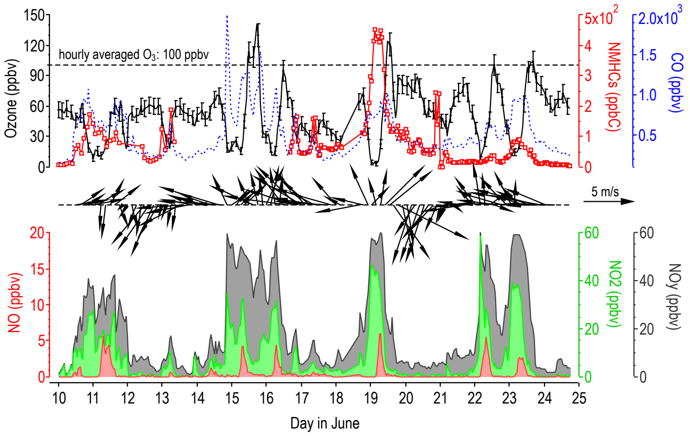
Reply to the comments of Anonymous Referee #1

We greatly appreciated the reviewer for providing insight comments and suggestions. We would like to modify our manuscript and clarify the discussions according to the specific comments. The detailed corrections are as follows:

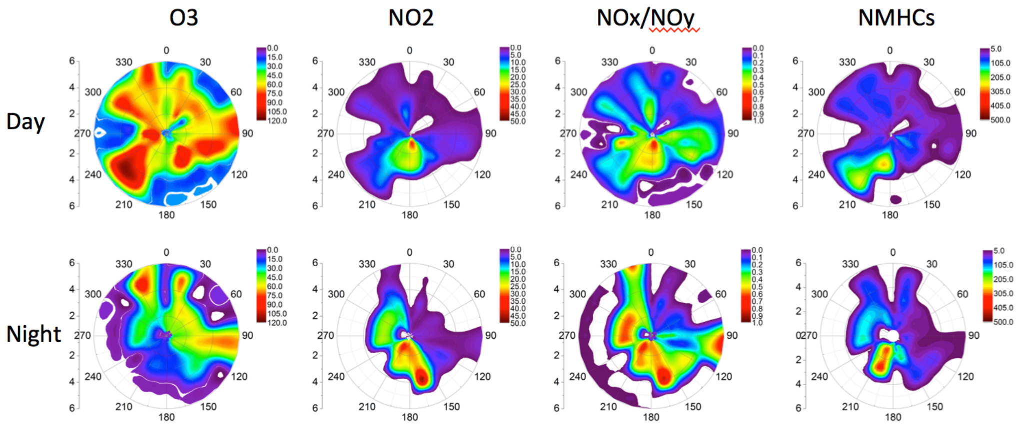
1. The paper argues that local anthropogenic emissions were very limited at the sampling site (page 30916, lines 26-27). It would be good to define what "local" means in this regard. The data in Figure 2 show that at high NOx, a very large fraction of NOy consists of NOx and ozone is titrated. This indicates that the sampled air masses at those times were only lightly aged, and had a significant contribution from local sources in addition to emissions from the Shanghai area 180 km South of the sampling site. The evidence thus appears to contradict the earlier statement that local anthropogenic emissions were very limited at the sampling site.

Reply: We will clarify this point in the revised manuscript according to referee's comments. Firstly, the NO$_x$ emission at the observation site was very limited because the field campaign was performed in a "Science and Technology Park" (almost vacant and has no residence) near seashore area. I will revise the expression in page 30916, line 26-27. We agree with referee's comments that, the sampled air masses consisted of large fraction of NO$_x$ and O$_3$ was titrated significantly, implying that the "local" source was greatly important. Within 40 km south of observation site, there are numbers of small county towns (e.g. Qidong, Latitude: 31.808°, Longitude: 121.658°), and the NO$_x$ emission from these towns seemed also contribute to the high concentration of NO$_x$ when southerly wind was prevailing (As shown in the revised Figure 2.). Additionally, to void misleading we will not point in particular to the impact of emission from Shanghai megacity on our observation because, except for Shanghai, there are numbers of big cities (population > 2 million) to the south of observation site (e.g. Natong, Latitude: 31.977°, Longitude: 120.900°, as shown in Figure 1), and the anthropogenic emission from industrial/residential area of these cities may also be responsible for the increase of NO$_x$ and NMHCs at the site.



2. Figure 4 is interesting, but the information about the dependence of trace gas concentrations on wind direction and speed is convoluted with the dependence of all those parameters on the time of day. I suggest making similar graphs for the day- and night- time separately, when the site is most likely impacted by different sources.

Reply: As suggested, the dependences of O$_3$, NO$_2$, NO$_x$/NO$_y$ and NMHCs on wind direction and speed for daytime (08:00 – 16:00 CST) and nighttime (19:00 – 05:00 CST) were plotted. As shown in the following figures, observation site was mostly impacted by the same source in the south in both daytime and nighttime. O$_3$ concentration in the daytime exceeded 100 ppb when air mass came from southwest. NO$_2$ and NMHCs also increase evidently; however at night, the O$_3$ concentration from southwest was extremely low, mostly due to titration reaction with NO$_2$ being emitted from urban area in the south. NO$_x$/NO$_y$ ratio from south at night was generally larger than 0.5, indicating that the air mass was not very aged.



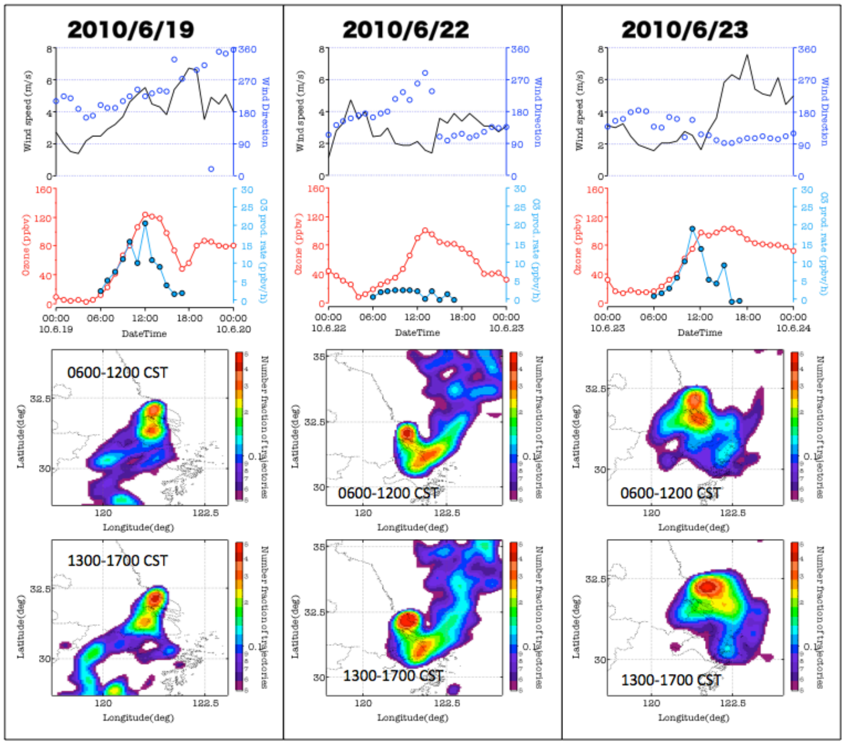
3. Section 5.1: it is interesting that the in-situ photochemical production rates of ozone are similar to the actual observed ozone increase rate during the day. However, this does not prove that a box model is adequate to explain the observations. I would weaken the conclusions of this section to reflect this. In addition, I would like to see more details on the diurnal variations in wind direction and speed to consider the relative importance of in-situ chemistry and transport. Given that the sampling site is on the coast, there may be land-sea breezes that could affect the ambient concentrations of trace species? In general, it is very dangerous to interpret observed increases in ozone at a ground site in terms of chemistry only, and the authors need to be much more cautious in this section.

Reply: We would like to follow reviewer's comments to revise the discussion in section 5.1. Diurnal variations of ambient O_x concentration, O_x net production rate on the basis of RACM2 model, and wind direction/speed were shown in the following figures. The relative importance of *in situ* photochemistry and direct transport differed from case to case.

The net photochemical production of O_x ($P(O_x)$) during the field campaign was calculated based on Eqs. (1) – (3) in the manuscript. As shown in Figure, when the site experienced prevailing moderate southwest winds (~ 4 m/s) on June 19th, 2010, the observed maximum O_x concentration was 128 ppbv at 12:00 CST, with a daytime increase of 79 ppbv. $P(O_x)$ also peaked (21 ppbv h^{-1}) at 12:00 CST with an accumulated photochemical production of O_x (06:00 – 12:00 CST) of 73 ppbv. This indicated that *in situ* photochemistry almost fully explained the buildup of O_x at the site in the morning. In the afternoon, the O_x concentration decreased quickly to 52 ppbv (at 17:00 CST) when it changed to a strong westerly wind (~ 6 m/s), and the total *in situ* photochemical production of O_x was only 27 ppbv, reflecting the importance of direct transport. Footprint calculations (Fig. 6f in the manuscript) indicated that O_x formation at the urban area to the south of the observation site had a significant contribution. Total O_x production was found to be 100 ppbv on a daily basis, and the 6-h (09:00 – 15:00 CST) averaged $P(O_x)$ was 13 ± 4 ppbv h^{-1}, which was similar to the observations (13 ppb h^{-1}) in central Tokyo areas during the summer (Kanaya *et al.,* 2008).

On June 23rd, 2010, the daytime maximum $P(O_x)$ was found to be 19 ppbv h^{-1} at 11:00 CST. The accumulated photochemical production of O_x (06:00 – 11:00 CST) at the site was 41 ppbv, similar to the observed increase of O_x (48 ppbv) during this period, which indicated that the buildup of O_x could be attributed to *in situ* photochemistry. During the afternoon, the wind direction shifted from south to east (the marine region) around 12:00 CST, and the wind speed increased from 2 to 6 m/s. Correspondingly, the $P(O_x)$ decreased sharply from 19 to 5 ppbv h^{-1}, and the ambient O_x concentration at the site continued to increase and finally exceeded 100 ppbv at 15:00 CST. On the same day, the observed O_x concentration at night reached 80 ppbv. This suggested that the photochemically produced O_3 in the YRDR during the daytime was preserved over the marine area and was transported directly to the observation site with an easterly sea breeze at night.

On the morning of June 22nd, 2010, the site experienced prevailing southwesterly winds, and the accumulated photochemical production of O_x (06:00 – 13:00 CST) was only 14 ppbv, accounting for one-fourth of the observed daytime O_x concentration buildup (60 ppbv), reflecting the importance of the direct transport of O_x.

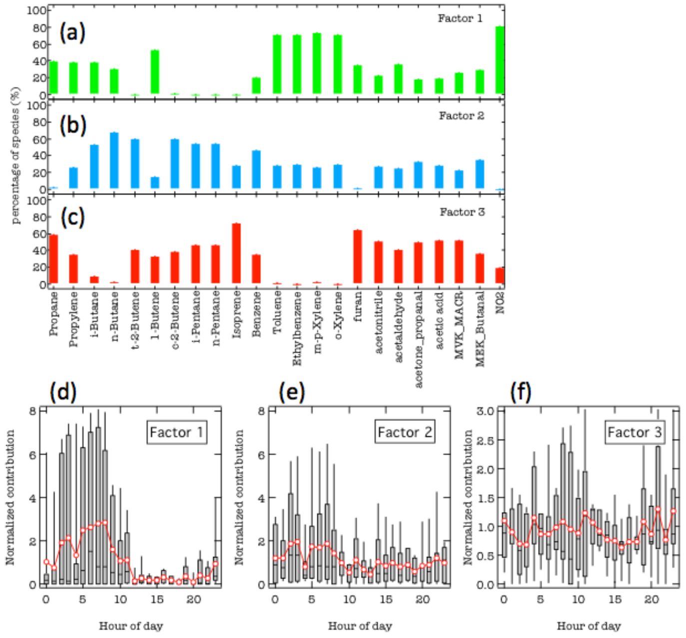


4. I found Figure 7 to be unclear. The isopleth in the background is clear, but what do the symbols, the symbol colors, the dashed and dotted lines, and the numbers along the x- and y-axes represent?

Reply: The circle and square symbols represent the calculated O_x production rate on the basis of RACM2 model on June 19th and June 23rd, respectively. The dashed lines indicate the temporal variations of NO_x on these days. NMHCs/NO_x ratios, marked as the red-dotted lines and numbers in the lower-left corner of the plot, are used to reflect of the controlling region of O_x production. We will clarify this in the figure caption of revised manuscript.

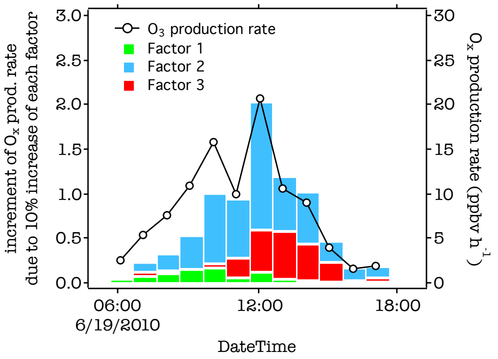
5. Section 5.4.1: I am worried that the other factors are influenced by chemical transformations in addition to different emission signatures. The authors have nicely shown that aged air masses from the Shanghai area are sampled at the site. In these air masses, the more reactive NMHCs will be more efficiently removed from the sampled air masses than the less reactive species. This chemistry leads to different time series for more and less reactive species 。 and PMF can pick up on those differences and attribute more and less reactive species into different factors even though they come from the same source. Indeed, in the authors' analysis, the more reactive aromatics fall into factor 1, the less reactive alkanes in factors 3 and 4, and the least reactive benzene and propane in factor 6. The issue was investigated in a recent paper [Yuan et al., 2012], and I think the authors need to discuss the results from their PMF analysis in light of this issue.

Reply: We will revise section 5.4.1 in the manuscript according to reviewer's insight comment. In the revised manuscript, we will reduce the PMF factor number from 6 to 3, and the previous explicit classification (such as vehicle, fuel evaporation etc.) is not used any more. The contribution of specified factor to the each specie (a-c), and diurnal variation (d-f) of each factor are shown in the following figure. Factor 1 showed an obvious diurnal variation and consisted of a large fraction of high-reactive species (e.g. C8-C9 aromatics, 1-butene) and NO_2. Factor 2 did not have diurnal variability and had an abundance of low-weight alkanes and fewer fractions of high-reactive species and NO_2. As reported previously (Yuan et al., 2012), NMHCs photochemistry could lead to different time series for more and less reactive species, and PMF can detect these differences and attribute more or fewer reactive species to different factors, even though they originate from the same source. Therefore, we cannot identify the emission sources for these two factors. Instead, we attributed factors 1 and 2 to less-processed and more-processed air masses. Factor 3 was considered an OBB-related source due to the pronounced contribution of furan and acetonitrile with mass fractions of 64% and 50%, respectively. This factor also accounted for 72% of total isoprene, 51% of MVK + MACR (oxidation production of isoprene), and 52% of acetic acid. As shown in previous studies (Kudo et al., 2014), factor 3 would retain the source characteristics, as the OBB occurred near the observation site.



6. Section 5.4.2: this analysis is not very convincing. In light of my previous comment, some of the source attribution may change. But also, what the analysis indicates to me is that the most reactive NMHCs simply do not reach the sampling site as efficiently in the afternoon. This does not imply that the more reactive NMHCs are less important for ozone formation in the afternoon. The same comment as before applies: it is dangerous to interpret the observed ozone in terms of in-situ formation only, whereas in reality the ozone at a site needs to be explained by a combination of the location and strength of the emissions, the transport of those emissions to the site and chemical transformations during the transport.

Reply: As shown in the revised manuscript, we followed referee's suggestions to discuss the importance of photochemical formation in the morning and afternoon, separately. During the heavy O_3 pollution case, the increase of O_x concentration in the morning could mostly be explained by *in situ* photochemistry, and the direct transport O_3 that formed in the upstream area was mostly responsible for the increase of O_x in the afternoon and night time at observation site. Here, we showed the relative importance of each factor on the *in situ* photochemical formation of O_x for a typical heavy O_3 pollution case on June 19th, 2010 (hourly-averaged O_3 concentration: 124 ± 5 ppbv and one minute value: 168 ppbv at 12:00 CST). As shown in following figure, the contribution of different factors to O_x production showed distinct variation patterns. Factor 2 was responsible for 60% (43 ppb) of the *in situ* photochemical production of O_x in the morning, followed by that of factor 1 (23%, 17 ppb), and the OBB-related factor only accounted for 17% of the total O_x production. In the afternoon, the relative importance of OBB-related factors increased and was responsible for 34% (12 ppb) of the photochemical O_x production.



Minor comments:

1. Caption of Figure 3: I think the definitions of the blue and red symbols are reversed, i.e. red represents high ozone days and blue low ozone days?

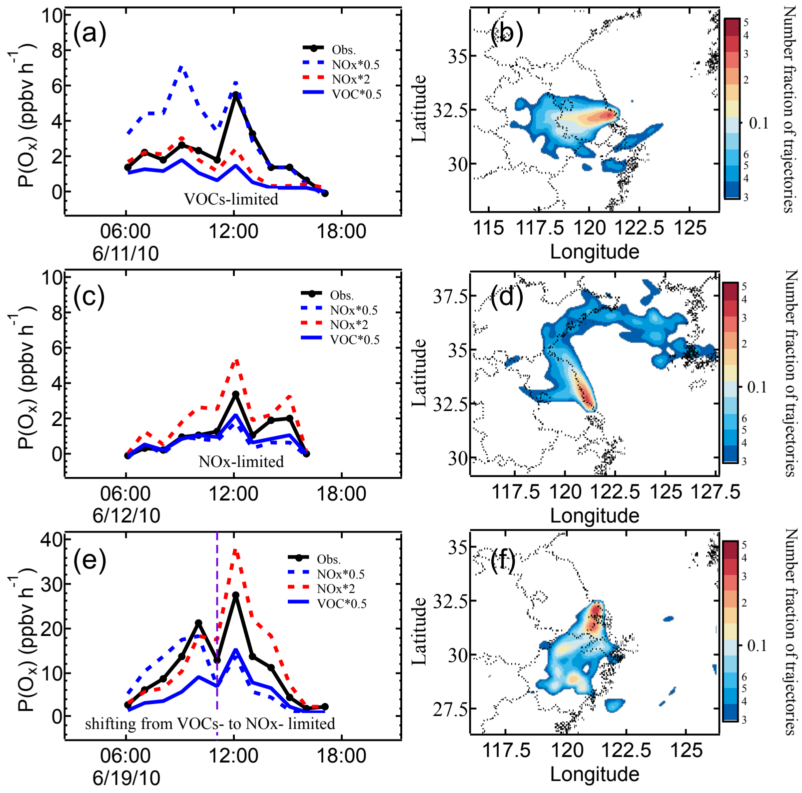
Reply: red and blue symbols in the figure 3 represented high- O_3 and low- O_3 days, we will corrected it in the revised manuscript.

2. Page 30922, line 22: "broader" instead of "boarder".

Reply: The misspelling of word will be corrected.

3. Figure 6: do the footprints on the right refer to specific sampling times for the days shown? Or are they an average for the time periods shown? This was not clear to me. Also, much of the important information is actually in the different scales used for the y-axes, i.e. ozone production ranges from 0-40 ppbv/hr on June 19, but only from 0-6 ppbv/hr on June 12. It would be good to point that out clearly to the reader as it is easily overlooked.

Reply: The footprints on the right refer to an average for the periods shown. For better review the sensitive regime of O_x production, y-axes of June 11th, and June 12th are the same (0-10 ppb h^{-1}), and y-axes of June 19th is 0-40 (ppbv h^{-1}). By the way, the residence time in the footprint plot will be changed to number fraction of backward trajectories in specific time period (0700- 1700 CST). The larger the fraction of number of trajectories in the grid the greater emission sources influence.



Reply to the comments of Anonymous Referee #2

We greatly appreciated the reviewer for providing insight comments and suggestions. We would like to modify our manuscript and clarify the discussions according to the specific comments. The detailed corrections are as follows:

1. P30917, L18, "Fifteen NMHCs species were detected . . . ...". At a site impacted by significant open biomass burning, the air should contain a lot of NMHCs and other VOCs with higher mixing ratios. Why only fifteen NMHCs were detected?

Reply: In the present study, NMHCs species were detected using an online gas chromatography-flame ionization detection (GC-FID) and gas chromatography-mass spectrometry (GC-MS). One of purpose of our field campaign was to clarify the contribution of OBB to O_3 pollution in YRDR. Therefore only main NMHC species relevant to O_3 chemistry was measured, and the detailed information about the measurements of NMHCs and OVOCs were published in the literature (Kudo, S., et al. (2014), Emissions of non-methane volatile organic compounds from open crop residue burning in the Yangtze River Delta region, China, J. Geophys. Res. Atmos., 119, 7684–7698, doi:10.1002/2013JD021044.)

2. P30918, L16, "The missing data were linearly interpolated". What was the portion of the missing data? Can the data interpolation impact your results?

Reply: The NMHCs species were measured at time resolution of 1~ 2 h, however the time step of input of RACM2 model was at time resolution of 5 minutes. For the convenience of model run, the data gap was linearly interpolated, and such treatment did not influence our results. During the observation period, the VOC data between June 13 and June 17 were missing because of instrumental maintenance. For the other high- O_3 pollution days, all the instruments were working well and the dataset was available for the RACM calculation. Actually, time interpolation of NMHCs data did impact our results.

3. P30919, equation (2), the summation sign in the last term is not necessary. However, it may be necessary to include reactions of NO2 with some peroxy radicals, such as peroxyacetyl radical

Reply: The summation sign in the equation (2) will be removed. As suggested by referee, we did sensitive tests by including the peroxy-acetynitrate formation reactions (ACO3 + NO2 → PAN, RCO3 + NO2 → PPN and ACP + NO2 → MPAN) and decomposition reaction (PAN → ACO3 + NO2 , PPN → RCO3 + NO2 and MPAN → ACP + NO2); however their influence on the simulation of O_3 production were ignorable. Here, we did not include these reactions in the present study.

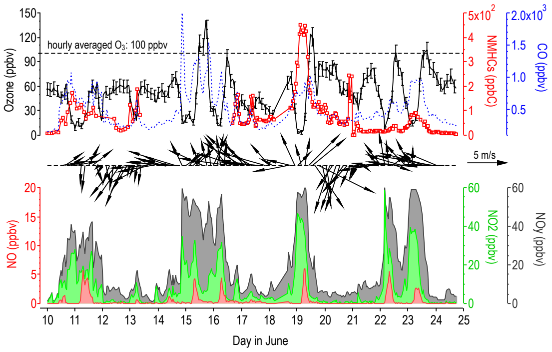
4. P30922, section 4.2, how did you know the measurements were impacted by OBB? Was the burning near the site? If so, NOx from the OBB plumes may also be significant so that the OBB impact may be more important than the urban plumes transported from about 100 km away

Reply: we would like to clarify this point in the manuscript according to referee's comments.
To the west of the observation site is the typical agriculture region, and open burning of agriculture residues sporadically occurred during our field campaign. Actually, the nearest open fires were about 1 km away from to the west of our observation site, and the flame and smoke was almost visible. The impact of OBB on the tropospheric carbonaceous aerosols and OVOCs loading have been reported in previous publications (1, 2).

1. Pan, X. L., et al., (2012), Emission ratio of carbonaceous aerosols observed near crop residual burning sources in a rural area of the Yangtze River Delta Region, China, **J. Geophys. Res.**, 117, D22304, doi:10.1029/2012JD018357.
2. Kudo, S., et al. (2014), Emissions of non- methane volatile organic compounds from open crop residue burning in the Yangtze River Delta region, China, **J. Geophys. Res.**, 119, 7684–7698, doi:10.1002/2013JD021044.)

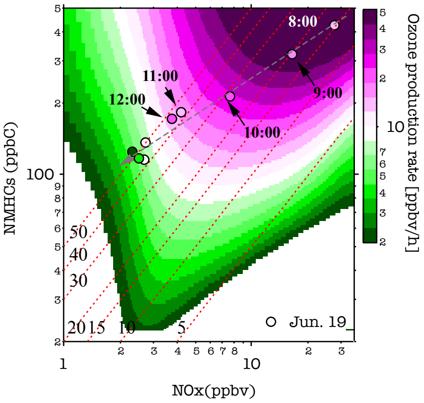
To avoid misleading, we would like to make it clear that, except for Shanghai, there are numbers of big cities (population > 2 million) in the south of observation site (Natong, Wuxi, Suzhou, as shown in Figure 1 in the manuscript), and the pollutants emitted from industrial/residential area of these cities were also responsible for the increase of NOx and NMHCs at the site. In the revised manuscript, we will use "transported from urban areas to the south", rather than specifying "Shanghai area" that is 100 km away.
As mentioned by referee, biomass combustions emit NO_x. However, it was hardly explain the long period (more than 12 hours), high troposphere NO_x concentrations (~50 ppbv). Firstly, OBB normally emitted substantial amount of CO due to incomplete combustion processes. Sudden and sharp increase of CO concentration (dashed line in Figure 2) indicated the impact of OBB on our observation. However, we did not observe evident increase of NO_x during OBB-impacting period.

Instead, the temporal variation of NO_x was more likely to reflect synoptic pollution processes. Secondly, we found that, NO_x concentration increased significantly when the air mass came from urban area to the south. Figure 2 illustrates the dependence of averaged NO_2 concentrations as a function of wind speed and direction in the day and at night. It was clear that the NO_2 emission from south was mostly responsible for the NO_2 at site, and the emission of NO_x from OBB was less important than that from anthropogenic emission in the urban area.



5. The paragraph between P30925 (L 26) and P30926 (L9), and Figures 6 and 7. I disagree with your statements. Based on Figures 6 and 7, I think O3 production on 19 June 2010 changed from VOCs-controlling to VOCs- and NOx-controlling, and that on 23 June 2010 was VOCs-controlling.

Reply: I will make it clear the discussion about the O_x production sensitivity, and we marked the time of day for the case on 19 June 2010. As shown in Figure 7, the O_x production in the morning was sensitive to both NO_x and VOCs; however decrease of NO_x concentration by 50% resulted in increase of O_3 (as shown in blue dash line), and decrease of VOCs concentration by 50% resulted in evident decrease of O_x production (blue line). It indicated that reduction VOCs in the morning was more effective to reduce the risk of high O_x pollution; therefore we defined this period as "VOCs-limited" and it was "NO_x-limited" in the afternoon. For the case on 23 June 2010, we could see similar variability of O_x productions by changing the NO_x or VOCs concentration, and O_x production regime shifted from VOCs- limited to NO_x- limited on 1200 CST. In the revised manuscript, we will remove the case on 23 June 2010 from the figure 7 because, the O_x production isopleth diagram was derived on the basis of a series of model runs that was constrained by the mixing ratio of O_3, NO_x, and VOCs, J values, and meteorological data observed at 10:00LST on 19 June. The O_3 production isopleth diagram on 23 June 2010 may be slightly different.



6. Section 5.4.1, are these results from Kudo et al. (2014)? The number of samples for the PMF analysis should be given. Very reactive species, such as isoprene, NO2, etc., were used in the PMF calculations. Such species may be largely reduced (NO2 may also be produced) during the transport from the sources to the observation site. How did you treat this influence? What is the impact of this problem on the results? You attribute Factor 5 to vehicular-exhaust, but we know that VOCs are co-emitted by vehicles. Are there coal-fired power plants in the surrounding area? If there are, they may be major contributor to NOx.

Reply: The result of PMF analysis in this study was not from Kudo et al.,' paper. In the revised manuscript, we will reduce the PMF factor number from 6 to 3, and the previous explicit classification (such as vehicle, fuel evaporation etc.) is not used any more. The data matrix for PMF was 206 rows (samples) × 23 columns (species). The uncertainty in

NMHC concentrations was determined as the sum of 15% of the concentration and half of the detection limit (the same as the descriptions in the PMF user's guide). Three potential factors were determined in this study. The contribution of specified factor to the each specie, and diurnal variation of each factor are shown in the supplementary materials. Factor 1 showed an obvious diurnal variation and consisted of a large fraction of high-reactive species (e.g. C8-C9 aromatics, 1-butene) and NO_2. Factor 2 did not have diurnal variability and had an abundance of low-weight alkanes and fewer fractions of high-reactive species and NO_2. As reported previously (Yuan et al., 2012), NMHCs photochemistry could lead to different time series for more and less reactive species, and PMF can detect these differences and attribute more or fewer reactive species to different factors, even though they originate from the same source. Therefore, we cannot identify the emission sources for these two factors. Instead, we attributed factors 1 and 2 to less-processed and more-processed air masses. Factor 3 was considered an OBB-related source due to the pronounced contribution of furan and acetonitrile with mass fractions of 64% and 50%, respectively. This factor also accounted for 72% of total isoprene, 51% of MVK + MACR (oxidation production of isoprene), and 52% of acetic acid. As shown in previous studies (Kudo et al., 2014), factor 3 would retain the source characteristics, as the OBB occurred near the observation site.

7. P30916, L8, delete "anthropogenic".

Reply: We delete "anthropogenic" in the revised manuscript

8. P30916, L15-16, "from the middle of May to the end of June". Which year? In other places you use "June 2010".

Reply: We will use "from the middle of May to the end of June, 2010" and I will keep consistency throughout the context.

9. P30916, L26-27, exchange the sentence "Local anthropogenic emissions in the Science and Technology park were very limited" with that after it.

Reply: We will clarify this sentence in the revised manuscript.

10. P30917, L16, delete "mass".

Reply: The word "mass" will be deleted in the revised manuscript.

11. P30917, L28, "Ionico" should be "Ionicon".

Reply: The word "Ionico" will be replaced with "Ionicon"

12. P30923, L8, delete "(not including oxygenated VOCs)". It is by definition clear that NHMC does not include oxygenated VOCs.

Reply: The sentence "not including oxygenated VOCs" will be deleted.

13. P30924, L19, "at 11:00 LST"? I think it is "at 12:00LST".

Reply: it will changed to "at 12:00 LST"

14. P30925, L3-4, how did obtain "the observed O3 production (151 ppbv)"?

Reply: We will change the discussion in this part. On June 22, increase of O_x concentration on a daily basis was 60 ppbv (sum of total mixing ratio of O_3 from 06:00 to 17:00, subtracting a background value ~25 ppbv), however only small fraction of O_3 (14 ppbv on a daily basis) was *in-situ* photochemically produced on the basis of RACM2 model. It suggested that O_3 in the daytime at observation site was owing to direct transport from upstream area.

15. Table 1, units are missing. And you cannot say "the mixing ratio of black carbon (BC)".

Reply: I will add a unit of black carbon concentration in Table 1.

Reply to the comments of Anonymous Referee #3

We greatly appreciated the reviewer for providing many insight comments and considerable suggestions. We would like to modify our manuscript and clarify the discussions according to the specific comments. The detailed replies are as follows.

1. P30914, L4: Please state the dates of the field campaign in the abstract (month/year/duration).

Reply: Observation period of field campaign will be added in the abstract.

P30914, L11: State the averaging period of the 100 ppbv (hourly?)

Reply: Averaging period is 60 minutes, and we will replace the sentence with "hourly averaged O_3 concentration at site frequently exceed 100 ppbv"

P30914, L20: Later in the paper we learn that 15 NMHCs were measured. Somewhere in the paper please compare your work to other studies of biomass burning to give a sense of what fraction of important ozone precursors were measured, and which were missing. Even in campaigns where 100+ NMHCs are measured from biomass burning, there are still dozens of unidentified compounds that can contribute to ozone formation. For this paper, which compounds were measured impacts how the '35 and 23%' are interpreted; the more NMHCs are measured probably the lower the solvent fraction becomes as the aromatic fraction becomes relatively less.

Reply: As suggested by referee, only limited NMHCs species were measured during our field campaign. To avoid misleading readers, we will make clear in the revised manuscript that there is dozens of NMHCs compounds was not measured during observation, and some of them also contributed to *in-situ* O_x photochemical formation. Considering this point, the contribution of different sources to O_x production will be reanalyzed. Another important point, the photochemical processes of more reactive VOC species during transport will affects the interpretation of PMF analysis, and the anthropogenic factor at different photochemical ages may mistakenly be attributed to independent sources. In the revised manuscript, we will reduce the PMF factor number from 6 to 3, and the previous explicit classification (such as vehicle, fuel evaporation etc.) is not used any more.

P30914, L11-13 and 21-22. L11-13 states that O3 production was large when there was substantial NOx, but L21-22 states the opposite, that large NOx in the morning inhibited O3 production. Please clarify.

Reply: the sentence in L12-13 will be replaced with "in particular when the site was characterized by a prevailing southerly wind that brought substantial amounts of NMHCs and NO_x emitted from urban area"

P30915, L25: On the other hand, ozone production can be suppressed on short timescales due to titration with NO when NOx levels are high. Following from the above comment, perhaps present a brief discussion of impacts of high NOx on O3.

Reply: we will add brief discussion about the titration reaction between NO and O_3 when NO_x concentrations are high in the introduction section. In the revised manuscript we will present a typical O_3 pollution case (on June 19, 2010) to demonstrate such effect, and the mixing ratio of O_3 decreased to only 4 ppbv when ambient NO_x concentration was the highest (57 ppbv).

P30916, L15: State which year the measurements were made.

Reply: the sentence will be replaced with "from middle of May to the end of June 2010"

P30916, L24: Minor point but it's 'CO' on L24 but 'carbon monoxide' on P30917 L6. Define carbon monoxide as CO the first time it's mentioned then use CO thereafter. Make sure everything is defined (e.g., NOy on P30917, L13).

Reply: "carbon monoxide" on P30917 L6 will be replaced with "CO", and definition of NO_y (NO_x, HNO_3, N_2O_5, PAN etc.) will be added.

P30917, L18: Somewhere early on in the manuscripts please state which specific NMHCs were measured (or refer the reader to Table 1). Which major O3 major precursors are included and which were not measured? How might this affect your analysis?

Reply: we will specify the NMHCs species which measured by GC-FID-MS at P30917, L18, and tell the reader that the unidentified NMHCs compounds (e.g. HC3: dimethylbutane; HC5: methyl-pentane, dimethyl-pentane, trimethyl-pentane; OLI: trans/cis-2-pentene, trans/cis-2-hexene) have some impact on the O_3 production calculation by RACM2 model, though their influence was limited.

P30917, L25: So presumably the 58-VOC mixture encompasses the 23 NMHCs/OVOCs presented here?

Reply: Yes.

P30917, L26: Please state how many oxygenated VOCs were measured (or refer the reader to Table 1). Change 'MCAR' to 'MACR'. Define all acronyms.

Reply: we will refer to Table 1 here and the misspelled acronyms "MCAR" will be replaced by "MACR".

P30917, L29: As well as the reference to Kudo et al., basic analytical information (precision, accuracy, etc.) still needs to be given here for the 15 NMHCs and 7 OVOCs.

Reply: The detection limit NMHCs by GC/MS was 0.002 – 0.005 ppbv at Signal/Noise ratio = 3, and detection limit of PTR-MS was estimated to be 0.01 – 0.08 ppbv. The uncertainty of measurements was estimated to be less than 15% in this study. We will mention basic analytical information in the revised manuscript.

P30918, L16: 'The missing data were linearly interpolated.' Please discuss what error/uncertainty this might introduce.

Reply: The NMHC species were measured at time resolution of less than 2 hours, however the time step of input of RACM2 model was at time resolution of 5 minutes. For the convenience of model run, the data gap was linearly interpolated, and such treatment did not influence our results. During the observation period, the VOC data between June 13 and June 17 were missing because of instrumental maintenance. For the other high- O_3 pollution days, all the instruments were working well and the dataset was available for the RACM calculation.

P30919, L5: Define RO2 and phi separately rather than together.

Reply: we will define RO_2 and phi separately in the revised manuscript.

P30920, L9: I believe this is the first time some specific NMHCs are being named (please name them sooner).

Reply: we will give brief introduction of observed NMHCs species earlier in the context.

P30920, L13: What is the 'G-space'? What is 'F-peak'? Try to ensure that the text is accessible to readers who are less familiar with PMF.

Reply: The G-Space shows scatter plots of one factor versus another factor, which can be used to assess rotational ambiguity as well as the relationship between source contributions. A more stable solution will have many samples with zero contributions on both axes, which provide greater stability in the PMF solution to less rotational ambiguity. A solution or combination of sources may also have no points on or near the axes, which results in greater rotational ambiguity (PMF v5.0 User guide). To avoid confusing readers who are less familiar with PMF, we would like to concise our analysis and remove the sentence in P30920, L13.

P30920, L20: Define 'sigma units'.

Reply: Hysplit model uses terrain following sigma coordinate, and 0.01 sigma unit is about 250 m.

P30920, L23: 'To better review the impact of emissions on the surface layer, only the footprint region of the air mass in units of residence time was demonstrated in the following analysis.' This sentence is a bit confusing because for me footprints and residence times do not intuitively go together, and the next sentence seems to define the footprint in terms of distance.

Reply: In the revised manuscript, we will use the number fractions of trajectories in the grids to represent their potential influence of anthropogenic emission.

P30921, L7: The reader still doesn't know which year the measurements were made.

Reply: The year "2010" will be added in the sentence.

P30921, L9-11: Is the 147 ppb O3 max also an hourly value? On L13 CO is presented as a daily average. On L16 it's not clear if the NOx and NOy means are hourly or daily. P30917 L20 says that the time resolution of the NMHC measurements is ~2 hrs . . . is the average value of 200 ppbC on P30921 L19 a 2-hour average? Is the 451 ppbC maximum 'at 05:00' actually a 2-hour average? Because many different averages are presented in this paragraph, please state the time period (hourly, daily, etc.) each time an average is cited.

Reply: Sorry for our confusing discussion in this paragraph. We will state the averaging period when averaged values are used. We will replace the discussion in section 4.1 with the followings:

Temporal variations of the mixing ratios of O_3, NMHCs, NO_x, NO_y, CO, and wind speed and direction are shown in Fig. 2. Statistics on the NMHC species are listed in Table 1. During the observation periods, five pollution episodes (June 10th to 12th, June 14th to 17th, June 18th to 20th, June 22nd, and June 23rd in 2010) were clearly identified based on NO_x variations, and on 4 different days (June 15th, June 19th, June 22nd, and June 23rd), the hourly averaged O_3 mixing ratio exceeded 100 ppbv (dashed line as shown in Fig. 2). The highest O_3 concentration (hourly mean: 140 ± 3 ppbv) occurred at 17:00 CST on June 15th, 2010. We found that the observation site was affected by the intensive open burning of crop residues in the surrounding agricultural according to the sharp increase in ambient CO levels. The hourly CO mixing ratio at 12:00 CST on June 15th was as high as 1309 ± 91 ppbv. O_3 precursors outflowed from the urban areas to the south also contributed to the build-up of ambient O_x at the site, because obvious enhancements in the NO_x mixing ratio (hourly mean: 22 ± 13 ppbv) were observed with the prevailing southerly wind (SE-SW sector), and the ambient O_3 mixing ratio decreased to almost zero due to titration reactions when the nitrogenous species were preferentially present in the early morning pollution days. The maximum concentration of NMHCs (hourly value: 443 ppbC) occurred at 07:00 CST on June 19th when the hourly averaged NO_x and NO_y mixing ratios were 42 ± 2 ppbv and 60 ± 1 ppbv, respectively, and the hourly O_3 mixing ratio was found to be 124 ± 5 ppbv at 12:00 CST due to strongly photochemical processes in the daytime.

P30921, L11, 16, 19, etc.: Add an error bar to this average (and all others).

Reply: Standard deviation will be added for each averaging calculation.

P30921, L12: What direction was the burning coming from?

Reply: Observation site was prevailing southerly wind, indicated that OBB to the south of site impact our observation.

P30921, L15-17: It may help to refer the reader to the wind roses in Figure 4 here; with- out wind direction plotted in Figure 2 it's difficult for the reader to connect the 'obvious enhancement' to southerly wind.

Reply: We will add wind information in the Figure 1.

P30921, L15: High O3 when air is transported from the megacity seems to contradict the previous sentence (highest O3 during intensive open burning).

Reply: We will revise the discussion as I replied in the answer to the previous question.

P30921, L25: This is the first time we're learning that the field campaign was in 2010. Please state the exact dates of the Expo so the reader can see the extent of the overlap.

Reply: We will mention the year that our field campaign was performed in the previous section in the context.

P30921, L25: This sentence states that traffic increased, but the next sentence mentions emission controls and P30927 L11 suggests that NOx and NMHC emissions were actually reduced during the Expo. So did traffic actually decrease?

Reply: As mentioned, strict emission control policy was implemented in Shanghai and Southern Jiangsu province during Shanghai Expo 2010 period; however we have no direct evidence that how much NO_x and NMHCs were reduced. To avoid misleading, we do not discuss the effects of emission controls in the revised manuscript in P30921 L24-P30922 L4.

P30922, L10: What does 'daily increment' mean? Do you mean amplitude? Same comment on L13-14.

Reply: We will replace the sentence with "… a predominant single peak distribution with a amplitude of 94 ppbv." on L10 and also on L13-14.

P30922, L14: Is '07:00' the average from '07:00-08:00'? Same question on L18-19. How the data are plotted can also be included in the Figure 3 caption. For NMHCs (Fig. 3g), if the time resolution is 2 hrs (P30917 L20) why are hourly data plotted?

Reply: The time label 07:00 LST means the average from 07:00 to 08:00 LST. We are sorry for misleading of the sentence on P30917 L20. Before 13 June 2010 the NMHCs species were measured at a time resolution of 2 hours, and the measurement interval was changed to 1 hour from 16 June 2010 to the end of the field campaign, that is why we could plot its hourly diurnal variation. We will clarify this point in the revised manuscript.

P30922, L17: '38.6' seems to have too many significant figures given the very large error bar shown in Fig. 3c. Please add the error here and adjust the sig figs accordingly. Also this peak appears to be for 4 a.m., not the broader peak from 0:00-8:00 a.m.; please revise the text. Likewise on L22 state that 57 ppbv is the hourly peak of the broader range.

Reply: We will revise this part according to suggestions as followings:

Diurnal variation in NO_2 at the site showed evident increase at night with a predominant peak (39 ± 18 ppbv) at 04:00 LST, evidently different from the features (two peaks at 09:00 and 18:00 LST during rush hour) observed at the urban site of the Shanghai megacity. The similar pattern was found for the diurnal variation of NO_y (Fig. 3d) on high- O_3 pollution days, which had a broader peak (57 ppbv) from 04:00-09:00 LST and gradually decreased due to photochemical consumption in the daytime.

P30922, L16-19: Please state that you have shifted from discussing the red line in the first half of the sentence to the gray line in the second half of the sentence; currently it is confusing because the reader does not know that the two peaks are no longer about the red line.

Reply: the second half of the sentence will be changed to "distinct from that (two peaks at 09:00 and 18:00 LST during rush hour, as shown by the gray line in Fig. 3c) observed at the urban site of the Shanghai megacity."

P30922, L20: Why do these different peaks suggest transport rather than in situ production? Also what is the typical transport time from Shanghai to the site?

Reply: We will clarify this point. As mentioned by referee, the titration reaction between NO and O_3 might contribute part of NO_2; however it was far from enough to explain such dramatic increase of NO_2 at night. As shown in Figure 1, during high-NO_x pollution period NO_x/NO_y ratio was normally larger than 0.5, it indicated that the air mass was just slightly aged, and the NO_x emission sources seemed not far away from the observation site. We found that there are numbers of small county towns (e.g. Qidong, Latitude: 31.808°, Longitude:121.658°) and mega-cities (e.g. Nantong, Latitude: 31.977°, Longitude:120.900°,) to the south of observation site, which may contributed to the high NO_x level at site. The transport time from the larger city was about less than 4 hours providing a moderate wind speed (3 m/s) . To avoid misleading, we discuss the source region case by case on the basis of footprint analysis, rather than the just transport from Shanghai city.

P30922, L21-25: In order for the general reader to follow these arguments you will need to define NOy and NOz earlier in the text (e.g. P30917 L13). Also add an error bar to 32.6 and reduce the number of significant figures as appropriate.

Reply: We will define NO_y and NO_z earlier in the text. The sentence in L25 will be replaced with "… with a maximum value of 33 ± 10 ppb at 12:00 LST"

P30922, L24: Fig. 3e shows NOx/NOy, not NOz. Similar comment for P30923, L1.

Reply: We will correct the mistakes.

P30923, L7: Add an error bar to the mean of 0.34 so the reader can see that it didn't vary.

Reply: The standard deviation (± 0.1) will be added.

P30923, L10: The morning error bars are large so 176.4 has too many significant figures. Please adjust and add its error bar. Also add an error bar to 48.6 and 62.8.

Reply: The standard deviation will be added. The sentence in L10 will be changed with "…, on the high- O_3 pollution days, total hourly averaged NMHCs concentration varied significantly (186 ± 200 ppbC) in the early morning (from 00:00 LST to 08:00 LST), and it gradually decreased to 65 ± 55 ppbC in the afternoon (from 12:00 LST to 18:00 LST). On low- O_3 pollution days, the NMHCs concentration was constantly low with a mean of 58 ± 46 ppbC, implying that contribution from the transport of urban emissions was limited."

P30924, L6-7: Please add error bars to each mean that is reported. Same comment on L20, 25. Same comment on P30926 L5-6.

Reply: The standard deviation will be added.

P30926, L7: The text makes it seem like the stagnant air mass persisted for 4 days (June 19-23) but Figure 2 indicates two separate pollution events on June 19 and 23. Should 'air mass was' be 'air masses were'?

Reply: We will change the sentence to "air masses were…"

P30926, L15: The runs are constrained by June 19 data but calculations are also presented for June 23 (L18). Were constraints from June 23 also used?

Reply: The sensitivity runs were constrained by data on June 19, because the site was prevailing southerly wind all the daytime. The footprint analysis indicated that the anthropogenic emission from urban area in the YRDR has great impact; On June 23, observation site was prevailing southerly wind in the morning. It changed to easterly wind in the afternoon, and the buildup of O_3 at site mostly resulted from direct transport of O_3 from marine area. Considering the representativeness of polluted air masses, we just use the data on June 19. To avoid misleading, we will remove the case on June 23 from the Figure 7.

P30926, L25: This sentence is confusing because 'which' should be about NOx but is actually about NMHC/NOx. Please rework. Also 30-50 compared to 5 seems much larger than 'slightly' up.

Reply: The sentence in L25 will be replaced with "… a substantial amount of NO_x, and NMHCs/NO_x ratio increased evidently up to 30-50 ppbC $ppbv^{-1}$ due to rapid NO_x photochemical loss."

P30927, L2-3: In both lines change 'radial' to 'radical'.

Reply: The mistake will be corrected.

P30927, L23: Is 1.4 similar to 2.0-2.2 (within the expected range) or is it actually statistically lower? An error bar on 1.4 would help (and on the values of 2.0 and 2.2 if they're available). P30927, L27: '74.4' and '55.4' are too precise. Also on P30928 L1 a 20% difference is not 'slightly' higher.

P30927, L27+: These arguments don't seem to be adding up. The fraction of isoprene explained by OBB (74%) is surprisingly high to me, and it is also is high compared to the YRDR inventory (54%; P30928 L2). Then on P30928 L3-4 the furan/isoprene ratio is too high, which is explained by isoprene being oxidized (so depleted). Please try to reconcile these arguments.

P30928, L6-7: How are you calculating the value of 3.5 from the Akagi paper? In their Table 1 column for crop residue they give an EF (g/kg) of 0.11 for furan and 0.38 for isoprene. So a furan/isoprene ratio would be 0.11/0.38 = 0.29 g/g. The mass ratio of 3.5 would be for isoprene/furan (0.38/0.11 = 3.5). I haven't checked the Christian paper; is it also isoprene/furan? Is the 16.5 presented here isoprene/furan or furan/isoprene? Anyway it seems that the differences could be even larger than what was presented.

P30928, L7: 'photo-oxidation of isoprene in the summer season' seems speculative. Akagi et al. state the sources of the crop residue data; were these measurements also performed in summer? If so then this argument may not hold. Please investigate.

P30928, L7-9: 'and the furan/isoprene ratio would be approximately equal to that of previous studies if MVK/MACR were taken into consideration'. Taken into account how? If you believe this to be the case please provide evidence.

P30928, L15: You could further this argument by checking if the i-butane/n-butane ratio is consistent with LPG or NG.

P30928, L24: What was the benzene loading on Factor 1? Were you surprised to see benzene load onto a different factor than TEX? Section 5.4.1: For each factor, each time a compound is associated with a specific source (acetonitrile for biomass burning, i-pentane for gas evaporation, NO2 for fossil fuel combustion, etc.) please provide a *reference.*

Replies to the comments from P30927 L23 – P30928 L24 together: We are very for unclear discussions in section 5.4.1. We will rework section 5.4.1 completely in the revised manuscript according to reviewer's insight comment. Firstly, since the identification of individual VOCs by PTR-MS is based on the mass number of protonated species, PTR-MS cannot distinguish isomers and is subject to fragments [de Gouw and Warneke, 2007]. PTR-MS signals at m/z 69 could be a mixture of isoprene and furan. Objectively distinguish them has always been a tricky issue. In this study, concentration of furan was calculated empirically as follows:

Furan (ppbv) = (m/z 69 (measured by PTR-MS) – Isoprene (measured by GC/MS)) × Isoprene sensitivity (PTR-MS) / Furan sensitivity (PTR-MS) → [Kudo et al., 2014]

Therefore, furan/isoprene ratio here actually includes large uncertainty. In the revised manuscript, we will delete the discussion of this part. To response to the comments of referee, mass ratio of furan to isoprene was 0.11/0.38 = 0.29 (Akagi et al., 2011) and 0.72/0.82 = 0.88 (Christian et al., 2012), and the sentence "The higher ratio in the present study was probably due to the photo-oxidation of isoprene in the summer season, and the furan/isoprene ratio would be approximately equal to that of previous studies if MVK/MACR were taken into consideration" will be deleted.

Another important point, as suggested by other referees and literature [Yuan et al., 2012], the photochemical processes of more reactive VOC species during transport will affects the interpretation of PMF analysis, and the anthropogenic factor at different photochemical ages may mistakenly be attributed to independent sources. In the revised manuscript, we will reduce the PMF factor number from 6 to 3, and the previous explicit classification (such as vehicle, fuel evaporation etc.) is not used any more. Factor 1 showed an obvious diurnal variation and consisted of a large fraction of high-reactive species (e.g. C8-C9 aromatics, 1-butene) and NO_2. Factor 2 did not have diurnal variability and had an abundance of low-weight alkanes and fewer fractions of high-reactive species and NO_2. As reported previously (Yuan et al., 2012), NMHCs photochemistry could lead to different time series for more and less reactive species, and PMF can detect these differences and attribute more or fewer reactive species to different factors, even though they originate from the same source. Therefore, we cannot identify the emission sources for these two factors. Instead, we attributed factors 1 and 2 to less-processed and more-processed air masses. Factor 3 was considered an OBB-related source due to the pronounced contribution of furan and acetonitrile with mass fractions of 64% and 50%, respectively. This factor also accounted for 72% of total isoprene, 51% of MVK + MACR (oxidation production of isoprene), and 52% of acetic acid. As shown in previous studies (Kudo et al., 2014), factor 3 would retain the source characteristics, as the OBB occurred near the observation site.

Accordingly, The discussion in this section will be modified in accordance to the new PMF classification. Here, we just showed the relative importance of each factor on the *in situ* photochemical formation of O_x for a typical heavy O_3 pollution case on June 19th, 2010 (hourly-averaged O_3 concentration: 124 ± 5 ppbv and one minute value: 168 ppbv at 12:00 CST). As shown in Fig. 8, the contribution of different sources to O_x production showed distinct variation patterns. Factor 2 was responsible for 60% (43 ppb) of the *in situ* photochemical production of O_x in the morning, followed by that of factor 1 (23%, 17 ppb), and the OBB-related factor only accounted for 17% of the total O_x production. In the afternoon, the relative importance of OBB-related factors increased and was responsible for 34% (12 ppb) of the photochemical O_x production.

Reference: Yuan, B. et al. (2012), Volatile organic compounds (VOCs) in urban air: how chemistry affects the interpretation of positive matrix factorization (PMF) analysis, J. Geophys. Res.-Atmos., 117, D24302, doi:10.1029/2012JD018236.

P30929, L1-20: The verb tense changes from present to past in this paragraph. Also sometimes using words like 'could' and 'may' makes it unclear if the statement is a result or a speculation (e.g. L10-13; L19-20). Please rework.

Reply: We will use past tenses in this paragraph.

P30929, L22: While you mean MVK+MACR (not MVK/MACR), earlier furan/isoprene was a fraction, so for clarity I suggest avoiding MVK/MACR if you don't mean a fraction and also to be consistent with L24-28. Same comment elsewhere in the paper and for other compound pairs.

Reply: We will use MVK+MACR in the manuscript.

P30930, L4: Are these mass-based ratios (like before with furan/isoprene)? If so please state this.

Reply: They are mass-based ratios, we will clarify this point.

P30930, L5-6: 'oxygenated VOCs (including MVK and MACR) might also be preferentially present in the OBB plumes'. This statement seems speculative. What evidence is there for this?

Reply: This sentence will be deleted.

P30930, L7: How were the numbers of 1.8 and 4.2 calculated? For example Akagi et al. don't provide MVK and MACR EFs for crop residue burning in their Table 1.

Reply: The discussion about MVK and MACR EFs in Akagi et al.,'s paper will be deleted. We will rework with this part as follows "... In the present study, the [MVK + MACR]/isoprene mass ratio was the highest, ranging from 3 to 25, suggesting that the impact of biogenic sources was not significant. Kudo et al., [2014] reported that observed normalized excess mixing ratios (NEMRs) of OVOCs enhanced with air mass age, implying that the isoprene might undergo further oxidization after being emitted."

P30931, L9: There are too many significant figures on these numbers from the literature (they can't be known that accurately). Are these annual averages? If so for what year?

Reply: We will reduce the significant figures and add the year (2007).

P30931, L13-14: Verb tense is changing again and the sentence is hard to follow. The paragraph is also a bit hard to follow . . . increased aerosols lead to decreased O3; O3 is assumed to increase if aerosols are reduced; aerosol reduction leads to decreased AOD but increased actinic flux; high aerosol leads to decreased JO(1D); etc. Just it seems to go back and forth between impacts of increasing and decreasing aerosols; perhaps there is a smoother way to present this.

Reply: We will use past tense in the sentence L14. We will rework with this paragraph as followings: "The incorporation effect of HO_2 loss into the RACM2 model calculation resulted in the O_3 production rate being reduced by 13 % on average. Huang et al. (2011) reported that primary anthropogenic emissions of $PM_{2.5}$ and PM_{10} in the YRDR were 1511Ggyr^{-1} and 3116Ggyr^{-1} in 2007, respectively, accounting for 11 and 17 % of the total emissions in China. It suggested that O_3 photochemical production has been depressed due to the presence of aerosol particles in the atmosphere. A study by (Gerasopoulos et al., 2013) found a 40 % reduction in JO^{1}D at a high solar zenith angle during high aerosol loading periods (AOD = 0.5– 0.7). Presumably, ambient O_3 concentrations could increase provided that particle emissions are significantly reduced while NO_x and NMHCs emission remain unchanged."

Table 1: Please also add max and min values and add units.

Reply: We will add max, min and units

Figure 4: The units for the mixing ratios of O3, NO etc. need to be given.

Reply: We will add the unit for the Figure 4.

Figure 6: The different lines also need to be defined in the legend (not just panel a).

Reply: We will add legend for a, c, e and g

Technical corrections:

P30914, L7: Suggest changing 'was observed' to 'was occurring'.

Reply: We will use "was occurring" as suggested.

P30914, L14: Change 'potential' to 'a potential'.

Reply: We will use "a potential"

P30915, L5: Change 'has' to 'have'.

Reply: We will use “have ” here.

P30915, L7: Omit ‘evident’.

Reply: The word “evident” will be removed

P30916, L1-4: The verb tense changes from present to past in this sentence.

Reply: We will use the same present tense.

P30916, L9: Change ‘have’ to ‘has’.

Reply: We will use “has”

P30916, L13: Change ‘Quantitive’ to ‘Quantitative’.

Reply: We will correct the misspell

P30916, L14: Change ‘measurement’ to ‘measurement campaign’.

Reply: We will adopt “measurement campaign”

P30916, L22: Change ‘area’ to ‘areas’.

Reply: We will use “areas” here

P30917, L18: Change ‘NMHCs’ to ‘NMHC’. Same comment on P30920, L7 and L11.

Reply: We will change NMHCs to NMHC.

P30917, L18: Remove ‘an’ for this sentence to work.

Reply: We will delete “an”.

P30918, L24: ‘since NO2 is known to exist at high concentrations comparable to O3 at the observation site’. The wording in this sentence is a bit confusing.

Reply: This sentence will be deleted in the revised manuscript.

P30920, L1: Do not capitalize ‘Where’. P30920, L6: Change ‘numbers of NMHCs sources’ to ‘number of NMHC sources’? P30920, L12: Change ‘alkane’ to ‘alkanes’.

Reply: We will use lower case “Where”; “alkane” will be changed to “alkanes”; P30920,L5-L8 will be deleted.

P30920, L13: Change ‘seed’ to ‘seeds’. P30920, L18: Capitalize all of ‘HYSPLIT’. P30921, L1: Define FLEXPART. P30921, L10: Change ‘dash’ to ‘dashed’. P30921, L10: Change ‘the Fig.’ to ‘Fig.’ P30921, L12: Change ‘agriculture’ to ‘agricultural’. P30922, L22: Change ‘boarder’ to ‘broader’. P30922, L23: Use a subscript in ‘NOz’.

Reply: We will change “seed” to “seeds”; “Hysplit” will be capitalized; We will provide hyperlinks for FLEXPART model; “dash” will be changed to “dashed”; article “the” will be deleted; the word “agriculture” will be changed to “agricultural”; The word “boarder” will be changed to “broader”; NOz will be changed to NO_z

P30923, L25: Change ‘e.g.’ to ‘i.e.’

Reply: We will remove “(e.g. shanghai)” because this information is not very necessary.

P30924, L12: Could change ‘Discussions’ to ‘Discussion’.

Reply: We will change “Discussions” to “Discussion”

P30925, L2: Change ‘productions’ to ‘production’.

Reply: We will change “productions” to “production”

P30925, L3: Omit ‘the’.

Reply: The article “the” will deleted.

P30925, L15-16: To avoid repetition change ‘observed at the observation site’ to ‘ob- served at the site’.

Reply: The word “observation” will be omitted.

P30926, L26: Change ‘the observation in’ to ‘previous observations in the’. P30927, L9: Change ‘from’ to ‘from the’. P30927, L20: ‘Alkane’ and ‘alkene’ should be plural. P30929, L4: Change ‘as’ to ‘to be’.

Reply: We will add “previous” in L26; article “the” will be added in L9; The discussion in section 5.4.1 will be changed, and we will use plural form of Alkane and alkene.

P30929, L15: Change ‘air mass was’ to ‘air masses were’. P30930, L10: Change ‘observation’ to ‘the observation’. P30930, L11-12: Change ‘little amount’ to ‘small amounts’.

Reply: “air mass was” will be changed to “air masses were”; “observation” will be changed to “the observation”; “little amount” will be changed to “small amounts”

P30930, L10-14: This is a run-on sentence. Please fix. P30930, L14-16: Please fix the grammar. P30930, L25: Omit ‘refers to the paper,’. P30931, L19: Change ‘will more’ to ‘will be more’. Figure 5: Caption needs a period.

Reply: We will rework with the L10-16; “refers to the paper” will be deleted; “will more” will be changed to “will be more”; a period will be added at the end.