

We thank the reviewers for their insights and helpful suggestions. We feel that their feedback has led to changes that have strengthened the quality of the revised manuscript. Please find below the original reviewer comments in black, our responses in blue, and changes to the manuscript in red.

## **Reviewer #2**

This paper presented the measurements of OH, HO<sub>2</sub> and XO<sub>2</sub> at the PROPHET site in July 2016. OH and HO<sub>2</sub> were measured by FAGE with dedicated efforts to minimize the interference. XO<sub>2</sub>, the sum of HO<sub>2</sub> and RO<sub>2</sub>, were measured by an ethane chemical amplification. The measured radical concentrations were consistent with previous field measurement in 2008 and 2009, considering the meteorological difference. The measurements were compared to box model calculations using RACM2-LIM1 and MCM3.3.1 as a standard test with updated information of isoprene oxidation mechanisms, which enhance the OH concentration by 30% and 20%, respectively, compared to those without LIM1 chemistry, namely RACM2 and MCM3.2. However, this standard models overpredicts OH, HO<sub>2</sub> and XO<sub>2</sub> more than 60%. RO<sub>2</sub>+RO<sub>2</sub> accretion reactions were added to reduce the model-measurement discrepancy. Further radical budget analysis and model sensitivity tests indicated the model overprediction was related the missing radical sinks.

This paper is well-written and structured reasonable. The discussion is justified. I recommend the paper published in ACP after minor revision. Here is a few suggestions that might help to improve the paper.

1. There were several radical measurements conducted at the PROPHET site. It would be very useful to show an overview plot to compare different field measurements. In selection 3.2 and 3.3, the measurements from 1998, 2008, 2009, and 2016 are compared in different position. For example, a bar plot summarizing the diurnal maximum OH, HO<sub>2</sub>, RO<sub>2</sub> concentrations together with temperature, BVOCs concentrations would help the readers to interpretate the consistency/difference between several campaigns.

We thank the reviewer for this helpful suggestion. We have added Table S3 to concisely summarize the field campaigns conducted at the PROPHET site and added the following sentence to page 11 of the revised manuscript.

**These measurements are summarized along with those from previous campaigns at the PROPHET site in Table S3**

2. As the authors suggested the radical sink related to surface loss may be relevant to the model overprediction, it would be interesting to have more quantitative analysis. One could calculate the probability of radical loss upon collision on the ground/canopy similar to the aerosol uptake. In this case, the surface area per volume can be derived from the ratio of ground/canopy area to the volume of mixing layer.

We agree with the reviewer that this section should be expanded, though studies investigating radical uptake on canopy surfaces are limited in comparison to studies involving HO<sub>2</sub> uptake to

aerosol surfaces. We have added a short paragraph on pages 24 and 25 of the revised manuscript that quantifies the uptake coefficients that are necessary to account for the missing HO<sub>2</sub> and isoprene RO<sub>2</sub> sink given a leaf area index of 3.8 m<sup>2</sup> m<sup>-2</sup> and assuming a mixing layer height of 1500 m.

Similar to the above discussion, radical loss to surfaces within the forest canopy can be estimated using Eq. 2 where  $A$  now represents the ratio of the canopy surface area to the height of the mixing layer. Previous measurements at the PROPHET site reported a leaf area index (LAI) of approximately 3.8 m<sup>2</sup> m<sup>-2</sup> (Ortega et al., 2007). Assuming a mixing layer height of 1500 m, this suggests that an HO<sub>2</sub> uptake coefficient of  $\gamma = 5 \times 10^{-4}$  would result in a first order loss rate of 0.013 s<sup>-1</sup>, which could account for the proposed missing HO<sub>2</sub> sink. This uptake coefficient is lower than those measured for many atmospheric aerosols, but is similar to measurements of HO<sub>2</sub> uptake on organic aerosols (Lakey et al., 2015). Similarly, an uptake coefficient of  $\gamma = 1.7 \times 10^{-3}$  for isoprene peroxy radicals would result in a first order loss rate of 0.024 s<sup>-1</sup> and could account for the missing radical sink. These results imply that loss to surfaces within the canopy could be a substantial radical loss mechanism in dense forests where low NO<sub>x</sub> mixing ratios result in longer peroxy radical lifetimes that are on the order of the transport time through the canopy.

Technical comments.

1. Please check the subscripts for HO<sub>2</sub>, RO<sub>2</sub>, XO<sub>2</sub> are other chemicals throughout the paper.

Thank you for noticing these mistakes. We have corrected several subscript formatting errors in the revised manuscript.

2. The name of RACM2, RACM2-LIM1, MCM3.2, MCM3.3.1 should be used properly. For example, Line 565, it should be RACM2-LIM1 instead of RACM-LIM1.

As suggested, we have corrected the references to each model mechanism to be more consistent. In the revised manuscript RACM2 models are referred to as RACM2, RACM2-LIM1, or RACM-ACC, and MCM models are referred to as MCM v3.2, MCM v3.3.1, or MCM-ACC.