We thank the two referees for their reviews of our revised MS. The Editor has asked us to respond to the comments in Report#1 from Anonymous Reviewer #3.

Below we reproduce the reviewer's comments in black normal type. Our response is given in blue type and changes to the MS are given in red type, unless the change is a major change (e.g. moving of material, removal of redundant material), in which case the revised MS with the tracked changes will indicate the changes that have been made (some minor changes were made once tracked changes were accepted).

Report #1 (Anonymous referee #3)

This manuscript describes observed OH, HO2, and RO2 radical concentrations and OH reactivity from the ICOZA campaign at Weybourne, UK, during July 2015 to investigate the degree of our process understanding regarding the atmospheric radicals and ozone formation. As this manuscript already experienced first round of reviews, I basically assumed or found that fundamental issues were mostly resolved. The budget analysis fully constrained by the observed terms is new and interesting. Nonetheless, I still identify two major issues as well as other minor points to consider for potential improvement, as listed below.

1. Consider removing redundant parts to achieve conciseness. I would recommend merging Discussion section to Results to avoid redundancy. For example, discussion about the RO2+NO reaction rate coefficient appears several times; from lines 796, 971, and 1076. Also, some auxiliary parts such as Section 4.3 could be moved to supplementary.

We agree that the combined manuscript can be significantly improved by further (and significant) shortening of the paper via removing redundancy as suggested by the reviewer. We have completely done away with the Discussion Section (Section 4) and renamed Section 3 "Results" as "Results and Discussion", and moved Section 4.3 to the SI as suggested (together with Table 4). In addition, upon moving the Discussion sections to the Results we were able to significantly shorten them (or in fact completely remove some parts). In fact we have gone further with some of the discussion, moving other sections to the SI, for example Section 4.1 and Section 4.2.

Please see the revised MS with the tracked changes showing to see how we have altered the document to achieve this. There is additional material now in the SI.

2. Considering CH3O2 is the main component of RO2 (line 1101, Figure S5), the k_14 rate coefficient of 2.3×10^{-11} cm3 molecule-1 s-1 (Line 403) might be too fast; four times faster than that for CH3O2 + HO2 (5.2×10^{-12} cm3 molecule-1 s-1). This might be the main reason why the analyzed budget for RO2 and HO2 is quite open. I believe this is treated adequately in the box model based on MCM and thus the gaps between the observed and modeled radical concentrations are valid; so the main conclusion of the manuscript may remain unchanged. However, many plots (e.g., Figures 9, 10 and 11) and the stated degrees of discrepancy would become different. The necessary shift in the RO2 + NO reaction rate coefficient to explain the observations that the authors propose (Line 747, pointing to section 3.10.1 instead of 3.11?) might become milder. Discussion in the lines 1009-1014 might also be affected.

We agree that using a single value for the RO2+HO2 rate coefficient for the budget analysis is a simplification. The treatment of this rate coefficient in the MCM is indeed more sophisticated. The rate coefficient for reaction of CH3O2+HO2 to form CH3OOH is 4.74×10^{-12} cm3 molecule-1 s-1 at 298K in the MCM. $k_{RO2+HO2} = 2.3 \times 20^{-11}$ cm3 molecule-1 s-1 is the generic RO2+HO2 rate coefficient which is used in the MCM for C3 RO2 and above (but this is scaled down by multiplying it by e.g. 0.52 for propane RO2, 0.63 for butane RO2 etc). Reducing $k_{RO2+HO2}$ in the budget analysis would reduce both DRO2 and DHO2, so would improve PRO2 and DRO2 agreement but would worsen agreement between PHO2 and DHO2. It would also likely close the PROx and DROx budget in the afternoon under SW conditions. Only reducing the rate of propagation of RO2 to HO2 can remedy both the PHO2 - DHO2 and PRO2-DRO2 discrepancies simultaneously and this is the focus of the discussion. Yes, by reducing this rate coefficient then the shift in the RO2+NO rate coefficient to explain the discrepancy in the budgets would in turn not need to be as much. We have altered the discussion accordingly in several places, and yes, it should have pointed to section 3.10.1 rather than to 3.11 (apologies, now corrected, although there is now only a Section 3.10).

For example in the Conclusion:

However, if the RO_2 + HO_2 rate coefficient were reduced then the reduction in the RO_2+NO rate coefficient to explain the discrepancy in the budgets would in turn not need to be by as much.

Minor but important issues:

3. Numbering of the equations and reactions need to be double checked. (E3) appears twice (Lines 293, 422). Line 452: k_4a, Line 456: the HO2+HO2 reaction appeared as R12 before and HO2+RO2 as R14. Line 693: E3 and E6? Line 703: E7 and 8 instead of E5-E6? Line 293: Some reaction rate coefficients are shown in a format as k_HO2+NO, instead of k_4a.

We apologise for equation (E3) appearing twice, this is a legacy of the merger of the two papers and not spotting all of the redundant material. We have checked for numbering of equations and reactions, and where they appear twice under different guises, or are incorrectly numbered, or are formatted inconsistently, we have harmonised in the revised MS. All equations now have k_N rather than by the name of the reaction. Please see the tracked changes document to see the changes in the equations.

4. Line 265: The 800m boundary layer height is kept constant. May it vary over a day? How well were the observed HCHO concentration levels reproduced by the model simulations predicting HCHO, with this boundary layer height assumption? Nighttime radical concentrations might be sensitive to this assumption.

The reviewer is correct in that the boundary layer will vary over a 24 hour period at this site. However, as there were no measurements of this parameter during this study, we have kept it constant. Regarding the model simulations of HCHO, in the SI, we have stated "The MCM-base model performance in simulating these carbonyls was assessed, where it was found that there was reasonable agreement for MVK+MACR on a diel average basis, but that HCHO concentrations were significantly overpredicted in the afternoon (data not shown)." It is possible that the model overprediction of HCHO may be due to an unrealistic boundary layer height in the afternoon, where it is likely be larger than 800m. It is worth noting though that modelled HO2 is still overpredicted in both the base and carb-constrained model, the latter where HCHO is a constraint used by the MCM

and so is not subject to any uncertainty in HCHO caused by an inappropriate boundary layer height used in the model. Nighttime radical concentrations (which in general are very low) might be sensitive to this boundary layer assumption, and we have stated this in the revised MS.

Section 3.4. "The nighttime modelling results might also be sensitive to the choice of boundary layer height, which was kept constant at 800 m in the model."

5. Line 708: HO2 to multi-ppbv is correct? (pptv?)

Yes, multi-ppbv level is correct.

Technical issues:

6. Line 155: The authors replied to a reviewer as 99.8% rather than 99.95%.

Sorry, this has been corrected now to 99.8%.

7. Line 340: Two different instruments were used for HONO observations. Which data did the authors use for the analysis?

We used the measurements of HONO made with the Long path absorption photometry (LOPAP) instrument. We have added a footnote in Table 1 to this effect.

[d] Used to constrain the MCM model

8. Line 360: Though stating all reaction rate constants are from MCM, it seems k_13 and k14 were rather set arbitrarily (line 403).

We address the point about the RO2+HO2 reaction above (R14, which is now R15). For the RO2+RO2 reaction (R13, which is now R14), the MCM uses k(CH3O2+RO2) = 3.5×10^{-13} cm3 molecule-1 s-1 (multiplied by a scaling factor (so at 298K is ~2.2E-13), then the scaling factor in the MCM varies with RO2. For the budget analysis a single value is indeed used, and we have stressed this further by adding to this sentence:

In this budget analysis, RO_2 radicals are treated as a single species, with generalised rate coefficients taken from the MCMv3.3.1: at 298 K and 1 atm.....

9. Line 688: Though the authors suggest missing ROx sources, this might be just from a too fast k_14 as mentioned above in the point 2.

We have added the following on this line to reflect this point (discussed above).

Alternatively, the rate constant for RO_2+HO_2, k_{15}, for which a single value is used in the MCM, may be too large for the mix of RO_2 present at the WAO.

10. Line 780. Where is the methane contribution to the OH loss?

We apologise that this was left out of the description of the OH loss, and also from Table 3 and Figure 13. A fixed concentration of 1900 ppm was used. The text has been modified as follows:

In terms of organic OH reactivity, methane (~ 10%, 12.5%, a constant mixing ratio of 1900 ppm was used)….

We have also been careful to be clear that missing OH reactivity is for the measured OH reactivity compared with the MCM modelled value, which includes unmeasured intermediates.

Figure 13 has been modified as follows:
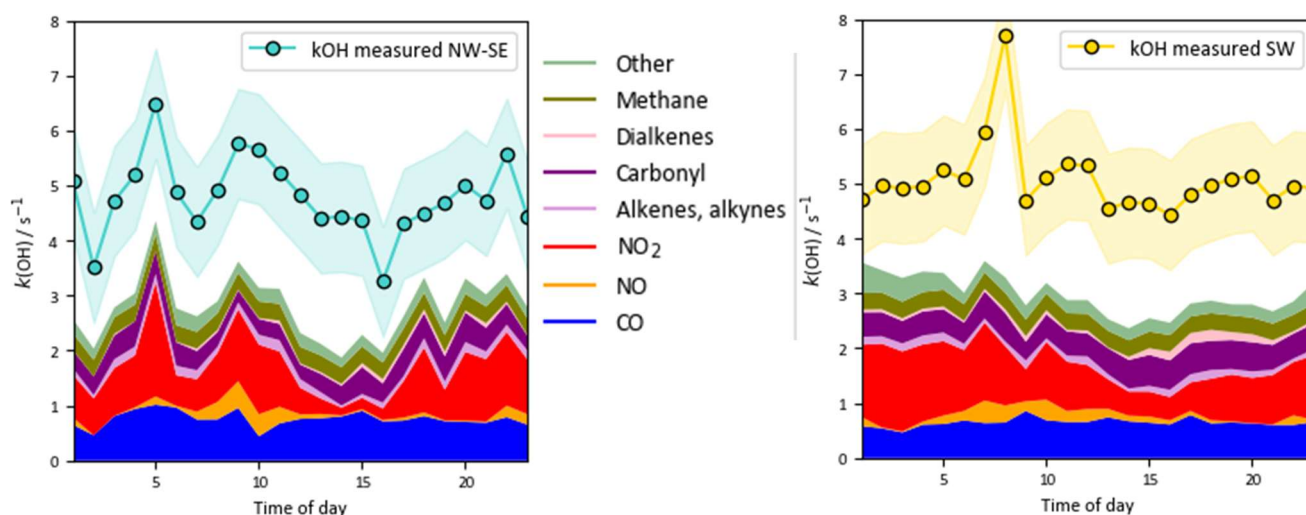
Figure 13. Median diel profiles of the OH reactivity calculated from measured reactants and comparison to measured OH reactivity, split according to wind direction. Average daytime contributions are given in Table 2. For interpretation of colours, please see the figure legend. Reactants in the "Other" class are listed in Table 2. The shaded area on measured k'_{OH} corresponds to the 1σ precision of ~1 s^{-1}. Model intermediates are not included here but their contributions are discussed in the text.

11. Line 855 and Section 3.11.2. How the relatively large uncertainties associated with the radical observations and with k_14 affect the conclusion here? If the estimated D(ROx) has large uncertainty, I cannot support this section.

We have decided on the basis of this comment, and also to further reduce the amount of material in the paper, to omit the section on the ozone production regime – Ln / Q, the analysis of which could be due to some uncertainty both due to the uncertainty in the radical measurements and in the chosen value of k_{14} (now k_{15}).

12. Lines 975-976. Smaller number of cited literature may be enough because this is not the main point of the study.

We agree that the referencing here is rather over-zealous, and so have reduced the number of references. The text now reads as:

The model HO$_2$ was somewhat reduced but the observations could still not be reconciled after inclusion of both HO$_2$ aerosol uptake (using γ_{HO2} = 1) and autoxidation chemistry (Bianchi et al.,

2019), which is now known to play a significant role in the gas phase oxidation of both BVOCs (e.g. Zha et al., 2017 and references therein) and anthropogenic VOCs (AVOCs) (Mehra et al., 2020 and references therein).

13. Figure 4, top panels. Can the authors point to the nighttime data for the observation panels too?

We thought about this, but for the observation panels the nighttime data are all clustered closer to the origin and given the density of points it is not possible to discern them easily, and no significant difference in slope is apparent (as we have already stated in the text). So it is not really possible to point to the nighttime data in a useful way. We have added to the caption the same wording as in the text:

For the model results, nighttime data exhibit a different RO_2 versus HO_2 slope (not observed in the observations).