Response to Reviewer 3

We would like to thank this Reviewer for their positive and constructive comments. We have addressed each of them below:

General comments

Section 3.2.3

- *I found Eqn 5 a little difficult to make sense of. The sentences "We note that the wind speed function is used here to represent surface tension of the sea surface microlayer (surface accumulation of organics). Higher wind speeds break this layer up, resulting in fewer organics being lofted into the atmosphere." are useful, but could some extra information be added to explain how the organic mass fraction responds to ocean chlorophyll-a and sea spray particle diameter.*

We have now included the following text about how the particle size and chlorophyll-a modulates the primary marine organic emissions.

Line 202:'Primary marine organic emissions are positively correlated to the seasonal cycle of CHL, acting as a proxy for biological productivity. The organic fraction of SSA is inversely related to the SSA particle size at sub-micron scales (the smaller the particle, the more organic fraction), while at super-micron sizes, the organic fraction is small and relativity constant.'

Figure 1

- *It is useful to see the climatologies plotted, however, it is not easy to see the differences between them. I appreciate that log scales are necessary, but would the authors consider using a more differentiated colour scale. Or perhaps using one of the data sets (maybe Kettle) as the "reference" and plotting the others as differences relative to the reference.*

We have altered to colour scale to try to make the colours less dark. We note that there is substantial literature on the differences between DMS climatologies, hence we have not made this comparison. We have added some more references to this effect in the text.

Line 216: 'Significant literature exists around the production and differences of DMS climatologies and we refer readers to these: (Hulswar et al. 2022, Lana et al. 2011, Zhou et al. 2024). '

Section 3.2.4

- *Could the authors please clarify whether the OM2 DMS simulation used the daily DMS values derived from the OM2 simulations or if a monthly mean of those daily values was used.*

We have rephrased this section to be more clear:

Line 226: 'We highlight that this DMS data set differs from the previous in that it is not a climatology - it is an annually varying dataset with daily resolution (we have changed the model to update DMS daily instead of monthly).'

Section 3.4, L298-300

- *Is there a reference, i.e. another piece of literature (not necessarily part of this work), where the performance of meteorology in the nudged model was assessed?*

We point this reviewer to two papers:

Uhe, P., & Thatcher, M. (2015). A spectral nudging method for the ACCESS1.3 atmospheric model. Geoscientific Model Development, 8(6), 1645–1658. https://doi.org/10.5194/GMD-8-1645-2015

Telford, P. J., Braesicke, P., Morgenstern, O., & Pyle, J. A. (2008). Atmospheric Chemistry and Physics Technical Note: Description and assessment of a nudged version of the new dynamics Unified Model. Atmos. Chem. Phys, 8, 1701–1712. www.atmos-chem-phys.net/8/1701/2008/

We have now included this in the text:

Line 360: '...given the model is nudged to ERA5, we expect the large scale flow to be accurate (Uhe & Thatcher 2015, Telford et al. 2008).'

Section 3.4.1, L319-322

- *"We also recognise that we have not performed a similar baseline filtering to the model (in part due to lack of radon in the model), but have applied the same baseline filtering to the model as what was developed for the observations." The above is not clear to me. I don't follow what the baseline filtering is, or the role Radon is playing.*

We apologise for the confusion here. We have now more clearly defined what is meant by 'baseline' and how radon is considered in defining this in Section 3.3.4 and referenced this in Section 3.4.1.

Line 210: 'The data presented here is the baseline filtered data (as described in Gras & Keywood, 2017). Baseline air is considered the worlds cleanest air, as unaltered by human activity as physically possible. At KCG, baseline air is identified as air that has come from the Southern Ocean where the wind direction was between 190 and 280 ° and the radon concentration (a marker of terrestrial influence) is below 100 mBq.'

Line 357: 'We also recognise that at KCG we have not performed a similar baseline filtering to the model data, in part due to lack of radon in the model (see Section 3.3.4 for details). Instead we we have matched the model data to the available daily mean baseline-filtered observations. '

Figures 3, 4, 5

- *Is there a reason for the offset horizontal lines in the subfigures? For example, in Figure 3a annual N10 is shown as horizontally offset coloured lines for the model experiments on the right hand side of the plot. Similarly in Figures d-g the lines representing seasonal mean N10 for the different model experiments are horizontally offset.*

Yes, we offset them just to make identifying different lines slightly easier, as some of them are very close together.

Section 4.1, L342

- *I don't think it's clear from the available data that the model mis-represents the seasonal minima at MI in May. The winter minimum N10 at MI appears to be more variable than at kennaook and Syowa and the model does not capture this. However, due to limited number of years of observations and model grid cell to point observation comparison, I think that it is difficult to conclude that the model mis-represents a seasonal minimum at MI in May.*

Yes, this is a good point. We have now altered the text as follows:

Line 387: 'The model's seasonal cycle is flat compared to the observations, indicating both missing sources of aerosol and missing seasonal processes. The control run does not capture the seasonal minima, which in the observations is shown in May after a steep decline through autumn, whilst for the model is shown in June (Figure 3b). Note that there are limited observations (only two seasonal cycles) and that there is greater observed variability (as shown by the shading) during winter. '

Section 4.1, L345

- *I'd argue that the model and observations both show less variation in winter. The values (for N10 etc) are smaller in winter compared with summer for both model and observations, so the variation as a fractional or percentage might better show if the variation was really much smaller in the observations.*

Yes, the text reads: 'The model again shows little variance in the winter periods, with larger variance in the summer.' which we think is in agreement with the Reviewers comment.

Section 4.1, L349

- *I agree that at Syowa the model does seem to simulate the minima too late compared with the obs. However, the model minima looks to extend from Jun-Aug, while the observed minima looks to be June (rather than May as stated).*

We have corrected this.

Line 394: 'Syowa has a minimum in June that is not captured by the model, which simulates the minima in August, although is generally low from May-August.'

Section 4.1, L365-367

- *Does ACCESS use VOC emissions ancillary files or calculate VOC emissions online? Can the authors say if there were there large VOC emissions over the first grid box? If there weren't large land-based VOC emissions over the first grid box, this might strengthen the argument that there are issues with the marine biogenics. Although I take the author's point that shifting the grid box did not help.*

ACCESS does use ancillary files for terrestrial VOC emissions (monoterpenes) (as per the CMIP protocol). The emissions over the grid box that KCG is located are small (by orders of magnitude) compared to other densely forested regions (e.g. the tropical and subtropical forests) however, they are not zero. In the text we have now re-iterated that we have attempted to filter out the days in which the terrestrial airmasses are experienced at KCG, but noting that removing this terrestrial influence all together from the model is difficult. We have ambitions to study these artefacts more fully with dedicated simulations beyond what we can accomplish with the existing simulations. We note that this section has changed somewhat due to other Reviewer comments.

Line 410: 'This large increase in small-sized aerosol maybe a result of several factors, including the relative simplicity of the GLOMAP-mode BL NPF scheme (a binary scheme outlined in Section 3.2.1), the influence of terrestrial airmasses (which contain emissions of VOCs that mediate the BL NPF, despite our efforts to filter these influences out) or aerosol pre-cursors. A more complex NPF scheme, such as those discussed in the Introduction may yield more realistic results, while greater investigation into the observed and modelled aerosol and aerosol precursors is called for in the region.'

Section 4.1, L380-384

- *Plots of aerosol size distribution would help diagnose how the model (re)distributes aerosol in the simulations.*

Yes we agree on this front. We have chosen not to show these results for two reasons. Firstly that we have only a few observed size distributions currently available to compare to, and second, our team is carrying out a body of work to process, quality control and harmonise 10+ years of observed size distributions, including for all the voyages used in this work. We are planning a significant body of work once these observations are published.

Section 4.2, L423

- *"...which could indeed be driven by sea spray, long range transport of aerosol" =¿ Add "or" before "long range transport of aerosol"*

Done

Section 4.2, L430-437

- *I agree that turning on the BL NPF has a small effect, but also worth noting that it's the only experiment that reduces CCN in MI and Syowa.*

The BL NPF simulation increases aerosol very marginally at MI and Syowa (the Reviewer may have gotten the lines mixed up (light red vs dark red, which we hope we have now rectified with clearer plots).

Section 4.2, L438-437

- *The contours in Figs 6-8 are quite hard to see. Could the authors consider changing the colour of these? Something like cyan might stand out more.*

We have tested out a range of colour combinations and the results are quite visually jarring. For this reason, we have retained the current colour ways, but have made the contour lines thicker.

References

- *Please update Bhatti et al., 2023 to the final version of the accepted maniscript.*

Done

Other points

- I agree that missing sources and mis-represented microphysics (probably aerosol and cloud) are large contributors to bias in cloud and RF over the Southern Ocean. Is there any reason to suspect loss processes (deposition) might be overestimated? Can the authors comment on how 'missing' marine VOC sources of VOCs (and possibly secondary organic aerosol, e.g. https://doi.org/10.1016/j.scitotenv.2021.145054) could affect clouds and climate over the Southern Ocean?

Yes, we agree with the Reviewer in that loss processes should also be examined. We have added some text around this in the Introduction.

Line 91: 'Aerosol sinks, and how they are modelled, are also a key source of uncertainty. Aerosol can be removed from the atmosphere via dry deposition or wet deposition. Dry deposition is difficult to measure and evaluate, however Regayre et al. (2020), after applying Southern Ocean observational constraints to a perturbed parameter ensemble, find that it is likely that a scaling factor for the accumulation mode dry deposition velocity in the Unified Model needs to be lower than the default value. This would result in a reduced sink of aerosol. Other observational studies have indicated that wet deposition (rain after coalescence of cloud droplets) is an important control of CCN variability in the Southern Ocean, particularly in relation to shallow convection (Alinejadtabrizi et al. 2024) and stratocumulus (Kang et al. 2024). Given the tendency for models to produce too much light rain (Stephens et al. 2010), it has been suggested that wet deposition may be overestimated in models (Kang et al. 2024). '

We have also added some comments about how marine VOCs may influence the Southern Ocean in our introduction:

Line 85: 'Additionally, marine volatile organic compounds (VOCs), such as isoprene, can reduce the atmospheric oxidative capacity by reacting with OH (as well as O_3 and NO_3 to a lesser degree) in the troposphere. Such VOCs, can also yield secondary organic aerosol and provide condensational mass, further influencing the clouds and climate. In the case of marine isoprene, this occurs on a much smaller scale than that of DMS (Yu et al. 2021), with isoprene concentrations being very low outside of phytoplankton blooms and biologically active coastal regions of the Southern Ocean Ferracci et al. (2021).'

- To help visualise the percentage changes in N10 etc across the model experiments the authors could consider including a matrix/table of percentage change colour coded to show an increase or decrease for the parameter (e.g. N10).

We appreciate the Reviewers suggestion here and have considered this carefully. Tables or matrixes that encapsulate all of our results are, bluntly, enormous. For this reason, we have pointed interested readers to our github page where this information can be found, but have not included it specifically here.

Line 367: 'Quantitative summaries of all our results can be found in the published code linked to this paper (see Code and Data Availability).':

Technical comments

- *Introduction, L22:*
- *Change "Aerosol affect. . . ." to "Aerosol affects. . . . ."*

Updated

- *Section 3.1.1, L141; Section 3.4.1, Line 310:*
- *A grammatical point, but I felt that "inline" should be "in line" in the text.*

We have updated all instances of this to be 'in line' with your suggestion

- *Section 3.3.1, L221: Not sure what 'underway' means here.*

We have defined 'underway' as 'automatic observations taken continuously while the ship is operating' and included this in the text

- *Section 3.3.3, L221: Change CO2 to CO2*

Done

- *Section 3.3.4, Heading and L265 (and Figure 3, 4, 5): I'm guessing that "kennaook" does use a lower case k, but in that case the legend in Fig 2 is the odd one out.*

We have updated these instances to be consistent.

- *Figure 3, 4, captions: "The monthly and annual median concentrations of N10 for at..." - Delete "for" or "at"*

Fixed

- *Fig 3-5, Captions L2: "For all, the 25th and 75th percentiles..." - Suggest adding "For all subfigures..." for clarity.*

Changed

- *Section 4.1, L360: I suggest a new sub-section here to report model simulations.*

We have added this for each of the Results (section 4) subsections.

- *Section 4.1: "For DJF, the BL NPF simulation is now overestimates the observations by 33%" - Please correct this.*

Fixed

- *Section 5, L518: Three best simulations rather than 4?*

Fixed

- *Section 5, Figure 7: Units on colour bar are g m-2. Units in caption are kg m-2.*

Fixed

- *Section 7, L639: Please change earth to Earth*

This sentence has been removed.