

Some comments/Requests from the Editor:

Referee 1.

In response to the referee you say:

"The observed-to-modeled HO₂ ratio shows little evidence of a NO-dependence. The reanalysis of Olson et al. (2006) explains the NO-dependence of the ratio discussed in Faloon et al., (1999), and the NO-dependence observed often in ground-based studies is not evident until NO exceeds 3-5 ppbv. Because the highest NO mixing ratio encountered in DC3 was about 3 ppbv, we would not expect to see this effect in the DC3 results."

Editor: Could you please give some very brief details in the revised MS about how the "re-analysis" by Olson et al. 2006 "explains" the NO_x dependence of the measured/modelled HO₂ ratio. Could you also please cite the other studies where substantial deviations observed during ground-based studies typically are not evident until NO exceeds 3 ppbv.

For your response to another of the comments:

"We agree that this is an interesting study to do, but it not within the scope of this paper. However, we did a quick check on this idea by calculating the observed-to-modeled HO₂ ratio, filtering it for altitude bins (i.e., 5-8 km, 8-10 km, >10 km), and solar zenith angle bins (i.e., 50o-60o, 60o-70o, 70o-80o), and then plotting it as a function of J(O(1D)). In all cases, the ratio was constant as a function of J(O(1D)). This observation indicates that using the measured photolysis frequencies accurately captures the HO_x production by photolysis as seen in Figure 4e."

Editor: Could you incorporate aspects of this response into the revised MS as it is an interesting finding you have stated.

Referee 1 also says:

page 8, line 6: The table showing the measurements is missing. It should be presented in the main paper. Accuracies and time resolution of the measurements should be given.

Editor: I agree with the referee that the Table belongs in the main paper. The table includes for example accuracy and time resolution of the main measurements which are modelled (OH, HO₂) and the species included in the model (e.g. VOCs) – these are central pieces of information. Also in Table 1 it is not clear what the references mean, as some of these are quite old, e.g. 1991. Could you please indicate in the caption what the reference actually refers to? For example, is it a description of the instrument used on the aircraft for the measurements? (this is not clear owing to the date of the reference being quite old in in some cases)

Referee 1 makes a point about there being additional VOCs in the convective outflows following rapid uplift...

.... is especially relevant in the outflow of convective clouds which can transport relatively short-lived species to higher altitudes in short time.

Referee 2 makes a similar comment about convective outflows where this may impact on the interference seen in the ATHOS instrument:

“On page 6, the OH and HO₂ interferences suffered by ATHOS are discussed and the authors state that interferences are only significant above forests and cities and are negligible outside of the planetary boundary layer. The authors should comment on whether this statement remains valid when sampling in and around convective clouds which can rapidly transport air from the surface. The correction applied to the HO₂ observations to account for RO₂ interferences is rudimentary and more details should be provided on the type of RO₂ species present. The correction will change with changing RO₂ species present and this relies on model predictions for RO₂ which is far from ideal”

Editor: Although in your response you discuss the method you use for correcting for RO₂ interferences, the MS still states that interferences are only significant just above forests and cities and is negligible above the PBL. Can you please modify this statement to say that interferences may also be important in the convective outflow at higher altitudes where VOC concentrations are higher owing to rapid uplift.

Referee 2 says:

Pg 14, line 1: What was recalibrated in the instrument? The phototube? The authors should comment on why they think it is appropriate to apply a calibration performed so long after the campaign.

Your response:

Looking back through the notes for DC3, we noticed that the procedure had not been followed completely and that the transmission of the window between the calibration lamp and the calibration flow tube, which is 0.86, had not been included in the calibration. We re-measured the window transmission and confirmed that it was still 0.86 and then applied this number to the OH and HO₂ calibration.

Editor: Please give some brief detail on the reason for recalibration in the revised MS as readers will be wondering the same as the reviewer.

Editor:

Other comments:

In the revised MS, you have the text:

We have rewritten Section 4.2, 2nd paragraph as

“In DC3, the DC-8 spent hours flying in anvils of the cumulus clouds, which consisted of ice particles. DC3 provides evidence that the HO₂ uptake on ice is small. These results are consistent with HO₂

results over the western Pacific Ocean (Olson et al., 2004) but not with those over the northern Atlantic (Jeaglé et al., 2000). In Mauldin et al. (1998), a large difference between the observed and modeled OH was found in clouds, but this difference may have been due to the lack of photolysis frequency measurements, which are crucial to test photochemistry in a cloudy environment. In DC3, the DC-8 spent essentially no time in liquid clouds, for which there is evidence of measurable HO₂ uptake (Olson et al., 2006; Whalley et al., 2015). Thus these DC3 results provide constraints of HO₂ uptake on aerosol and ice particles, but not on liquid water particles.”

Editor: The following paper ought to be cited and briefly discussed here, as it shows that the HO₂ concentration is reduced when the aircraft sampled clouds, and the concentration was reduced more than expected owing to the reduction in J(O¹D), providing some evidence for heterogeneous uptake which was not included in the calculation of [HO₂]. The liquid water measurements shows these are primarily liquid clouds.

Commane, R., Floquet, C. F. A., Ingham, T., Stone, D., Evans, M. J., and Heard, D. E.: Observations of OH and HO₂ radicals over West Africa, *Atmos. Chem. Phys.*, 10, 8783-8801, <https://doi.org/10.5194/acp-10-8783-2010>, 2010.

Also:

““Modeled RO₂ is primarily CH₃O₂ and CH₃CHO₂ above 5 km.”

Change to CH₃CH₂O₂