We would like to thank the referee for their useful comments. We address these in bold below:

Referee 1:

This papers reports on measurements of the total OH reactivity (kOH) in the megacity London. Measurements of kOH are sparse. Therefore, the presentation of these measurements are important. Measurements of kOH are completed in this campaign by the detection of a large set of different VOCs. The authors nicely show, to which extend contributions of all measured OH reactants can explain measured OH reactivity. If also contributions of unmeasured oxygenated VOCs that are calculated by models are taken into account, the authors can show that kOH is explained. As the authors emphasize, missing reactivity that is discussed in literature always needs to be seen in the context of the completeness of measurement of OH reactants. These messages are clearly presented in the manuscript. However, the focus of the paper on these points could be improved specifically in the second part of the manuscript. The authors should consider the following points:

(1) I assume that the measurement site was close to sources of VOCs. This may have consequences for the applicability of a box model, which assumes that oxygenated VOCs can accumulate. If the transport time between the source and the site is short, concentrations of products could be over-estimated.

We have run the model both constrained and unconstrained to OVOCs that were measured (acetaldehyde, acetone as well as formaldehyde which is discussed in the paper) and find that the model has a tendency to under-estimates the concentration of these species, suggesting that there may be direct emissions of these species which are not considered in the model when run unconstrained.Therefore, unknown direct emissions of these species which would lead to model under-predictions may outweigh any expected model overprediction if full steady-state is not reached owing to short times between emission of parent VOCs and the site. We have discussed the sensitivity of the model predictions to the deposition term for all the unconstrained model species which determines the steady state concentrations reached and have shown the model to be reasonably insensitive to this.

(2) In my opinion, the presentation of instruments that are used for the analysis is not well-balanced. The major progress of this work is that a large set of VOC measurements was detected. However, the authors present their FAGE system in much greater detail than their GC systems. The authors may want to move Fig. 4 in the context of a more detailed description of these instruments. It is not clear to me, if all details of the FAGE system that are currently given are required.

The recent paper by Dunmore and co-authors (ACP, 15, 9983-9996, 2015) which we reference in the Experimental section details the VOC measurements and so we only provide a basic overview of these measurements in this paper. We do explicitly list all the VOCs individually included in the model in Table 1, however, which we feel is the key information for this manuscript. In the revised manuscript we will add further details on the effective saturation concentration range and sampling resolution of the

GC systems. The details of the FAGE instrumentation was included as we present OH and RO2 measurements from this instrument, however, in light of points (3) and (4) we will modify the FAGE details provided.

(3) The authors discuss results of model calculations for HCHO, OH, HO2, and RO2. This discussion seems partly misplaced in this paper, especially because the authors announce to publish details of the comparison of modelled and measured radical concentrations in a separate paper. The context of the discussion is not always clear here and the information that is given to understand the results is not sufficient. I understand that the authors want to emphasize the importance of the increased number of VOC species that could be detected in this campaign. In this context, the discussion of HCHO might be justified. However, I do not see that the discussion of OH and RO2 makes much sense here. Only sparse information is given concerning important other species like for example ozone, HONO, details of NOx, HO2, CO that are needed to understand the production and destruction of radicals. Specifically, the model run of OH which is constrained to measured HO2 requires at least to show the diurnal profile of measured HO2 and NO. I understand that this will be the topic of separate paper, but then I would suggest to remove the discussion here and to focus the paper on kOH.

In this section of the paper we were aiming to demonstrate the impact of under-estimating OH reactivity on predicted OH (and, hence, the oxidising capacity), on the predicted HCHO and also on predicted ozone production. We think it is important to show the impact of this as many of the earlier publications which have compared modelled or calculated OH reactivity to observations have often constrained the model or calculation with only a limited VOC suite.

The aim of this section was not to model the observed OH and so, as the referee suggests, we will remove the OH model-measurement comparison (figure 9). We propose to restrict our discussion on the impact of under-estimating OH reactivity simply to stating the % change in modelled OH as a function of the three OH reactivity scenarios.

We also agree that it is not necessary to compare [RO_2] calculated from OH reactivity to the observed [RO_2] and so will remove Figure 10 (and remove the associated discussion) from the revised manuscript. We think it is important and relevant to discuss the impact the extra OH reactivity (that derives from the extended VOC suite or model generated intermediates) on ozone production, however, so we propose to still include discussion on the calculation of [RO_2]ss.

(4) The authors use measured OH to calculate steady state RO2, that is then used to calculate ozone production rate. It seems a bit arbitrary to use measured OH, but not to use measured RO2 instead of calculated RO2. If the calculated RO2 concentration is used, the reader needs more quantitative information, if simplifications in this calculation are justified.

To evaluate the impact of the additional OH reactivity which derives from the extended VOC suite and model generated intermediates (which are not always considered when modelling or calculating OH reactivity) on ozone production we need to estimate the additional [RO_2] that derives from these species. Using the observed [RO_2] would not allow us to evaluate this.

(5) Concerning the simplification of the calculation of the ozone production rate, it is not clear, why the authors do not include ozone production from HO2.

As our aim was to show the impact on ozone production that arises from under-estimating the VOC fraction which contributes to the total OH reactivity, we chose not to include ozone production from HO_2. We hope the revisions proposed above will make our main aims clearer in the revised manuscript.

In my opinion, the discussion of the ozone production rate does not fit the topic of the paper well, because the only conclusion is that radical concentrations needs to be correct in the model. If RO2 is measured like in this campaign, however, there is no need to use kOH at all.

The aim was not to model (or calculate) ozone production rate – if we wanted to do this we would use our observations of RO_2 and HO_2. The aim was to show the impact on ozone production of under-estimating the VOC fraction of the total OH reactivity. Substantial ozone production was observed during the campaign and understanding the influence of more-complex VOCs (which are not traditionally measured) and modelled intermediates have on this pollutant is important and a relevant discussion point.

(6) Fig. 8 is mostly showing results that are already included in other figures. I would suggest to reduce this figure to the new aspects (physical loss, HO2 constrained model).

We will modify this figure accordingly.

Why do the authors use the model run, in which HO2 is constrained? If I understand the point correctly, the purpose is to get the modelled OH correct. In this case, however, the use of measured OH as constraint would make much more sense.

Yes, we constrained the model to HO_2 to allow the model to predict OH correctly. We chose not to constrain to the measured OH so we could evaluate the model’s ability to predict OH for diurnal comparisons presented in Figure 9. Regardless of the method chosen to reproduce the observed OH by the model, the impact of reducing the model OH on OH reactivity will be the same.

(7) Technical remark regarding figures: The font size and thickness of lines may be not adequate in the final publication for one-column figures.

We will assess this point at the production stage.