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Interactive comment on “Effects of Marine Organic Aerosols as Sources of Immersion-Mode Ice Nucleating Particles on High Latitude Mixed-Phase Clouds” *by* Xi Zhao et al.

Anonymous Referee #2

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The manuscript titled: “Effects of Marine Organic Aerosols as Sources of Immersion-Mode Ice Nucleating Particles on High Latitude Mixed-Phase Clouds” discusses the impacts of adding marine organic aerosols (MOA) into the Community Atmosphere Model (version 6) on cloud properties. The study shows that introducing MOA as an aerosol species leads to a higher concentration of available CCN and INP, which results in different cloud properties. In contrast to the title of the study, the authors find that MOA has a much larger effect on the CCN concentration and related cloud radiative forcing, relative to INP and associated cloud radiative forcing. Nevertheless, the study does show that adding MOA increases the number of INPs and that MOA-INP are likely the most important INP species over the Southern Ocean, especially at heights below

400 hPa. I would like to commend the authors on a very well written and thorough study. However, I found the implementation of the emission scheme quite confusing and vague. I also have some additional comments below.

General comments:

In regards to the implementation of the MOA emission, it is not very clear how the MOA particles are handled. Firstly, how is the mixing state of the MOA determined (i.e. internally or externally mixed)? Does this just depend on the size mode of the emitted sea salt aerosol? If yes, how does the mass of the emitted MOA impact the resulting size of the sea salt and therefore the mixing state. Secondly, for the externally mixed MOA, how is the size versus number determined? Perhaps I am completely misunderstanding how this is done but please clarify in the methods.

When considering the internally mixed MOA with sea salt, is a freezing point depression considered when using the ice nucleation parametrizations? Based on the mass fractions of MOA relative to sea salt for the majority of the particles, sea salt appears to be the dominant component of the aerosol and I expect this to significantly lower the freezing efficiency of the MOA.

How does the lower hygroscopicity of the MOA relative to sea salt, factor into the available CCN numbers? Perhaps this will become clearer once the number and size of the MOA and its mixing state is explained more clearly.

A major point of the paper is that the addition of MOA greatly improves the representation of INP over the Southern Ocean and to some extent over the Arctic. However, when looking at the comparisons to the field observations, it could be argued that the N12 scheme does better at predicting observed INP concentrations. Therefore, perhaps it is better to compare the influence of adding MOA as an INP to the N12 scheme. I understand that this comparison will not be as straightforward but it seems a bit unfair to say that MOA is an important INP by comparing to D15, which clearly underestimates the observed INP concentrations and as the authors mention, only considers dust particles larger than 500 nm. Nevertheless, I support the author's conclusion that MOA is likely an important INP species over remote regions such as the Southern Ocean. However, I think comparing to D15 may be making MOA out to be more important than it is.

As the Southern Ocean is where the largest changes in INP and CCN are observed, I find it quite surprising how little mention there is of sea ice extent. In theory, and to some degree is seems like some of the reported values in the study show this, the emission of MOA should be greatly reduced during the austral winter and early spring. In fact, I think the handling of sea ice in the model should be more clearly discussed and the seasonal influence on emission of MOA would warrant its own figure (perhaps in the supplement).

Minor comments:

Line 186-192: As I am not so familiar with MAM4, it is unclear to me where the SSA aerosol falls into one of the six aerosol species? Does it count as sea salt and are the cited parametrization used to determine the size distribution of sea salt?

Line 218-223: The G11 scheme requires the input of the chlorophyll concentration. This may show my ignorance but is chlorophyll predicted in the model or is it taken from fixed look up tables? If this comes from look up tables, does it account for seasonal variability and if so, how is the chlorophyll concentration over the Southern Ocean determined during the austral winter when satellite data for chlorophyll is limited to lower latitudes (below ∼55 degrees)?

Section 2.2.2: This section is a bit confusing as it is unclear which method was used here. Is the MOA emitted in the externally-mixed or internally-mixed approach? Based on the authors, it sounds like the Burrows et al, 2018 approach where the MOA is internally mixed should be used. Is that what is done here? If yes, and if the MOA is added to the sea salt fraction, does this not lead to an overall reduction in the hygroscopicity of the sea salt aerosols (or a freezing point depression)? Or is the increase in sea salt

mass due to the MOA fraction make the resulting particles large enough to overcome the reduction in hygroscopicity of the particles to act as CCN? Or does the MOA fraction also increase the number of sea salt+MOA particles. If the latter is the case how is the increase in number concentration handled as the MOA always scales with sea salt mass. Perhaps it would be worthwhile to clarify how the inclusion of the MOA fraction to the different size modes (i.e. Aitken, Accumulation, Coarse, Primary) acts to increase the number and size of the sea salt aerosol and lowers its overall hygroscopicity.

Table 1: Please explain the names of the aerosol species in MAM4. Perhaps these are standard in the modelling community but would be helpful for the reader to know what the acronyms stand for easily. That being said perhaps the same acronyms can be used in Table 1 and 2 for consistency and easy reference.

Section 2.2.2a: Please explain how the TOC is derived for the W15 scheme. Furthermore, is the TOC estimated from the sea surface or derived from the fraction emitted MOA? Later it is stated that it comes from the surface when expaliaing why W15 may be overestimating but please add that here.

Section 2.2.2b: Again here is it not immediately clear how this parametrization is implemented. Is this just dependent on temperature or is the MOA fraction somehow utilized here? Is the derived ns value applied to the total surface area and number concentration of the sea spray aerosol to activate a certain number of particles into ice crystals and if so what aerosol size modes are used? Also, how is the freezing point depression handled. Afterall, the particles are primarily composed of sea salt (at least the internally mixed ones)

Line 396-399: How do these studies justify such low fractions of MOA to sea salt when the laboratory studies report much higher fractions of MOA to SSA in the literature previously cited in the paper (ie. Prather et al, 2013 and Facchini et al, 2008) or are the large contributions of other compounds than MOA and sea salt in SSA? Furthermore, when looking at Figure 2, the fraction of MOA in SSA is much higher than the values reported in this section. Granted Figure 2 appears in a region of high MOA emission, however it is unclear what SSA emission looks like globally. Perhaps it would be worthwhile to plot MOA/SSA emissions as an addition to Fig. S1.

Line 401-406: I find the switch between SSA and sea salt mass rather distracting when discussing the comparison of MOA fractions. Consider making this consistent as to my understanding, SSA is MOA+Sea salt and so it is just a different way of comparing the same values (e.g. MOA/(MOA+Sea salt) or MOA/Sea salt).

Table 5: Perhaps it would be worthwhile to also show the change in mean hygroscopicity of the emitted aerosols as the overall burdens do not seem to change much. Considering some of my comments on section 2.2.2, it would also be worthwhile to see the change in mean number and size of the sea salt and MOA aerosols emitted. I know this greatly varies by region but it would make it clearer to see how adding MOA to the model impacts the number of potential available particles to act as CCN. Also, as previously mentioned, based on the literature discussion, why is the fraction of MOA (MOA/Sea Salt) emission so much lower than reported values of around a few percent for particles larger than 1 micron and even higher for smaller particles?

Line 462: Consider revising to state that impact on clouds via CCN will be discussed next as the INP section follows section 3.2

Figure 3: Is there a reason why such a decrease is observed over the Tibetan plateau? Is there a way to add hatching for regions where the changes between simulations are significant or are these changes significant because they are the averages over the nine years?

Line 514-515: I would argue that the N12 parametrization does the best job of predicting the INP concentrations across the entire temperature range as shown in Figure 4. This is mischaracterized in these sentences. In fact based on the ability of N12, it would be possible to argue that including MOA emission is not needed to accurately predict the observed INP concentrations.

Line 521-523: Following up in the previous comment, the fact that combining M18 and D15 still under predicts, shows that the MOA addition is not as good as just using the N12 parametrization. And one could argue that when using the entire size distribution for dust nucleation, the majority of INPs would accurately simulated.

Figure 5: How are the parametrizations drawn for the flight campaigns (e.g. Socrates), where INP measured aboard the aircraft are sometimes collected at different altitudes?

Line 540-551 and Figure 6: Why was INPs at -25 C and 950 hPa used for this analysis here? For cloud formation, this seems highly irrelevant as it is rarely -25 C at 950 hPa especially in the regions where MOA is expected to be important (over ice-free regions of the Ocean). In fact, how common is it for MPCs to exist at this height. This seems like an unfair height for showing the importance of MOA as INPs as it is a height where MOA concentrations are extremely high due to being within the boundary layer and at a temperature where the ability of MOA to freeze is essentially maximized based on field measurement techniques used at temps above ∼-30 C.

Line 552-561: I generally agree with the explanation for the observed differences shown in Fig.6d. However, how does the model handle the sea ice coverage over the Southern Ocean during Austral winter? As the sea ice extent should extend as far north as approximately 60 S. Therefore, it important to know how the model handles the emission of MOA during the austral winter and spring months, especially as the sea ice extent is prescribed based on climatology (see lines 354-356). Does this mean that the entire year assumes a constant sea ice extent or does it change based on season. Depending on how this is handled, it could have large implications for both the INP and CCN distribution due to MOA emissions.

Figure 7: How does the model produce ice nucleation or even MOA at such high latitudes in the Southern Hemisphere at the surface when the center of the Antarctic ice sheet is ∼3000 m (700 hPa)?

Line 579-581: Mentioning seasonal dependence is quite interesting especially concerning my previous comment about the sea-ice extent. Therefore, I think it would be worthwhile to show how the emission of MOA changes over the Southern Ocean between austral summer and winter and how this influences the freezing rates.

Line 590-592: What does this increase in percent mean? Can you report the change in the number of CDNUMC? Also the fact that there is a difference between austral summer and winter might point to a change in the sea ice extent in the model as well as biological activity.

Line 599: why do you switch to an isotherm of -15 now? Previously the -25 isotherm was used and the figures also have the -20 isotherm. Perhaps it is better to be consistent and choose one isotherm throughout or at least explain why different isotherms are chosen.

Line 628-630: Could this also be due to sea ice extent?

Line 655-661: Discussion on the differences between bubble bursting (which is implemented in the model) and jet drops (which is not) does not seem necessary. Perhaps it is just fine to just mention that more observations/ fundamental understanding are needed for implementation of jet drops as is not clear why the differences in size of the emitted aerosols matter here. If this is important, please expand on why that could make a significant difference to the observed results and overall importance of MOA.

Editorial comments:

Line 47: “replace to” with “and” as the sentence should read: “temperature is between -38 and 0...”

Line 49: Please consider rephrasing the sentence to read: “INPs have different characteristics depending on their composition and origin” as it does not make sense as it is written.

Line 52: Consider adding some citations when you mention the uncertainty in the ability of black carbon to act as INPs. To my understanding, the evidence is mounting that BC

is irrelevant in the MPC region.

Line 74: Please add Ault et al, 2013 to the reference list.

Line 123: Please add “method” or something similar after: “[Chl-a]-based”

Figure 1: Fix unit for micro

Figure 3: The 0.1 % supersaturation is not showing up well. Also, the longitude representation (248 W) seems a bit odd, but perhaps that’s just a personal preference. In the Figure caption, it might be nice to state that percent change in surface CCN comes from comparing B14-BASE.

Figure 4: Consider flipping the color bar so that the warmest temperatures are at the top and the coldest at the bottom.

Figure 6: Please fix panel d to be consistent with the other panels.

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