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Interactive comment on "Global relevance of marine organic aerosols as ice nucleating particles" *by* Wan Ting Katty Huang et al.

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Review of Huang et al. "Global relevance of marine organic aerosols as ice nucleating particles"

This paper addresses a question that is currently of some interest to the atmospheric ice nucleation community: the roles of marine organic aerosol (MOA), as contrasted with dust, in contributing to heterogeneous freezing in clouds and impacting cloud properties and the Earth's radiative budget.

Consistent with previous studies, the authors find that MOA frequently contributes the majority of mixed-phase cloud freezing nuclei at lower altitudes and high latitudes, especially over the Southern Ocean, with the freezing contribution from MOA greater

than that from dust on up to 40% of days under a variety of sensitivity cases.

In addition, the authors show that the relative importance of marine organic matter and dust as ice nuclei depends strongly on the choice of ice nucleation parameterizations for the two particle classes. This motivates a need for further experimental work to understand and quantify the factors that contribute to the observed variability in the INP activity of both dust and MOA, particularly at warmer temperatures.

Two published parameterizations of MOA INP (Wilson et al., 2015, drawn from sea surface microlayer samples, and DeMott et al., 2016, from laboratory-generated sea spray) are compared and shown to produce very different contributions of MOA to atmospheric freezing, although the percentage of time that MOA contribute the majority of INP is similar in each case.

Among other sensitivity experiments, the authors compare the impacts on cloud ice and liquid properties in simulations where MOA does not trigger freezing (noMOAfrz) and where it triggers freezing at a high rate (MOA100). The freezing induced by MOA is shown to produce a small (3%-9%), but statistically significant, decrease in zonal annual mean in-cloud mean ice crystal radius in near-surface clouds over the Southern Ocean. The changes in aerosols and aerosol-cloud effects are shown to result in statistically significant radiative cooling that is almost entirely attributable to an increase in outgoing longwave (terrestrial) radiation at high Southern latitudes, and that arises from a synergistic interaction of MOA acting as CCN and as scattering aerosol, and MOA acting as INP. MOA that do not act as INP, by contrast, are shown to produce a small zonal mean increase in reflected solar radiation, consistent with previous studies on the simulated effect of non-ice-nucleating MOA.

I have one comment that I feel needs to be seriously considered before publication, and which may require some corrections to one simulation. I am unclear on whether the DeMott et al. (2016) experimental data were interpreted and applied in the most appropriate manner. These data are best understood as characterizing the INP activity

of the total sea spray aerosol (including both salt and MOA), but a fit to these data appears to have been to only the MOA portion of the model's sea spray. If I have understood correctly what the authors did here, this one sensitivity case should be corrected before publication, as it will lead to a potentially significant underestimate of the globally-distributed concentrations of MOA INP that are implied by DeMott et al. (2016) and could create some confusion in the community about the appropriate interpretation of these data.

After the comments have been addressed, I strongly recommend this paper for publication. This study will be the most careful and comprehensive treatment of the global significance of MOA as INP to date. It constitutes an important contribution that advances the community's understanding of atmospheric INP sources, and of the importance of various uncertainties impacting the simulation of their contributions to freezing in clouds. In particular, it goes beyond most previous studies on the topic in several ways, in particular: (1) by comparing different observationally-derived representations of MOA INP activity for the first time in an atmospheric model, (2) by simultaneously considering the sensitivity to different parameterizations of sea spray emissions and of dust INP activity, and (3) by going beyond prediction of temperature-dependent INP concentrations to also investigate simulated impacts on clouds.

Major comments: 1. The most important comment I have is on the interpretation of the DeMott et al. (2016) dataset. DeMott et al. (2016) reported CFDC measurements of the ice nucleation activity of laboratory-generated sea spray aerosol particles that were composed of a combination of salt and organic matter. It's not completely clear to me whether the fit to the DeMott data (as presented in Fig 2) accounts for this in some way, but if I am understanding correctly, a fit was produced from the data in Fig 3 of DeMott et al. (2016), or a subset of those data, and then applied only to the surface area of the MOA portion of the sea spray aerosol - effectively, the surface area that the sea spray would have if the salt were removed (Eq. 4). It would be more appropriate to treat the DeMott data as representing the INP activity of the total sea spray surface

area of particles including both salt and MOA, and to then apply the fit to that total surface area as simulated by the model. This would produce an INP activity that is greater than what is calculated when only considering the organic portion of the sea spray, but still substantially less than what would be calculated using the Wilson et al. (2015) relationship. This needs to be clarified and discussed, and if am understanding correctly what was done, I think the calculations in this one sensitivity case need to be corrected in order to accurately represent the conclusions that can be drawn from the existing experimental data.

2. I'm aware that ECHAM-HAM cloud microphysics is continually being developed further and improved. To provide some perspective the simulated cloud impacts, it would be very helpful if the authors could comment a little further on the current level of confidence in the model's ability to credibly simulate mixed-phase cloud microphysics. In particular, the authors note that the model has a weak sensitivity to heterogeneous ice nucleation, but it's not completely clear what "weak" means here - i.e. is this in comparison to other models, to observational constraints, or to prior "expert judgement"? I recognize that this is the subject of another manuscript in preparation, but I think it is also important to contextualizing the results presented in this paper.

3. It would be helpful to see comparisons of the MOA100 versus noMOAfrz cloud properties on a seasonal basis, rather than only an annual mean basis. One would expect the strongest effects to occur in summertime at high latitudes (especially Southern hemisphere DJF), when longwave and shortwave radiation fluxes are greater. Could this explain why there are relatively large, and statistically significant effects on annual zonal mean LW and SW radiation at high Southern latitudes (Fig 11) despite limited effects of MOA INP on annual zonal mean cloud properties (Fig 9)?

Minor comments: 1. Figure 12: in the discussion of this figure, it is pointed out that using the statistical significance criterion applied here, some differences that are identified as statistically significant in the 10-year free-running simulation are no longer identified as significant in the 20-year free-running simulation, which the authors attribute to nat-

ural variability arising from dynamical feedbacks in the two simulations. How sensitive is this finding to the choice of significance threshold? To what extent does this finding hold if a stricter significance criterion is applied (e.g., $p < 1\%$ or $p < 0.1\%$ instead of $p < 5\%$)?

2. Appendix A: For the comparison with Mace Head and Amsterdam Island data, arguably it might be more appropriate to compare the model's MOA with observed water-insoluble organic carbon (as opposed to total organic carbon) – which would actually would produce a better agreement with the Rinaldi et al. (2013) parameterization, at least for the Amsterdam Island seasonal cycle. I don't think this matters for the conclusions of the paper, but it might be worth mentioning.

3. Appendix A: Rinaldi et al. (2013) is one of the better available chlorophyll-based MOA parameterizations and is clearly an appropriate choice for this study.

However, I think it would be appropriate to explicitly point out to non-specialist readers that considering the large differences between various models, it is not possible to infer from this figure that previous studies (Burrows et al., 2013; Vergara-Temprado et al., 2016) either significantly over- or underestimated MOA INP concentrations, compared to the current paper. This is already alluded to on p. 5, l. 3-5, but it should be pointed out again with respect to this specific analysis. Both previous studies documented that they produced OC concentrations consistent with the value typically observed in the remote marine atmosphere. In addition, the previous studies and Huang et al. have all pointed out that the uncertainty in modelling MOA INP concentrations is currently driven primarily by uncertainty regarding the INP activity of MOA, which is uncertain to at least two orders of magnitude. While important uncertainties remain in modelling of atmospheric MOA concentrations, these are comparatively well-constrained by observations of boundary-layer MOA concentrations from coastal stations and ships.

Typographical / technical comments: 1. P. 9, l. 19-20: "the total sea spray is split . . . following Long et al. (2011)." I don't fully understand this sentence. Please reword for

clarity.

2. Maybe it's just my printer, but the hatching on some figures (e.g., Figure 9) is really difficult to see. Can this be improved?

3. P. 21, l. 5: ". . .further diagnostics are performed. . ." ("is" -> "are")

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