

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) Limited size distribution We agree that many important physico-chemical properties of Aitken mode particles were extremely simplified or ignored due to size distribution we used. This limitation was discussed between P&R and Raes et al. (1995) in detail, and this was also added to our revised manuscript in the discussion section. In the revised discussion, we tried to focus on the main purpose of our study. Our main concern was to assess a relative contribution to CCN concentration by DMS and wind generated sea salt. Instead of using multi-sectional aerosol model, we wanted to test other important physical processes in the MBL, such as competition between DMS CCN and SS SSCN, DMS flux, SS CCN from climatological mean wind speed, condensation sink in terms of accommodation coefficient. Though our study used limited aerosol size distribution, and thus dynamics and growth of the recently formed parti-

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cles were simplified, we believe that our result should be reflected to the global climate change scenarios. We used parameterised nucleation rate from P&R and tuned this rate using a coefficient like the work by Katoshevski et al. (1999), 10^{10} . We agree that this is not an ideal approach compared with Pirjola et al. (2000) which used binary and ternary nucleation rate, but sensitivity test of this factor showed that this nucleation can not affect CCN concentration in MBL. For example, a sensitivity test of this empirical factor showed 10 % increase in CCN concentration with 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. As for the particle growth from the first to second mode, we calculated growth rate and it is only $9 \text{ particles cm}^{-3} \text{ day}^{-1}$. This means that CCN formation growth by condensation of Aitken particles contribute very little to the population of CCN in MBL. We think your concern over the growth process has already been included in our approach.

(2) FT aerosol contribution to MBL CCN: We focused on DMS and SS rather than FT entrainment. What we have tried was to assess the role of the entrainment in the competition between DMS CCN and SS CCN. The FT entrainment process of our approach is like this; we assumed a constant Aitken and accumulation mode concentration in FT and we calculated the flux using entrainment velocity. The accumulation mode aerosol flux was small compared with the Aitken mode aerosol. Our approach also assumed that entrained aerosol experience condensation growth and other processes and eventually some of them make CCN. We defined these as FT CCN, strictly speaking, CCN which resulted from the FT entrainment process. Though the chemical processes in the MBL were simplified in our study, we believe that we visualized the effect of entrainment on MBL CCN as the way you have commented. We revised our manuscript to make these features clearer to the readers. In abstract, we replaced 'CCN from FT' by 'aerosol from FT', and 'FT CCN' by 'FT aerosol'. We revised sec-

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tion 3.2.2 as 'FT may explain at least half of the CCN formation in MBL'. In addition, a sentence in conclusions was revised as 'In general, the FT aerosol and SS CCN similarly contribute to the MBL CCN concentration.' The chemical composition of the FT aerosol is beyond the scope of our research, so we are reluctant to define FT aerosol as simply NSS CCN. As we carefully clarified in the revised manuscript, FT CCN of which the meaning in our study is CCN formed as a result of FT-MBL entrainment and small amount of accumulation mode aerosol directly entrainment to MBL. Instead, we categorized MBL CCN into 2 groups V NSS CCN and SS CCN in section 3.3 where we especially simulated a relationship between MBL CCN and various seawater DMS concentration and wind speeds.

(3)Sensitivity tests: We built up a table as you recommended. The DMS flux model shows different values according to parameters used. As for DMS flux calculation parameters, we used corrected L&M method by Turner et al. (1996) for DMS. We acknowledge that our DMS flux might be larger by a factor of about 2 if we used parameter by Wanninkhof (1992), but our main arguments are that MBL CCN concentration is less dependent on DMS flux itself than the previous results such as Pandis et al. (1994) and Lawrence (1993). We believe that the Fig. 10, which showed a relatively weak relationship between DMS flux and MBL CCN concentration, can replace the DMS flux parameter sensitivity test, and it is not needed to discuss on these transfer velocity parameters in detail for our study. As for SO₂ heterogeneous oxidation, we indirectly tested sensitivity by changing cloud frequency in MBL. More detailed discussion on this process have been added to section 3.4 Table below, which was revised in our manuscript, is the sensitivity test for important parameters in our study.

(4) Accommodation coefficient sensitivity test: We used the value of 0.70 as sulfuric acid accommodation coefficient. The reason we used 0.02 in the sensitivity test is that we wanted to find a reason why P&R's original work showed linear relationship between DMS flux and MBL CCN. Because their original work used 0.02 as the default value from Van Dingenen and Raes (1991), P&R model seriously underestimated the condensation of sulfuric acid onto aerosols. We also agree that the value 0.02 is unre-

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alistic based on the reference we used (Jefferson et al., 1997). We argue that sulfuric acid accommodation coefficient is one of the important factor when modelling the contribution on DMS derived sulfur compounds to MBL CCN system, and this argument could have been driven from this sensitivity test.

(5)FT aerosol specification: This was specified in the revised manuscript as "These entrainment of aerosols from the FT into the MBL was added to the P&R model as Eq. (5). Katoshevski et al (1999) adopted the FT aerosol concentration from Weber and McMurry (1996), and this data was also used in our research. The approach in our study assumed that there are fluxes of DMS out of MBL and other DMS derived sulfur compounds have same concentration for both MBL and FT. "

<Technical comments> (1)The paper by Turner et al. (1996) was added to the list of references. (2)We have decided to use Aitken mode instead of nucleation mode aerosols.

<Revised list of references> Pandis S. N., Russell L. M., and Seinfeld J. H., Reply to Comment on "The relationship between DMS flux and CCN concentration in remote marine regions", J. Geophys. Res., 100, 14357-14358, 1995. Raes F. and Van Dingenen R., Comment on "The relationship between DMS flux and CCN concentration in remote marine regions". J. Geophys. Res., 100, 14355-14356, 1995. Turner, S.M., Malin, G., Nightingale, P.D. and Liss, P., Seasonal variation of dimethylsulphide in the North Sea and an assessment of fluxes to the atmosphere, Mar. Chem., 54, 245-262, 1996.

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Parameter	Default value	Tested value	CCN	Change in CCN [%]
Sea water DMS concentration	2 nmol l ⁻¹	1	105	-17
		4	152	+21
Wind speed at 10 m height	8 m s ⁻¹	4	76	-40
		12	194	+54
MBL height	1000 m	500	126	0
		1500	120	-5
Entrainment velocity	0.36 cm s ⁻¹	0.18	122	-3
		0.54	123	-2
Relative Humidity	0.8	0.7	119	-6
		0.9	141	+12
Precipitation frequency	1 / 10 days	1/5	113	-10
Cloud frequency	1.0 day ⁻¹	0.5	137	+9
		1.5	120	-5
(OH)g max value	5 10 ⁶ molecule cm ⁻³	2.5 10 ⁶	104	-17
		7.5 10 ⁶	142	+13
SO ₂ yield rate from DMS oxidation	0.9	0.7	118	-6
		1.0	130	+3
SO ₂ deposition velocity	0.5 cm s ⁻¹	0.25	130	+3
		1.00	121	-4
H ₂ SO ₄ deposition velocity	1.0 cm s ⁻¹	0.5	126	0
		1.5	126	0
Accommodation coefficient	0.70	0.35	120	-5
		0.90	129	+2
D1	0.023 micrometer	0.001	77	-39
Da	0.1 micrometer	0.05	190	+51
D2	0.6 micrometer	1.0	116	-8
Aitken nuclei deposition velocity	0.04 cm s ⁻¹	0.02	127	+1
		0.08	125	-1
CCN deposition velocity	0.06 cm s ⁻¹	0.03	131	+4
		0.12	118	-6
Nucleation empirical coefficient	10	5	126	0
		15	139	+10

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