

Dear Editor,

Thank you for taking the time to handle the review of our article. We would also like to thank the reviewers for their helpful advice, which has resulted in a much improved manuscript. Please find a list of reviewer comments, along with author responses and manuscript changes below. All page and line numbers refer to the discussion paper version. Figures/tables labelled 'Manuscript Figure/Table' are to appear in the manuscript, and figures/tables labelled 'Response Figure/Table' do not appear in the revised manuscript.

Kind regards,

L. B. Hande, C. Engler, C. Hoose and I. Tegen.

Response to anonymous reviewer 1.

Reviewer Comment:

Pg. 32075 and throughout. Data and parameterisations have only been produced for winter and summer. Was there a specific reason for omitting spring and autumn? It makes the parameterisation a bit incomplete and I would recommend including spring and autumn parameterisations or recommendations for which months to use the summer/winter parameterisations.

Author Response:

Winter and summer were initially chosen as they represent seasons with contrasting aerosol and meteorological conditions. For completeness we have included spring and autumn seasons in the revised manuscript. Here, summer refers to Dec 2007, Jan 2008 and Feb 2008, spring refers to Mar, Apr and May of 2008, Winter includes Jun, Jul and August of 2008, and finally Autumn includes Sep, Oct and Nov of 2008. Additionally, a yearly parameterisation has been provided for use in longer simulations. This includes data from Dec 2007 through to Nov 2008.

Manuscript Changes:

Figure 1 now includes the dust and INP profiles for spring and autumn. The profiles show the median, and 5th and 95th percentiles for all seasons. The discussion in section 2 now includes references to the dust and INP concentrations for Autumn and Spring. A discussion of the difference between the mean and median profiles is provided in the text in section 2, as these vertical profiles are not shown.

Pg 32075, line 28: Added: 'The winter season refers to December 2007, January 2008 and February 2008, Spring includes March to May of 2008, Winter includes June to August of 2008, and finally Autumn refers to September to November of 2008.'

Reviewer Comment:

Pg. 32089, Fig 1. Please explain why INP concentrations decrease so sharply at higher levels, as this is counter intuitive. For example, the summer dust surface area/number concentration shows no significant trend with altitude, but the number of INP goes down at high altitude. Until the fraction frozen predicted by the Niemand et al 2012 and/or Steinke et al. 2014 parameterisations is 1 (i.e. $n_{dust} = n_{ice}$) the ice concentration should continue to increase. Please ensure this is a real effect and not a statistical issue as the parameterisation values and shape are based on it.

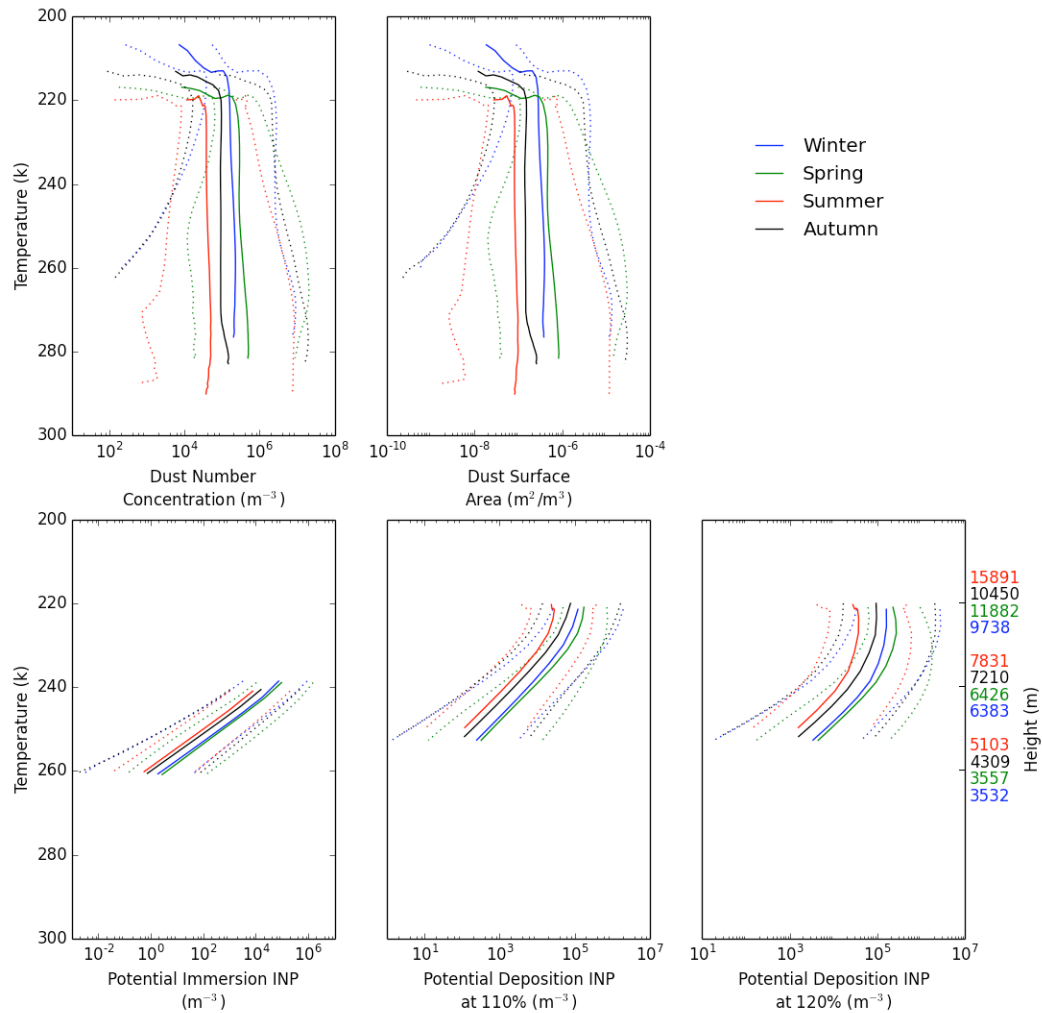
Author Response:

The reviewer is correct. The method used to calculate the statistics produced the artificial decrease in INP concentrations at higher altitudes. The method has now been modified, so that the statistics for the dust number concentration are calculated for each size bin separately. From these, and using the mean temperature, the statistics for dust surface area, immersion and deposition INPs are calculated. The percentiles for the dust concentration and the mean temperature for the relevant months are used to calculate the percentiles for the INP concentrations. This has been explained in the revised manuscript.

This approach has a number of advantages over the first approach. The resulting INP profiles are now easier to interpret and relate to the dust number concentration profiles. Secondly, the INPs are more appropriately constrained by the mean temperature profile, particularly for the mean vertical profiles. However the shape of the profiles has now changed, which requires a modification to the form of the parameterisation. This also has the advantage of simplifying the parameterization by reducing the number of equations and parameters defining the INP concentrations. This is addressed in more detail in the reviewer comment relating to the parameterizations.

Manuscript Changes:

Figure 1:



Manuscript Figure 1: Median (solid), 5th and 95th percentiles (dotted) of total dust number concentration, total dust surface area, potential immersion INP, and potential deposition INP at RH_{ice} of 110% and 120% over Europe for all seasons. The right height axis shows the average model height levels at three temperatures (260, 240, and 220 K) for all seasons.

Manuscript Text: The text has been updated in all the relevant sections (abstract, section 2, and conclusions) with a description of Figure 1 above.

Reviewer Comment:

Pg. 32078. Parameterisations. Were any other forms considered for the parameterisation? I agree that the median INP are a function of concentration and temperature, and hence proportional to altitude. However I would wonder if the multiplication of two lines would be a simpler representation, e.g. an equation representing the fraction frozen vs z multiplied by the trend of dust vs z . I also think a temperature dependent form of the parameterisations needs to be provided, as most of the models that would use this parameterisation will carry temperature information. This is especially important since the 4 K range the 25-75 % temperatures cover would result in around an order of magnitude shift in INP concentrations.

Author Response:

The form of the parameterisation has now changed, and is best described by a modified exponential function. This is possibly the easiest and simplest implementation, since there is only one equation with 4 parameters. The only disadvantage is that the maximum INP concentrations are no longer naturally constrained, and if the new formulation is misused, this could lead to unrealistically high concentrations at cold temperatures. The importance of limiting the maximum concentrations is emphasized in the text. The authors suggest to use $C_{INP}(273.15)$ at lower temperatures for immersion freezing, and $C_{INP}(220)$ for colder temperatures with deposition nucleation.

Also, the parameterisation is now a function of temperature, instead of height. We also provide suggested scaling factors for the 5th and 95th percentiles, as shown in the last two columns of table 1. Giving INP concentrations as a function of temperature now includes two different dependencies, namely that on temperature as well as the concentration dependency on altitude.

Manuscript Changes:

Equation (1):

$$C_{INP}(T_K) = A \exp[-B(T_K - T_{min})^C]$$

T_k is the model temperature in Kelvin, and the free parameters are defined in Table (1).

Pg 32078: Added: ‘Finally, it is recommended to use $C_{INP}(273.15)$ for temperatures colder than 273.15 K for immersion freezing, and $C_{INP}(220)$ for temperatures colder than 220 K for deposition nucleation, in order to prevent zero ice nuclei concentrations.

Table (1):

Immersion	A (m⁻³)	B	C	T_{max} (K)	T_{min} (K)	5th PSF	95th PSF
Winter	1.0259e5	0.2073	1.2873	261.15	237.15	0.04	12.06
Spring	1.5684e5	0.2466	1.2293	261.15	237.15	0.10	17.82
Summer	2.9694e4	0.2813	1.1778	261.15	237.15	0.13	27.28
Autumn	4.9920e4	0.2622	1.2044	261.15	237.15	0.06	31.38
Year	8.1909e4	0.2290	1.2553	261.15	237.15	0.10	17.14

Deposition							
Winter	1.2663e5	0.0194	1.6943	253	220	0.17	15.25
Spring	1.7836e5	0.0075	2.0341	253	220	0.24	5.87
Summer	2.6543e4	0.0020	2.5128	253	220	0.22	12.88
Autumn	7.7167e4	0.0406	1.4705	253	220	0.16	22.19
Year	9.6108e4	0.0113	1.8890	253	220	0.22	12.00

Manuscript Table 1: Parameters defining equation (1), for immersion and deposition INPs (at RH_{ice} =110%). The percentile scaling factors (PSF) for the 5th and 95th percentiles are provided.

Reviewer Comment:

Pg. 32073, line 4-5 and Pg. 32075, lines 10-16. In the same year as Hoose and Möhler, 2012, there was another ice nucleation laboratory review (Murray et al., 2012). In the interests of impartiality, this should also be referenced.

Author Response:

Thanks, this reference has been added.

Manuscript Changes:

The reference has been added to the suggested sections.

Reviewer Comment:

Pg. 32073, line 17. Please do not use ‘and references therein’ for referencing specific details, especially in a journal that does not have a reference limit.

Author Response:

No problem.

Manuscript Changes:

‘And references therein’ has been removed.

Reviewer Comment:

Pg. 32073, line 21-22. Being pedantic, immersion and condensation are separate pathways. Yes there is debate as to whether the process actually causing ice nucleation is different, but the pathway from dry particle to ice is different.

Author Response:

This sentence has been modified to include condensation freezing as a separate pathway.

Manuscript Changes:

Pg 32073, line 21 – 22: ‘Ice nucleation in the atmosphere takes place via four different pathways: immersion, condensation, deposition, and contact freezing.’

Reviewer Comment:

Pg. 32073, line 25-29. You might also consider referencing the parameterisations produced by Phillips (e.g. Phillips et al., 2008) in here somewhere.

Author Response:

Thanks, the reference has been added.

Manuscript Changes:

The reference has been added to the text.

Reviewer Comment:

Pg. 32073-32077. Section 2 in general is lacking in detail. A list of specifics is provided below but should not be viewed as exhaustive.

Author Response:

Details have been added, primarily through the following comments.

Manuscript Changes:

See the following comments.

Reviewer Comment:

Pg. 32074, line14. Please list the actual dust particle size bins. 5 bins for 0.1-24 μm seems a bit coarse – has an investigation been done to see if there are any effects from this bin scheme?

Author Response:

The dust particle sizes have been listed in the manuscript.

Manuscript Changes:

Added: ‘... in 5 size bins (0.1 – 0.3 μm , 0.3 – 0.9 μm , 0.9 – 2.6 μm , 2.6 – 7.9 μm , 7.9 – 24 μm).’

Reviewer Comment:

Pg. 32074, line 17. Is soil temperature taken into account for the dust emission? Significant parts of the domain will be below freezing for large periods of time. Also, how important is the in domain dust emission compared with advected dust?

Author Response:

The description of the model set up has been expanded to include a discussion of soil dust temperature, and alternative dust sources.

Manuscript Changes:

Pg 32074, line 17: Added: ‘While soil temperature is not directly included in the dust

emission scheme, snow-covered gridcells are excluded as dust sources. Transported dust from the Sahara (which are the focus of this study) are considered to be the main source for INP. Local soil dust sources are considered less important, as their emission fluxes are low and they generally remain in the boundary layer.’

Reviewer Comment:

Pg. 32074, lines 22-27. Please clarify – were these evaluations performed with exactly the same setup as the model runs used in the current work?

Author Response:

Yes, the same model set up was used. This has been explicitly stated in the manuscript.

Manuscript Changes:

Pg 32074, line 25. Added: ‘The evaluation of the dust model results shown in Tegen et al (2013) were performed with the same model setup as described in this work.’

Reviewer Comment:

Pg. 32075, line 3 and elsewhere. I would caution against referring to this as specifically desert dust; natural dust or soil dust is likely more appropriate. Of the samples used by Niemand et al., 2012 one is from an agricultural valley (Canaries Island Dust), one is from 50 Km north of Cairo: the Nile Delta (‘Saharan’ Dust), and one is from an unknown location (Israeli Dust – collected from a dust storm of unknown origin). No other description such as the organic content or mineralogy of these dusts was given.

Author Response:

References to the Niemand et al. (2012) parameterisation have been changed to ‘natural dust’

Manuscript Changes:

Pg 32075, line3: ‘desert dust’ changed to ‘natural dust’
Pg 32082, line 20: ‘desert dust’ changed to ‘natural dust’

Reviewer Comment:

Pg. 32075, lines 10-16. *‘While there are indications that Arizona Test Dust is a more efficient ice nucleus than natural desert dust particles at the higher end of this temperature range, their behaviour is comparable at temperatures below 238 K (Hoose and Möhler, 2012; Hiranuma et al., 2014)’*

I have issues with this statement. It is not borne out by the reference to Hoose and Möhler, 2012 –

Fig. 3 of that paper shows that for sub-micron particles ATD and natural dusts are similar above 238 K and ATD much better (i.e. lower RH) below 238 K, whereas in super-micron particles the natural dusts are better at all temperatures. Also, from a purely

geological point of view using Hiranuma et al., 2014, which refers to Hematite as a desert/atmospheric dust proxy, is probably not appropriate. See the author response to comments/discussion on the ACPD version of that paper. References to any temperature threshold used, 238 K or otherwise, need to be made in the figures, especially figure 1.

Author Response:

We would like to clarify which results of the cited references we were referring to. Figure 3 of Hoose & Möhler (2012) does not give a reliable answer to this question, because it shows data from instruments with different detection limits and for experiments with different thresholds defining the “onset” of ice nucleation. A more quantitative investigation of the activity of ATD versus natural dusts is shown in Fig. 8 and Fig. 14 of Hoose and Möhler (2012), inserted below, comparing the onset saturation ratio for comparable experimental conditions and the ice nucleation active site density, respectively. The results summarized in these two figures show that ATD is found to be more active than some desert dusts, but not all of them. For Hiranuma et al (2014), we were referring to the ATD and natural dust isopleths of ice nucleation active site densities from previous experiments shown in their Fig. 3 (also inserted below), not to the results on hematite. As the ATD and desert dust data shown here are also included in Hoose & Möhler (2012).

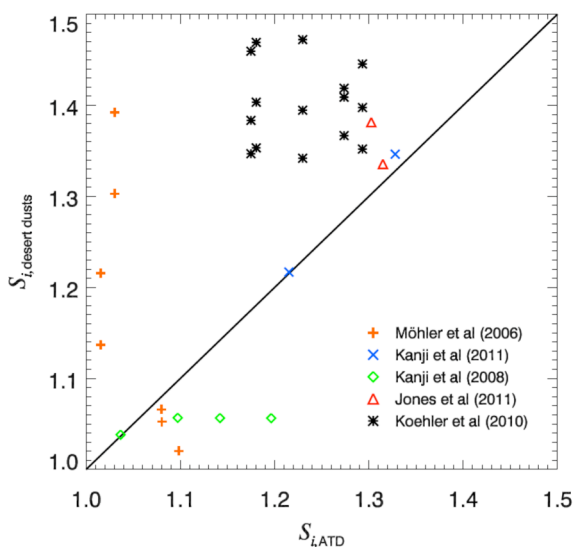


Fig. 8. Saturation ratios at nucleation onset for Arizona test dust and natural desert dusts (Saharan, Asian, Canary Island and Owens Lake dust) under comparable experimental conditions and interpolated to the same temperatures, respectively. The data by Koehler et al. (2010) are for dry dispersed particles of 200 nm size. The data by Kanji et al. (2011) are for UT-CFDC measurements sampled from the aerosol preparation and characterization chamber for ATD and Canary Island dust, which have similar size distributions. The data by Kanji et al. (2008) are interpolated to the same total particle surface areas. The data by Jones et al. (2011) are from the CSU-CFDC instrument and for water-subsaturated conditions.

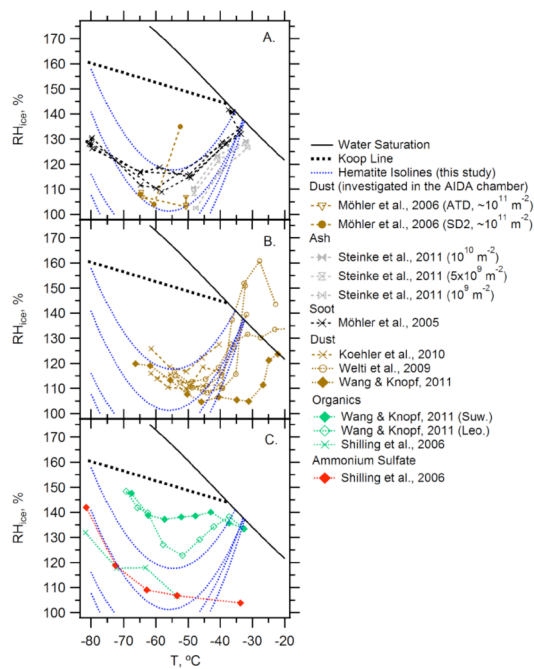


Figure 3. Ice nucleation onset T and RH_{ice} of previously published data (a AIDA studies, b dust, and c ammonium sulfate and organics) shown together with the isolines of hematite particles from the present study (10^{12} m^{-2} (top)– 10^9 m^{-2} (bottom)).

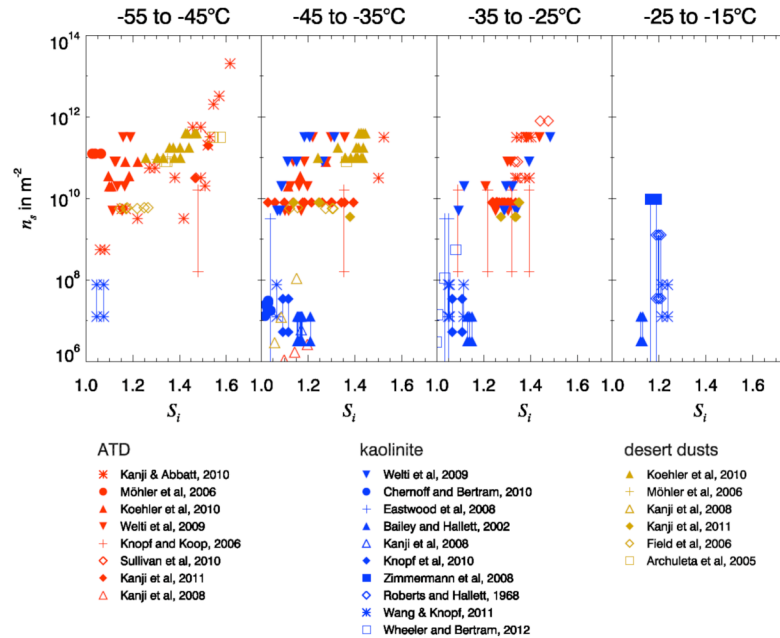


Fig. 14. INAS densities for ATD, kaolinite and desert dusts for deposition nucleation.

Response Figure 1: Hoose and Möhler (2012), Fig 8, 3, and 14.

Manuscript Changes:

Pg 32075, line 13: The reference to Hiranuma et al. (2014) has been removed.

Reviewer Comment:

Pg. 32075 line 25 – Pg. 23076 line 25. Please provide more detail on how the different means and medians are calculated and related to each other. For example, is the median potential INP calculated from the median particle surface area and concentration, or using the absolute particle surface area and concentration and then calculating the median using time/spatial data? Is the particle surface area calculated using a spherical assumption with the bin-centre radius? Is the 25 - 75% range for INP concentration estimated using the 25-75% range of the dust surface area/concentration, the temperature or both? The way these paragraphs are written implies that deposition is more important (i.e. has more INP) than immersion, but the deposition maximums are at a much lower temperature – it would be helpful clarify by adding a comment about the lower temperatures for deposition.

Author Response:

The method used to calculate the statistics has been clarified in the manuscript.

Manuscript Changes:

Pg 32076, line 2: ‘The dust number concentration statistics were firstly computed for a whole month. Then, in order to calculate seasonal (yearly) statistics, the monthly

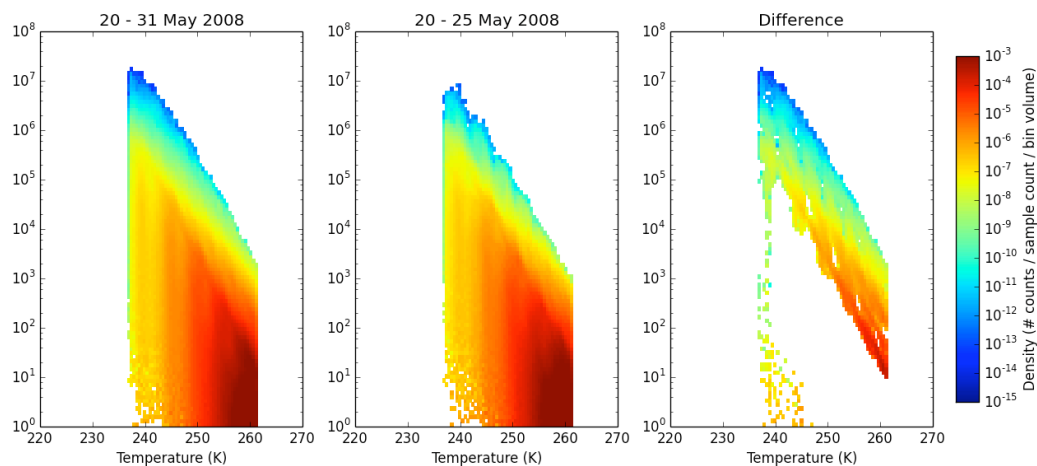
statistics were averaged over a three (twelve) month period. Using this, the dust surface area statistics were calculated by assuming spherical particles with the bin centre radius. The INP statistics were then calculated by using the respective dust number concentration statistic as an input to the parameterisation, along with the mean temperature for that season.'

Reviewer Comment:

Pg. 32076 line 13-14. Have you checked the residuals to see if infrequent significant dust events is the cause of a high mean? The mean values are generally neglected in the remainder of the manuscript, which should be justified better. If the parameterisations that follow are for background INP might it not be better to remove outliers?

Author Response:

Analysing the residuals wouldn't indicate significant dust events by themselves. It just indicates departures from the parameterisation. However, in order to maximize the likelihood of departures from the parameterisation being due to significant dust events, the residuals were analysed for a few days around a known significant dust event in late May 2008. The figure below shows the 2D histogram for immersion freezing INPs from 20th – 31st May 2008 (including the dust event), then from 20th – 25th May 2008 (non-dust event) and the difference between the two (panel 1 – panel 2).



Response Figure 2: 2d Histograms for immersion freezing for 20th – 31st May 2008, 20th – 25th May 2008, and the difference between the two (panel 1 – panel 2).

The effect of the significant dust event is most obviously seen in the last panel, indicating the large dust event does contribute mostly to the high INP concentrations over all temperatures. Note that the colorbar is logarithmic, which means the negative values are not plotted, however these negative values do occur at lower concentrations in the histogram, indicated by the white region. The large positive values at high INP concentrations indicate that significant dust events produce larger amounts of high

concentrations INPs, and fewer INPs in lower concentrations. Therefore, the median concentrations are a better representation of the background dust concentrations.

Removing the outliers from the analysis is not necessary, since the median concentrations are not affected by the outliers. The median profiles are representative of the background conditions and will be used to define the parameterisation.

Manuscript Changes:

Pg 32078, line 16: ‘Since the parameterisation uses the median concentrations, it is representative of the background INP concentrations. An analysis of the residuals showed that during a large dust event, INPs are produced at high concentrations over all temperatures compared to a non-dust event, therefore the mean concentrations are not used here since these would be heavily influenced by large dust outbreaks.’

Reviewer Comment:

Pg. 32076, line 17. To save your readers from being obliged to look up and read the Bangert paper, please also provide the values.

Author Response:

No problem, the dust concentrations from between 235 – 273 K were estimated from Bangert et al. (2012) figure 6.

Manuscript Changes:

Pg 32076, line 18: ‘... but close to modeled concentrations of over 10^7 m⁻³ reported by Bangert et al. (2012)...’

Reviewer Comment:

Pg. 32077 line12. As suggested 2 comments above, the presence of significant outliers can be confirmed by analysing the residuals.

Author Response:

The analysis of residuals has been addressed 2 comments above.

Manuscript Changes:

None.

Reviewer Comment:

Pg. 32077 line 17. Why was an altitude of 5 km chosen?

Author Response:

The altitude has been changed to 6 km. This altitude was chosen because during winter median INP concentrations in both the immersion and deposition modes are sufficiently high to draw meaningful conclusions about the horizontal homogeneity. See figure 1.

Manuscript Changes:

Pg 32077, line 17: ‘... 6 km above the terrain, which is where most INPs are located.’

Reviewer Comment:

Pg. 32077 line 19. This is indeed a remarkable sounding result. Are there any observations to back this up?

Author Response:

One could confirm this with satellite or lidar observations using a method published by Mamouri and Ansmann (2014), however this is a bit of a digression for our work here. From the Bangert et al. (2012) model results, one can see there is still significant dust concentrations in the north of Germany between 235 - 273 K. These are daily means from during a dust event, so one can't draw any conclusions about the concentrations on seasonal timescales, however one can see that it is possible to have significant dust concentration in the north of our domain.

Furthermore, the south of the domain is approximately 3000 km from the source region, and the domain is less than about 900 km in the north direction. Therefore the gradient within the domain shouldn't be large since the whole region is quite far from the source. Also, we have shown number concentration, and if one plotted the mass concentrations then maybe there would be more of a notable gradient. Finally, the settling and washout timescales are typically quite long compared to the transport timescale through the domain.

Manuscript Changes:

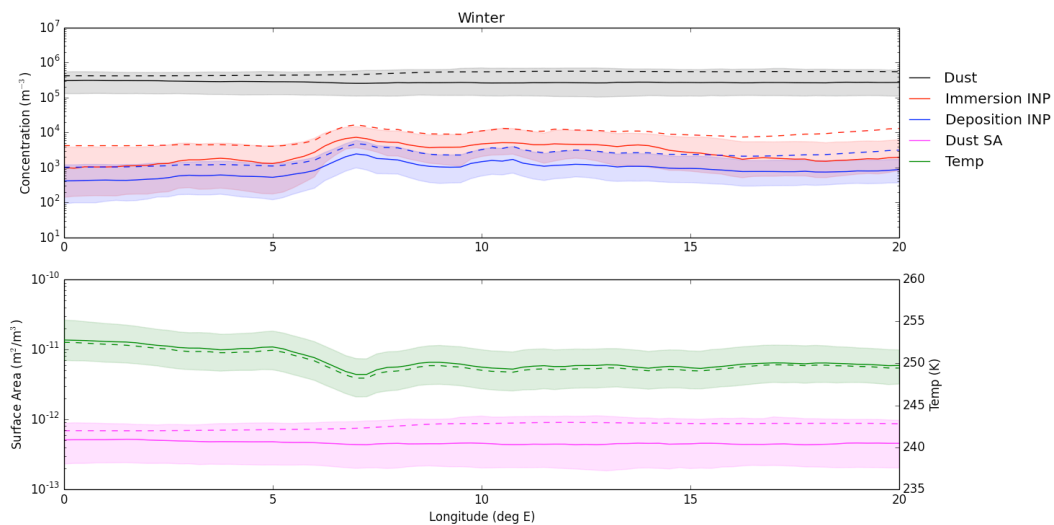
None.

Reviewer Comment:

Pg. 32077 line 29 – Pg. 32078 line 2. I agree that in terms of latitude the dust concentration is largely constant throughout your domain. However with the prevailing winds and a major source of advected dust in the domain being from the west, I would think that the same comparison for longitude is needed before it can be said that dust concentration is constant within the domain.

Author Response:

We've included the dust, INP and temperature statistics as a function of longitude below:



Response Figure 3: Top panel: Median (solid) and mean (dashed) total dust number concentration (black), potential immersion INP (red), and potential deposition INP at 110% (blue) at 5 km above terrain over Europe for winter. Bottom panel: Median (solid) and mean (dashed) total dust surface area (magenta) and temperature (green). The area between the 25th and the 75th percentiles is shaded.

Once again, one can see the dust is very constant as a function of longitude. There is an increase in variability of dust closer to the eastern side of the domain. This may indicate, as the reviewer suggested, that dust can be transported from the west into the domain. However the significant dust events enter from the south, for example the well documented case in late May 2008.

The INP concentrations are therefore fairly constant with longitude also. There is an enhancement in INP concentrations at 7 deg E, which is the beginning of the alpine regions, and corresponds to a decrease in temperature of a little over 3 deg K in 2 deg of longitude. This enhancement is relatively small, from INP concentrations of about 1500 m⁻³ to 7000 m⁻³ for immersion freezing. Therefore, we can conclude that INP concentrations are more-or-less constant throughout the whole domain, and there is a slight enhancement in ice nucleation in the alpine regions.

Manuscript Changes:

Pg 32077, line 29: ‘The variability of dust and INP as a function of longitude was also investigated, and a similarly small amount of variability was found (not shown). In the immersion mode, an enhancement in the median INP concentrations from about 1500 m⁻³ to 7000 m⁻³ was found around 7 deg E. This corresponds to a decrease in temperature of about 3 K at the western edge of the alps.’

Reviewer Comment:

Pg. 32078, line 12. Please clarify the identity of σ . Is this the usual definition of SD (Standard Deviation)? If so I would suggest this is not appropriate for your purpose, as using the standard deviation not only requires a normal distribution (which apparently

you do not have) but is also derived from the mean – should it be used in combination with the median?

Author Response:

The parameterisation has now changed, and the comment is no longer relevant.

Manuscript Changes:

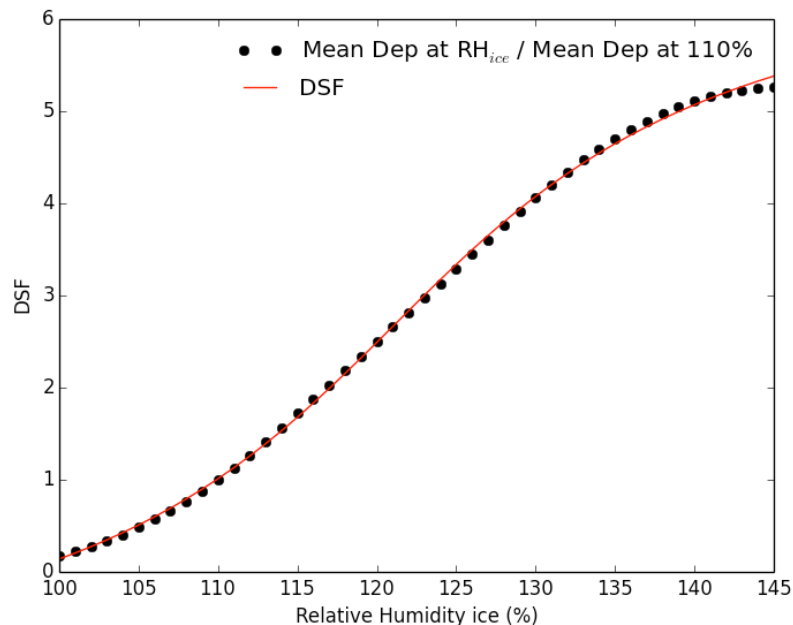
None.

Reviewer Comment:

Pg. 32080, lines 10-14. This is a little confusing/concerning. The INP concentrations in Fig 1 are calculated using the Steinke et al. 2014 parameterisation. The DSF equation 6 is derived directly from the same function. Surely the results from the two should be the same?

Author Response:

We have decided to use a new method to calculate the DSF. Since we are now calculating median INP concentrations by applying the parameterizations with the median dust concentrations, this allows us to calculate deposition INPs at many values of RH_{ice} quickly, and then compute the DSF directly from the model data. The DSF is defined as the ratio of mean deposition INP at a given RH_{ice} to the mean deposition INPs at 110%. The figure below shows the model computed DSF for a range of RH_{ice} values.



Response Figure 4: Deposition Scaling Factor computed from model data (black dots), and a best fit line (red line).

One can fit a line of the form:

$$DSF(RH_{ice}) = a \times \arctan(b \times (RH_{ice} - 100) + c) + d$$

with $a = 2.7626$, $b = 0.0621$, $c = -1.3107$, and $d = 2.6789$, shown as the red line in the above figure. These factors were determined by a best fit. Then the deposition INP concentrations are simply:

$$C_{INP}(T_K, RH_{ice}) = C_{INP}(T_K) \times DSF(RH_{ice})$$

This formulation is a significant improvement over the previous method, as it is directly calculated from the model data. As can be seen from the above figure, the fit provided by the DSF equation is excellent, and provides an easy way to incorporate a RH_{ice} dependence into the parameterisation for deposition nucleation.

The DSF begins to depart from the above relationship at a RH_{ice} > 145% . Here, the DSF is at a constant value of 5.2, however the DSF given by the above equation still increases slowly. However, observations show in-cirrus cloud RH_{ice} values higher than 145% are exceptionally rare (Haag et al. (2003)).

Manuscript Changes:

Pg 32079, lines 1 – 12: ‘A deposition scaling factor (DSF) was defined as the ratio of mean deposition INPs at a given RH_{ice} to the mean deposition INPs at RH_{ice} = 110%, and calculated from the model data for RH_{ice} from 100-145%. This showed an increase in mean deposition concentrations that followed the form:

$$DSF(RH_{ice}) = a \times \arctan(b \times (RH_{ice} - 100) + c) + d$$

where $a = 2.7626$, $b = 0.0621$, $c = -1.3107$, and $d = 2.6789$, and were determined by a best fit. Finally, the scaled INP concentrations due to deposition nucleation, are approximately:

$$C_{INP}(T_K, RH_{ice}) = C_{INP}(T_K) \times DSF(RH_{ice})$$

where $C_{INP}(T_K)$ is the concentration of deposition INPs at RH_{ice}=110%, given by equation (1), and the approximation is most valid for small activated fractions.’

Reviewer Comment:

Pg. 32081. Data from Conen et al. 2012 and Klein et al. 2010 appear in Fig. 4 but are not mentioned in the discussion and probably should be.

Author Response:

Observations from Klein et al. (2010) appear in pg. 32081, lines 7 – 8. This is in reference to the higher INP concentrations from during a Saharan dust event lying at the edge of the results from the Niemand parameterisation. A brief discussion of the

observations from Conen et al. (2012) has been added.

Manuscript Changes:

Pg 32081, line 18. ‘Most of the data from Conen et al. (2012) was also taken at temperatures warmer than 260 K, indicating dust can nucleate ice at temperatures warmer than the Niemand et al. (2012) parameterisation. Nevertheless, the concentrations are the same as the parameterisation at 260 K.’

Reviewer Comment:

Pg. 32081, lines 9-10. Could a reason for the different temperature sensitivities be speculated upon?

Author Response:

Observations from DeMott et al (2010) used to derive this best fit function are from many locations (from remote to urban locations), and include many chemically different INPs. There is also a large amount of variability in INP concentrations at colder temperatures, meaning that the fit is very sensitive to which data is included.

DeMott et al. (2010) suggest most of the variability in their fit is due to ‘aerosol chemical composition or other factors’. This has been noted in the manuscript.

Manuscript Changes:

Pg 32081, line 10. ‘... than DeMott et al. (2010) indicate, however it’s important to note that the best fit provided by these authors is dependant on aerosol composition, amongst other things.’

Reviewer Comment:

Pg. 32081, line 14. I think that this sentence need to be rearranged - the INP are not at higher temperatures, the observations are.

Author Response:

That’s what we mean to say. Nevertheless, the sentence has been restated to try to clarify any confusion.

Manuscript Changes:

Pg 32081, line 14: ‘Most of the observed INPs are at temperatures warmer...’

Reviewer Comment:

Pg. 32081, lines 15-18. Discussion on Joly et al. 2014 could be expanded to include implications. Do the authors think this implies that Joly et al. 2014 was actually measuring dust, Niemand et al. 2012 was measuring biological, both, or that composition is not that important?

Author Response:

The authors don't want to engage in uninformed speculation. The results from Joly et al. (2014) indicate the measurements were of biological INPs, and the measurements from Niemand et al. (2012) were of dust INPs. We just mean to say that it is interesting the concentrations from these two different INPs are roughly the same, and occur at roughly the same temperature.

Manuscript Changes:

None.

Reviewer Comment:

Pg. 32081, lines 21-27. This discussion is weakened by not including and considering the observational conditions. E.g. DeMott et al. 2010 was in the condensation mode at 240 K, which fits with Fig 4 nicely.

Author Response:

Thanks, this has been noted in the text.

Manuscript Changes:

Pg 32081, line 23: '...INP concentrations can reach over 10^5 m⁻³ at 240 K in the condensation mode.'

Reviewer Comment:

Pg. 32089, Fig 1. It would be helpful to readers who look at the figures before reading in depth if you define what Europe means, either in the caption or with a domain-description figure.

Author Response:

Done.

Manuscript Changes:

Fig 1 Caption: '... over Europe (44-60 N and 0-20 E)...'

Reviewer Comment:

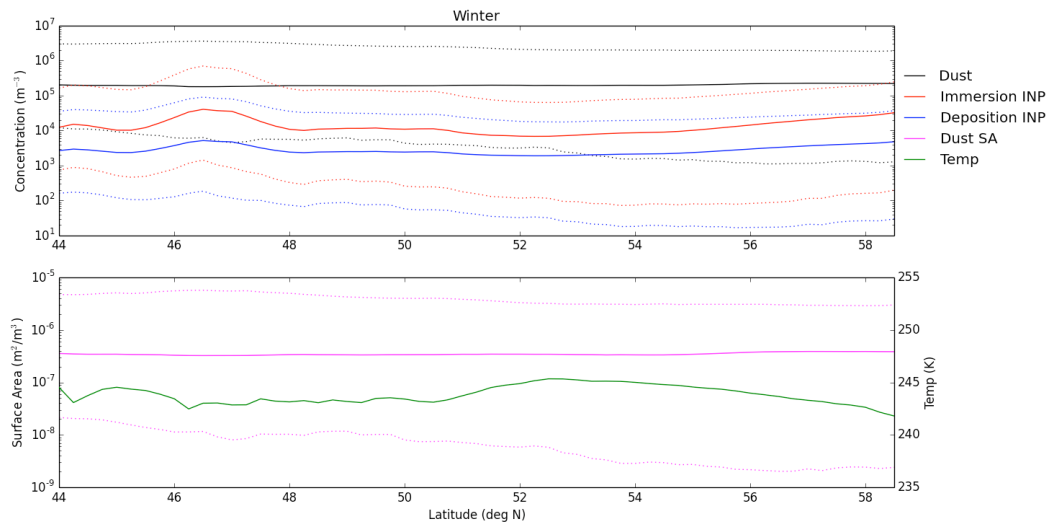
Pg. 32090, Fig 2. The scale in the upper panel runs from 10^1 to 10^7 , whereas the data fits between 10^2 and 10^6 . Since the aim is to instruct the reader that there is no significant variation with latitude, adjusting the y-axis would make this clearer.

Author Response:

The statistics used to generate this figure have been recalculated to be consistent with the new method used. The new figure is shown below.

Manuscript Changes:

Figure 2:



Manuscript Figure 2: Top panel: Median (solid), 5th and 95th percentiles (dotted) for total dust number concentration (black), potential immersion INP (red), and potential deposition INP at 110% (blue) at 6 km above terrain over Europe for winter.

Bottom panel: Median (solid) and mean (dashed) total dust surface area (magenta) and temperature (green). Note the alpine regions are between about 46-48 deg N.

Reviewer Comment:

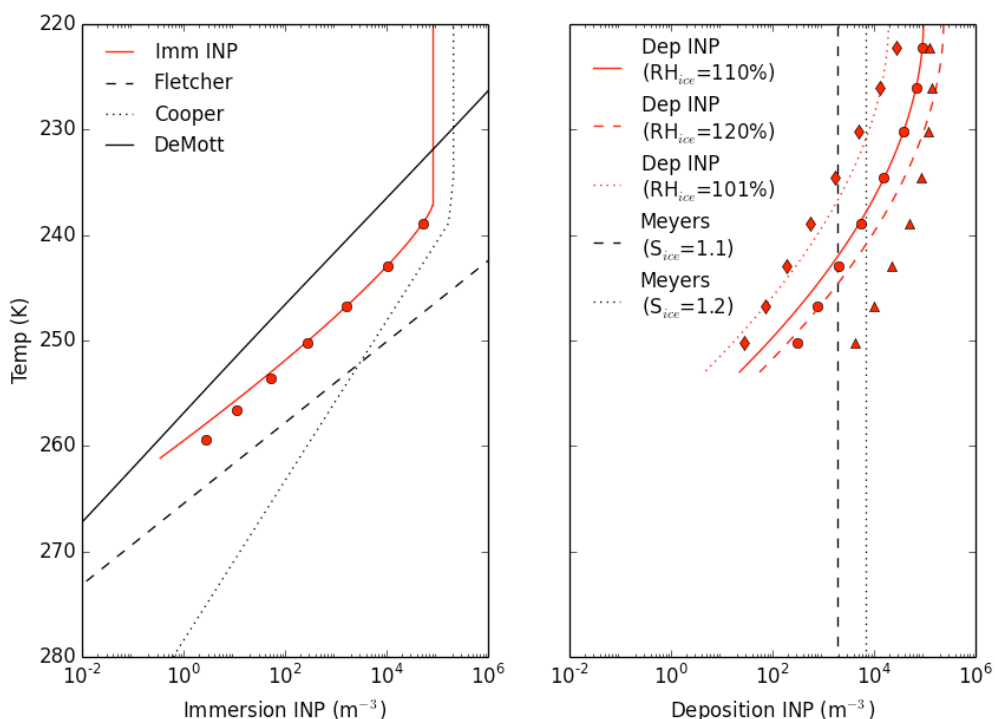
Pg. 32091, Fig 3. Please consider adding the model data from Fig. 1 to this figure, as it would make it easier to see the quality of the parameterisations.

Author Response:

The model data used to develop the parameterisation has been included in this figure.

Manuscript Changes:

Figure 3:



Manuscript Figure 3: (LEFT): Immersion INP concentrations from equation (1) for using yearly parameters (red). The red circles represent the model data. The black dashed line is the Fletcher et al. (1962) parameterisation, the dotted line represents the Cooper (1986) parameterisation, and the solid line represents DeMott et al. (2014) parameterisation. (RIGHT): Deposition INP from equation (3) for using yearly parameters (red), at $RH_{ice} = 110\%$ (solid) and $RH_{ice} = 120\%$ (dotted). The black vertical dashed (dotted) line represents concentrations from Meyers et al. (1992) at $S_{ice} = 1.1$ ($S_{ice} = 1.2$). The red circles (triangles / diamonds) represent the model data at $RH_{ice} = 110\%$ ($RH_{ice} = 120\% / 101\%$)

Reviewer Comment:

Pg. 32092, Fig 4. I could not find any reference to the left panel in the text. Could you speculate as to the cause of the wave-like feature in the left panel, and also the horizontal/diagonal bands in the data? Please also consider changing the symbols as they print very badly in the discussions version and are not discernible.

Author Response:

A brief description of the deposition INP histogram is already in the manuscript, pg 32081, lines 10 - 13. The wave-like feature and the bands in the histogram are due to the discrete nature of the model levels. For deposition nucleation, which occurs at colder temperatures (higher altitude), the terrain following model levels become flatter. Therefore, it is simply more likely to find data at certain temperatures corresponding to these model levels.

Manuscript Changes:

None.

Reviewer Comment:

Pg. 32074, line 10-27. This section needs more references.

Author Response:

No problem, more references have been added to this section.

Manuscript Changes:

Pg 32074, line 11: Added: 'Steppeler et al. (2003)'

Pg 32074, line 12: Added: 'Wolke et al. (2004)'

Pg 32074, line 17: Added: 'Tegen et al. (2002)'

Reviewer Comment:

Pg. 32075, line9. Replace 'a' with 'at'.

Author Response:

Done, thanks.

Manuscript Changes:

Pg 32075, line 9: '...is active at colder temperatures...'

Reviewer Comment:

Pg. 32075, lines 17-21. The concept of potential INP is not new and should be referenced (Murray et al., 2012; Atkinson et al., 2013).

Author Response:

This has been noted in the text.

Manuscript Changes:

Pg 32075, line 23: 'This concept of potential INP has been used previously by others (Murray et al. 2012).'

Reviewer Comment:

Pg. 32076, line 27, Pg. 32077 line 2. The values for deposition INP (10-3 and 10-4) do not match figure1, please check.

Author Response:

These values were updated in the manuscript.

Manuscript Changes:

Pg 32076: All values for immersion and deposition median and means have been updated.

Reviewer Comment:

Pg. 32078, Eq. 2. Should $\varphi(t)dt$ be $\varphi(z)dz$? If not, please define t .

Author Response:

This comment is no longer relevant since the form of the parameterisation has changed.

Manuscript Changes:

None.

Reviewer Comment:

Pg. 32079, line 24. Insert: ...suggested by Fletcher...

Author Response:

Done, thanks.

Manuscript Changes:

Pg. 32079, line 24: ‘... than those suggested by Fletcher et al. (1962)...’

Reviewer Comment:

Pg. 23080, lines 20-25. Please check your use of past-present tense. Specifically: ...observations *were* typically... and ...DeMott et al. (2010) developed...

Author Response:

Thanks, the tense has been changed, as suggested.

Manuscript Changes:

Pg 23080, line 20: ‘These observations were typically for...’

Pg 23080, line 24: ‘DeMott et al. (2010) developed a parameterisation...’

Reviewer Comment:

Pg. 32081, lines 19-21. Please provide a reference for this sentence, and clarify if the first half of this sentence refers to the parameterisation results or field observations.

Author Response:

This sentence refers to the parameterisation results, and as such there is no other reference. The second half of the sentence refers to the observations from Klein et al. (2010) in the immersion mode. This has been clarified in the text.

Manuscript Changes:

Pg 32081, line 19: ‘The results from the immersion parameterisation suggest that high INP concentrations greater than 10^6 m^{-3} are only produced at temperatures less than 250 K,...’

Response to anonymous reviewer 2.

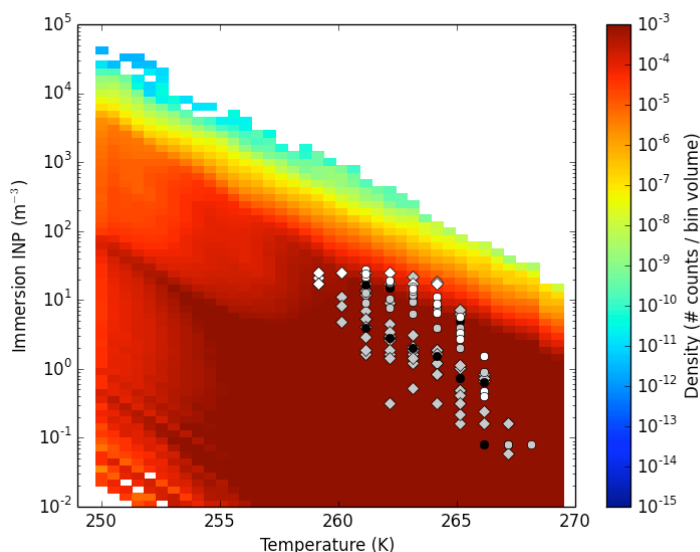
Reviewer Comment:

Would it be possible to show a model-obs comparison on an x-y plot using the data presented in Fig 4 (particularly for immersion freezing)? I recognize that the Niemand et al. (2012) parameterization is only valid over a specific temperature range and these observations are mostly at warmer temperatures, but it would be interesting to know whether extrapolating to the warmer temperature would produce good agreement with the observations. From Fig. 4, it appears that it likely would agree well.

Author Response:

A direct comparison between the model derived INP concentrations and the observations would not be helpful, since we can't reliably temporally and spatially match the observations to the model data.

To address the main question of whether the Niemand et al. (2012) parameterisation can be extended to higher temperatures, we have computed a similar histogram which extends to higher temperatures of 270 K, and down to lower concentrations of 10^{-2} m^{-3} . This is shown below, and indicates that the Niemand et al. (2012) parameterisation can be extended to 270 K. This provides further confidence in the results presented within the manuscript, however since the parameterisation is not valid at these warmer temperatures, the authors don't wish to include this result in the manuscript.



Response Figure 5: Potential immersion INPs for July 2008, compared to observations. Observations are shown from Conen et al. (2012) (black circles: dust from North Italy, grey circles: dust from North Africa/Switzerland, white circles: dust from Switzerland/South Germany), Joly et al. (2014) (grey diamonds: within detection limit, white diamonds: at detection limit)

Manuscript Changes:

None.

Reviewer Comment:

Figure 4 would have a more intuitive interpretation (to my mind) if it were normalized such that the integral of each temperature bin were unity. On p. 32081, l. 3, it is stated that “From Fig. 4, most immersion INPs are occurring at temperatures warmer than 250 K”, but doesn’t the figure simply show that the temperature in the model domain is almost always warmer than 250 K? It would be more useful to know, for a fixed temperature, the probability of different values of potential INP occurring.

Author Response:

Figure 4 actually is already normalized to the total samples in a temperature bin. The colour bar label is wrong and probably contributed to the confusion. The label should read ‘bin count / sample count / bin area’. This has been explicitly stated in the manuscript.

Manuscript Changes:

Pg 32080, line 26: ‘Figure 4 shows a 2-D histogram of normalized potential INP concentrations...’

Pg 32092: Figure Caption: ‘(LEFT): Normalised potential depositions..., (RIGHT): normalized potential immersion...’

Figure 4: The y-axis now extends down to concentrations of 10^{-2} m^{-3} .

Reviewer Comment:

The direct comparison with the DeMott (2010) parameterization in Fig. 4 could be misleading. First, the histogram convolves the temperature dependence of IN activation with the distribution of dust particle in the atmosphere. Both particle concentrations and temperatures will be higher at lower altitudes, and both impact the histogram. The DeMott et al. parameterization is also a function of particle concentration, but in this figure the particle concentration has been set to a constant value (I assume), so it is not straightforward to compare these two variables. The comparison would be more straightforward if the histogram were normalized as suggested in the previous comment. Furthermore, the DeMott et al. (2010) parameterization is not composition-specific, while the parameterization applied in this paper is applicable only to dust aerosol. Tobo et al. (2013) and DeMott et al. (2015) have published coefficients for this parameterization that are specific to dust, which would be more appropriate for this comparison. The dust-specific comparison should be used either in addition to, or in lieu of, the DeMott et al. (2010) parameterization, and it should be mentioned in the figure caption that a fixed particle concentration was used for the parameterization.

Author Response:

The dashed line shown in Fig 4 is not the DeMott et al. (2010) parameterisation, it's the best fit to the observations presented in the paper. This was unclear in the manuscript, and it has been clarified. The idea to directly compare the DeMott et al. (2015) parameterisation for dust aerosols is nice, and this has been included in Fig 3, left panel, using the yearly mean temperature and median dust concentrations.

Manuscript Changes:

Pg 32080, line 24: 'DeMott et al. (2010) provide a best fit function to a number of observations from outside Europe, which is used here as an additional evaluation tool.'

Figure 3: Revised version of figure 3 is shown above.

Reviewer Comment:

The same issue also affects the comparison with the observed INP concentrations. I believe the temperatures of all of these observations are instrument temperatures, not ambient temperatures (which should be pointed out to the reader). But, here they are being compared with a histogram of INP concentrations that is based on the number of INP active at ambient temperatures, which is not an apples-to-apples comparison, for the reasons mentioned above.

Author Response:

Thanks, this is an important point which has been clarified in the manuscript.

Manuscript Changes:

Pg 32081, line 2: ‘Finally, note that the temperatures of the observations are instrument temperatures, and the model INPs are calculated at the modeled ambient temperature in the grid box.’

Reviewer Comment:

p. 32075, l. 9: “this parameterization is active a[t] colder temperatures : : :” ! does this mean that the parameterization is only valid in the range given, or that particles are IN active at those temperatures? Or that this parameterization is applied in the model when the conditions are within the specified range?

Author Response:

Correct, the deposition parameterisation (as well as the immersion parameterisation) are only valid within the stated temperature limits. These limits represent the temperatures investigated during the AIDA experiments used to derive the parameterizations. It’s still an open question as to whether the parameterizations can be extrapolated to temperatures outside these ranges, but the histogram presented above goes some way to answering this.

Observations indicate that particles can be INP active at temperatures outside these ranges, particularly for immersion freezing at temperatures warmer than the upper limit of the Niemand et al. (2012) parameterisation (eg, see Figure 4).

The last part is also correct, the parameterizations are only applied to the modeled data within the respective temperature ranges. However, we recommend that constant INP concentrations are used at colder temperatures to prevent zero INP concentrations.

Manuscript Changes:

Pg 32075, line 21: ‘...calculation of potential INPs, and only if the temperature was within the range of the relevant parameterisation.’

Reviewer Comment:

p. 32075, l. 17-24: does “potential immersion INP concentration” here mean the ambient temperature in each model grid box was used to calculate the number of immersion INP active at that temperature? (and the same question applies for the “potential deposition INP concentration”)

Author Response:

Yes correct. This has been clarified in the manuscript.

Manuscript Changes:

Pg 32075, line 21: ‘The ambient temperature in each model grid box was used for the calculation of potential INPs,...’

Reviewer Comment:

p. 32072, l. 4: “it’s” -> “its”, omit comma after “concentrations”

Author Response:

Done.

Manuscript Changes:

Pg 32072, line 4: ‘... concentrations, and it’s seasonal...’ changed to ‘...concentrations and its seasonal...’

Reviewer Comment:

p. 32074, l. 20: “Modell” -> “Model”

Author Response:

Thanks.

Manuscript Changes:

Pg 32074, line 20: ‘Modell’ changed to ‘Model’

Reviewer Comment:

p. 32075, l. 9: “active a” -> “ active at”

Author Response:

Thanks.

Manuscript Changes:

Pg 32075, line 9: ‘active a’ changed to ‘active at’

Reviewer Comment:

p. 32077, l. 25: “northern most” -> “northernmost”

Author Response:

Done!

Manuscript Changes:

Pg. 32077, line 25: ‘northern most’ changed to ‘northernmost’

Reviewer Comment:

p. 32082, l. 22: typo in “parameterisations”

Author Response:

Thanks, it’s been corrected.

Manuscript Changes:

Pg 32082, line 22: 'paramterisations' changed to 'parameterisations'