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Atmospheric
Chemistry
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Discussions

Interactive comment on "Low sensitivity of cloud condensation nuclei to changes in the sea-air flux of dimethyl-sulphide" *by* M. T. Woodhouse et al.

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The authors thank the reviewer, Roland von Glasow, for a thorough and thought provoking review. Specific points are addressed below, reviewer's comments in bold.

The authors ignore to mention that DMS is also oxidised by BrO, Cl and O3 (in cloud water) which can be very relevant as even concluded in a study by one of the co-authors, Boucher et al., ACP, 2003. I think this should be added in a few model runs or at least discussed in some detail as neglecting these reactions introduces further uncertainties as the reaction products are different for these pathways and often don't lead to the formation of new particles but rather to the growth of existing particles (see e.g., von Glasow, Env. Chem., 2007). It is important to point out that H2SO4 is the only sulphur species that is involved in

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the formation of new particles.

In ongoing work with a coupled chemistry version of GLOMAP (with BrO), the response of CCN to DMS is similar to that described here. Breider et al. (in prep.) calculate a global annual mean relative sensitivity of 0.08, compared to 0.05 here. We are not able to comment on the role of Cl in DMS oxidation.

Additionally, the sources and chemistry, and therefore the role of BrO and Cl, are poorly understood on the global scale and subject to quite large errors.

We suggest that the role of O_3 in aqueous phase DMS oxidation is small, as most DMS oxidation occurs in the cloud free troposphere.

p. 3722, l. 15 - 20: The use of offline oxidants for DMS and SO2 obviously comes with the limitation that any reduction in the concentration of these oxidants by DMS and SO2 is ignored and hence the oxidation is potentially overestimated. I would expect a comment on this. Is this a problem for this study (esp. for NO3) and what regions are especially prone to this?

The lifetime of DMS with the coupled model is approximately double that of the offline model, when BrO is neglected in both. However when BrO is included in the coupled model, the lifetime is significantly decreased. This is ongoing work by another member of the GLOMAP group. NO_3 appears to be the oxidant with the biggest error in the offline model, especially in the SH.

This is now highlighted in the manuscript. It is difficult to speculate exactly how large an error the offline oxidants introduce to the study. However, experiments perturbing the flux of DMS in the coupled model produce similar results to the offline model, giving us confidence.

p. 3723, l. 14 - 21: As all scenarios are described in Table 1 and not only CLIM 2 and CLIM3 I would start this section with something like: "All scenarios are listed in Table 1, here we give more details for each of these scenarios."

Agreed, text modified.

p. 3725, l. 5 - 10: I think the "global warming scenario" should be described in more detail at this point. What is the relevant temperature increase and how are the DMS fields linked to the rise in atmospheric CO_2?

The effect on climate and temperature of the CO_2 increase is poorly described in the two relevant references, we are therefore not able to quote a relevant temperature increase. Differences in atmospheric factors (e.g. air temperature, winds) from the global warming scenario are applied to ocean model forcing fields, driving the globally warmed scenario.

The manuscript has been updated to include more detail, though a more complete description is not possible.

p. 3728, l. 8 - 12: I assume it is unavoidable to use different years for the meteorology driving the model and the years of which you have DMS data from these locations but it would be good to briefly discuss the limitations of this comparison.

The comparisons are made to multi-year observations, the range being shown by the error bars in Fig. 5. Interannual variation in DMS fluxes resulting from wind speed variability is discussed later in the paper. This is now referred to in the manuscript in the atmospheric DMS observations section, and the interannual variability in atmospheric DMS concentration is also highlighted.

p. 3732, l. 3 - 15: Doesn't the lack of presence of sub-mircon sea salt particles in your model limit the extent to which you can draw conclusions? If I remember correctly, previous results from your group have shown some interesting nonlinearities plus the absolute number of CCN would be affected as well. Please discuss whether/how the lack of small sea salt particles limits the applicability of your model study to the overall question of this paper. Also: could you please

show a figure that shows the sensitivities that you discuss in this paragraph?

We include the contribution from ultrafine sea-salt from the Mårtensson scheme because the contribution these emissions make when calculating relative CCN differences are important over some regions. Merikanto et al. (2009, Impact of nucleation on global CCN, ACP 9, 21: 8601) show that the flux of aerosol number into the marine boundary layer from the free troposphere is not affected by primary particles emitted at the surface. Additionally, Korhonen et al. (2008) show that the free troposphere is the main source of DMS-derived CCN. This suggests that the free-tropospheric aerosol and sea spray are effectively decoupled, and that it is acceptable to include the ultrafine sea spray non-interactively as we have done.

We have added words to this effect to the manuscript.

A figure showing relative sensitivities is now also included.

Figure 2 and 3: I think these figures could be merged.

A possibility, we will review this during any further typesetting of the manuscript.

Fig 4, caption: What do you mean by "land is not included in the meaning."?

When calculating global and hemispheric mean differences, the land area is not considered. The differences over land would be zero, and would reduce the calculated mean. What we are interested in is the mean change in DMS emissions from the ocean.

By 'meaning' we meant 'averaging'.

The caption has been updated to clarify this.

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