

Review 2

We thank the second Reviewer for his/her useful comments, which help improving the manuscript. We reproduce below the comments in italics with our answers for each of them. The modified text is presented in blue. Line numbers refer to the first submitted version of the paper.

General comments:

- *This paper presents an evaluation of ocean DMS in CMIP6 models over the historical period, and discusses their projected changes by the late 21st century under SSP585. To my knowledge, no previous work on DMS has been done using CMIP6 models. Therefore, this paper provides useful insights into the current state of DMS represented in the latest generation of ESMs. I recommend publication after major revisions, addressing my general and specific comments below.*

The historical evaluation is very extensive, but maybe a bit too extensive to be included in the main text. I do not suggest to delete anything, but I do suggest to move some content into Supplementary Information (SI). One suggestion is to move Section 3.1.2 and associated figure/table (Figure 5 & Table 5) into SI. I particularly pick on this section because: (1) this section compares the models with L11 only, which is now considered to be outdated (i.e. G18 and W20 are better replacements); and (2) this section compares over small biogeographical regions, in which global models are not necessarily expected to perform well. I think Figure 3 is just sufficient for regional evaluation of these coarse-resolution models.

We carefully examined this suggestion to move Section 3.1.2 into SI. However, we believe that it must remain in main text for several reasons that we explain hereafter. First, this Section leads to an important conclusion that the model skills in reproducing observed seasonal cycles depend on the overall location, with better skills in mid and high latitudes regions (polar and westerly biomes) than in low latitudes regions (trade biome, and low latitude coastal provinces). Second, this Section introduces the methodology of analysis based on biogeographical provinces and biomes, which is further used in Section 4 (Figures 10 and 13). We believe that the presentation of the ocean partitioning (first paragraph of Section 3.1.2, coloured map in Figure 5) must be presented in the main text. Reviewer #2 points out that Figure 3 also provides a regional evaluation which is sufficient for our analysis. Figure 3 indeed presents a zonal average of DMS concentration. However, zonally averaged results do not allow a biome analysis such as that presented in Section 3.1.2, which brings useful insights about the model performance. We also highlight that as compared to Figure 3, Figure 5 and Table 5 provide an evaluation metric (correlation coefficients) which is helpful to draw the conclusions on factual elements. Last, Reviewer #2 also mention that "L11 (...) is now

considered to be outdated” to justify that Section 3.1.2 should be moved to SI. However, as stated in the last paragraph of this Section (l. 385-389), the same provinces- and biomes-based analysis carried out with W20 as the reference climatology, and provided in the SI, results in similar conclusions regarding the models skills. We thus believe that the comparison against L11 is meaningful and should be kept in main text.

- *In addition to the environmental variables considered in the paper, I suggest to consider three additional variables for analysis: pH, MLD, and SST. Projected changes in these variables might play a substantial or additive role, as they influence directly or indirectly the DMS concentration and flux, as parameterised in the models. The Arctic might have experienced greater changes in these variables, so it is worthwhile checking these variables.*

These three variables can indeed play a role on DMS concentration and flux, however a detailed sensitivity analysis of each of them is above the scope of this study. Furthermore, we believe that it would not necessarily be relevant to support the main conclusions of the paper, regarding the multimodel evaluation and assessment of future trends. To explain further our point, we first tackle the question of the relation of these three variables with the DMS concentration. The sensitivity study carried out by Wang et al. (2020, Fig. 9) shows a negative correlation between MLD and DMS concentration, which is quite straightforward (dilution effect, as explicitly accounted for in the parameterisation of Aranami and Tsunogai (2004) as used in MIROC-ES2L). In this sensitivity study by Wang et al. (2020), the effect of SST increase is more balanced and depends primarily on the latitude, with a decrease in DMS at low latitudes but an increase for higher latitudes, especially in the Southern Ocean. However, Wang et al. (2020) show that individual linear regressions for both variables explain very little ($< 10\%$) of the observed variance in DMS concentration (7 % for MLD, 2 % for SST). Thus, both variables are expected to have little predictive ability to infer the evolution of DMS concentration. To some extent, this is confirmed for SST, since all models unanimously predict an increase in ssp585 simulations, but disagree on the trend of DMS concentration. Conversely, the study of Wang et al. (2020) shows that SST and MLD are good predictors for DMS when they are associated to other biological and climatological variables in the neural networks. Regarding the pH, only CNRM-ESM2 accounted for a dependency of DMS concentration onto this variable, which prevents from any multimodel comparison.

Regarding the DMS emission to the atmosphere, we agree that there is a positive dependence of the flux on the SST, at least when a formulation of the Schmidt number for DMS as a function of temperature is used (such as that provided by Wanninkhof, 2014). We had already mentioned this effect in the paper at L504. For clarity, we added an additional Figure in the SI (Fig. SI-7) showing the timeseries of SST for both historical and ssp585 simulations as modelled by the four evaluated models. We updated

a sentence accordingly to refer to this new Figure (at L505): "This is likely explained by the positive dependence of the gas-exchange parameterisations on the SST, which is sharply increasing in ssp585 simulations (see the timeseries of modelled SST in Fig. SI-7)."

0.1 Specific comments:

- *L48: replace "last" with "latest".*

The entire sentence has been changed.

- *L49: I'm not sure if "unprecedented" is an appropriate term here, considering that: (1) Tesdal et al. (2016) have incorporated more products (measurement/empirical/prognostic approaches) in their assessment; and (2) there are only 4 ESMs in CMIP6 that simulated ocean DMS. Has this number increased/decreased from CMIP5?*

We agree with your comments. The entire sentence has been changed, and "unprecedented" is no longer used. To answer your question about the change between CMIP5 and CMIP6 models, interactive modelling of DMS is indeed a new feature in several CMIP6 models and no CMIP5 models include prognostic DMS (see Séférian et al., 2020, Table 2).

- *Sec.2.1.1: Given the dependence of DMSP (DMS) production rate on phytoplankton species, I suggest to list the types of phytoplankton and cellular quota (sulfur to carbon/CHL ratios) specified in all of the 4 models.*

We chose not to provide additional details on the DMS production in this paper. Readers are referred to the specific references.

- *L98&L101: I'm confused about pH dependency. In L98, it says DMS release is computed as a function of pH. In L101, it says it has not been activated in CMIP6 runs. So which one is used for this paper? If it has not been activated, the word "pH" should be removed from L98.*

Indeed, the pH dependency is implemented in NorESM2, but was not activated in CMIP6 runs. We removed "pH" at L98 as suggested. We also rephrased the sentence L100 from:

"The tunable pH dependency was not present in the original parameterisation of Kloster et al. (2006), and has not been activated in CMIP6 runs (Tjiputra et al., 2020)"

to:

"Although a tunable pH dependency, that was not present in the original parameterisation of Kloster et al. (2006), has been implemented in NorESM2, it has not been activated in CMIP6 runs (Tjiputra et al., 2020)."

- *L142: Is there plan to publish the DMS data for MIROC-ES2L on ESGF nodes in the near future?*

MIROC-ES2L data have now been uploaded on ESGF nodes. We updated the sentence at L142 to remove the note about MIROC-ES2L data: "All datasets were downloaded from ESGF nodes, ~~except for the DMS variables of MIROC-ES2L, which were obtained directly from the MIROC team.~~" We also updated Table 2 and the relevant figures: all ancillary variables from MIROC-ES2L in ssp585 simulations now account for the same number of realisations (10).

- *L158: What about mixed layer depth (MLD)? Given its direct effect on DMS in diagnostic models, I think MLD should be assessed in addition to Chl.*

Please see our answer to your general comment. Also, MLD is available for only 3 over 4 models, with limits the potential for a multimodel evaluation.

- *L159: In addition to these, I suggest to show pH of the models whose DMS depends on pH. The parameterisation of Six et al. (2016) has quite strong pH effect, so this might play an important role in some regions like the Arctic. Figures can go into SI.*

See our answer to general comment. CNRM-ESM2-1 is the only model where pH dependency onto DMS concentration is implemented and used in CMIP6 simulations. However, the overall effect of this has been found to be limited.

- *L163: I am not sure if I get this correct. Is MMM calculated by averaging the ensemble means of the 4 models? Or is it calculated by averaging the ensembles of the 4 models (11 + 10 + 3 + 16 for historical)? I think it is the former, but it is not clear from this sentence.*

The MMM is indeed calculated by averaging the ensemble means of the four models. We rephrased the sentence at L163 as follows:

"In the following, the multi-model ensemble mean (hereafter MMM) is computed from the various model outputs using an equal weight for each model, irrespective of the number of realisations."

now reads:

~~"In the following, the multi-model ensemble mean (hereafter MMM) is calculated by averaging the ensemble means of the four models using an equal weight for each model, regardless of the number of realisations."~~

- *Figure 2: For readability, indicate in the caption whether these differences represent model-minus-obs or obs-minus-model.*

We added "model minus climatology" in the caption.

- *L365: why is it "striking" that models do well in these regions?*

We replaced "striking" by "noteworthy".

- *L392: Instead of text, it might be helpful to visualise the different wind-speed-based parameterisations used by these models. Consider creating a*

simple plot like Figure 2 of Ho et al. (2006) (but do this for Schmidt number for DMS).

Ho et al. (2006): *Measurements of air-sea gas exchange at high wind speeds in the Southern Ocean: Implications for global parameterizations*, *GRL*, 10.1029/2006GL026817

Thank you for pointing this specific figure in the Ho et al. (2006) paper. It is indeed very informative. However, although it appears fully appropriate in Ho et al. (2006) whose purpose is air-sea exchange parameterisation, it is not the case of our paper. We now refer to similar figures from the original papers. The modified version of our text now reads (L404):

”The reader is referred to Figures 12 of Nightingale et al. (2000) and Figure 2 of Wanninkhof (2014) to illustrate the relationships between wind speed and exchange coefficients for the three gas transfer parameterisations of interest here.

- L410: *In addition to wind, would temperature bias play a role in modifying flux via solubility/diffusivity? SST figures could be added to SI.*

In the evaluated models, the solubility of DMS (reverse flux from the atmosphere to the ocean) is neglected. However, the diffusivity is indeed dependent onto SST, thus a temperature bias can affect the flux calculation. However, we evaluate that this bias is small as compared to the overall uncertainty of flux parameterisations, and to the large flux range obtained with various existing parameterisations. To give a rough estimation, using a 2 K SST bias, the difference in Schmidt number is around $\sim 5\%$ at 20 °C, while the difference of emission depending on the flux parameterisation is in the 35–40 % range (see Tesdal et al., 2016, Fig. 7). We added a sentence at L404: **A bias in modelled SST can thus contribute to the bias in flux calculation, but is estimated to be smaller than the uncertainty of flux parameterisation.**

- *Figures 6&8: I suggest to add a subplot showing the results of Wang et al. (2020), which I assume are better obs-based products than L11/CAMS19? Without them, it just gives an impression that model are performing badly compared to the obs (L11/CAMS19). I think they compare better with Wang et al. (2020), and this point should be made clear in these figures.*

We considered your suggestion but decided not to implement it for two reasons. First, we do not have in hand a gridded flux dataset from the G18 DMS concentration data, so including fluxes from W20 and not including fluxes from G18 would have been in our opinion bringing forward W20, which we have no reason for doing. Second, in agreement with previous studies such as that of Tesdal et al. (2016), our work shows that to first order, the global DMS emission depends on global mean DMS concentration, with a significant impact of the chosen flux parameterisation. Conversely, the patterns of DMS concentration are of second importance. Thus, while a detailed comparison of the fields of DMS concentration from

models and climatology is of major interest, it seems that a comparison of the flux fields is slightly redundant.

- *L465: The 4 CMIP6 models differ in the flux parameterisation, so the finding here does not confirm the conclusion of Tesdal et al. (2016) that global emission is roughly linearly dependent upon global mean concentration for “a given flux parameterisation”.*

Thank you for this remark. We modified the original text:

”MIROC-ES2L has an intermediate emission value of 18.4 Tg S year⁻¹ which ranges in the low end of other studies also using the flux parameterisation of Nightingale et al. (2000). However, MIROC-ES2L has the lowest mean DMS concentration among the models, thus confirming the conclusion of Tesdal et al. (2016, Fig. 8) who show that global emission is roughly linearly dependant upon global mean concentration for a given flux parameterisation.”

to: ”MIROC-ES2L has an intermediate emission value of 18.4 Tg S year⁻¹ which ranges in the low end of other studies also using the flux parameterisation of Nightingale et al. (2000). This is consistent with the rather low global mean DMS concentration in MIROC-ES2L (1.77 nM, Table 3), and the finding of Tesdal et al. (2016, Fig. 8) that to first order the global mean concentration of DMS determines the global mean flux.”

- *L546: I recommend two papers from Wang et al. (2018), which incorporates perhaps more DMS producers than the 4 CMIP6 models, including Phaeocystis.*

Wang et al. (2018): Impacts of Shifts in Phytoplankton Community on Clouds and Climate via the Sulfur Cycle, Global Biogeochemical Cycles, 10.1029/2017GB005862

Wang et al. (2018): Influence of dimethyl sulfide on the carbon cycle and biological production, Biogeochemistry, 10.1007/s10533-018-0430-5

Thank you for suggesting these references. The first one is mostly dedicated to an assessment of the radiative effect of the sulfur cycle, and is not closely related to our study. Conversely, the second reference is well suited for the comparison. We added the following sentence at L556:

Wang et al. (2018) performed simulations with the Community Earth System Model (CESM) with RCP8.5 scenario, which computes a global decrease in DMS flux of -8.1 % in 2100, with significant spatial variability. These findings are consistent with those of Kloster et al. (2007), and agree qualitatively with the results obtained with NorESM2-LM despite the projected DMS decrease is only half that found by Wang et al. (2018).

- *Figures 13,14,15: For understanding what each colour represents easily, could you plot a legend in one of the subplots? I know the colours are described in figure caption, but it is easier with a legend.*

We have added a frame with a legend in Figs. 13, 14 and 15

- *L601: I think this paragraph deserves a bit more discussion. The strong relationship between DMS and Chl/NPP is probably true for a given phytoplankton species (and therefore, this relationship holds for in situ observations of a particular phytoplankton bloom or relatively simple-complexity phytoplankton models). However, should this really be the case at global scale where different phytoplankton species dominate in different regions and phytoplankton have a wide range of DMS production rates (i.e. cellular quota; Stefels et al. 2007)? I understand that this point leads to the conclusion in the subsequent paragraph, but I think the reality of the DMS-Chl/NPP relationship is highly variable regionally due to the diversity of phytoplankton species, which should be acknowledged.*

NB: our response below also answers the question raised by Martí Galí in Review #1, General comment #2.

The ambition of our paper, quite specific here and that could appear modest in comparison to existing much more specialised literature, is to broadly assess how some CMIP6 climate models behave in terms of DMS ocean surface concentrations and DMS fluxes, both in the current climate and in the rest of the century.

We are indeed lacking a large-scale observational database that would enable us to draw robust conclusions on the relationship between NPP and DMS concentrations, or emissions, at the scale of global oceans.

We have reworded this part of our text, noting, as you said, that a number of observational studies have highlighted such relationship, at a local scale though, and complementing these local-scale studies with the recent studies of Uhlig et al. (2019) and of Osman et al. (2019) that have been conducted at a basin scale.

However, we think that there are other lines of evidence, other than observations, on the existence of such relationship.

Firstly, previous modelling work of Bopp et al. (2003) and of Kloster et al. (2007) show that the response of the marine biology (i.e., declining NPP) is one of the prominent drivers of changes in DMS emissions. Although the current generation of the PISCES and HAMOCC models derive from previous model versions, key processes have been revised and updated. These changes have implications on model performances and on future projections as reported and documented in Séférian et al. (2020) and Kwiatkowski et al. (2020). In consequence, our work shines light of an emergent property of marine biogeochemical models linking changes in NPP and changes in DMS that is robust across model generations.

Secondly, factorial experiments conducted by Wang et al. (2020) using an artificial neural network show that a 10 % decrease of Chl *a*, a proxy for NPP, leads to a reduction in DMS concentration in large open-ocean domains.

We acknowledge though that a number of studies observed no correlation between DMS and Chl *a*, reflecting the complex mechanisms that control

DMS concentrations and fluxes (e.g., Wang et al., 2020, and references therein).

We thus replaced the text : "Local in situ observations (e.g., Simó et al., 2002; Becagli et al., 2016) have shown positive correlations between NPP and DMSP, and the link between DMSP and DMS concentration has been described in several studies (e.g., Stefels, 2000; Yoch, 2002; Asher et al., 2017; Lizotte et al., 2017). The first group of models (CNRM-ESM2-1, NorESM2-LM and UKESM1-0-LL) thus captures a relationship which is consistent with such ocean field experiments, while the response simulated in MIROC-ES2L is not consistent with the current understanding of the DMSP production pathways by marine phytoplankton (Stefels et al., 2007)."

with

"Although the limited current knowledge about the NPP-DMSP-DMS relationships hampers our ability to constrain this emergent property, several lines of evidence tend to suggest that there is a positive correlation between NPP and DMS concentration. Firstly, noting that some studies observed no correlation between DMS and Chl *a* (e.g., Wang et al., 2020, and references therein), a number of other studies showed positive correlations between NPP and DMS production: the link between NPP and DMSP is highlighted at the local scale (e.g., Simó et al., 2002) and at a basin wide scale (e.g., Uhlig et al., 2019), that between NPP and DMS concentration again at a basin wide scale in Osman et al. (2019), and the link between DMSP and DMS concentration has been described in several studies (e.g., Stefels, 2000; Yoch, 2002; Asher et al., 2017; Lizotte et al., 2017). Secondly, factorial experiments conducted by Wang et al. (2020) using an artificial neural network show that a 10 % decrease of Chl *a* leads to a reduction in DMS concentration in large open-ocean domains. Finally, previous modelling work of Bopp et al. (2003) and of Kloster et al. (2007) show that the response of the marine biology (i.e., declining NPP) is one of the prominent drivers of changes in DMS emissions. The first group of models..."

The framework you describe in Galí and Simó (2015) and that one could apply to these CMIP6 models is largely beyond the scope of our article. Not to mention all the distinct variables involved in your analysis that are not part of the official CMIP6 data request, and thus are not available for a comparable analysis. However, we cite Galí and Simó (2015) in our conclusions as a way forward to progress in DMS climate modelling. The text at L742 now reads:

"Overall, our work shows that there is a major uncertainty in low-latitude ocean where the change in DMS concentration results from the interplay of marine biology factors with many other environmental drivers (e.g., temperature, salinity, stratification, nutrient availability, acidification, large-scale circulation), which and all may affect in both directions the trends

in DMS concentration (Wang et al., 2020). Further analysis to disentangle the role of these factors is required, for instance along the lines of the meta-analysis of Galí and Simó (2015) that specifically addresses the issue of the "summer paradox". This would require important coordination among modellers to work in a multi-model perspective as only a few CMIP6 models include DMS and their DMS-related outputs are limited and insufficient at present to conduct such analysis. In turn, "

- *L622: Briefly state what the conclusions are.*

We left the sentence as is, as the rest of the section largely presents when CMIP6 models agree (or not) with the analysis of Galí et al. (2019).

- *L650: I don't really understand the latter part of this sentence: "the specific role ... are clearly visible." I think this latter part can be deleted, and combine the earlier part with the previous sentence, i.e. "Comparing the time series ... variables, especially when considering ... (dashed lines)."*

We updated the text according to your suggestion.

- *Figure 15: DMS emissions at 100 % are indicated only for two models?*

Thank you for this remark, the 100 % was indeed missing for one model, we updated the Figure.

- *Section 5: I think this section should be named as "Discussion and Conclusions", as it is quite extensive for just Conclusions.*

We prefer to keep the current title for this section, since there is no further discussion of our results.

- *L694: I understand L11 has sampling biases. However, should W20 have similar sampling biases because it also relies on the same dataset (well, twice more) for both training and evaluation (L196-204)? So unless W20 accounts for a preferential sampling of DMS-productive conditions incorporated into the dataset (L189), how can we conclude that W20 does not suffer from similar sampling biases as in L11?*

Two reasons can explain this. First, W20 handles the extreme DMS values differently than L11: while L11 removed data that were above the 99.9 percentile (with the indication that "The 0.1 % eliminated were seawater DMS concentrations greater than 148 nM"), W20 removed "ultralow (< 0.1 nM) and ultrahigh (> 100 nM) DMS measurements". The second reason is L11 and W20 rely on very different methodologies. In L11, the "first guess" field is obtained by extrapolating the averaged available measurements to the entire Longhurst province (see Fig. 1 in Lana et al., 2011). This can lead to important bias if the available measurements are not representative enough of the actual province. Conversely, the ANN developed by W20 uses the actual conditions (SST, SSS, nutrients, etc.) at the measurement location. The values computed by the neural networks account for the variable conditions, and even if the measurements were

carried out in DMS productive area, the ANN is expected to account for this, and compute lower DMS concentrations in low-productivity places. To clarify, we added the following elements in the text:

L171: "... the largest values above the 99.9 percentile are removed (i.e., values above 148 nM)."

L197: "This study relies on the same database of in-situ DMS measurements, which now contains twice as many measurements (over 93k after removing concentrations below 0.1 nM and above 100 nM) as in the study of Lana et al. (2011)."

Regarding the second argument, in line with the remarks and suggestions of Martí Galí in Review #1 (see our answer to his general comment #1), we elaborated and clarified the text about potential biases in L11 as follows:

L185: "Thus, as pointed out by Tesdal et al. (2016), small scale features are transformed into large scale ones by the interpolation procedure, and anomalous values observed at local scale could induce bias when extrapolated across data-sparse regions. This is illustrated by Hayashida et al. (2020), who show that the entire Arctic region in L11 is based on extremely limited data (0–4 % areal coverage north of 60°N). The resulting extrapolation of open water DMS concentration to sea-ice covered areas, where primary production is presumably lower, may lead to a positive bias in L11."

We also rephrased the sentence at L694 to refer to both sampling and extrapolation biases, as follows: "As concluded by previous authors (see for instance Galí et al., 2018, Sect. 4.1), the widely used L11 climatology likely overestimates climatological surface DMS concentration at the spatial resolution of climate models due to the combination of scarce and biased sampling."

- *L719: Briefly state what the conclusions are.*

We rephrased the sentence as follows:

"Our analyses using CMIP6 ESMs confirm the conclusions of Tesdal et al. (2016) that global DMS emission depends primarily on global mean surface ocean DMS concentration, while the spatial distribution of DMS concentration and the parameterisation of ocean-atmosphere exchange coefficient are of secondary importance. Our study further demonstrate that to first order, changes in marine global DMS concentration determine the evolution of the global DMS emission to the atmosphere."

- *L725: I don't think the word "overcome" is appropriate here. Overcome suggests one effect counteracts and defeats another effect. The trend of DMS concentration is neutral (neither increasing/decreasing; Figure 14 bottom panel), so it's just that the positive trend of ice-free extent drives the trend of DMS emission.*

Please mind that Fig. 14 shows only the 1950–2014 period, during which the trends are indeed weak or absent. Conversely, there is a marked pro-

jected trend over the 21th century (ssp585 simulations, see Fig. 10). In the Polar N biome, depending on the model, the trend in DMS concentration is weakly negative (UKESM), weakly positive (CNRM and MIROC) and markedly positive (NorESM) but for all four models, the trend in DMS emission is strongly positive due to the sea-ice retreat. We thus believe that the word "overcome" is appropriate.

- *L750: Data availability for the CMIP6 models should also be mentioned here.*

We added the following sentence: "Datasets from CMIP6 simulations are available from every ESGF node, such as <https://esgf-node.ipsl.upmc.fr/search/cmip6-ipsl/> (last accessed 14 April 2021)."

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