Reply to reviewer's comments of Lennartz et al. (2015):

Modelling marine emissions and atmospheric distributions of halocarbons and DMS: the influence of prescribed water concentration vs. prescribed emissions, ACPD

Reply to Reviewer #1, S.M. Elliott

First of all we would like to thank Dr. Elliott for the helpful review that addresses important scientific questions in our study. We would be looking forward to interacting in the future.

Reviewer (bold font): Thank you for asking me to review the manuscript "Modeling Marine Emissions and atmospheric distributions of VSLS", by Lennartz et al. The analysis in the paper is strong, and I personally found no technical errors. The work represents an important step towards fully coupled marine-atmospheric biogeochemistry modeling, which is one of the keys to building next generation Earth System Models (ESM). The authors have adopted a careful, incremental and defensible approach to the problem of com-puting sea-air trace gas fluxes in a more consistent manner. The paper is simple and effective. I wish I had personally conceived of such an elegant study. By extension the research raises interesting science and modeling questions bearing on my own work, which lies in the area of biogeochemical ESM development. Let me touch upon these issues here in the sense of an on-line discussion, while noting that it is not at all critical for the authors to address them directly before publication.

General –The overall theme here is that gas fluxes computed from surface ocean concentration distributions will improve simulation consistency, relative to the usual and standard emission data sets. But dissolved concentrations need not necessarily be climatologies. The argument can be pushed a step further –if surface water distributions are computed from dynamic on-line biogeochemistry then the entire marine system becomes unified. This is in fact the major driving force for our own model development in the U.S. Department of Energy climate system code. The possibility is opened for full CLAW-like feedback studies.

Line 70 –The importance of iodine to stratospheric ozone chemistry is mentioned as a motivation. My understanding, however, is that this particular heavy element may in fact be of even greater importance in the troposphere. Its lower atmospheric relevance occurs in the context of nucleation and coastal aerosol composition. My favorite references on the subject come from the O'Dowd and Saiz-Lopez groups. Organo-iodine compounds are apparently produced with particular intensity by the ice algae, so that there may be links through polar aerosol and cloud chemistry to albedo amplification.

Indeed, it is correct that due to the short lifetime, the direct impact of CH_3I is larger on the troposphere than on the stratosphere. Additionally, with CH_3I bringing most of the iodine from the ocean to the atmosphere, it contributes to the total loading of the iodine in the atmosphere and thus can influence tropospheric and stratospheric chemistry by product gas injection as well. We thank Dr. Elliott for the suggested reference and added it to the paper:

“Iodine oxides, which can be product gases of CH_3I are likely to contribute to nucleation and growth of secondary marine aerosol production (O'Dowd and De Leeuw, 2007). “ p. 17556, l. 21

Lines 161 and 243 –I am gratified to see that the authors have the courage to cite classic references like Liss and Slater 1974, or even Wilke and Change 1955. I grew up with these papers and agree that the pioneers should be continually recognized.

Line 182 and elsewhere –Beginning with the treatment of rain effects, I was reminded of a traditional obstacle to effective sea-air gas transfer modeling. A lingering question is, what processes contribute the largest uncertainties to dynamic flux estimation? I believe the answer has always been and remains the same. The effect of organics and surfactants on physical properties of the ocean interface drives error bars of order a factor of three in either direction. Hence the total uncertainty can approach an order of magnitude at some wind speeds. This difficulty is implicit in almost any transfer study and sometimes it is even stated directly. One often finds the information buried deep in a discussion section, since it is viewed partly as an intractable embarrassment. But Nelson Frew of WHOI began to unravel the real physical chemical issues involved beginning in the middle 1990s. In our DOE-ESM effort, we are now simulating global distributions of chemically resolved marine surfactants, initially for purposes of computing primary organic aerosol sources from bubble breaking. This involves the detailed simulation of generalized biomacromolecules and polymers from within the familiar DOC. We are hoping to make the Frew connections to laminar layer barrier and viscosity effects in the very near future. I would be interested in interacting with Lennartz and company on this topic.

As Dr. Elliott states correctly, a large uncertainty of flux estimations originates from the difficulties in including the effect of surfactants in gas transfer velocity parameterizations. We therefore included the following statement:

“Other factors that are known to influence air-sea gas exchange, such as the presence of surfactants, but parameterizations including that effect are only marginally explored (e.g. Tsai and Liu, 2003) and require global distributions of surfactants that are currently not available. First steps of including surfactants in global models are currently discussed (Elliott et al., 2014; Burrows et al., 2014).”, p. 17561, l. 16.

Line 342 –the effect of including real atmospheric DMS concentrations is surprisingly and disturbingly large. But this of course is the point of the entire exercise. To conclude, let me summarize as follows: The paper Lennartz et al. is scientifically important, complete and understandable. It aligns in several interesting ways with my own work on unified biogeochemical systems modeling, and so I have been a very receptive audience. Any criticisms or suggestions that I can offer are quite minor. As I moved through the text I found certain phrases for which the English might be improved or made more standard. These number perhaps a few per page. But in fact while I was finishing up my reading, I reflected on the potential edits and decided that they are unimportant. The work is timely to the ESM community and so it should not be delayed. I will send my long list of small recommendations only if specifically requested to do so. Please get this one in the literature as soon as possible, and encourage Lennartz plus coworkers to be in touch with me

Reply to Reviewer #2, Anonymous

We thank the anonymous reviewer for her/his helpful comments that further improve the visualization of the results and for pointing out an important reference for observational DMS data to compare our model output.

In this study, the authors implemented the online calculated emission module of very short-lived trace gases into one atmospheric chemistry model (EMAC) with prescribed seawater concentration (referred as the PWC method). The objective is to evaluate this method by comparing with the one prescribing sea-to-air emission fluxes (referred as the PE method) and observations. They concluded that the PWC method is more accurate in computing atmospheric mixing ratio of relevant trace gases than the PE method. They also investigated the uncertainties of online calculated emission associated with different air-sea transfer velocity parameterization. Generally speaking, this is a nice modeling study appropriate for the scope of ACP. I have a few minor comments listed below. A minor revision is recommended.

Specific comments: 1. Since authors mentioned one of the biggest benefits of the PWC method is to determine both direction and magnitude of air-sea exchange fluxes given the concentration gradient, It would be very interesting for authors to show a spatial map where the negative or positive fluxes reflecting either deposition or emission are in the PWC method. I think this is of great importance to be distinct from the method prescribing non-negative emission fluxes.

In figure 4, the colorbar and its range was adapted to show regions of emission or deposition more clearly.

2. Page 17566, line 17-24, the authors mainly compared modeled DMS with ship and air craft measurements and stated that "no data from ground based time series stations is available". I disagree. To the best of my knowledge, Ayers et al. (1995) provided a long time series data of DMS in Cape Grim from 1988 to 1993. Sciare et al. (2000) discussed a time-series data of DMS observed in the Amsterdam Island in the Indian Ocean. These data might be useful for the authors to validate their model in terms of the seasonal variation of DMS predicted from either PWC or PE method similar to Figure 7/8/9.

We thank the reviewer for pointing us to the DMS time series data. We have added an additional figure to the manuscript with published DMS data from two time series station, one on Cape Grim and one on Amsterdam Island (Figure 9, now previous Fig. 9 shifted to Fig. 10).

"Additionally, DMS data from 2 time series stations, Cape Grim, Australia, (Ayers et al., 1995), 1990-1993, and Amsterdam Island in the Indian Ocean (Sciare et al., 2000), 1990-1999, was used for comparison (Table 2). ", p. 17566, l. 21

Deleted: "Four ship campaigns were chosen for comparison of DMS, since long-term measurements of atmospheric mixing ratios of DMS are not available. In addition, no observations from time series stations are available, which makes an analysis of seasonality as done for the halocarbons difficult." P. 17571, l. 15-18

Added:

“The observed seasonality of DMS mixing ratios at Amsterdam island is well reflected in the simulations except for the summer months, where PWC and PE overestimate the monthly mean by a factor of up to 4.6 (PWC) and 6.7 (PE) (Fig. 10). At Amsterdam Island, the simulated annual mean atmospheric mixing ratio of 180.7 ppt in the PWC set-up agrees very well with observed annual mean of 181.2 ppt, whereas the simulated annual mean in the PE set-up is 268.5 ppt. At Cape Grim, the results of the two set-ups do not differ that much, and both simulations underestimate the mixing ratios measured during austral summer. ”

Reply to anonymous reviewer #4

We thank the anonymous reviewer #4 for her/his detailed and very comprehensive review that helped to improve the clarity and the strength of the argumentation.

Overall comments:

This manuscript describes numerical experiments assessing the uncertainty in emissions of DMS and halocarbons, conducted using recent climatologies of ocean water DMS concentrations (Lana et al., 2011) and halocarbons (Ziska et al., 2013), and online calculation of emissions using a parameterization of the air-sea transfer velocity. Simulations with online emissions are compared that use eight different parameterizations of the transfer velocity (2-year simulations + 1 year spin-up); prescribed emissions are also compared with default online emissions (23-year simulations). Careful intercomparisons of model parameterizations within the same model system are highly valuable and important in improving understanding of differences in performance of different parameterizations. The study is well-designed and has been carefully and thoughfully carried out. The results are mostly well-presented and discussed, but some additional information is needed to clearly show the results of the model-observation comparison. In particular, the presentation of error metrics should be improved, and alternative error metrics for characterizing the model performance should be considered. Also, a direct comparison of the observations and subsampled model output should be provided in a figure. After these and the remaining detailed comments below have been addressed, I would recommend this paper for publication.

Major comments:

1. p. 17564, l. 11-12: The simulations comparing the effects of different transfer velocity parameterizations are each two-year simulations, with one year of spin-up. Two years may not be long enough to obtain a good statistics. Please provide results from simulations lasting at least five years (plus spin-up time), or justify why two years is sufficient for this study.

The reviewer raises an important point about the length of the simulation with respect to the statistical output. To obtain a reasonable amount of data to compare against observations, we performed runs ranging longer than 20 years when evaluating PWC and PE. For the comparison of the k-parameterizations, we compare between two model runs and not against observations. The largest difference between the compared runs is the k-parameterizations based on surface wind speed and sea surface temperatures, and both wind and sea surface temperature are very similar in all 2-year runs due to the nudging. The difference between the k-parameterizations on the global annual mean is not time dependent, and because of the short tropospheric lifetime of the gases, we believe that the differences between the k-parameterizations can be detected in simulation periods of 2 years.

2. p. 17564, l. 19 - p. 17565, l. 2: Please provide some brief information for the reader about how these different transfer velocity parameterizations were developed, e.g., are they based on laboratory or field observations?

The paragraph where the different parameterizations were introduced (p. 17561, l. 3-l. 15) has been changed to provide the suggested information on the original methodology used (i.e. experiments

conducted in the field or in the laboratory). A more detailed discussion would be beyond the scope of the manuscript, but all of the original references are provided.

Revised text:

"The water-side transfer velocitiy k_w is often parametrized in relation to wind speed with linear (e.g. Liss and Merlivat, 1986), quadratic (e.g. Ho et al., 2006) or cubic (e.g. Wanninkhof and McGillis, 1999) dependencies. Differences between these parameterizations arise from different techniques to determine k_w. The kw parameterizations tested in our study result from tracer release experiments in wind tanks (Liss and Merlivat, 1986), from deliberate tracer techniques in the open ocean (Nightingale et al., 2001, Ho et al., 2006) or from direct flux measurements using eddy covariance (Wanninkhof et al, 1999, Marandino et al., 2009, Bell et al., 2013). Additional drivers of gas exchange, e.g. bubble mediated transfer (e.g. Asher and Wanninkhof, 1998) and enhancement in the presence of rain (e.g. Ho et al., 2004) are discussed. Bubble mediated transfer has been suggested to be influential for gases with low solubilities, since they more quickly escape from the liquid phase into the bubbles. Asher and Wanninkhof (1998) reanalysed data from a dual tracer experiment and found a better fit when bubble mediated gas transfer was considered in the flux calculations. Bubbles are more easily transported to the surface and released to the atmosphere, thereby adding to the total flux. Rain is believed to increase the flux under calm wind conditions due to an alteration of the sea surface, which was tested in a dual tracer experiment in the laboratory (Ho et al., 2004). "

3. When model resolution is increased, a greater amount of wind gustiness can be represented. For emission parameterizations with a non-linear dependence on wind speed, this leads to resolution dependencies in the emissions. How much do the online-calculated transfer velocities in this study depend on model resolution, and were these parameterizations previously developed / tuned for use at a particular spatial resolution?

The effect of model resolution was investigated according to the reviewer's comment and we found only minor differences between simulations in the T42 grid and a finer resolution, the T106 grid. Results for this resolution test are now included in the supplementary material. Since we are comparing emissions on a global scale, the mean averages between the different resolutions are compared.

We added to the manuscript:

Supplementary material: S-Table 2

"The effect of resolution on the results tested with a finer resolution (T106) was only minor (see S-Tab. 2, supplementary material)." P. 17559, l. 12

4. Table 5: I am finding the row labels in this table confusing – it needs to be explained better what the numbers represent. For instance, I expected the row "Total ship" to equal the sum of the four rows above it, but it doesn't. It is unclear which rows are absolute differences expressed as ppt, and which are relative differences expressed as percentages. This may seem comparatively minor, but I am listing it as a "major issue" here because it makes it difficult to understand what results were obtained. The caption seems to indicate that some of the statistics presented here are "normalized mean bias" (i.e., sum (model – obs) / sum (obs) x 100%). If that's correct, please use this standard terminology for clarity. The normalized mean bias suffers from the difficulty that it is asymmetric with regards to overestimation (which is unbounded) and underestimation (which is

bounded by 100%). It would be valuable (and should require minimal additional effort) to also provide additional a performance statistic such as the mean normalized fractional bias (Yu et al., 2006), which is a statistic of relative bias that is symmetric to relative values of overestimation and underestimation.

We provided scatterplots of the aircraft data as suggested (see answer above), but would prefer to include them in the supplement with a reference in the main text to limit the length of the manuscript. With the data that was illustrated in the scatterplots (see 5. In this reply), we performed the error metrics according to Yu et al. (2006) as suggested by the reviewer. We performed them for all substances and replaced Table 5 with a new table for all substances, including the centered bias and the relative bias as suggested by the reviewer. We also used the terminology from Yu et al. (2006) as suggested.

Added text:

"However, the PWC reduces discrepancies within both ship and aircraft campaigns by a factor of 2 (Tab. 5), as the mixing ratio is overestimated by a factor of 0.61 in PWC as opposed to 1.31 in PE." P. 17571, l. 20

The equations to compute the statistics are also included in the supplements now for the convenience of the reader.

5. Please add one or more scatterplots showing subsampled model output versus observations. This is especially important for the ship and aircraft observations, since the paper currently doesn't include any figure showing the values of these observations, or how they compare with the model. However, it would be useful for the ground-based observations as well.

We provided scatterplots of the aircraft data as suggested, but would prefer to include them in the supplement with a reference in the main text to limit the length of the manuscript. We included the following on p. 17569, l. 12:

"A scatterplot for direct comparison between model output and aircraft or ship observations is provided in the supplements in S-Fig. 2."

6. Comparison of various transfer velocity parameterizations: how well does each parameterization compare to observations? Can any conclusions be drawn about which parameterization is most realistic and which should be used?

The reviewer touches an important point with this comment, but conclusions about the choice of k_w-parameterizations are beyond the scope of our study. We aim to compare the effects of two different methods of considering marine emissions in atmospheric climate models. The climatologies used in this study are based on measurements which are extrapolated to a global grid. We therefore believe that the uncertainty that is attached to these climatologies does not allow for conclusions of the different parameterizations, because deviation between observed and modelled mixing ratios cannot be exclusively related to the parameterizations but also to the concentration input in the first place.

7. Model setup: What feedbacks processes of the VSLS compounds onto climate (via radiation, clouds) are represented in the model configuration used? Do the atmospheric chemistry reaction

mechnisms used here interact with aerosols, and which aerosol model / microphysical and chemical representations were used?

The simulations were performed with a simplified chemistry using prescribed rates for efficiency reasons. This approach is reasonable, because the resulting atmospheric lifetimes of the gases agree well with previous studies. However, that means that feedbacks between online computed emissions and radiation or clouds have not yet been tested. Also, no aerosol model was used. Since one of the goals of this study is to compare the emissions with (PWC) or without (PE) taking the current state of the atmosphere into account, the feedback on atmospheric chemistry does not influence the main results presented here. Future studies will consider this point.

Minor and technical comments:

p. 17557, l. 18: "Compared to" -> "In contrast to"

changed as suggested

p. 17559, l.4: "EMAC/MESSy" -> "ECHAM/MESSy"

changed as suggested

p. 17559, l. 11: The model resolution used was T42L39, which is reasonable but at the lower end of the resolutions typically used for global modelling – how sensitive are the processes modelled here (air-sea gas exchange, atmospheric transport and chemistry) anticipated to be to increases in model resolution?

see reply to major comment #3

p. 17559, l. 24-26: "Photolysis rates for VSLS were calculated by the TOMCAT CTM" – Since the results will depend strongly on these rates, please describe this in a little bit more detail here, so that readers can get a quick idea of what was this photolysis rate product represents. In particular, what is the time/spatial resolution (e.g., are the rates that are used monthly means?), and are photolysis rates entirely prescribed, or is there some ability for them to respond to online, prognostically calculated variables (particularly radiative transfer)? How is the use of prescribed (rather than online-calculated) photolysis rates expected to affect the results?

We have added more details of the TOMCAT simulation used to generate the photolysis rates to the text. The revised text is as follows:

"Monthly mean photolysis rates for VSLS were calculated by the TOMCAT CTM which has been used extensively to examine the tropospheric chemistry of VSLS (e.g. Hossaini et al., 2013). These fields were provided at a horizontal resolution of 2.8 x 2.8 degrees (longitude x latitude) and on 60 vertical levels (surface to ~60 km). TOMCAT calculates photolysis rates online using the code of Hough (1988) which considers both direct and scattered radiation. Within TOMCAT, this scheme is supplied with surface albedo, monthly mean climatological cloud fields and ozone and temperature profiles. The photolysis rates have recently been used and evaluated as part of the ongoing TRANSCOM-VSLS model intercomparison project (http://www.transcom-vsls.com)."

p. 17563, l. 8: please mention whether the regridding performed using the "extensive" regridding algorithm (conserving global mass). Please also mention the spatial resolution of the original

datasets, and their time resolution. When applying the datasets in the online calculation of fluxes, are the prescribed ocean concentrations fields interpolated in time to the model time step?

For regridding, the ncregrid algorithm (Joeckel et al., 2006) was used, using the intensive algorithm, as concentration is an intensive quantity.

Added: p. 17563, l. 1: "The emission climatology from Z13 is based on constant water and atmospheric concentrations extrapolated from ~5,000 measurements, using 6-hourly ERA-Interim wind, pressure and sea surface temperature fields and the Nightingale et al. (2000) parameterization for water-side transfer velocity. "

p. 17563, l. 9: "The climatologies, prescribing emissions and concentrations of the gases of interest (CH_2Br_2, $CHBr_3$, CH_3I and DMS) were regridded to the T42 grid of EMAC with ncregrid (Jöckel et al., 2006a), which is in all four cases coarser than the original grid described in Z13 & L11 (1°x1° in both)."

Concerning the interpolation to the time step: Z13 is an annual climatology and thus stays constant in each time step. For L11 monthly climatologies of emissions and concentrations were available,and they were not interpolated with respect to time.

p. 17563, l. 17-18: "opposite to"-> "as opposed to" or "in contrast to"

changed "opposite to" to "as opposed to"

p. 17564, l. 19: "overview on" -> "overview of"

changed as suggested

p. 17564, l. 10: "sensitivity towards"

changed as suggested

p. 17565, l. 9-10: "These two parameterization for kw were added to the submodule code of AIRSEA." Will the implementations of the parameterizations be made available to the public by contributing them back to EMAC for future released versions? (also please note the typo in this sentence).

p. 17565, l. 9-10: typo corrected. The code is made available to the community and is part of the most recent release (MESSy 2.52).

Added: "Both newly implemented parameterizations are part of the most recent release MESSy 2.52." p. 17565, l. 16.

p. 17565, l. 13: "until the wind speed of " could perhaps be changed to "at wind speeds below"

changed as suggested

p. 17566, l. 6: "same location of" -> "same location as"

changed as suggested

p. 17568, l. 19-20: "respond stronger" -> "respond more strongly"

changed as suggested

p. 17570, l. 24: “both and” -> “either or”

changed as suggested

p. 17572, l. 8: “eight 2 year” -> “eight 2-year”

changed as suggested

p. 17572, l. 9: “Largest uncertainty” -> “The largest uncertainty”

changed as suggested

p. 17573, l. 4: The sentence beginning with “White cap coverage” needs revision.

The sentence was revised to “The parameterisation based on white-cap coverage (A98) also has small but ambivalent effects on the global flux for the different compounds (simulation 8, Tab. 4). ”

p. 17574, l. 22: instead of “uncertainties”, the term “relative differences” (or similar) should be used for clarity. These are not really uncertainties so much as differences between the results of different parameterizations.

changed “uncertainties” to “relative differences”. Thanks for pointing that out.

Table 4: why not convert the parameterization for simulation 10 into cm/h for better comparability?

k_{720} for simulation 10 is now converted to cm/hr

Figure 6: Are these zonal means? Please clarify.

Inserted into caption: “Model output was subsampled at locations and times of observations and binned for direct comparison.”

Figure 7: Are the standard deviations here the standard deviations of monthly mean values? Please clarify. Please also remind the reader here (i.e. in the caption) how many / which years of observations were used.

added “ standard deviations of monthly means” and “Monthly time series of at least 7 years were averaged, the exact periods are listed in table 4.”

Figure 10: Taylor diagrams are calculated from centered statistics and can obscure information about the mean bias. Please also print the mean bias and uncentered RMSE on the plot for the reader’s information / reference.

We included the reviewer’s suggestions in the new Table 5 that now includes the biases and error metrics for all compounds.

Reply to reviewer Philip Cameron-Smith

We thank Dr. Cameron-Smith for his detailed comments that helped to improve the clarity of the manuscript.

The manuscript by Lennartz et al. is well written and well designed. It primarily studies the value of using ocean concentrations to generate trace-gas fluxes to the atmosphere that are more physically consistent than specifying the fluxes directly (which is what is commonly done for chemistry transport models). I did not expect the improvements to be as large as shown here, so I think this work will be important for many atmospheric chemistry modeling groups. I am surprised that a study like this hasn't been done before, but I am not aware of one.

I do not have any major suggestions.

I have one general comment: There were a number of times when there appeared to be minor duplication of information or comments in different parts of the manuscript. Sometimes this is useful for the reader, and I didn't notice any major cases, however I suggest the authors look for opportunities to eliminate duplicative text.

Below is a list of minor suggestions for the consideration of the authors:

p17555, line 10: Add "the" before "ocean".

Changed as suggested

p17555, line 11: Add "in the" before "atmosphere".

Changed as suggested

p17555, line 17: Expand acronym "VSLS".

p. 17555, l. 17: acronym expanded to: "… dampen or even invert the fluxes (i.e. deposition instead of emissions) of very short lived substances (VSLS)."

p17555, line 26 & 27: "k" is not defined in abstract. I suggest rewording to eliminate mention of "k" in the abstract.

Changed "Calculating emissions online also enables effective testing of different air-sea transfer velocity parameterizations *k*, …" into "Calculating emissions online also enables effective testing of different air-sea transfer velocity (*k*) parameterizations,…"

p17556, line 12: Only DMS is discussed in the rest of the manuscript, so I suggest removing the other sulfur species.

p. 17556, l. 12: Since we want to highlight that there are other sulfur compounds emitted, but DMS is quantitatively the most important, we left the sentence but added: "Thus, we focus on DMS in this study."

p17556, line 17: Delete "effectively"

changed as suggested

p17557, line 29: DMS emissions were also modelled in one of my papers. However, since I have a conflict of interest, I leave it entirely up to the authors to determine whether it is appropriate to mention it. This paper calculated DMS emissions from a coupled atmosphere-ocean model, with ocean biogeochemistry but no atmospheric chemistry. There was no comparison of the fluxes with observations.

The reviewer raises an important point about the coupling of ocean and atmospheric models to evaluate ocean fluxes. We haven't mentioned this because our focus lies on how to treat oceanic emissions in stand-alone atmospheric models. To better clarify this, we modified the paragraph to:

"Oceanic DMS emissions have been evaluated in coupled ocean-atmosphere models (Kloster et al., 2006, Cameron-Smith et al., 2011) or modelled online during a test for the implementation of different submodels (Kerkweg et al., 2006b). In our study, the focus lies on how to consider oceanic emissions in an stand-alone atmospheric model, and uses the most updated DMS concentrations available (Lana et al., 2011). Additionally, we compare the output of the two methods with observations from aircraft and ship campaigns."

p17558, line 20: Clarify what is meant by "comparable set-up".

To explain "comparable set-up", we changed "To obtain a comparable set-up, we use water concentration climatologies and corresponding emissions climatologies by Ziska et al. (2013) for halocarbons and Lana et al. (2011) for DMS." Into "To compare the simulation set-up with prescribed emissions to the set-up with prescribed water concentrations, we used the same concentration climatologies that were used to create the emission climatologies. In our study, these concentration and corresponding emission climatologies were published by Ziska et al (2013) for the halocarbons and Lana et al (2011) for DMS."

p17558, line 27: Replace "towards" with "to".

Changed as suggested

p17559, line 7: Typo "submodule"

changed "submodul" into "submodule"

p17559, line 14: Replace "on" with "of".

Changed as suggested

p17560, line 5: Modify to "...167 days, which was found in Hossaini...".

modified to "…167 days, which was found in Hossaini…"

p17560, line 11: A DMS lifetime of 3 days seems long, and is longer than the 1 day mentioned in section 1. Is this a typo?

Thanks for raising the point of the DMS lifetime. To better clarify this point, we changed the manuscript at two locations. Since the lifetime of DMS varies due to the presence of hydroxyl and nitrate radicals, we rather provide the range of lifetimes in the introduction rather than one number:

“DMS has a shorter lifetime of 11 min to 46 h (Barnes et al., 2006; Osthoff et al., 2009) compared to CH_3I. Despite the short lifetime, there is potential even for the very short lived DMS to be transported to the tropical tropopause layer (TTL) in convective hot spot regions (Marandino et al., 2013a; Marandino et al., 2013b).” p. 17557, l.1.

“The tropical lifetime of DMS in our study ranges between less than 1 day and up to 3 days, and is thus within but at the higher end of the range of 11 min to 46 hr (see introduction).” P. 17560, l. 11

p17560, line 26: I assume T is “air temperature”. If so, I suggest adding “air” before “temperature”.

changed to “air temperature”, also in equation (2)

p17561, line 19: Rephrase to remove the ‘e.g.’.

rephrased to “The parameterizations of k_{air} according to Kerkweg et al. (2006a, eq. 3 and 4 therein) assumes a dependency on the friction velocity and surface wind speed, and is considered in the AIRSEA submodel.”

p17562, line 24: I assume the ‘/’ is intended to mean ‘and’. Since ‘/’ can have multiple meanings, I suggest replacing with ‘&’ throughout the manuscript.

l. 24 and after: replaced “Z13/L11” by “Z13 & L11”

p17563, line 17: I find this sentence confusing. I suggest it be rephrased.

p. 17563, l. 17: changed “The mean wind speed in the model was 7.51 m s^{-1}, 4.7% larger than in Z13 and 2.7% larger than in L11.” into

“The mean wind speed in the EMAC simulations (PWC, PE) was 7.51 m s^{-1}, which is slightly larger than the wind speed used to calculate the emission climatologies in Z13 (EMAC is 4.7% larger) and L11 (EMAC is 2.7% larger).” Thanks for pointing out this ambiguity.

p17565, lines 4-8: I find this sentence confusing. I suggest it be rephrased.

p. 17565, l. 4-8: changed “Two additional simulations were performed, with a different k_w-parameterization, used here only for DMS. These parameterizations have been derived from in-situ eddy covariance measurements and deviate from previously published parameterizations, because the transfer velocity does not increase at wind speeds higher than 11 m s^{-1} (Bell et al., 2013) or because a linear relationship to wind speed is suggested (Marandino et al., 2009). ” into

“Two additional simulations including only DMS were performed to test the effect of two recently published parameterizations of k_w. These two parameterizations have been derived from in-situ DMS eddy covariance measurements and deviate from previously published parameterizations. Bell et al. (2003) observed that the transfer velocity does not increase at wind speeds higher than 11 m s^{-1}. Marandino et al. (2009) found a linear dependency between wind speed and the transfer velocity k_w for DMS.”

p17565, line 9: Typo “parameterizations”.

typo in “parameterizations” corrected

p17566, line 10: Modify to "...1990s onwards..." and p17566, line 10: Rephrase sentence, since a couple of the stations didn't start until 2002 or 2004.

p. 17566, l. 10: modified to "Nine coastal ground stations from NOAA/ESRL, where halocarbons have been measured by the NOAA global flask sampling network starting from 1990-2004, from the database HalOcAt (Ziska et al., 2013), were chosen for comparison due to their location close to the coast (Tab. 2)."

p17566, line 22: Typo "aircraft".

p. 17566, l. 22: typo in aircraft corrected

p17567, line 4: The sentence is a little confusing. In particular, it isn't clear what 'differ' is referring to.

p. 17567, l. 4: changed "The long-term mean of global emissions (1990-2013, simulation 1 in Tab. 1) based on PWC differ for the four gases tested and varies between +11% (CHBr3) to -28% (CH_2Br_2) (Tab. 3), but yield globally a similar spatial pattern of emissions as Z13/L11 (Fig. 4 and 5)."

Into

"The long-term mean of global emissions (1990-2013, simulation 1 in Tab. 1) based on PWC is different from the offline calculated emission climatologies for all four gases. The magnitude of this difference varies between the gases +11% (CHBr3) to -28% (CH_2Br_2) (Tab. 3). The global spatial pattern of the PWC emissions is similar to the spatial patterns in Z13 & L11 (Fig. 4 and 5)."

p17567, line 16: Clarify which two approaches are being referred to.

changed to: "The main differences between PE and PWC result from…"

p17568, line 17: Typo "ratios".

typo corrected

p17568, line 19: Replace "stronger" with "more strongly".

replaced as suggested

p17572, line 2: This sentence essentially duplicates the previous sentence.

deleted sentence

p17572, line 8: Typo "2-year".

typo corrected

p17572, line 9: Start sentence with "The".

"The" inserted

p17572, lines 23-26: Long sentence. Consider rephrasing.

changed “The k_w parameterization adding flux under calm conditions due to precipitation (simulation 7 in Tab. 4) resulted in a 4 % (CH_2Br_2) to 6% (DMS) additional flux (Tab. 4) to the atmosphere for all the compounds compared to the reference flux using H06 alone (simulation 6, Tab. 4).” Into

“The k_w parameterization in simulation 7 (Tab. 4) increases the flux under calm conditions due to precipitation. This increase ranged from 4 % (CH_2Br_2) to 6% (DMS) (Tab. 4) when compared to the reference flux using H06 alone (simulation 6, Tab. 4).”

p17573, line 1: The ‘/’ is confusing. Should it be “~” ?

“/” changed to “ ~ ”

p17573, line 5: “an own” does not make sense to me. Suggest rephrasing.

p. 17573, l. 5: changed “White cap coverage as an own parameterization according to A98 also has small but ambivalent effects on the global flux for the different compounds (simulation 8, Tab. 4).”

into

“The parameterisation based on white-cap coverage (A98) also has small but ambivalent effects on the global flux for the different compounds (simulation 8, Tab. 4). “

p17573, line 11: It might be less confusing to change “reduced” to “changed”, and make the numbers negative.

changed as suggested

p17573, lines 23-27: This sentence partially duplicates the previous sentence.

deleted “Marine emissions are thus modelled more consistently, as the concentration gradient that determines the direction and the magnitude of the emissions is in agreement with the modelled atmospheric boundary layer mixing ratio and the prescribed ocean surface concentration of the gas.”

p17574, lines 27- : This is a long sentence. Suggest rephrasing.

changed “The approach of modelling emissions online was successfully applied for the very short-lived halocarbons for the first time and was based on the submodel AIRSEA coupled to EMAC by Pozzer et al. (2006).” Into “The approach of modelling emissions online was successfully applied for the very short-lived halocarbons for the first time. The approach refers to the submodel AIRSEA coupled to EMAC by Pozzer et al. (2006).”

p17575, lines 6-7: Suggest deleting ‘on one hand’ and ‘on the other hand’.

changed as suggested

p17575, line 26: Suggest deleting ‘would like to’.

changed as suggested

Table 3: Replace ‘a oceanic’ with ‘an oceanic’.

changed as suggested

Table 4: Replace ‘wind speed in’ with ‘wind speed at’.

changed as suggested

Figure 2: Add units to graph for k660 axis, caption: Switch ‘Marandino et al.’ and ‘M09’ for consistency.

Figure 2: caption changed as suggested, units added to y-axis