Review of Sensitivity of cloud condensation nuclei to regional changes in dimethyl-sulphide emissions

Woodhouse et al. (2012) ACPD

Overall:

This paper attempts to examine how efficiently CCN are produced when DMS emissions are changed in different oceanic regions. It was found that the global CCN production per unit mass of sulfur emitted varies by more than a factor of 20 depending on which oceanic region the change in DMS emission flux occurs. The variation in CCN production efficiency was found to depend on CCN production (DMS oxidation, SO_2 oxidation, nucleation and growth) and removal (deposition) processes. The paper is well written and the topic (the role of DMS in climate regulation) is timely and relevant to ACP. I recommend following revisions for the paper prior to publication.

General Comments:

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. 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.
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.

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.

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 SO_2 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 SO_2 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 SO_2, and neglecting the dust effects may lead to the considerable uncertainties.

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?

Specific Comments:

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

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?

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.

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

References:

de Leeuw, G., E. L Andreas, M. D. Anguelova, C. W. Fairall, E. R. Lewis, C. O'Dowd, M. Schulz, and S. E. Schwartz (2011), Production flux of sea spray aerosol, Rev. Geophys., 49, RG2001, doi:10.1029/2010RG000349.