We appreciate the reviewers for their constructive comments and suggestions. Below we respond to each comment; reviewer comments are shown in black, our response is in *red italics*, and revised text is in blue. The line numbers we refer to are within the tracked changes document.

Response to Referee #1

I appreciate the authors for their thorough responses to my previous review. The manuscript has been improved substantially. I am happy to recommend 'accepted subject to technical corrections'. Below are my suggested minor/editorial corrections for Abstract and Conclusions:

We thank the reviewer for their comments. The abstract and conclusions sections have been re-written as per the request from Reviewer 2. Therefore, many of these sentences have been re-phrased or deleted. Where this is the case, I will refer to it as 'changed'.

L1: Replace 'complex and dynamic and driven' by 'complex, dynamic, and driven'

Done

L2: Replace 'leads' by 'lead'

Changed

L4: Remove 'nudged to observations'

Done

L5: Add a few words to describe what has been 'tested' (the sensitivity)

L6 now reads:

We tested the sensitivity of atmospheric DMS to four seawater DMS data sets and three DMS transfer velocity parameterizations.

L12: Replace 'DMS emission' by 'flux parameterization' because emission depends on source (source x flux parameterization)

L14 - This sentence has been changed to:

We find that the choice of oceanic DMS data set has a larger influence on atmospheric DMS than the choice of DMS transfer velocity.

L17: Either remove the sentence 'As a precursor …' or move it to the beginning of the abstract, as it is a background information and not the finding of this study

We have removed this sentence.

L352: Add the year to 'Gali et al'

done

L367: Replace 'B17' by 'Blomquist et al. (2017)' for clarity

done

Response to Referee #2

The revised version of this paper has addressed many of the questions raised by me and the other reviewer and does read better. However, I think the writing can be further improved to tease out the key messages/findings. This is especially important in the title, abstract, and conclusions, but is also relevant to the other sections.

Climatology of seawater DMS concentrations such as Lana et al and Hulswar et al, containing monthly concentration field for each grid cell, have been used historically in many ESMs to drive DMS flux. Doing so requires a choice of the gas transfer velocity (K), which comes with significant uncertainty. The use of satellite Chla (and other ancillary parameters) also allow for a description of the interannual variability in seawater DMS concentration and so flux, which cannot be captured by using a climatology.

The main motivations of this work (according to my reading of the paper, and according to the authors' replier to reviewers) are to see 1) how sensitive are DMS flux and concentrations (in water and in air) to these different options of K and DMS concentration fields, and 2) the impact of interannual variability on atmospheric DMS concentration. These motivations are fine, but the abstract/conclusion do not currently reflect point 2 (to me the key motivation).

Currently the abstract/conclusion really focus on the finding that a linear wind speed dependence in K is superior to a quadratic dependence. While this is a useful message (especially to the earth system modellers), it really isn't a new finding – in situ observations more than a decade ago show that K of DMS and wind speed has a near linear relationship.

The current conclusion also states that by combining satellite Chla with a seawater DMS parameterization (Anderson et al 2001), one captures the spatial/temporal variability in atmospheric/seawater DMS better than using a climatology. To me, it's clear that this approach would provide more interannual variability than a climatology. However, based on the evidence shown it's not obvious that this approach captures the spatial variability 'better' than the climatology. As seen in Fig 2, the seawater DMS concentration field driven by Chla/Anderson is very different from the observation-based climatology. And the limited comparisons with in situ observations do not yield clear cut evidence to me that MODIS-DMS is 'better' or more accurate (but better than MEDUSA? Probably). For example it seems to give a lower mean (in seawater DMS, DMS flux, and atmospheric DMS) than the observation-based climatology. Are the authors arguing that the climatology itself is biased high?

This discrepancy is probably in part due to the decision of using the Anderson parametrization, which is >20 years out of date and does not represent the state-of-the-art. I would've liked to see the authors testing some more recent parametrizations of seawater DMS concentrations. If for whatever reason the authors cannot do so, fine, but then the authors should refrain from statements that suggest MOIDS-DMS captures the mean or spatial variability in seawater DMS better than climatology. They should focus more on the interannual variability component.

I suggest the authors to rewrite the abstract/conclusion with the following components (and revise the paper to reflect/back up these points):

- Sensitivities of DMS concentrations (water/air) and flux to the different parametrizations of K and seawater DMS concentration fields

- Various validations using in situ observations

- Then the key take home messages:

1) That use a linear wind speed dependence in K (based on DMS measurements) is better than using a quadratic dependence. Again, a useful message, but don't labour over it.

2) the seawater DMS concentration field driven by Chla/Anderson is very different from the observation-based climatology. Probably need (to incorporate) more observations to say conclusively which is more accurate

3) interannual variability (a key motivation for their MODIS-DMS approach). The authors seem to suggest currently in the conclusion that it's not very important, but I don't think their estimates capture the entire interannual variability. See detailed comments below.

4) Atmospheric impact: the authors seem to suggest that Nd/CCN/AOD are not very sensitive to the different DMS emissions, but I think Figure 9 shows otherwise. While none of the emission options explains the gaps between simulations and satellite derived Nd/CCN/AOD, among the simulations themselves I think there's quite a range in especially Nd and CCN, which is worth stating.

Finally, they should probably acknowledge that future work should adopt more recent parametrization of seawater DMS concentration.

We thank the reviewer for this comprehensive overview and detailed suggestions on improvements to the conclusion and abstract. To better reflect our key motivation and results within the conclusion and abstract we have focused more on including inter-annual variability of DMS, and the sensitivity of different oceanic DMS data sets and transfer velocities used to atmospheric DMS concentrations. We have also re-written the abstract to fit the ACP requirements of around 250 words and focussed the abstract.

Changes from these specific comments are located in the tracked changes document for:

Lines 396 to 412 for the conclusion and lines 1 to 29 for the abstract.

Specific comments.

Overall, the writing is still not very precise. Please be specific whether the sentences are talking about DMS concentrations (water/air) or flux. And make clear distinctions between DMS flux (~=K * [DMS]seawater) and DMS transfer velocity (K).

We have focused the paper on the main results, especially in the conclusion.

Title. To many, the word 'source' can be synonymous with 'emission'. Here the authors really mean 'surface seawater concentration'

We have changed the term 'source' in the title with 'oceanic DMS concentrations'.

Line 9. 'Same gas transfer velocity', not 'same sea-to-air flux'

done

Line 10. DMS FLUX varies …

Changed from 'DMS varies' to 'DMS emissions vary'.

Line 11. 'different gas transfer velocity', not 'differing sea-to-air flux'

done

Line 12. The phrase 'DMS source' implies a flux to me. But the authors actually mean a different seawater DMS concentration dataset

This has now been changed to oceanic DMS data set

Line 16. 'recently developed transfer velocity parametrizations' is presumably a linear one? The last two sentences have some repetition

We have added 'linear' to this sentence.

Line 17-18 this could be moved to near the top of the abstract

We have removed this sentence as recommended by Reviewer 1

Line 18-20 I find it strange that one of the main messages in the abstract is that the seawater DMS datasets and transfer velocity parametrizations are poorly constrained. Isn't the point of this paper to improve the representations of these processes?

L21 This sentence has now been changed to better reflect that it is poorly constrained within current climate models:

This work highlights that the seawater DMS data sets and transfer velocity parameterizations for DMS currently used in climate models are poorly constrained for the Southern Ocean region.

Line 29-30; 35-37. These descriptions of marine DMS cycling are pretty vague. I suggest for line 29, remove the 'controlled by marine biota statement', and move that to line ~36. More accurately, phytoplankton produces DMSP, and bacteria+ phytoplankton consume DMSP to produce DMS. Thus one expects some correspondence between chlorophyll a and DMS, but not a perfect one.

L41 We have modified to sentence to read as:

Marine biogenic activity, controlled by marine biota, plays a key role in chlorophyll-a (chl-a) production and is considered to be a key driver of oceanic DMS production (e.g. Uhlig et al., 2019; Townsend and Keller, 1996; Anderson et al., 2001; Deppeler and Davidson, 2017).

Line 41-42. Not exactly. Zooplankton graze on phytoplankton, which releases DMSP/DMS. Dacey and Wakeham, 1986

L46 This sentence has been changed to make our point more clear.

The CNRM-ESM2-1 and NorESM2-LM models use a prognostic approach, closely related to zooplankton and dimethylsulfoniopropionate abundance, which is a precursor of oceanic DMS (Seland et al., 2020; Séférian et al., 2019)

Line 46. Also mention Hulswar et al 2022 here

L52 done

Line 54. This seems a good place to introduce the flux equation = K * deltaC

L61 We have added in the equation below:

$$DMS_{flux} = K \times \Delta C = K\,(DMS_w - DMS_a) \qquad (1)$$

ΔC represents the concentration gradient across the air-sea interface where DMSwα is the concentration of DMS in water, and DMSa is the concentration in the air but is negligible as this concentration is substantially smaller than that of seawater.

Line 121. By how much? Be quantitative

L132: We have added to this sentence:

In general, the MODIS-aqua Ocean Color chl-a retrieval underestimates Southern Ocean chlorophyll concentrations by up to 25% (Zeng et al., 2016; Haëntjens et al., 2017; Jena, 2017; Gregg and Casey, 2007; Johnson et al., 2013).

Line 126. Liss and Merlivat is piece-wise linear, to be precise

L137: his has been added to the sentence.

LM86 is a piece-wise linear equation and the default parameterization within UKESM1 (Sellar et al., 2019) and was evaluated in combination with all oceanic DMS data sets.

Line 139. Indicate unit for T

L146 T has already been defined previously on line 136 as:

The Schmidt number represents the viscosity/diffusion properties of a gas, varying with respect to sea surface temperature (T in ◦C).

Eq.4 this is an unusual way of representing Liss and Merlivat 1986, and I'm not sure that they're correct.

Equation for U <3.6 m/s is ok.

For 3.6<U10<13 m/s, it should be (2.85U10-9.65)*(600/Sc)^1/2

For U10>13 m/s, it should be (5.8U10-49.3)* 600/Sc)^1/2

L150 We have now changed the formula in the manuscript to as written above.

Section 2.4.1. please be specific when talking about datasets. Are they datasets of seawater DMS concentration, atmospheric concentration, or both (+fluxes)?

L164 They are just seawater DMS and atmospheric DMS concentrations that we use. We have changed the section title to read:

2.4.1 Oceanic DMS and Atmospheric DMS Datasets

Line 178. 'daily-averaged observations' of CDNC from MODIS?

Daily-averaged CDNC observations derived from MODIS data (Grosvenor et al. 2018; Bennartz & Rausch 2018) were used to calculate CDNC values, based on MODIS satellite data.

Line 180. Here and elsewhere. Phases such as 'we used Choudhury and Tesche (2023)' are too colloquial and inexact. Do the authors mean. E.g. 'we used observations from Choudhury and Tesche (2023)'?

L190 Thank you for the suggestion, this has been changed now to read it as:

Finally, to evaluate cloud condensation nuclei (CCN), we used observations from Choudhury and Tesche (2023) at 818 m, in comparison with simulated CCN at 800 m.

Line 202, 223, 249 etc. Fig number not specified

These have now been fixed

Line 248. 'aligns well' seems generous. MODIS-DMS clearly underestimates relative to observations for the TAN cruise. For SOAP, MODIS-DMS does cover a large range of variability, but its mean is also overestimated.

L258 Based on this comment, we have altered the wording of the sentence:

Having established that oceanic DMS from the MODIS-DMS simulation compares reasonably with summertime observational voyages as seen in Figure 3, 4, we now assess the sensitivity of atmospheric DMS to various sea-to-air transfer functions (Figure 5, A4).

Line 250. How's 'Southern Ocean' defined here?

L31 The definition of the Southern Ocean has been added to the beginning of the introduction.

The representation of aerosols over the Southern Ocean (40 ˚S to 60 ˚S) is a large source of uncertainty in climate models due to the lack of observational data and large seasonal variability (Revell et al., 2019).

Line 253, 266, 268. By 'source', it's more exact to say seawater concentration

Where 'source' has been used, has been changed to 'oceanic DMS data set'.

Line 271. Why are the authors choosing to compare their flux estimates with Webb et al. 2019 observations, which were from the coastal zone? The comparison is bound to be poor.

Line 270-283. I don't find these comparisons very insightful, because 1) these previous observation-based flux estimates probably used different K parameterizations (would be slightly more useful to just compare seawater DMS concentration), and 2) many of these observations seem outside the 10-

year simulation window, so I don't know how the authors are able to make direct comparisons (e.g. how were SST, U10, etc treated?).

We have deleted those paragraphs comparing our simulations with previous observations between lines 270 to 283.

Line 286. References here should really be consistent with line 60.

We have added Yang et al., 2011 so that these references are consistent with line 60.

Line 298. Not clear to me what insights these comparisons offer exactly. That variability is greater near the coast? Or high DMS concentrations are associated with sea ice? Keep in mind due to atmospheric transport and loss, atmospheric DMS and seawater DMS concentrations usually correlate very poorly. Here atmospheric DMS measured/modelled at the coast mostly originated from further upwind.

We believe keeping the comparisons between observations across the literature and our simulations from our manuscript would provide insight into how variable the high latitude Southern Ocean atmospheric DMS can be during the summer. As we compare our simulations with observational stations at these higher latitudes provides useful multi-year variability comparisons to our simulations. We show that most oceanic DMS data sets produce much higher atmospheric DMS concentrations over these latitudes than what has been observed. Additionally, we show that using a transfer velocity derived from DMS observations also has better comparisons to the observations than using the other transfer velocities. It also allows an insight into how the model performs during the summer, when sea ice melt occurs, relative to what has been observed.

Line 311, 318. Fig number?

Fixed

Line 320, 321. If I understand correctly, the authors compared MODIS-DMS vs. a climatology of MODIS-DMS (e.g. ~climatology of Chla). To drive flux, in both sets of simulations the other parameters (wind speed, SST) were allowed to be time-varying and contained interannual variability. If so, this comparison underestimates the total interannual variability in DMS flux. The high r2 (0.92) is surely driven in part by the fact that both sets of simulations used the same wind speed, SST, etc. To estimate the total interannual variability in DMS flux, one should probably compare against a climatology of FLUX driven by DMS-MODIS.

The extra simulation shown in Figure 7 is in response to reviewer 1, which was to evaluate the difference induced by the choice of surface seawater DMS data set. We aimed to retain all parameters (SST, wind, etc) to assess the differences of using a climatology of oceanic DMS vs time-series from this parameterisation. This analysis evaluates different years of biological activity (oceanic DMS blooms) and whether there are statistical differences to DMS in the atmosphere compared with a climatology.

Line 329 the wind?

L351 Changed to:

The oceanic DMS signal in the atmosphere is strong but includes large fluctuations driven by the wind variations.

Line 332. This seems in contrast to results from Revell et al. 2019 (line 30). Not quite sure about the phrase ‘little change’ here. Can authors show panels a c e in Fig. 9 on linear, rather than log scale? By eye there seems to be significant change in at least Nd and CCN. Just because the model (still) severely underestimates Nd and CCN relative to observations (line 339), it doesn’t mean that the DMS emission doesn’t matter. But rather (probably) the model is missing some other aerosol sources in the Southern Ocean.

Also, would be useful to see maps of simulated Nd/CCN/AOD vs. satellite derived observations. Given the fact that climatology and MODIS-DMS yield very different maps of seawater DMS concentrations, can comparisons with maps of Nd/CCN/AOD yield some clues about which is more realistic?

L354 This paragraph has been altered to include the spread from our simulation, consistent with the previous sections in the manuscript. We decided to retain the log y scale as it better captures the entire distribution of all data sets, including the higher values, rather than the linear scale.

We have also added a new figure for the supplementary materials (Figure S6) showing the DJF spatial distribution of all the oceanic DMS data sets and the observations for AOD, CCN, and CDNC.

It now reads:

Figure 9 and Figure S6 show the effect on cloud and aerosol properties of changing the atmospheric DMS distribution. Changing the atmospheric DMS concentration yields a spread across all our simulations for AOD, CDNC, and CCN by 6%, 15%, and 11%, respectively, over the austral summer Southern Ocean. As DMS predominately oxidises into sulfate within the smaller aerosol modes, it has a smaller influence on the AOD than the larger modes from sea-salt aerosol (Mulcahy et al., 2020). However, these smaller aerosols influence cloud seeding as our simulations show. Changes to the oceanic DMS data set increase the spread in simulated CCN and CDNC over the Southern Ocean rather than changing the DMS emissions, which is consistent with our findings for atmospheric DMS concentrations. Altering the oceanic DMS data set produces a 73% greater change in AOD than altering the DMS emissions over the Southern Ocean, emphasizing the role of the ocean in producing atmospheric DMS. Box plots of AOD, CCN, and CDNC (Figure 9e, a, c) show that the simulations do not capture the maxima in CDNC, CCN or AOD over the Southern Ocean.

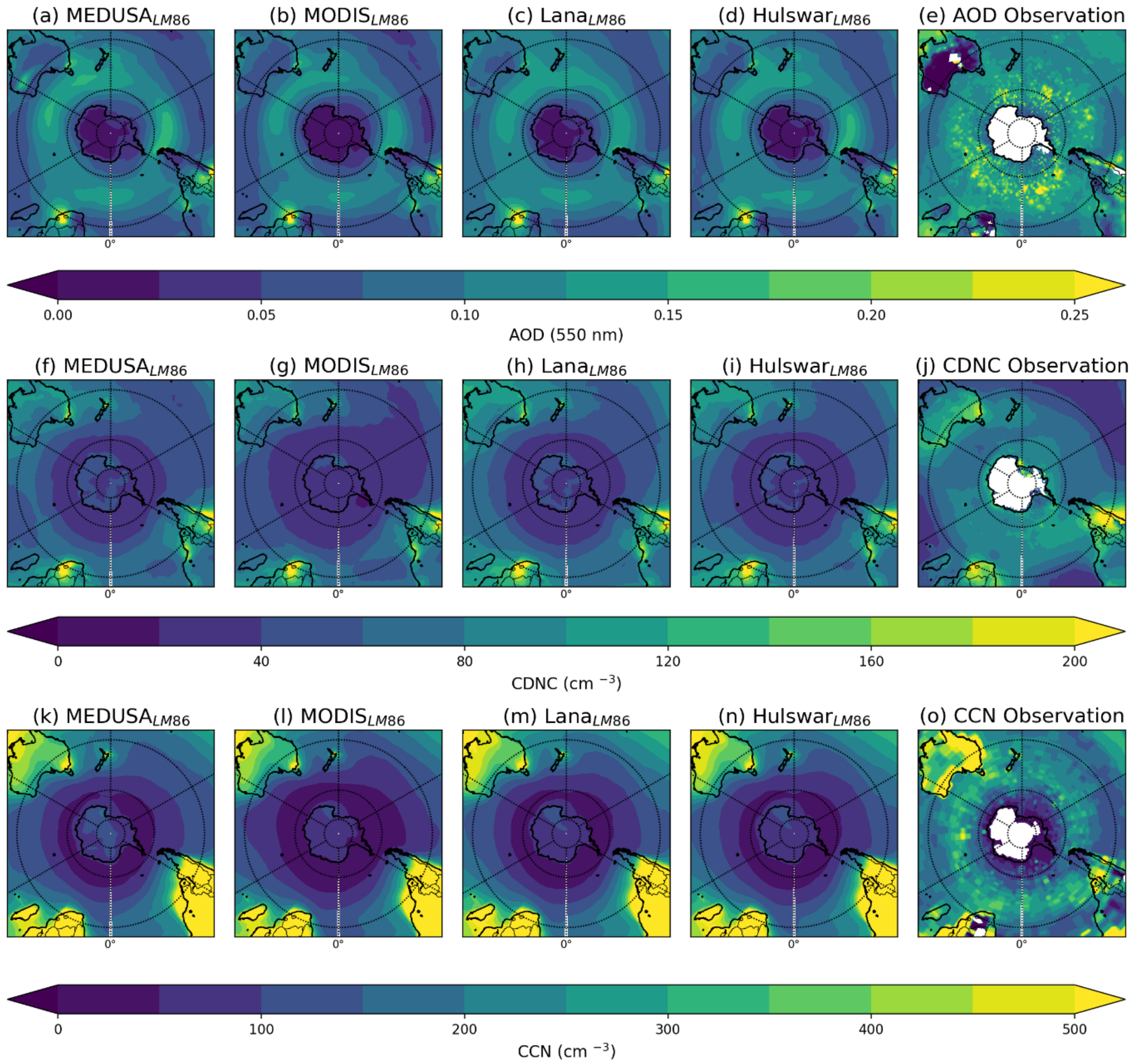

Figure S6. Spatial distribution of each oceanic DMS data set using the Liss and Merlivat (1986) transfer velocity parameterization for (a – d) AOD, (f – i) CDNC, and (k – n) CCN. The observations are from (e) MODIS AOD satellite retrieval, (j) Grosvenor et al. (2018), and (o) the from Choudhury and Tesche (2023).

Line 335. What do authors mean by 'changing the mean DMS emissions'?

We meant changing the DMS emissions, with the 'mean' now removed from the manuscript.

Line 336. I don't understand what this sentence means

L362 This sentence has been changed to:

Altering the oceanic DMS data set produces a 73% greater change in AOD than altering the DMS emissions over the Southern Ocean, emphasizing the role of the ocean in producing atmospheric DMS.

Line 340. Again, I think it's more accurate to use 'seawater DMS concentration' instead of 'source' here, as to many 'source' = emission

Done

Line 343-344. This sentence makes it sound like the MODIS-DMS is the truth. Probably better to just say that MODIS-DMS simulates lower DMS concentration

This sentence has been removed as part of re-writing the conclusion section.

Line 349. If this is the case, why not just use the climatology (e.g. Hulswar)?

Although our analysis suggests that using a climatology instead of time-series for oceanic DMS may not provide differences across large scale, we also show that capturing spatial variability of oceanic DMS is very important. Therefore, careful consideration of the spatial variability from the climatology is needed. We demonstrate the usefulness of using chlorophyll-a satellite retrieval within an oceanic DMS algorithm to capture a high spatial variability within the Southern Ocean. Although we do suggest that using the newly developed climatology may present good comparisons with observations.

Line 352. 'good spatial representation' doesn't seem like the best choice of words, as MODIS-DMS (Fig 2 b) looks very different from the Lana and Hulswar climatology. Do the authors feel that MODIS-DMS give a more realistic representation of the spatial distribution of DMS than the climatology? And if so, what's their evidence?

L384 We have changed this sentence to more appropriately highlight MODIS-DMS:

We show how using chlorophyll-a data from the MODIS-aqua satellites offers an alternative spatial representation of oceanic DMS based on the chlorophyll-a distribution.

Line 353-356. This sentence could be moved to the beginning of conclusion, as justification for this work

This sentence has now been moved to the beginning of conclusion.

Line 357, 359, 361. 'transfer velocity parametrization', not 'flux parametrization''

Done

Line 358. 'transfer velocity parametrization', not 'sea-to-air parametrization'

Done

Line 363. 'transfer velocity parametrization', not 'DMS parametrization'

Done

Line 366. Specify what these atmospheric DMS concentrations are, for the entire southern ocean? During which months/years? Or during the comparison periods?

'During austral summer over the Southern Ocean' has been added to the beginning of the paragraph.

Line 367. Again, be specific with language. 'Transfer velocity parametrization' is more appropriate than 'DMS-specific relationships'

L412 We have changed this sentence to read:

In future, we recommend that models use up-to-date transfer velocity parametrization specific to DMS such as Blomquist et al. (2017).

I can't find the appendix.

We apologise that you couldn’t find the appendix. Although we attached the supplementary materials with the document upon uploading these revisions.