

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.

J. Pierce (Referee)

jeffrey.robert.pierce@gmail.com

Received and published: 23 March 2010

Review of “Low sensitivity of cloud condensation nuclei to changes in the sea-air flux of dimethyl-sulphide” by Woodhouse et al. 2010.

This paper uses a global CTM with aerosol microphysics to compare the sensitivity of DMS emissions and CCN concentrations due to 1) uncertainties in oceanic DMS concentrations, 2) changes in oceanic DMS concentrations due to climate change, and 3) interannual variability in wind speed. The authors do not assess the impact of wind speed changes due to climate change on DMS fluxes and CCN concentrations, but make strong recommendations to assess this is future work.

The major conclusions I took from this paper are 1) prognostic models of oceanic

C854

DMS concentrations predict very minor changes in DMS with climate change relative to present-day uncertainties in oceanic DMS and 2) CCN concentrations are not highly sensitive to changes in the DMS flux, thus 3) CCN concentrations do not change much from climate-change-driven DMS changes.

The paper is well written, is well within the scope of ACP and contributes new knowledge to our understanding of the CLAW hypothesis. I believe most people involved with aerosol microphysics had a hunch that the sensitivity of CCN to DMS changes would be small, but it is very good to see a detailed quantification of this. I definitely recommend this paper to be published in ACP once several comments and consistency issues have been addressed.

Specific comments

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).

2. P3722, l20: The competition between condensational growth and coagulation 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)).

C855

3. What nucleation scheme(s) were used. Because nucleation/growth is so important to the sensitivity, this should be included.
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.
5. P3724, l20: CLIM6 used PlankTOM5 too.
6. Top of P3727 and Section 3.6: I am curious to know if it is 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.
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 CLIM 6 have lower fluxes than CLIM1. I suggest going carefully through this section and making sure its consistent with Figure 4.
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.
9. Figure 6: I would find it useful if a 3rd figure panel that showed the fractional contri-

C856

bution of DMS to CCN was shown too.

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 coagulation scavenging is not very fast but not large enough to be subject to in-cloud 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.

11. Section 3.5 and Figure 4: Please discuss why you have negative absolute sensi-

C857

tivities in some cases (see my point above).

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.

13. P3732, I3: "... CCN contributions from sub-micron sea-salt must be taken into account ..." I don't entirely understand why the contribution of UF sea-salt (Martensson) 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).

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 3717, 2010.