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Seasonal variability of Saharan desert dust and ice nucleating particles over Europe

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Abstract

Dust aerosols are thought to be the main contributor to atmospheric ice nucleation. While there are case studies supporting this, a climatological sense of the importance of dust to atmospheric ice nucleating particle (INP) concentrations, and it's seasonal variability over Europe is lacking. Here, we use a mesoscale model to estimate Saharan dust concentrations over Europe in winter and summer of 2007–2008. There are large differences in median dust concentrations between seasons, with the highest concentrations and highest variability in the lowest 4 km. Laboratory based ice nucleation parameterisations are applied to these dust number concentrations to calculate the potential INP resulting from immersion freezing and deposition nucleation on these dust particles. The potential INP concentrations generally increase with height due to decreasing temperatures in the lower and mid-troposphere and exhibit a maximum in the upper troposphere where INP concentrations decrease again with altitude due to decreasing dust concentrations. The potential INP profiles exhibit similarly large differences between seasons, with the highest concentrations in winter (median potential immersion INP concentrations up to 10^3 m^{-3}, median potential deposition INP concentrations at 120 % relative humidity with respect to ice up to 10^5 m^{-3}) occurring closer to the ground for both nucleation modes. Using these results, a best-fit function is provided to estimate the potential INPs for use in limited-area models, which is representative of the normal background INP concentrations over Europe. A statistical evaluation of the results against field and laboratory measurements indicates that the INP concentrations are in close agreement with observations.

1 Introduction

Atmospheric aerosols have an important influence on cloud properties through the direct and indirect aerosol effects, however there is significant uncertainty in quantifying both of these. Considering only the indirect effects, the ice phase has a particularly

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strong influence on cloud properties by affecting cloud lifetime and precipitation processes (Lohmann and Feichter, 2005; Boucher et al., 2013). The ice nucleating ability of many aerosols has been experimentally determined through both field (e.g., Cozic et al., 2008; Conen et al., 2012; Joly et al., 2014) and laboratory studies (Hoose and Möhler, 2012, and references therein). Mineral dust has been identified as a major contributor to atmospheric ice nucleation at temperatures relevant for mixed phase and cirrus clouds (Heintzenberg et al., 1996; DeMott et al., 2003; Atkinson et al., 2013). During large Saharan dust outbreaks, model results suggest that dust aerosol concentrations can reach $10^7\,\mathrm{m}^{-3}$ over Europe (Bangert et al., 2012), but it also appears dust dominates the normal background ice nucleating particle (INP) and ice residual composition in the absence of these large dust events (Targino et al., 2006; Prenni et al., 2009; Kamphus et al., 2010; Cziczo et al., 2013). This lends some weight to the notion that dust can have an important indirect effect on clouds (Sassen, 2002; Sassen et al., 2003) on seasonal timescales.

Other important ice nucleating aerosols are soot and biological particles (Pratt et al., 2009), however their contribution to ice nucleation is on average lower than that of dust (Hoose et al., 2010, and references therein). Case studies of the impact of dust events on INP concentrations in Europe have been performed (Klein et al., 2010; Chou et al., 2011), however, climatological estimates of dust number concentrations and the resulting INP concentrations, as well as an understanding of their seasonal variability, remain elusive.

Ice nucleation in the atmosphere takes place via three different pathways: immersion/condensation, deposition, and contact freezing. Efforts to parameterise these processes for use in models have relied on either empirical evidence or a theoretical approach, yielding a wide variety of parameterisations (e.g., Fletcher et al., 1962; Cooper, 1986; Meyers et al., 1992; DeMott et al., 2010). Typically, these parameterisations are independent of the aerosol type, however more recently, dust aerosol specific parameterisations have begun to emerge (Niemand et al., 2012; Steinke et al., 2014; DeMott et al., 2014; Hiranuma et al., 2014).

The aims of this manuscript are straightforward. Firstly, we will quantify the background dust number concentrations in Europe during different seasons using model data from December 2007–August 2008. This will allow the quantification of the potential INP concentrations resulting from immersion freezing and deposition nucleation on these particles, using two new parameterisations specific to dust aerosols. These results will then be used to develop a best-fit function which can be used to estimate immersion and deposition INP concentrations in regional climate and numerical weather prediction models, and for process studies. Finally, a statistical comparison with available observations will be presented.

2   Saharan dust and INP concentrations

The COnsortium for Small-scale MOdelling (COSMO) meteorological model coupled to the MUlti-Scale Chemistry Aerosol Transport (MUSCAT) was used to simulate the generation and transport of Saharan desert dust to Europe for December 2007–August 2008. The model was configured to simulate dust in 5 size bins from 0.1–24 μm. The dust model uses a horizontal grid resolution of 28 km and 40 vertical layers. Dust emission fluxes depend on surface wind friction velocities, surface roughness, soil particle size distribution, and soil moisture in unvegetated areas. Dust advection is computed by a third-order upstream scheme, particle removal is computed considering dry and wet deposition processes. COSMO simulations were initialised with analysis fields from the global model GME (Global Modell of the DWD) and the lateral boundary conditions updated 6 hourly. The simulations were re-initialised every 48 h to keep the modelled meteorology close to the analysis fields. The model results have been extensively evaluated with field measurements of ground-based and airborne measurements of dust concentrations as well as size distribution, aerosol optical thickness and lidar backscatter and extinction (Heinold et al., 2009, 2011; Tegen et al., 2013). It performed well in a regional model intercomparison of an observed event in the Bodele depression in Chad (Todd et al., 2008).

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The simulated dust number concentrations were used to estimate the potential immersion and deposition INP. Niemand et al. (2012) provides a parameterisation for immersion freezing on desert dust particles. This work is derived from experiments on a variety of dust types carried out at the Aerosol Interaction and Dynamics in the Atmosphere (AIDA) cloud chamber facility, and is valid between temperatures of 261–237 K at or above water saturation. The parameterisation is a function of the dust particle surface area and the temperature. Similarly, a parameterisation for deposition freezing on Arizona Test Dust was experimentally determined by Steinke et al. (2014) from AIDA measurements. This parameterisation is active a colder temperatures of between 253–220 K, and above ice supersaturation. 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.e. in the cirrus regime where deposition nucleation is most relevant. At present, a comparable parameterisation based on laboratory experiments for natural desert dusts covering the entire required temperature and humidity range is not available.

The potential immersion INP concentration is simply the parameterised immersion INP concentration irrespective of the relative humidity with respect to water. Similarly, the potential deposition INP concentrations are calculated from the simulated dust concentrations and the parameterisation from Steinke et al. (2014) at a prescribed relative humidity with respect to ice (RH_{ice}), irrespective of the actual in-situ RH_{ice}. The potential immersion and deposition INP concentrations are presented here in order to be independent of the resolved and parameterised clouds in COSMO–MUSCAT. The domain considered here is over central Europe, between 44–60° N and 0–20° E.

Figure 1 shows the horizontal mean, median, and the 25th and 75th percentiles for temperature, total dust surface area, total dust number concentration, and the parameterised potential immersion and deposition INP concentrations for winter (DJF) and summer (JJA) of 2007–2008. The potential deposition INP concentrations are calculated using a constant RH_{ice} of 110 %. To account for a range of possible in cloud

relative humidities, potential deposition nucleation is also shown at 120 %. The vertical coordinate in Fig. 1 is height above the terrain.

Temperatures are approximately 12 K colder in winter, and the tropopause is over 10 km in both seasons. Qualitatively, the dust surface area and the dust number con-
centrations have similar vertical profiles, which implies there is no significant change in the size distribution with height. The highest concentrations of dust are below 4 km, with median values in summer around 5×10^4 m^{-3}, and in winter over 2×10^5 m^{-3}. It is also in this region where there is the largest amount of variability in dust concentra-
tions, particularly in summer. The 75th percentile value is up to 40 times larger than the median in summer, and 3 times larger than the median during winter. Between 4 km and the tropopause, median dust concentrations in summer are about an order of magnitude lower than those in winter. The mean values of dust are 30 times larger than the median values in summer, and 5 times larger in winter, implying that infrequent but significant dust events contribute a lot to the mean values.

Looking at the more extreme values, the maximum in the 95th percentile (not shown) of the dust number concentration is around 5.8×10^6 m^{-3} in the lowest 2 km. This is still lower than, but close to those reported by Bangert et al. (2012) during a large Saharan dust event during May 2008.

These dust number concentrations translate into maximum median potential INP concentrations in the immersion mode of 4×10^3 m^{-3} at an altitude of 6.5 km above the terrain height. Median INP concentrations are higher, and occur at lower alti-
tudes in winter than summer. The summertime maximum concentration is just over 2.6×10^3 m^{-3} at just below 8 km above the terrain. The area bounded by the 25th and 75th percentiles spans more than an order of magnitude at the maximum concentration for both seasons.

In the deposition mode, median potential INP concentrations at RH$_{ice}$ of 110 % with respect to ice are up to 7×10^4 m^{-3} at about 8.5 km during winter, which is also the sea-
son during which the highest concentrations occur. Again, due to the colder air tem-
peratures, the maximum in the potential deposition concentrations occur about 2 km

lower in the winter compared to the summer months. The summertime maximum is around 1.6×10^3 m^{-3}. The 25th and 75th percentiles indicate slightly less variability in the deposition mode for both seasons.

The final panel in Fig. 1 shows potential deposition INP concentrations at RH$_{ice}$ of 120 %. Here, the profiles closely resemble the potential deposition INP at 110 %, but just shifted to higher concentrations. Now the maximum median concentrations are approximately twice as large as those at 110 %.

The mean values for both freezing modes covers a much larger vertical range than the median values, from close to the surface up to the tropopause. Furthermore, the maximum means in the immersion mode are an order of magnitude larger than the maximum medians. The difference is less in the deposition mode. This again suggests the mean concentrations are dominated by a few very significant events of large INP concentrations. The peak in INP concentrations is due to there being less dust available for nucleation at higher altitudes.

Figure 2 shows the median, 25th and 75th percentiles for the meriodionally averaged total dust number concentration, potential immersion, and potential deposition INP concentrations, as a function of latitude and at an altitude of 5 km above the terrain. The bottom panel also shows total dust surface area, and temperature at 5 km. The dust concentrations and total surface area are remarkably constant with increasing latitude. The bottom panel shows some variability in temperature. There is about a 3.5 K drop in temperature over the high alpine regions around 47° N, and another decrease in temperature north of 53° N. This means that there is an amplification of INP concentrations in these colder regions. In the immersion mode, median potential concentrations change from a maximum of 8×10^3 over the high terrain, down to 1×10^3 m^{-3} a few degrees further north, and then increase again to about 5×10^3 m^{-3} at the northern most point of the domain. A similar change occurs in the deposition mode, with maximum and minimum median potential concentrations of 2×10^3 and 4×10^2 m^{-3} respectively. At higher altitudes, where most of the INPs are located, there is a smaller amount of variability with latitude (not shown). This implies that the potential immersion and

deposition vertical profiles presented in Fig. 1 are a suitable representation of INP concentrations over the whole domain considered here.

The shape of the median INP profiles in Fig. 1 resembles that of a skewed normal distribution. Therefore, the median profiles can be described by applying a normal probability density function:

$$\phi(z) = \frac{1}{\sigma\sqrt{2\pi}}\exp\left[\frac{-(z-\mu)^2}{2\sigma^2}\right] \tag{1}$$

with a cumulative distribution function given by:

$$\Phi(\alpha z) = \int_{-\infty}^{\alpha z}\phi(t)\mathrm{d}t = \frac{1}{2}\left[1 + \mathrm{erf}\left(\frac{\alpha\left(\frac{z-\mu}{\sigma}\right)}{\sqrt{2}}\right)\right] \tag{2}$$

where z is the height in meters, and erf is the error function. Then the INP concentration (m^{-3}) as a function of height (m) is given by:

$$C_{\text{INP}}(z) = 2a\phi(z)\Phi(\alpha z) \tag{3}$$

In these series of equations, σ is the SD, μ is the median, α is the shape parameter defining the skewness, and a is a scale factor. These parameters are shown in Table 1 for each case of potential immersion and potential deposition INPs in winter and summer. Using Eqs. (1) to (3) along with these parameters, it is possible to specify realistic median INP concentrations for model simulations over Europe. The model diagnosed moisture or the parameterised cloud occurrence must be used to define supersaturated conditions with respect to water for immersion INPs, and with respect to ice for deposition INPs. The median INP number concentrations are used to define these series of equations in order to provide a more realistic representation of the background INP concentrations, independent of large Saharan dust events.

Since the Steinke et al. (2014) parameterisation is a function of both temperature and supersaturation, the skewed normal distribution for deposition nucleation provided here can be simply scaled to the model diagnosed RH_{ice}. Combining Eqs. (1) and (2) from Steinke et al. (2014), and expressing in terms of RH_{ice}, gives:

$$n_s(T, RH_{ice}) = 1.88 \times 10^5 \exp(0.2659(-(T - 273.2) + (RH_{ice} - 100))) \tag{4}$$

Therefore, a deposition scaling factor (DSF) as a function of RH_{ice} can be defined as:

$$DSF(RH_{ice}) = \frac{n_s(T, RH_{ice})}{n_s(T, RH_{ice} = 110)} \tag{5}$$

$$= \exp(0.2659(RH_{ice} - 100) - 2.659) \tag{6}$$

Finally, the scaled INP concentrations due to deposition nucleation, are approximately:

$$C_{INP}(z, RH_{ice}) \approx C_{INP}(z) \times DSF(RH_{ice}) \tag{7}$$

where $C_{INP}(z)$ is the concentration of deposition INPs at $RH_{ice}=110\%$, given by Eq. (3), and the approximation is most valid for small activated fractions.

Figure 3 shows the INP concentrations derived from Eqs. (3) and (7) at $RH_{ice} = 120\%$, compared to the Meyers et al. (1992) parameterisation for deposition nucleation, and the Fletcher et al. (1962) and Cooper (1986) parameterisations for immersion freezing. For the Fletcher et al. (1962) and Cooper (1986) formulations, the mean winter temperature was used. The limits on the height of the INP estimates are indicated by the solid sections of the best-fit curves. This represents the height range of the model data used to construct the best-fit function. In order to prevent sharp discontinuities in the vertical extent of the modelled INP, these limits may be disregarded, provided INPs are limited to below the tropopause.

For immersion freezing, both parameterisations are larger than the wintertime INP concentrations presented here. The summertime concentrations shown here are many orders of magnitude lower at 5 km than those suggested Fletcher et al. (1962) or

Cooper (1986). Calculating INP concentrations from Eq. (3) has the advantage of capturing a peak in concentrations, and not overpredicting INPs at colder temperatures.

The Meyers et al. (1992) deposition parameterisation is a function of supersaturation with respect to ice, therefore the INP concentrations resulting from nucleation at supersaturation of 1.1 (1.2) is shown as the dashed (dotted) line. At a supersaturation of 1.1, there is good agreement with the maximum summertime concentration. Maximum concentrations in winter are about an order of magnitude higher than Meyers et al. (1992) suggest. At higher supersaturations, the concentrations calculated from Eq. (7) are higher than the Meyers et al. (1992) parameterisation.

Applying Eq. (6) with RH_{ice} of 120 % gives DSF of 14. However, Fig. 1 indicates there are only about twice as many INPs at the higher relative humidity. At higher supersaturations and for the largest particles, the activated fraction is already approaching 1, hence Eq. (6) will overpredict INP concentrations. At RH_{ice} of 115 %, the DSF is 3.8, whereas the parameterised INP concentrations are 1.6 times greater. Observations suggest that in cirrus clouds, RH_{ice} is mostly below 120 % (Haag et al., 2003), meaning the lower RH_{ice} values where the DSF works better are most relevant to observed cirrus clouds.

3 Evaluation

There are case studies investigating ice nucleation over Europe at specific locations, under a variety of atmospheric conditions. These observations are typically for only a few weeks at a time, so climatological time series of ice nuclei are not yet available. Observations presented in several recent studies (Chou et al., 2011; Conen et al., 2012; Joly et al., 2014; Klein et al., 2010) will be used to make a statistical comparison with the results presented here. From other observations outside of Europe, DeMott et al. (2010) develop a parameterisation for the INP number, which is a function of both temperature and aerosol number. Figure 4 shows a 2-D histogram of potential immersion INP concentration in 0.5 K bins for the whole domain in July 2008. Overlayed on

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the figure are the observations from selected field studies, as well as the parameterisation suggested by DeMott et al. (2010).

From Fig. 4, most immersion INPs are occurring at temperatures warmer than 250 K, with concentrations typically less than $10^2\,\mathrm{m}^{-3}$. The Niemand et al. (2012) parameterisation produces most of the INPs with concentrations about 1–2 orders of magnitude lower than DeMott et al. (2010) suggest. Almost all the observations fall within the range of the parameterised immersion INPs, and note that the observations from Klein et al. (2010) were taken during a Saharan dust outbreak resulting in higher than normal INP concentrations. The immersion parameterisation also shows a greater sensitivity to temperature than DeMott et al. (2010) indicate. Observations at colder temperatures from Chou et al. (2011) fall in the middle of the range of concentrations given by the deposition nucleation parameterisation. Again, during Saharan dust events these observations indicate higher concentrations of INPs.

The observations show most of the INPs are at temperatures warmer than the immersion parameterisation allows. According to Joly et al. (2014), most of the measured INPs are biological in origin. These INPs are not considered in this study. However, it is interesting to note that the parameterised dust INP concentrations agree well with the Joly et al. (2014) data at 260 K.

High INP concentrations greater than $10^6\,\mathrm{m}^{-3}$ are only produced at temperatures less than 250 K for immersion freezing, and only observations from during a Saharan dust event suggest concentrations this high. In addition to this, observations shown by DeMott et al. (2010) from the Pacific Dust Experiment suggest INP concentrations can reach over $10^5\,\mathrm{m}^{-3}$. DeMott et al. (2003) presents observations from aircraft measurements of INPs in an air mass which originated from North Africa. At temperatures above the homogeneous freezing threshold, INPs were present in concentrations up to $10^6\,\mathrm{m}^{-3}$. This implies that the INP concentrations presented here are in broad agreement with available observations.

4 Conclusions

The COSMO–MUSCAT model was used to simulate the generation and transport of Saharan desert dust to Europe during December 2007–August 2008. Maximum median dust concentrations are around 5×10^4 m^{-3} in the lowest 4 km during summer, with higher median concentrations in winter of up to 2×10^5 m^{-3}. There is a significant amount of variability in dust concentrations, particularly in summer. The resulting potential immersion INP reach maximum median concentrations of 4×10^3 m^{-3} at 6.5 km during winter. During the summer months concentrations are lower, and the maximum occurs about 2 km higher than in winter. INP concentrations in the deposition mode for RH$_{ice}$ of 110 % reach over 10^5 m^{-3} at around 8.5 km in winter, with lower concentrations occurring at higher altitudes during summer.

Since the median concentrations are constant with latitude, the median profiles of INP concentrations are representative of the background INP concentrations over the whole domain considered here. Therefore, using these results, a mathematical model based on a skewed normal distribution is provided to estimate the INP concentrations as a function of altitude for immersion freezing, and as a function of altitude and RH$_{ice}$ for deposition nucleation. The deposition scaling factor works best for values of RH$_{ice}$ less than about 115 %. This can be applied to process studies and regional climate simulations over Europe wishing to include a realistic description of ice formed from immersion freezing and deposition nucleation on desert dust particles.

The new estimates of INP concentrations were compared to commonly used aerosol independent paramterisations for immersion freezing and deposition nucleation. The peak concentrations are smaller than the earlier parameterisations for immersion freezing, and mostly larger for deposition nucleation. The approach presented here captures a much more realistic vertical and seasonal variability, thus providing an extra level of utility for model simulations over Europe. A statistical evaluation with available observations indicates the Niemand et al. (2012) and Steinke et al. (2014) parameterisations

produces most of the INPs at similar concentrations to what the observations suggest, providing confidence in the results presented here.

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Table 1. Parameters defining Eqs. (1) to (3), for immersion and deposition INPs (at RH_{ice} = 110 %) for winter and summer.

	σ (m)	μ (m)	α	a
Winter Imm	749.106	6771.768	−2.943	5.268×10^6
Winter Dep	1078.682	8999.464	−2.721	1.168×10^8
Summer Imm	518.604	7959.074	−2.278	2.210×10^6
Summer Dep	1198.783	10 499.219	−0.674	4.628×10^6

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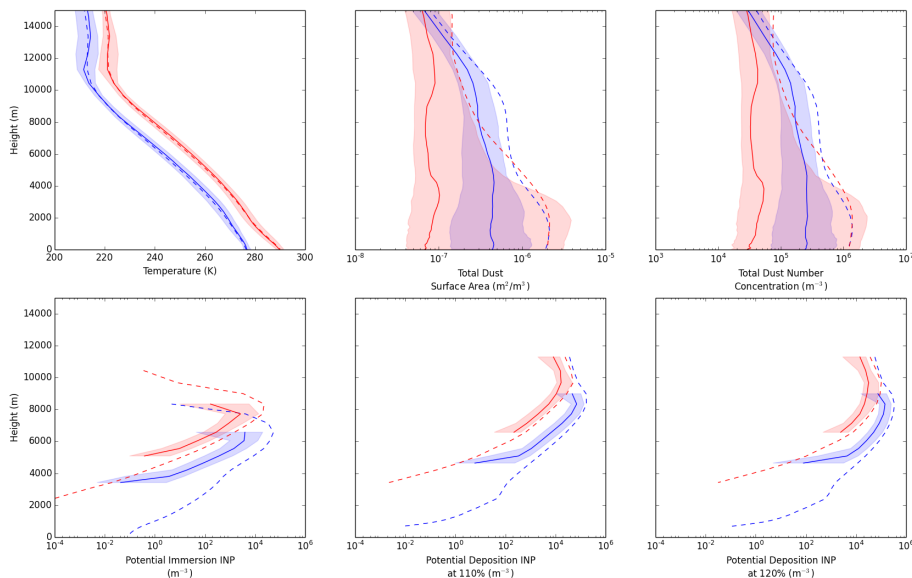
Figure 1. Median (solid) and mean (dashed) temperature, total dust surface area, total dust number concentration, potential immersion INP, and potential deposition INP at RH_{ice} of 110 and 120 % over Europe for summer (red) and winter (blue). The area between the 25th and the 75th percentiles is shaded.

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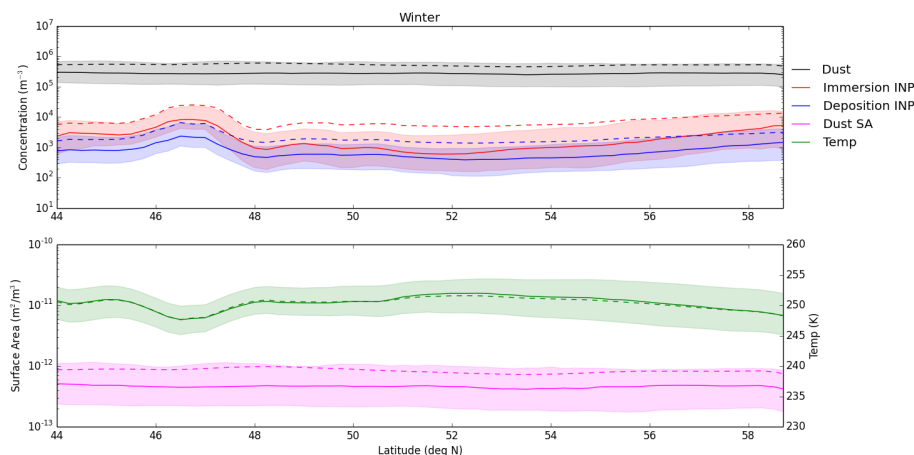
Figure 2. Top panel: median (solid) and mean (dashed) total dust number concentration (black), potential immersion INP (red), and potential deposition INP at 110 % (blue) at 5 km above terrain over Europe for winter. Bottom panel: median (solid) and mean (dashed) total dust surface area (magenta) and temperature (green). The area between the 25th and the 75th percentiles is shaded. Note the alpine regions are between about 46–48° N.
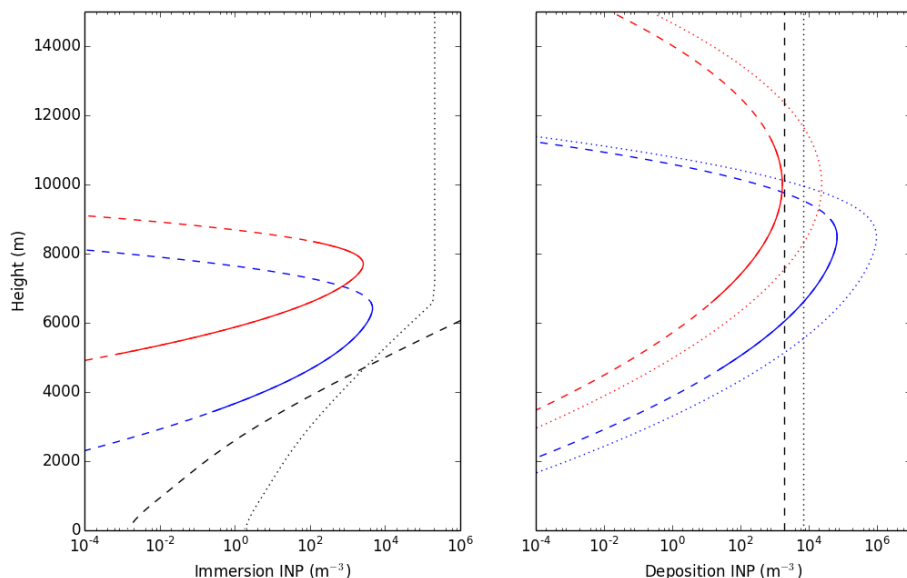
Figure 3. (LEFT): immersion INP concentrations from Eq. (3) for winter (blue) and summer (red). The solid section of the lines is the INP concentrations over the height range used to define the best-fit function. The black dashed line is the Fletcher et al. (1962) parameterisation, and the dotted line represents the Cooper (1986) parameterisation. (RIGHT): deposition INP from Eq. (7) for winter (blue) and summer (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 $S_{ice} = 1.1$ ($S_{ice} = 1.2$).
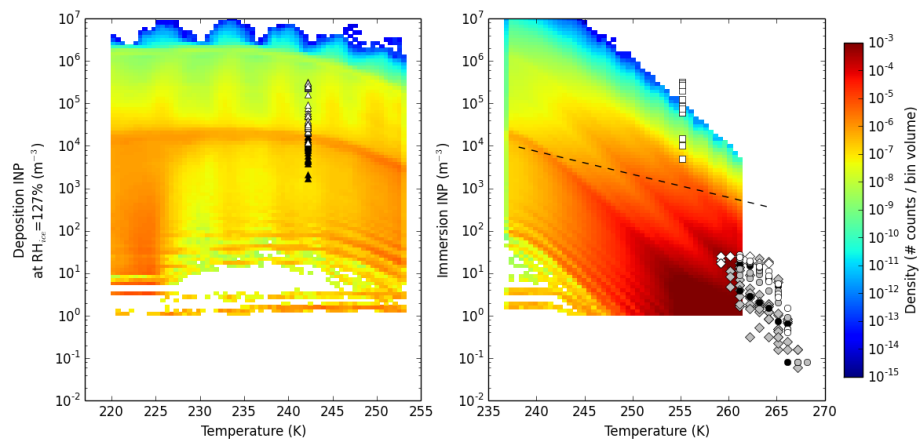
Figure 4. (LEFT): potential deposition INPs at $RH_{ice} = 127\%$, (RIGHT): potential immersion INPs for July 2008, compared to observations. Observations are shown from Chou et al. (2011) (white triangles: Saharan dust event, black triangles: non-dust event), Conen et al. (2012) (black circles: dust from North Italy, grey circles: dust from North Africa/Switzerland, white circles: dust from Switzerland/South Germany), DeMott et al. (2010) (dashed line), Joly et al. (2014) (grey diamonds: within detection limit, white diamonds: at detection limit), and Klein et al. (2010) (white squares).