Response to comment #1

We would like to thank the reviewer for their useful and constructive comments. Our response and the subsequent modifications to the paper are structured as follows:

Blue for the reviewer comment

Normal text for our answers

Bold for the changes in the manuscript

-Page 2, lines14-19: I think there are some contradictions in these sentences: The authors mention the parameterization by DeMott (2010), which is dependent on the INP concentration (greater than a certain size). Then shortly after it is stated that, "However,studies have shown that cloud are sensitive to INP concentrations. " This is already included in the DeMott (2010) parameterization. What DeMott (2010) does not take into account is the variation in nucleation properties (as stated to be an important factor in the previous sentence). I think the sentences on these lines need to be rephrased.

Further, the sentence on line 29-30 is almost identical to the sentence on line 17-18.

We have rephrased these lines to clarify that these parameterizations currently do not represent the differences in the ice nucleating properties of different aerosol species. We also deleted the last sentence.

The current representation of heterogeneous freezing in climate models and operational numerical weather prediction models is usually based on parameterizations that depend on the temperature (Young et al. 1974, Meyers et al. 1992) or the size distribution of aerosol particles as well as the temperature (DeMott et al. 2010). These parameterizations treat aerosol particles all around the globe and across seasons as having the same ice-nucleating properties irrespective of the aerosol chemical composition. Representing these differences may lead to a better simulation of INP concentrations, thereby improving the representation of mixed-phase clouds.

-Page 3, line 12. I suggest including references to Marcolli et al. (2007) and Eidhammer et al. (2009), who also included distributions of contact angles in their studies.

References added.

-Page 3, line 15-16 and many other places: There are many citations where the parentheses are misplaced, such as for the Vali et al. (2015) citation. Here it should be "..approximation (Vali et al. 2015) in which the time …" Other places, such as page 5, line 6, it should be "…model described in Mann et al. (2010). " Please go trough the manuscript and fix all misplaces parentheses.

Done

-Page 3, line 33. I suggest including reference to Koehler et al. (2010), which also conducted studies of the ice nucleation ability of dust.

Done

-Page 3, line 32: I am confused by this sentence: Atkinson et al. (2013) found that a mineral component of desert dust, is responsible for most of ice nucleating activity of mineral dust aerosols. Should it be "..activity of desert dust aerosols"?

This has been rephrased to: **Atkinson et al. (2013) found that K-feldspars are far more effective at nucleating ice than any of the other minerals in desert dust.**

Page 4, line 1: What does "this type of mineral" exactly refer to?

Refers to K-feldspar, we have rephrased this sentence

Therefore the representation of K-feldspar in atmospheric models…

-Page 5, line 13-14: By Nucleation scavenging is suppressed for ice clouds, is it meant that it is not included, meaning that the ice nucleation parameterization is only based on temperature and not INP concentration. This should be explicitly stated. Also, by stating assumed to glaciate at -15C, is it implied that below -15C, the clouds comprise only of solid hydrometeors, and not mixed?

This refers to the model assumptions that we need to make in order to represent nucleation scavenging of aerosol particles in our model. As we are using a chemical transport model, aerosols do not interact with clouds in any way (other than being scavenged by precipitation) and all the meteorological fields (including cloud fields) come from ECMWF reanalysis. With these fields, we can predict the concentrations of different aerosol species and from the concentrations we calculate offline (after the model simulation) the INP concentrations. The discussion of the nucleation scavenging assumptions is included in Browse et al (2012), so we refer to it for a more detailed description.

Nucleation scavenging is suppressed for ice clouds, which are assumed to glaciate at -15ºC. A discussion of the nucleation scavenging assumptions in our model is included in Browse et al. (2012).

-Page 5, line 21: using “accurately” by stating that the model has been shown to reproduce dust concentration accurately is a strong statement. I suggest rephrasing/rewording.

Reworded to ‘within an order of magnitude’

The model has been shown to reproduce dust mass concentrations within an order of magnitude

-Page 14, line 1: Are Figure 6 zonal averages?

Yes. Clarification has been added to figure caption

-Page 14: line 12: Please give a range for the mixed-phased range.

Done, added: “(0°C to -37°C)”

-Page 22, EqA1. I suggest moving Eq.A1 up to line 1, page 22, where the equation is first mentioned.
Done

Technical comments
Page 1 line 2: Replace “of their properties” to “of the cloud properties”
Done

Page 1, line 15: replace “
. . .
Southern Ocean at some time of the year” with
“
. . .
Southern Ocean at some part of the year”
Done

Page 2, line 10: remove “other”
Done

Page 2, line 27: “In future” should be “In the future”
Done

Page 4, line 6: Replace “;” with “and”
Done

Page 5, line 7: Suggest replacing “resolution“ with “gridspacing”
Done

Page 6, line 3: Southern Ocean is “a” . Remove “a”
Done

Page 6, line 34: Include: “
. . .
..based parameterizations such as in Rinaldi et al. (2013)
and Gnatt et al. (2011) but scaled
. . .
”

Done

Page 8, line 12: I suggest replacing potassium feldspar with K-feldspar for consistency.

Done

Page 12, line 6: 5a should be in parenthesis.

Done

Page 14, line 22: Fig. 6 should be in parenthesis.

Done

Page 22, line 11 and 12. Missing parentheses before Fig.11a and Fig.11 b

Done

Page 25, line 6: Replace Where with Here.

Done

Page 25, line 13: switch : “be therefore” with “therefore be

Done

Response to comment #2

We would like to thank the reviewer for their useful and constructive comments. Our response and the subsequent modifications to the paper are structured as follows:

Blue for the reviewer comment

Normal text for our answers

Bold for the changes in the manuscript

- 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 field 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?

The referee's logic is correct, up to the point where they state "*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 field observations)*." This is not correct. The parameterisations in the literature we refer to simply treat all aerosol identically in all locations or simply assume a temperature dependent concentration of INP. Hence, for example, the Meyers scheme 'predicts' the same INP spectrum over the ocean as over the land. We stress, that we have developed a global model of INP concentrations using a global aerosol model, hence can link aerosol specific INP properties to specific aerosol species.

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?

A discussion of this hypothesis is included in DeMott et al. (2016). Additionally Figure 8 (a,b,c) shows an overestimation of INP in marine environments (triangles) when using 3 commonly used parameterizations.

In order to clarify the arguments here we have extended the discussion:
“Over the Southern Ocean clouds tend to persist in a supercooled state more commonly than models predict (Bodas-Salcedo et al., 2014), which might be related to very low INP concentrations in this region.”

To:
Over the Southern Ocean clouds tend to persist in a supercooled state more commonly than models predict (Bodas-Salcedo et al., 2014), which might be related to very low INP concentrations in this region (Bigg et al. 1973; DeMott et al. 2016). It has been shown that less INP in the Southern Ocean lead to less ice and more supercooled water in model clouds, with a significant impact on the radiative properties of the clouds (Tan et al. 2016).

- 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.

The referee is incorrect in the statement that “average (single) value for the density of active sites for the particle population is used”. The parameterisation is a temperature dependent function describing the cumulative density of active sites which become active on decreasing temperature. In this model, a specific site has a characteristic temperature at which it nucleates ice and each particle has a distinct population of active sites. If we applied an average density of active sites, then all particles that possessed that site would trigger freezing at the same temperature. The cited experimental work shows that materials have a spectrum of nucleation sites. Assuming a single contact angle also means that each particle (of the same size) has the same probability of nucleating ice and according to classical theory, nucleation will occur over a narrow range of temperatures. Experimental work suggests that this is not the case and that there is a distribution of sites.

A very important distinction between using a single contact angle parameterisation and the singular description is that when using classical nucleation theory with a single contact angle, eventually all your aerosol particles will freeze, as time dependence is the main factor that drives nucleation. That is in contrast with the singular description and many laboratory studies that show how just a fraction of particles nucleate. This topic has been widely discussed in the past so, and we have referred the interested reader to Appendix 2 and the references therein for a more detailed discussion.

We have improved the clarity of the discussion here by changing: “The ice nucleating efficiency using the singular description is defined by a density of active sites, which is a function of the temperature and usually of the surface area (ns),”

To **“The ice nucleating efficiency using the singular description is defined by a temperature dependent density (i.e. per unit surface area) of active sites, (ns(T) which represents a spectrum of active sites with variable characteristic ice nucleation temperatures. The temperature dependent number of active sites can also be normalised to another parameter characteristic of the aerosol population (such as mass or volume) (Murray et al., 2012). From this density of active sites, one can calculate what fraction of the particles will nucleate ice at a certain temperature (See Appendix:2)”**

- 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.

Figure 7 was calculated using daily values of the temperature and concentration in order to account for the large temperature dependence of the simulated INP concentrations. Figure 8, however, does not depend on the modelled temperatures, as the temperature used to calculate INP is that corresponding to each observation and this is independent of the ambient local temperature. In order words, for an observation at a temperature T1, we calculated the predicted INP concentration with the annual mean concentrations of aerosols given by our model and the temperature at which the observation was done (T1).
We have modified the figure caption in order to clarify this concept.

For each individual observation, we calculated the INP concentration at the temperature corresponding to the temperature that aerosol particles were exposed to in the INP instruments.

- 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 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).

We have done some changes to address this comment.

First, we have included in figure 8 the datapoints outside the temperature range of the different parameterizations with semitransparent markers. This is done in order to have a visual comparison of how the parameterizations will look like if they are extrapolated.

Then, we have added in Table 1 the same statistical values as before, but also for the other two aspects (all the temperature range and just for the shared temperature range).

With these changes, we think that the 3 main aspects are addressed. Overall. The changes are very minor to the plots, with relatively few data points being added.
We added in the text:

When the parameterizations are extrapolated outside their temperature range, they still perform similarly.

Looking at the performance of the different ways of representing INP within the smallest temperature range shared by the all the parameterizations (-12 to -25C), our representation of INP is able to reproduce 61.6% of the datapoints within and order of magnitude and 78.7% within 1.5 orders of magnitude. These values are greater than the obtained when using the other 3 parameterizations used for this study (Table.1)

The caption of Table 1 has changed as well:

Statistical performance of the different parameterizations. Pt1 and Pt1.5 are the percentages of datapoints reproduced within an order of magnitude and 1.5 orders of magnitude in the temperature range of every parameterization. The number of datapoints used for calculating these values is shown under the 'Datapoints' column. The values with * show the same calculation but including

datapoints outside the temperature range of the parameterizations. These values give an idea of the performance that you would expect if you extrapolate the parameterizations in a climate model. The values with ** are for datapoints within the smallest temperature range shared by the 4 parameterizations (-12C to -25C). The correlation coefficient has been calculated with the logarithm of the values as INP concentrations vary logarithmically with temperature.

Minor remarks and typos:

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

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

A better representation of mixed-phase clouds in climate models has been shown to be important for climate prediction.

- Page 2, line 29: Is it proven that freezing is a main model bias?
It has been proven that freezing is poorly represented in climate models (see McCoy et al., 2015 figure 1). This reference is given in the text.

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

- Page 3, line 6: You could add more references here.
Added Sesartic et al (2013) and Lohmann et al. (2006)

- Page 4, line 6: Replace ";" by "and".
Done
- Page 4, line 9: Is it Pseudonana instead of Psuedonana?
Yes, corrected

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

- Page 4, line 20: Please state which other studies.
The other studies are cited in the following sentences, I have rephrased the sentence to connect it with the following sentences.
Further evidence for the biological origin of marine INP is the heat sensitivity of some types of organic INP, i.e. the temperature at which they nucleate ice is reduced after heating to 100C (Wilson et al. 2015, Schnell et al. 1975, Schnell et al. 1976).

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

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

As we are using a chemical transport model, all the meteorological fields are obtained from ECMWF, including clouds, and our aerosols do not feedback in clouds. Because of this reason, we have to assume a temperature for representing in-cloud scavenging of aerosol particles in ice and liquid clouds. A more detailed evaluation of this assumption as well as a detailed description of the in-cloud scavenging scheme is described in Browse et al. (2012).
We have inserted:
'A discussion of the nucleation scavenging assumptions in our model is included in Browse et al. (2012)'

- 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.

Most k-feldspar samples have ice nucleating abilities that agree with each other within a factor of 6. This factor is substantially larger than a factor of 2.

…ice nucleating ability of K-feldspar such as differences in the density of active sites of different types of K-feldspar (around a factor of 6) (Harrison et al. (2016).

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

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

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

…processes, or model grid and temporal resolution, as well as uncertainties related to the organic mass fraction parameterization.

- 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

A more in deep explanation of the dependence of WIOM with windspeed is included in Gantt et al (2011) (figure 1). We have rephrased this section to clarify

The development of our new organic mass fraction parameterization, explained in detail in Appendix A , assumes that the organic mass fraction of the sea-spray particles depends on wind speed and the chlorophyll content of seawater. The organic emission parameterization includes a positive dependence of WIOM mass fraction on chlorophyll (O'Dowd et al., 2015; Rinaldi et al., 2013; Gantt et al., 2011), but a negative dependence on wind speed.Thus, the WIOM is essentially diluted in the sea spray particles when the total sea spray emission flux is high, which may be caused by a limited supply of organic material in the surface ocean but effectively limitless salt (Gantt et al 2011). This parameterization is similar to previous chlorophyll based parameterizations such as Rinaldi et al.(2013) and Gantt et al (2011) but scaled in order to fit the observations in Amsterdam Island and Mace Head when applied in our model.

.

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

Done

- 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.

Done

- 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?

No, it refers to the concentration of sub-micron marine organic aerosol mass at the surface. We have changed the description

Annual mean mass concentration of sub-micron marine organic (μg m^{-3}) aerosol at surface level

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

Done

- 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.

Added the range of temperatures

Hence, when considering a deep convective cloud where air is moved vertically through all the mixed-phase range of temperatures…

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

It is an example color scale referring to temperatures decreasing from 0 to -37C.

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

Changed from Figure 4b to Figure 4 (bottom)

- 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.

Figure 5a shows a similar picture but for surface level. A comparison of the influence of both marine organic and K-feldspar for all heights is done in figures 6 and 7.

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

No, the bottom panel shows the annual mean INP concentration active at local ambient temperature.

- 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.).

It is a temperature at which many atmospheric observations of INP are made, so they could be compared with what this paper predicts. We stress, that $[INP]_{15}$ is independent of local ambient temperature.

In the figure caption we have added a statement:

We show $[INP]_T$ for a *T* of -15°C because this is a temperature used by many instruments. The number of INPs that activate to ice crystals ($[INP]_{ambient}$) at the surface will be zero over much of the globe, because these particles will only

become important at high altitudes. Surface concentrations are show because this is where most observations of atmospheric INP concentrations are made.

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

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

- 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?
We use the surface concentration as it is where most INP observation are made. Figure 6 show the vertical profiles of INP ambient.

We have addressed this in the caption of Fig 5 (see above).

- Fig. 5a: What is the white spot in the plot (bottomleft)?
It was a concentration range that was outside the colorbar range. Now it is corrected.

- Fig 6: Add a label to the colorscale. Which variable is plotted?
The description of the variable plotted ($[INP]_{ambient}$) is defined in the caption of the figure. We have added it to the colorscale label

- Page 14, line 1, 4 and 5 and caption Fig. 6: You plot seasons and not separate months- adapt the wording.
We have changed the 'monthly' to **'seasonal'**

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

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

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

Done

- Fig. 6: It would be more consistent with the following analysis if you would give the INP conc. in 1/l instead of 1/m3.

We prefer to keep the units in 1/m3 in figure 6, as using 1/l would make the numbers too small affecting the quality of the image as the plot becomes too messy.

- 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).
We have modified the plots to avoid overlap between the labels of the contour lines, but would prefer to maintain one plot since it makes the comparison more direct.

- Fig. 7: Why do you have values in the temperature range below -26◦C?
The concentrations at temperatures colder than the limit of the parameterizations are set as the value at the limiting temperature as explained in Page 11,line 4:

“The concentrations of [INP]ambient at temperatures colder than the temperature limit of the parameterizations (for K-feldspar: -25oC and marine organics: -27oC) is set at the value defined by the concentration at the limiting temperature of each parameterization. This is consistent with studies that caution against extrapolating singular parameterizations outside the range where measurements were made.”

- Fig. 7: Especially in the third plot there are INP values even below -40◦C- you should explain these "artefacts" or whatever it is.
The black lines in Figure 7 represent seasonal mean isotherms. Some of the values are below those lines because of day-to-day temperature variability.
We have rephrased the caption to clarify that they are seasonal mean isotherms.

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

- 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?
Because variations in aerosol emissions could be different for different aerosol species. This will imply that particles with very different ice nucleating abilities will be emitted in different amounts and hence the change in the INP concentrations will not necessarily be proportional to the change in the total emitted aerosol amount. We have rephrased this sentence to clarify the concept.

…so they may not capture variations and long-term trends since different aerosol types have different ice nucleating abilities.

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

- 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.
The reference refers to the value of the correlation coefficient.

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

- Table 1: Why is the correlation coefficient calculated for the logarithm of the values? Please explain shortly in the manuscript.
It is calculated with the logarithm of the values as they vary logarithmically with temperature. Explanation added to caption.

The correlation coefficient has been calculated with the logarithm of the values as INP concentrations vary logarithmically with temperature

- 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.
Done

- Fig. 8 f is not mentioned in the text. Is this figure necessary? It would need some further explanation to be easy understandable.
There was a mistake in the text. In page 17 line 24, where it says Figure 8g it should say Figure 8f. There is where the figure was mentioned. It is been solved now that figure 8 has been divided in 2.

- Fig. 8 and Fig. 9 and Fig. 10: Axis labels etc . are quite small font.
We have increased the font of the figures and divided figure 8 into 2 different figure so it improves its aspect.
- Fig. 8: Label b is truncated.
Checked

- Fig. 8 label: Add which simulated and observed variable it is.
Added [INP]

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

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

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

- Page 21, line 24: What kind of measurements would be needed? It would be helpful to elaborate that in 1-2 more sentences.
Expanded:
In addition, more measurements in the ambient atmosphere for different environments and seasons are necessary to better evaluate and constrain models. Among those, exploratory studies about the composition and type of ice nucleating particles in terrestrial environments at high temperatures will be crucial to determine which species need to be included in models.

- Page 22, line 26: Please explain this formula a bit more.
The derivation of the equation has been added

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

- Fig. 11 b) is not explained.
We have modified the caption to include the required information:
a) OMF compared as a function of chlorophyll-a content and surface wind speed for the monthly mean values across the year in both stations. The size of the points represent the mean chlorophyll-a content of the grid-boxes related previously to every station (Fig.10), the colour of the points is related to the wind speed of those grid-boxes. b) Shows the performance of the parameterization for reproducing the OMF calculated with the simulated concentration of sub-micron sea-salt and the observed values of WIOM.
- Fig. 11, caption: add space between the fit parameters. Add a "." at the end of the

caption.
Done
- Appendix B: How do you get from Eq. B2 to B3?
By adding f(0,λ)- f(0,λ) (equals 0) into the right side of the equation. The first f(0,λ) goes inside the sumatory (note the change in the stating value of the sumatory).

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

- Appendix B: What does the last section mean in your model context?
It means that for aerosol species with small number of active sites per particles (λ<0.1 always) we can reduce the complexity of the calculation. This method is used for marine organic aerosols as stated in Page 9 line 4

- Table 2: Remove brackets around the references.
Done

- 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.
The dataset was obtained from the BACCHUS project. The values are published, but the data was taken straight from the dataset.

We added a clarification:
The datasets obtained through the BACCHUS project database are labelled as "BACCHUS" in table 2.

General remarks:
- The citations are not done consistent- sometimes brackets are used where there 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.
We have gone through that again. This was an issue with transferring text between latex and word.

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

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

- 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.
Checked.

- Reduce the space between the single letters within your variables INP and ff, that increases the readability.
Done
- Be consistent with writing OMF as a variable in italic or not, e.g. page 24, line 3.
Done

~~Jesús Vergara Temprado~~

Contribution of feldspar and marine organic aerosols to global ice nucleating particle concentrations

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Abstract. ~~Ice nucleating~~ Ice-nucleating particles (INP) are known to affect the amount of ice in mixed-phase clouds, thereby influencing many of their properties. The atmospheric INP concentration changes by orders of magnitude from terrestrial to marine environments, which typically contain much lower concentrations. Many modelling studies use parameterizations for heterogeneous ice nucleation and cloud ice processes that do not account for this difference because they were developed based on ~~measurements predominantly from~~ INP measurements made predominantly in terrestrial environments. Errors in the assumed INP concentration will influence the simulated amount of ice in mixed-phase clouds, leading to errors in top-of-atmosphere radiative flux and ultimately the climate sensitivity of ~~climate models~~the model. Here we develop a global model of INP concentrations relevant for mixed-phase clouds based on laboratory and field measurements of ice nucleation by K-feldspar (an ice-active component of desert dust) and marine organic aerosols (from sea spray). The simulated global distribution of INP concentrations based on these two-species agrees much better with currently available ambient measurements than when INP concentrations are assumed to depend only on temperature or particle size. Underestimation of INP concentrations in some terrestrial locations may be due to neglect of INP from other terrestrial sources. Our model indicates that, on a monthly ~~or yearly~~ average basis, desert dusts dominate the contribution to the INP population over much of the world, but marine organics become increasingly important ~~in the world's~~ over remote oceans and ~~can dominate in~~ they dominate over the Southern Ocean~~at some time of the year. Furthermore, we show that~~. However, day-to-day variability is important~~and since~~. Because desert dust aerosol tends to be sporadic, marine ~~organics~~ organic aerosols dominate the INP population on many days per month ~~in~~ over much of the mid and ~~high latitude~~ high-latitude northern hemisphere. This study advances our understanding of which aerosol

species need to be included in order to adequately describe the global and regional distribution of INP in models, which will guide ice nucleation researchers on where to focus future laboratory and field work.

1 Introduction

In the absence of aerosol particles which can act as ~~ice nucleating~~ ice-nucleating particles (INP), liquid water droplets can supercool to temperatures below -37^oC (Riechers et al., 2013; Herbert et al., 2015). It is well-known that ice formation frequently occurs at much higher temperatures in many clouds~~around the globe~~, indicating that INP are ~~present to a greater or lesser extent depending on the location and the aerosol properties~~ prevalent in the atmosphere (Choi et al., 2010; Rosenfeld et al., 2011). In supercooled and mixed-phase clouds (containing ice and water) INP cause clouds to glaciate, which leads to changes in many cloud properties such as cloud lifetime, their radiative effect on the atmosphere, and the formation of precipitation through the Wegener–Bergeron–Findeisen process (Murphy and Koop, 2005; Korolev, 2007) and possibly ~~other~~ cloud ice multiplication processes (Hallett and Mossop, 1974). In the mixed-phase cloud regime the dominant freezing mechanism is thought to be through INP that are immersed within cloud droplets, known as immersion freezing (Westbrook and Illingworth, 2011; Field et al., 2012; Murray et al., 2012). Hence, this is the pathway we focus on in this study.

~~The current representation of heterogeneous~~ Heterogeneous freezing in climate models and operational numerical weather prediction models is usually based on parameterizations that depend on the temperature (Young, 1974; Meyers et al., 1992) or the size distribution of aerosol particles as well as the temperature (DeMott et al., 2010). ~~However, these~~ These parameterizations treat aerosol particles all around the globe and across seasons as having the same ~~ice nucleating properties . However, studies have shown that clouds are sensitive to INP concentrations, which could affect the radiative balance of the atmosphere (Zeng et al., 2009; Hoose et al., 2010b; DeMott et al., 2010; Wang et al., 2014; Tan et al., 2016) .~~ ice-nucleating properties irrespective of the aerosol chemical composition. This is an unrealistic assumption that may affect the realism of mixed-phase clouds in models.

Over the Southern Ocean clouds tend to persist in a supercooled state more commonly than models predict (Bodas-Salcedo et al., 2014), which might be related to the very low INP concentrations that exist in this region ~~.~~ but which are not simulated in models (Bigg, 1973; DeMott et al., 2016) . It has been shown that less INP in the Southern Ocean lead to less ice and more supercooled water in model clouds, with a significant impact on the radiative properties of the clouds (Tan et al., 2016) . The variability among different models in the representation of cloud glaciation can lead to differences of ~~10s~~ tens of degrees in the temperature at which clouds glaciate (McCoy et al., 2015b). A ~~poor~~ better representation of mixed-phase clouds in climate models ~~has been shown to be~~ is important for climate prediction. For example, Tan et al. (2016) concluded that the response of global mean surface temperature to a doubling of CO_2 is more than one degree greater when mixed-phase clouds are better represented. This cloud-phase feedback is particularly sensitive to the amount of supercooled liquid in Southern Ocean mixed-phase clouds where most current models are biased relative to ~~observations~~ measurements (McCoy et al., 2015b).

In the future, regional and global climate models will include improved representations of cloud processes (Bauer et al., 2015), including ice processes, so an improved representation of heterogeneous ice nucleation will be required to make the

models more physically realistic and correct some of the main ~~model~~ biases. In particular, studies have shown that clouds are sensitive to INP concentrations, which could affect the radiative balance of the atmosphere (Zeng et al., 2009; Hoose et al., 2010b; DeMott et al., 2010; Wang et al., 2014; Tan et al., 2016). The reliability of such studies will depend on being able to relate the changes in cloud properties to emitted aerosol species so that we can attribute future changes in weather and climate to particular aerosol sources. ~~A similar approach has been used in global aerosol models~~ Global aerosol models have for many years ~~, enabling~~ been based on transported aerosol species from different sources, which enables aerosol radiative forcing to be related to anthropogenic and natural emissions and their effects on ~~aerosols and~~ cloud droplet formation (Ghan and Schwartz, 2007; Rap et al., 2013; Carslaw et al., 2013; Kodros et al., 2015). Our ability to achieve the same level of realism for ice formation has been much more difficult to achieve, partly because it has been challenging to identify species-specific ~~ice nucleating~~ ice-nucleating properties (Hoose and Möhler, 2012; Murray et al., 2012) and model them on a global scale.

Previous studies have simulated heterogeneous ice nucleation on a global scale accounting for different aerosol species ~~(Hoose et al., 2010b, a; Spracklen and Heald, 2014)~~ (Lohmann and Diehl, 2006; Hoose et al., 2010b, a; Sesartic et al., 2013; Spracklen an These studies used classical nucleation theory to calculate nucleation rates using contact angles derived from laboratory data for each INP species. This approach has the advantage that the time dependence of ice nucleation is represented, but when a single contact angle is used to describe ice nucleation by a single aerosol species, particle-to-particle variability is not represented (Herbert et al., 2014). Classical nucleation theory can be extended with a distribution of contact angles to account for differences in the ~~ice nucleating~~ ice-nucleating ability between different particles within the same material ~~(Niedermeier et al., 2011; Broadley et al., 2012; Herbert et al., 2014)~~ (Marcolli et al., 2007; Eidhammer et al., 2009; Niedermeier et al., 2(has been applied in models (Wang et al., 2014). In addition, it has been shown that representation of the time evolution of the distribution of contact angles is necessary to improve the representation of ice formation in a cloud-resolving model under some conditions using classical nucleation theory (Savre and Ekman, 2015).

The alternative to describing ice nucleation by classical nucleation theory is to use a singular approximation ~~Vali et al. (2015)~~ (Vali et al., which the time dependence of nucleation is assumed to be of secondary importance compared to the particle-to-particle variability. This approach has been used to define the population of INP in previous model studies (Niemand et al., 2012; Atkinson et al., 2013; Wilson et al., 2015). ~~The ice nucleating~~

The ice-nucleating efficiency using the singular description is defined by a ~~density~~ temperature dependent density (i.e. per unit surface area) of active sites, ~~which is a function of the temperature and usually of the surface area (n_s), or~~ ($n_s(T)$) which represents a spectrum of active sites with variable characteristic ice nucleation temperatures. The temperature dependent number of active sites can also be normalised to another parameter characteristic of the aerosol population (such as mass or volume) (Murray et al., 2012). ~~This~~ From this density of active sites, one can calculate what fraction of the particles will nucleate ice at a certain temperature (See Appendix:B)

The singular description of ice nucleation is consistent with many laboratory studies showing that particle-to-particle variability is the main factor driving the ~~observed~~ measured spectrum of INP concentrations with temperature (Vali, 2008; Herbert et al., 2014; Vali and Snider, 2015) for most of the known atmospherically relevant ~~ice nucleating~~ ice-nucleating species. However, it should be borne in mind that time dependence could play a role in long-lived stable mixed-phase clouds where ice

crystals are produced over a long period of time (Morrison et al., 2011; Murray et al., 2011; Westbrook and Illingworth, 2013; Herbert et al., 2014). Nevertheless, the singular approach for ice nucleation can be used to approximate INP concentrations, which can be calculated with knowledge of the number, size distribution and density of active sites of the relevant INP species.

~~Among the different aerosol species, mineral~~ Mineral dust is considered to be the dominant ~~ice nucleating~~ ice-nucleating species in many parts of the world (Hoose et al., 2010b; Ardon-Dryer and Levin, 2014; DeMott et al., 2015; Boose et al., 2016). Satellite ~~observations~~ measurements have shown a negative correlation between the amount of supercooled water and dust concentration (Choi et al., 2010), suggesting that dust ~~might be~~ is important for cloud glaciation. The ~~ice nucleating~~ ice-nucleating ability of dust has been quantified in ~~a number of studies (Niemand et al., 2012; Broadley et al., 2012; Augustin-Bauditz et al., 2014)~~ several studies (Koehler et al., 2010; Niemand et al., 2012; Broadley et al., 2012; Augustin-Bauditz et al., 2014) . Atkinson et al. (2013) found that ~~a mineral component of~~ K-feldspars are far more effective at nucleating ice than any of the other minerals in desert dust, ~~is responsible for most of ice nucleating activity of mineral dust aerosols. Several more recent studies agree with the results shown in (Atkinson et al., 2013) such as~~ which is supported by several later studies (Wex et al., 2014; Harrison et al., 2016; Zolles et al., 2015; Emersic et al., 2015; O'Sullivan et al., 2014; Niedermeier et al., 2015; Whale et al., 2014). Therefore the representation of ~~this type of mineral~~ K-feldspar in atmospheric models is important in order to obtain a realistic representation of ice nucleation by mineral dust. We have previously represented ice nucleation on a global scale by K-feldspar aerosols (Wilson et al., 2015; Atkinson et al., 2013). In this study we will take a similar approach to estimate the contribution of K-feldspar aerosol to global INP concentrations.

~~Phytoplankton and some~~ Some marine aerosol particles ~~might act as ice nucleating~~ act as ice-nucleating particles. Early evidence for a relationship between phytoplankton and marine INP was found by ~~Schnell and Vali (1975, 1976)~~ Schnell and Vali (1975) and Schnell and Vali (1976) , who observed active INP at temperatures as high as -4^oC in ~~re-suspended~~ resuspended biological material, largely from phytoplankton ~~,~~ filtered from bulk sea water. A relationship between the amount of biological material and the ~~concentration of INP was also observed in seawater~~ INP concentration was also measured in sea water and fog water by Schnell (1977). More recent studies have ~~observed~~ measured ice nucleation by *Thalassiosira* ~~*Psuedonana*~~*Pseudonana* (a ubiquitous species of phytoplankton) diatom cells ~~–~~(Knopf et al., 2010; Alpert et al., 2011) and exudates (Wilson et al., 2015). However, these studies ~~observed~~ measured ice nucleation at significantly lower temperatures than those ~~observed by Schnell and Vali (Schnell and Vali, 1975, 1976)~~ measured by Schnell and Vali (1975) and Schnell and Vali (1976) , suggesting that more active INP could be associated with phytoplankton material in the ocean. This would be supported by a previous ~~observation of ice nucleating~~ measurement of ice-nucleating bacteria associated with phytoplankton cultures ~~(Fall and Schnell, 1985)~~(Fall and Schnell, 1985) . Further evidence for the biological origin of marine INP is the heat sensitivity of some types of organic INP, i.e. the temperature at which they nucleate ice is reduced after heating to 100^oC (Wilson et al., 2015; Schnell and Vali, 1975, 1976). The likelihood of a marine source of INP was highlighted in studies that ~~observed~~ measured INP concentrations in ~~marine~~ environments remote from other sources of INP (Bigg, 1973; Schnell, 1982; Rosinski et al., 1986; Bigg, 1996; Rosinski et al., 1987, 1988). ~~Using the results from these early studies, Burrows et al. (2013) produced the~~

The first global simulation of marine INP concentrations ~~and a comparison with dust INP concentrations, and~~ (Burrows et al., 2013) sugge that marine organics were likely to ~~dominate the INP population~~ be the dominant source of INPs over remote marine regions

such as the Southern Ocean. Other studies provide further strong evidence that there is a marine source of atmospheric INP with biological origin. For example, INP production associated with phytoplankton blooms has been ~~observed~~ measured in laboratory experiments that use artificially generated sea spray aerosol from wave and bubble tanks (Wang et al., 2015; DeMott et al., 2016). DeMott et al. (2016) ~~observed~~ measured that the INP concentrations in laboratory-generated sea spray were consistent with measurements made by Bigg (1973) as well as with measurements of ambient INP concentrations in marine-influenced air. Wilson et al. (2015) found that the sea surface microlayer is enriched in INP compared to sub-surface seawater at the same locations. The sea surface microlayer is enriched in surface active organic material similar to that found in sea spray (Cochran et al., 2016; Gantt et al., 2011; Quinn et al., 2014; Aller et al., 2005; Orellana et al., 2011; Russell et al., 2010; Cunliffe et al., 2013). A correlation between total organic carbon content and the temperature at which microlayer droplets froze was ~~observed~~ measured (Wilson et al., 2015).

All the above evidence suggests the existence of a marine organic source of ~~ice nucleating~~ ice-nucleating particles that we will attempt to represent in this paper.

Here we conduct a modelling study of global immersion mode INP concentrations based on recently developed laboratory-based parameterizations of the ~~ice nucleating~~ ice-nucleating ability of two species: marine organic matter and potassium feldspar (K-feldspar). The objectives of our study are to: (i) determine the ability of laboratory-measured INP efficiencies to explain the global distribution of INP concentrations as a function of activation temperature; (ii) quantify the relative importance of these two sources of INP in different locations; (iii) determine what fraction of global INP concentrations can be explained by these two ~~major~~ sources and (iv) determine whether, within model and measurement uncertainties, we can use the model results to draw conclusions about additional important sources of INP.

2 Methods

2.1 Global modelling

We use the GLOMAP-mode global aerosol model described in (Mann et al., 2010). The model has a horizontal latitude-longitude ~~resolution~~ gridspacing of 2.8^ox2.8^o and 31 pressure levels from the surface to ~~10hPa~~10 hPa. The species represented in the baseline version are sulphate, sea-salt, black carbon, particulate organic matter and dust. In this study we focus on the representation of two species of relevance to INP: the K-feldspar component of dust and the organic component of primary marine sea spray aerosols. Aerosol chemical component mass concentrations and the particle number concentration are represented by seven internally mixed log-normal modes (four soluble and three insoluble). Aerosol microphysical processes in the model include nucleation of new particles by gas-to-particle conversion, growth by coagulation and condensation of low-volatility gases, dry deposition at the surface and below-cloud (impaction) and in-cloud (nucleation) wet scavenging. Nucleation scavenging is suppressed for ice clouds~~(~~, which are assumed to glaciate at -15^oC~~).~~. A discussion of the nucleation scavenging assumptions in our model is included in Browse et al. (2012) . Scavenging of aerosols by marine drizzle clouds is also included in the model to improve the predicted concentration in polar regions, as shown in Browse et al. (2012). The model uses wind, temperature and humidity fields from the European Centre for Medium-Range Weather Forecasts (ECMWF). We

ran the model from the year 2000 to 2001 in order to reach a steady state aerosol distribution before running the model and then used data from 2001 to 2002.

2.2 Representation of feldspar

Feldspar is emitted in the model as a fraction of the mass of dust (derived from AEROCOM emissions (Dentener and Kinne, 2006)). The model has been shown to reproduce dust ~~concentrations accurately~~ mass concentrations within an order of magnitude (Mann et al., 2010; Huneeus and Schulz, 2011). The fraction of feldspar emitted is assumed to be equal to the fraction by mass of this mineral found in the soils in the arid emission regions. This assumption has been shown to be a close approximation to the fraction of the mineral emitted in the form of aerosols (Lafon et al., 2004; Nickovic et al., 2012). However, new studies suggest that there is a difference between the fraction of the minerals found in the soil after wet sieving and the aerosolized fraction (Perlwitz et al., 2015). This difference is considered to be small (around a factor of 2) compared to other errors in our representation of the ~~ice nucleating~~ ice-nucleating ability of K-feldspar such as differences in the density of active sites of different types of K-feldspar (around a factor of 6) (Harrison et al., 2016).

Feldspar is emitted into the insoluble accumulation and coarse modes with fractions corresponding to the clay and silt size range (Lafon et al., 2004; Nickovic et al., 2012), similar to the method followed in Atkinson et al. (2013). However, once in the atmosphere, dust particles (including feldspar) are aged by condensation of sulphates and secondary organic aerosol material and moved into the soluble modes, which are subject to wet scavenging. This process was not represented in Atkinson et al. (2013), and was likely one of the causes of the overestimation of dust concentrations in remote locations as discussed in Atkinson et al. (2013). With this wet scavenging process active, the concentration of feldspar in remote places such as the Southern Ocean is ~~a~~ several orders of magnitude smaller than the concentrations simulated by Atkinson et al. (2013). However, the concentrations closer to source regions are very similar to Atkinson et al. (2013). ~~Compared with other minerals, feldspar~~

Feldspar tends to reside in the larger particles ~~as~~ because it is found mainly in the silt fraction ($~~r > 1 \mu m~~ r \geq 1\,\mu$m) (Claquin et al., 1999). ~~Due to its size, it is rapidly removed~~ It is therefore removed more rapidly from the atmosphere compared with other ~~mineral species such as those corresponding to~~ minerals that occur preferentially in the clay fraction ~~as~~ because removal by dry deposition increases with particle size. Relatively rapid scavenging of large feldspar-containing particles means that it is transported shorter distances compared with smaller dust particles~~(that are less rich in feldspar)~~.

2.3 Representation of marine organic aerosols

Submicron marine organic aerosols are usually parameterized by relating the organic mass fraction observed in sea-spray to some variables such as seawater chlorophyll content or wind speed (O'Dowd et al., 2015; Rinaldi et al., 2013; Gantt et al., 2011). With those parameterizations, the flux of marine organic mass can be calculated in a model with the flux of sub-micron sea-salt following Eq.A5 (see Appendix). The performance of any parameterization in reproducing observations of marine organic mass concentrations will therefore depend on the emission fluxes of submicron sea-spray, which is a highly uncertain model-dependent process. Mann et al. (2014) showed that models can have differences of more than a factor of six in the simulated concentration of particles with a diameter larger than 100nm in the Southern Ocean. Other uncertainties affecting

the modelled concentrations of marine organic aerosols can arise from removal processes or some other aspect of the model such as the parameterization of convection and cloud microphysical processes, or model grid and temporal resolution, as well as uncertainties related to the organic mass fraction parameterization. Therefore, the performance of any parameterization in an aerosol model will be affected by the uncertainties related to these processes. It is therefore necessary to evaluate and adjust the modelled marine organic concentrations to match observations.

To represent primary marine organic aerosols in GLOMAP-mode, we developed a parameterization of the organic mass fraction of submicron sea spray particles and adjusted it to fit the observations of water insoluble organic matter (WIOM) at Amsterdam Island (37.48^oS, 77.34^oE) and Mace Head (53.33^oN, 9.9^oW). We use observations from only these two stations due to the limited availability of long-term measurements of marine WIOM. It is thought that most primary marine organic emissions are formed of water-insoluble components (Facchini et al., 2008). The marine organic component is assumed to be internally mixed with sea-salt. The sea-salt emissions in our model are dependent on the surface wind speed (10m above the surface) and follow the parameterization of (Gong, 2003), which is an extension of (Monahan et al., 1986). The development of our new organic mass fraction parameterization, explained in detail in Appendix:A, assumes that the organic mass fraction of the sea-spray particles depends on wind speed and the chlorophyll content of seawater. ~~In order to match the seasonal cycle of WIOM at these two sites, the~~ The organic emission parameterization includes a positive dependence of WIOM mass fraction on chlorophyll (O'Dowd et al., 2015; Rinaldi et al., 2013; Gantt et al., 2011), but a negative dependence on wind speed. Thus, the WIOM is essentially diluted in the sea spray particles when the total sea spray emission flux is high, which may be caused by a limited supply of organic material in the surface ocean but effectively limitless salt (Gantt et al., 2011) . This parameterization is similar to previous chlorophyll based parameterizations such as ~~Rinaldi et al. (2013); Gantt et al. (2011) but scaled~~ Rinaldi et al. (2013) and Gantt et al. (2011) but adjusted in order to fit the observations in Amsterdam Island and Mace Head when applied in our model. Our model agrees with the observed WIOM concentrations within a factor of two (Fig. 1) which is a small factor compared with other uncertainties related to the calculation of INP concentrations such as the uncertainty related to the parameterization of the number of INP per gram of organic carbon in sea-water (around an order of magnitude) ~~Wilson et al. (2015)~~ (Wilson et al., 2015) .

The mixed organic-salt sea spray particles are emitted into the accumulation mode and treated as water-soluble particles with respect to their CCN activity, and hence they are removed by nucleation scavenging when they enter a precipitating cloud. This treatment of primary marine organic mass as internally mixed with sea-salt and being able to activate to cloud droplets is consistent with other previous studies (Vignati et al., 2010; Burrows et al., 2013; Orellana et al., 2011; Ovadnevaite et al., 2011; Fuentes et al., 2011; Partanen et al., 2014). Simulated surface concentrations of marine organic aerosol mass are shown in Figure 2.

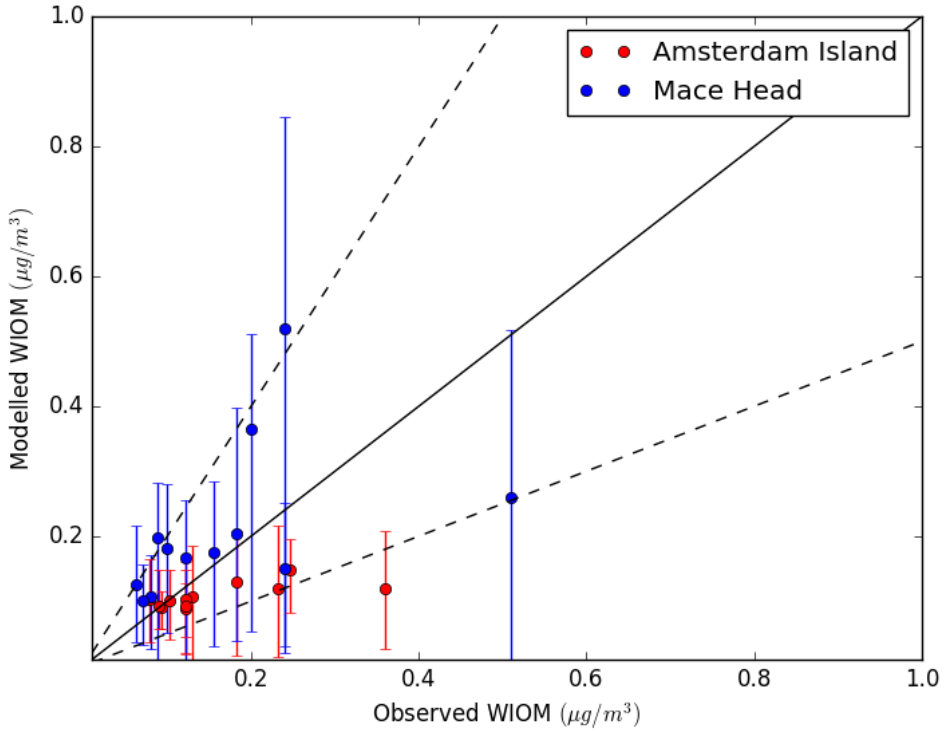

Figure 1. Evaluation of modelled water-insoluble organic matter (WIOM) mass concentration with monthly mean observations at Mace Head $(53.33^{\circ}N, 9.9^{\circ}W)$ and Amsterdam Island $(37.48^{o}S, 77.34^{o}E)$. The dashed lines correspond to a factor of two difference between modelled and observed values. The error bars correspond to the simulated daily variability within a month (maximum and minimum values). Variability in the observed values is not shown because the measurements were made with filter samples which were collected over 1 week, and therefore they do not represent the day-to-day variability.

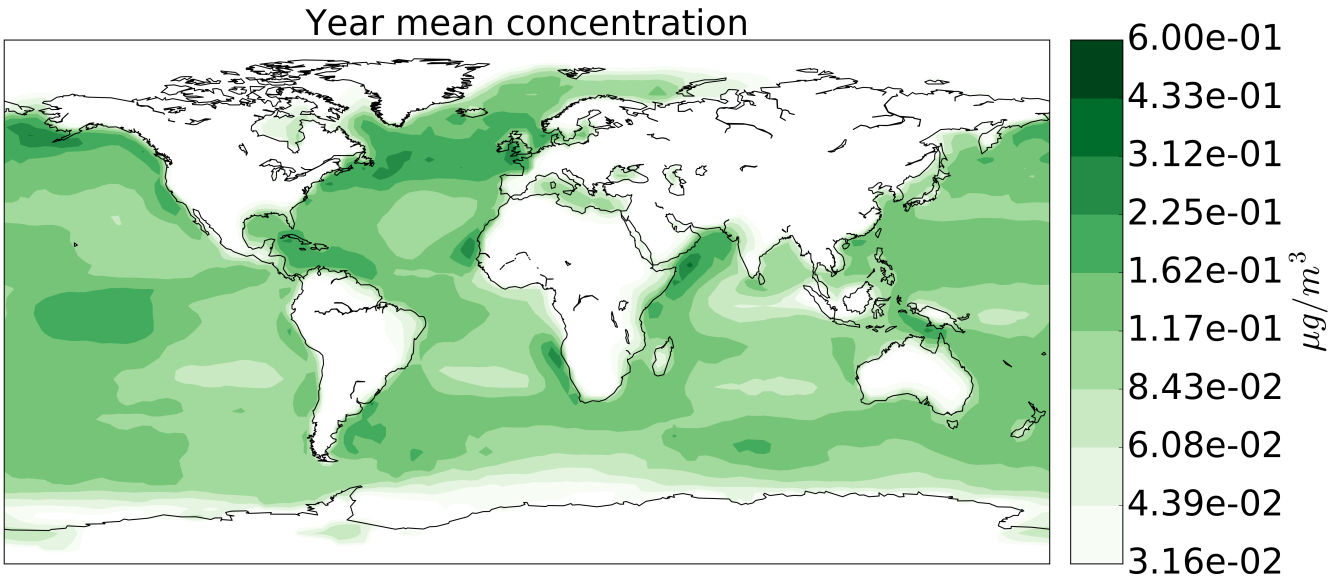

Figure 2. Annual mean ~~surface~~ modelled mass concentration of sub-micron marine organic ~~aerosols (WIOM)~~ aerosol mass at surface level

2.4 Calculation of INP concentrations

To quantify INP concentrations from the modelled aerosol distributions we use the singular description. This method assumes that the time dependence of ice nucleation plays a secondary role and that specific particles have a characteristic temperature at which they nucleate ice. The spectrum of ~~ice nucleating~~ ice-nucleating properties is often represented as a surface area density of active sites dependent on temperature, which is appropriate for solid particles like dust (Atkinson et al., 2013). For marine organic material the active site density is defined per unit mass of organic material in the particle (Wilson et al., 2015).

The method for calculating ~~ice nucleating~~ ice-nucleating particle concentrations from the simulated aerosol size distributions is explained in Appendix: B.

To represent the ~~ice nucleating~~ ice-nucleating ability of K-feldspar we assume that 35% of the total feldspar is K-feldspar, as assumed in (Atkinson et al., 2013), then we apply the parameterization for n_s shown in (Atkinson et al., 2013). ~~By using this parameterization we assume that the~~ Our method assumes that different varieties of K-feldspar nucleate ice with the same efficiency. Different studies have shown that the values of n_s for most types of ~~potassium feldspar~~ K-feldspar tend to agree with the values shown in (Atkinson et al., 2013) within a factor of two to four (Harrison et al., 2016; Emersic et al., 2015; O'Sullivan et al., 2014; Zolles et al., 2015; Niedermeier et al., 2015; Whale et al., 2014). However, it should be borne in mind that a minority of feldspar samples are either much more active or much less active than indicated by ~~the parameterization defined by (Atkinson et al., 2013)~~ Atkinson et al. (2013) . Nevertheless, the Atkinson parameterization is a good approximation of the majority of K-feldspars that have been studied in the laboratory. Assuming that feldspar particles are externally mixed in terms of their mineralogy, we can use the laboratory parameterizations to calculate the INP concentration for each soluble mode, following Eq.B9, as a function of activation temperature (see Appendix:B for the derivation).

For marine organic aerosols, we use the parameterization shown in (Wilson et al., 2015), and apply it to our distributions of simulated marine organic aerosol mass. We are assuming that the organic material found in the sea-surface microlayer is representative of the organic material in sea-spray aerosols and that this material has the same ~~ice nucleating~~ ice-nucleating ability as sea-surface microlayer material. For marine organic particles the density of active sites per particle is always small ($\lambda < 0.1$ see Appendix: B) for the whole temperature range covered by the parameterization (-6 to -27^oC) and all realistic sizes of particle (submicron particles). This means that we can calculate the INP concentration in a simplified way following Eq.1 (Appendix: B for the derivation).

$$[INP](T) \approx \lambda(T) \cdot [N] \tag{1}$$

It should be noted that extrapolating this parameterization to lower temperatures, or for bigger particles, may lead to unrealistically high concentrations of INP because Eq.1 is no longer valid.

~~It is important to bear in mind~~ There are two distinct ways of presenting simulated INP concentrations – either according to the concentration that an INP counter would measure or the concentration of potential INP under ambient conditions. An INP is defined as a particle which has the potential to nucleate ice if exposed to a specific set of conditions (much like a CCN is defined at a specific super-saturation). For the immersion/condensation mode, the INP concentration we quote are for water saturation and for a defined activation temperature. ~~Hence, there is the question of which activation temperature is most appropriate for displaying the model data. In Figure 4 we illustrate two distinct ways of displaying the model data.~~ The two ways of quoting INP concentrations (~~$[INP]$, where square brackets indicate concentration) are to quote $[INP]_T$~~ Figure 4) are at a specific activation temperature ~~(T) or to quote $[INP]_{ambient}$~~ T ($[INP]_T$), which is appropriate for comparing with an INP instrument set to measure at that temperature, or $[INP]_{ambient}$ where the activation temperature is set as the local ambient temperature.

In Figure 4~~a~~ (top) we show the INP concentration for ~~a specific activation temperature ($[INP]_T$)~~ an activation temperature of -20^oC ($[INP]_{-20}$) at the 600 hPa pressure level, which is what would be measured by an INP instrument set to this temperature. Throughout much of the globe, especially through the tropics, the temperature at this pressure level will never reach -20^oC, so the INP at this altitude that can be active at -20^oC or warmer would not fulfil their potential to nucleate ice. However, if the air at a particular altitude were drawn into a convective system the INP it contains would activate higher in the cloud. Hence, when considering a deep convective cloud where air is moved vertically through all the mixed-phase range of temperatures, it is the spectrum of ~~$[INP]_T$~~ $[INP]_T$ (a spectrum over activation temperature) which is the pertinent quantity (see Fig. 3 for an illustration). In addition, when comparing measurements of ~~$[INP]$~~ $[INP]$ concentration to our modelled ~~$[INP]$~~ $[INP]$, we compare these quantities at specific activation temperatures. Hence, Figure 4~~a provides the $[INP]$~~ (top) provides the $[INP]$ to compare to a measurement of ~~$[INP]$~~ $[INP]$ where the activation temperature in a measurement was -20^oC.

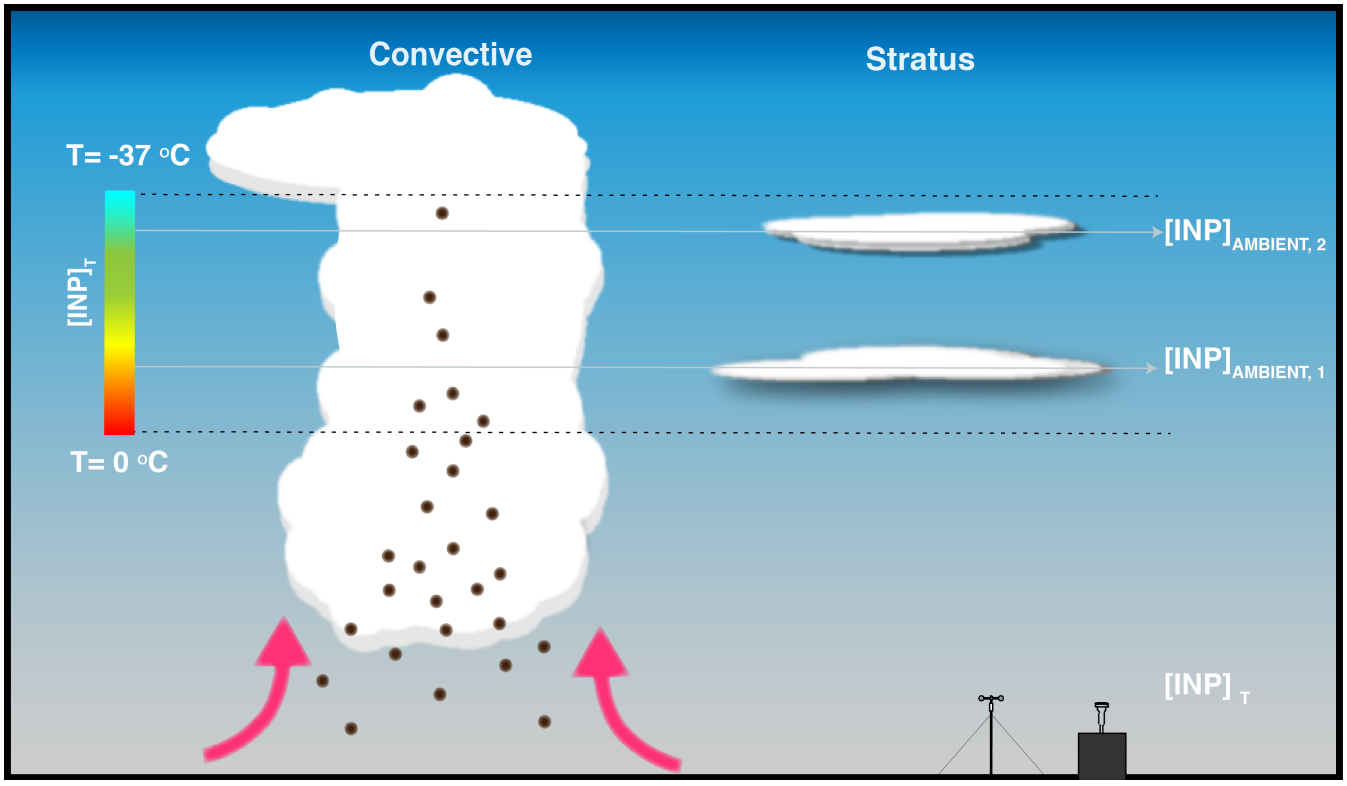

Figure 3. Illustration of the two ways in which we display INP concentrations. It is important to bear in mind that INP are defined as particles with the potential to nucleate ice and their concentration is quoted for a specific set of conditions. ~~$[INP]_{ambient}$~~ $[INP]_{ambient}$, where ambient denotes the local atmospheric temperature, is a useful way of looking at the INP concentration relevant to non-deep convective mixed-phase clouds. ~~$[INP]_T$~~ $[INP]_T$ on the other hand, has utility in representing the spectrum of INP concentrations over temperature that will influence clouds with a large vertical extent such as deep-convective systems. Moreover ~~$[INP]_T$~~ $[INP]_T$ is the relevant quantity when comparing modelled and observed INP concentrations, since measurements are made by exposing particles to controlled temperatures within the instrumentation.

In Figure 4~~b~~ (bottom) we plot the ~~$[INP]$~~ $[INP]$ where the activation temperature is set to the local atmospheric temperature. ~~$[INP]_{ambient}$~~ $[INP]_{ambient}$ is useful to identify regions in the atmosphere where we might expect cloud glaciation in stratus type mixed-phase clouds. Non-deep convective clouds with minimal vertical extent, such as altostratus, altocumulus or high latitude stratus, form in air parcels which have not been vertically transported large distances, in contrast to deep convection. Based on Figure 4~~b~~(bottom), we would expect K-feldspar to contribute much more to mid-latitude, mid-level (600 hPa), mixed-phase clouds in the Northern Hemisphere than in the Southern Hemisphere.

Both ~~$[INP]_T$ and $[INP]_{ambient}$~~ $[INP]_T$ and $[INP]_{ambient}$ are useful ways of looking at the global INP distribution, but in order to understand the impact of these INP species on clouds, we would need a model where the INP fields are coupled to cloud microphysics and dynamics. This is beyond the remit of this study, where our goal is to understand the global distribution of INP and evaluate the model against measurements.

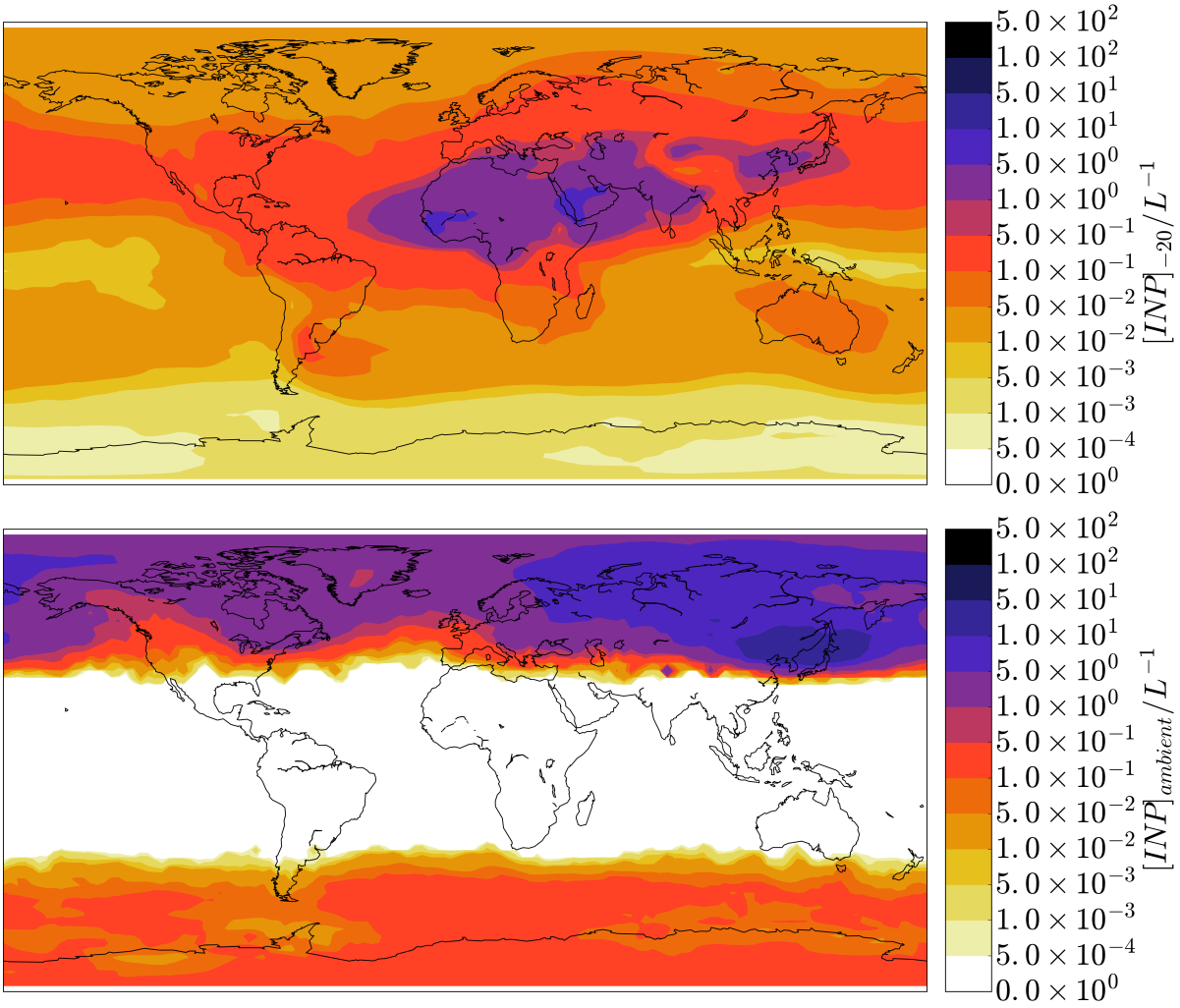

Figure 4. Annual mean K-feldspar INP distribution using GLOMAP-mode at a pressure level of 600hpa. Top panel shows the concentration of ~~ice nucleating~~ ice-nucleating particles active at a temperature of ~~-20C~~ -20^oC ($~~[INP]_T~~[INP]_T$) whereas the bottom panel shows the INP concentration at local ambient temperature ($~~[INP]_{ambient}~~[INP]_{ambient}$).

~~In order to calculate~~ $~~[INP]_{ambient}~~$To calculate $[INP]_{ambient}$, we use the daily mean temperatures obtained from ECMWF and the daily mean concentrations (mass and number concentrations) predicted by the model. ~~With these values we can then~~ The daily values are then averaged to calculate monthly and annual mean values of INP. The concentrations of $~~[INP]_{ambient}$ ~~at temperatures colder~~ $[INP]_{ambient}$ at temperatures lower than the temperature limit of the parameterizations (for K-feldspar: -25^oC and marine organics: -27^oC) is set at the value defined by the concentration at the limiting temperature of each parameterization. This is consistent with studies that caution against extrapolating singular parameterizations outside the range where measurements were made. For example, Niedermeier et al. (2015) showed that the density of active sites on the surface of

K-feldspar particles plateaus below about -25^oC and a simple extrapolation of the parameterization of Atkinson et al. (2013) would lead to substantial errors.

3 Results

3.1 Simulated global INP distributions

Simulated INP concentrations at the surface are shown in Figure 5 for an activation temperature of -15^oC. Feldspar dominates the INP concentration in environments influenced by terrestrial dust emission sources such as the Sahara and the Asian dust belt. However, concentrations fall rapidly with distance away from dust sources because the large size feldspar-containing dust particles are rapidly removed from the atmosphere ~~5a~~(Fig. 5a). The concentrations of INP from K-feldspar and marine organics are summarized in Figure 5c and comparison with panels 5a and 5b reveals that INP from deserts far outnumber INP from sea spray throughout much of the low and mid-latitudes, which are strongly influenced by desert dust, but marine organics become more important over the world's remote oceans, such as the Southern Ocean.

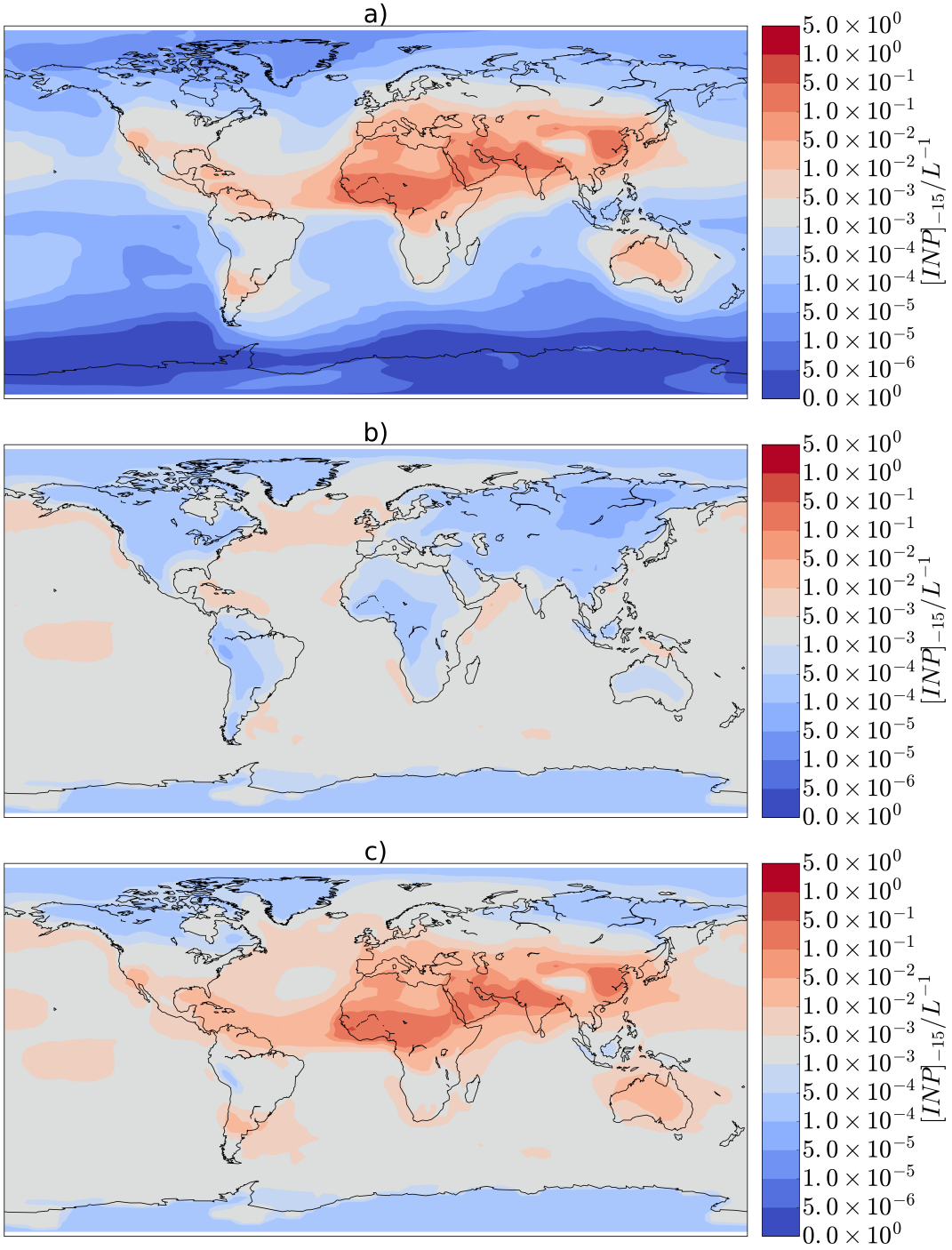

Figure 5. ~~Yearly~~ Annual mean distributions of ~~ice nucleating~~ ice-nucleating particles concentrations, for an activation temperature of -15^{o}C. Based on feldspar (a) and marine organics (b). (c) shows the total INP concentration obtained by summing the INP concentrations from K-feldspar and marine organics. We show $[INP]_T$ for a T of -15^{o}C because this is a temperature used by many instruments. The number of INP that activate to ice crystals ($[INP]_{ambient}$) at the surface will be zero over much of the globe, because these particles will only become important at high altitudes. Surface concentrations are shown because this is where most observations of atmospheric INP concentrations are made.

Figure 6 shows the ~~$[INP]_{ambient}$~~ $[INP]_{ambient}$ concentration of marine organics and K-feldspar for the different ~~months of the year~~seasons. Feldspar dominates ~~$[INP]_{ambient}$~~ $[INP]_{ambient}$ on a monthly mean basis across the northern hemisphere, while marine organic aerosols, tend to be important in southern high latitudes, such as those corresponding to the Southern Ocean and Antarctica.

The ~~monthly~~ seasonal mean results in Fig. 6 have to be interpreted with caution since high dust concentrations are often associated with episodic dust plumes. Hence, the ~~monthly~~ seasonal mean may not reflect the relative contributions of desert dust and sea spray INP on a day-to-day basis. In addition, day-to-day fluctuations in temperature can drive large changes in ~~$[INP]_{ambient}$~~ $[INP]_{ambient}$ which are not necessarily representative of the typical concentrations of active ~~ice nucleating~~ ice-nucleating particles, but will greatly affect the monthly mean value of ~~$[INP]_{ambient}$~~$[INP]_{ambient}$, as the INP concentration increases exponentially with temperature. To account for such variability, Fig.7 shows the percentage of days per season when the concentration of ~~$[INP]_{ambient}$~~ $[INP]_{ambient}$ from marine organics is greater than the concentration from K-feldspar. Overall, over the northern hemisphere, marine organic INP concentrations are greater than K-feldspar INP concentrations between 10% and 30% of the days when the temperature is within the mixed-phase range $(0^oC$ to $-37^oC)$ and the total concentration of ~~$[INP]_{ambient}$~~ $[INP]_{ambient}$ is larger than $10^{-4}L^{-1}$. This large influence of marine organic INP is hidden when looking at the monthly mean values shown in Figure 6 as the feldspar monthly mean concentrations are dominated by short periods when a dust plume occurs. It is striking that the contribution of marine organics is more important than K-feldspar on a significant fraction of days in the Northern Hemisphere because in these zonal mean plots we are averaging across the Eurasian and North American continents where the influence of marine organics is minor. In fact, Fig.7 ~~suggest~~ suggests that marine organics are more important than K-feldspar in the North Atlantic, for example, on 10-40 % of days at 600 hPa.

In the Southern Hemisphere, the dominance of marine organic aerosols is more ~~consistent. Both on~~ prevalent. On a monthly mean basis and on the large majority of days, marine organic aerosols are the dominant INP from March through to November (Fig.6b-d). On the other hand, K-feldspar cannot always be ruled out as an important source of INP in the southern high latitudes in the period from March to November, since there are still several days per month (10 to 60%) when the concentration of transported K-feldspar INP ~~, particularly from South American and Australian sources,~~ dominates over marine organics (Fig.6). Conversely, during December to February at southern high latitudes, K- feldspar mineral dust is more important on more days than marine organic aerosols (Fig.7a). This is related to higher dust concentrations during the austral summer.

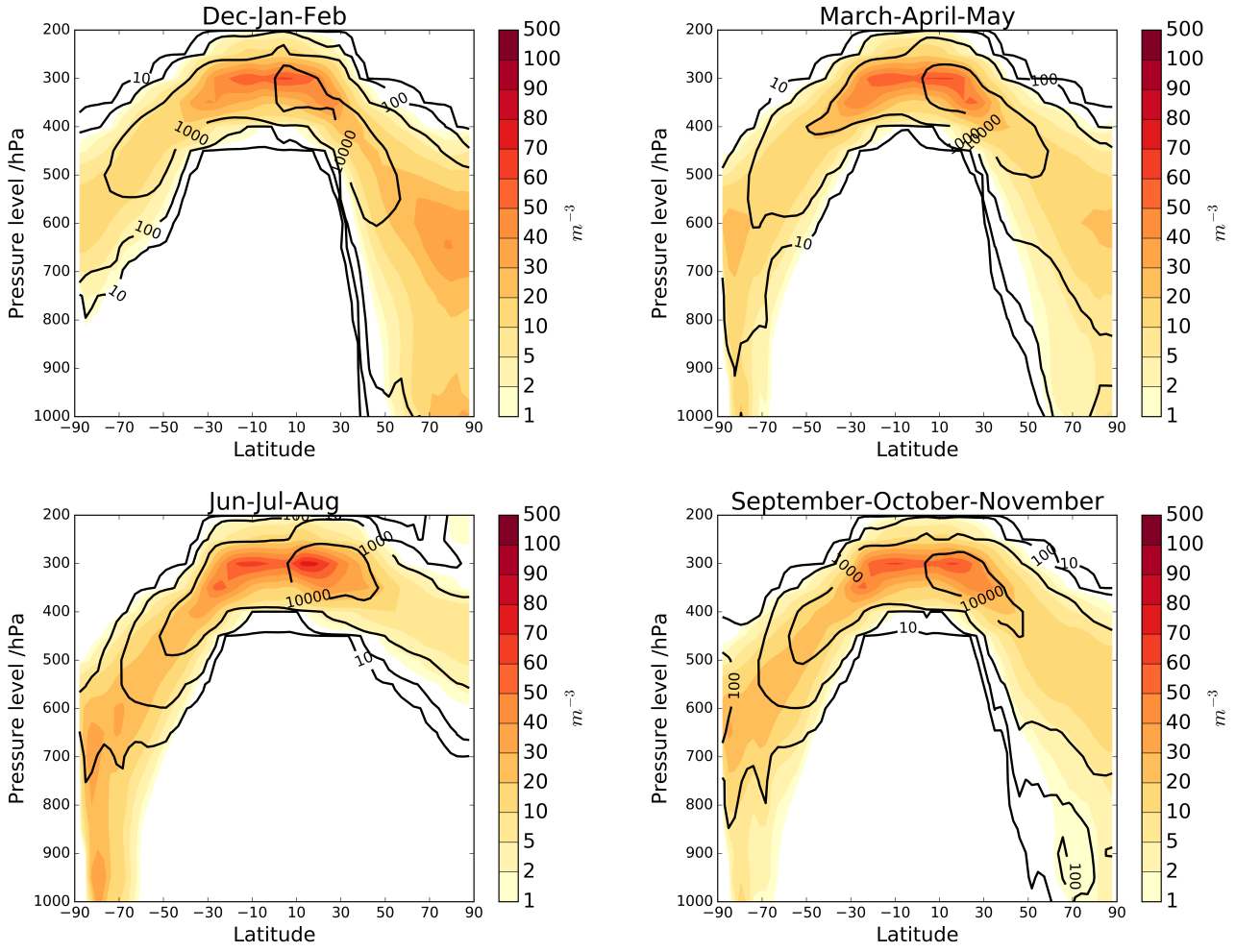

Figure 6. Zonal mean profiles of ~~$[INP]_{ambient}$~~ $[INP]_{ambient}$ for every month of the year. The black contour lines correspond to the INP concentration of K-feldspar aerosols (m^{-3}), while the colormap shows the INP concentration of marine organic aerosol The values correspond to ~~monthly~~ seasonal mean values calculated using daily concentrations and temperatures and averaged across latitudes.

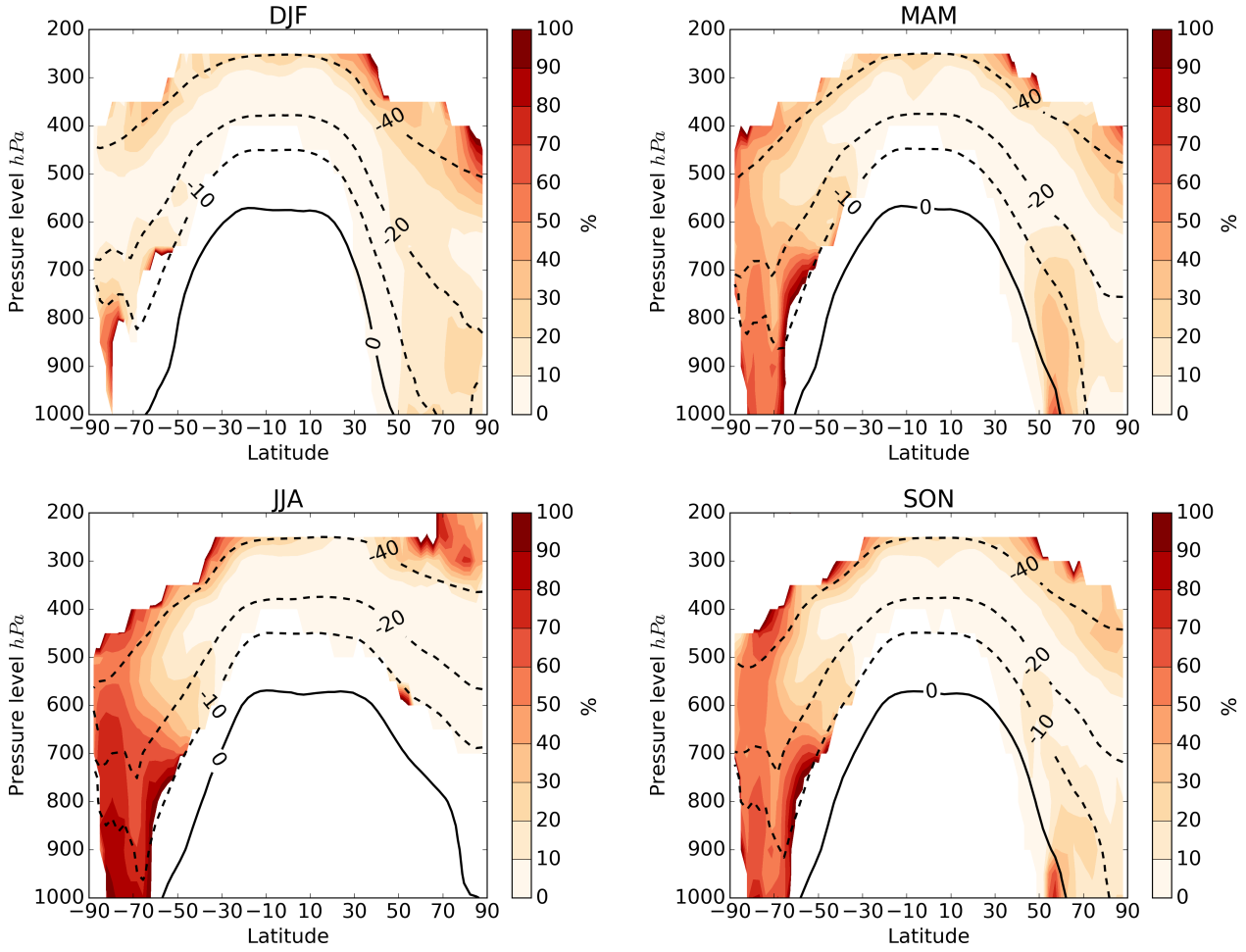

Figure 7. Percentage of days when ~~$[INP]_{ambient}$~~ $[INP]_{ambient}$ from marine organic aerosols is greater than from K-feldspar. The number of days have been calculated only for times and locations where the total ~~$[INP]_{ambient}$~~ $[INP]_{ambient}$ concentration is larger than $0.1m^{-3}$. The black contour lines represent seasonal mean isotherms in degrees centigrade.

3.2 Comparison with observations and other parameterizations

Some climate models determine heterogeneous freezing using parameterizations that depend only on the temperature, ~~(McCoy et al., 2015a)~~ as the scheme of Meyers et al. (1992). This type of parameterization does not account for spatial or temporal variations in the aerosol loading and does not differentiate between different aerosol species, both of which actually determine INP concentrations. Other parameterizations such as (DeMott et al., 2010) use empirical evidence from extensive atmospheric measurements to define INP concentrations in terms of the aerosol particle concentration above a defined size.

Such parameterizations implicitly account for the fact that many INP-active species are present in larger particles, such as in dust (Niemand et al., 2012) and biological particles (Tobo et al., 2013). In addition, larger particles are more likely to

carry nano-scale or smaller ice active materials (O'Sullivan et al., 2015). Nevertheless, size-based parameterizations of INP concentration do not account for the source of the particles or differences between marine and terrestrial aerosols,so they may not capture variations and long-term trends driven by changes in aerosol emissions since different aerosol types have different ice-nucleating abilities.

In Figure 8 we compare several singular INP parameterizations with observations. Panel ~~b~~ a compares the observed values of ~~$[INP]_T$~~ $[INP]_T$ to those predicted by the scheme of Meyers et al. (1992), which relates ~~$[INP]_T$~~ $[INP]_T$ to temperature and is independent of aerosol properties. This is clearly a poor representation of many INP measurements in the atmosphere (Table ~~.~~1). Fig. 8~~c shows the $[INP]_T$~~ b shows the $[INP]_T$ predicted by the parameterization of DeMott et al. (2010), in which ~~$[INP]_T$~~ $[INP]_T$ is predicted on the basis of the concentration of particles larger than 0.5 ~~μm~~ μm diameter, $n_{aer,0.5}$, and temperature. This parameterization ~~has a similar performance to Meyers et al. (1992) as it still~~ tends to overpredict ~~$[INP]_T$,~~ ~~although~~ $[INP]_T$, although multiplicative scaling of the ~~predicted values by multiplying them by a factor to fit the observations might~~ simulated values would greatly improve its performance as it has a better correlation coefficient (~~Tab.~~Table 1). We also note that in our analysis we use the annual mean $n_{aer,0.5}$ from our model (without the contribution of sea-salt aerosols), whereas DeMott et al. (2010) used $n_{aer,0.5}$ from measurements coincident with their INP measurements and obtained a better representation of the ~~$[INP]_T$~~ $[INP]_T$ data (some of which is included in Figure 8). Sulphate aerosols contribute significantly to the simulated $n_{aer,0.5}$ in remote places impacting DeMott et al. (2010) over oceans.

Fig.8~~d~~ c shows how our model compares with observed ~~$[INP]_T$~~ $[INP]_T$ using the desert dust parameterization from Niemand et al. (Niemand et al., 2012) (with no additional marine organic INP). In this case some observations are overestimated by a factor 100-1000, especially those in marine regions (triangles). This overprediction is partly caused by the implicit assumption that all components of dust particle nucleate ice with the same efficiency. Feldspars exist mainly in the large dust particles (silt fraction) so they are not transported as efficiently to remote locations as the clay minerals, consequently transported desert dust is less important as an INP in remote locations.

Finally, we compare our two-species representation of INP with the same ~~$[INP]_T$~~ $[INP]_T$ dataset Fig.8~~e-g~~d. The observations used in this comparison are within the range of temperatures of the parameterizations (-5 to -27^oC). In this case our representation of INP (~~8e~~Fig. 8d) is able to reproduce 56.7% of the observations within an order of magnitude and 74% within 1.5 orders of magnitude ~~Tab. 1.~~ (Table 1). When the parameterizations are extrapolated outside their temperature range, they still perform similarly (Table 1).Looking at the performance of the different ways of representing INP within the smallest temperature range shared by the all the parameterizations (-12 to -25^oC), our representation of INP is able to reproduce 61.6% of the datapoints within and order of magnitude and 78.7% within 1.5 orders of magnitude. These values are greater than the obtained when using the other 3 parameterizations used for this study (Table 1).

The contributions of K-feldspar and marine organics to the simulated INP concentrations of each data point are illustrated in Figure ~~8g~~9b. Marine organics explain more than 90% of the INP concentrations in Marine-influenced environments and some terrestrial environments with low concentrations of INP (corresponding to high temperature observations). K-Feldspar, however, explains most of the observations in terrestrial regions. The large biases observed when using species independent parameterizations over marine regions are largely corrected, as most marine influenced INP concentrations are simulated within

Parameterization	Temperature range	Datapoins	Pt1	Pt1.5	R ~~(log)~~	Pt1*	Pt1.5*	R *	Pt1**	Pt1.5**	R *
Meyers et al. (1992)	0^oC to -37^oC	479	35.5%	51%	0.57	35.5%	51%	0.57	27.4%	39.4%	0.4
DeMott et al. (2010)	0^oC to -37^oC	479	24%	39.2%	~~0.672~~ 0.67	24%	39.2%	0.67	23.6%	38.3%	0.4
Niemand et al. (2012)	-12^oC to -33^oC	438	33.7%	53%	0.58	31.7%	53%	0.64	46.9%	70%	0.4
Marine + Kfeldspar	-6^oC to -25^oC	354	56.7%	74%	~~0.625~~ 0.62	54.9%	75.9%	0.64	61.6%	78.7%	0.5

Table 1. Statistical performance of the different parameterizations. Pt1 and Pt1.5 are the percentages of datapoints reproduced within an order of magnitude and 1.5 orders of magnitude in the temperature range of every parameterization. The number of datapoints used for calculating these values is shown under the 'Datapoints' column. The values with * show the same calculation but including datapoints outside the temperature range of the parameterizations. These values give an idea of the performance that you would expect if you extrapolate the parameterizations in a climate model. The values with ** are for datapoints within the smallest temperature range shared by the 4 parameterizations (-12^oC to -25^oC). The correlation coefficient has been calculated with the logarithm of the values as INP concentrations vary logarithmically with temperature.

an order of magnitude (72% of marine points), although ~~,~~some biases are still apparent. Figure 10 shows the location and temperature of the observations with a bias greater than 1.5 orders of magnitude. Figure 10a suggests that the main positive bias occurs at low temperatures (<-20^oC) in locations far form K-feldspar emission sources , where it is transported. It is possible that processes such as atmospheric aging by acids play a role in modifying the efficiency of K-feldspar aerosols in nucleating ice (Augustin-Bauditz et al., 2014) or that we overestimate the amount of feldspar particles that are transported. One possible explanation for this is that we do not model the preferential removal of INP during cloud glaciation, hence K-feldspar aerosol transported over long distances may contain fewer INP than our model simulations (Stopelli et al., 2015; Haga et al., 2014, 2013). Fig.10b shows that the model underestimates high-temperature INP concentrations (~ -5 to -15^oK) over terrestrial locations, which might indicate that we are missing some terrestrial source that affects the INP concentration. Some of the possible candidates for these particles could be bacteria (Möhler et al., 2008; Hartmann et al., 2013; Maki and Willoughby, 1978) , fungal material (O'Sullivan et al., 2015, 2016; Fröhlich-Nowoisky et al., 2015; Pouleur et al., 1992; Morris et al., 2013), agricultural dust (O'Sullivan et al., 2014; Tobo et al., 2014; Garcia et al., 2012) or biological nanoscale fragments attached to mineral dust particles (O'Sullivan et al., 2015, 2016; Pummer et al., 2015; Fröhlich-Nowoisky et al., 2015). However, size-resolved INP measurements in several terrestrial locations suggest that a large proportion (40%- 90%) of INP are commonly associated with larger particles (diameter > 2.5 ~~μm~~μm) (Mason et al., 2016). Such large particles are likely to have short atmospheric lifetimes, so they are less likely to be transported to cloud altitudes than smaller particles and are more likely to be transported shorter distances. In summary, the overall agreement between the two-species model and observations is good, but there are significant discrepancies. These discrepancies indicate that processes such as aging and preferential INP in-cloud removal ~~are~~ could be important and also that we ~~are~~ could be missing high temperature terrestrial sources of INP in the model.

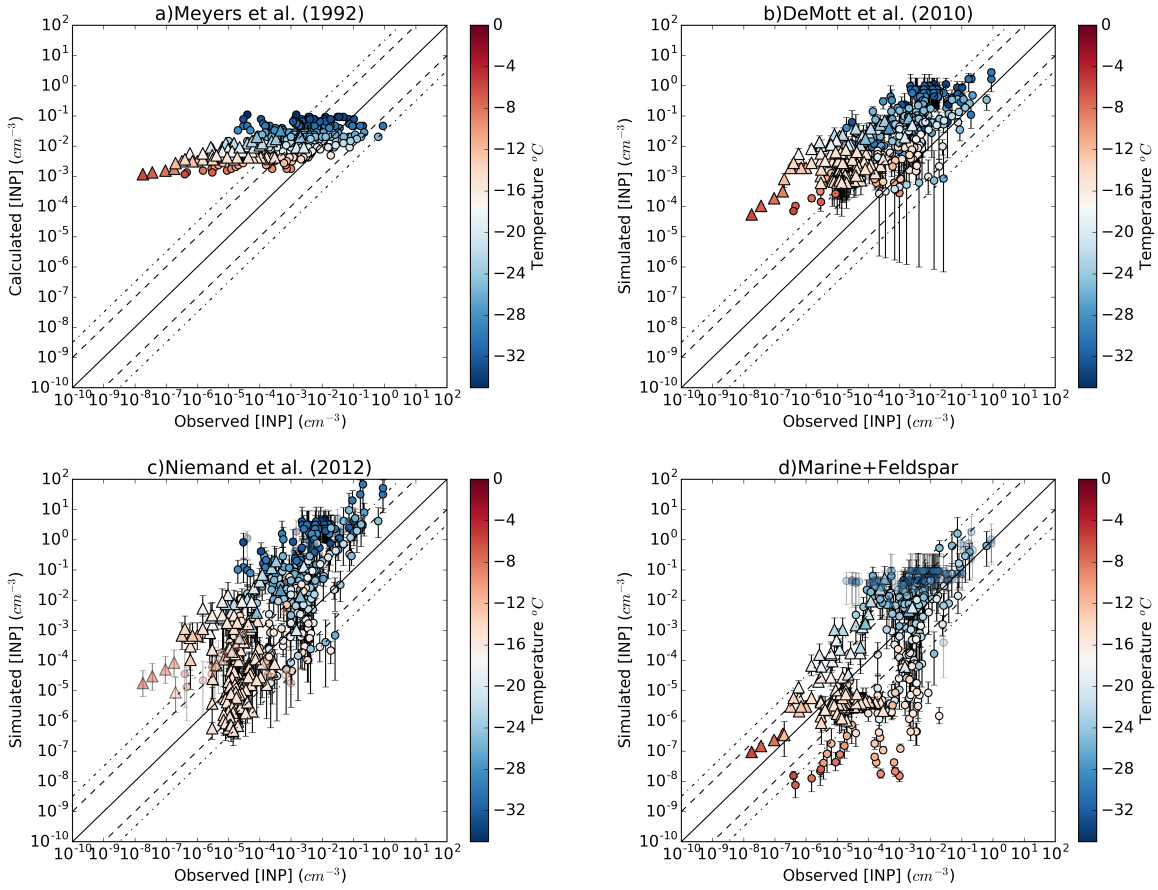

Figure 8. Comparison of the performance of a variety of INP parameterizations tested against field measurements. ~~a)Location of the data used for comparison. (a-g~~(a-d) Modeled INP concentration values when using: ~~b~~a) Meyers parameterization (Meyers et al., 1992) ~~c~~b) DeMott's parameterization (DeMott et al., 2010) combined with a global aerosol simulation using GLOMAP-mode, ~~d~~c) Niemand dust parameterization (Niemand et al., 2012), ~~e~~d) Our two-species based representation based on feldspar (Atkinson et al., 2013) and marine organic aerosols (Wilson et al., 2015). . ~~f) Same as e) but showing the relative contribution (in orders of magnitude) of each aerosol species to the simulated concentration. g) Same as e) but distinguishing between the different campaigns shown in a)(with the same colours and symbols).~~ Triangles represent marine influenced regions and points terrestrial environments. The light shaded points in d) and e) are for datapoints outside the temperature range of the parameterizations. The dashed lines represent one order of magnitude of difference between modelled and observed and the dashed-dotted lines 1.5 orders of magnitude. The simulated values correspond to an annual mean concentration and the error bars correspond to the simulated seasonality of INP calculated with monthly mean values. For ~~Niemand's dust parameterization (Niemand et al., 2012) the range of data is within the range of temperatures shown in Niemand et al. (2012) (-12 to -33^oC~~each individual observation, we calculated the INP concentration at the temperature corresponding to the temperature that aerosol particles were exposed to in the INP instruments. The locations of the datapoint are shown in figure 9.

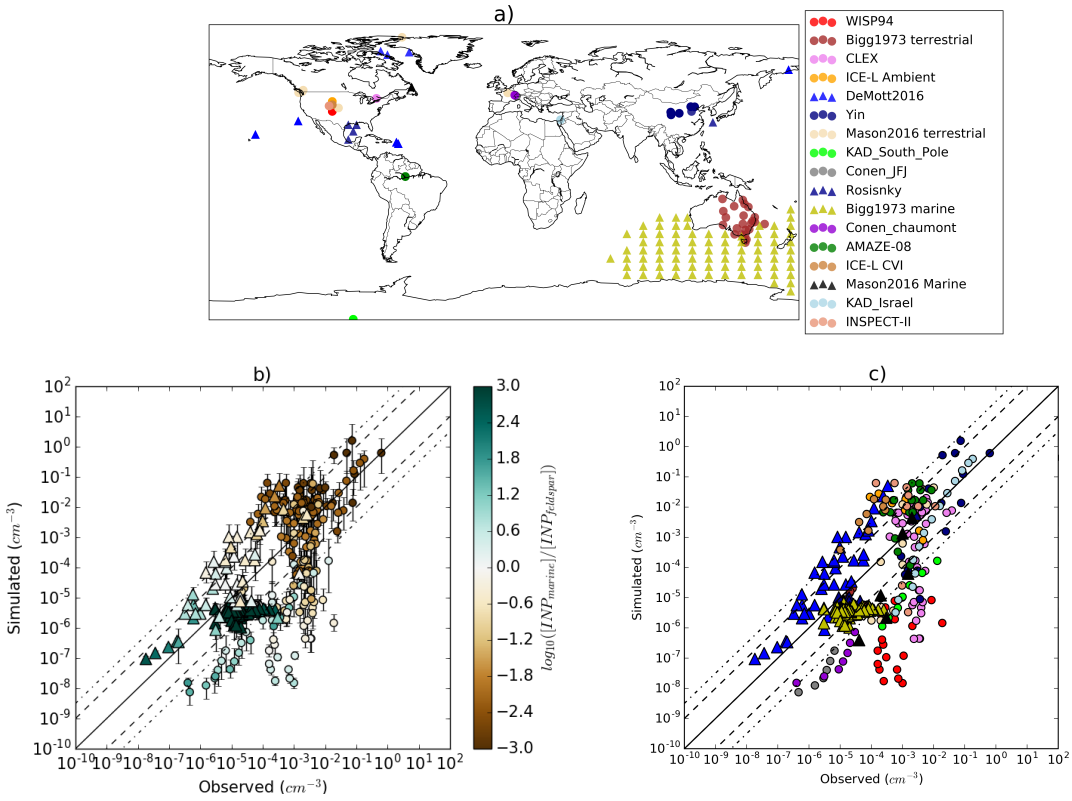

Figure 9. a) Location of the data used for comparison in figure 8. b) Same as figure 8d but showing the relative contribution (in orders of magnitude) of each aerosol species to the simulated concentration. c) Same as b but distinguishing between the different campaigns shown in a (with the same colours and symbols). References to the datasets used are shown in Appendix: C

4 Conclusions

This study is a step towards the inclusion of ~~ice nucleating~~ ice-nucleating particles in weather and climate models in a way that accounts for the aerosol chemical composition using laboratory- derived parameterizations under the singular description. ~~By using a representation of INP based on K-feldspar and marine organic aerosols, we can compare the relative importance of these two species.~~ We find that marine organic aerosols dominate the concentration of INP in remote locations like the Southern Ocean on many days, whereas feldspar particles are the dominant species for ice nucleation in places influenced by the terrestrial emission sources. However, even over northern hemisphere regions influenced by dust, marine organic INP concentrations exceed K-feldspar INP concentrations on 10-30% of the days when the temperature is within the mixed-phase range and the total concentration of INP is larger than 10^{-4} L^{-1}. Similarly, K-feldspar cannot be ruled out as an important source of INP in the southern high latitudes because there are several days per month when the concentration of transported K-feldspar INP dominates over the prevailing marine organics.

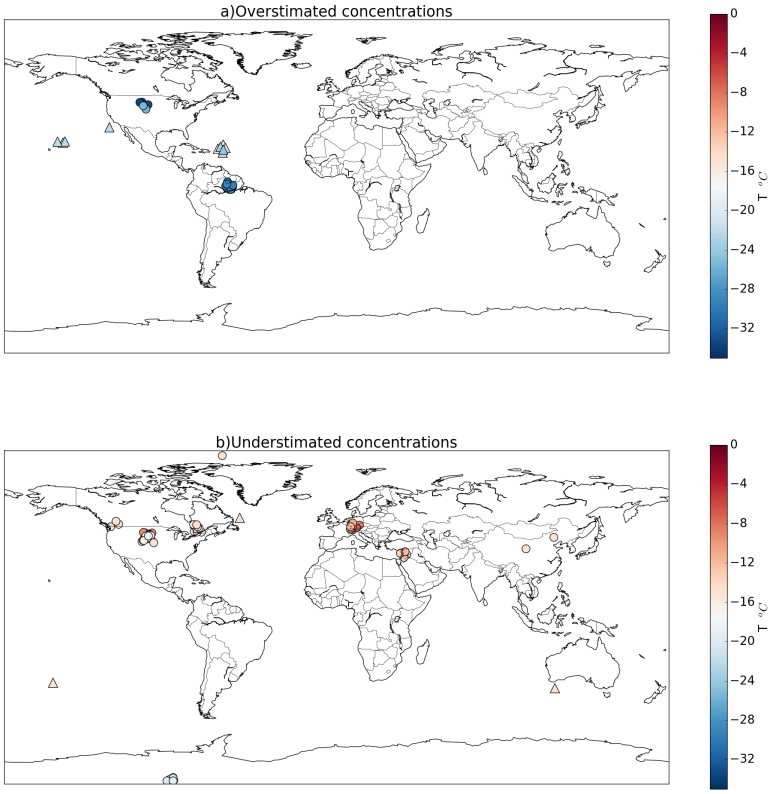

Figure 10. Overestimation and underestimation places according to our two species based parameterization of INP (Atkinson et al., 2013; Wilson et al., 2015). a) Shows the places where we overestimate the values of INP by more than 1.5 orders of magnitude. b) Similar to a but for places where the concentration is underestimated by more than 1.5 orders of magnitude. The location of the points have been moved randomly in the plot for purpose of visualization so it can be seen when the bias affect to a single data point or a whole dataset.

K-feldspar in our model can reproduce 70% of the observations of INP in terrestrial locations at low temperatures ($T < -15^oC$) within 1.5 orders of magnitude. Because K-feldspar is mainly a coarse aerosol type, it is scavenged more rapidly than the clay fraction of desert dusts, and therefore has substantially smaller influence on remote marine environments in contrast with Atkinson et al. (2013) where dust was not subject to wet removal. For remote locations, we find that marine organic aerosols acting as INP are able to reproduce a majority (80%) of the observations within an order of magnitude.

Our model of INP based on emitted and transported aerosol species provides a reasonable explanation of measured global INP concentrations, but there are some important biases. The two-species model overestimates by around 1.5 orders of magnitude the concentrations of INP in marine locations that are influenced by the transport of K-feldspar-containing dust particles, although it is difficult to draw firm conclusions from the small number of observations. Nevertheless, the bias points to the possible importance of missing processes, such as the effect of atmospheric processing of feldspar particles, a preferential

scavenging of INP as proposed in (Stopelli et al., 2015), or a possible overestimation of the transport of this aerosol type. The model also underestimates measured INP concentrations at high temperatures in some terrestrial locations. This bias is most likely to be explained by neglecting the contribution of some terrestrial biogenic aerosol species such as soil dust, fungal spores and bacteria. The model bias is large at the surface, but some studies show that some of these species are not important for ice nucleation once in the atmosphere (Spracklen and Heald, 2014; Hoose et al., 2010a) because of their low simulated concentrations above the surface for heterogeneous ice nucleation. These species however, could be important for triggering secondary production ice processes, such as the Hallet-Mossop process, due to its high nucleation temperatures. In addition, other unknown sources of ~~ice nucleating~~ ice-nucleating particles, such as biological fragments attached to mineral dust particles (O'Sullivan et al., 2015, 2016), could help explain underestimated INP concentrations in the model.

In summary, our results suggest that the inclusion of both marine organic and feldspar emissions are required to accurately simulate global INP concentrations. However, there are still large uncertainties to be resolved, such as the importance of acid coating affecting the INP ability of K-feldspar (Wex et al., 2014; Sullivan et al., 2010) or the relative importance of soot for ice nucleation in the atmosphere, which could lead to a possible anthropogenic effect on clouds.

Finally, we suggest that further experimental studies on the ~~ice nucleating~~ ice-nucleating ability of different aerosol species, followed by modelling studies of their importance in the atmosphere, will be crucial for determining the possible importance of other species for ice nucleation under atmospheric conditions. In addition, more measurements in the ambient atmosphere for different environments and seasons are necessary to better evaluate and constrain models. Among those, exploratory studies about the composition and type of ice-nucleating particles in terrestrial environments at high temperatures will be crucial to determine which species need to be included in models.

Appendix A: Marine organic emissions

In order to represent the distribution of sub-micron marine organics aerosols, first we simulate the distribution of sea-salt aerosols (SS) with GLOMAP-mode for the year 2001. Then we look at the correlation between the monthly mean emission flux of sea-salt particles in the accumulation mode ~~$(100nm < r < 1\mu m)$~~ $(100nm < r < 1\,\mu\mathrm{m})$ and the monthly mean surface concentration of sub-micron sea-salt in the grid-boxes corresponding to Mace Head and Amsterdam Island. We then take the grid-boxes that score a correlation $R > 0.9$ and assume that, as a first order approach, the emissions of these grid-boxes will drive the concentrations of sub-micron sea-spray in their corresponding stations (Fig.11). Once these grid-boxes are identified for every station, we calculate the organic mass fraction (OMF) in surface air (lowest model layer) at both stations with modelled concentrations of sea-spray and measured concentrations of water insoluble organic matter (WIOM) following Eq.A1.

$$OMF = \frac{[WIOM]}{[SS_{mass}] + [WIOM]} \tag{A1}$$

The WIOM in Mace Head data is obtained from (Rinaldi et al., 2013) by averaging measurements corresponding to a few days (from 5 to 14 days) in every month. For Amsterdam island, WIOM is derived from (Sciare et al., 2009) using a factor of

1.9 to convert from water insoluble organic carbon to WIOM (Burrows et al., 2013). The chlorophyll-a maps used correspond to monthly mean values obtained from GLOBCOLOUR (Maritorena and Siegel, 2005), which made use of data from 3 different satellites to merge their chlorophyll-a maps and produce a final product with an enhanced global coverage.

$$OMF = \frac{[WIOM]}{[SS_{mass}] + [WIOM]}$$

In order to develop a parameterization of the organic mass fraction to be used in both hemispheres we use the monthly mean values of the chlorophyll-a content in the grid-boxes previously related to each station, together with the monthly mean reanalysis (ECMWF) wind speed at 10 meters over the surface (U10M) of these grid-boxes and relate these two variables to the organic mass fraction previously calculated (Fig.12 a). We then fit the OMF to a two-dimensional equation with the wind speed and chlorophyll-a content as variables (Fig.12 b). This gives us a parameterization of the OMF emitted with sub-micron sea-spray that can fit our model. In order to avoid unrealistic OMF values due to extrapolation, we limit the maximum value of our OMF to be 0.85.

The mass flux of marine organic material can be then ~~calculated as~~from the sea-salt flux following Eq.A5:

$$Flux_{total} = Flux_{SS} + Flux_{WIOM} \tag{A2}$$

$$Flux_{WIOM} = OMF \cdot Flux_{total} \tag{A3}$$

$$Flux_{total} = \frac{Flux_{WIOM}}{OMF} \tag{A4}$$

$$Flux_{WIOM} = \frac{Flux_{SS} \cdot OMF}{1 - OMF} \tag{A5}$$

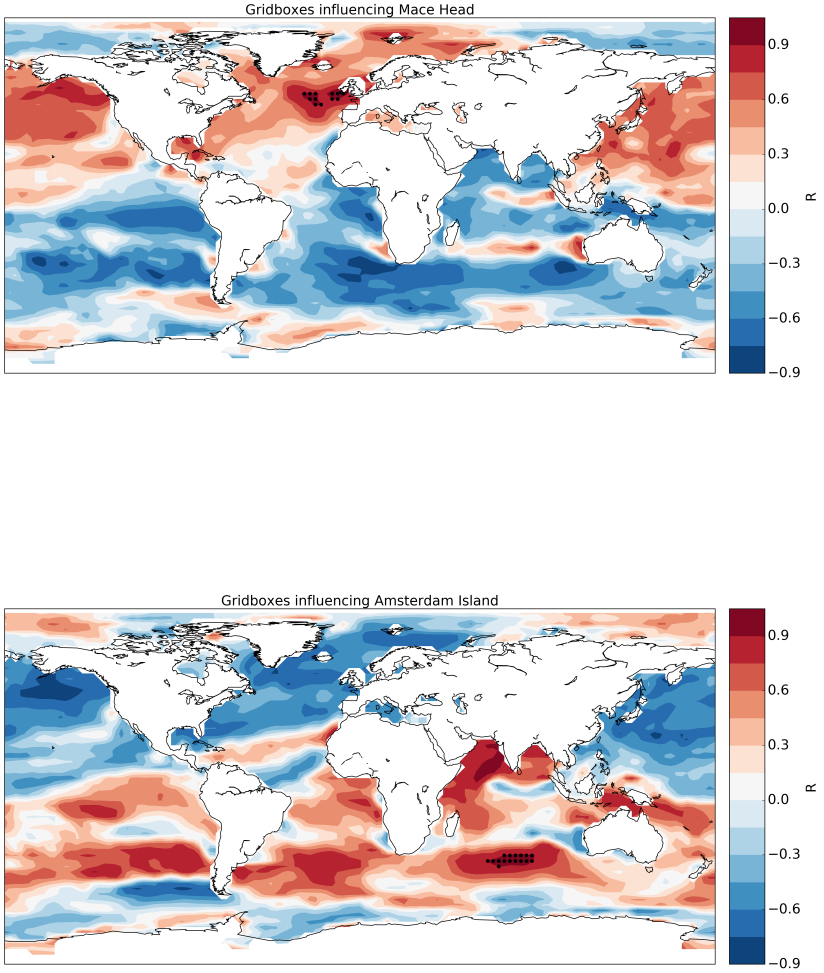

Figure 11. Linear correlation values between the monthly emission of sub-micron sea-spray and their monthly concentrations in Top panel: Mace Head, Bottom pannel: Amsterdam Island . The dots represent the grid-boxes that we relate to every station because of having a value R>0.9

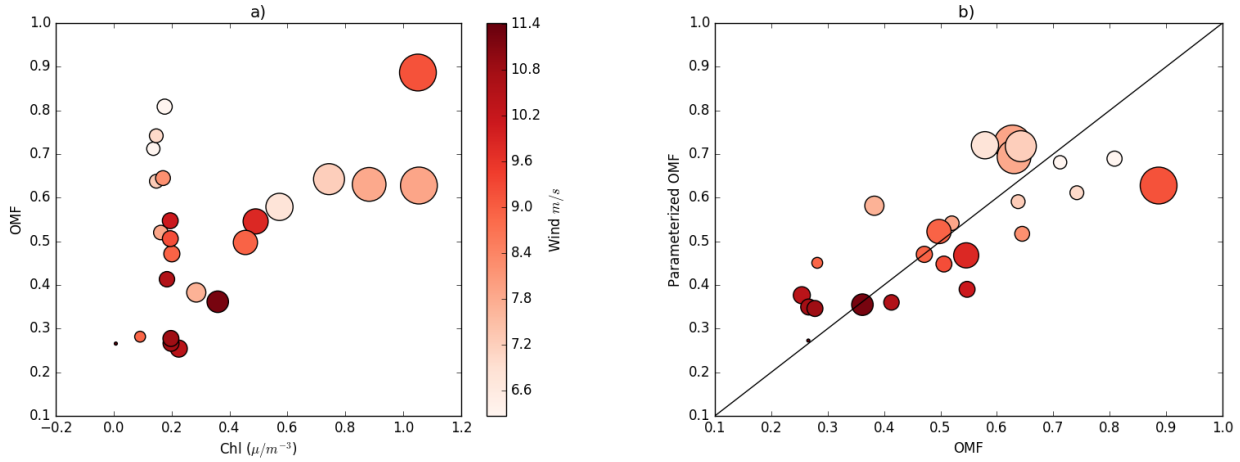

Figure 12. a) OMF compared as a function of chlorophyll-a content and surface wind speed for the monthly mean values in both stations. The size of the points represent the mean chlorophyll-a content of the grid-boxes related previously to every station (Fig.11), the colour of the points is related to the wind speed of those grid-boxes. b) Shows the performance of the parameterization for reproducing the OMF calculated with the simulated concentration of sub-micron sea-salt and the observed values of WIOM. The parameterization for the OMF is ~~$OMF = A * [CHL(mg/m^3)] + B * [U10M(m/s)]^C + D$~~ $\mathrm{OMF} = A * [CHL(mg/m^3)] + B * [U10M(m/s)]^C + D$ with $A = 0.241$, $B = -7.503$, $C = 0.075$, $D = 9.274$.

Appendix B: Calculation of INP concentrations

Assuming that the active sites from which ice nucleation can occur under the singular description are randomly distributed in the aerosol population, the probability of one particle to have a certain number of active sites (k) can be represented by the poisson distribution Eq.B1.

$$f(k,\lambda) = \frac{e^{-\lambda}\lambda^{k}}{k!} \tag{B1}$$

~~Where~~ Here f is the probability of having k active sites in a particle and λ represents the expected value of active sites per particle at a certain temperature (T). We can calculate the probability of a particle immersed in a supercooled water droplet to freeze it (P), as the sum of the probability of having 1 or more active sites in it Eq. B2:

$$P = \sum_{k=1}^{\infty} f(k,\lambda) \tag{B2}$$

As the sum from $k = 0$ to $k = \infty$ of Eq. B1 has to be equal to 1, we can also represent this sum as Eq. B3:

$$P = \sum_{k=0}^{\infty} f(k,\lambda) - f(0,\lambda) = 1 - e^{-\lambda} \tag{B3}$$

If we have a distribution of particles of the same size and same density of active sites, this probability P will be the same for all of them, and so the fraction of supercooled water droplets that will freeze know as fraction frozen (~~ff~~ff), will ~~be therefore~~ therefore be:

$$ff = 1 - e^{-\lambda} \tag{B4}$$

We can then calculate the INP concentration as:

$$[INP] = ff \cdot [N] \tag{B5}$$

where $[N]$ represent the concentration of a certain type of aerosol. For the case in which we have a density of active sites distributed across the surface area of a particle depending on temperature $n_s(T)$, we can calculate λ for a particle of radius r as:

$$\lambda(r,T) = 4\pi r^2 \cdot n_s(T) \tag{B6}$$

Hence:

$$ff(r,T) = 1 - e^{-n_s(T)\cdot 4\pi r^2} \tag{B7}$$

In GLOMAP-mode, the size distribution of aerosols is represented in log-normal modes, and their probability density function PDF is given by:

$$PDF(r) = \frac{1}{r \cdot ln(\sigma) \cdot \sqrt{2\pi}} \cdot e^{\frac{-(ln(r)-ln(r_m))^2}{2\cdot ln(\sigma)^2}} \tag{B8}$$

where r_m is the mean radius of the mode and σ the standard deviation of the mode.

The INP concentration is therefore the integral across all the possible values of r for every mode, and it will change for every temperature:

$$[INP]_{mode}(T) = \int_0^\infty (1 - e^{-4\cdot\pi\cdot r^2\cdot n_s(T)}) \cdot N \cdot \frac{1}{r \cdot ln(\sigma) \cdot \sqrt{2\pi}} \cdot e^{\frac{-(ln(r)-ln(r_m))^2}{2\cdot ln(\sigma)^2}}\, dr \tag{B9}$$

In our case, we consider that just the soluble modes can activate into water droplets, so the total INP concentration is the sum of the concentrations for every soluble mode.

In the special case of having a value of λ small ($\lambda < 0.1$) , we can approximate the value of the fraction frozen (ff) using a 1st order Taylor series centred in 0:

$$ff \approx ff_{\lambda=o} + \frac{1}{1!}\frac{\partial ff}{\partial \lambda}\bigg|_{\lambda=0} \cdot \lambda + ... \tag{B10}$$

$$ff_{\lambda=0} = 1 - e^0 = 0 \tag{B11}$$

$$\frac{\partial ff}{\partial \lambda}\bigg|_{\lambda=0} = \Big[-e^{-\lambda}\cdot(-1)\Big]_{\lambda=0} \cdot \lambda = 1 \cdot \lambda \tag{B12}$$

$$ff \approx \lambda \tag{B13}$$

In other words, if the number of active sites is small compared with the number of particles, we can approximate the number of particles having one or more actives sites, to the number of active sites. And the INP concentration can be calculated as:

$$[INP](T) \approx \lambda(T) \cdot [N] \tag{B14}$$

Campaign/dataset	Location	Marine or Terrestrial	Data points	References
Bigg73	Australia	Terrestrial	24	~~(Bigg, 1973)~~ Bigg (1973)
CLEX	East Canada	Terrestrial	60	~~(DeMott et al., 2010)~~ DeMott et al. (2010)
Yin	China	Terrestrial	21	~~(Yin et al., 2012)~~ Yin et al. (2012)
ICE-L Ambient	Central USA	Terrestrial	31	~~(DeMott et al., 2010)~~ DeMott et al. (2010)
DeMott2016	Marine locations	Marine	44	~~(DeMott et al., 2016)~~ DeMott et al. (2016)
Conen_JFJ	Jungfraujoch	Terrestrial	6	BACCHUS ~~(Conen et al., 2015)~~ Conen et al. (2015
Mason2016 terrestrial	Terrestrial locations	Terrestrial	15	~~(Mason et al., 2016)~~ Mason et al. (2016)
KAD_South_Pole	South Pole	Terrestrial	8	BACCHUS ~~(Ardon-Dryer et al., 2011)~~ Ardon-Dryer et al.
ICE-L CVI	Central USA	Terrestrial	27	~~(DeMott et al., 2010)~~ DeMott et al. (2010)
Rosisnky	Gulf of Mexico	Marine	5	~~(Rosinski et al., 1988)~~ Rosinski et al. (1988)
Bigg1973	Southern Ocean	Marine	102	~~(Bigg, 1973)~~ Bigg (1973)
Conen_chaumont	Chaumont	Terrestrial	7	BACCHUS ~~(Conen et al., 2015)~~ Conen et al. (2015
AMAZE-08	Amazon rainforest	Terrestrial	63	~~(DeMott et al., 2010)~~ DeMott et al. (2010)
INSPECT-I	Central USA	Terrestrial	13	~~(DeMott et al., 2010)~~ DeMott et al. (2010)
Mason2016 Marine	Marine locations	Marine	6	~~(Mason et al., 2016)~~ Mason et al. (2016)
KAD_Israel	Jerusalem	Terrestrial	16	BACCHUS ~~(Ardon-Dryer and Levin, 2014)~~ Ardon-Dryer and
INSPECT-II	Central USA	Terrestrial	11	~~(DeMott et al., 2010)~~ DeMott et al. (2010)

Table 2. Table of the datasets used for this study.

Appendix C: INP dataset

The dataset used in this study is a compilation of published dataset and unpublished data provided by different groups collaborating to the BACCHUS dataset of INP (http://www.bacchus-env.eu/in/index.php). ~~We contacted all the researchers with condensation-immersion freezing INP data advertised in the BACCHUS dataset at the moment of doing this study~~Table 2 shows a summary of the datasets. We note that an study from Bigg (1996) reported INP concentrations in the high Artic. We could not include it in our database as the exact locations could not be obtain. However we note that the range of concentrations reported by Bigg (1996) (from 13 m^{-3} to 2.9 m^{-3} at -15C^o) are close to our simulated values using feldspar and marine organics (from 7.4 m^{-3} to 0.1 m^{-3}) during the months as the campaign (August to October). The datasets ~~used are listed~~ obtained through the BACCHUS project database are labelled as "BACCHUS" in table 2. The datasets corresponding to long term measurements in a single location were re-sized to account for a single data point at every temperature. This is done in order to avoid statistical over-weighting of a single location or campaign.

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References

Aller, J. Y., Kuznetsova, M. R., Jahns, C. J., and Kemp, P. F.: The sea surface microlayer as a source of viral and bacterial enrichment in marine aerosols, Journal of Aerosol Science, 36, 801–812, doi:10.1016/j.jaerosci.2004.10.012, http://linkinghub.elsevier.com/retrieve/pii/S0021850204003829, 2005.

Alpert, P. A., Aller, J. Y., and Knopf, D. A.: Ice nucleation from aqueous NaCl droplets with and without marine diatoms, Atmospheric Chemistry and Physics, 11, 5539–5555, doi:10.5194/acp-11-5539-2011, http://www.atmos-chem-phys.net/11/5539/2011/, 2011.

Ardon-Dryer, K. and Levin, Z.: Ground-based measurements of immersion freezing in the eastern Mediterranean, Atmospheric Chemistry and Physics, 14, 5217–5231, doi:10.5194/acp-14-5217-2014, http://www.atmos-chem-phys.net/14/5217/2014/, 2014.

Ardon-Dryer, K., Levin, Z., and Lawson, R. P.: Characteristics of immersion freezing nuclei at the South Pole station in Antarctica, Atmospheric Chemistry and Physics, 11, 4015–4024, doi:10.5194/acp-11-4015-2011, 2011.

Atkinson, J. D., Murray, B. J., Woodhouse, M. T., Whale, T. F., Baustian, K. J., Carslaw, K. S., Dobbie, S., O'Sullivan, D., and Malkin, T. L.: The importance of feldspar for ice nucleation by mineral dust in mixed-phase clouds., Nature, 498, 355–8, doi:10.1038/nature12278, http://www.ncbi.nlm.nih.gov/pubmed/23760484, 2013.

Augustin-Bauditz, S., Wex, H., Kanter, S., Ebert, M., Niedermeier, D., Stolz, F., Prager, a., and Stratmann, F.: The immersion mode ice nucleation behavior of mineral dusts: A comparison of different pure and surface modified dusts, Geophysical Research Letters, 41, 7375–7382, doi:10.1002/2014GL061317, 2014.

Bauer, P., Thorpe, A., and Brunet, G.: The quiet revolution of numerical weather prediction, Nature, 525, 47–55, doi:10.1038/nature14956, http://www.nature.com/doifinder/10.1038/nature14956, 2015.

Bigg, E. K.: Ice Nucleus Concentrations in Remote Areas, Journal of the Atmospheric Sciences, 30, 1153–1157, doi:10.1175/1520-0469(1973)030<1153:INCIRA>2.0.CO;2, 1973.

Bigg, E. K.: Ice forming nuclei in the high Arctic, Tellus B, 48, 223–233, doi:10.1034/j.1600-0889.1996.t01-1-00007.x, http://www.tellusb.net/index.php/tellusb/article/view/15888, 1996.

Bodas-Salcedo, a., Williams, K. D., Ringer, M. a., Beau, I., Cole, J. N. S., Dufresne, J. L., Koshiro, T., Stevens, B., Wang, Z., and Yokohata, T.: Origins of the solar radiation biases over the Southern Ocean in CFMIP2 models, Journal of Climate, 27, 41–56, doi:10.1175/JCLI-D-13-00169.1, 2014.

Boose, Y., Sierau, B., García, M. I., Rodríguez, S., Alastuey, A., Linke, C., Schnaiter, M., Kupiszewski, P., Kanji, Z. A., and Lohmann, U.: Ice nucleating particles in the Saharan Air Layer, Atmospheric Chemistry and Physics, 16, 9067–9087, doi:10.5194/acp-16-9067-2016, http://www.atmos-chem-phys.net/16/9067/2016/, 2016.

Broadley, S. L., Murray, B. J., Herbert, R. J., Atkinson, J. D., Dobbie, S., Malkin, T. L., Condliffe, E., and Neve, L.: Immersion mode heterogeneous ice nucleation by an illite rich powder representative of atmospheric mineral dust, Atmospheric Chemistry and Physics, 12, 287–307, doi:10.5194/acp-12-287-2012, http://www.atmos-chem-phys.net/12/287/2012/, 2012.

Browse, J., Carslaw, K. S., Arnold, S. R., Pringle, K., and Boucher, O.: The scavenging processes controlling the seasonal cycle in Arctic sulphate and black carbon aerosol, Atmospheric Chemistry and Physics, 12, 6775–6798, doi:10.5194/acp-12-6775-2012, http://www.atmos-chem-phys.net/12/6775/2012/, 2012.

Burrows, S. M., Hoose, C., Pöschl, U., and Lawrence, M. G.: Ice nuclei in marine air: biogenic particles or dust?, Atmospheric Chemistry and Physics, 13, 245–267, doi:10.5194/acp-13-245-2013, http://www.atmos-chem-phys.net/13/245/2013/, 2013.

Carslaw, K. S., Lee, L. a., Reddington, C. L., Pringle, K. J., Rap, a., Forster, P. M., Mann, G. W., Spracklen, D. V., Woodhouse, M. T., Regayre, L. a., and Pierce, J. R.: Large contribution of natural aerosols to uncertainty in indirect forcing., Nature, 503, 67–71, doi:10.1038/nature12674, http://www.ncbi.nlm.nih.gov/pubmed/24201280, 2013.

Choi, Y.-S., Lindzen, R. S., Ho, C.-H., and Kim, J.: Space observations of cold-cloud phase change., Proceedings of the National Academy of Sciences of the United States of America, 107, 11 211–6, doi:10.1073/pnas.1006241107, http://www.pubmedcentral.nih.gov/articlerender.fcgi?artid=2895094{\&}tool=pmcentrez{\&}rendertype=abstract, 2010.

Claquin, T., Schulz, M., and Balkanski, Y. J.: Modeling the mineralogy of atmospheric dust sources, Journal of Geophysical Research, 104, 22 243, doi:10.1029/1999JD900416, 1999.

Cochran, R. E., Jayarathne, T., Stone, E. A., and Grassian, V. H.: Selectivity Across the Interface: A Test of Surface Activity in the Composition of Organic-Enriched Aerosols from Bubble Bursting, The Journal of Physical Chemistry Letters, 7, 1692–1696, doi:10.1021/acs.jpclett.6b00489, http://dx.doi.org/10.1021/acs.jpclett.6b00489http://pubs.acs.org/doi/abs/10.1021/acs.jpclett.6b00489, 2016.

Conen, F., Rodríguez, S., Hüglin, C., Henne, S., Herrmann, E., Bukowiecki, N., and Alewell, C.: Atmospheric ice nuclei at the high-altitude observatory Jungfraujoch, Switzerland, Tellus B, 67, 1–10, doi:10.3402/tellusb.v67.25014, http://www.tellusb.net/index.php/tellusb/article/view/25014, 2015.

Cunliffe, M., Engel, A., Frka, S., Gašparović, B., Guitart, C., Murrell, J. C., Salter, M., Stolle, C., Upstill-Goddard, R., and Wurl, O.: Sea surface microlayers: A unified physicochemical and biological perspective of the air ocean interface, Progress in Oceanography, 109, 104–116, doi:10.1016/j.pocean.2012.08.004, http://linkinghub.elsevier.com/retrieve/pii/S0079661112000924, 2013.

DeMott, P. J., Prenni, a. J., Liu, X., Kreidenweis, S. M., Petters, M. D., Twohy, C. H., Richardson, M. S., Eidhammer, T., and Rogers, D. C.: Predicting global atmospheric ice nuclei distributions and their impacts on climate., Proceedings of the National Academy of Sciences of the United States of America, 107, 11 217–11 222, doi:10.1073/pnas.0910818107, 2010.

DeMott, P. J., Prenni, A. J., McMeeking, G. R., Sullivan, R. C., Petters, M. D., Tobo, Y., Niemand, M., Möhler, O., Snider, J. R., Wang, Z., and Kreidenweis, S. M.: Integrating laboratory and field data to quantify the immersion freezing ice nucleation activity of mineral dust particles, Atmospheric Chemistry and Physics, 15, 393–409, doi:10.5194/acp-15-393-2015, http://www.atmos-chem-phys.net/15/393/2015/, 2015.

DeMott, P. J., Hill, T. C., McCluskey, C. S., Prather, K. A., Collins, D. B., Sullivan, R. C., Ruppel, M. J., Mason, R. H., Irish, V. E., Lee, T., et al.: Sea spray aerosol as a unique source of ice nucleating particles, Proceedings of the National Academy of Sciences, 113, 5797–5803, 2016.

Dentener, F. and Kinne, S.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, … and Physics, pp. 4321–4344, http://www.atmos-chem-phys.net/6/4321/2006/acp-6-4321-2006.pdf, 2006.

Eidhammer, T., DeMott, P. J., and Kreidenweis, S. M.: A comparison of heterogeneous ice nucleation parameterizations using a parcel model framework, Journal of Geophysical Research, 114, D06 202, doi:10.1029/2008JD011095, http://doi.wiley.com/10.1029/2008JD011095, 2009.

Emersic, C., Connolly, P. J., Boult, S., Campana, M., and Li, Z.: Investigating the discrepancy between wet-suspension- and dry-dispersion-derived ice nucleation efficiency of mineral particles, Atmospheric Chemistry and Physics, 15, 11 311–11 326, doi:10.5194/acp-15-11311-2015, http://www.atmos-chem-phys.net/15/11311/2015/, 2015.

Facchini, M. C., Rinaldi, M., Decesari, S., Carbone, C., Finessi, E., Mircea, M., Fuzzi, S., Ceburnis, D., Flanagan, R., Nilsson, E. D., de Leeuw, G., Martino, M., Woeltjen, J., and O'Dowd, C. D.: Primary submicron marine aerosol dominated by insoluble organic colloids

and aggregates, Geophysical Research Letters, 35, L17 814, doi:10.1029/2008GL034210, http://doi.wiley.com/10.1029/2008GL034210, 2008.

Fall, R. and Schnell, R. C.: Association of an ice-nucleating pseudomonad with cultures of the marine dinoflagellate, <I>Heterocapsa niei</I>, Journal of Marine Research, 43, 257–265, doi:10.1357/002224085788437370, http://openurl.ingenta.com/content/xref?genre=article\&issn=0022-2402\&volume=43\&issue=1\&spage=257, 1985.

Field, P. R., Heymsfield, A. J., Shipway, B. J., DeMott, P. J., Pratt, K. a., Rogers, D. C., Stith, J., and Prather, K. a.: Ice in Clouds Experiment Layer Clouds. Part II: Testing Characteristics of Heterogeneous Ice Formation in Lee Wave Clouds, Journal of the Atmospheric Sciences, 69, 1066–1079, doi:10.1175/JAS-D-11-026.1, http://journals.ametsoc.org/doi/abs/10.1175/JAS-D-11-026.1, 2012.

Fröhlich-Nowoisky, J., Hill, T. C. J., Pummer, B. G., Yordanova, P., Franc, G. D., and Pöschl, U.: Ice nucleation activity in the widespread soil fungus Mortierella alpina, Biogeosciences, 12, 1057–1071, doi:10.5194/bg-12-1057-2015, http://www.biogeosciences.net/12/1057/2015/, 2015.

Fuentes, E., Coe, H., Green, D., and McFiggans, G.: On the impacts of phytoplankton-derived organic matter on the properties of the primary marine aerosol Part 2: Composition, hygroscopicity and cloud condensation activity, Atmospheric Chemistry and Physics, 11, 2585–2602, doi:10.5194/acp-11-2585-2011, http://www.atmos-chem-phys.net/11/2585/2011/, 2011.

Gantt, B., Meskhidze, N., Facchini, M. C., Rinaldi, M., Ceburnis, D., and O'Dowd, C. D.: Wind speed dependent size-resolved parameterization for the organic mass fraction of sea spray aerosol, Atmospheric Chemistry and Physics, 11, 8777–8790, doi:10.5194/acp-11-8777-2011, 2011.

Garcia, E., Hill, T. C. J., Prenni, A. J., DeMott, P. J., Franc, G. D., and Kreidenweis, S. M.: Biogenic ice nuclei in boundary layer air over two U.S. High Plains agricultural regions, Journal of Geophysical Research: Atmospheres, 117, n/a–n/a, doi:10.1029/2012JD018343, http://doi.wiley.com/10.1029/2012JD018343, 2012.

Ghan, S. J. and Schwartz, S. E.: Aerosol Properties and Processes: A Path from Field and Laboratory Measurements to Global Climate Models, Bulletin of the American Meteorological Society, 88, 1059–1083, doi:10.1175/BAMS-88-7-1059, http://journals.ametsoc.org/doi/abs/10.1175/BAMS-88-7-1059, 2007.

Gong, S. L.: A parameterization of sea-salt aerosol source function for sub- and super-micron particles, Global Biogeochemical Cycles, 17, n/a–n/a, doi:10.1029/2003GB002079, http://doi.wiley.com/10.1029/2003GB002079, 2003.

Haga, D., Burrows, S., Iannone, R., Wheeler, M., Mason, R., Chen, J., Polishchuk, E., Pöschl, U., and Bertram, A.: Ice nucleation by fungal spores from the classes Agaricomycetes, Ustilaginomycetes, and Eurotiomycetes, and the effect on the atmospheric transport of these spores, Atmos. Chem. Phys., 14, 8611–8630, doi:doi:10.5194/acp-14-8611-2014, http://www.atmos-chem-phys.net/14/8611/2014/acp-14-8611-2014.html, 2014.

Haga, D. I., Iannone, R., Wheeler, M. J., Mason, R., Polishchuk, E. A., Fetch, T., Van Der Kamp, B. J., McKendry, I. G., and Bertram, A. K.: Ice nucleation properties of rust and bunt fungal spores and their transport to high altitudes, where they can cause heterogeneous freezing, Journal of Geophysical Research Atmospheres, 118, 7260–7272, doi:10.1002/jgrd.50556, 2013.

Hallett, J. and Mossop, S. C.: Production of secondary ice particles during the riming process, doi:10.1038/249026a0, 1974.

Harrison, A. D., Whale, T. F., Carpenter, M. A., Holden, M. A., Neve, L., O'Sullivan, D., Vergara Temprado, J., and Murray, B. J.: Not all feldspars are equal: a survey of ice nucleating properties across the feldspar group of minerals, Atmospheric Chemistry and Physics, 16, 10 927–10 940, doi:10.5194/acp-16-10927-2016, http://www.atmos-chem-phys.net/16/10927/2016/, 2016.

Hartmann, S., Augustin, S., Clauss, T., Wex, H., Šantl Temkiv, T., Voigtländer, J., Niedermeier, D., and Stratmann, F.: Immersion freezing of ice nucleation active protein complexes, Atmospheric Chemistry and Physics, 13, 5751–5766, doi:10.5194/acp-13-5751-2013, http://www.atmos-chem-phys.net/13/5751/2013/, 2013.

Herbert, R. J., Murray, B. J., Whale, T. F., Dobbie, S. J., and Atkinson, J. D.: Representing time-dependent freezing behaviour in immersion mode ice nucleation, Atmospheric Chemistry and Physics, 14, 8501–8520, doi:10.5194/acp-14-8501-2014, http://www.atmos-chem-phys.net/14/8501/2014/, 2014.

Herbert, R. J., Murray, B. J., Dobbie, S. J., and Koop, T.: Sensitivity of liquid clouds to homogenous freezing parameterizations, Geophysical Research Letters, 42, 1599–1605, doi:10.1002/2014GL062729, http://doi.wiley.com/10.1002/2014GL062729, 2015.

Hoose, C. and Möhler, O.: Heterogeneous ice nucleation on atmospheric aerosols: A review of results from laboratory experiments, doi:10.5194/acp-12-9817-2012, http://www.atmos-chem-phys.net/12/9817/2012/, 2012.

Hoose, C., Kristjánsson, J. E., and Burrows, S. M.: How important is biological ice nucleation in clouds on a global scale?, doi:10.1088/1748-9326/5/2/024009, http://stacks.iop.org/1748-9326/5/i=2/a=024009?key=crossref.7c17e416dc1fe6881839166d060f7a64, 2010a.

Hoose, C., Kristjánsson, J. E., Chen, J.-P., and Hazra, A.: A Classical-Theory-Based Parameterization of Heterogeneous Ice Nucleation by Mineral Dust, Soot, and Biological Particles in a Global Climate Model, Journal of the Atmospheric Sciences, 67, 2483–2503, doi:10.1175/2010JAS3425.1, http://journals.ametsoc.org/doi/abs/10.1175/2010JAS3425.1, 2010b.

Huneeus, N. and Schulz, M.: Global dust model intercomparison in AeroCom phase I, Atmospheric Chemistry and Physics, 11, 7781–7816, doi:10.5194/acp-11-7781-2011, http://www.atmos-chem-phys.net/11/7781/2011/https://escholarship.org/uc/item/6g03v2hm.pdf, 2011.

Knopf, D. a., Alpert, P. a., Wang, B., and Aller, J. Y.: Stimulation of ice nucleation by marine diatoms, Nature Geoscience, 4, 88–90, doi:10.1038/ngeo1037, http://www.nature.com/doifinder/10.1038/ngeo1037, 2010.

Kodros, J. K., Scott, C. E., Farina, S. C., Lee, Y. H., L'Orange, C., Volckens, J., and Pierce, J. R.: Uncertainties in global aerosols and climate effects due to biofuel emissions, Atmospheric Chemistry and Physics, 15, 8577–8596, doi:10.5194/acp-15-8577-2015, 2015.

Koehler, K. A., Kreidenweis, S. M., DeMott, P. J., Petters, M. D., Prenni, A. J., and Möhler, O.: Laboratory investigations of the impact of mineral dust aerosol on cold cloud formation, Atmospheric Chemistry and Physics, 10, 11 955–11 968, doi:10.5194/acp-10-11955-2010, http://www.atmos-chem-phys.net/10/11955/2010/, 2010.

Korolev, A.: Limitations of the Wegener-Bergeron-Findeisen Mechanism in the Evolution of Mixed-Phase Clouds, Journal of the Atmospheric Sciences, 64, 3372–3375, doi:10.1175/JAS4035.1, 2007.

Lafon, S., Rajot, J. L., Alfaro, S. C., and Gaudichet, A.: Quantification of iron oxides in desert aerosol, Atmospheric Environment, 38, 1211–1218, doi:10.1016/j.atmosenv.2003.11.006, 2004.

Lohmann, U. and Diehl, K.: Sensitivity studies of the importance of dust ice nuclei for the indirect aerosol effect on stratiform mixed-phase clouds, Journal of the Atmospheric Sciences, pp. 968–982, http://journals.ametsoc.org/doi/abs/10.1175/JAS3662.1, 2006.

Maki, L. R. and Willoughby, K. J.: Bacteria as Biogenic Sources of Freezing Nuclei, Journal of Applied Meteorology, 17, 1049–1053, doi:10.1175/1520-0450(1978)017<1049:BABSOF>2.0.CO;2, 1978.

Mann, G. W., Carslaw, K. S., Spracklen, D. V., Ridley, D. a., Manktelow, P. T., Chipperfield, M. P., Pickering, S. J., and Johnson, C. E.: Description and evaluation of GLOMAP-mode: a modal global aerosol microphysics model for the UKCA composition-climate model, Geoscientific Model Development, 3, 519–551, doi:10.5194/gmd-3-519-2010, http://www.geosci-model-dev.net/3/519/2010/, 2010.

Mann, G. W., Carslaw, K. S., Reddington, C. L., Pringle, K. J., Schulz, M., Asmi, A., Spracklen, D. V., Ridley, D. A., Woodhouse, M. T., Lee, L. A., Zhang, K., Ghan, S. J., Easter, R. C., Liu, X., Stier, P., Lee, Y. H., Adams, P. J., Tost, H., Lelieveld, J., Bauer, S. E., Tsigaridis, K., Van Noije, T. P. C., Strunk, A., Vignati, E., Bellouin, N., Dalvi, M., Johnson, C. E., Bergman, T., Kokkola, H., Von Salzen, K., Yu, F.,

Luo, G., Petzold, A., Heintzenberg, J., Clarke, A., Ogren, J. A., Gras, J., Baltensperger, U., Kaminski, U., Jennings, S. G., O'Dowd, C. D., Harrison, R. M., Beddows, D. C. S., Kulmala, M., Viisanen, Y., Ulevicius, V., Mihalopoulos, N., Zdimal, V., Fiebig, M., Hansson, H. C., Swietlicki, E., and Henzing, J. S.: Intercomparison and evaluation of global aerosol microphysical properties among AeroCom models of a range of complexity, Atmospheric Chemistry and Physics, 14, 4679–4713, doi:10.5194/acp-14-4679-2014, 2014.

Marcolli, C., Gedamke, S., Peter, T., and Zobrist, B.: Efficiency of immersion mode ice nucleation on surrogates of mineral dust, Atmospheric Chemistry and Physics Discussions, 7, 9687–9716, doi:10.5194/acpd-7-9687-2007, http://www.atmos-chem-phys-discuss.net/7/9687/2007/, 2007.

Maritorena, S. and Siegel, D. a.: Consistent merging of satellite ocean color data sets using a bio-optical model, Remote Sensing of Environment, 94, 429–440, doi:10.1016/j.rse.2004.08.014, http://linkinghub.elsevier.com/retrieve/pii/S0034425704002834, 2005.

Mason, R. H., Si, M., Chou, C., Irish, V. E., Dickie, R., Elizondo, P., Wong, R., Brintnell, M., Elsasser, M., Lassar, W. M., Pierce, K. M., Leaitch, W. R., MacDonald, A. M., Platt, A., Toom-Sauntry, D., Sarda-Estève, R., Schiller, C. L., Suski, K. J., Hill, T. C. J., Abbatt, J. P. D., Huffman, J. A., DeMott, P. J., and Bertram, A. K.: Size-resolved measurements of ice-nucleating particles at six locations in North America and one in Europe, Atmospheric Chemistry and Physics, 16, 1637–1651, doi:10.5194/acp-16-1637-2016, http://www.atmos-chem-phys.net/16/1637/2016/, 2016.

McCoy, D. T., Burrows, S. M., Wood, R., Grosvenor, D. P., Elliott, S. M., Ma, P.-L., Rasch, P. J., and Hartmann, D. L.: Natural aerosols explain seasonal and spatial patterns of Southern Ocean cloud albedo, Science Advances, 1, e1500 157–e1500 157, doi:10.1126/sciadv.1500157, http://advances.sciencemag.org/content/1/6/e1500157.abstract, 2015a.

McCoy, D. T., Hartmann, D. L., Zelinka, M. D., Ceppi, P., and Grosvenor, D. P.: Mixed-phase cloud physics and Southern Ocean cloud feedback in climate models, Journal of Geophysical Research: Atmospheres, 120, 9539–9554, doi:10.1002/2015JD023603.Received, 2015b.

Meyers, M. P., DeMott, P. J., and Cotton, W. R.: New Primary Ice-Nucleation Parameterizations in an Explicit Cloud Model, Journal of Applied Meteorology, 31, 708–721, doi:10.1175/1520-0450(1992)031<0708:NPINPI>2.0.CO;2, 1992.

Möhler, O., Georgakopoulos, D. G., Morris, C. E., Benz, S., Ebert, V., Hunsmann, S., Saathoff, H., Schnaiter, M., and Wagner, R.: Heterogeneous ice nucleation activity of bacteria: new laboratory experiments at simulated cloud conditions, Biogeosciences Discussions, 5, 1445–1468, doi:10.5194/bgd-5-1445-2008, http://www.biogeosciences.net/5/1425/http://www.biogeosciences-discuss.net/5/1445/2008/, 2008.

Monahan, E. C., Spiel, D. E., and Davidson, K. L.: A Model of Marine Aerosol Generation Via Whitecaps and Wave Disruption, pp. 167–174, Springer Netherlands, Dordrecht, doi:10.1007/978-94-009-4668-2_16, 1986.

Morris, C. E., Sands, D. C., Glaux, C., Samsatly, J., Asaad, S., Moukahel, A. R., Gonçalves, F. L. T., and Bigg, E. K.: Urediospores of rust fungi are ice nucleation active at 10 C and harbor ice nucleation active bacteria, Atmospheric Chemistry and Physics, 13, 4223–4233, doi:10.5194/acp-13-4223-2013, http://www.atmos-chem-phys.net/13/4223/2013/, 2013.

Morrison, H., de Boer, G., Feingold, G., Harrington, J., Shupe, M. D., and Sulia, K.: Resilience of persistent Arctic mixed-phase clouds, Nature Geoscience, 5, 11–17, doi:10.1038/ngeo1332, http://dx.doi.org/10.1038/ngeo1332, 2011.

Murphy, D. M. and Koop, T.: Review of the vapour pressures of ice and supercooled water for atmospheric applications, Quarterly Journal of the Royal Meteorological Society, 131, 1539–1565, doi:10.1256/qj.04.94, 2005.

Murray, B. J., Broadley, S. L., Wilson, T. W., Atkinson, J. D., and Wills, R. H.: Heterogeneous freezing of water droplets containing kaolinite particles, Atmospheric Chemistry and Physics, 11, 4191–4207, doi:10.5194/acp-11-4191-2011, http://www.atmos-chem-phys.net/11/4191/2011/, 2011.

Murray, B. J., O'Sullivan, D., Atkinson, J. D., and Webb, M. E.: Ice nucleation by particles immersed in supercooled cloud droplets, Chemical Society Reviews, 41, 6519, doi:10.1039/c2cs35200a, http://www.ncbi.nlm.nih.gov/pubmed/22932664, 2012.

Nickovic, S., Vukovic, a., Vujadinovic, M., Djurdjevic, V., and Pejanovic, G.: Technical Note: High-resolution mineralogical database of dust-productive soils for atmospheric dust modeling, Atmospheric Chemistry and Physics, 12, 845–855, doi:10.5194/acp-12-845-2012, http://www.atmos-chem-phys.net/12/845/2012/acp-12-845-2012.html, 2012.

Niedermeier, D., Shaw, R. A., Hartmann, S., Wex, H., Clauss, T., Voigtländer, J., and Stratmann, F.: Heterogeneous ice nucleation: exploring the transition from stochastic to singular freezing behavior, Atmospheric Chemistry and Physics, 11, 8767–8775, doi:10.5194/acp-11-8767-2011, http://www.atmos-chem-phys.net/11/8767/2011/, 2011.

Niedermeier, D., Augustin-bauditz, S., Hartmann, S., Wex, H., and Stratmann, F.: Can we define an asymptotic value for the ice active surface site density for heterogeneous ice nucleation ?, Journal of Geophysical Research: Atmospheres, 120, 5036–5046, doi:10.1002/2014JD022814.The, 2015.

Niemand, M., Möhler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., Skrotzki, J., and Leisner, T.: A Particle-Surface-Area-Based Parameterization of Immersion Freezing on Desert Dust Particles, Journal of the Atmospheric Sciences, 69, 3077–3092, doi:10.1175/JAS-D-11-0249.1, http://journals.ametsoc.org/doi/abs/10.1175/JAS-D-11-0249.1, 2012.

O'Dowd, C., Ceburnis, D., Ovadnevaite, J., Bialek, J., Stengel, D. B., Zacharias, M., Nitschke, U., Connan, S., Rinaldi, M., Fuzzi, S., Decesari, S., Cristina Facchini, M., Marullo, S., Santoleri, R., Dell'Anno, A., Corinaldesi, C., Tangherlini, M., and Danovaro, R.: Connecting marine productivity to sea-spray via nanoscale biological processes: Phytoplankton Dance or Death Disco?, Scientific reports, 5, 14 883, doi:10.1038/srep14883, http://www.nature.com/srep/2015/151014/srep14883/full/srep14883.html, 2015.

Orellana, M. V., Matrai, P. A., Leck, C., Rauschenberg, C. D., Lee, A. M., and Coz, E.: Marine microgels as a source of cloud condensation nuclei in the high Arctic, Proceedings of the National Academy of Sciences, 108, 13 612–13 617, doi:10.1073/pnas.1102457108, http://www.pnas.org/content/108/33/13612http://www.pnas.org/cgi/doi/10.1073/pnas.1102457108, 2011.

O'Sullivan, D., Murray, B. J., Malkin, T. L., Whale, T. F., Umo, N. S., Atkinson, J. D., Price, H. C., Baustian, K. J., Browse, J., and Webb, M. E.: Ice nucleation by fertile soil dusts: relative importance of mineral and biogenic components, Atmospheric Chemistry and Physics, 14, 1853–1867, doi:10.5194/acp-14-1853-2014, http://www.atmos-chem-phys.net/14/1853/2014/, 2014.

O'Sullivan, D., Murray, B. J., Ross, J. F., Whale, T. F., Price, H. C., Atkinson, J. D., Umo, N. S., and Webb, M. E.: The relevance of nanoscale biological fragments for ice nucleation in clouds, Scientific Reports, 5, 8082, doi:10.1038/srep08082, http://www.nature.com/doifinder/10.1038/srep08082, 2015.

O'Sullivan, D., Murray, B. J., Ross, J. F., and Webb, M. E.: The adsorption of fungal ice-nucleating proteins on mineral dusts: a terrestrial reservoir of atmospheric ice-nucleating particles, Atmospheric Chemistry and Physics, 16, 7879–7887, doi:10.5194/acp-16-7879-2016, http://www.atmos-chem-phys.net/16/7879/2016/, 2016.

Ovadnevaite, J., O'Dowd, C., Dall'Osto, M., Ceburnis, D., Worsnop, D. R., and Berresheim, H.: Detecting high contributions of primary organic matter to marine aerosol: A case study, Geophysical Research Letters, 38, n/a–n/a, doi:10.1029/2010GL046083, http://doi.wiley.com/10.1029/2010GL046083, 2011.

Partanen, I., Dunne, E. M., Bergman, T., Laakso, a., Kokkola, H., Ovadnevaite, J., Sogacheva, L., Baisnée, D., Sciare, J., Manders, a., O'Dowd, C., de Leeuw, G., and Korhonen, H.: Global modelling of direct and indirect effects of sea spray aerosol using a source function encapsulating wave state, Atmospheric Chemistry and Physics, 14, 11 731–11 752, doi:10.5194/acp-14-11731-2014, http://www.atmos-chem-phys.net/14/11731/2014/, 2014.

Perlwitz, J. P., Pérez García-Pando, C., and Miller, R. L.: Predicting the mineral composition of dust aerosols – Part 1: Representing key processes, Atmospheric Chemistry and Physics, 15, 11 593–11 627, doi:10.5194/acp-15-11593-2015, http://www.atmos-chem-phys.net/15/11593/2015/, 2015.

Pouleur, S., Richard, C., Martin, J., and Antoun, H.: Ice nucleation activity in Fusarium acuminatum and Fusarium avenaceum, Applied and Environmental Microbiology, 58, 2960–2964, 1992.

Pummer, B. G., Budke, C., Augustin-Bauditz, S., Niedermeier, D., Felgitsch, L., Kampf, C. J., Huber, R. G., Liedl, K. R., Loerting, T., Moschen, T., Schauperl, M., Tollinger, M., Morris, C. E., Wex, H., Grothe, H., Pöschl, U., Koop, T., and Fröhlich-Nowoisky, J.: Ice nucleation by water-soluble macromolecules, Atmospheric Chemistry and Physics, 15, 4077–4091, doi:10.5194/acp-15-4077-2015, http://www.atmos-chem-phys.net/15/4077/2015/, 2015.

Quinn, P. K., Bates, T. S., Schulz, K. S., Coffman, D. J., Frossard, a. a., Russell, L. M., Keene, W. C., and Kieber, D. J.: Contribution of sea surface carbon pool to organic matter enrichment in sea spray aerosol, Nature Geoscience, 7, 228–232, doi:10.1038/ngeo2092, http://www.nature.com/doifinder/10.1038/ngeo2092, 2014.

Rap, A., Scott, C. E., Spracklen, D. V., Bellouin, N., Forster, P. M., Carslaw, K. S., Schmidt, A., and Mann, G.: Natural aerosol direct and indirect radiative effects, Geophysical Research Letters, 40, 3297–3301, doi:10.1002/grl.50441, 2013.

Riechers, B., Wittbracht, F., Hütten, A., and Koop, T.: The homogeneous ice nucleation rate of water droplets produced in a microfluidic device and the role of temperature uncertainty, Physical Chemistry Chemical Physics, 15, 5873, doi:10.1039/c3cp42437e, http://www.ncbi.nlm.nih.gov/pubmed/23486888http://xlink.rsc.org/?DOI=c3cp42437e, 2013.

Rinaldi, M., Fuzzi, S., Decesari, S., Marullo, S., Santoleri, R., Provenzale, A., von Hardenberg, J., Ceburnis, D., Vaishya, A., O'Dowd, C. D., and Facchini, M. C.: Is chlorophyll- a the best surrogate for organic matter enrichment in submicron primary marine aerosol?, Journal of Geophysical Research: Atmospheres, 118, 4964–4973, doi:10.1002/jgrd.50417, http://doi.wiley.com/10.1002/jgrd.50417, 2013.

Rosenfeld, D., Yu, X., Liu, G., Xu, X., Zhu, Y., Yue, Z., Dai, J., Dong, Z., Dong, Y., and Peng, Y.: Glaciation temperatures of convective clouds ingesting desert dust, air pollution and smoke from forest fires, Geophysical Research Letters, 38, n/a–n/a, doi:10.1029/2011GL049423, http://doi.wiley.com/10.1029/2011GL049423, 2011.

Rosinski, J., Haagenson, P., Nagamoto, C., and Parungo, F.: Ice-forming nuclei of maritime origin, Journal of Aerosol Science, 17, 23–46, doi:10.1016/0021-8502(86)90004-2, 1986.

Rosinski, J., Haagenson, P., Nagamoto, C., and Parungo, F.: Nature of ice-forming nuclei in marine air masses, Journal of Aerosol Science, 18, 291–309, doi:10.1016/0021-8502(87)90024-3, http://linkinghub.elsevier.com/retrieve/pii/0021850287900243, 1987.

Rosinski, J., Haagenson, P., Nagamoto, C., Quintana, B., Parungo, F., and Hoyt, S.: Ice-forming nuclei in air masses over the Gulf of Mexico, Journal of Aerosol Science, 19, 539–551, doi:10.1016/0021-8502(88)90206-6, http://linkinghub.elsevier.com/retrieve/pii/0021850288902066, 1988.

Russell, L. M., Hawkins, L. N., Frossard, A. a., Quinn, P. K., and Bates, T. S.: Carbohydrate-like composition of submicron atmospheric particles and their production from ocean bubble bursting., Proceedings of the National Academy of Sciences of the United States of America, 107, 6652–7, doi:10.1073/pnas.0908905107, http://www.pubmedcentral.nih.gov/articlerender.fcgi?artid=2872374{\&}tool=pmcentrez{\&}rendertype=abstract, 2010.

Savre, J. and Ekman, a. M. L.: A theory-based parameterization for heterogeneous ice nucleation and implications for the simulation of ice processes in atmospheric models, Journal of Geophysical Research: Atmospheres, 120, 4937–4961, doi:10.1002/2014JD023000, http://doi.wiley.com/10.1002/2014JD023000, 2015.

Schnell, R. C.: Ice Nuclei in Seawater, Fog Water and Marine Air off the Coast of Nova Scotia: Summer 1975, Journal of the Atmospheric Sciences, 34, 1299–1305, doi:10.1175/1520-0469(1977)034<1299:INISFW>2.0.CO;2, 1977.

Schnell, R. C.: Airborne ice nucleus measurements around the Hawaiian Islands, Journal of Geophysical Research, 87, 8886, doi:10.1029/JC087iC11p08886, http://doi.wiley.com/10.1029/JC087iC11p08886, 1982.

Schnell, R. C. and Vali, G.: Freezing nuclei in marine waters, Tellus, 27, 321–323, doi:10.1111/j.2153-3490.1975.tb01682.x, http://tellusa.net/index.php/tellusa/article/view/9911, 1975.

Schnell, R. C. and Vali, G.: Biogenic Ice Nuclei: Part I. Terrestrial and Marine Sources, Journal of the Atmospheric Sciences, 33, 1554–1564, doi:10.1175/1520-0469(1976)033<1554:BINPIT>2.0.CO;2, 1976.

Sciare, J., Favez, O., Sarda-Estève, R., Oikonomou, K., Cachier, H., and Kazan, V.: Long-term observations of carbonaceous aerosols in the Austral Ocean atmosphere: Evidence of a biogenic marine organic source, Journal of Geophysical Research, 114, D15 302, doi:10.1029/2009JD011998, http://doi.wiley.com/10.1029/2009JD011998, 2009.

Sesartic, A., Lohmann, U., and Storelvmo, T.: Modelling the impact of fungal spore ice nuclei on clouds and precipitation, Environmental Research Letters, 8, 014 029, doi:10.1088/1748-9326/8/1/014029, http://stacks.iop.org/1748-9326/8/i=1/a=014029?key=crossref.88a72028f40efa4d07daaef5c00b97b5{%}5Cnhttp://iopscience.iop.org/1748-9326/8/1/014029/article/http://stacks.iop.org/1748-9326/8/i=1/a=014029?key=crossref.88a72028f40efa4d07daaef5c00b97b5, 2013.

Spracklen, D. V. and Heald, C. L.: The contribution of fungal spores and bacteria to regional and global aerosol number and ice nucleation immersion freezing rates, Atmospheric Chemistry and Physics, 14, 9051–9059, doi:10.5194/acp-14-9051-2014, http://www.atmos-chem-phys.net/14/9051/2014/, 2014.

Stopelli, E., Conen, F., Morris, C. E., Herrmann, E., Bukowiecki, N., and Alewell, C.: Ice nucleation active particles are efficiently removed by precipitating clouds, Scientific Reports, 5, 16 433, doi:10.1038/srep16433, http://www.nature.com/articles/srep16433, 2015.

Sullivan, R. C., Miñambres, L., DeMott, P. J., Prenni, A. J., Carrico, C. M., Levin, E. J. T., and Kreidenweis, S. M.: Chemical processing does not always impair heterogeneous ice nucleation of mineral dust particles, Geophysical Research Letters, 37, n/a–n/a, doi:10.1029/2010GL045540, http://doi.wiley.com/10.1029/2010GL045540, 2010.

Tan, I., Storelvmo, T., and Zelinka, M. D.: Observational constraints on mixed-phase clouds imply higher climate sensitivity, Science, 352, 224–228, doi:10.1126/science.aad5300, 2016.

Tobo, Y., Prenni, A. J., DeMott, P. J., Huffman, J. A., McCluskey, C. S., Tian, G., Pöhlker, C., Pöschl, U., and Kreidenweis, S. M.: Biological aerosol particles as a key determinant of ice nuclei populations in a forest ecosystem, Journal of Geophysical Research: Atmospheres, 118, 10,100–10,110, doi:10.1002/jgrd.50801, http://doi.wiley.com/10.1002/jgrd.50801, 2013.

Tobo, Y., Demott, P. J., Hill, T. C. J., Prenni, A. J., Swoboda-Colberg, N. G., Franc, G. D., and Kreidenweis, S. M.: Organic matter matters for ice nuclei of agricultural soil origin, Atmospheric Chemistry and Physics, 14, 8521–8531, doi:10.5194/acp-14-8521-2014, 2014.

Vali, G.: Repeatability and randomness in heterogeneous freezing nucleation, Atmospheric Chemistry and Physics, 8, 5017–5031, doi:10.5194/acp-8-5017-2008, http://www.atmos-chem-phys.net/8/5017/2008/, 2008.

Vali, G. and Snider, J. R.: Time-dependent freezing rate parcel model, Atmospheric Chemistry and Physics, 15, 2071–2079, doi:10.5194/acp-15-2071-2015, http://www.atmos-chem-phys.net/15/2071/2015/, 2015.

Vali, G., DeMott, P. J., Möhler, O., and Whale, T. F.: Technical Note: A proposal for ice nucleation terminology, Atmospheric Chemistry and Physics, 15, 10 263–10 270, doi:10.5194/acp-15-10263-2015, http://www.atmos-chem-phys.net/15/10263/2015/, 2015.

Vignati, E., Facchini, M., Rinaldi, M., Scannell, C., Ceburnis, D., Sciare, J., Kanakidou, M., Myriokefalitakis, S., Dentener, F., and O'Dowd, C.: Global scale emission and distribution of sea-spray aerosol: Sea-salt and organic enrichment, Atmospheric Environment, 44, 670–677, doi:10.1016/j.atmosenv.2009.11.013, http://linkinghub.elsevier.com/retrieve/pii/S1352231009009571, 2010.

Wang, X., Sultana, C. M., Trueblood, J., Hill, T. C. J., Malfatti, F., Lee, C., Laskina, O., Moore, K. a., Beall, C. M., McCluskey, C. S., Cornwell, G. C., Zhou, Y., Cox, J. L., Pendergraft, M. a., Santander, M. V., Bertram, T. H., Cappa, C. D., Azam, F., DeMott, P. J., Grassian, V. H., and Prather, K. a.: Microbial Control of Sea Spray Aerosol Composition: A Tale of Two Blooms, ACS Central Science, 1, 124–131, doi:10.1021/acscentsci.5b00148, http://dx.doi.org/10.1021/acscentsci.5b00148http://pubs.acs.org/doi/abs/10.1021/acscentsci.5b00148, 2015.

Wang, Y., Liu, X., Hoose, C., and Wang, B.: Different contact angle distributions for heterogeneous ice nucleation in the Community Atmospheric Model version 5, Atmospheric Chemistry and Physics, 14, 10 411–10 430, doi:10.5194/acp-14-10411-2014, http://www.atmos-chem-phys.net/14/10411/2014/, 2014.

Westbrook, C. D. and Illingworth, a. J.: Evidence that ice forms primarily in supercooled liquid clouds at temperatures >-27C, Geophysical Research Letters, 38, 1–4, doi:10.1029/2011GL048021, 2011.

Westbrook, C. D. and Illingworth, A. J.: The formation of ice in a long-lived supercooled layer cloud, Quarterly Journal of the Royal Meteorological Society, 139, 2209–2221, doi:10.1002/qj.2096, http://doi.wiley.com/10.1002/qj.2096, 2013.

Wex, H., DeMott, P. J., Tobo, Y., Hartmann, S., Rösch, M., Clauss, T., Tomsche, L., Niedermeier, D., and Stratmann, F.: Kaolinite particles as ice nuclei: learning from the use of different kaolinite samples and different coatings, Atmospheric Chemistry and Physics, 14, 5529–5546, doi:10.5194/acp-14-5529-2014, http://www.atmos-chem-phys.net/14/5529/2014/, 2014.

Whale, T. F., Murray, B. J., O'Sullivan, D., Umo, N. S., Baustian, K. J., Atkinson, J. D., and Morris, G. J.: A technique for quantifying heterogeneous ice nucleation in microlitre supercooled water droplets, Atmospheric Measurement Techniques Discussions, 7, 9509–9536, doi:10.5194/amtd-7-9509-2014, http://www.atmos-meas-tech-discuss.net/7/9509/2014/, 2014.

Wilson, T. W., Ladino, L. a., Alpert, P. a., Breckels, M. N., Brooks, I. M., Browse, J., Burrows, S. M., Carslaw, K. S., Huffman, J. A., Judd, C., Kilthau, W. P., Mason, R. H., McFiggans, G., Miller, L. a., Nájera, J. J., Polishchuk, E., Rae, S., Schiller, C. L., Si, M., Vergara Temprado, J., Whale, T. F., Wong, J. P. S., Wurl, O., Yakobi-Hancock, J. D., Abbatt, J. P. D., Aller, J. Y., Bertram, A. K., Knopf, D. a., and Murray, B. J.: A marine biogenic source of atmospheric ice-nucleating particles, Nature, 525, 234–238, doi:10.1038/nature14986, http://www.nature.com/doifinder/10.1038/nature14986, 2015.

Yin, J., Wang, D., and Zhai, G.: An evaluation of ice nuclei characteristics from the long-term measurement data over North China, Asia-Pacific Journal of Atmospheric Sciences, 48, 197–204, doi:10.1007/s13143-012-0020-8, http://link.springer.com/10.1007/s13143-012-0020-8, 2012.

Young, K. C.: A Numerical Simulation of Wintertime, Orographic Precipitation: Part I. Description of Model Microphysics and Numerical Techniques, Journal of the Atmospheric Sciences, 31, 1735–1748, 1974.

Zeng, X., Tao, W.-K., Zhang, M., Hou, a. Y., Xie, S., Lang, S., Li, X., Starr, D. O., Li, X., and Simpson, J.: An indirect effect of ice nuclei on atmospheric radiation, Journal of the Atmospheric Sciences, 66, 41–61, doi:10.1175/2008JAS2778.1, 2009.

Zolles, T., Burkart, J., Häusler, T., Pummer, B., Hitzenberger, R., and Grothe, H.: Identification of ice nucleation active sites on feldspar dust particles, Journal of Physical Chemistry A, 119, 2692–2700, doi:10.1021/jp509839x, 2015.