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Atmospheric Chemistry and Physics 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, Jeff Pierce, for the time taken to undertake a very thorough and clear review. Specific points are addressed below, reviewer's comments in bold.

**1. p 3719, l12: "... results in the production of SO2, MSA and H2SO4. These can condense on existing aerosol particles or nucleate to form new H2SO4 particles." SO2 doesn't really condense on existing aerosol partitions (it may equilibrium partition into haze particles) and isn't involved with nucleation. MSA condenses to aerosols, but I haven't heard of it being involved in nucleation (I could be wrong about this though).**



Sentence clarified to read 'Sulphuric acid (H2SO4) can condense on existing aerosol particles...'

**2. P3722, l20: The competition between condensational growth and coagulational scavenging should be a primary factor in the sensitivity of CCN concentrations to DMS fluxes. Generally, high spectral resolution bin models (e.g. Spracklin et al., 2005) are thought to be the best for simulating the numerics of this. For this reason, I am curious as to why a modal scheme was used. On the other hand, since global microphysics models all differ in their predictions of the condensation/coagulation sinks and condensation rates, issues of the numerical schemes may only be of 2nd-order importance.**

**Still, I would like to see a short discussion (a couple sentences) on how well the modal microphysics scheme predictions growth/loss of nucleated particles compared to the sectional model (even if this is explored in the Mann et al. (2010)).**

Multiple years of simulations were required in this study, and the sectional version of the GLOMAP model (Spracklen et al., 2005) would have been too computationally expensive to run. The modal scheme is well suited to the study however, as it contains the same aerosol microphysics (including competition between condensation and coagulation) as the sectional scheme.

Comparisons (Graham Mann, in prep.) suggest that the modal and sectional versions of GLOMAP compare very well. Further work is also being undertaken to test the responses of both schemes to specific perturbations. Again, significant differences in response of the two schemes are not expected.

A sentence has been added to the paper to highlight this.

**3. What nucleation scheme(s) were used. Because nucleation/growth is so important to the sensitivity, this should be included.**

Kulmala et al., 1998. Now included in text.

**4. P3722, l24: Dust is ignored. Since dust can dominate aerosol surface area in much of the subtropical regions, I wonder if the large sensitivity of CCN concentrations to DMS in these regions (e.g. Figure 6b) would be reduced because of this. I would guess that much of the H2SO4 could condense onto the dust. A paper with 2 common authors with this paper looked at this recently (Manktelow, P.T. et al., The impact of dust on sulfate aerosol, CN and CCN during an East Asian dust storm, ACP 2010.), so I assume you have better insight into this than I do.**

Manktelow et al. (2010) find that 'fine sulphate' (diameter <1.3 $\mu$m) decreases by only $\sim$10% (in the middle of a large dust storm) when dust is included in the model, suggesting that sulphate CCN formation is not strongly influenced by dust. There are changes in the division of mass between the nucleation and Aitken, and coarser modes however.

Coarse sulphate (>1.0 $\mu$m) is affected more greatly when dust is included, though this aerosol size range makes a small contribution to CCN. We therefore believe that neglecting dust in the present simulations will not significantly affect the calculated CCN number concentrations and sensitivities.

We now include a sentence in the model description highlighting the Manktelow et al., 2010 findings.

**5. P3724, l20: CLIM6 used PlankTOM5 too.**

'CLIM6' now included in parentheses in text.

**6. Top of P3727 and Section 3.6: I am curious to know if it is it likely that the choice of sea-air flux parameterization would greatly effect the sensitivity of CCN to future DMS changes. Is the DMS flux linearly proportional to the oceanic DMS concentrations in all parameterizations? It makes sense that this would be the case, but if it isn't true for all cases, that may cause bigger differences.**



The DMS flux is linearly proportional to the oceanic DMS concentrations, for all parameterisations. The different flux parameterisations differ most at high wind speeds. If a large DMS change was co-located with an area of high wind speed, the CCN change could be greater in one parameterisation than another. This does not affect the calculated sensitivity however.

**7. P3727 l7-21 and Figure 4: There seems to be inconsistencies between the text and figure. Line 11, Ann DMS NH, CLIM6 is not the only scheme with a lower flux and than CLIM1. CLIM2 and CLIM3 do too. Line 18, December DMS NH. CLIM3 and CLIM6 have lower fluxes than CLIM1. I suggest going carefully through this section and making sure its consistent with Figure 4.**

This section caused confusion, and has been significantly rewritten. Additionally, the relative differences have been recalculated, and the text and figures are now consistent. The recalculated relative differences result in a new relative sensitivity of 0.05 (changed from 0.11). This updated value does not affect our conclusions. The manuscript has been updated where required to take into account the revised numbers.

**8. Section 3.2 and Figure 5: There is much discussion in the text about CLIM4 results having a high bias against observations. However, in Figure 5, it is the CLIM3 line that has the high bias, not CLIM4. It makes sense that CLIM4 is the case that should have the high bias. I believe that the legend in Figure 5 is incorrect. Also, the line-styles (e.g. the length of dashes) in the legend don't exactly match what is in the figure.**

The legend has been corrected; linestyles match.

**9. Figure 6: I would find it useful if a 3rd figure panel that showed the fractional contribution of DMS to CCN was shown too.**

Figure 6 has been updated so that absolute and fractional contribution of DMS to CCN is now shown.

**10. P3730, l8: Can you please discuss briefly the physical reason why CCN concentrations increase when DMS emissions are turned off in Korhonen? This has implications to why you get negative absolute sensitivities of CCN to DMS changes in your study later. I can see two possible reasons for this inverse CLAW, but may be missing something:**

**a) DMS emissions allow for nucleation to occur (or occur more quickly) causing a nucleation mode to appear and compete for condensate with the UF sea salt. Under some circumstances, this added competition could reduce the total number of particles that actually grow to CCN sizes. However, the DMS emissions also contribute extra condensational growth material (H2SO4), so this seems unlikely.**

**b) This is a possible consequence of model configuration, which may or may not be an issue in your model. This increase of CCN due to turning off of DMS could occur if the size cutoff for in-cloud wet scavenging in the model is larger than the size cutoff for CCN used here. Lets say that your in-cloud wet scavenging cutoff is at 50 nm, but your CCN cutoff is at 35 nm. The particles between 35 and 50 nm would count as CCN, but would also have a very long lifetime since they are large enough that coagulational scavenging is not very fast but not large enough to be subject to incloud wet scavenging. When DMS emissions are present, the DMS-derived H2SO4 is present to grow these particles to 50 nm where they quickly rain out of the atmosphere. In the absence of DMS, these particles may be stuck much longer in the 35-50 nm size range and contribute to high CCN concentrations without risk of in-cloud scavenging. If this is true for your model setup, this is somewhat of an inconsistency because we are calling particles CCN, but not allowing them to wet scavenge.**

**There may be other reasons for the increase in CCN when DMS is turned off, so please add discussion of this to the paper.**

Korhonen et al. (2008) term this the 'inverse-CLAW' effect, and suggest the following



to explain it. DMS emissions are very low during winter, the input of nucleated aerosol from the free troposphere was shown to be quite weak. However, $SO_2$ in the boundary layer can be taken up by cloud droplets and oxidized to sulphate. This increases the aerosol size, potentially making them large enough to be nucleation scavenged (which in the model affects aerosol with a wet radius larger than 100 nm).

In GLOMAP we do indeed assume that the wet scavenging radius is larger than the CCN activation radius. This is physically plausible because precipitation tends to favour the large tail of the drop distribution. So when DMS is included and aerosol particles grow more in the BL the net effect can be more scavenging and a negative CLAW effect (negative dCCN/dDMS) unless the DMS causes additional (nucleated) particles to be entrained from the FT. Because the FT source is sometimes weak (especially in winter as shown by Korhonen et al., 2008), it is possible to have a net negative CLAW effect.

**11. Section 3.5 and Figure 4: Please discuss why you have negative absolute sensitivities in some cases (see my point above).**

Three of the four negative absolute sensitivities in Fig. 4 are due to decreases in DMS flux and associated CCN increases. The efficiency of conversion of DMS to CCN is not expected to be the same for all locations (this is under investigation in a seperate study). This, combined with the spatial inhomogeneity of DMS flux changes (Fig. 3), can lead to a hemispheric mean decrease in DMS emissions, but corresponding increase in CCN number concentration (or vice versa).

For example, if a large decrease in DMS flux coincides with an area where DMS to CCN conversion is quite inefficient, there will be only a small decrease in CCN number concentration. On the other hand, consider a small increase in DMS flux, but a strong response in CCN; this leads to an increase in CCN number concentration. The global mean difference for DMS flux could therefore be negative, but if the increases in DMS flux were coincident with an area that efficiently produces CCN from DMS, the global mean CCN difference could be positive. This combination gives a negative CCN

sensitivity, when considered hemispherically or globally.

It should be noted that the negative sensitivities only occur where the DMS flux difference is small.

The manuscript now includes a discussion of this.

**12. Figure 4: It is interesting that the sensitivity CCN to DMS in CLIM3 is strongly negative in the SH annually, but positive in the SH both in the summer and winter. There must be something very different happening in the autumn and spring.**

The negative annual mean sensitivity of CLIM3 in the SH (Fig. 4) is the result of a small negative change in DMS flux leading to a moderate change in CCN number concentration.

The CLIM3 SH mean difference in DMS flux is low positive in June, and moderately negative in December. It is possible to directly compare the DMS flux and CCN number concentration changes between different time and spatial domains in Fig. 4, but comparing the calculated CCN sensitivities in this way is not as straightforward. In other words, the SH annual DMS flux differences is a product of the DMS flux differences from June and December (together with the other months), likewise for annual CCN. However, the annual CCN sensitivity is not a product of the June and December CCN sensitivities; it is a product of the annual DMS differences and annual CCN differences.

Teasing out information about months not shown in this plot therefore requires caution.

**13. P3732, l3: ".. CCN contributions from sub-micron sea-salt must be taken into account : : :" I don't entirely understand why the contribution of UF sea-salt (Martennson) must be taken into account here (or just only this section). 1) The Gong scheme used already includes sub-micron sea-salt already (though not much UF sea-salt). 2) If we were confident that the Martensson emissions parameterization is correct, why wasn't it used for the entire paper? I understand**



**that in this case you are evaluating the relative CCN changes and UF sea-salt increases the CCN in remote marine regions, but the UF sea-salt particles contribute to the condensation/coagulation sink and thus would effect your results throughout the entire paper. 3) The CCN comparisons you show at Cape Grimm are better without UF sea-salt emissions (given, this is just one location).**

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) find 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.

This justification is now also included in the manuscript.