

Interactive comment on “Modelling the contribution of sea salt and dimethyl sulfide derived aerosol to marine CCN” by “Y. J. Yoon and P. Brimblecombe”

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1. Introduction was revised according to the comments. Historical debates over the 2 issues, the ability of sea salt to contribute to CCN concentration and DMS nucleation ability in MBL, and this made the aim of our study in the manuscript clearer. Here are summary and answer to the questions. Blanchard and Cipriano (1987) argued that the background SS CCN concentration is 15 - 20 cm⁻³ and postulated that the biological regulation of the climate is less obvious than CLAW hypothesis. More recently, using a volatility technique, O'Dowd and Smith (1993) and O'Dowd et al. (1999a) succeeded in distinguishing non-sea salt accumulation mode aerosol from sea salt CCN and quantified the accumulation mode sea salt aerosol concentrations as a function of wind speeds up to 17 m s⁻¹. They also argued that sea salt aerosols are well mixed

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throughout the MBL because these aerosols have similar radii to sulfate aerosols. Murphy et al. (1998) also supported these arguments. They measured chemical composition of accumulation mode marine aerosols and found that almost all particles larger than 0.13 micrometer contained sea salt under unpolluted conditions. Katoshevski et al. (1999) and Pirjola et al. (2000) have challenged the dependency of marine CCN concentration on DMS flux. Katoshevski et al. (1999) argued that aerosol concentration in MBL is very sensitive to the entrainment of free tropospheric aerosols and sea salt emission. They proposed that a prediction of relative contribution to MBL CCN by sea salt and DMS derived sulfate is important to elucidate the relationship between marine biota and climate. Pirjola et al. (2000) also elucidated that DMS flux in MBL alone cannot explain new particle formation, instead they suggested that additional condensable species other than DMS-derived sulfuric acid is required to help thermodynamically stable sulfate clusters grow into detectable particle size.

2. A discussion section was added as the section 3.5. According to the purpose of this research, this discussion focussed more on relative role of DMS and SS in CCN formation than on detailed aerosol formation aspects. Here are the answers and reply to the points raised by the referee 1.

1) Limited size resolved spectrum: This issue was discussed in detail between P&R and Raes et al. as the referee pointed out. The main purpose of our study was to assess a relative contribution of DMS and SS to MBL CCN, and finally build up or support a hypothesis in terms of climatic effect. We agree that our approach extremely simplified the physico-chemical process in CCN formation. On the other hand, this simple infra-structure of the model made us easily add or test other important physical processes in the MBL, estimating DMS flux, modelling SS CCN from climatological mean wind speed, testing a sensitivity of condensation sink in terms of accommodation coefficient, etc. We believe that our simple and fast approach is an useful tool when cooperated into a global climate model and eventually test various scenarios arising from anthropogenic emissions or global wind speed fluctuation.

2) Total lack of coarse mode SS: This was discussed in the discussion section, as

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”Condensation of sulfuric acid onto super micron jet drop sea salt particles, which was not considered in this study, is nearly three times greater than that onto accumulation mode distribution according to a calculation by O’Dowd et al. (1997). This means that even our study might have overestimated the role of DMS in CCN formation in MBL due to the lack of sulfuric acid condensation onto super-micron sea salt particles. This fact also supports the importance of selection of accommodation coefficient when modeling the CCN concentration in marine environment as suggested by Pandis et al. (1995).”

3) Nucleation rate in our study: We used the parameterised nucleation rate from P&R and tuned this by using 1010. A sensitivity test showed that this factor could not affect MBL CCN variation seriously when compared with the mean wind speed fluctuation. Based on this test, we wanted to concentrate more on SS than on nucleation itself. A detailed discussion in conjunction with the limited size distribution in our approach was added as "The P&R model, which we adapted and modified for this study, is composed of only two aerosol size bins, and this simplified approach has been discussed in detail between Raes and Van Dingenen (1995) and Pandis et al. (1995). Because of the simplified aerosol size distribution, some dynamical information might have not been included in this study. This approach might not be able to fully reflect the nucleation of new particles and their growth from 0.001 micrometer to the first bin (0.023 - 0.1 micrometer), though we tried to tune the nucleation rate by adopting an empirical enhancement factor 10^{10} - see reaction (4) in Pandis et al. (1994) - from Katoshevski et al. (1999). As already been discussed by Katoshevski et al. (1999) and Pirjola et al. (2000), theoretical binary nucleation rate has failed to reproduce the MBL aerosol. Pirjola et al. (2000) also elucidated that even ternary nucleation can hardly contribute to the general MBL aerosol concentration except for the case when there is a significant removal of MBL aerosol such as due to precipitation. It is likely that whether a classical nucleation or tuned rate is used, this does not effect the MBL CCN variation seriously when compared with the effects due to mean wind speed. For example, a sensitivity test of this empirical factor showed 10 % increase in CCN concentration with

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the value 10^{15} , and no change at all with 10^5 , respectively. By contrast, changes in wind speed from 8 m s⁻¹ to 4 m s⁻¹ and from 8 m s⁻¹ to 12 m s⁻¹ showed CCN concentrations to decrease by 40 % and increase by 54 %, respectively. It is obvious that the contribution of DMS to MBL CCN is suppressed by wind speed because of direct contribution of sea salt to CCN concentration."

4) DMS(g) concentration in MBL: Our result showed averaged DMS(g) concentration as 85 ppt for the default simulation (a typical condition with sea water DMS concentration 2 nmol l⁻¹ and wind speed 8 m s⁻¹). We believe that this average value and diurnal variation is in good agreement with field measurements based on the references included. Because we did not consider nighttime DMS(g) reactions with NO₃, the average concentration might be slightly overestimated, but still this effect surely is a minor in the remote marine environment. Our result showed that the DMS concentration of 100 ppt can be reached before sun rise, but rapidly decreased soon after the reaction started when sun rise. Our default case simulation showed that DMS(g) concentration of the order of 100 ppt cannot be sustained for many hours, and we do not believe we overestimated DMS(g) seriously. When compared with the work by Pirjola et al. (2000), we calculated DMS flux after considering wind speed, which also affected SS CCN concentration. As a result, some high DMS(g) concentration assumption (high seawater DMS concentration and high wind speed) does not necessarily mean that DMS derived nucleation dominated CCN in MBL, this was visualised in Fig. 8 in our revised version.

5) sulfuric acid concentration vs measured value: In section 3.1, modelled mean sulfuric acid concentration, 0.05 ppt, and diurnal variation in default case simulation was validated as "A measurement by Weber et al. (1998) also reported the typical sulfuric acid concentration in MBL as approximately 0.1 ppt. Though another measurements by Weber et. al. (1997) are available from continental sites, the sulfuric acid concentration showed similar diurnal variation to our result. "

6) Aqueous phase oxidation of DMS derived SO₂: In P&R the aqueous phase oxidation of SO₂ was parameterised simply in terms of cloud frequency and SO₂ con-

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centration. In discussion, the aqueous phase reaction of SO₂ was mentioned as "The DMS derived SO₂ also efficiently react in sea salt cloud droplets (O'Dowd et al., 1997), then reduce the production rate of NSS CCN. Though DMS flux as a function of wind speed was not dealt with in O'Dowd et al. (1999b) as in our approach, they also postulated a reduced role of DMS in cloud droplet formation."

7) Cape Grim data: In section 3.4, this was added as "The activation diameter of CCN is dependent on the supersaturation, and the lower supersaturation means larger activation diameter limit and this leads less CCN available in the MBL as shown in Cape Grim data. This effect was not included in the work reported here."

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