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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Contribution of feldspar and marine organic aerosols to global ice nucleating particle concentrations" *by* Jesús Vergara Temprado et al.

Anonymous Referee #2

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The manuscript "Contribution of feldspar and marine organic aerosols to global ice nucleating particle concentrations" by Vergara-Temprado et al. 2016 investigates the question if an aerosol specific freezing parameterization scheme using feldspar as a terrestrial source of INP and marine organic aerosols as a marine source of INP can better represent the overall global INP distribution in comparison to simple non aerosol specific freezing schemes. They retrieve better results with their scheme compared to earlier easier schemes. Additionally, they investigate the role of the two species used as INP (feldspar and marine organic aerosols). They show that feldspar dominated the global INP population. Nevertheless, they demonstrate that marine organic aerosols are an important INP species, especially over the Southern ocean. The manuscript is

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Discussion paper

well written. However, some of the analysis needs some further testing and possible improvements. It should be published after major revisions.

Major remarks:

- The argumentation in the abstract and in the introduction is partly not convincing and should be explained a bit clearer or rephrased. Specifically the authors claim that because of a difference in terrestrial and marine INP concentrations, INP species specific parameterization schemes are needed instead of schemes that predominantly stem from terrestrial sources. But if a scheme was developed for terrestrial sources and does not account for marine sources, the INP concentration represented in the model would be different for marine and terrestrial sources as well (it would be smaller above marine regions as shown by the fiels observations). Maybe the argumentation should be split up in two aspects: 1.) Why is it important to account for aerosol species in a freezing parameterization scheme (in general), 2.) Why is it important to also include marine sources? Additionally, the argumentation about the underprediction of the persistance of supercooled clouds oder the Southern ocean and the connection to low INP concentrations (page 2, line 20) could be explained better- is the hyposesis that models overestimate INP concentrations over the Southern ocean which leads to a faster glaciation of the clouds? Can you add references for this hypothesis, e.g. showing an overestimation of INP of models over the Southern ocean?

- Some statements of the singular approximation (in comparison to CNT) sound missleading: you write that the time-dependence is of secondary importance compared to the particle-to-particles variability in case of the singular approximation. When using a simple ns-approach, with one set of fit parameters for one species the particle-to-particle variability is also not really considered. Instead of using an average (single) contact angle for one particle population, an average (single) value for the density of active sites for the particle population is used. I do not see where and how the particle-to-particle variability is better represented in the ns-scheme compared to CNT.

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- It is not always clear what kind of model output is used for the analysis. While Fig. 7 seems to be based on daily values, Fig. 8 seems to be calculated using annual means (of n_aer,0.5 and probably also the size of the dust particles for the Niemand scheme). Using annual means for the calculations of the INP concentrations could be meaningless. Freezing is very sensitive to variability in temperature etc.. The INP concentrations should be calculated on a model timestep level and then averaged. If that is already done like this in the manuscript, please explain the methodology better. If it is not done like this, the methodology should be thought through again. It should be shown for one example at least that using annually averages does not influence the result.

- The way the global INP dataset is used and the results are analysed can lead to biases, because it is not used in a uniform way for all parameterization schemes. There are three aspects one could investigate using the dataset, but depending on the aspect the use of the dataset should be different:

1.) Evaluating the parameterization schemes:

To evaluate how well a specific parameterization scheme represent the INP conc. the simulated values should be compared to the observed values only within the valid temperature range of the parameterization scheme. That is what was done in this study. However, that does not tell one how good the parameterization scheme works in a model context where it is used over the whole temperature range (see 3.)).

2.) Comparing the "ability" of the different parameterization schemes within each other:

If one would like to compare how different parameterization schemes compare to each other, the comparison should be done for the same temperature range (in this case the smallest defined temperature range of the parameterization schemes). If they are compared not using the same temperature range it could be that the result does not only show the difference of the parameterization schemes but also other aspects, e.g. one parameterization schemes lacks the INP in high temperature regimes, where another

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scheme is not defined (and therefore the R^2 is not affected). Using different temperature ranges could lead to a bias towards the scheme with the best defined validity temperature range. E.g. looking at the comparison done in this study, the DeMott et al. 2010 scheme would achieve a much better score if the temperature range between 0 and -4°C would not be taken into account.

3.) Evaluating the model performance:

Finally what is interesting in a model context is how good a specific parameterization scheme is able to represent the global INP concentrations. Also if a parameterization scheme is only defined for a certain temperature range the INP concentration has to be simulated for the whole temperature range. In the presented scheme that means that the INP conc. is 0 above -6°C. If one would like to evaluate the performance of a model using this scheme also the INP conc. above -6°C have to be compared to the simulated one (in this case the simulated conc. being 0).

This manuscript shows aspect Nr. 1, but does not really evaluate the other aspects in a correct way. It is reasonable to define parameterization schemes only for a specific temperature range, but is has to be considered that the schemes are later on in a model context used over the whole temperature range and should give reasonable results for the whole range (also if they are not extrapolated).

Minor remarks and typos:

- Page 1, line 4: Remove space before . .

- Page 2, line 22: "A poor representation ... is important..." sounds missleading.

- Page 2, line 29: Is it proven that freezing is a main model bias?

- One name is missspelled in one citation: Instead of Schenell and Vali 1975, it has to be Schnell and Vali 1975.

- Page 3, line 6: You could add more references here.

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- Page 4, line 6: Replace ";" by "and".

- Page 4, line 9: Is it Pseudonana instead of Psuedonana?

- Page 4, line 16. Add . after citation.

- Page 4, line 20: Please state which other studies.

- Page 5, line 2: Skip "major" (you do not know if that are the two major sources).

- Page 5, line 14: What do you mean by saying the clouds are assumed to glaciate at -15°C?

- Page 5, line 27: Please elaborate how large the difference would be in case of different types of feldspar compared to the difference between soil/aerosolized feldspar fraction.

- Page 6, line 3: Remove "a".

- Page 6, line 13: Add . after bracket.

- Page 6, line 18: The OMF parameterization does not cause uncertainty? Or why is this not mentioned?

- Page 6, line 29: It also has physical reasons why WIOM depends pos. on chlorophyl and neg. on wind speed. How you write it, it sounds like this is only due to fitting the observations. Please rephrase and maybe elaborate with 1-2 more sentences.

- Page 7, line 4: Add . after bracket.

- Fig. 1: You could color the errorbars in the same color as the data points to make it easier to differentiate the two locations, especially where WIOM is small.

- Fig. 2: I do not understand the unit of the variable plotted here (or the variable)- is it the accumulated mass of sub-micron marine organics over the whole column?

- Page 8, line 12: Add an "a" after "within".

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- Page 9, line 22: Higher in the cloud refers to which temperature? Maybe you could explain that a bit more, it might noch be obvious for every reader.

- Fig. 3: What does the color scale mean next to [INP]_T?

- Page 10, line 1: The reference has to be Figure 4 not 4b.

- Fig. 4: Did you also plot this figure for a different height to check if the picture would then look different? E.g. it could be that the dust distribution is more "present" in the lower figure for a different height. That would be an interesting aspect to look at and mention in the manuscript.

- Fig. 4: Does the lower figure indirectly shows that the temperature in the Arctic is always below -20°C at 600 hPa?

- Page 12, line 3: You should explain why you chose an activation temperature of -15°C, that is quite low for the surface (where you want to simulate the INP conc.).

- Page 12, line 5: Add "dust" in front of "sources".

- Page 12, line 6: Put brackets around "5 a".

- Fig. 5: Does it make sense to use the surface concentration for this plot? Wouldn't it be more reasonable to do the simulations at a higher altitude?

- Fig. 5a: What is the white spot in the plot (bottomleft)?

- Fig 6: Add a label to the colorscale. Which variable is plotted?

- Page 14, line 1, 4 and 5 and caption Fig. 6: You plot seasons and not separate months- adapt the wording.

- Page 14, line 4: Add a space between "Fig." and "6".

- Page 14, line 18: More consistent with what?

- Page 14, line 22: Add brackets around "Fig. 6".

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- Fig. 6: It would be more consistent with the following analysis if you would give the INP conc. in 1/l instead of 1/m^3.

- Fig. 6: Instead of the black contour lines you could also display two plots next to each other, that is maybe better readable. In the second plot the labels of the contour lines are difficult to read (overlap).

- Fig. 7: Why do you have values in the temperature range below -26°C?

- Fig. 7: Especially in the third plot there are INP values even below -40°C- you should explain these "artefacts" or whatever it is.

- Caption Fig. 7: Add a space between label "ambient" and "concentration" (line 2).

- Page 17, line 1: Other schemes indirectly capture the source since large particles sediment and are more predominant close to the source region. Why is only a species-differentiating scheme able to capture variations and long-term trends?

- Page 17, line 6: Add a space between "Table" and "1" (remove the . or write Tab.). Add a space between "Fig." and "8c".

- Page 17, line 10: There is no improvement shown in Tab. 1 (the unscaled values or not shown)? Eather add it in the table, or remove the reference to the table.

- Page 17, line 23: Add brackets around "Tab. 1".

- Table 1: Why is the correlation coefficient calculated for the logarithm of the values? Please explain shortly in the manuscript.

- Page 18, line 10: Since you do not know if preferential INP in-cloud removal is important you should change "are" to "could be". Same in line 11 for the terrestrial source of INP.

- Fig. 8 f is not mentioned in the text. Is this figure necessary? It would need some further explanation to be easy understandable.

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- Fig. 8 and Fig. 9 and Fig. 10: Axis labels etc . are quite small font.

- Fig. 8: Label b is truncated.

- Fig. 8 label: Add which simulated and observed variable it is.

- Fig. 8 caption, line 5: Remove one ".".

- Fig. 8 caption, last line: Add a ".".

- Fig. 9 caption, line 2: Add a bracket after "a".

- Page 21, line 24: What kind of measurements would be needed? It would be helpful to elaborate that in 1-2 more sentences.

- Page 22, line 26: Please explain this formula a bit more.

- Fig. 11: Are that yearly mean values or for which time period is the comparison/relation plotted?

- Fig. 11 b) is not explained.

- Fig. 11, caption: add space between the fit parameters. Add a "." at the end of the caption.

- Appendix B: How do you get from Eq. B2 to B3?

- Page 26, line 1: Do you refer to size distribution when you write "distribution"?

- Appendix B: What does the last section mean in your model context?

- Table 2: Remove brackets around the references.

- Table 2: Are the references unpublished where you did add the label "BACCHUS"? Otherwise I do not understand why this is labeled like this and what it means.

General remarks:

- The citations are not done consistent- sometimes brackets are used where there

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should not be, sometimes brackets are missing, e.g. at page 3 line 15 brackets are missing vs. at page 2 line 34 brackets should be removed. Please thoroughly go through the citations again.

- Please add a space between numbers and units, e.g. page 5 line 7: 10 hPa.

- Units should not be italic, e.g. page 6 line 5.

- The naming of the modell is not consistent throughout the paper, sometimes you write GLOMAP, sometimes GLOMAP-mode. This should be explained (if the names are different on purpose) or made consistent.

- Reduce the space between the single letters within your variables INP and ff, that increases the readability.

- Be consistent with writing OMF as a variable in italic or not, e.g. page 24, line 3.

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