

Several comments by referee #2 have not or only very (too) briefly been addressed in the revised manuscript. They are cited below (re-ordered by topic). In addition, I have several additional comments that need to be addressed before the manuscript can be accepted for publication.

Previous comments by Referee #2:

- 3) [...] In their response, the authors also say that scavenging of all soluble species is turned off in their sensitivity study and that this is done globally. This is not clear from the text, which remained unchanged as “To investigate this assumption we performed a sensitivity study with EMAC excluding scavenging.” The authors should update the text to provide more detail to the reader. Finally the authors do not address whether the EMAC very large underestimate in ROOH can be explained by excessive wet removal during transport. It seems that their sensitivity study should be able to answer this question in a straightforward way.

Authors' response:

Please note that large enhancements of MHP and larger hydroperoxides were not found in the sensitivity study with switched-off scavenging, indicating that the strong underestimation by the model of those species is not due to an overestimation of wet removal in convective clouds.

We also change the text on line 455:

To investigate this assumption we performed a sensitivity study with the wet scavenging for all soluble species being switched-off globally.

- Lines 477-480 “Convective injection of H₂O₂ (and MHP) into the upper troposphere over India most likely forms a pool of hydroperoxides in the upper troposphere that subsequently influences the western AMA, giving rise to a significant longitudinal gradient of H₂O₂ and MHP mixing ratios, with increasing values towards the center of the AMA. It is likely that at least a large part of UHP is due to additional MHP from an up-wind source.” I suggest replacing MHP with ROOH as ROOH is the only quantity that is measured. Also, the conclusions should reflect the fact that EMAC significantly underestimates ROOH, but that the underestimate in H₂O₂ is not quite as large. The conclusion lacks any mention of the sensitivity study results.

Authors' response:

We changed the text to:

Convective injection of H₂O₂ (and potentially MHP) into the upper troposphere over India most likely forms a pool of hydroperoxides in the upper troposphere that subsequently influences the western AMA, giving rise to a significant longitudinal gradient of H₂O₂ and ROOH mixing ratios, with increasing values towards the center of the AMA. It is likely that at least a large part of UHP_{PSS} is due to additional MHP from an up-wind source. A sensitivity study using EMAC with no scavenging tends to reproduce the observed longitudinal gradients in H₂O₂, although it does not increase the level of ROOH. The reasons for this different behavior are unclear.

Editor comment 1: Both of these referee comments address the scavenging (or the lack of it) of hydroperoxides. Your response is very brief. Given that EMAC only considers three hydroperoxides, could it be that the properties relevant for scavenging (solubility) are not characteristic for the

majority of the hydroperoxides? Or if it is not scavenging, what other sources processes could occur?

Previous comment by Referee #2:

Abstract line 30-33: “Deviations to EMAC simulations are most likely due to uncertainties in the scavenging efficiencies for individual hydroperoxides in deep convective transport to the upper troposphere, corroborated by a sensitivity study.” And text lines 410-411 “Differences between observation and EMAC simulation could potentially arise due to uncertainties in the scavenging efficiency for H₂O₂, as the chemistry does not seem to be a dominant cause of uncertainty.” From the response to the reviewers it seems that another explanation is the too weak convective transport in the model due to its relatively coarse resolution. The authors quoted an underestimate in CO. This should be noted in the abstract and text.

Authors’ response:

We do not state that convection is too weak. Instead as mentioned above the low resolution of the model leads to an underestimation of species with local source in the inflow region of convection (e.g. CH₄).

Editor comment 2: Please add this information to the manuscript

Previous comment by Referee #2:

Lines 132-140. Given that the ROOH measurement is non-specific, I find it confusing that the LOD and uncertainties are given for MHP. Shouldn’t they be given for ROOH, which is actually what is measured? Please clarify the text.

Due to the fact, that the exact composition of ROOH_{obs} is unknown, and the solubility of different ROOH species can be quite variable, a detection limit for ROOH_{obs} cannot be given. Instead, we calculate an upper limit of the detection limit, assuming that all ROOH_{obs} consists of MHP, the species with the smallest solubility.

Authors’ response:

We changed the text to (page 5, line 136): and 9 – 52 pptv for ROOH (median 23 pptv) assuming that ROOH_{obs} is composed of MHP only.

Editor comment 3: Add this information on the solubility to the manuscript.

Previous comment by Referee #2:

Lines 358-360. I already pointed out in my first review that the statement “The deviations from unity in the slope are within the combined uncertainties of measured and steady-state estimations of H₂O₂ (51%)” is quite wrong. The authors concurred by saying in their reply that 82% of the points are OUTSIDE the range of uncertainties, but haven’t updated the text to correct their misleading statement.

Authors’ response:

See above. Changed to:

The regression coefficient R² is 0.26 with most of the H₂O_{2PSS} mixing ratios (75%) varying between 0 and 65 pptv with a median value of 15 pptv, while the H₂O_{2obs} extend over a larger range mainly between 10–210 pptv with a median of 150 pptv and thus 10 times higher than for steady-state, which can also be seen in the histograms in Figure 13.

Editor comment 4: Please add the information to the text that > 80% of all points are outside the range of uncertainty.

Previous comment by Referee #2:

Table 2 and Section 4.3.2. [...] I suggest that the authors add a column for ROOH in Table 2 and discuss it in the text. The comparison between observed and EMAC ROOH is shown in Figure 17 and shows a very large underestimate of EMAC (factor of 5-10?). A similar suggesting was made by the other reviewer and not addressed by the authors.

Authors' response:

We added a sentence to Section 4.3.2: In general EMAC tends to strongly underestimate measured total hydroperoxide in all air masses by a factor of 5 -10.

Editor comment 5: Given that both referees suggested to add a column to Table 2 reporting the ROOH values from EMAC and observations, your response is not sufficient. It seems that based on your discussion, you should be able to add these values.

Previous comment by Referee #2:

Lines 455-457 and Figure 19. It would be useful to include a panel comparison modeled and observed ROOH in Figure 19. It seems that removing scavenging in EMAC results in between agreement for H₂O₂, this begs the question as to whether it also leads to improved agreement with ROOH. Please address this.

Authors' response:

See our comment above.

Editor comment 6: Your response to the referee comment is too brief. I assume that the results for ROOH(EMAC) are not exactly identical with and without scavenging. I second the referee's suggestion to contrast the small predicted effect of ROOH scavenging to the much larger one by H₂O₂ by adding a panel for ROOH in Figure 19. Could you estimate which scavenging coefficient would be needed for ROOH to reach a better model/observation agreement?

Previous comment by Referee #2:

Lines 477-480 "Convective injection of H₂O₂ (and MHP) into the upper troposphere over India most likely forms a pool of hydroperoxides in the upper troposphere that subsequently influences the western AMA, giving rise to a significant longitudinal gradient of H₂O₂ and MHP mixing ratios, with increasing values towards the center of the AMA. It is likely that at least a large part of UHP is due to additional MHP from an up-wind source." I suggest replacing MHP with ROOH as ROOH is the only quantity that is measured. Also, the conclusions should reflect the fact that EMAC significantly underestimates ROOH, but that the underestimate in H₂O₂ is not quite as large. The conclusion lacks any mention of the sensitivity study results.

Authors' response:

We changed the text to:

Convective injection of H₂O₂ (and potentially MHP) into the upper troposphere over India most likely forms a pool of hydroperoxides in the upper troposphere that subsequently influences the western AMA, giving rise to a significant longitudinal gradient of H₂O₂ and ROOH mixing ratios, with increasing values towards the center of the AMA. It is likely that at least a large part of UHP_{pss} is due to additional MHP from an up-wind source. A sensitivity study using EMAC with no scavenging tends

to reproduce the observed longitudinal gradients in H₂O₂, although it does not increase the level of ROOH. The reasons for this different behavior are unclear.

Editor comment 7: Your response is unclear and confusing. The referee had suggested to replace MHP in this context by ROOH. Your response implies that the large underestimate of ROOH is exclusively due to MHP. Why do you exclude the possibility that other ROOH are underestimated?

Additional editor comments

8) I appreciate that you added indices (PSS, EMAC, obs) to the species names. However, they do not seem correct at all places and sometimes they are even confusing. Some examples are listed below. Please check the complete manuscript for their use.

- l. 23: 'We observed enhanced mixing ratios of [...] MHP(PSS), UHP(PSS)' – is consistent with the previous sentence ('Observations are compared to photostationary calculations...')

- l. 212 and Equation 5: Shouldn't it be H₂O₂(PSS) and not H₂O₂(obs)?

9) l. 30 and l. 31: 'Deviations' is very qualitative. Please quantify the value and state whether there is a consistent trend (over- or underestimate) between model results and observations.

10) l. 134 and l. 138: Clarify 'to be mainly MHP' or 'MHP only'?

11) l. 234/5: Should this read

'The combination of Eq. 5 and 12 **results** in Eq. 13 which was used to calculate the MHP^{PSS} concentrations **based on** the observed mixing ratios during OMO.'?

12) l. 244: 'To estimate the composition of the organic hydroperoxides...' should be replaced by 'To estimate the contribution of MHP to the total organic hydroperoxides...' (or similar) because your estimate does not yield any further information on the composition of ROOH.

13) Equation 15: 1) The index obs should be before exponent, i.e. [HO₂]^{obs 2}

2) Specify the reaction denoted by k_{OH} (i.e. similar to Eq.-13 etc k_{OH+}...)

14) l. 323-325: I do not understand this sentence. Why does the fact that you see a correlation between UHP(PSS) and ROOH(obs) imply that this is 'mainly due to UHP(PSS)'? How does the correlation of MHP(PSS) look like?

15) l. 371/2: The text here is confusing. Should it read

'For the SH the model simulated H₂O₂^{EMAC} mixing ratios (**272 pptv**) are four times higher than in the NH background (**66 pptv**),'

16) l. 373: Unless I misunderstand this sentence or the table, shouldn't it be '(**100 pptv** to 211 pptv) (Table 2).'??

17) l. 374: 'measured' should be removed here

18) l. 378 and 379: For both EMAC and observations, the NH background and AMA values show about a 10% difference. I suggest rewording the sentence in this regard rather than saying 'almost identical' versus 'small difference', respectively.

19) l. 379-381: Please add the predicted values to this sentence so it is easier to follow the data in the table.

20) l. 388-390: These sentences do not read well. You mix relative differences ('factor of four') with absolute differences ('200 ppt'). Either use only the most meaningful difference or report both relative and absolute differences.

21) l. 400: should 'blue' be 'purple'?

22) l. 409: Where can one see the 'rather large deviation of 150 pptv'? The average difference between the PSS values and observations in Figure 16 look less than that.

23) l. 414: 'Assuming that MHP is also enhanced in the outflow of deep convection...' – Is this assumption based on literature (if so, add reference(s)) or on observations in the current study?

24) l. 426: Table 2 shows a value of 100 pptv, not 115 pptv.

25) l. 456: Add 'EMAC' to H₂O₂.

26) l. 478 and l. 483: This is contradictory: You define UHP as all organic hydroperoxides except MHP. The sentence in l. 478 seems consistent with this definition 'a large contribution of an unidentified organic hydroperoxide (UHPPSS)' whereas the later text 'a large part of UHPPSS is due to additional MHP from an up-wind source' is inconsistent. Please make sure that you use the definitions of ROOH, UHP, MHP consistently.