 and 15), I suggest the authors plot the ratios between observations and model simulations, instead of absolute values.

We think differences in the spatial patterns between observations and model are sufficiently depicted in the original figures, which allow us to use the same spatial resolution provided in the original data or model output. Thus, we prefer using the submitted figures for our paper.

Lines 3-8 on page 9366, I am curious why the model predict different peak values and regions from observations? Also why all five simulations predict relative low NO2 column (lines 15-17) over Burma in December?

For March, the WRF-Chem  $NO_2$  column mostly reflects the biomass burning emissions pattern (Figure 3), while for December WRF-Chem is more similar to the anthropogenic emissions (Figure 4). The OMI  $NO_2$  column does not show the high  $NO_2$  over northern Thailand and Burma where the model has high biomass burning emissions in March. To explain this difference, WRF-Chem fire emissions could be too high, or OMI may miss high  $NO_2$  because of cloud interfering with the instrument's view. In situ measurements would allow us to evaluate better the performance of the model. The anthropogenic emissions in Burma are lower than the surrounding regions and are possibly too low. We have added this discussion to the text.

Lines 8-11 on page 9366, it seems that shipping emissions explain a lot of variations among model simulations with different inventories (also lines 1 and 10 on page 9356, line 18 on page 9362, and line 22 on page 9365). I am wondering whether it is necessary to separate shipping emissions in each inventory and show them in Table 1.

The shipping emissions (listed in the Table below) for our model domain from each inventory do vary, especially between RETRO and the other two inventories. Shipping emissions are important in near coastal regions. For example, Huszar et al. (2010) ACP found that the contribution of ship-induced surface  $NO_x$  to the total  $NO_x$  is 10-30% in the North Atlantic coastal regions. In addition to adding ship emissions to Table 1, we have modified the text in the  $NO_2$  evaluation section to the following.

"The largest variation ... a result of both low  $NO_2$  mixing ratios from the MOZART boundary conditions and different estimates for shipping emissions among the different inventories (Table 2, Figs. 3 and 4). Both the RETRO ship emissions, which are 75-80% smaller than INTEX-B and MACCity ship emissions, and the SEAC4RS only simulation, which does not have ship emissions contribute to the variation. When the MACCity ship emissions are combined with the SEAC4RS emissions (MACCity/SEAC4RS), the agreement with OMI NO<sub>2</sub> column is much better than the SEAC4RS only simulation.

		E_CO (mo	$le km^{-2} hr^{-1})$		E_NO (mole km <sup>-2</sup> hr <sup>-1</sup> )			
	March		December		March		December	
Emission Inventory	Total Ship		Total	Ship	Total	Ship	Total	Ship
RETRO – 2000	410,840	3,404	496,860	3,364	30,590	5,097	39,320	5,186
INTEX-B – 2006	396,170	5,888	406,240	5,785	27,410	3,273	29,640	3,301
MACCity – 2010	436,750	3,569	454,250	3,717	27,440	3,980	28,280	5,138

Table. Total and shipping CO and NO emissions for the model domain.

Section 5.2.2 and 5.2.3, the authors compared monthly-average correlation coefficients and biases across model results with five different emission inventories, and then concluded that "none of the anthropogenic emission inventories are better than the others" (lines 25-26 on page 9346). I am wondering whether it is possible to do some paired difference tests and show the significance level if the modeled results are independent on the type of emission inventory?

Thank you for the suggestion. Doing paired difference tests will strengthen our conclusions. We apply Kruskal-Wallis non-parametric test, which is used for comparing more than two samples that are independent, or not related, to analysis whether there is significance level for each model simulation or not. The p-values from each simulation from different emission inventory were shown in table below. The p-values of ozone are greater than 0.05, so there is no statistical significant difference between each data set for both March and December (Table below). There is a statistically significant difference between model and observations for CO at some sites in both March and December (Table below). For example Khonkaen, Saraburi, Nonthaburi and Chonburi have p-values < 0.05, while Chiang Mai CO has a statistically significant difference only in December. Suratthani, which is further from urban emissions, is the only site where there is no statistical difference for CO.

After doing this statistical analysis, we have modified our conclusion and abstract to state the following. "None of the anthropogenic emission inventories are better than the others for predicting  $O_3$  surface mixing ratios. However, the simulations with different anthropogenic emission inventories do differ in their predictions of CO surface mixing ratios producing variations of ~30% for March and 10-20% for December at Thai surface monitoring sites.

Species	es CM		K	K	SRB		NTB		CBR		SRT	
	Mar	Dec	Mar	Dec	Mar	Dec	Mar	Dec	Mar	Dec	Mar	Dec
Ozone	0.846	0.409	0.526	0.557	0.726	0.416	0.576	0.576	0.783	0.77	0.03	0.397
СО	0.593	0.0007	0.024	0.023	0.0001	0.0001	0.002	0.039	0.0001	0.072	0.272	0.307

Table. p-values at the Thai monitoring stations using the Kruskal-Wallis non-parametric test.

Section 5.2.4, how do the model results compare to each other when different emission inventories are used to predict NO2 columns?

In the original manuscript, we included a sentence (page 9366, lines 8-11) stating, "The largest variation among the model simulations occurs in this region near Indonesia and is a result of both low  $NO_2$  mixing ratios from the MOZART boundary conditions and different estimates for shipping emissions among the different inventories." We now add to this information that the  $NO_2$  columns from the different WRF-Chem simulations have a similar pattern and magnitude, and have put this information in a separate paragraph.

<Editorial Comments>

- 1. Use carbon monoxide or CO (ozone or O3) consistently
- 2. Is the abbreviation for Chonburi CB (line 23 page 9362) or CBR (Fig. 9)? Be consistent.
- 3. Line 9 on page 9364, Table 4, instead of Table 5, shows correlation coefficients.
- 4. Line 10 on page 9364, Table 5, instead of Table 6, shows O3 biases.
- 5. Show units in Tables 3 and 5.

Thank you for the editorial comments. We have made these changes.

#### **Response to the 2<sup>nd</sup> Referee**

We very much appreciate the comments and suggestions from the referee. Incorporation of the referee's comments has greatly improved our paper. Below is a detailed response to the comments. The referee's comments are in regular font and our responses are in **bold** font.

1. A major conclusion of the paper is that there is severe underestimate of CO and over estimate of ozone by WRF-Chem, and this cannot be fixed by using different emission inventories. For the underestimate of CO, the authors provide two possible reasons: (1) vertical distribution of biomass burning plumes (2) missing CO sources such as biofuel and trash burning. But this is not satisfying as vertical distribution of bb plumes does not help the underestimate of CO in December when biomass burning emission is relatively small. The more interesting question is, how much increase on emission is needed to reproduce those CO surface observations? Kopacz et al. [2010] recommends a large increase of current CO anthropogenic emission has an uncertainty of ±185%. It seems more interesting that the authors can scale up their emission inventories by a factor of 2-3, to see if the bias in CO can be eliminated. Another possible reason is the model chemistry. A recent paper by Mao et al. [2013] suggests that the heterogeneous process can also help to improved modeled CO particularly over Southeast Asia. It seems to me that further discussion is warranted on these hypotheses.

We thank the reviewer for these comments. They are good points. We conducted sensitivity simulations using higher CO and NO emissions, based on the uncertainties reported for the INTEX-B inventory. Two sensitivity simulations were done with CO and NO emissions 2x and 1.4x higher for both INTEX-B and MACCity emissions INTEX-B 2xCO 1.4xNO (results shown below are marked and MACCity 2xCO 1.4xNO for these two simulations). Another two sensitivity simulations were done with only CO emissions increased by 2x (NO emissions remained the same as original inventory) for both INTEX-B and MACCity emissions (results shown below are marked INTEX-B 2xCO and MACCity 2xCO for these two simulations). The sensitivity simulations were performed for March when biomass burning is a major contribution to the results. The results were compared to the six ground sites from Pollution Control Department in Thailand including Chiang Mai (CM), Khonkaen (KK), Sarabuti (SRB), Nonthaburi (NTB), Chonburi (CBR) and Surat Thani (SRT).

The higher emissions improve agreement for both  $O_3$  and CO concentrations at the 6 monitoring sites. For example, the  $O_3$  prediction from the increased emission simulations, on average, improved the correlation term by ~18% and reduced the bias from 24 ppbv to 8 ppbv. The high emissions simulations decreased, on average, the correlation for CO surface mixing ratios by 23--34%, but reduced the average bias from 250-264 ppbv to 184-224 ppbv. Interestingly, the high emissions simulations produced too much CO at Chiang Mai (CM) by over 400 ppbv, yet the  $O_3$  bias at CM was reduced to 2-4 ppbv (from 38-40 ppbv). This suggests that either the CO emissions

from biomass burning are too high, or co-emitted VOCs should have a higher emissions rate. The SRB site, downwind of Bangkok, went from too little CO (bias = -150 ppbv for INTEX-B) to too much CO (bias = 173 ppbv for INTEX-B high emissions simulations) with only a 2 ppbv decrease in bias of  $O_3$ . CO at SRT changed very little, because SRT is located away from urban and biomass-burning regions. At the same time, KK, NTB and CBR all have a better correspondence to observations as shown by the decreased bias. However, WRF-Chem still underpredicts CO at these sites. The higher emissions slightly improved the prediction of  $NO_2$  mixing ratios increasing the correlation coefficient by 18% but not changing the bias on average. By comparing the simulation with increased CO and NO emissions to the simulation with only increase CO emissions, the results for  $O_3$  and CO are very similar indicating thzat increased CO emissions caused the decrease in  $O_3$  concentrations. We have now added this discussion to the paper (Section 6).

We also include text in Section 5.2.2 regarding the effect of heterogeneous chemistry on CO and  $O_3$ , citing Mao et al. (2013). However, we did not perform any sensitivity simulations of heterogeneous chemistry because it would require major changes to WRF-Chem and is better suited for a future study.

"However, the underprediction of CO could also be due to low anthropogenic emissions (discussed further in Section 5), a high planetary boundary layer height, which would cause dilution of surface mixing ratios, and/or missing chemistry such as heterogeneous chemistry in the model. Mao et al. (2013) suggest uptake of HO<sub>2</sub> to aerosols undergo reaction with transition metal ions to convert HO<sub>2</sub> to H<sub>2</sub>O, removing hydrogen oxides from the atmosphere. They show that this proposed mechanism decreases OH at the surface, as simulated by the GEOS-Chem model, and consequently increases CO mixing ratios by 20-30 ppbv. While a 20-30 ppbv increase in CO over Thailand will not remove the high CO bias in our simulation, this heterogeneous chemistry may explain some of the underprediction of CO."

	KK	SRB	NTB	CBR	SRT
Mar	Mar	Mar	Mar	Mar	Mar
0.74	0.72	0.70	0.45	0.05	0.44
0.68	0.71	0.27	0.66	0.80	0.53
0.67	0.71	0.28	0.66	0.79	0.52
0.68	0.71	0.69	0.44	0.02	0.43
0.63	0.69	0.26	0.65	0.82	0.49
0.62	0.68	0.29	0.65	0.81	0.49
	0.74 0.68 0.67 0.68 0.63	0.74 0.72   0.68 0.71   0.67 0.71   0.68 0.71   0.63 0.69	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	

Monthly-average correlation coefficients of daytime (00, 06, 12 UTC) ozone.

Ozone Correlation Average INTEX-B = 0.52, Average INTEX-B-2xCO1.4xNO = 0.61, Difference: +17% Ozone Correlation Average INTEX-B = 0.52, Average INTEX-B-2xCO = 0.61, Difference: +17% Ozone Correlation Average MACCity = 0.50, Average MACCity-2xCO1.4xNO = 0.59, Difference: +18% Ozone Correlation Average MACCity = 0.50, Average MACCity-2xCO = 0.59, Difference: +18%

Emission Inventories	СМ	KK	SRB	NTB	CBR	SRT
	Mar	Mar	Mar	Mar	Mar	Mar
INTEX-B	37.76	32.62	15.31	18.54	23.64	14.57
INTEX-B_2xCO_1.4xNO	-3.72	8.92	14.63	3.45	-5.01	14.57
INTEX-B_2xCO	-2.65	8.64	13.77	2.71	-6.45	11.97
MACCity	39.67	30.24	14.62	18.53	24.88	15.76
MACCity_2xCO_1.4xNO	3.00	8.89	13.50	2.00	-5.00	18
MACCity_2xCO	2.13	8.17	12.66	1.38	-6.32	15

Monthly-average biases of daytime (00, 06, 12 UTC) ozone (ppb)..

Ozone Biases Average INTEX-B = 23.74, Average INTEX-B-2xCO1.4xNO = 8.38, Difference: -65% Ozone Biases Average INTEX-B = 23.74, Average INTEX-B-2xCO = 7.70, Difference: -68% Ozone Biases Average MACCity = 23.95, Average MACCity-2xCO1.4xNO = 8.39, Difference: -65% Ozone Biases Average MACCity = 23.95, Average MACCity-2xCO = 7.61, Difference: -68%

Monthly-average correlation coefficients	(r) of davtime (00, 06,	12 UTC) carbon monoxide.
		<b>12</b> e i e, eu son monomue e

Emission Inventories	СМ	KK	SRB	NTB	CBR	SRT
	Mar	Mar	Mar	Mar	Mar	Mar
INTEX-B	0.51	0.42	0.17	0.33	0.44	0.58
INTEX-B_2xCO_1.4xNO	0.46	0.40	0.09	-0.05	0.09	0.57
INTEX-B_2xCO	0.46	0.40	0.08	-0.06	0.08	0.58
MACCity	0.50	0.45	0.11	0.26	0.43	0.55
MACCity_2xCO_1.4xNO	0.45	0.40	-0.04	-0.22	-0.07	0.56
MACCity_2xCO	0.44	0.40	-0.04	-0.22	-0.08	0.55

CO Correlation Average INTEX-B = 0.41, Average INTEX-B-2xCO1.4xNO = 0.27, Difference: -34% CO Correlation Average INTEX-B = 0.41, Average INTEX-B-2xCO = 0.27, Difference: -34% CO Correlation Average MACCity = 0.38, Average MACCity-2xCO1.4xNO = 0.29, Difference: -23% CO Correlation Average MACCity = 0.38, Average MACCity-2x = 0.29, Difference: -23%

Monthly-average biases of daytime (0	10, 06, 12 UTC) carbon mo	noxide (ppb).
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Emission Inventories	СМ	KK	SRB	NTB	CBR	SRT
	Mar	Mar	Mar	Mar	Mar	Mar
INTEX-B	-16	-316	-150	-662	-203	-234
INTEX-B_2xCO_1.4xNO	414	-105	173	-373	-80	-196
INTEX-B_2xCO	414	-105	173	-375	-80	-194
MACCity	-18	-292	-116	-636	-195	-238
MACCity_2xCO_1.4xNO	418	-70	295	-109	-13	-200
MACCity_2xCO	412	-69	297	-107	-12	-197

CO Biases Average INTEX-B = 263.5, Average INTEX-B-2xCO1.4xNO = 223.5, Difference: -15% CO Biases Average INTEX-B = 263.5, Average INTEX-B-2xCO = 223.5, Difference: -15% CO Biases Average MACCity = 249.2, Average MACCity-2xCO1.4xNO = 184.2, Difference: -26% CO Biases Average MACCity = 249.2, Average MACCity-2xCO = 184.2, Difference: -26%

#### Monthly-average correlation coefficients (r) of daytime (00, 06, 12 UTC) nitrogen dioxide.

Emission Inventories	СМ	KK	SRB	NTB	CBR	SRT

	Mar	Mar	Mar	Mar	Mar	Mar			
INTEX-B	0.63	0.04	-0.11	-0.14	-0.02	0.37			
INTEX-B_2xCO_1.4xNO	0.69	-0.07	-0.15	-0.20	0.05	0.42			
INTEX-B_2x	0.70	-0.06	-0.15	-0.21	0.05	0.43			
MACCity	0.64	0.04	-0.09	-0.16	-0.08	0.36			
MACCity_2xCO_1.4xNO	0.70	-0.08	-0.19	-0.26	0.003	0.43			
MACCity_2xCO	0.69	-0.07	-0.20	-0.26	-0.0001	0.42			
NO <sub>2</sub> Correlation Average INTEX-B = 0.22, Average INTEX-B-2xCO1.4xNO = 0.26, Difference: 18%									
$NO_2$ Correlation Average INTEX-B = 0.22, Average INTEX-B-2xCO = 0.26, Difference: 18%									
NO <sub>2</sub> Correlation Average MACCity = 0.23, Average MACCity-2xCO1.4xNO = 0.28, Difference: 21%									
NO <sub>2</sub> Correlation Average N	ACCity = (	0.23, Average N	MACCity-2xC0	= 0.	28, Difference:	21%			

Monthly-average biases of daytime (00, 06, 12 UTC) nitrogen dioxide (ppb).

Emission Inventories	СМ	KK	SRB	NTB	CBR	SRT
	Mar	Mar	Mar	Mar	Mar	Mar
INTEX-B	14	-19	132	-14	91	-6
INTEX-B_2xCO_1.4xNO	24	-14	137	-10	93	-5
INTEX-B_2xCO	19	-17	134	-12	92	-6
MACCity	-15	-21	132	-13	92	-6
MACCity_2xCO_1.4xNO	4	-17	136	-9	96	-6
MACCity_2xCO	-26	-32	120	-34	79	-7

NO<sub>2</sub> Biases Average INTEX-B = 46, Average INTEX-B-2xCO1.4xNO = 47, Difference: +2.2%

 $NO_2$  Biases Average INTEX-B = 46, Average INTEX-B-2xCO = 47, Difference: +2.2%  $NO_2$  Biases Average MACCity = 45, Average MACCity-2xCO1.4xNO = 45, Difference: 0%

 $NO_2$  Biases Average MACCity = 45, Average MACCity-2xCO

- 2xCO = 50, Difference: +11%
- 2. The authors seem to have ignored the overestimate of ozone in the discussion section. Some insights are needed to this bias.

The original manuscript had a small discussion about the overestimate of ozone. This discussion brought out the use of the plume rise feature for the biomass burning emissions. We show that emitting biomass-burning emissions in just the surface layer (which is a more common occurrence in Southeast Asia than fires with plumes reaching higher elevations) improves the  $O_3$  predictions. After conducting the additional sensitivity simulations, we have added discussion on the effect of increased CO and NO emissions on improving  $O_3$  predictions.

3. I agree with reviewer #1 that, a table with detailed comparison for each inventory, including seasonality, year, total amount etc., would help the reader to understand the difference among these inventories.

We have added a table listing the source sectors for each of the emission inventories, and have modified the table listing the total emissions. We also discuss the difference in

## seasonality between RETRO and MACCity. We hope with these changes the reader is better able to understand differences between the emission inventories.

4. Comparison with satellite. The discussions on satellite comparison are very brief and problematic. My understanding is that MOPITT signal is very weak for the surface air due to low thermal contrast between surface air and the surface itself [Deeter et al., 2007]. What is the uncertainty level for the MOPITT retrieval for surface CO? Also it seems that MOPITT CO is less than all modeled CO, and even further away from those ground site observations. This makes the reader suspicious of the quality of MOPITT retrieval on surface CO.

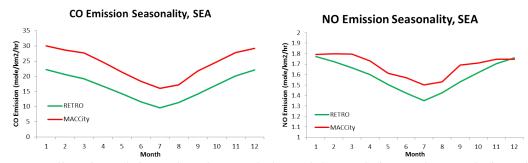
The new version of MOPITT data (Version 6; Deeter et al. 2011; 2012; 2013; Worden et al. 2010), which we used in this paper, has improved near surface CO retrievals. This improvement is accomplished by using near-infrared and thermal-infrared observations simultaneously to enhance the retrieval sensitivity of CO in the lower troposphere. Please see the MOPITT V6 User's Guide at <a href="http://www2.acd.ucar.edu/sites/default/files/mopitt/v6\_users\_guide\_201309.pdf">http://www2.acd.ucar.edu/sites/default/files/mopitt/v6\_users\_guide\_201309.pdf</a> and the cited references for more information.

Deeter et al., JGR 2013: <u>http://onlinelibrary.wiley.com/doi/10.1002/jgrd.50272/abstract</u> Deeter et al., JGR 2012: <u>http://onlinelibrary.wiley.com/doi/10.1029/2012JD017553/abstract</u> Deeter et al., JGR 2011: <u>http://onlinelibrary.wiley.com/doi/10.1029/2011JD015703/abstract</u> Worden et al., JGR 2010: doi:10.1029/2010JD014242

5. Model spin up. Given the long lifetime of CO and ozone (particularly in winter) how was the model spun up for each simulation with different emission inventories? Did the WRF-Chem have a few months to spin up, or just use the same boundary condition from MOZART at the beginning of March and December of 2008? This needs to be clarified. If there is bigger discrepancy among those inventories in other months (February for example), how will that affect the modeled CO or ozone?

The WRF-Chem simulations were spun up for a 2-week period for each simulation. The two weeks should be long enough to remove the effect of initial conditions because there is plenty of time for emissions to control surface mixing ratios and transport to affect mixing ratios in the free troposphere. The seasonality follows the RETRO emissions seasonality for the RETRO, INTEX-B, and SEAC4RS simulations and follows the MACCity emissions seasonality for the MACCity and MACCity/SEAC4RS simulations. The seasonalities of RETRO and MACCity CO emissions are quite similar (Figure below). We do not expect the February or November CO emissions to affect the results of the paper. The seasonalities of RETRO and MACCity NO emissions are somewhat different when comparing March NO emissions to February. A bigger difference is seen for NO emissions in November compared to December. These differences could introduce uncertainties in the O<sub>3</sub> results for the

region. Therefore we have now included a short discussion on these differences in the paper (section 6).



**Figure.** Seasonality of CO (left panel) and NO emissions (right panel) for Southeast Asia from the RETRO (green) and MACCity (red) emission inventories.

### **Relevant Changes Made to the Manuscript**

1. By conducting a statistical significance test, our conclusion has changed. The last 2 sentences of the abstract (p. 2) now say that the different emission inventories predict different CO surface mixing ratios but similar  $O_3$  mixing ratios.

2. The fourth paragraph of the Introduction (p. 4-5) was revised to include major innovations besides the study region.

3. Section 2 (p. 6) has added information to clarify that the model simulations began 2 weeks before the focus time period of either March or December.

4. Section 3 introduction and Sect. 3.1 (p. 7-8), which describe the emissions inventories used, now include more information on activity sectors contributing to the inventory and a table listing these sectors has been cited.

5. Section 3.1 (p. 9) has additional information on the MACCity emissions uncertainties (top of page) and the shipping emissions in each inventory (bottom of page).

6. Section 3.1 (p.10) also includes new text elaborating on the seasonal cycle of both the MACCity emissions and the RETRO emissions and how those seasonal cycles compare to each other. This text is in the last paragraph of the section (p. 10).

7. Section 3.2 compares the emission inventories to each other and to other inventories not included in this study. The 1<sup>st</sup> paragraph (top of p. 11) revised the comparison between 2000 and 2012 emission scenarios to use MACCity/SEAC4RS instead of SEAC4RS emission. The 3<sup>rd</sup> paragraph (p.11) of Sect. 3.2 has been revised to present a clearer description of how our inventories compare to the REAS inventory.

8. In Section 4.2 (p. 14) we have clarified which MOPITT dataset we are using and explained that versions 5 and 6 have improved sensitivity for near-surface CO.

9. In Section 5.2.1 (p. 16), we clarified how we estimate the role of biomass burning emissions on results, define what is meant by "variation" (i.e. standard deviation among model results), and clarified that these variations are due to the different anthropogenic emissions.

10. In Section 5.2.2 (p. 17), we revised the paragraph on the CO evaluation with Thai monitoring sites to give more explanation as to why the CO is underpredicted (following reviewer's comments) and included the statistical significance of the results.

11. In Section 5.2.2 (p. 18), we added information stating that V6 of MOPITT data has improved sensitivity near the surface.

12. In Section 5.2.3 (p. 19), we added information on the statistical significance of  $O_3$  comparisons with Thai monitoring stations.

13. In Section 5.2.4 (p. 20), we added an explanation as to why the spatial pattern of the  $NO_2$  column is high in certain regions and low in others and more explanation of why modeled  $NO_2$  column differs from OMI  $NO_2$ .

14. In Section 6 (p. 22), the discussion of CO underprediction and  $O_3$  overprediction is expanded, adding information on sensitivity simulations where CO emissions are increased and where CO and NO emissions are increased.

15. In the Conclusions section (p.25), we added the results of the sensitivity simulations and the statistical significance of the comparison with monitoring stations.

16. Eight references have been added to the Reference section.

17. We have added a table (Table 1, p. 51) listing the emission sectors for each inventory.