

## **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 emissions over Southeast Asia, by almost a factor of 2. The authors also state in the text that CO emission has an uncertainty of  $\pm 185\%$ . 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 (results shown below are marked INTEX-B\_2xCO\_1.4xNO 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<sub>3</sub> and CO concentrations at the 6 monitoring sites. For example, the O<sub>3</sub> 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<sub>3</sub> 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<sub>3</sub>. 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<sub>2</sub> 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<sub>3</sub> and CO are very similar indicating that increased CO emissions caused the decrease in O<sub>3</sub> 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<sub>3</sub>, 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.”

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

Emission Inventories	CM	KK	SRB	NTB	CBR	SRT
	Mar	Mar	Mar	Mar	Mar	Mar
INTEX-B	0.74	0.72	0.70	0.45	0.05	0.44
INTEX-B_2xCO_1.4xNO	0.68	0.71	0.27	0.66	0.80	0.53
INTEX-B_2xCO	0.67	0.71	0.28	0.66	0.79	0.52
MACCity	0.68	0.71	0.69	0.44	0.02	0.43
MACCity_2xCO_1.4xNO	0.63	0.69	0.26	0.65	0.82	0.49
MACCity_2xCO	0.62	0.68	0.29	0.65	0.81	0.49

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%

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

Emission Inventories	CM	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

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 daytime (00, 06, 12 UTC) carbon monoxide .**

Emission Inventories	CM	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-2xCO = 0.29, Difference: -23%

**Monthly-average biases of daytime (00, 06, 12 UTC) carbon monoxide (ppb).**

Emission Inventories	CM	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	CM	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<sub>2</sub> 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 MACCity = 0.23, Average MACCity-2xCO = 0.28, Difference: 21%

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

Emission Inventories	CM	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<sub>2</sub> Biases Average INTEX-B = 46, Average INTEX-B-2xCO = 47, Difference: +2.2%  
 NO<sub>2</sub> Biases Average MACCity = 45, Average MACCity-2xCO1.4xNO = 45, Difference: 0%  
 NO<sub>2</sub> Biases Average MACCity = 45, Average MACCity-2xCO = 50, Difference: +11%

- 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<sub>3</sub> predictions. After conducting the additional sensitivity simulations, we have added discussion on the effect of increased CO and NO emissions on improving O<sub>3</sub> predictions.**

- 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.**

- 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 [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) and the cited references for more information.**

Deeter et al., JGR

2013: <http://onlinelibrary.wiley.com/doi/10.1002/jgrd.50272/abstract>

Deeter et al., JGR

2012: <http://onlinelibrary.wiley.com/doi/10.1029/2012JD017553/abstract>

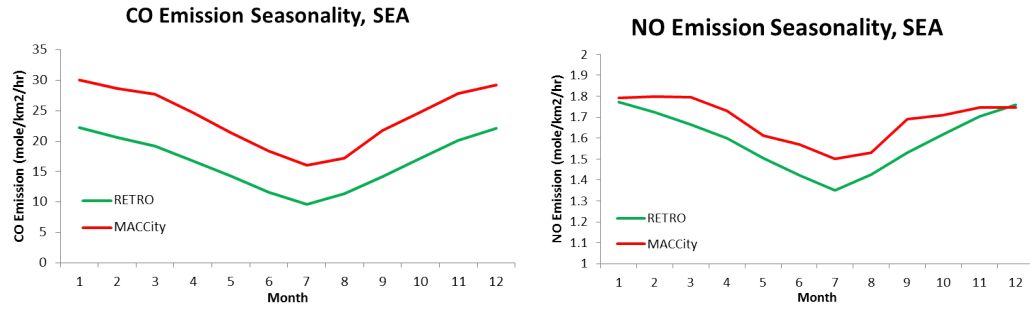
Deeter et al., JGR

2011: <http://onlinelibrary.wiley.com/doi/10.1029/2011JD015703/abstract>

Worden et al., JGR 2010: [doi:10.1029/2010JD014242](http://onlinelibrary.wiley.com/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.