Editor Decision: Reconsider after minor revisions (Editor review) (29 May 2015) by Dr Rupert Holzinger
Comments to the Author:
Please address the reviewer comment of the second round. In addition I find one answer (see below) to a comment raised by referee#1 a bit vague. Please provide information about the artifact to justify removal of these data.

Referre # comment:
"Fig 4, why do the model H2O2 concentrations become negative early on March 14?"

Response: "The negative H2O2 mixing ratios in Fig. 4 are an artefact and have been removed. In the revised manuscript figures 3, 4 and 6 that were affected by negative model results have been revised.".

Further comment:

The numerical solution of the ordinary differential equation (ODE) system describing the kinetic system occasionally produces small negative overshoots. These are corrected in the model system to keep the positive definiteness.
The resulting mass conservation violation is diagnosed for error control and the correction is performed after the model output is written, in order to be able to track where and when this happens.
Thus we had to remove the negative values from the primary model output.

Reviewer #1

Suggestions for revision or reasons for rejection (will be published if the paper is accepted for final publication)
The revised version addresses my main concerns. Below are a few minor suggestions for the authors to consider.

138-140: the uncertainty in the MHP sampling efficiency is ambiguous as written. The efficiency is given as a percent (60%), and its uncertainty is also given as a percent (30%). So it's unclear whether you mean the error is 30% of 60% (i.e., 0.60 +/- 0.18), or 30% in an absolute sense (i.e., 0.60 +/- 0.30). I guess it should be the latter. Please clarify the writing here.

As discussed in the manuscript the sampling efficiency for MHP is estimated to be 60%, cannot be higher than that for H2O2 (95 %) and is unlikely smaller than 30 %, thus yielding an uncertainty of +/- 30 %. This clearly indicates that the uncertainty is 60 +/- 30 %.

Abstract (21-22), line 344, and conclusion (430-432): I don't really see why you claim that the downward mixing of H2O2 and CH3OOH around midnight on 3/16 is well captured by the model. From Figures 4, 5, and 7 it seems that the model captures the fact that it occurs, but strongly underestimates the magnitude of the effect on H2O2 and CH3OOH mixing ratios. In my opinion more careful wording is needed here.

We changed wording to "qualitatively reproduced".

Figure quality (resolution, font size in Figs 4-6 too small) can be improved

We changed the font size of these figures.

320-321: “During the night dry deposition is the only loss mechanism” … unless there is rain or fog, of course

Line 318 – 319 already states that dry deposition is the dominant loss process under the present conditions, i.e. with no clouds or rain.

326-335: just a comment, but it appears that restricting the maximum deposition velocity to 0.6 does not improve the overall performance of the model: the underestimate is reduced during the first phase of the campaign in Fig 4, but now we have a very large overestimate in the middle phase.

The purpose of the sensitivity study is to identify potential sources of deviations between model simulations and observations and not to fine tune the model for a perfect agreement. Even the base run overestimates H2O2 during the middle phase, which could be due to an earlier start of downward mixing of free tropospheric air into the marine boundary layer in the model simulation.

Reviewer #2

Suggestions for revision or reasons for rejection (will be published if the paper is accepted for final publication)

Dear editor, author, co-authors;
Overall most of my specific comments have been well addressed but there are still two issues that I would like to raise.

1) Regarding the issue on the too large H2O2 dry deposition velocities; this appears to be mainly a unit/conversion issue and where the maximum H2O2 dry deposition velocities are 1.8 cm s-1. The sensitivity analysis that has been done has been based on reducing the maximum wind speed use for the calculation of dry deposition to 5 m s-1 (by the way, I assume that EMAC reproduced quite well the actually observed meteorological conditions during the measurement campaign). I guess that this does not only apply to the H2O2 dry deposition rate but to all dry deposition velocities which implies that also the chemical regime, affecting the H2O2 chemical production might have substantially changed. The maximum VdH2O2 is then 0.6 cm s-1 but it raises the question why the VdH2O2 would be that small when wind speeds in those regions can be large as 10-20 m s-1. Recognizing the fact that calculation of the turbulent transport term is pretty straightforward, it implies that there should be a substantial surface resistance against H2O2 uptake by the oceanic surface layer and wonder what this could be where the solubility cannot be the limiting factor or? Consequently, although the conducted sensitivity analysis suggests that a strongly reduced VdH2O2 results in a much better agreement between the model and the observations, there is no viable explanation why VdH2O2 should be that small. There is an extensive discussion about the dry deposition process in the discussion section but think that this "finding" on requiring a substantially smaller VdH2O2 to explain discrepancies between model and observations is conflicting with common-sense estimates of VdH2O2 for the conditions encountered in the region of interest. This really requires an effort to measure in-situ H2O2 dry deposition fluxes. This could be further addressed in the discussion.

We agree with the referee, that a maximum DdH2O2 of 0.6 cm/s at high wind speeds in excess of 10 m/s is smaller than expected, but keep in mind that this is a sensitivity study to identify potential causes of differences between model simulations and observations. We don't know any direct measurements of the deposition velocities of H2O2 as a function of wind speed in the marine boundary layer to compare our sensitivity study to. It is also true, that restriction of the deposition velocities to 0.6 cm/s affects other species, but we don't see how this will significantly affect the H2O2 production rate. Ozone as a precursor of HO2 is only marginally affected by the change in deposition velocity (due to the low wind speed dependency of its deposition velocity and a surface loss of HO2 itself is not considered in the model.

2) I raised the comment on the potential role of entrainment of FT air masses in explaining both the underestimation of O3 and H2O2. In the response it is then referred to the fact that MHP is also affected by entrainment and where a potential enhanced role of FT entrainment would make the comparison worse for MHP which is already largely overestimated. But further reading about some of the potential issues on the MHP measurements and also recognizing the fact that the deposition process for MHP is really poorly constrained by measurements, I am not convinced that MHP is the proper tracer to exclude a potential explanation of the discrepancies associated with the entrainment process. Here it might be more optimal to rely directly on analysis of some of the physical properties that can indicate about the role of entrainment, e.g., moisture.

Actually, we consider MHP as an excellent tracer for downward mixing of free tropospheric air into the MBL. As figure 5 shows, the case study on March 16 indicates a 5-fold increase in MHP in this downward mixing event, similar to the increase in H2O2 itself. This nicely demonstrates the high sensitivity of the MBL boundary layer MHP concentration to transport from the FT.

Some remaining minor comment: In the revised version the following statement has been included also in response to the comments I raised.

“The deposition velocity calculated by the model for O3 does not depend on the wind speed and is about 0.05 cm/s, indicating that the deposition loss is limited by the ocean uptake resistance.”

The O3 dry deposition over the ocean still depends on wind speed (actually as it appears now it depends stronger on it than previously known due to the role of waterside turbulent transport) but its effect is minor. Consequently, I suggest to rephrase to:

“The deposition velocity calculated by the model for O3 hardly depends on the wind speed being small as ~0.05 cm/s, indicating that the deposition loss is limited by the ocean uptake resistance.”

We changed the manuscript accordingly.