

## General Comments

This paper presents OH reactivity measurements in Beijing and Heshan, China using the CRM technique. Missing OH reactivity was found in both studies and its impact on ozone production efficiency was assessed using a box model. The scope of this work is important and can improve our understanding of ozone production and I think this work is worth publishing. My big concern is the uncertainty in the OH reactivity measurement, which may reduce the significance of missing OH reactivity on the ozone production. Another concern is that in the ozone production efficiency (OPE) calculation, the NO<sub>x</sub> loss due to organic nitrate formation is omitted, i.e., only the formation of nitric acid from OH + NO<sub>2</sub> is considered. This could overestimate the OPE depending on the relative importance of organic nitrate production over nitric acid production in these two environments. Overall I found the manuscript needs much improvement in English. The authors need to clarify many things in several parts of the manuscript (see Special Comments below), especially the discussion of the effect of the measurement uncertainty on importance of missing OH reactivity and the clarification on how the measurement corrections were done due to interference of humidity and NO. In addition, I would ask the authors to consider the following special comments in their revision.

## Special Comments

1. L.2: The country should be added, i.e. “Case studies in Beijing and Heshan, China”
2. L.22-23: with a detection limit of 5 s<sup>-1</sup> stated in L.198, it is not possible that two decimals in the OH reactivity values can be significant. Integer numbers probably enough.
3. L.23: it should read “Measurements in Beijing presented ...”
4. L.25-26: need to define missing OH reactivity here. I can guess it is the difference between measured OH reactivity and OH reactivity calculated from measured OH reactants. If so, state so.
5. L.32: “However, the model failed to explain the missing reactivity in Heshan,”, but was the box model able to explain the missing OH reactivity in Beijing?
6. L.35-36: it should read “...when the model is constrained by the measured reactivity...”.
7. L.48: remove “researches” or change it to “calculations”.
8. L.70: “...the 75% missing reactivity in Paris in MEGAPOLI under continental air masses influences”, need a reference for this statement. It should be mentioned that missing OH reactivity in each study depends heavily on the completeness of measurement suite, especially VOC species, so the next paragraph can follow.
9. L.89-90: change “...in one case could increase reactivity by over 50%...” to “...in one case which could increase reactivity by over 50%...”.
10. L.91: an increasing concern.
11. L. 92: What is the ozone level for Grade II of China National Ambient Air Quality Standards?
12. L.94-95: “...it appears there is an increasing trend for ozone in Beijing and other area...” for what time frame? Recent years?

13. L.104: change those to which.
14. L.106: from the total OH reactivity.
15. L.109: change "...two intensive observation datasets conducted..." to "...data from two intensive field studies conducted..."
16. L.114-116: consider to change this sentence to: "The possible missing reactivity and its importance for the ozone production calculation are discussed."
17. L.125-126: consider to change this to: "a 14.9 m Teflon inlet with an outer (I assume) diameter of 3/8 inch..."
18. L.136: Some impurities in dry air and nitrogen could also be photolyzed.
19. L.142-143: total ambient (?) OH reactivity is calculated as...
20. In Fig. 2, the color for symbols with different standards is not clearly shown. Consider to use different symbols and/or change to different colors for symbols and lines with a better color contrast.
21. L.155-156: What is the "uncertainty range for all calibrations"?
22. L.157-165 about correction due to humidity: it is not clear how exactly this correction was done based on Figure S2, where no labels for x and y axes are given so we really do not know what is plotted here. If the pyrrole signal versus relative humidity are plotted, why there are negative values?
23. L.166-182 about the correction due to NO: in Fig. 3 the y axis is labeled as delta reactivity. Is this the difference between calculated (standards) and measured OH reactivity? In the legends of the figure, there are reactivity numbers (60/120/18 s<sup>-1</sup>) and I assume these are based on the OH reactivity calculated from the contents in the standard gases. If this is correct, why can the delta reactivity be 300-600 s<sup>-1</sup>? As stated in L.173-174, the "measured" reactivity decreased as the NO mixing ratio increased. If so, the measured OH reactivity should be lower than the calculated values and the difference should be also less than these numbers (60/120/18 s<sup>-1</sup>). Please clarify.
24. L.192-193: Was the correction associated with HONO interference also applied to the measurements in both sites, or to the measurements in Heshan only, since it looks like there is no HONO measurements in Beijing from Table S1.
25. L.198: 2σ instead of 2δ.
26. L.198-201: is the uncertainty of 20% for 1σ or 2σ? Shouldn't the uncertainty associated with NO correction be taken into account?
27. L.248-251: it is not clear what output results from the box model was used in the calculations, time dependent results or steady-state result? The authors mentioned both a time-dependent mode of 5 min and steady-state conditions with a 3-day spin-up time. Please clarify.
28. L.263: P(NO<sub>z</sub>) should also include the production rates of organic nitrates which can be calculated using the box model results. Depending the fraction of organic production rate in the total NO<sub>x</sub> consumption rate, the OPE could be significantly over-estimated.
29. L.272: include units for the measurement results in Heshan. Are the errors standard deviations?
30. L.279: (O<sup>1</sup>D).
31. L.281: is 93 ppbv for hourly or 8-hour maximum?
32. L.282-284: How come that VOC concentrations in Beijing and Heshan are the same (i.e., Table S3 and Table S4 are identical)? Alkanes made up over 60% of the summed VOCs in Beijing. Is this in terms of concentration or VOC reactivity? Please clarify.

33. L.294: photochemical age is mentioned here but it is not defined until next paragraph. It's not presented in Fig. 6-7 either.
34. L.314: please define LTC.
35. L.325: again the two decimals are not significant considering the relatively large uncertainty of  $5 \text{ s}^{-1}$  in the measurements. Please correct all reported numbers for OH reactivity.
36. L.330-331: the morning rush hour peak could be because of a shallow boundary layer.
37. L.335-338: I don't think the little variations in OH reactivity on clean day can fully explain the less variability of OH reactivity in Heshan and in Beijing. It's probably because the air sampled in Heshan is more aged (as the authors have discussed) and regionally mixed than in Beijing.
38. L.353-354: please give the absolute missing OH reactivity values in  $\text{s}^{-1}$  for both location. A comparison between the missing OH reactivity and the combined uncertainty of measured ( $5 \text{ s}^{-1}$ ) and calculated (from the measured species) OH reactivity is needed in order to see if the missing OH reactivity is significant. The uncertainty of measured OH reactivity should be also discussed somewhere in Section 4 when the contribution to the missing reactivity is discussed.
39. L.357: the entire campaign.
40. L.363-364: "the relative reactivity compared to NMHCs mixing ratios were higher." Higher than what?
41. L.399: the sentence, "We found only one dataset in 2005 measured by NOAA (Liu et al., 2009)." is not clear to me. One dataset of what?
42. L.400-402 and Fig, 11 lower panel: missing OH reactivity is plotted against the reactivity assumed from 4 branched alkenes. What are these 4 alkenes; are they representative for the missing alkenes; and how is the calculation performed? Please clarify.
43. L.402-403: consider to change this to: "even with the mixing ratios of the 4 branched alkenes measured in Beijing in 2005, the reactivity..."
44. L.419-420: need a reference for the statement that the mixing ratios of branched alkenes could be lower than 0.1 ppbv. The site in Beijing is only a few hundred meters from major roads and can easily get influenced by vehicle emissions.
45. L.425-426 and Table S5: not sure if I understand the "major secondary contributors to modeled reactivity. Why only these species are listed in Table S5? Are they related unmeasured intermediates that are calculated in the model? Need definitions for the acronyms in the model (ALD, DCB, etc.).
46. L.460-462: It's not clear to me how the remaining missing reactivity were allocated into different intermediates. Are these intermediates constrained (remain constant) in the box model run in the second scenario?
47. L.466: on average. Also please give absolute values, i.e., increase from XX to YY.
48. Section 4.4: again, the OPE needs to be recalculated by including the production rate of organic nitrates in P(NO<sub>z</sub>). This may change the picture currently shown in Fig. 14.
49. L.497-499: Need to include the modeled OH reactivity in Fig. 11. Without this, it is hard to assess this statement that missing OH reactivity can be reconciled with modeled intermediates that were not measured. Also this statement seems in contrast with the statement in L.395-399, where the author stated that unmeasured primary VOCs, especially branched alkenes, are responsible for the missing OH reactivity. Please clarify.

50. L.507-510: probably add a sentence stating that efforts to reduce the uncertainty of OH reactivity measurements based on the CRM technique to increase the confidence of results as shown in this work.
51. Fig. 5: please give the Grade II of National Ambient Air Quality Standard for ozone and PM<sub>2.5</sub> in the caption. Also it seems the two red lines in Fig. 5-a and Fig. 5-b are different, one above 80ppbv and the other below 80 ppbv. Please clarify.
52. Fig. 6: the yellow (or brown) lines show the NO fraction (not percentage) in NO<sub>x</sub>. Please correct it.
53. Fig. 11: please plot the modeled OH reactivity, the same as in Fig. 12. Is the gray area along the red line showing the uncertainty of the measurement? If so, please state this in the caption.
54. Fig. 14: references for OPEs in other studies should be given.
55. Again, Table S5 and Table S6 are identical.
56. P.3 of the supplement: in Alkenes, ethane should be ethene.