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Atmospheric Chemistry and Physics Discussions

# *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 2.

We thank the reviewer for the time taken to review our manuscript. The comments are appreciated and have served to improve the manuscript.

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

**I still have doubts about the treatment of new particle formation in this work. Yu and Luo (Atmosphere, 2010) pointed out that enhanced DMS emission and photochemistry during the austral summer season lead to significant new parti-**



**cle formation via ion-mediated nucleation (IMN) and much higher particle number concentrations over Antarctica and surrounding oceans. In their sensitivity studies, they found binary homogeneous nucleation remarkably underestimated CN10 number concentration at the German Antarctic station Neumayer. They concluded that the downward entrainment of new particles formed in the free troposphere is not able to account for the CN10 values observed in the Antarctic boundary layer. What do the authors think about the contribution of DMS to CCN over Antarctica and surrounding oceans? Will it impact the sensitivity of surface layer CCN to regional changes in DMS emissions?**

Merikanto et al. (2009) quantify the contributions of primary emission, boundary layer nucleation and free tropospheric nucleation to total particle number on a global scale. Fig. 6 in Merikanto et al. (2009) shows only $\sim$25% of the total number at Neumayer is explained by boundary layer nucleation. The disparity between the Yu and Luo (2010) study and the Merikanto et al. (2009) study suggests there is some uncertainty over the exact contribution of boundary layer nucleation (or ion-mediated nucleation) to total number concentration.

The present study considers CCN rather than total particle number concentration. Fig. 8 of Merikanto et al. (2009) shows the contribution of boundary layer nucleation to CCN concentrations. Around the Antarctic coastline, boundary layer nucleation contributes a maximum of 15% to CCN. In the Southern Ocean, boundary layer nucleation contributes less than 5% to CCN concentration.

The small contribution of boundary layer nucleation to CCN on a global scale (and particularly around Antarctica) strongly suggests that CCN sensitivity is not influenced by boundary layer nucleation. This assumption is particularly robust in the Southern Ocean, where Merikanto et al. (2009) show that most CCN ($> 85$%) are derived from primary (sea-salt) emissions.

**Figure 2b indicated that the change of DMS emission at SP2 patch can impact**

**CCN concentrations over a number of different regions which include anthropogenic polluted areas such as Argentina and South Africa. Many studies have shown that binary homogeneous nucleation cannot explain the boundary layer nucleation events observed in these polluted areas. If boundary layer nucleation is considered, will the sensitivity of surface layer CCN to regional changes in DMS emissions be changed?**

As is the case in the Antarctic / Southern Ocean region, the contribution of boundary layer nucleation to CCN concentration in South America is $\sim$5% according to Fig. 8 in Merikanto et al. (2009). Additionally, South America is also a strong source of primary particles. We are therefore very confident that boundary layer nucleation would not substantially change the CCN sensitivity.

**The author shown that the global mean sensitivities of surface layer CCN to regional changes in DMS emissions at SP1 and NA2 are much higher than other patches. Can the authors give more detailed discussion and explanation on this finding?**

Fig. 5 has been updated to include the aqueous-phase oxidation sensitivities - they were omitted in error.

Patches SP1 and NA2 have high CCN sensitivities in December because the rate of aqueous-phase oxidation in that region is relatively low (see Figs. 5 and 6). As concluded in the manuscript, a low rate of aqueous-phase oxidation promotes formation of new CCN via nucleation and growth. Also of interest is patch NA3 in December, which has a high CCN sensitivity, but an aqueous-phase oxidation sensitivity near the mean. In the case of NA3, the outflow of biomass burning aerosol from North Africa provides an abundant source of Aitken mode particles that can be aged and grown to CCN-relevant size.

The following text has been added to the manuscript: 'The link between CCN sensitivity and aqueous-phase oxidation is clear in the patches with the two highest CCN



sensitivities (SP1 and NA2 in December). In both of these patches, the aqueous-phase oxidation sensitivity is low, and so does not limit formation of new CCN. Also of interest is patch NA3, situated in the biomass burning outflow from North Africa. Patch NA3 has a high CCN sensitivity, but an aqueous-phase oxidation sensitivity near the mean. It is likely that the high CCN sensitivity of NA3 is the result of the biomass burning aerosol providing an abundant source of Aitken mode particles that are aged and grown to CCN-relevant size.'

**The typical cloud base height for low cloud is around 300-500 m. However, the authors limited the study to only surface layer CCN which usually locates lower than the cloud base height. I would like to suggest the authors to present the changes of column CCN concentration within boundary layer.**

CCN concentration (not column-integrated) is the relevant quantity when considering the aerosol indirect effects. The column-integrated CCN concentration is also strongly influenced by the depth of the boundary layer, and so is not an appropriate quantity for reporting the present results. The boundary-layer can be considered well-mixed, such that surface level CCN concentrations are representative of cloudbase CCN concentrations. Woodhouse et al., (2010) quote surface level CCN concentrations, and we wish to be consistent in the present study. Finally, surface CCN concentrations are most relevant to the majority of CCN concentration observations, as the observations tend to be at or near the surface.