

Dear editor,  
thank you very much for editing our paper. Find below the replies to the three reviewers.

## Reply to Reviewer 1

### General changes

- We improved the structure and readability of the paper. We restructured the paper by introducing a new section structure, divided into “Measurements”, “Modeling” and “Effect on chemical species”. The introduction has been expanded considerably by moving paragraphs from later sections at the beginning of the paper. The appendixes have been integrated into the main text, with the exception of Appendix A, which is quite long and contains details not of interest to everyone. Several paragraphs have been moved to positions where they fit better into the context. A “Conclusions” section has been added. A description of the sections has been added to the introduction. The abstract has been extended. Some more specific changes can be found at the end of this reply under “Additional changes”.

### Your comments

- **“... broader introduction to the basic climatology and history of the study region...”** and **“... extensive discussion of results in light of previous similar investigations ...”**

We have considerably extended the discussion of many aspects (e.g. more measurements are discussed or interannual variability is discussed, see also specific answers to other points and to the other reviewers) and added a considerable number of new references. Please understand that we do not want to write a review paper here or to repeat information in detail that has already been published elsewhere. We would like to keep our study focussed.

- **“... broader introduction to the relevant chemistry of atmospheric halogens considered to be important...”**

We have expanded the introduction to the CH<sub>2</sub>Br<sub>2</sub> calculations considerably (now in Section 5.1) and added several new references. The chemistry which is relevant here is quite simple: The most important species by far are CHBr<sub>3</sub> and CH<sub>2</sub>Br<sub>2</sub> (short-lived chlorine species are only a minor contributor to ozone loss, since the abundance of stratospheric chlorine is much larger than that of stratospheric bromine). CHBr<sub>3</sub> is mainly destroyed by photolysis, while CH<sub>2</sub>Br<sub>2</sub> is mainly destroyed by reaction with OH, which is the reason why it is treated here. The chemistry of the product gases is complicated and still not well understood, and involves washout and heterogenous reactions. But the amount of product gases can be seen as an upper limit for the amount of bromine that does not

reach the stratosphere, since only soluble species that are washed out do not reach the stratosphere in the end.

- **“mere speculation (as, for example, the brief hint at the modification of the stratospheric sulfate layer being sustained by moderate volcanic activity and increasing SO<sub>2</sub> emissions in the region and its relation to global climate, i.e., the Solomon et al. paper and WMO study)”**

We have considerably toned down our introductory remarks on these more speculative potential implications of our main findings and have focussed the paper on the main aspects.

- **“It lacks in comprehensiveness and fails to set the observations in a proper modelling context in comparison with previous field and model studies.”**

We have incorporated a considerably broader discussion on several topics into the paper (see above). The incorporation of the appendices into the main text should also help.

- **“There is no real Conclusions section”**

We have added a section “Conclusions”. Much of the material in the previous “Discussion and Conclusions” section was incorporated in the main body of the paper.

- **“And why are there five appendices? Are these considered to be less important, although they include four critical figures and key details on data sources, measurements, and modelling?”**

We have reduced the number of appendixes to one and have integrated the other appendixes and figures into the main text.

- **“The abstract mentions halogen emissions from kelp and seaweed farming as potentially important sources for reactive halogens in the stratosphere. Nothing about this statement is further substantiated in the text, not even maps of primary production or chl-a are included.”**

We have removed this statement from the abstract. This is only a minor side aspect of the paper and should not be mentioned in the abstract. Also, we have now added some more information in the introduction to the new section 5.1. We cited two references in the old manuscript which give more details (WMO, 2011 and Martinez-Aviles) and have added several other references that discuss the topic of halogen emission sources in detail (e.g. Liang et al., Warwick et al., Hossaini et al.). We have replaced the two references Quack et al. and Pyle et al. by more appropriate ones. We don't think it makes sense to show maps of primary production or chlorophyll here. We think it is clearly outside the scope of this study to add a detailed study on emissions of halogenated species here. This topic

has been covered elsewhere in great detail (see our new references), there are still large uncertainties and it would be a completely new and largely unrelated topic compared to our main topic of OH abundances. We would like to keep the study focussed.

- **“And what about seaweed farming? Any data from the study area or a survey of worldwide seaweed farming growth and related halogen emission estimates?”**

We removed any mention of this from the abstract. It was unfortunate that we had mentioned this in our original abstract, which led to some misunderstanding. This is really only a side remark in the paper. We think this issue is clearly outside the scope of the paper and would like to refer to the relevant citations here, see above.

- **“Likewise, no tabulated data on moderate volcanic activities and anthropogenic SO<sub>2</sub> emission trends in the East Asia/West Pacific region are provided.”**

This would be beyond the scope of this study. The new abstract should make it much clearer that this study is about the ozone and OH minima in the tropical West Pacific. Some discussion of the potential implications for halogenated species or sulfur is included, but these are not central for the paper.

- **“I am also missing a thorough analysis in relation to the warm pool climatology of the region, and in particular its potential relation to ENSO (2009 was an El Nino year, although anomalous SST were not reaching the study area (see, e.g., Kim et al.). The related large-scale atmospheric circulation patterns developing in ENSO causing advection of low O<sub>3</sub>, low OH air masses to the western Pacific mentioned in the paper should be thoroughly investigated! Is this a persistent feature throughout each year, or occurring seasonally, or just in relation with ENSO events, etc.?”**

Figure A4 (now Figure 5) shows that the position of the minimum of the ozone column follows the eastward shift of the warm pool during El Nino conditions to some extent, but that the minimum is very stable. The stability was also shortly discussed in the manuscript on page 28873, lines 9–11. We have now added some more discussion in the text in Section 2.3 (new manuscript).

It was said in the old manuscript that interannual variability for the density distributions is limited (page 28880, line 2). The corresponding part has now been moved into the main text. We have added figures that show the interannual variability of the density distributions of Figs. 4d and e (Figs. 7 and 8) and have added a discussion in the text. The main features, as a clear maximum between 10 S–10 N and 120 and 180 W are very robust.

- **“One previous model study comprising a large data set from multiple NASA field experiments over the Pacific, specifically PEM-West and PEM-Tropics, also including direct airborne measurements of OH in this region, should certainly be referenced and discussed for comparison of model results: Liang et al.”**

We have cited and discussed the Liang study now in the introduction to the new section 5.1. A direct comparison of our more conceptual model studies with the results of Liang et al. is not possible without access to the Liang model data. E.g. we use values relative to the boundary mixing ratios here, and Liang uses absolute values.

A discussion of the OH measurements carried out during the PEM-Tropics B campaign has been added. We have cited Tan et al. (2001) as a reference. The reference Liang et al. does neither show nor discuss measurements of OH.

- **Figure 4:** The figure is not reproduced in the correct size in the “printer-friendly” version of the manuscript. In our original manuscript, this was a big panel covering a complete page. This will be corrected in the final version.
- **Figure A2:** We have removed the Figure and the corresponding paragraph. A very similar Figure is shown in Ridder et al. (2012, Fig. 5, see citation in the paper). Instead, we added discussion on the FTIR measurements to the new section 2.1 and added discussion on the good agreement between model, FTIR and ozone sondes visible in Fig. 5 of Ridder et al. to the new Section 3.

#### **Additional changes**

- Changed “Much of our understanding of transport of short-lived species into the stratosphere is based on studies that assume fixed uniform lifetimes” to “Some important studies were based on fixed uniform lifetimes of OH in the past”.
- We rephrased section 2.1 to discuss more ozone measurements and to discuss the CEPEX measurements in more detail. In particular, we added a reference to the Appendix, where we propose that there is a low bias in the CEPEX measurements compared to our measurements. We added discussion on additional ozone sonde measurements (Fujiwara et al., Takashima et al.).
- We have split Figure 4 (old manuscript) into two Figures. These are the Figures 9 and 10 in the new manuscript.
- Figure A4 (now Figure 5) was blurry. A new version is included in the new manuscript.

- A new Figure 6 showing OH profiles from the model run and discussion in the text comparing these profiles to the PEM-Tropics B measurements has been added.
- In the description of the back trajectories, the information that the trajectories were started in January was missing and has been added.
- Added discussion of OH modeling uncertainties.

## Reply to Martin Manning

### General changes

- We improved the structure and readability of the paper. We restructured the paper by introducing a new section structure, divided into “Measurements”, “Modeling” and “Effect on chemical species”. The introduction has been expanded considerably by moving paragraphs from later sections at the beginning of the paper. The appendixes have been integrated into the main text, with the exception of Appendix A, which is quite long and contains details not of interest to everyone. Several paragraphs have been moved to positions where they fit better into the context. A “Conclusions” section has been added. A description of the sections has been added to the introduction. The abstract has been extended. Some more specific changes can be found at the end of this reply under “Additional changes”.

### Specific comments

- Page 28870, line 25: Thanks for the additional references. We have added both references to the text.
- Page 28871, line 9: We have followed all your recommendations here: The reference to Naik et al. has been added. A sentence referring to the systematic differences has been added. In addition we have considerably extended the introduction by moving several paragraphs from later sections into the introduction. That includes the paragraph about the West Pacific as entry point for stratospheric air and the reference to Fueglistaler et al.
- Page 28871, line 10: We cleaned up the structure of the paper to achieve a clearer separation between the different topics. There are now several new sections and subsections for measurements, modeling and chemistry. We have included most of the appendixes in the main text now.
- Page 28871, line 20: We moved discussion about the entry region into the stratosphere to the introduction.
- Page 28872, line 1: Only one appendix is left in the new version, which avoids the ambiguity.

- Page 28872, line 9–10: The Appendix is Appendix E. The Appendix has been integrated into the main text now.
- Page 28872, line 11–15: Unfortunately it is not easily possible to be more quantitative here or to make a quantitative assessment about the relative roles of loss in the marine boundary layer versus loss in the free troposphere. This would require a cloud resolving modelling study with interactive tropospheric chemistry, which is far beyond the scope of the current paper. We use the words “most likely explanation” to make clear that this statement is not based on such a rigorous model study.
- Page 28872, line 16–28: The Appendix has been integrated into the main text just below this paragraph. We hope that resolves the issue.
- Page 28872, line 28: Actually the key point we want to make here is that the low OH was actually seen in a measurement at upper tropospheric levels. Otherwise the discussion of OH in our paper is purely based on model results and we find it important to support that by this observation. We do only have the data that is shown in the Figure – this is the only occasion where the ER-2 has probed a patch of air that originated from the “OH minimum” area. Potential mixing of this patch of air with air from higher latitudes could only increase OH. Therefore our purely qualitative point about very low OH in this air mass is robust.
- Page 28873, line 1–11: We have reorganized the whole paper. This is hopefully much clearer now.
- Page 28873, line 12–13: Added this information.
- Page 28874, line 6–17: There is a misunderstanding here: The ATLAS run used here includes no chemistry, although ATLAS has a (stratospheric) chemistry module. The results of the chemistry run from GEOS-Chem are interpolated on the ATLAS trajectories here. This important information was missing in Appendix D and has now been added. There are two reasons for this approach: 1) A Lagrangian model like ATLAS is better suited to calculations of air mass origin. This is easily done with back trajectories, but difficult in an Eulerian model like GEOS-Chem. 2) ATLAS contains no detailed tropospheric chemistry scheme, so we have to use GEOS-Chem here.

We have moved the text of Appendix D, which clarifies the approach, into this section. Additionally, we have added some more information on how the 57% value was calculated.

- Page 28874, line 12–17: This was not intended to imply that advection of OH is a significant factor. We hope that the changes described in the last comment clarify what we have done. Nevertheless, the statement is correct as written here: It is not said that OH is advected, but that other species which ascend into the stratosphere, like CH<sub>2</sub>Br<sub>2</sub> or SO<sub>2</sub>, will encounter these OH values (no matter how these OH values are maintained).

- Page 28874, line 13: Changed.
- Page 28874, line 21: The model was explained in Appendix E, but unfortunately the reference to the appendix was missing here. We have now restructured the text and introduced a new section which aggregates all text dealing with the CH<sub>2</sub>Br<sub>2</sub> and SO<sub>2</sub> results. The text of Appendix E has also been moved into this section, so that the model is explained close to this paragraph. We hope that clarifies what we have done. The simple box model was specifically written to model CH<sub>2</sub>Br<sub>2</sub> and only contains two reactions.
- Page 28875, line 8–12: We have restructured the text and there is now a separate subsection for the SO<sub>2</sub> results. We have changed “our findings” to “model calculations” to avoid misunderstandings. These changes hopefully clarify things.
- Page 28875, line 11–12: Changed as suggested (also in the abstract, which includes the same sentence).
- Page 28876, line 1–8: This paragraph deals with the conditions after volcanic eruptions, while the paragraph before deals with the background conditions without volcanic eruptions. We do not think that we are repeating points here, the subjects of the paragraphs seem sufficiently different to us.
- Page 28876, line 9–14: Thank you for this suggestion. We have added a final statement along this line.
- Appendixes: We have moved the text of the appendixes into the main text (except for Appendix A).
- Appendix A: –
- Appendix B: Added a remark that there is some seasonal variation in OH abundances. The measurements from Wennberg et al. are from the same campaign, but the paper show only data below 50 degree solar zenith angle, which excludes the measurements that show very low OH.
- Appendix C: We have added a reference to the Data User’s Guide of TES. The TES data of Fig. A4 are mentioned in the main text (Page 28873, lines 9–11), but the only given reference there is the Appendix. We have now moved the Appendix into the main text and replaced the reference to the Appendix by a reference to the Figure. That should make it much clearer.
- Page 28879, line 21: We have changed that to “vertical motion”. The vertical coordinate is pressure at the surface and potential temperature above the tropopause and slowly transforms between these coordinates inbetween. Likewise, the “wind” matching these coordinates transforms from pure vertical winds at the surface to pure heating rates at the tropopause.

- Appendix E: –

### **Additional changes**

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- We have removed Figure A2 and the corresponding paragraph and moved the discussion into the main text.
- Added discussion on interannual variability and relation to ENSO, including two figures.

### **Reply to Reviewer 3**

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## Comments

- 1) Based on our experience with ECC ozonesondes we agree that assuming a very rapid drop of the background current after launch is an extreme approach. But as we discuss in the paper, there is a broad range of suggestions for background current corrections in the literature, which also includes the Voemel and Diaz paper. In this situation we think showing our “robust upper limit”, which is based on not subtracting any background current is useful. You may be right that this is a fairly conservative approach to establish the upper limit of our measurements but we think this is a particular strength of our study – our findings are robust even for the most extreme assumption on in flight background current decline, since we find extremely low ozone mixing ratios throughout the troposphere even then.
- 2) Overall we toned down these conclusions considerably and have focussed the study more on the main findings, which are much more robust. We have formulated the statement that the changes in aerosol may lead to more global warming more carefully and have removed it from the abstract. We do not claim that “the perturbed chemistry is global”. It is clearly stated that the “perturbed” chemistry is restricted to the West Pacific. Nevertheless, since the main source region of stratospheric air is in the West Pacific, changes in composition in the West Pacific troposphere can have a disproportionally large and global effect on stratospheric composition. Also, the paper does not claim that the “entire” photochemical balance of the lower stratosphere is “dominated” by the chemistry of the Western Pacific. We have reworded parts of the paper to avoid such a misunderstanding.

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