Reply to referee #1

First of all, we would like to express our appreciation to the referee for their work on reviewing the manuscript and providing the constructive and valuable comments.

Secondly, we would like to respond to the two major comments suggested by the referee.

The first is that the simulations presented lack other aerosol species, in particular sulfate, when the vast majority of other microphysical aerosol models routinely represent them. This seems to negate somewhat the advantages of using a microphysical model, as coagulation and condensation are ignored. These may be important in some instances for growing sub-CCN size aerosol to CCN sizes, with implications for removal (and therefore mass, number and optical proper ties). The lack of other aerosol species also drastically limits the number of observations that can be use to evaluate the model.

My other comment would be that the manuscript may be better suited to Geoscientific Model Development than Atmospheric Chemistry and Physics, as it evaluates a recent (hybrid) parameterisation rather than fundamentally changing the way we think about sea-salt parameterisations. Nevertheless, the work is a useful contribution, and I would recommend publication after consideration of the points listed below.

We are aware that many models treat multiple types of aerosols. Our group is in the process of combining several types of aerosols, and have other papers discussing sulfates, dust, and smoke for instance with the same model. However, we also think there is value in isolating the individual aerosols and concentrating on getting them right individually. It can be distracting to try to do all of the aerosol types at once. We now note some effects likely due to added marine aerosols in the conclusion of our paper, and in the paragraph below.

"Obviously sea-salt is not an isolated aerosol species in the marine environment. Sulphate and organic aerosols as well as their gaseous precursors co-exist in the marine environment. Sea-salt dominates the coarse-mode marine aerosol. Its large surface area as well as large pH value facilitates the condensation of precursor gases, such as SO_2, and their subsequent oxidation . The consumption of precursor gases inhibits the nucleation of sulfate aerosols. Coagulation of sulfate aerosols with sea-salt aerosol will change the marine CN spectrum, which in turn influences the CCN activation and removal processes. We are aware that the interactions between sea-salt and other marine aerosol species influence the emission, removal, and optical properties of the sea-salt aerosols. These interactions are not currently included in our simulation because we also think there is value in isolating the individual aerosols and concentrating on getting them right individually. Further model improvements are needed such as treating the condensation of sulphate precursors on sea-salt particles, considering the coagulation of sulfate and sea-salt particles, and assessing the organic sea spray emission, among others. This work is the basis for future studies we plan of marine aerosol direct and indirect effects using the coupled CAM/CARMA model. "

We did not have the goal of defining new parameterizations, these require much new observational information, for example to determine if there is temperature dependence to the source function as some models suggest. Rather our goal is to determine if we can reproduce the data available using a combination of available source functions. We do think our work is new in the sense that we are the first to show we can simulate mass, optical depth and number in agreement with (limited) data in a climate model. Many models have only simulated one aspect of the observations, often mass, in comparison with data. We think it is essential to show models can simulate all three properties for them to be most useful for climate simulations.

Now we list our point-to-point response to the specific comments:

Particle swelling: may be more accurately described as hygroscopic growth?

We change to hygroscopic growth as suggested.

p24502, l8: '...we used...', recommend using the present tense when describing your study and its results.

We use present tense as recommended.

Is there a reference for the CARMA model? Or is this the first time it has been described? If the latter is the case, it should be stated.

Description of the development of CARMA is available in Toon et al., 1988 (p24502, l26). The CAM coupled model is first described in Bardeen et al., 2008 (p24503, l7). Su and Toon (2011 and 2009) describe its application to dust. We have added the latter references to the paper.

p24503, l19: 'Coagulation is not considered in the model since the low number concentrations and short lifetime of SSA indicate that it is not an important process'. Is this true even at ultrafine sizes of SSA, where concentrations may be considerably higher?

Sea salt particles usually have a lifetime of about a week before they are removed by wet scavenging, so coagulation is not very important. However, we have done a simulation including coagulation and find the relatively small number of small radius SSA is not strongly influenced by coagulation. For example at Midway Island we find the number of particles is reduced from 146 to 142 cm^{-3} when we include coagulation. This change is a "line width" on a size distribution plot. We now describe this simulation in the paper.

p24504, last paragraph: could add couple of sentences in here describing how number fluxes shown in Fig. 1 vary with wind speed.

We added "The number concentration for all the source functions shown in Fig. 1 increases with wind speed while the shape of the spectrum is unchanged, except for the Caffrey source function. The shape of the Caffrey function will change above 9 m s^{-1} to include spume particles as the result of introduction Smith et al. (1993) source function. These wind speed dependencies are illustrated in Fig. 2"

p24514, l8: Does assumption of 100% of SSA residing in cloud water not mean that removal is overestimated, particularly in number? According to Fig. 1, there is emission of sea-salt aerosol down to 0.01 um radius. These aerosol are too small to be activated (e.g. at supersaturation of 0.1%), and therefore presumably should also not be nucleation scavenged. This would clearly have a dramatic impact on the number size distribution. Perhaps this assumption is necessary as there is no coagulation nor condensation (due to lack of other aerosol species) represented in your simulations. This is perhaps a key limitation in the assessment of the ability of the model to reproduce number concentrations and the size distribution at smaller aerosol radii.

We use solubility of 0.3, 0.5, and 0.8 to test the model, which means 30%, 50%, and 80% of SSA resides in the cloud water. We rephrase the description of the solubility factor in the text.

We have tried a size-dependent in-cloud scavenging ratio (solubility factor) and the below-cloud scavenging coefficient (Henning et al., 2004; Dana&Hales, 1976). The size-dependent scheme considers both the aerosol and rain droplet sizes. However, the efficiency of removal, which is dominated by Brownian motion for the smaller particle sizes in our model is not significantly affected. Moreover, the small sea salt aerosols likely have sulfate on them and therefore are more likely to be in the CCN size range as total aerosols than our model suggests. However, the idea you suggest will be important to reconsider when we add sulfate to the model.

p24515, l10: 'Solubility factor'. I don't think this parameter has very much to do with solubility, and should therefore be renamed. As the authors state in the sentence before, the assumption is that all SSA are in cloud water for the purposes of nucleation scavenging, and therefore that all aerosol are soluble. Perhaps a better term might be 'cloud to rainwater conversion factor'. Presumably this factor is per timestep?

As indicated by the referee, 'cloud to rainwater conversion factor' is a more precise description of this parameter. However, "Solubility factor" is the term used by the CAM community, thus we prefer to keep this terminology for conventional purpose. This factor is per timestep.

p24514, l19: That lifetimes are tuned also means that burdens and removal are tuned, which has implications for the conclusions about how well the source parameterisations reproduce the observations.

We only tuned the rainout lifetime, which mostly impacts smaller particles. The larger ones are removed by falling out. Hence the mass and optical depth are not greatly impacted by the rainout as discussed in Fig 21. By simulating optical depth, mass and number we can determine the behavior of source functions across the range of sizes. For example, as seen from Table 3, the Gong source function systematically underestimates the optical depth, even using solubility factor of 0.3. This is largely because the optical depth is controlled by sedimentation. It is unfortunate that observational data on the rainout rate is not really available. The literature on this subject is sparse and largely based on qualitative arguments. We set our rainout time to be similar to a large group of other models, so they should all find results similar to ours assuming their wind speeds are similar. We do not tune any of the source parameterizations ,those are based on laboratory or observational work. Since the burden has to be consistent with both the production rate and the removal rate we expect the results to be internally consistent if the burdens are correctly simulated.

p24515, l7: It would be useful to describe how the Savoie & Prospero (1977) results were obtained. Are they indicative of sea-salt mass only, or likely to be influenced by other aerosol components?

We now described the SP data in more detail. "The sodium mass is measured by flame atomic absorption with a one-standard deviation uncertainty of 2%. The mass of sodium is then multiplied by 3.252 to retrieve the mass of sodium chloride. Uncertainty may arise from the different samplers they used and the varying locations from the shoreline at different sites. For further details of the source of uncertainties, refer to Savoie et al. (1994). To minimize island effects on their data, SP used wind sensors to control the sampler pumps so that the wind during the measurements was off the ocean at a speed greater than 1 ms^{-1}.
"

p24515, l25, and Fig. 6: I think it would also be useful to show the Gong results in this plot. How well Gong reproduces the seasonal variation should also be commented on.

As shown in Table 3, the Gong source function also shows reasonable agreement with the observations. Below we attached Figure C1 showing both the CMS and Gong source function results. We didn't' add Gong to Figure 6 since it would be too busy. In term of mass concentration, Gong basically captures the seasonal variation of the SP data as the CMS source function does. The Gong source function produces lower mass concentration than the CMS source function, especially at the maximums when high wind speeds are high.

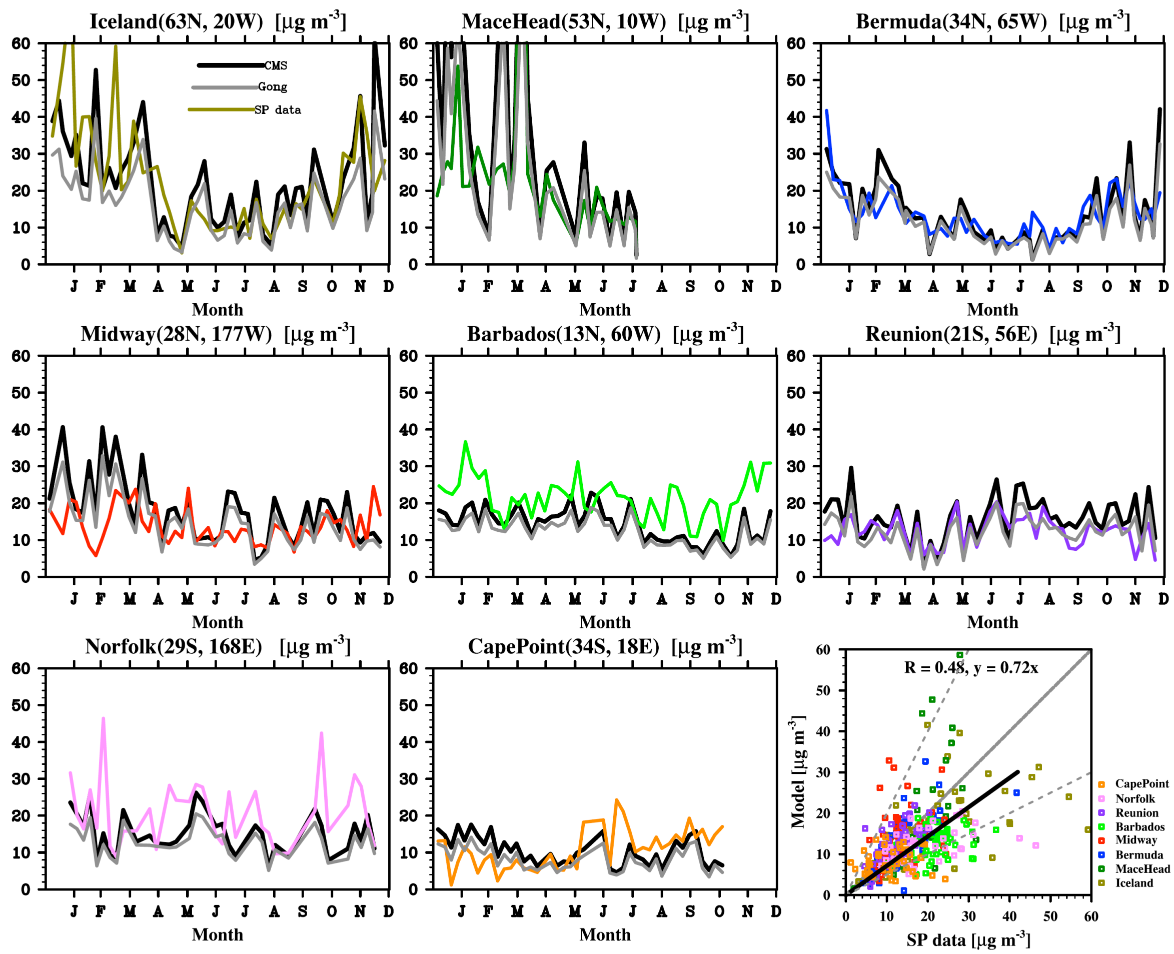

Figure C1. Seasonal variations and scatter plot of mass concentrations in the marine boundary layer comparing the model results to the measurements at eight coastal sites by the University of Miami global network (SP data, Savoie and Prospero (1977)) in 1994. We present results from the CMS (black) and Gong (grey) source function with a solubility factor of 0.5. The scatter plot is for the results of the Gong source function. The solid line is the total linear fit to all the data. The grey short dash line is the one-to-one line and the grey long dashed lines are the one-to-two and two-to-one lines.

Section 3.2.2: The implications (for the CMS parameterisation / model) of this section are not clear to me. My impression from the text is that, under certain conditions, optical depth from sea-salt can be estimated directly from wind speed. This then gives two estimates of optical depth (the other

calculated in the model). The two estimates may give similar results, but I don't think this comparison can be used to evaluate model skill.

The optical depth due to sea salt is not easily separated from the influence of the other aerosol species. Since sea-salt optical depth has been observed as a function of wind speed, it is one of the few ways to see if the modeled optical depth agrees with observed optical depth under the same wind speed conditions. Unfortunately if you look at other models you will see there is no reliable database for sea salt optical depth. Using the wind speed dependence is at least a step forward in finding a suitable database. Further observations that single out the sea salt optical depth and its wind speed dependence would be useful. We have changed the wording in this section so that it is clearer that Mulcahy is not global data, just an interesting observation in one place, that our model suggests will work in other places.

p24523, l9: 'This mode is probably due to sulfate and organic aerosols from the oceans or pollution aerosols that are not represented in the model.' I agree, though does this also have implications for Sect. 3.2.1, 'Comparison with AERONET optical depth'?

We are comparing AERONET coarse-mode optical depth in Midway Island. This mode should be mainly sea salt optical depth.

Fig. 14: The mode from sulfate and organic aerosol in Fig. 14 may be reduced in winter, when organic emissions including DMS are likely to be at their minimum. Could you also show a plot for December or January, and comment on this?

We replace the March plot with January plot. We do see a relatively smaller fine-mode in January which could be due to the decreased organic and DMS emission in January.

p24525, l1: It is also possible that these 'uncertainties' may alter the shape

of the size distribution also.

That is possible. However, the shape of the source functions does not depend on wind speed except for spume while the flux is a strong function of wind speed. Hence one expects the shape to be less variable than the absolute abundance. One learns different things looking at the shape than looking at the absolute abundance.

p24526, l17 and Fig. 18: I'm not sure that this is a new result, it has been well described before. This paragraph and figure could be removed.

Although it is commonly agreed that mass and number are dominated by difference size ranges, it is not very often mentioned that the mass and optical depth are dominated by different sizes. Climate models usually reproduce mass and they assume optical depth should also be well represented. We would like to show in Fig. 18 that it is not an absolutely correct assumption.

p24527, l5: 'The high optical depths near Peru are due to the effect of the Andes Mountains on the NCEP wind field.' This is interesting, has this been described / investigated anywhere?

This is through conversation with Dr. J.F. Lamarque at NCAR who produced the NCEP input for this investigation.

Technical corrections

We have made all of the changes suggested.

p24500, l6: 'We aimed at finding...' should be 'We aimed to find...'

p24500, l11: 'the research...' should be 'this research...'

p24500, l24: Sea-salt or sea salt? Need to be consistent with hyphenation throughout.

p24501, l14: fine-mode and ultrafine-mode, specify sizes

p24501, l16: 'SSA particles activate...' should be 'SSA particles can serve...'

p24501, l21: '...as small as 0.01 um...', radius or diameter?

p24503, l7: 'advective' should be 'advected'.

p24503, l14: '2_ _ 2.5_', state which is longitude and which is latitude.

p24504, l13: 'They are...' should be 'There are...'

p24505, l6: 'Martensson et al' should be 'Mårtensson et al', and elsewhere

p24505, l29: 'stands for' should be 'is'

p24505, l29: 'However, as shown in Fig. 1, Caffrey et al., (2006)'s number flux is about one magnitude higher below 0.1 um compared to Clarke et al. (2006).' Suggest clarifying to 'However, as shown in Fig. 1, below 0.1 um Caffrey et al., (2006)'s number flux is about one magnitude higher than Clarke et al. (2006).'

p24506, l11: 'see' should be 'show'

p24506, l14: 'fit demands' should be 'fit the demands'.

p24508, l7: Suggest deletion of 'therefore',

p24508, l8: Sentence 'The wind field in CAM...' is repeated information. Suggest that following sentence 'The model runs in an offline mode...' could be moved to Model Description section.

p24508, l24: Weibull wind speed distribution, again repeated information

p24509, l6: 'is the a two-parameter'

p24510, l10: '...and we accept vg we just calculated.' should be '... and we accept vg as in Table 2'?

Eq. 13: not all terms are defined?

p24510, l16: ‘Associating the formula of vg and Re,’ can be deleted.

p24510, l16: ‘gravitational sedimentation velocity’ is same as ‘fall velocity’? Should be consistent and stick to one term.

p24510, l20: ‘vg varies a little with location since the wet radius depends on location.’ Expand on this? Do you mean wet radius depends on humidity, which varies with location?

p24511, l6: ‘...where the constant of proportionality vd is called...’ could be changed to ‘...where vd is...’

p24511, l9: ‘We use the method described in Zhang et al. (2001)’... to calculate dry deposition velocity

p24511, l22: ‘It is determined’. What is determined, Rs?

p24511, l24: ‘...respectively.’, add reference to Table 2 at the end of this sentence?

We add reference to Table 2 at the end of the paragraph.

p24512, l10: ‘The dry deposition...’ should be ‘Dry deposition...’

p24514, l10: ‘percentage’ should be ‘fraction’

p24515, l8: ‘Since...’ should be ‘As...’

p24516, l6: ‘...than CMS...’ should be ‘...than the CMS...’

p24516, l7: ‘Note that we did not apply...’, this sentence needs rephrasing.

This sentence is deleted.

p24516, l10: ‘...results to the CMS...’ should be ‘...results to the CMS source function...’

p24516, l18: ‘since’ should be ‘as’

p24516, l18: ‘production’ should be ‘source’

p24516, l19: Please provide IMPROVE reference

p24520, l22: ‘The wind speed dependence...’, of Total number? SSA number?

Eq. 24: Terms need defining.

p24521, l19: ‘Again, the model and the data are in different years, which could also bring in some of the discrepancies.’ to ‘Again, the model and

the data are in different years, which could introduce some discrepancies.'

This sentence is deleted.

p24522, l6: '...different than the data,' to '...different to the data,'

p24522, l14: 'radius' to 'radii'

p24524, l11: 'Vigniti' should be 'Vignati'

p24525, l15: 'rage' should be 'range'

p24525, l22: Generally not good idea to start a sentence with a number, suggest rephrasing

p24527, l3: 'removing' should be 'removal'

p24527, l14: 'among' should be between 'between'

p24527, l29: 'most' should be 'mostly'

p24528, l16: 'SSA model' could be 'SSA source function'?

We mean to say this is a model simulating sea salt.

p24529, l10: 'tends' should be 'tend'

"Roaring Forties" is a name, so we prefer to use 'tends'

Fig. 1: In legend, 'Caffery' should be 'Caffrey'?

Corrected.

Fig. 5: 'Resident Time' should be 'Residence Time' on y-axis

Corrected.

Fig. 6: Could you also show the results using the Gong parameterisation? Another useful statistic to calculate would be the normalised mean bias: ((model –observation) / observation).

As show in Fig C1, we include Gong parameterisation in the plot. The bias is included in the text. The normalized mean bias is 0.340, 0.145, -0.009 using CMS source function with solubility factor of 0.3, 0.5, and 0.8, respectively. The normalized mean bias is 0.036, -0.089, and -0.197 using Gong source function solubility factor of 0.3, 0.5, and 0.8, respectively. The normalized mean bias measures the relative difference between the model and the observation. It does not tell one the correlation between the model and the observation. The regression fit measures how representative the model is to the observation. The two statics could result in different conclusion about which gives the best fit. Anyway, both statics shows that both source functions perform reasonably well except CMS source function with solubility factor of 0.8.

Fig. 7: Would be useful to see a scatter plot as in Fig. 6, with statistics including bias.

The scatter plot is shown as in Figure C2. The bias is 2.52. The correlation (R) is reasonably well meaning that the spatial distribution is reasonably modeled. Most overestimation of the model is in the coastal site where a global model with very coarse resolution is not good enough to represent the on-shore transport. Also as we mentioned in the manuscript, the IMPROVE data underestimates the sea-salt mass. Here in Fig. 7, we would like to illustrate that the removal process is reasonable simulated by comparing the gradient of the mass concentration over continent.

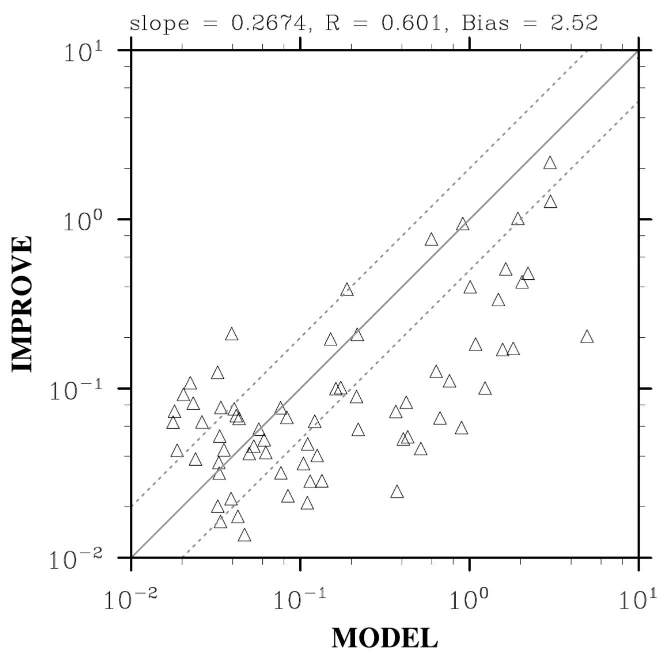

Figure C2. Scatter plot of the Modeled mass concentration in the surface layer and IMPROVE dataset. Each triangle represents a site (or grid in the model) we compared in Fig. 7. The model using the CMS source function with solubility factor of 0.5, unit: μg m^{-3}. The normalized bias, the slope of the regression line and the correlation (R) are shown. The solid line is the one-to-one line and the short dash lines are one-to-two and two-to-one lines.

Fig. 9: Please be more specific than 'roaring forties', give latitude band.

The latitude band is given.

Fig. 9: 'The dash lines...' should be 'The dashed lines...' in the caption.

Corrected

Fig. 10: Also calculate bias.

The bias is included in the text. The normalized mean bias is 0.311, -0.044, using CMS source function with solubility of 0.3, 0.5, and 0.8. The normalized mean bias is -0.336, -0.510, and -0.621 using Gong source function with solubility of 0.3, 0.5, and 0.8.

Fig. 11: Are the 'measurements' actually from Eq. 24?

Yes, the coefficients (a_0 and N_0) can be referred to O'Dowd and Smith (1993).

Fig. 12: Need to mention in figure caption the 10 cm^{-3} offset.

Corrected.

Fig. 20: In caption, need to mention that plot shows surface level number.

Corrected.

Reference:

Caffrey, P. F., Hoppel, W. A., and Shi, J. J.: A one-dimensional sectional aerosol model integrated with mesoscale meteorological data to study marine boundary layer aerosol dynamics, J. Geophys. Res., 111, D24201, doi 10.1029/2006jd007237, 2006.

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O'Dowd, C. D., Smith, M. H., Consterdine, I. E., and Lowe, J. A.: Marine aerosol, sea-salt, and the marine sulphur cycle: A short review, Atmos. Environ., 31, 73-80, 1997.

Savoie, D.L., J.M. Prospero, R. Arimoto, R. A. Duce: Non-sea-salt sulfate and methanesulfonate at American Samoa, J. Geophys. Res., 99(D2), 3587-3596, 1994.

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Smith, M. H., Park, P. M., and Consterdine, I. E.: Marine aerosol concentrations and estimated fluxes over the sea, Q. J. Roy. Meteor. Soc., 119, 809-824, 1993.