

**Answer to: Interactive comment on “Impact of the South Asian monsoon outflow on atmospheric hydroperoxides in the upper troposphere” by Bettina Hottmann et al.
Anonymous Referee #2**

**Please note the colour code
(black: RC, red: AC, blue: changes in manuscript)**

The paper describes aircraft measurements of hydroperoxide compounds and supporting observations taken during the Oxidation Mechanism Observation (OMO) mission. These measurements are analyzed alongside photochemical steady state calculations, trajectory modeling, and global model simulations to understand the source regions of the air sampled in the upper troposphere. The major findings are that hydroperoxide mixing ratios are enhanced in the Asian Monsoon Anticyclone (AMA) compared to the background Northern Hemisphere (NH) mixing ratios, but highest hydrogen peroxide (H₂O₂) and methyl hydrogen peroxide (CH₃OOH) mixing ratios were found in the background Southern Hemisphere (SH). The authors attribute the high mixing ratios in the AMA to upwind convection, using a sensitivity simulation with the global model EMAC to support this claim.

It is interesting to discover the higher-than-expected hydroperoxide mixing ratios in the Asian Monsoon Anticyclone, and then learn what caused these high mixing ratios. I find that the study provided hints as to the cause of the high mixing ratios, but did not provide complete attribution. The analysis would benefit from conducting box model chemistry calculations to fully understand the processes affecting hydroperoxide mixing ratios. In addition, there are a number of items that need further work as detailed below.

Specific Science Comments

1. *The abstract should be more quantitative in their claim about enhancements in the AMA versus NH background. Line 22 states that observations show enhanced mixing ratios for H₂O₂, MHP (CH₃OOH), and UHP (unidentified hydroperoxides) in the AMA relative to the NH background. However, Figure 4 shows perhaps a small enhancement (10-20%) of H₂O₂, which is within the uncertainty (25%) of the measurements and no enhancement of MHP. There is only substantial enhancement (78%) of UHP. More convincing are the histograms in Figure 9 that show median values of H₂O₂ in the AMA to be 55% higher than those in the NH background, but MHP median values are quite similar between AMA and NH background. Again, the UHP median value is clearly enhanced in the AMA.*

Abstract changed to:

Line 22: We observed enhanced mixing ratios of H₂O₂ (45%), MHP (9%) and UHP (136%) in the AMA relative to the northern hemispheric background. Highest concentrations for H₂O₂ and MHP of 211 ppbv and 152 ppbv, respectively were found in the tropics outside the AMA, while for UHP, with 208 pptv highest concentrations were found within the AMA. In general, the observed concentrations are higher than steady-state calculations and EMAC simulations. Especially in the AMA, EMAC underestimates the H₂O₂ (medians: 71 pptv vs. 164 pptv) and ROOH (medians: 25 pptv vs. 278 pptv) mixing ratios.

2. *Introduction. Consider adding more information on the flow patterns of the Asian monsoon. A good resource for this information is Lawrence and Lelieveld ACP (2010).*

Introduction changed to:

Line 66: So far we know that the updrafts of the summer monsoon deep convection can effectively transport insoluble pollutants from the surface to the upper troposphere and there these polluted air masses can be transported over a long distance (Lawrence and Lelieveld, 2010). Thus the Asian summer monsoon has a strong influence on the upper troposphere (UT)

and the lower stratosphere (Randel et al., 2010; Gettelman et al., 2004) and it is important to study its physical and chemical properties in greater detail.

3. Section 3.1 describes the hydroperoxide measurements. The method measures total peroxides which is the sum of H_2O_2 , CH_3OOH , and other organic peroxides. The method uses a catalase to destroy H_2O_2 allowing the ability to infer H_2O_2 (i.e. total peroxides minus $ROOH$ gives H_2O_2 , where $ROOH$ represents the sum of organic peroxides). $ROOH$ is assumed to be mainly CH_3OOH . To determine CH_3OOH , a photostationary-state chemistry approximation is used based on measurements of OH , HO_2 , CO , CH_4 , NO , and photolysis rates. It is unclear why it is valid to estimate CH_3OOH from photostationary steady-state when the chemical lifetimes of CH_3OOH and H_2O_2 are a few days (as stated on line 381, page 13). It would be better to describe the measurement technique as measuring total peroxides, inferring H_2O_2 , and estimating MHP_{PSS} with the remaining $ROOH$ being called unidentified hydroperoxides (UHP). Then, when the authors suggest that most of the UHP is CH_3OOH , then they can use MHP without further notation.

As mentioned in the experimental section, the $ROOH$ measurement is unspecific and due to the different solubilities of hydroperoxides qualitative. In order to estimate the amount of MHP, which according to previous measurements is expected to be the dominant (if not the only) $ROOH$ component, we calculated the amount of MHP from a photo-stationary state calculation (as well as H_2O_2). The difference between $ROOH$ and PSS MHP is unexplained or unaccounted for, thus we named it UHP. As discussed in the paper UHP can be due to an unidentified hydroperoxide (e.g. PAA), additional MHP due to advection or a combination of both. Since no specific $ROOH$ measurements were made nor does the 3D-model indicate substantial amount of hydroperoxides other than MHP, we cannot finally decide on the nature (or composition) of the UHP.

4. Lines 295-305. The correlation between UHP and acetone is very strong, but there's no explanation on what the cause and effect may be. I suggest further analysis on this result. Even stating acetone photolysis produces methyl peroxy radical which can react with HO_2 to form CH_3OOH is good, but more interesting would be box model calculations.

We calculated the production of MHP from acetone and acetaldehyde. For the AMA appr. 40 pptv MHP per day can be formed through this reaction.

Section changed to.

L347: Enhanced mixing ratios of hydroperoxides are typically associated with enhanced acetone mixing ratios, especially for PSS-UHP. Our calculation of the production of MHP from the photolysis of acetone and the reaction of acetaldehyde (from EMAC) with OH shows that per day appr. 40 pptv MHP can be formed within the AMA. The lifetime of MHP was calculated to be around 1.5 days. Thus not all of the PSS-UHP in the AMA (median 210 pptv) can be accounted for MHP that was chemically produced from VOCs in the AMA.

5. Section 4.3. I think assuming photostationary steady-state for H_2O_2 and CH_3OOH interferes with the comparisons described in Section 4.3.

a. While, it is useful to point out the large discrepancy between observed H_2O_2 and H_2O_2 estimated by photostationary steady-state, it needs an explanation of why there is such a discrepancy (Lines 333-340).

The discrepancy is mainly due to transport phenomena especially deep convection over India.

We added to Section 4.3.1.:

L363: The discrepancy between in situ and PSS-H₂O₂ shows that the local PSS does not account all main contributions of H₂O₂ even though all chemical reactions are included. Thus transport phenomena like deep convection seem to play a key role (see 4.3.3).

b. In Section 4.3.2, I do not find it useful to compare the ranges of the observations and EMAC results. For example, EMAC clearly underpredicts CH₃OOH mixing ratios most of the time, but the range of EMAC results overlaps with the observations. It may be better to discuss medians or simply describe that most EMAC CH₃OOH is < 50 pptv for NH background and AMA air, while most observations range from level of detection to 120 pptv.

Section changed to:

Line 375: EMAC mainly simulates MHP mixing ratios lower than 50 pptv for background and AMA, while PSS-MHP ranges from LOD–140 pptv.

c. The photostationary steady-state assumption interferes with the comparisons of the MHP and UHP observations with EMAC. If H₂O₂ and CH₃OOH are not in photostationary steady-state, then it would make more sense to compare total organic peroxides between observations and EMAC results. The differences between model results and observations need an explanation of why they are different.

In sum we compare total observed peroxides with EMAC-the mixing ratios are just splitted into PSS-MHP from local CH₄ oxidation and other organic hydroperoxides. If this is MHP from other chemical sources (e.g. from acetone photolysis or reaction of acetaldehyde and OH), transported MHP or if it is another hydroperoxide like PAA cannot be verified.

Reasons for the differences between the model and observations are given in Lines 432–440:

“Although there is rather good agreement between EMAC simulations and observations for all the species that affect the local photochemical budget of H₂O₂, EMAC significantly exceeds PSS calculation for H₂O₂. This is an indication that an additional H₂O₂ source is accounted for in the global model and that the local photo-stationary-state assumption is not fulfilled. The additional source is attributed to transport associated with deep convection over India, yielding in an upwind source of H₂O₂ that is significant throughout the western part of the AMA. In the AMA, clouds are absent, so that gas phase photochemical processes may determine the lifetime of H₂O₂. Based on observed OH levels and photolysis frequencies during OMO the H₂O₂ lifetime in the upper troposphere is of the order of several days, sufficiently long for the excess H₂O₂ to reach the western parts of the AMA, producing the observed longitudinal H₂O₂ gradient observed in both observations and EMAC simulations (Figure 16).”

Sections changed to:

Line 454: Differences between H₂O₂ observations and EMAC simulations are most likely due to an overestimation of scavenging in the model as also pointed out by Klippel et al., (2011). To investigate this assumption we performed a sensitivity study with EMAC excluding scavenging. The result is shown in Figure 19. The H₂O₂ mixing ratios significantly increase with longitude by a factor of 3–4 and thus to the level of observed H₂O₂.

Line 458: There is a rather large uncertainty regarding the scavenging efficiency of MHP in deep convection (Barth et al., 2016). For the Trace A campaign Mari et al. (2000) found observed (modelled) enhancement ratios of post-convective to pre-convective mixing ratios of 11 (9.5) for MHP and 1.9 (1.2) for H₂O₂. Such efficient transport in the Indian Summer Monsoon would yield a strong source of upper tropospheric MHP explaining the large enhancement of ROOH in the AMA described here. It seems that a large part of the PSS-UHP is actually MHP advected

throughout the AMA after deep convective transport over India. In the EMAC simulations the transport of MHP is less efficient and thus EMAC-MHP is lower than PSS-MHP and PSS-UHP.
d. Shouldn't a conclusion be that assuming photostationary steady-state can be inappropriate?

Yes we would like to stress this in the conclusions. Local photochemistry does not explain the high mixing ratios that we found. Thus transport must play a substantial role especially since other chemical formation of MHP is insufficient. We assume that deep convection is the reason for the higher concentrations in the upper troposphere.

6. Lines 379-385. This discussion, to me, is not well supported and contains a lot of suppositions. This manuscript is relying on one paper (Bozem et al., 2017) to say H₂O₂ is enhanced in convective outflow regions compared to the background upper troposphere. Yet Bozem et al. (2017) found an unusually high H₂O₂ mixing ratio in convective outflow (1.25 ppbv) which is not found in other studies. Snow et al. (2007) and Barth et al. (2016) both show that H₂O₂ is depleted in convective outflow compared to background upper troposphere. We do not know what H₂O₂ mixing ratios are like in convective outflow near the convection that transports constituents into the AMA, but we can make use of the array of literature from past studies to guide us for what to expect and what further analysis is needed. This leads to my next comment.

We now show the inconsistent results from both sides.

Section changed to:

L403: Previous studies present results that are difficult to reconcile. Snow et al. (2007) and Barth et al. (2016) for example both show that H₂O₂ is depleted in convective outflow compared to background upper troposphere. In contrast, other studies found that deep convection can be a source of H₂O₂ in the upper troposphere (e.g. Jaeglé et al., 1997; Prather and Jacob, 1997; Mari et al., 2003; Bozem et al., 2017).

7. There will always be some variability in peroxides scavenging efficiencies and uncertainty in these scavenging efficiencies due to the complex processes associated with convection and chemistry. However, there were also 100s km (multiple days) of transit between the convection in northern India and the measurements over the Arabian Sea. What chemistry occurred during this transit? If it is true that the observations are reflecting chemistry in convective outflow then one would also expect other volatile organic compounds, CO, and CH₄ to have been lofted in the convection. I would recommend conducting a number of box model calculations (e.g. Pickering et al., JGR, 1992; Apel et al., ACP, 2012, etc) to learn what chemical transformations are affecting the peroxides. Further, this box model can more definitively provide information on what unidentified hydroperoxides are.

We do not see any chance to get more information on the organic hydroperoxides since PAN and acetaldehyde were not measured during the campaign. Calculations based on EMAC acetaldehyde and observed acetone are added (see above).

8. Lines 405-409. I do not think the typhoon Mireille case is suitable to compare to this paper's results. Typhoon Mireille occurred over the Western Pacific ingesting air from Oceania (Preston et al., JGR, 2019) in the early 1990s and not the South Asia region of the mid 2010s.

We do not want to exactly compare the Typhoon study with our study. We just want to mention that such phenomena are already known from previous studies.

9. I am surprised there is no mention of past literature on peroxides, peroxy radicals and convection and how these current results compare to those previous findings. Some papers to

discuss are Jaeglé et al., GRL, 1997, Prather and Jacob, GRL, 1997, Crawford et al., JGR, 1999).

Some of these studies are now mentioned (see 6.).

Organization, Clarity, Technical Comments

1. Line 140 should include a list of all species measured. It should state OH and HO₂ instead of HO_x, and include NO.

Section changed to:

L144: For this study CO, CH₄, OH, HO₂, O₃, Acetone, NO, NO_y, JH₂O₂ and JMHP data measured by other instruments have been used for data interpretation, steady-state calculations and interference corrections (see section 3.1). A complete list of all measured compounds can be found in Lelieveld et al., 2018.

2. Lines 192-195. Shouldn't pressure and temperature measurements also be listed? These state parameters must be needed for calculating rate constants and air density.

Section changed to:

L196: Latitude, longitude and altitude data as well as temperature and pressure were collected with the BAHAMAS (BASIC HALO Measurement And Sensor system) instrument. More detailed information about the installation of scientific instruments and mission flights can be found on <http://www.halo.dlr.de/science/missions/omo/omo.html>.

3. Line 209. It would be good to explain why the CH₃O₂ to HO₂ ratio is needed. It would also be good to define P(HO₂) and P(CH₃O₂).

P(CH₃O₂) and P(HO₂) are defined in eq. 12. As it is analogous to P(H₂O₂) and P(MHP) we thought this is enough explanation. If not-we can change it.

4. Line 217. Why is a scale height needed? Why not use pressure measurements from the aircraft along with temperature to get air density that can then be translated to N₂?

Section changed to:

L220: For the calculations of the rate coefficients the mean temperature of 259.18 K, the mean altitude of 10,992.8 m and the mean pressure of 22,932.9 Pa were used.

5. Line 221. Isn't there a H₂O dependence to the self-reaction of HO₂? See, for example, JPL (2015). <https://jpldataeval.jpl.nasa.gov/>

Yes there is a [H₂O] dependence. But in this case it was neglected because of the low RH in the upper troposphere. An example: For 100 ppm H₂O a factor of $\{1+1.4 \times 10^{-21} \times [\text{H}_2\text{O}] \times \exp(2200/T)\}$ and thus 1.004 needs to be included to the calculation.

([http://iupac.pole-ether.fr/htdocs/datasheets/pdf/HOx14_2HO2_\(M\).pdf](http://iupac.pole-ether.fr/htdocs/datasheets/pdf/HOx14_2HO2_(M).pdf))

Section changed to:

L222: As the relative humidity is very low in the upper troposphere the water dependence in eq. 11 was neglected.

6. Line 229. Can you show or quantify the contribution of CH₄+OH and CO+OH to the total CH₃O₂ and HO₂ production, respectively? If the hypothesis is that the higher AMA mixing ratios

are due to convective transport, then HCHO, CH₃OOH, and other VOCs will be elevated compared to background mixing ratios and their chemistry may be more important than assumed here.

Section changed to:

Line 234: This is justified by the generally low mixing ratios of these species at high altitudes. Measurements of HCHO with the TRISTAR instrument yielded values below the detection limit of 30 pptv, and although acetaldehyde was not measured, we assume that its mixing ratio is within a factor of two of those of HCHO.

7. Lines 245-248. What grid spacing is used for EMAC? Perhaps a short summary of the configuration could be given in supplemental material. Further, how do the authors analyzed EMAC output to provide comparisons to aircraft observations? Are the model results interpolated in time and space to the aircraft location? Or are values from the nearest grid point and model output time (which, I assume, is every hour)?

The EMAC simulation was made for the flight track of the aircraft. EMAC data were compared to measurements and calculations for the same time and thus location in 2.8°x2.8° grids of the aircraft. Therefore the corresponding values out of the 1 minute means were used. EMAC offers one value every 12 minutes.

Line 253: For this study EMAC simulations were performed for the OMO flight tracks in 2.8°x2.8° grids with a time resolution of 12 minutes. Detailed specifications and results have been published previously (Lelieveld et al., 2018; Tomsche et al., 2019).

Line 276: To compare the simulations from EMAC with measured and PSS calculated data, the corresponding values (out of the 60-second-means) were used at the given times from EMAC.

8. Lines 249-255. What is the procedure when the back-trajectory encounters convection? Does FLEXPART have a means to represent convective transport? Or are the trajectories stop when convection is encountered?

Convective transport can be simulated in FLEXPART with the convection parameterization by Emanuel and Zivkovic-Rothman (1999). To represent moist convection realistically in models, the parametrization includes cloud microphysical processes, the physics of entrainment and mixing, as well as large scale control of ensemble convective activity. It builds on temperature and humidity fields to provide mass flux information (Stohl et al., 2005). The back trajectories in the present paper are calculated with the convective parametrization. Further the Lagrangian particle dispersion model FLEXPART produces so called centroid trajectories, which found on cluster analysis. These trajectories are comparable to traditional trajectories, but include convection via the centroid of all particles per time step.

Section changed to:

L256: Ten-day back-trajectories were calculated along the flight path using FLEXPART to identify the air mass origin (Tomsche et al., 2019). Convective transport can be simulated in FLEXPART with the convection parameterization by Emanuel and Zivkovic-Rothman (1999). To represent moist convection realistically in models, the parametrization includes cloud microphysical processes, the physics of entrainment and mixing, as well as large scale control of ensemble convective activity. It builds on temperature and humidity fields to provide mass flux information (Stohl et al., 2005). The back trajectories in the present paper are calculated with the convective parametrization. Further, the Lagrangian particle dispersion model FLEXPART produces so called centroid trajectories, which found on cluster analysis. These trajectories are

comparable to traditional trajectories, but include convection via the centroid of all particles per time step.

References:

Emanuel, K. A. and Zivkovic-Rothman, M. (1999). Development and Evaluation of a Convection Scheme for Use in Climate Models. *Journal of the Atmospheric Sciences*, 56(11):1766-1782.

Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G.: Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, *Atmos. Chem. Phys.*, 5, 2461–2474, <https://doi.org/10.5194/acp-5-2461-2005>, 2005.

9. Line 253-254. Since methane and its use for identifying AMA air via a threshold value is discussed on these lines, it would be useful to combine the first paragraph of section 4.1 with this information. Or move lines 252-255 to section 4.1.

Sentences shifted to 3.5

Line 267: Thus a threshold of $\text{CH}_4 \geq 1879.8$ ppbv was used to distinguish between air masses influenced by the monsoon ($\text{CH}_4 \geq 1879.8$ ppbv), the SH background ($\text{CH}_4 < 1820$ ppbv) and the NH background ($1820 \text{ ppbv} \leq \text{CH}_4 < 1879.8$ ppbv) (Tomsche et al., 2019).

10. Line 262. It is not clear which species concentrations are binned into 10 pptv segments. Please specify which species.

Section changed to:

L275: For the histograms the concentrations of all species shown were binned into samples with a width of 10 pptv, starting the plots with the lowest bin.

11. Line 265. "Case study: flight 17" is not very descriptive to the general audience. Consider using a heading that mentions the date and location of the flight.

Header changed to:

L278: 4.2 Case study: Flight 17 from Gan to Bahrain (10.08.2015)

12. Line 266. There should also be a short description of the flight, again mentioning date, but perhaps adding weather conditions (cloudy anywhere?) and location of the anticyclone, etc. You might want to add this description to section 2.

Yes there were some clouds but as the focus is on the upper troposphere and the convection took place before and somewhere else it was not mentioned.

Section changed to:

L279: In a case study analyzing flight 17 from 10th of August 2015, the method used to determine the origin of the measured air masses and a quantification and comparison of measured and simulated mixing ratios of H₂O₂, PSS-MHP and PSS-UHP is presented.

L286: Figure 3 shows the time series for measured H₂O₂ during the flight at the time steps given from the frequency of EMAC output (orange circles).

13. Line 272. → *Tomsche et al. (2019)* and on line 273, remove (*Tomsche et al. 2019*).

Section changed to:

L285: Tomsche et al. 2019 showed that the measured air in the AMA was affected by deep convection over India resulting in methane mixing ratios above the threshold.

14. Line 274. *State what EMAC time step is. I imagine this is the frequency of model output.*

Section changed to:

L286: Figure 3 shows the time series for measured H₂O₂ during the flight at the time steps given from the frequency of EMAC output (orange circles).

15. Line 279-280. *Clarify that it is EMAC model data.*

Section changed to:

L292: One hour later the EMAC model data decrease to 416 pptv and the in situ data increase to 214 pptv.

16. Line 282. → *last period of the flight at the higher altitude,*

Section changed to:

L293: During the following hour until around 8:00 UTC and thus at higher altitude, both mixing ratios increase with the modelled data showing a much stronger increase up to approximately 800 pptv while the in situ data increase only to 230 pptv.

17. Line 293. *I think it would be better to say “temporal pattern” rather than “evolution” as there is no following an air parcel in time in the figure.*

Section changed to:

L306: The *in situ* H₂O₂ mixing ratios show a similar temporal pattern and mixing ratio levels to those of PSS-UHP over the Arabian Sea and the Arabian Peninsula, with values in the range of 140–243 pptv.

18. Line 318. → *is found. Air masses*

Section changed to:

L334: For PSS-MHP (Figure 9, middle panel) the frequency distribution in the NH background shows a maximum at 30–40 pptv (green). For AMA influenced air a sharp maximum at 50–70 pptv (red) is found. Air masses from the SH exhibit a rather flat distribution with a maximum at values of 40–50 pptv and a median of 152 pptv (blue).

19. Line 333. → *photostationary steady-state*

Section changed to:

L355: A scatter plot of the results from the H₂O₂ photostationary steady-state calculation based on observed HO_x data in the UT (eq. 15) is shown in Figure 12.

20. Line 372. *It should be degrees E (not O), and please mention the red box in the figure.*

Section changed to:

L395: In Figure 16 observations, steady-state calculations and EMAC simulations for upper tropospheric (9–15 km) H₂O₂ are displayed as a function of longitude from west to east (20–30 °N, 36–60 °E, according to the red box in Figure 15).

21. Line 373. The observations are in orange (not yellow).

Section changed to:

L297: The observations (orange) show roughly a 100% increase of in situ H₂O₂ from west to east (90 pptv to 175 pptv), similar to simulation with EMAC (black), although absolute mixing ratio levels in EMAC-H₂O₂ are smaller (61 pptv to 121 pptv).

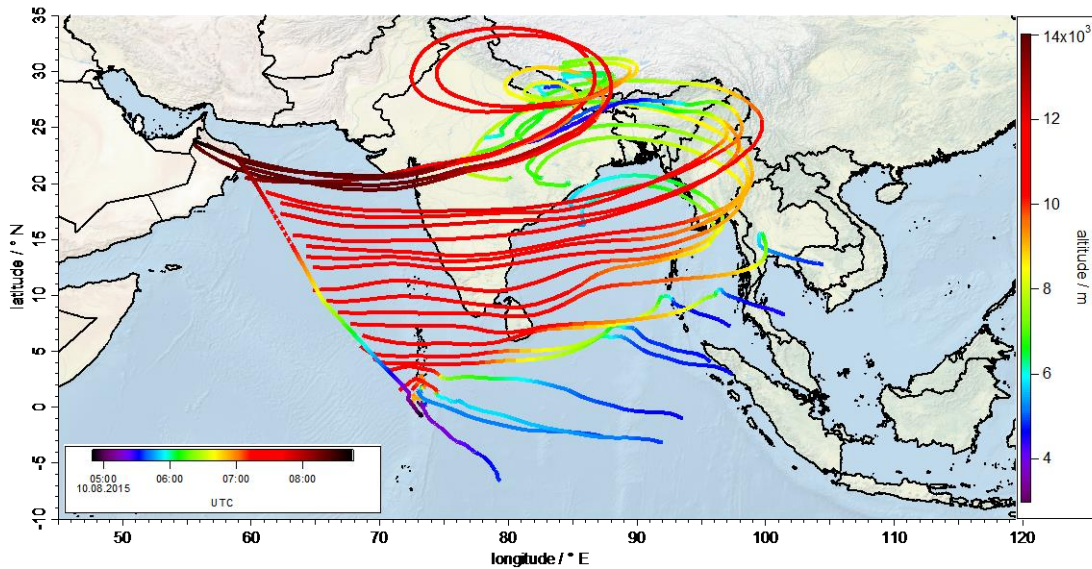
Figures and Table

1. Consider putting some figures into one with panels.

As the other referee asked for more figures we want to decide this at the end.

2. Figure 2. Please add number of days for back trajectories to caption. It would be helpful to mark each hour (text box of time) along the flight track so one can connect the map to the time series.

Figure changed to:



3. Figure 3 and others. It would help to say “EMAC modelled” for clarity. And “dataconstrained calculated” should be “photostationary steady state calculated”. At least be consistent from figure to figure and figure to text with nomenclature.

Changed to:

Figure 3: Time series of measured (orange circles), PSS calculated (blue crosses) and modelled (grey triangles) H₂O₂ mixing ratios for flight 17. The brown line shows the altitude, the colored bar on top indicates the origin of air masses according to the methane mixing ratio classification: for SH blue, NH green and monsoon red.

Figure 4: Time series of hydroperoxide mixing ratios during flight 17. The mixing ratios of in situ H₂O₂ (orange circles), PSS-MHP (purple triangles) and PSS-UHP (black crosses) are shown.

The brown line shows the altitude, the colored bar on top indicates the origin of air masses according to the methane mixing ratio classification: for SH blue, NH green and monsoon red.

Figure 5: Time series of PSS-UHP (black crosses) and in situ acetone (green circles) mixing ratios during flight 17. The brown line shows the altitude, the colored bar on top indicates the origin of air masses according to the methane mixing ratio classification: for SH blue, NH green and monsoon red.

Figure 6: Scatter plots of measured acetone and in situ H₂O₂ (left), in situ ROOH (middle) and PSS-UHP (right) during flight 17. The black lines represent the least orthogonal distance fits with regression coefficients of 0.99, 0.98 and 0.99.

Figure 7: Scatter plots of in situ H₂O₂ (left), in situ ROOH (middle) and PSS-UHP (right) and NO/NO_y ratio during flight 17.

Figure 8: All flight positions in the upper troposphere ($p < 300$ hPa) during OMO as a function of (a) in situ H₂O₂ on top, (b) PSS-MHP in the middle and (c) PSS-UHP at the bottom.

Figure 9: Histograms of in situ H₂O₂ (top), PSS-MHP (middle) and PSS-UHP (bottom) mixing ratios during the OMO campaign for NH background (green), SH (blue) and monsoon (red) air masses.

Figure 10: Scatter plots of in situ acetone and in situ H₂O₂ (left), in situ ROOH (middle) and PSS-UHP (right) in the UT (purple squares) and especially in the AMA (red circles). The black lines represent the least orthogonal distance fit with linear regression coefficients of 0.96 (H₂O₂), 0.97 (ROOH) and 0.96 (UHP).

Figure 11: Scatter plots of NO/NO_y and in situ H₂O₂ (left), in situ ROOH (middle) and PSS-UHP (right) in the UT (blue triangles) and especially in the AMA (red circles).

Figure 12: Scatter plot of in situ and PSS calculated H₂O₂ mixing ratios (red) with the 1:1 (black), 1:2 and 2:1 (both green) lines.

Figure 13: Histograms of in situ (top) and PSS (bottom) H₂O₂ mixing ratios (bars) and the associated medians (lines).

Figure 14: Histograms of in situ and EMAC H₂O₂ (top), PSS and EMAC-MHP (middle) and PSS and EMAC-UHP (bottom) mixing ratios during the OMO campaign for NH background (green), SH (blue) and AMA (red) air masses.

Figure 16: Longitudinal trends of in situ H₂O₂ mixing ratios (orange circles), EMAC-H₂O₂ (black triangles) and PSS H₂O₂ (purple crosses). The data are shown in the light colors while the darker ones represent the medians.

Figure 17: Longitudinal trends of in situ ROOH mixing ratios (green asterisks), EMAC-ROOH (blue plus signs) and PSS mixing ratios for MHP (pink triangles) and UHP (black crosses) as well as EMAC-MHP (yellow squares). The data are shown in the light colors while the darker ones represent the medians.

Figure 18: Scatter plot of in situ and EMAC HO₂ data (left) and the OH data (right) (both red) with the 1:1 (black), 1:2 and 2:1 (both green) lines. The blue line shows the calculated least orthogonal distance fit.

Figure 19: Longitudinal trends of in situ H₂O₂ mixing ratios (orange circles), EMAC (black triangles) and the sensitivity study without scavenging in EMAC (blue circles). The data are shown in the light colors while the darker ones represent the medians.

4. *Figure 4. Purple triangles look red in my version.*

Might be due to the printer used.

5. *Figures 5, 6 and 7 could be combined. The lilac colored squares should be darker.*

As figures 6 and 7 are now made of 3 scatter plots we do not combine these figures.

6. *Figure 8. Please note in the figure caption that you are showing data only at <300 hPa.*

Caption changed to:

Figure 8: All flight positions in the upper troposphere ($p < 300$ hPa) during OMO as a function of (a) in situ H₂O₂ on top, (b) PSS-MHP in the middle and (c) PSS-UHP at the bottom.

7. *Figures 10, 11, and 12 could be combined. The lilac colored squares should be darker.*

As figures 10 and 11 are now made of 3 scatter plots we do not combine these figures.

8. *Figure 12 figure caption should say "photostationary steady state calculated".*

Caption changed to:

Figure 12: Scatter plot of in situ and PSS calculated H₂O₂ mixing ratios (red) with the 1:1 (black), 1:2 and 2:1 (both green) lines.

9. *Figure 14 axes labels and legends need to be larger. Please explain what vertical lines are in the caption.*

Vertical lines are explained in the legend-these are medians. Font size now increased.

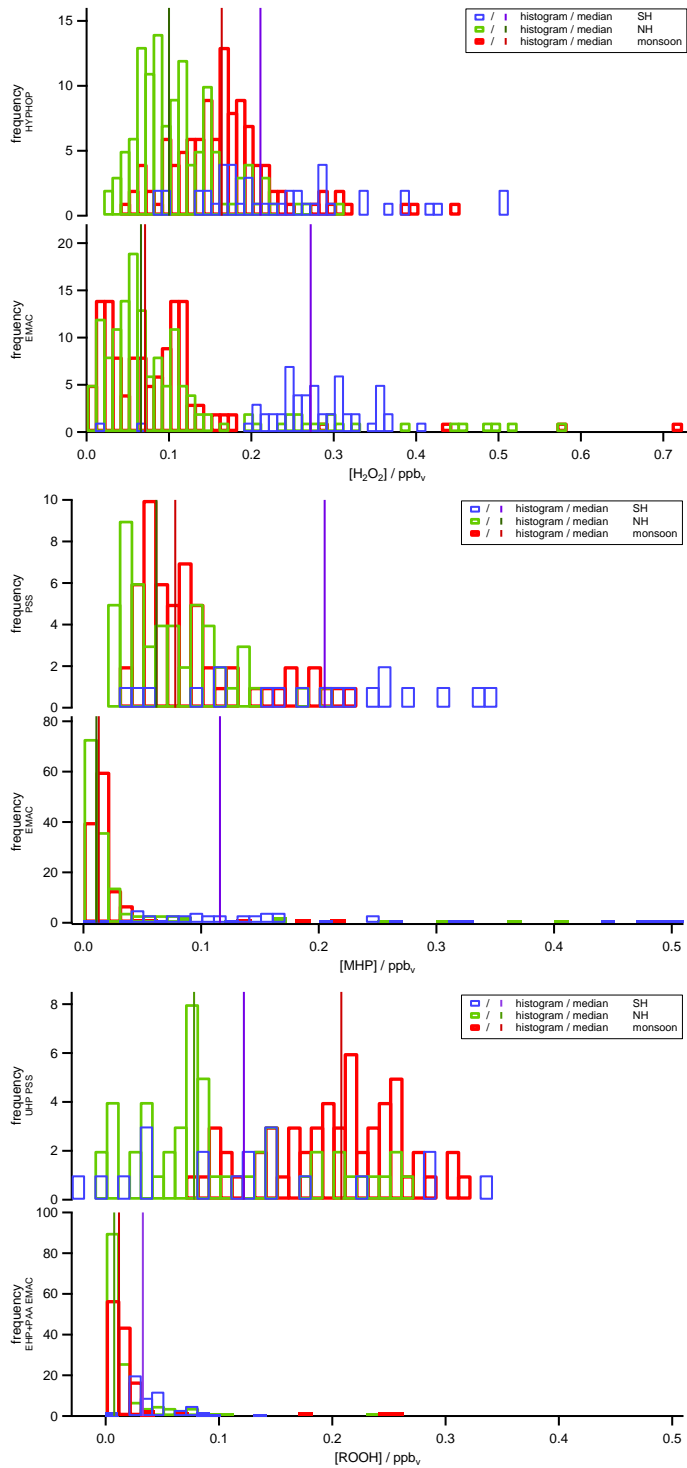


Figure 1: Histograms of observed and modelled H_2O_2 (top), MHP (middle) and UHP (bottom) mixing ratios during the OMO campaign for NH background (green), SH (blue) and AMA (red) air masses.

10. Figure 15. Please explain what the red box is in the caption.

Caption changed to:

Figure 15: Location of measurements used for the longitudinal gradient study (red box) out of all flight tracks (black).

11. Figure 16. The photostationary steady state calculated markers are purple not blue.

Caption changed to:

Figure 16: Longitudinal trends of in situ H₂O₂ mixing ratios (orange circles), EMAC-H₂O₂ (black triangles) and PSS H₂O₂ (purple crosses). The data are shown in the light colors while the darker ones represent the medians.

12. Figure 17. The MHP mixing ratios look more like pink than purple.

Caption changed to:

Figure 17: Longitudinal trends of in situ ROOH mixing ratios (green asterisks), EMAC-ROOH (blue plus signs) and PSS mixing ratios for MHP (pink triangles) and UHP (black crosses) as well as EMAC-MHP (yellow squares). The data are shown in the light colors while the darker ones represent the medians.

13. Table 1. Are these values from all flights? Please say so in the caption. "calc." is not a good heading. I suggest PSS estimate.

Table changed to:

Table 2: Comparison of H₂O₂, MHP and UHP mixing ratios in the upper troposphere from EMAC, measurements and PSS calculations.

region	median	[H ₂ O ₂]/ppt _v		[MHP]/ppt _v		[UHP]/ppt _v	
		EMAC	HYPHOP	EMAC	PSS	EMAC	PSS
NH background	median	66	100	11	64	8	78
	range	6–576	20–301	2–408	21–202	1–238	LOD–261
	avg±sdev	102±110	110±53	28±58	75±42	18±31	103±77
monsoon	median	71	164	13	70	12	208
	range	8–714	46–446	2–216	37–220	1–259	80–311
	avg±sdev	84±92	167±69	18±28	92±49	18±34	199±59
SH background	median	272	211	116	152	33	122
	range	15–409	85–510	2–502	40–346	1–132	LOD–334
	avg±sdev	272±68	238±105	155±125	191±95	42±24	125±102