

Interactive comment on “Sensitivity of cloud condensation nuclei to regional changes in dimethyl-sulphide emissions” by M. T. Woodhouse et al.

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Author’s response to reviewer 1.

We thank the reviewer for the time taken to review our manuscript. We appreciate the comments, and have used them to improve the manuscript.

Specific points are addressed below, reviewer’s comments in bold.

Please be more specific regarding the definition of sea-salt vs. sea spray. To my knowledge Gong (2003) parameterization considers "sea-salt", not sea spray. If sea spray was used in the model, please explain the mechanism for sea-to-air

transport of surface-active organic material of biogenic origin (e.g., de Leeuw et al., 2011). If not, please replace "sea spray" by "sea-salt" in the text.

We represent sea-salt (as sodium chloride) and not sea-spray in the model. We have updated the manuscript accordingly.

Also indicate, if possible, how the consideration of primary and secondary organic aerosol of marine origin may have influenced the results of the current study.

Our simulations don't include a marine organic matter source (primary or secondary). We find that DMS produces CCN mainly through nucleation in the free troposphere (consistent with Korhonen et al., 2008). Consequently, although it is difficult to speculate on the impact of marine organic aerosol sources on our results, we do not expect our results to change significantly. We also note that Spracklen et al. (2008, GRL 35: L12811) found that a flux of 8 Tg a⁻¹ of organic material (primary and secondary combined) from the oceans explains coastal OC observations. Contrast the 8 Tg a⁻¹ of marine OC emissions with the total sea-salt emission (on the order of 8,000 Tg a⁻¹) and it is unlikely that neglecting the marine OC component will affect the findings of our study.

There seem to be further inconsistencies regarding the submicron sea-spray. On Pg. 27400, Ln. 10 text reads: "Sea-spray emissions are calculated online in the model using the Gong (2003) parameterisation between 0.035 and 30.0 μm dry radius". However, on Pg. 27403, Ln. 25 it is stated that "The model simulations here do not include emissions of sub-micron sea-spray." Please explain.

Our simulations use the Gong (2003) parameterization to calculate bin-resolved emissions of sea-salt between 0.035 and 30.0 μm dry radius. These emissions are then applied to the accumulation mode (bins $<1\mu\text{m}$) and coarse mode (bins $>1\mu\text{m}$). We agree that the wording is confusing, and have omitted the line 'The model simulations here do not include emissions of sub-micron sea-spray' from the manuscript.

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The current study often compares findings to that of Woodhouse et al. (2010). The June and December combined hemispheric mean (I assume authors meant CCN) in current study is 80 vs. 63 in Woodhouse et al. (2010), mean summer and winter hemisphere CCN sensitivities of 75 and 82 vs. 47 and 78 Woodhouse et al. (2010). If these differences are significant, please explain the causes for the discrepancies in the two model studies. The reader should not be expected to know Woodhouse et al. (2010) study in order to understand the current one.

As noted in the introduction, the current study builds naturally on the results of Woodhouse et al. (2010), where we note a weak response in CCN to changes in DMS flux. The variation in CCN sensitivities between the two studies is small (already noted in the conclusions), but explained by the different form of the DMS perturbations introduced (global changes in Woodhouse et al., 2010, vs. discrete patches in the present study). The sentence in question now reads ‘The June and December combined hemispheric mean CCN sensitivity is 80..., slightly higher but comparable to the 63... mean calculated in Woodhouse et al. (2010)’.

Pg. 27400, Ln. 11. Discussion regarding the minor effect of dust by referencing the study of Manktelow et al. (2010) is misleading. Manktelow et al. (2010) used low uptake coefficient of SO₂ on dust because of a "strong calcium component" of the Asian dust. The same uptake coefficient may not be used for all chemical composition of dust derived from different source regions (see references in Manktelow et al. (2010)). Furthermore, due to high sulfate concentration downwind from East Asia Manktelow et al. (2010) assumed that all SO₂ molecules adsorbed onto dust produce sulfate and that surface saturation occurs once the dust is coated in a mono-molecular sulfate layer. Will the same be true over the Southern Ocean? If yes, that means that significant fraction of e.g., South Australian and Patagonian dust will be coated by DMS-derived SO₂, and neglecting the dust effects may lead to the considerable uncertainties.

The Manktelow et al. (2010) study considers an extreme dust event from a major dust

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source region. Dust concentrations (and in particular, dust surface area) in the southern hemisphere are not likely to reach the levels noted in Manktelow et al. (2010). We therefore suggest that even if a higher uptake coefficient was used, the impact in the Southern Ocean would be minimal. We also note that Lee et al. (2009) used a global aerosol microphysics model to quantify the effect of dust on CCN, finding negligible effects in almost all regions except the main dust outflow region from North Africa, where 10-20% decreases in CCN concentration were found (due to the additional condensation sink from the dust). We have included the Lee et al. (2009) reference in the manuscript.

One of the main contributions of the current study is to elucidate the importance of variability in phytoplankton abundance/speciation and wind speed on marine boundary layer CCN budget. In that view I would like to see more discussion regarding the importance of the variability in DMS concentrations (i.e., +0.5, +1.0, +2.0, +5.0 nM, etc). How realistic is the examined increase in DMS concentration? What is a probable range in DMS concentration due to variability in phytoplankton speciation and abundance?

The context of the specified patch changes is a valuable addition to the manuscript. The following text has been added to the manuscript:

‘Surface ocean DMS concentrations are highly variable with season and location. Concentrations range from < 0.1 to > 50.0 nM, the highest concentrations occurring in the high-latitude oceans during summer. Annual mean DMS concentrations from the Kettle & Andreae (2000) observational climatology are shown in Fig. [New figure]. Typical annual mean sea-surface DMS concentrations are on the order of a few nanomols. Sea-surface DMS concentrations have been observed to increase by up to 8.5 nM in response to iron addition experiments (Boyd et al., 2007), but such changes are short-lived and affect only a small area. Vallina et al. (2007) found that sea-surface DMS concentrations increased locally by up to 0.5 nM in response to a global warming scenario with a marine ecosystem model.’

We also now show the annual mean DMS concentration as specified in Kettle & Andreae (2000) in a new figure.

Pg. 27396, Ln. 18. Please explain what you mean by "changes in phytoplankton". Is it speciation, abundance or something else?

We don't limit our conclusions to any single change in the phytoplankton population. Any change in the characteristics of the phytoplankton population (speciation, distribution, abundance) has the potential to affect the DMS flux and thereby CCN abundance. A more complete discussion of potential changes to the phytoplankton population is beyond this manuscript.

Pg. 27397, Ln.12. Please be more specific when discussing "The direction" of the CLAW feedback. Is this about the absolute value of the climate forcing?

We refer to whether the CLAW feedback is positive (reinforces the initial change) or negative (in opposition to the initial change). The sentence now reads 'The direction (positive or negative) of the CLAW feedback is not certain however'.

Pg. 27401, Ln. 1. I believe the statement "Merikanto et al. (2009) showed that binary nucleation accounts for ~ 90% of CCN in the marine boundary layer" is inaccurate. Merikanto et al. (2009) showed that in the marine boundary layer 55% of CCN (0.2%) were from nucleation, with 45% entrained from the free troposphere and 10% nucleated directly in the boundary layer.

The sentence as originally written was inaccurate. The text has been updated with the addition of the words 'and primary emissions': 'Since Merikanto et al., (2009) showed that binary nucleation and primary emissions account for ~ 90 % of CCN in the marine boundary layer...'

Please use negative 1:1 line on Figs. 6 and 7 for negative r values.

Figures 6 and 7 have been updated.

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