

Allen Bertam - ACP Editor,

Dear Allen,

We have revised the manuscript “Size-dependent ice nucleation by airborne particles during dust events in the Eastern Mediterranean” by Reicher et al., in view of the constructive and valuable referees’ comments as outlined in detail below. Here attached please find the point by point response to both referees.

We thank the referees for their comments. Consequently, we have revised the introduction section to include wider background and literature, clarifications of few technical details, and a more robust comparison of our findings with recent literature. The corrections and additions support our motivation for this study, as well as our conclusion and scientific contribution.

We enclose also the revised manuscript and a revised manuscript where we have highlighted the modified text.

It is our belief that that the manuscript was improved and hope that it will be considered for publication in Atmospheric Chemistry and Physics.

Best regards,

on behalf of all coauthors,

Yinon Rudich and Naama Reicher

*The authors are grateful to both of the referees for their positive feedback on the manuscript, and for their valuable and constructive corrections and suggestions that have improved the manuscript. Below we address each of the comments listed in red font. Our answer listed in black font and revised text is listed in green font. Number of lines in our answers are based on the revised manuscript.*

### Referee 1:

Review of “Size-dependent ice nucleation by airborne particles during dust events in the Eastern Mediterranean” by Reicher et al. General comment This study investigated the ice-nucleating abilities of size-segregated mineral dust particles from seven different events in the Eastern Mediterranean. The ice-nucleating abilities of the collected samples were tested using two different techniques the Weizmann Supercooled Droplets Observation on Microarray (WISDOM) and Bielefeld Ice Nucleation ARraY (BINARY). The authors found a large variability in the INP concentration as a function of the aerosol source. Additionally, supermicron particles were found to be an important contributor to the INP concentration. The authors report that the current data is in agreement with literature data and provided new parametrizations as a function of particle size. This is an interesting and sound manuscript. The present results are important for the ice nucleation community and can be useful for modelers. The experiments were well designed and were properly executed. The paper nicely fits with the ACP scope and it can be accepted for its publication after the following points are properly addressed.

### Major Comment

1. The introduction is too short and important information is missing. For example, it is missing what are the characteristics of mineral dust that makes them good INPs. Also, based on the vast literature data, how aging can modify the ice nucleating abilities of mineral dust particles. How much dust is transported to the Mediterranean annually, and when does it happen. It may be important to briefly mention that the presence of mineral dust not only affect cloud formation. What other impacts can be assigned to the presence of mineral dust?

The introduction was revised and extended, and now includes more details and references to previous studies.

2. I found the inter-comparison of the current data with literature data very selective. I suggest to add other previous studies to provide more robust and solid conclusions.

We agree that adding more literature data would probably increase the robustness of our conclusions. There are many studies that provide INP concentrations, but  $n_s$  values for mineral-dust dominated aerosols in the immersion freezing mode are lacking. Figure 9(a) now includes two studies which measured airborne particles for relatively long time period in the Eastern Mediterranean. Within the sampling period, few Saharan dust events occurred. To Figure 9(b) we added parameterization by Niemand et al (2012) for desert dust, and by Niedermeier et al (2015) for K-feldspar particles.

3. Given that particle size is the focus of this manuscript, I am surprised this was not deeply discussed in the manuscript. What was found before this study in terms of particle size vs. ice nucleating abilities of different aerosol particles? Is the current data in agreement with previous studies? What fraction of the INPs measured in the current study correspond to super-micron size particles?

We now added previous findings to our discussion in section 3.3 (line#251):

“Previous studies also pointed out the significant contribution of supermicron particles to the INP population. Mason et al. (2016) studied the immersion freezing abilities of airborne particles in North America and Europe, and found that supermicron particles dominated the freezing, especially at relatively higher temperature (258 K). Recent measurements in a coastal tropical site conducted by Ladino et al. (2019) also found high concentrations of INPs at relatively high temperatures ( $> 258$  K) due to supermicron particles. In these studies, however, mineral dust is not expected to dominate the samples, and bioaerosol particles are thought to dominate the freezing at the higher temperatures ( $> 258$  K). At lower temperatures (below 253 K), Ladino et al. (2019) suggested that mineral dust dominated the freezing. Moreover, DeMott et al. (2010) found that INP concentrations are correlated with particles  $> 0.5$   $\mu\text{m}$ . Other studies, such as Rosinski et al. (1986) and Huffman et al. (2013), also found that supermicron

particles were responsible for most of the INP population in some cases, while when changing the freezing mode that was analysed or the measurement meteorological conditions, their contribution was reduced. Vali (1966) in contrast, found that submicron particles dominate freezing in hail melt samples.”

We also updated the conclusions following these additional information (line#416):

“In general, supermicron particles contributed the most to the INP concentration, in agreement with other previous studies (Mason et al., 2016; Huffman et al., 2013; Ladino et al., 2019). However, our current study is probably the only case where mineral dust dominated the samples. Nevertheless, all of these studies highlight the importance of the supermicron size class of AMD for atmospheric ice nucleation.”

#### 4. I suggest to improve the English in the revised version.

The manuscript has been thoroughly edited and corrected. We believe that this has improved the readability.

#### Minor comments

##### 1. Lines 132-133: What is the reason of using two different cooling rates, and why the transition was made at 263K?

The advantage of using two different cooling rates is that the experiment will run faster. Usually, down to 263 K, no freezing activity was observed anyway, and, therefore, there was no need for a high temperature resolution of 1 K per minute. Hence, cooling to 263 K was achieved in 3.5 minutes instead of cooling for 35 minutes (from room temperature ~ 298 K).

The text was revised to explain that (line#159):

“The device was first cooled at a faster constant rate of 10 K min<sup>-1</sup> from room temperature to 263 K, since freezing events were not expected and indeed were never observed in that temperature range. Then a constant cooling rate of 1 K min<sup>-1</sup> was used until all the droplets froze.”

2. Line 224 and along the text + Figures: I suggest change the units to L-1 as this is commonly used in the ice nucleation community.

Changed.

3. Lines 246-247: “indicates that the supermicron particles are better INP than the submicron ones”. Is this in agreement with literature? Please add a proper discussion here.

This is now detailed in the text as shown in point #3 above.

4. Lines 238-239: I am wondering if the agreement will be improved if the parametrization is based on total particles and not divided by size. This may be a better comparison with literature data

As seen in Figure 6, the  $n_s$  values of the larger particles ( $D_{50}=5.6$ ) are normally larger than those of the other particle classes, and – since the graph is on a log scale – summing up would not change the  $n_s$  values much.

5. What is the effect of marine aerosol. The back trajectories show that there is chance that marine particles can affect the ice nucleating abilities of the Saharan dust particles. This is not mentioned/discussed at all.

In lines #302-306 we speculated that the passage of dust over the sea could affect its activity and explain why in some cases submicron particles are less efficient than supermicron and in some cases similar. However, we have no way to prove that there was interaction with the marine aerosol and other factors, such as how long it remained over the sea. Information about possible interactions and the effects of the Mediterranean sea is now added to the introduction (line#72):

“Levin et al. (1996) found that AMD particles transported over the Mediterranean Sea were often coated with sulphate and other soluble materials, which could affect clouds’ microphysical properties and can eventually result in enhanced ice nucleation.”

6. Line 348: I suggest to add a little discussion here on how large or small is this number in comparison to the INP concentrations reported in other environments or parts of the planet (e.g., Polar regions, marine, agricultural, tropics, etc).

Since INP concentration is not normalised quantity and can be affected by technical issues, such as the amount of material that was immersed in the analysed droplets or the freezing mode that was used, in the revised text we compare our ice-active site density ( $n_s$ ) data, which is more robust, to recent studies, as detailed above in point #2 in major comments.

7. Figure 9: what of the samples tested in Boose et al. (2016b) are shown here? Please add this to the main text as well.

We show the four samples of airborne particles. It is detailed in the text in lines #356-357: “Boose et al. (2016b) analysed airborne particles which were deposited in the Eastern Mediterranean region in Egypt, Cyprus and the Peloponnese (Greece) during dust events. Boose et al. (2016b) also sampled airborne particles during dust events over Tenerife, off West Africa.”

#### Technical comments

Line 34: Add a reference after “troposphere” and after “climate”.

Added

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Line 36: “Key properties” such as?

In the revised version we have added details on the “key” properties in the introduction line#52. We did not want to repeat this information, so at that point of the text we added the clarification word ‘surface’ before “key properties”.

Line 37: ...in THE characterization.

Added.

Line 40: “most prominent”. What does it mean?

We meant to say that it is very dominant and common in the Earth atmosphere. We have reworded the text to “abundant” (line#41):

“One of the most abundant INP in the atmosphere is mineral dust..”

Line 45-46: “Field observations have identified an increase in INP concentrations and ice clouds formation in the presence of mineral dust”. Are these the only 2 studies that found this?

Additional appropriate references were added (line#43).

Line 51: Add a reference after “calcite”.

Added (line#57).

Line 55: “and suggested its importance for atmospheric ice formation”. This reads a bit awkward.

We rephrased the sentence (line#61-64):

“Traditionally, clay minerals were thought to be responsible for atmospheric ice nucleation because they compose much of the dust fraction. However, using standard mineral particles, Atkinson et al. (2013) showed that K-feldspar is the most efficient type, and suggested that it could dominate atmospheric ice formation at relatively high temperatures, above 258 K.”

Line 56: “quartz mineral phases”. What does it mean?

It means different types of the quartz mineral, but to avoid confusion, we deleted the word “phases”.

Line 61-62: “While there are only few measurements of AMD near source regions”. Just Price et al. (2018)?

Added (line#80)

Line 63: Please add more studies together with the Niemand et al., (2012) study.

Added (line#82)

Line 64: Add a reference after “AMD”.

Added (line#83).

Line 64: Surface-sampled natural...

Rephrased (line#81-82):

“..freezing properties of natural dust or soil samples collected from deserts or standard dust particles.”

Line 65: “to laboratory processes” such as?

Rephrased (line#85-86):

“possibly due to laboratory processes, such as milling or sieving, that were applied to the natural dust samples and may have enhanced its activity”

Line 72-77: This belongs to the methodology section.

The methodology details removed from the introduction and the paragraph was rephrased (line#97-101):

“The ability of the collected particles to initiate immersion freezing was studied using the Weizmann Supercooled Droplets Observation on a Microarray (WISDOM) instrument (Reicher et al., 2018), and one of the dust events was studied using the Bielefeld Ice Nucleation ARraY (BINARY) instrument (Budke



and Koop, 2015). We characterized the concentrations and the density of ice nucleation active sites (INAS) of AMD in different size-classes for several different dust cases, as well as combined recent literature and available AMD data to understand how well AMD is represented in models based on recent parameterizations. “

**Line 88: I think it is redundant to write MOUDI impactor. I suggest to briefly describe the MOUDI principle.**

A short description of the MOUDI impactor is now added to the text (line#112-116):  
“The MOUDI is a 10-stage impactor with 18  $\mu\text{m}$  cut-point inlet stage followed by size segregating stages with cut points ( $D_{50}$ ) between 0.056 and 10  $\mu\text{m}$  in aerodynamic diameter (Marple et al., 1991). The particles are collected on the different stages as function of their aerodynamic diameter. The collection efficiency for each particle size is described in Marple et al. (1991). Sampling time ranged between 17 and 48 h with a 30  $\text{L min}^{-1}$  sample flow rate, similarly to previous studies (Huffman et al., 2013;Mason et al., 2015).”

**Line 107: What is close?**

The distance is now specified in the text (line#131-133):

“Concentrations of particles with aerodynamic diameters smaller than 10  $\mu\text{m}$  ( $\text{PM}_{10}$ ) were measured in the Rehovot station, located about 1 km from our sampling site.”

**Line 112: “particle surface area assumed sphericity and diameter was taken as the midpoint of the GRIMM’s channels”. This is unclear.**

The text was revised to allow a better description of the method (line#136-138):

“In order to estimate the total surface area that was collected on the different stages, we assumed that the particles are spheres and used the diameter of the GRIMM midpoint of the GRIMM’s channels as the particle’s diameter.”

Line 120: “optical diameter > 17.5  $\mu\text{m}$  are assumed to be collected”. Is it a good assumption?

Yes it is a good assumption that is based on the collection efficiency of the MOUDI.

Line 122: “of 0.5  $\mu\text{m}$ ”. Optical diameter?

Yes. Now specified in the text as well (line#147-148):

...” For example, a small fraction of particles with 0.5  $\mu\text{m}$  optical diameter are collected on stage”..

Line 123: “The initial particle concentration that was used”. What does it mean?

Since we analyzed a filter that collected particles for a few hours, we summed up all the OPC readings to obtain the total number size distribution of the particles that were collected, and this is the initial particle concentration.

Line 126: “Immersion freezing properties”. What does it mean?

Rephrased to “Immersion freezing abilities of the sampled ambient particles” (line#152).

Line 128: “30 sec” should be 30 s.

Corrected.

Lines 132-133: “10 K min<sup>-1</sup>” should be 10 K min<sup>-1</sup>.

Corrected.

Line 145: “ice nuclei” should be INP.

Corrected.

Line 145: Do the authors refer to water when talking about a solvent?

Yes, in this study the solvent it is water. It is detailed in Eq. #1.

Line 157: “ice nucleating particle” should be INP.

Corrected.

Line 170: “travelled directly to the sampling site” from where?

We meant to emphasize that in some cases there was a short and direct path to the sampling site, while in others, the path was longer, for example, where the air mass was deflected to the sampling site while travelled in a different path. We rephrased the sentence (line#199):

“...the air masses travelled directly to the sampling site from the source region.”

Line 212: “burnings” remove the “s”.

Corrected.

Line 217: initiated?

Corrected.

Line 218: Add a reference after “concentrations”

This is a claim we made.

Line 239: “increased with the particle size” delete “the”.

Corrected.

Line 240: “the activity was similar” between what?

The activity of the dust in the three different stages is similar, considering the measurement uncertainties. The sentence was rephrased (line#279-280):

“The highest  $n_s$  values were observed in the supermicron range  $D_{50}=5.6 \mu\text{m}$ , followed by  $D_{50}=3.2, 1.8$  and  $1.0 \mu\text{m}$ . The activity of the latter three classes was similar within measurement uncertainties.”

Line 247: “implying they have better active sites”. Better or more? What do the authors mean with better?

‘better’ is now explained in the text (line#287-288):

“they have more active sites or/and active sites that nucleate ice at higher temperatures.”.

Line 256: “important ice-inducing component”. What does it mean?

It means that from a mixture of minerals that the particles are composed of, there is one component that dominates the observed ice nucleation (i.e., these particles are the most active ones).

Line 262-264: I found this quite speculative.

We updated the text so that it will be clear that this is our speculation (line#302-306):

“For example, we propose that the passage of SDS1 and SDS2 over the Mediterranean Sea can contribute to their reduced activity in the submicron range, while for the MDS event, a shorter and relatively direct transport path resulted in less atmospheric processing. Although speculative, these considerations may possibly explain why the freezing activity of submicron particles converged with those of the supermicron particles, but we acknowledge that further measurements are needed to confirm these suggestions.”

Line 324: “Boose et al. (2016)” should be Boose et al. (2016b).

Corrected.

Lines 330 and 333: “(R – square)”. Fix this.

Corrected.

Line 364: Add a reference after “distance”.

Added (line#421).

Line 365: Add a reference after “scale”.

Added (line#423).

References: DOIs are missing and either use the full name or abbreviated name of the journals. Need to be consistent.

Corrected.

Figure 3: “distributions averaged over the entire sampling periods” events?

For each event, the average size distribution is shown. Clarified now in the text:  
“..distributions averaged over the entire sampling periods of the events as monitored by GRIMM OPC during the studied events.”

Figure 4 and 5: Change the units of the INP concentration to L-1 .

Changed.

## Referee 2:

### General statement

This paper presents results of an experimental investigation of the ice-nucleating properties of aerosol particles sampled from the atmosphere over Israel. Five episodes with mineral dust being transported from the deserts of Northern Africa and the Middle East and one case with clear sky were sampled by MOUDI in six size-classes. Aqueous extracts of the samples were analysed by the droplet freezing devices WISDOM and BINARY. Atmospheric mineral dust (AMD) is next to sea salt the largest constituent of atmospheric aerosol, and a major ice nucleating agent. Several previous studies have conducted size-resolved INP measurements. It is evident since long ago (e.g. Georgii and Kleinjung, *Jour. des Recherches Atmosphériques*, 145-156, 1967) that ice nucleating particles (INP) are mostly large particles. This is also found here, and no surprise. However, the new size-resolved data allow a much more detailed understanding of ice nucleation by AMD. Supermicron particles are shown to nucleate at warmer temperatures and to contain higher numbers of INP than submicron particles, even if normalized to the aerosol surface (expressed as surface densities  $n_s$  of INP). The authors conclude from their  $n_s(T)$  curves of the different events and size classes, as well as from the overlap with published  $n_s$  data for minerals, that feldspars dominate the freezing induced by supermicron particles. Quartz dominates ice nucleation

by submicron particles. From the comparison with published data it is further concluded that current parameterizations of  $n_s(T)$  overestimate the activity of airborne dust. As a consequence, the authors derive a new, size-dependent parameterization from their data. The present manuscript is not just another study on ice nucleation by mineral dust. Its size-resolving approach yields substantial and valuable new information. Including the particle size and the modification of mineral dust during transport in the parameterization – such as done here – will help to improve the modelling of cloud glaciation and related effects. The work as a whole is sound and perfectly suited to the scope of the journal. The advanced experimental methods are well documented. The data are well presented and convincingly interpreted in the light of current knowledge and literature. The manuscript as a whole is crafted very well. I recommend publication after some minor adjustments described below.

### Major comments

Chapters 2.7.1 and 2.7.2 / sonication times: WISDOM sonicates 90 seconds, whereas BINARY does 30 minutes, accompanied by a 20°C warming. I presume the good agreement of both methods suggests that the effect of this different treatment is negligible?

Yes, we do not think the different sonication protocols have any significant effect on the ice nucleating properties of the dust. The sonication used for preparing the suspensions analyzed in WISDOM was a dry sonicator and, hence, this is more intense than the sonicator used for suspension preparation for the BINARY experiments, which was a bath sonicator (much of the energy is lost during the sonication to the water bath). This is the reason for the chosen longer sonication time in the BINARY. We have added some additional explanatory text to section 2.7.1 accordingly (line#153-154):

“Immersion freezing activity of the sampled ambient mineral dust was measured using suspensions of the collected particles that were extracted from the filters by dry sonication (VialTweeter, model UP200St; Hielcher). This type of sonication method is more effective than the ultrasonic bath in which most of the energy is dissipates in the surrounding water.”

There are some laboratory studies on ice nucleation of minerals that might be cited. Consider to mention and discuss these where relevant, either in the introduction or among the results: 1) Welti et al., Ice nucleation properties of K-feldspar polymorphs and plagioclase Feldspars, *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp2018-1271>, 2019 2) Archuleta et al., Ice nucleation by surrogates for atmospheric mineral dust and mineral dust/sulfate particles at cirrus temperatures, *Atmos. Chem. Phys.*, 5, 2617–2634, 2005 3) Lüönd et al., Experimental study on the ice nucleation ability of size-selected kaolinite particles in the immersion mode, *J. Geophys. Res.*, 115, D14201, doi:10.1029/2009JD012959, 2010

Thank you. These references were relevant and have been added to the text (line#51)

Line 161: please spend a few words on how  $A$  was derived from primary data.

It is explained in section 2.5 (line#134), and we added a clarification in the text (line#189-190):

“.. $A$  is the surface area immersed in a single droplet of the experiment, based on the total surface area of particles in the suspension.”

Line 229: I cannot see from Fig.4 that “SyDS2 has a weaker size dependence in comparison to the other dust events . . .” (smaller spread of curves for a given  $T$ ), as you state in line 229.



For SyDS2, in the supermicron range, particles in the stage of  $D_{50}=3.2\ \mu\text{m}$  had lower activity (and also different slope) than in the stage of  $D_{50}=1.8\ \mu\text{m}$ , and in the warmer temperatures (above 247 K), also lower activity than particles in the stage of  $D_{50}=1.0\ \mu\text{m}$ . In the submicron range, particles of stage of  $D_{50}=0.6\ \mu\text{m}$  had lower activity than particles in the stage of  $D_{50}=0.3\ \mu\text{m}$ . In the other events, the dependency of the ice nucleation activity in the particle size was kept, and normally larger particles were more active than the smaller ones.

Figures 6 and 7: The clear case CSDS has the highest  $n_s$  of all data. How is this interpreted? Is the aged tropospheric background aerosol more active than "fresh" mineral dust plumes? Wouldn't that be an interesting result that needs discussion?

The CSDS event had less surface area in the suspension than the rest of the SDS events. We believe that this is the reason for the higher  $n_s$  values: when there is less active material within the droplets, then the droplets experience higher supercooling because there are fewer active sites at warmer temperatures. Therefore, some of the droplets were allowed to cool to colder temperature. As the temperature becomes colder, higher numbers of nucleation sites were activated, and therefore higher  $n_s$  values. In Figure 7 for example, CSDS collapse to the same activity of the other events, and the activity is similar ( $D_{50} = 5.6, 3.2, 1.8, 0.6, 0.3$ ) or smaller than most of the events ( $D_{50} = 1.0$ ).

#### Minor comments:

Line 183: maybe add (MDS) after "Another event"?

Yes, it is more logical to introduce the event here, now the text was revised (line#213):

"Another event was defined as a "mixed dust" event (MDS), because it was more complicated and included contributions of different sources."

Line 184: I believe “west” or “southwest” is better than “south”

Corrected to ‘southwest’ (line#215).

Line 211: although it is without consequences, the reader will be interested to know whether the fires are around Rehovot or farther away. Can you say a word on this?

We added to the text the information about the radius in which the fires occurred (line#241):

“Note that prior to and during this event, a series of biomass burning events occurred in Israel extending to about 100 km north and 50 km east of the sampling site.”

Line 219: maybe add “to ice nucleation” after “supermicron particles”?

Added.

Line 234: You write: “.. ranged from  $10^{-3}$  to almost 1 . . .”; I read exactly  $10^{-1}$  as upper bound.

Thank you, this is indeed correct, the value is almost 0.3 and this is closer to  $10^{-1}$  than to 1. Value was changed in the text from 1 to  $10^2$  (on the Liter scale) (Line#263-264).

Line 322: I believe it is “ice cloud formation” or “formation of ice clouds”, instead of “ice clouds formation”

Corrected to “ice cloud formation” (Line#375).

Line 368: Isn't it “emphasizes”, because it is related to “overprediction” (singular)?

Yes, thank you, corrected.

### Technical items

Line 217: Typo: "Ice nucleation is initiated . . ." instead of "initiates"

Corrected.

Figure 6, CSDS: The diagonal line is missing in the graph.

Fixed.

Figure 9a) delete "r" in the graph's legend, now it says "(submicron class)"

Fixed.

Figure 9: add a) and b) to the left and right graphs

Fixed.

# 1 **Size-dependent ice nucleation by airborne particles during dust events** 2 **in the Eastern Mediterranean**

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11 **Abstract.** Prediction of cloud ice formation in climate models remains a challenge, partly due to the complexity of ice-related  
12 processes. Mineral dust is a prominent aerosol in the troposphere and is an important contributor to ice nucleation in mixed  
13 phase clouds, as dust can initiate ice heterogeneously at relatively low supercooling conditions. We characterized the ice  
14 nucleation properties of size-segregated mineral dust sampled during dust events in the Eastern Mediterranean. The sampling  
15 site allowed to compare between the properties of airborne dust from several sources with diverse mineralogy that passed over  
16 different atmospheric paths. We focused on particles with six size-classes, determined by the Micro-Orifice Uniform Deposit  
17 Impactor (MOUDI) cut-off sizes: 5.6, 3.2, 1.8, 1.0, 0.6 and 0.3  $\mu\text{m}$ . Ice nucleation experiments were conducted in the  
18 Weizmann Supercooled Droplets Observation on Microarray (WISDOM) setup, where the particles are immersed in nanoliter  
19 droplets using a microfluidics technique. We observed that the activity of airborne particles depended on their size-class, where  
20 supermicron and submicron particles had different activities, possibly due to different composition. The concentrations of ice  
21 nucleating particles and the density of active sites ( $n_s$ ) increased with the particle size and particles concentration. The  
22 supermicron particles in different dust events showed similar activity, which may indicate that freezing was dominated by  
23 common mineralogical components. Combining recent data of airborne mineral dust, we show that current predictions, which  
24 are based on natural dust or standard mineral dust, overestimate the activity of airborne dust, especially for the submicron  
25 class, and therefore we suggest to include information of particle size in order to increase the accuracy of ice formation  
26 modelling and, thus, in weather and climate predictions.

## 27 **1 Introduction**

28 Cloud droplets can supercool to 238 K before homogeneous freezing occurs (Koop and Murray, 2016; Rosenfeld and Woodley,  
29 2000). At warmer temperatures, heterogeneous ice nucleation (HIN), where the presence of aerosol particles lowers the  
30 required energy barrier to form a stable ice nucleus is the common pathway of ice formation (Murray et al., 2012; Pruppacher  
31 and Klett, 1997; Khvorostyanov and Curry, 2004; Hoose and Möhler, 2012). These ice-nucleating particles (INPs) can be

32 activated at sub-zero temperatures and subsequently lower humidity conditions, mainly by interaction with supercooled  
33 droplets. INPs are relatively rare particles and comprise only about  $10^{-5}$  of the total ambient particles in the free troposphere  
34 (Rogers et al., 1998). Yet, their interaction with clouds can greatly influence climate (Gettelman et al., 2012; Tan et al.,  
35 2016; Lohmann and Feichter, 2005). Therefore it is important to represent them well in weather and climate models (DeMott  
36 et al., 2010). Currently, ice formation is a source of great uncertainty in cloud and climate models, partly due to the complexity  
37 of ice processes and the insufficient understanding of the key surface properties which determine an INP (IPCC, 2013). To  
38 improve the predictions of models, a great effort is invested in the characterization of INPs and in the development of  
39 parametrizations based on their physical and chemical properties (Cantrell and Heymsfield, 2005; Niemand et al., 2012; Ullrich  
40 et al., 2017).

41 One of the most abundant INP in the atmosphere is mineral dust, which originates in dryland zones, such as deserts (Middleton,  
42 2017; DeMott et al., 2003b). Field observations have identified an increase in INP concentrations and ice clouds formation in  
43 the presence of mineral dust (Ansmann et al., 2008; Rosenfeld et al., 2001; DeMott et al., 2003b; Czikzo et al., 2004; Sassen et  
44 al., 2003). Ice residuals often contain mineral particles (Czikzo et al., 2013; Czikzo and Froyd, 2014; Twohy and Poellot, 2005).  
45 Mineral dust has high spatial and temporal variability, impacting atmospheric, oceanic, biological, terrestrial and human  
46 systems (Garrison et al., 2003; Gat et al., 2017; Jickells et al., 2005; Mahowald et al., 2014; Mazar et al., 2016; Middleton and  
47 Goudie, 2001). Each year, gigatonnes of dust are transported globally over long distances, dominating the atmospheric aerosol  
48 mass and aerosol optical depth (AOD) (Chiapello et al., 1999; Tegen and Fung, 1994; Ben-Ami et al., 2010; Prospero,  
49 1999; Koren et al., 2006). Though the exact property of an aerosol that determines its ice nucleation ability remains unclear, it  
50 was consistently shown that the mineral composition plays an important role (Kanji et al., 2017), and that for a certain mineral  
51 type, larger particles are more effective heterogeneous ice nuclei than the small ones (Archuleta et al., 2005; Lüönd et al.,  
52 2010; Welti et al., 2019). Local surface features such as steps, cracks and cavities, a close match of the surface lattice with that  
53 of ice, or surface hydroxyl groups (Freedman, 2015; Marcolli, 2014; Zielke et al., 2015; Kiselev et al., 2017; Fletcher,  
54 1969; Tunega et al., 2004; Anderson and Hallett, 1976; Pruppacher and Klett, 1997) are believed to be the responsible factors  
55 for the ice nucleation ability of mineral surfaces.

56 Natural mineral dust particles are often chemically similar but differ in their mineralogy (Engelbrecht et al., 2009), and the  
57 particles are often composed of a mixture of minerals (internally mixed), such as clays, quartz, feldspars and calcites (Claquin  
58 et al., 1999). Other common minerals are palygorskite, hematite, halite, gypsum, gibbsite and goethite (Ganor et al.,  
59 1991; Perlwitz et al., 2015; Kandler et al., 2007; Mahowald et al., 2014). The mineralogy of mineral dust is set by its source  
60 region and is considered to be an important factor that determines its freezing characteristics (Zimmermann et al.,  
61 2008; Augustin-Bauditz et al., 2014). Traditionally, clay minerals were thought to be responsible for atmospheric ice nucleation  
62 because they compose much of the dust fraction. However, using standard mineral particles, Atkinson et al. (2013) showed  
63 that K-feldspar is the most efficient type, and suggested that it could dominate atmospheric ice formation at relatively high

64 temperatures, above 258 K. This was further supported by measurements of natural mineral dust from desert surfaces  
65 worldwide, where the importance of quartz mineral was also indicated (Boose et al., 2016b).

66 Airborne mineral dust (AMD) can experience chemical and physical modifications during its atmospheric transport that may  
67 alter dust's ability to nucleate ice (Kanji et al., 2013). It was shown that atmospheric aging processes can change the size, the  
68 morphology and the surface chemistry of the particles. For example, adsorption of organic components on AMD (Murphy et  
69 al., 2006;DeMott et al., 2003a;Falkovich et al., 2004) or coatings of nitrates, chlorides and sulphates which enhance the  
70 hygroscopicity of the particles (Krueger et al., 2004;Laskin et al., 2005;Li and Shao, 2009). Levin et al. (1996) found that  
71 AMD particles transported over the Mediterranean Sea were often coated with sulphate and other soluble materials, which  
72 affect clouds' microphysical properties and can eventually result in enhanced ice nucleation. In addition, mineral dust carries  
73 biological components, such as bacteria and fungi, which are known to have the ability to induce ice nucleation at relatively  
74 high temperatures (Gat et al., 2017;Mazar et al., 2016;Pratt et al., 2009;O'Sullivan et al., 2016). Further modifications that can  
75 occur during AMD atmospheric transport are the differentiation of size and mineralogy. These can occur due to gravitational  
76 sedimentation, for example, where larger particles sediment faster than the smaller ones. Near source regions, dust samples  
77 were richer in components that are more abundant in the coarse fractions, such as quartz and potassium feldspars, while in  
78 remote locations, higher amount of clay minerals and sodium/calcium feldspar were observed (Murray et al., 2012;Schepanski,  
79 2018).

80 While there are only few measurements of AMD close to source regions (Price et al., 2018;Boose et al., 2016a;Ardon-Dryer  
81 and Levin, 2014;Schrod et al., 2017), parameterizations of ice formation in climate models are often based on the freezing  
82 properties of natural dust or soil samples collected from deserts or standard dust particles (Niemand et al., 2012;Connolly et  
83 al., 2009;Ullrich et al., 2017;Atkinson et al., 2013;Broadley et al., 2012), that may not sufficiently represent AMD (Boose et  
84 al., 2016b;Spichtinger and Cziezo, 2010). Natural dust samples showed higher ice nucleation ability than AMD samples,  
85 possibly due to atmospheric processing of AMD that may lead to deactivation, and possibly due to laboratory processes, such  
86 as milling or sieving, that were applied to the natural dust samples and may have enhanced its activity (Boose et al., 2016b).

87 In this study, we sampled airborne particles during dust events in the Eastern Mediterranean and investigated their ice  
88 nucleation abilities. The Eastern Mediterranean is located in the strip of the world's main deserts, and experiences transport of  
89 desert dust from different sources. The main source is the Sahara Desert in North Africa. It is estimated that about 100 million  
90 tons of dust per year is lifted from the Sahara towards the Eastern Mediterranean, during late winter and spring (Ganor,  
91 1994;Ganor and Mamane, 1982;Ganor et al., 2010). In autumn, local dust is transported, commonly from the Arabian Peninsula  
92 and the Syrian Desert (Dayan et al., 1991;Ganor, 1994). The dust events are often associated with the regional Eastern  
93 Mediterranean synoptic systems, such as winter lows and Red-Sea troughs (Ganor et al., 2010). Our sampling site was located  
94 in Israel, where Saharan dust is transported over North Africa and/or the Mediterranean Sea, and Syrian and Arabian dust is

95 transported over land from the east (Ganor et al., 1991). These distinct sources and paths allow investigating the ice nucleation  
96 properties of AMD with diverse origins and transport paths.

97 The ability of the collected particles to initiate immersion freezing was studied using the Weizmann Supercooled Droplets  
98 Observation on a Microarray (WISDOM) instrument (Reicher et al., 2018), and one of the dust events was studied using the  
99 Bielefeld Ice Nucleation ARraY (BINaRY) instrument (Budke and Koop, 2015). We characterized the concentrations and the  
100 density of ice nucleation active sites (INAS) of AMD in different size-classes for several dust cases, as well as combined recent  
101 literature and available AMD data to understand how well AMD is represented in models based on recent parameterizations.

102

## 103 **2 Data and Methods**

### 104 **2.1 Sampling**

105 Airborne particles were sampled during six dust events in 2016 and 2017, detailed in Table 1. Sampling started when the  
106 visibility reduced due to increasing concentrations of particulate matter (PM). The sampling site is located on a roof of a three-  
107 story building in Rehovot, Israel (31.9N, 34.8E about 80m AMSL). The location is often impacted by mineral dust storms,  
108 transported from nearby and distant geographical locations, mainly from the Sahara and Arabia deserts, and less frequently  
109 from the Syrian Desert, depending on the season and the synoptic conditions (Dayan, 1986;Ganor et al., 2010;Kalderon-Asael  
110 et al., 2009).

111 Particles were collected on polycarbonate filters (47mm cyclopor, 0.1  $\mu\text{m}$  isopores; Whatmann), using the Micro-Orifice  
112 Uniform Deposit Impactor (MOUDI) (model 110-R). The MOUDI is a 10-stage impactor with 18  $\mu\text{m}$  cut-point inlet stage  
113 followed by size segregating stages with cut points ( $D_{50}$ ) between 0.056 and 10  $\mu\text{m}$  in aerodynamic diameter (Marple et al.,  
114 1991). The particles are collected on the different stages as function of their aerodynamic diameter. The collection efficiency  
115 for each particle size is described in Marple et al. (1991). Sampling time ranged between 17 and 48 h with a 30 L  $\text{min}^{-1}$  sample  
116 flow rate, similarly to previous studies (Huffman et al., 2013;Mason et al., 2015).

### 117 **2.2 Air mass Back Trajectories**

118 Back trajectories were calculated by a Lagrangian method, using LAGRANTO 2.0 (Sprenger and Wernli, 2015). The  
119 calculation of air mass trajectories was based on wind data from the European Centre for Medium-Range Weather Forecasts  
120 ERA-Interim reanalysis (Dee et al., 2011), available every 6 h, at  $1^\circ \times 1^\circ$  horizontal grid and 60 vertical hybrid levels. For each  
121 6-h time step during each event, 72 h back trajectories were calculated, from all available data grid points with pressure larger  
122 than 850 hPa, resulting in 11 trajectories, which end their path in the lower troposphere for each calculation. In a second step,

123 the Eulerian densities of the resulting trajectories were computed by gridding the trajectories for each event, smoothed by  
124 using a radius of 100 km and interpolated to 1 h. Finally, the trajectory density was summed over the entire event duration and  
125 normalized by the maximum trajectory count.

### 126 **2.3 Dust Column Mass Density Maps**

127 Time averaged maps of dust column mass density (hourly  $0.5^\circ \times 0.625^\circ$ ) reanalysis data were obtained from the Modern-Era  
128 Retrospective analysis for Research and Applications (MERRA-2). Maps were produced using NASA's Global Modeling and  
129 Assimilation Office (GMAO) (Gelaro et al., 2017), for a period of up to 72 h prior to the sampled event.

### 130 **2.4 Particulate Matter Data**

131 Particulate matter mass data were obtained from the Israeli Ministry of Environment website. Concentrations of particles with  
132 aerodynamic diameters smaller than  $10 \mu\text{m}$  ( $\text{PM}_{10}$ ) were measured in the Rehovot station, located about 1 km from our  
133 sampling site. The 5-minutes mean data was used to calculate peak and mean concentrations of the sampled dust events.

### 134 **2.5 Particle Number-Size and Surface Area distributions**

135 Particle size distribution and concentrations between  $0.25$  and  $32 \mu\text{m}$  was measured on site by an optical particle counter  
136 (OPC; GRIMM Technologies model 1.109), in parallel to the MOUDI sampling. In order to estimate the total surface area that  
137 was collected on the different stages, we assumed that the particles are spheres and used the diameter of the GRIMM midpoint  
138 of the different GRIMM's channels as the particles' diameter.

### 139 **2.6 Conversion of GRIMM channels to MOUDI stages**

140 To determine the total surface area collected on MOUDI's filter, a conversion matrix between the GRIMM channels and the  
141 MOUDI stages was applied. The conversion was based on the particle collection efficiency curves of the MOUDI and inter-  
142 stage particle losses reported in Marple et al. (1991). Figure 1 demonstrates the fraction of particles that are collected on the  
143 stages based on their aerodynamic diameter. Freezing analyses focused on stage #2 ( $D_{50} = 5.6 \mu\text{m}$ ), stage #3 ( $D_{50} = 3.2 \mu\text{m}$ ),  
144 stage #4 ( $D_{50} = 1.8 \mu\text{m}$ ), stage #5 ( $D_{50} = 1.0 \mu\text{m}$ ), stage #6 ( $D_{50} = 0.6 \mu\text{m}$ ) and stage #7 ( $D_{50} = 0.3 \mu\text{m}$ ). For example, most of  
145 the particles with an optical diameter  $> 8.5 \mu\text{m}$  will be collected on stage #2 ( $D_{50}=5.6 \mu\text{m}$ ), whereas all the particles with an  
146 optical diameter  $> 17.5 \mu\text{m}$  are assumed to be collected on former stages (inlet and stage #1). In some cases, particles in a  
147 certain size are likely to impact on two different MOUDI stages. For example, a small fraction of particles with  $0.5 \mu\text{m}$  optical  
148 diameter are collected on stage #5 ( $D_{50}=0.6 \mu\text{m}$ ), and most of them impact on stage #6 ( $D_{50}=0.3 \mu\text{m}$ ). The initial particle  
149 concentration that was used is the accumulated sum of all particles for the entire sampling period.



## 150 2.7 Ice Freezing Experiments and Quantification

### 151 2.7.1 WISDOM

152 Immersion freezing activity of the sampled ambient mineral dust was measured using suspensions of the collected particles  
153 that were extracted from the filters by dry sonication (VialTweeter, model UP200St; Hielcher). This type of sonication method  
154 is more effective than the ultrasonic bath in which most of the energy is dissipated in the surrounding water. A quarter filter  
155 was inserted into a 1.5 ml Eppendorf vial with 0.3 ml deionized water, and sonicated in three 30 s cycles, to avoid heating  
156 produced during intense sonication. The suspension was immediately used for droplet production and freezing experiments in  
157 WISDOM as detailed in Reicher et al. (2018). Briefly, an array of 0.5 nL monodispersed droplets (~100 μm diameter,  
158 suspended in an oil mixture) was generated in a microfluidic device that was cooled by a commercial cooling stage (THMS600,  
159 Linkam) under a microscope (BX-51 with 10X magnification, Olympus) coupled to a CCD camera. The device was first  
160 cooled at a faster constant rate of 10 K min<sup>-1</sup> from room temperature to 263 K, since freezing events were not expected and  
161 indeed were never observed in that temperature range. Then a constant cooling rate of 1 K min<sup>-1</sup> was used until all the droplets  
162 froze. The temperature uncertainty was ± 0.3 K, based on error propagation between the calibrated droplet temperature and  
163 the uncertainty of the temperature sensor that is located in the cooling stage (see Reicher et al. (2018) for more details).

### 164 2.7.2 BINARY

165 The Bielefeld Ice Nucleation ARraY (BINARY) is an optical freezing array of droplets pipetted on a hydrophobic substrate in  
166 separated sealed compartments and cooled in a Linkam cooling stage (LTS120) (Budke and Koop, 2015). In the present study  
167 an array of 64 droplets of 0.6 μL was employed. Suspensions were prepared by extracting a quarter filter in 1.5 ml of double-  
168 distilled water (that is, 5 times more diluted than WISDOM suspensions), using a bath sonicator (Elma Transsonic Digital, TP  
169 670/H) for 30 min. The bath temperature increased during sonication from about 288 to 308 K. The obtained suspensions  
170 were used directly and further diluted (1:10) for another set of measurements with reduced surface area of the particles in the  
171 droplets. For the freezing experiments, the droplets were cooled at a rate of 1 K min<sup>-1</sup>. Temperature uncertainty was ±0.3 K.

### 172 2.7.3 Quantification of Freezing Properties

173 The cumulative concentration of INP present in a volume of solvent,  $V$ , at temperature  $T$ , was derived using the fraction of  
174 frozen droplets ( $f_{ice}(T)$ ), that was obtained directly from the freezing experiments (Vali, 1971):

$$175 K(T) = \frac{-\ln(1-f_{ice}(T))}{V} \quad [cm^{-3} \text{ of water}] \quad (1).$$

176 For control experiments, a quarter of blank filter was immersed in pure water, similarly to freezing experiments of the airborne  
177 samples, and the concentration of the background impurities ( $K_{imp}(T)$ ) were subtracted from the concentrations that were  
178 detected for airborne samples.

179 The atmospheric concentrations of INP per unit volume of air as a function of temperature,  $INP(T)$ , were determined by  
180 incorporating the sampling and solvent parameters into Eq. 2 (Hader et al., 2014):

$$181 \quad INP(T) = \left( K(T) - K_{imp}(T) \right) \frac{V_{solvent}}{f \cdot V_{air}} \quad [L^{-1} \text{ air}] \quad (2),$$

182

183 where  $V_{solvent}$  is the volume of the water used for extraction,  $V_{air}$  is the total sampled air volume, and  $f$  is the fraction of filter  
184 that was used in the extraction.

185 For comparison of ice nucleation activity of the different dust events, the INP concentration in the liquid was converted to the  
186 number of active sites per unit surface area of INPs, i.e., the surface density of sites  $n_s$  active above temperature,  $T$  (Vali,  
187 1971):

$$188 \quad n_s(T) = \frac{-\ln(1-f_{ice}(T))}{A} \quad [m^{-2}] \quad (3),$$

189 where  $A$  is the surface area immersed in a single droplet of the experiment, based on the total surface area of particles in the  
190 suspension.

## 191 2.8 Scanning Electron Microscopy

192 A quarter of selected filters were coated with Iridium for analysing the chemical composition of airborne particles using a  
193 scanning electron microscope (SEM; Supra 55VP, LEO) equipped with an Energy-dispersive X-ray spectroscopy (EDX)  
194 detector for elemental microanalysis. The analysis was done at a voltage of 5 kV using the Quantax software (Bruker).

## 195 3 Results and Discussion

### 196 3.1 Air mass Back Trajectories and the Origin of the Dust Storms

197 The density of air mass back trajectories for 72-h period prior to the sampling for all events are shown in Figure 2. The sampling  
198 site and the surrounding main deserts are shown as well. During the sampled events, the air mass trajectories were diverse. In  
199 some cases, the air masses travelled directly to the sampling site from the source region, while in other cases, they travelled  
200 longer distance. In most events, the air mass had either easterly or westerly component, and were often concentrated in the  
201 same geographical area.

202 The dust origins were identified based on back trajectory analysis, integrated with reanalysis data of remote sensing of  
203 atmospheric dust. We followed the dust mass concentration prior to the sampling period, as detailed in Figure S1. Locations

204 that contained high levels of suspended dust and overlapped with the air mass trajectories were identified as the possible  
205 sources of dust. The green contours in Figure 2 represent the assigned dust origin for each sampled event based on the  
206 reanalysis data. Note that in two events, there was no overlap between the dust origin and air mass trajectories. These events  
207 will be further discussed below. Two events, denoted by SDS1 and SDS2, originated in North Sahara Desert. The source of  
208 SDS1 was near the border of Egypt and Libya, and the source of SDS2 was in Egypt, east of SDS1. The dust travelled over  
209 the Mediterranean Sea and was potentially affected by the marine environment, possibly obtaining a sea salt or anthropogenic  
210 sulfate coating (Levin et al., 1996). Two other events, denoted by SyDS1 and SyDS2, originated from the Syrian Desert, from  
211 western Iraq and southern Syria. Compared to the Saharan events, the dust mass density in the Syrian Desert events was  
212 relatively low.

213 Another event was defined as a “mixed dust” event (MDS), because it was more complicated and included contributions of  
214 different sources: the analysis indicates that there is one possible dust origin east of the sampling site in the Syrian Desert, and  
215 another one southwest of the sampling site in the Sahara Desert. However, the air mass trajectories did not overlap with the  
216 Saharan dust origin, but indicated that the air mass was transported from the Red Sea. Further analysis of the air mass  
217 trajectories prior to the sampling period in the Red Sea showed that both Sahara and Arabia dusts were transported to the Red  
218 Sea (see the supplementary part, Figure S2(a)). Another event did not show overlap between the air mass trajectories and the  
219 dust origin. Further analysis of air mass back trajectories in the days prior to the sampling period showed that dust was  
220 transported to the Mediterranean Sea from the region of Libya in the Sahara Desert, towards Turkey, and was deflected  
221 eastward by westerly winds to the sampling site (see the supplementary part, Figure S2(b)). The dusty air masses rapidly  
222 cleared up, and relatively non-dusty air masses arrived at the sampling site, as inferred from  $PM_{10}$  concentrations and the OPC  
223 size distributions, see section 3.2. This event was defined as “clean and Saharan dust storm” and denoted by CSDS. Table 1  
224 summarizes the sampled events, their sampling periods, and the peak and mean  $PM_{10}$  concentrations during sampling. Peak  
225 values ranged from  $67 \mu g m^{-3}$  in CSDS and  $132 \mu g m^{-3}$  in SyDS1, to  $717 \mu g m^{-3}$  in SDS2, which was the strongest dust event  
226 in this study. In SDS1, MDS and SyDS2, the values ranged between  $\sim 300$  to  $400 \mu g m^{-3}$ . When comparing the mean  $PM_{10}$   
227 concentrations during the entire sampling periods, CSDS was categorized as a non-dusty event, with the lowest concentrations  
228 of  $30 \pm 13 \mu g m^{-3}$ , i.e. below the threshold of  $42 \mu g m^{-3}$  for dusty conditions (Krasnov et al., 2014). The mean values in the rest  
229 of the events ranged from  $76$  to  $206 \mu g m^{-3}$ , and were therefore categorized as dust storms.

### 230 3.2 Particle Number-Size Distributions

231 Figure 3(a) describes the mean particle number-size distributions of sampled air during the dust events, as was detected by the  
232 GRIMM OPC. The lowest channel of the GRIMM includes particles that are larger than  $0.25 \mu m$ . This channel possibly  
233 underestimates the total particle count since the counting efficiency is less than 100%.

234 The number-size distributions had similar patterns in all the events. The highest particle number concentrations were in the  
235 submicron size range, decreasing towards larger particles. Events SDS1, SDS2, and MDS had a rather similar particle  
236 concentration distribution. Event SyDS1 showed similar particle concentrations in the submicron range, but the particle  
237 concentrations in the supermicron range were about an order of magnitude lower, which was also apparent in the PM<sub>10</sub> data.  
238 CSDS, a predominantly non-dusty event, had the lowest particle concentrations in comparison to the rest of the sampled events,  
239 as was also indicated by the PM<sub>10</sub> data. In the SyDS2 event, exceptionally high concentrations in the supermicron range above  
240 3 μm were observed, and the peak extended towards larger particle sizes, combined with relatively high particle concentrations.  
241 Note that prior to and during this event, a series of biomass burning events occurred in Israel extending to about 100 km north  
242 and 50 km east of the sampling site. Therefore, this peak may include also contributions from biomass burning particles. This  
243 is further supported by the SEM-EDX analysis of the filters from this event, which in comparison with the other events,  
244 contained super-aggregates in the supermicron range, typically observed in biomass burning emissions (Chakrabarty et al.,  
245 2014), with distinct morphologies and elemental composition (shown in the supplementary part in Figure S3).

246 The surface-area-size distributions shown in Figure 3(b) compare the contribution of supermicron and submicron particles to  
247 the available ambient surface area. Ice nucleation initiated on the surface of the particles, and therefore, their surface area  
248 concentration is an important parameter in addition to number concentrations. Here it is clearly seen that the potential  
249 contribution of the supermicron particles to the ice nucleation may be significant when compared to the submicron particles,  
250 although their number concentrations were up to two orders of magnitude lower.

### 251 3.3 Airborne INP Concentrations

252 The cumulative INP concentration spectra for the six dust events are shown in Figure 4. In each event, different particle size  
253 classes are marked by different color. Freezing was observed between 255 and 238 K, and the INP concentrations spanned  
254 four orders of magnitude from 10<sup>-1</sup> to 10<sup>3</sup> L<sup>-1</sup> of air.

255 A particle size dependence of the freezing temperature and INP concentration was observed. Larger particles froze at warmer  
256 temperatures with higher number of INPs. The variation between the six size-classes ranged from 1 to 2 orders of magnitude,  
257 and in some cases the smallest particles had similar behavior to the large ones. For example, in event SDS2, size-classes  
258 D<sub>50</sub>=0.6 μm and D<sub>50</sub>=0.3 μm were less ice-active than the rest of the size-classes, while in MDS, all size classes showed similar  
259 activity. As an exception, event SyDS2 showed a weaker size dependence in comparison to the other dust events, and in some  
260 size-classes, lower INP concentrations. In comparison, in the relatively non-dusty event CSDS, the variability between the  
261 different size classes was higher, especially at lower temperatures. In Figure 5, similarly to Figure 4, INP concentrations are  
262 presented, but arranged according to the different size classes. The variability within each size class was relatively high and  
263 spans over 2 orders of magnitude; for example, at size class D<sub>50</sub>=0.3 μm near 245 K, INP concentration ranged from about 1  
264 to almost 10<sup>2</sup> L<sup>-1</sup> of air. It is clearly seen that INP concentrations in dusty conditions (SDS1, SDS2, MDS and SyDS1) were

265 higher than in non-dusty conditions (CSDS) for the supermicron range, but similar in the submicron range. Previous studies  
266 also pointed out the significant contribution of supermicron particles to the INP population. Mason et al. (2016) studied the  
267 immersion freezing abilities of airborne particles in North America and Europe, and found that supermicron particles  
268 dominated the freezing, especially at relatively higher temperature (258 K). Recent measurements in a coastal tropical site  
269 conducted by Ladino et al. (2019) also found high concentrations of INPs at relatively high temperatures ( $> 258$  K) due to  
270 supermicron particles. In these studies, however, mineral dust is not expected to dominate the samples, and bioaerosol particles  
271 are thought to dominate the freezing at the higher temperatures ( $> 258$  K). At lower temperatures (below 253 K), Ladino et al.  
272 (2019) suggested that mineral dust dominated the freezing. Moreover, DeMott et al. (2010) found that INP concentrations are  
273 correlated with particles  $> 0.5 \mu\text{m}$ . Other studies, such as Rosinski et al. (1986) and Huffman et al. (2013), also found that  
274 supermicron particles were responsible for most of the INP population in some cases, while when changing the freezing mode  
275 that was analysed or the measurement meteorological conditions, their contribution was reduced. Vali (1966) in contrast, found  
276 that submicron particles dominate freezing in hail melt samples.

### 277 3.4 Size-Dependence of Ice Active Site Density ( $n_s(T)$ )

278 Figure 6 presents the  $n_s(T)$  curves for the different dust events spanning a range of  $10^6 \text{ m}^{-2}$  at 253K to  $10^{11} \text{ m}^{-2}$  at 238 K. In  
279 general,  $n_s(T)$  increased with particle size. The highest  $n_s$  values were observed in the supermicron range  $D_{50}=5.6 \mu\text{m}$ ,  
280 followed by  $D_{50}=3.2, 1.8$  and  $1.0 \mu\text{m}$ . The activity of the latter three classes was similar within measurement uncertainties. In  
281 the submicron range, stages  $D_{50}=0.6$  and  $0.3 \mu\text{m}$ , the  $n_s(T)$  values were lower than in the supermicron range and showed  
282 higher variability between the different events, except for the MDS event, that had similar activity in the submicron and the  
283 supermicron range. While INP concentrations may generally vary due to experimental parameters, such as particle  
284 concentration in the droplet or droplet size,  $n_s(T)$  accounts for these differences since it is normalized by the total surface area  
285 of particles immersed in the droplet. Therefore, the effect of particle size diminishes using the  $n_s(T)$  curves, if the particles'  
286 ice-nucleation ability is indeed similar. Hence, the analysis presented in Figure 6 indicates that the supermicron particles are  
287 better INP than the submicron ones, implying they have more active sites or/and active sites that nucleate ice at higher  
288 temperatures.

289 Figure 7 displays the same  $n_s(T)$  curves as Figure 6, but now arranged according to the different size-classes. It is observed  
290 that in the supermicron range, all  $n_s(T)$  curves from the different events merge (with the exception of SyDS2) suggesting that  
291 freezing was dominated by a common component. While the freezing activity decreases with decreasing particle size, the  
292 shape of the curves is preserved, suggesting that the abundance of this common component decreases with particle size. One  
293 possible explanation for this observation may be mineralogy segregation, known to occur with particle size: larger particles  
294 contain more primary minerals, such as K-feldspar, whereas smaller particles contain more secondary minerals, such as clays  
295 and quartz that are common in all particle sizes (Perlwitz et al., 2015;Claquin et al., 1999). Therefore, the reduced activity in  
296 the submicron range and the higher variability between the dust events, especially at  $D_{50}=0.3 \mu\text{m}$ , may be attributed to a

297 different mineralogical composition of the particles, or to the lack of the important ice-inducing component. Alternatively, it  
298 is also possible that the submicron particles are mixed with other particle types, that are more common in this size range, such  
299 as urban pollution (Weijun et al., 2016), and therefore freezing may not be dominated exclusively by mineral dust. Moreover,  
300 due to their larger surface-to-volume ratio, submicron particles are more sensitive to atmospheric processing than supermicron  
301 particles, which can lead to further deactivation of their ice active sites (Boose et al., 2016a). These considerations may explain  
302 the variability in the activity between different events. For example, we propose that the passage of SDS1 and SDS2 over the  
303 Mediterranean Sea can contribute to their reduced activity in the submicron range, while for the MDS event, a shorter and  
304 relatively direct transport path resulted in less atmospheric processing. Although speculative, these considerations may  
305 possibly explain why the freezing activity of submicron particles converged with those of the supermicron particles, but we  
306 acknowledge that further measurements are needed to confirm these suggestions.

307 In Figure 7, we also compare a few relevant  $n_s(T)$  curves of standard minerals, as derived by Atkinson et al. (2013) and  
308 Niedermeier et al. (2015), together with our measured  $n_s(T)$  curves. The standard curves of K-, Na/Ca-feldspar and quartz  
309 were scaled to the estimated fraction of these minerals in AMD (see Table S1), and are typically used for prediction of AMD  
310 ice nucleating activity. A good agreement of the absolute  $n_s$  values was observed in the relevant temperature range, and the  
311 slopes of the curves were similar to those of the feldspars, especially for the supermicron range. A good agreement was also  
312 observed with the standard  $n_s(T)$  curve of quartz, suggesting that it contributes to freezing of the submicron particles in the  
313 lower temperature range. Note that the standard  $n_s(T)$  curves of clay minerals and calcite were not plotted here despite their  
314 large abundance in AMD, because there was no overlap with the ice nucleation activity in this study. Only the freezing activity  
315 of the largest particles ( $D_{50}=5.6 \mu\text{m}$ ) overlapped with the K-feldspar prediction of Atkinson et al. (2013), indicating that this  
316 prediction possibly overestimates the freezing activity of the entire size distribution of AMD. For the particles in the size range  
317 of  $3.2 < D_{50} < 1.0 \mu\text{m}$ , there is an overlap in activity with the K-feldspar prediction of Niedermeier et al. (2015) and Na/Ca-  
318 feldspar of Atkinson et al. (2013). However, in all cases, the feldspars predictions overestimate the freezing activity of AMD  
319 in the submicron range.

320 The  $n_s(T)$  curves of SyDS2 displays moderate slopes and lower IN activity in comparison with the other dust events, in all  
321 size classes, except for the smallest particles with  $D_{50}=0.3 \mu\text{m}$ . As was already mentioned, these particles were most likely  
322 mixed with smoke particles from biomass-burning events that occurred during the same period, and the filters from this event  
323 were covered with super-aggregate particles in the supermicron size, rich with potassium, similar to particles seen in other  
324 biomass burning events (Chakrabarty et al., 2014).

### 325 **3.5 Comparison of WISDOM and BINARY measurements for event CSDS**

326 A complementary analysis for the CSDS event using BINARY is shown in Figure 8. BINARY probes droplets with larger  
327 volumes and, thus, it is more sensitive to less common ice-nucleating sites that may not show a signal in WISDOM. In the

328 BINARY experiments, two suspensions were tested, with different dilution factors, for extending our sensitivity. The higher  
329 total dust surface area per droplet sample that was investigated in the BINARY experiments, yellow markers in Figure 8,  
330 demonstrates the warmest freezing temperatures, ranging from 255 to 246 K, and the  $n_s(T)$  values ranged from  $10^6$  to  $10^9$  m<sup>-2</sup>.  
331 The 1:10 diluted samples (purple markers) showed freezing at lower temperatures, ranging from about 251 to 244 K, with  
332 higher  $n_s(T)$  values ranging from  $10^8$  to  $10^{11}$  m<sup>-2</sup>. In some of the dilute cases of the BINARY experiments, the data were at  
333 the limit of the background impurities (see supplementary part, Figure S5). In order to include only data that are significantly  
334 different from the background, a criterion was set, in which only those data points that are larger by at least two standard  
335 deviations than the mean background impurities were further considered in Figure 8. If data were below that threshold, they  
336 were considered as not significant and thus were removed (e.g., the data of the  $D_{50}=0.6$  and  $0.3$   $\mu\text{m}$  for the diluted BINARY  
337 samples).

338 Figure 8 shows a very good agreement between the BINARY and WISDOM data, because the  $n_s(T)$  curves merged nicely  
339 onto each other for each size-class. Whereas BINARY was more sensitive than WISDOM to the warmer and relatively rare  
340 active sites, WISDOM detected the more common active sites in the low temperature range. Overall, the dependence of the  
341 freezing activity temperature range on the immersed surface area per droplet is well demonstrated here, where a reduction in  
342 the surface area of the different experiments (WISDOM < BINARY diluted < BINARY) decreased the probability to observe  
343 freezing at the higher temperatures. This was also demonstrated previously in studies of standard mineral dust (Broadley et  
344 al., 2012; Marcolli et al., 2007; Reicher et al., 2018). Overall, the data shown in Figure 8 indicate the added value when using  
345 experimental techniques of different sensitivity for the purpose of measuring the concentration and active site density of INP  
346 in field studies (e.g., Atkinson et al. (2013); Chen et al. (2018); Harrison et al. (2018)).

347

### 348 3.6 Comparison of Super- and Submicron ranges with AMD Measurements and Predictions

349 The particle surface area that was used to derive  $n_s(T)$  represents the total airborne particles that were collected for each  
350 sample, regardless of particle composition. When mineral dust dominated the composition, as in a dust event case (see for  
351 example Figure S4 in the supplementary part), we treat  $n_s(T)$  as representative for AMD freezing. Figure 9(a) compiles the  
352  $n_s(T)$  results of AMD from a few recent studies that focused on airborne particles (albeit not size-selected) during dust events.  
353 Results from our current study, excluding the events SyDS2 and CSDS that were not dominated by AMD, are presented  
354 alongside those of Price et al. (2018) and Boose et al. (2016b). Price et al. (2018) collected airborne particles in flights west of  
355 the Sahara Desert over the tropical Atlantic at altitudes of up to 3.5 km. Boose et al. (2016b) analysed airborne particles which  
356 were deposited in the Eastern Mediterranean region in Egypt, Cyprus and the Peloponnese (Greece) during dust events. Boose  
357 et al. (2016b) also sampled airborne particles during dust events over Tenerife, off West Africa. In addition, we present  
358 measurements which were also conducted in the Eastern Mediterranean region in Cyprus. Schrod et al. (2017) measured INP

359 in the lower troposphere using an unmanned aircraft system and Gong et al. (2019) measured INP at ground level. Both studies  
360 measured the immersion freezing of the sampled particles during different atmospheric conditions that included few dust  
361 plumes from the Sahara. Note that here we present only immersion/condensation freezing measurements by Schrod et al.  
362 (2017) and not the entire data . Also note that the presented data is not necessarily dominated by mineral dust, in contrast to  
363 the current study or to Price et al. (2018) and Boose et al. (2016b). The specific cases where the samples were taken during  
364 passage of dust plumes and are possibly dominated by mineral dust are marked in Figure 9(a) in green for Schrod et al. (2017)  
365 and cyan for Gong et al. (2019). The supermicron data presented in this paper is about 1 to 2 orders of magnitude higher, while  
366 our submicron data is in relatively good agreement with Schrod et al. (2017), except for the lowest temperature (243 K) points  
367 where 1 to 3 orders of magnitude differences were observed. The Gong et al. (2019) data are lower in 1 to 3 orders of magnitude  
368 but there is some overlapping with this study and with Price et al. (2018).

369 This compilation of the data that was dominated by mineral dust (i.e., this study, Price et al. (2018) and Boose et al. (2016b))  
370 shows that  $n_s(T)$  curves from the different studies exhibit great similarities over a wide range of temperatures (236 - 265 K)  
371 for dust from different locations and geographic sources, with varying atmospheric paths and altitudes. This similarity may  
372 have significant implications for modelling ice nucleation activity by AMD, since it suggests that parameterizations can be  
373 simplified, for example by neglecting the complication of accounting for mineralogy of different geographical sources. Due  
374 to the different behaviour of submicron and supermicron particles, we also suggest that accounting for the particle size class  
375 will improve the prediction of ice cloud formation. For that purpose, we derived two basic parameterizations (Eq.4), for  
376 supermicron and submicron particles, based on the combined AMD data (including data from this study, Price et al. (2018)  
377 and Boose et al. (2016b), and excluding SyDS2), which cover a wide range of temperatures, and spread more than 5 orders of  
378 magnitudes in  $n_s(T)$  values. These parameterizations are the best mathematical fit for a Hill-type equation, which is normally  
379 used for fitting S-shaped data as they are observed in this compilation:

$$380 \quad n_s(T) = \exp[y_0 + a/(b + \exp[(T - 248)/c])] [m^{-2}] \quad (4)$$

381 where the coefficients (95% confidence bounds) for supermicron range particles are set to:

$$382 \quad y_0 = 11.47 \quad (10.97, 11.98), a = 24.00 \quad (22.01, 25.99), b = 1.53 \quad (1.35, 1.70), \text{ and } c = 4.54 \quad (4.06, 5.02),$$

$$383 \quad T \in [236K, 266K] \quad (R^2 = 0.93).$$

384 and for submicron range:

$$385 \quad y_0 = 9.48 \quad (8.19, 10.76), a = 23.00 \quad (20.23, 25.77), b = 1.34 \quad (1.10, 1.57), \text{ and } c = 7.38 \quad (5.84, 8.92),$$

$$386 \quad T \in [238K, 266K] \quad (R^2 = 0.93).$$

387 Parameterizations for each individual size class can be found in Table S2 in the supplementary part.



388 In Figure 9(b), the parameterizations derived here are presented next to the recent parameterizations of ice nucleation of desert  
389 dust by Ullrich et al. (2017) and Niemand et al. (2012). These parameterizations are based predominantly on natural surface-  
390 collected dust samples, but also contained one sample of AMD from Israel, and agrees within an order of magnitude with our  
391 supermicron data in the low-temperature range (243 - 247 K), but overpredicts  $n_s(T)$  by more than an order of magnitude  
392 when compared to our submicron data and to the Price et al. (2018) data at warmer temperatures (247-259 K). This emphasizes  
393 that AMD ice nucleation may not be correctly represented when based on desert dust sampled from the surface, consistent  
394 with the conclusions of Boose et al. (2016b) who showed that the average freezing activity of AMD is reduced when compared  
395 to the activity of surface-collected desert dust. K-feldspar parameterizations by Atkinson et al. (2013) and Niedermeier et al.  
396 (2015) are also shown here, and as mentioned before, overpredicts the freezing activity of AMD at temperatures lower than  
397 about 255 K.

#### 398 4 Conclusions

399 We characterized the INP activity of particles collected during several mineral dust events in the Eastern Mediterranean. Dust  
400 from the Sahara Desert, the major source for atmospheric dust, together with dust from the Arabian and Syrian deserts were  
401 included. Six size classes were studied that cover both the super- and submicron size ranges. The INP concentrations ranged  
402 from  $10^{-1} \text{ L}^{-1}$  of air in the relatively weak dust events to  $10^3 \text{ L}^{-1}$  of air in the strongest event. The  $n_s$  values ranged from  $10^6$  to  
403  $10^{11} \text{ m}^{-2}$  in the temperature range of 238 – 255 K. A size dependence was observed, both in the INP concentration and in  $n_s$   
404 values. Larger particles were more active INP, exhibited higher INP concentrations and a higher number of nucleating sites  
405 per surface area at higher temperatures. Comparison between freezing results of WISDOM with BINARY showed good  
406 agreement, strengthened previous studies that observed how the freezing activity could depend on technical properties and  
407 limitations of the used instrumentation, and therefore emphasize the importance of using complementary instruments.

408 The dust events studied here represent a range of dust loads, different dust origins and atmospheric paths. Yet, the supermicron  
409 particles in these events exhibited similar freezing abilities. This may indicate that there is a unique component that is  
410 responsible for freezing activity, as was previously suggested (Atkinson et al., 2013;Boose et al., 2016b;Kaufmann et al.,  
411 2016;Price et al., 2018). Our measurements showed that the activity of the supermicron particles was in the range of standard  
412 particles of feldspar mineral, and that the activity of the submicron particles was in the range of standard quartz. Therefore,  
413 we suggest that these may be the two most important components that dominate the freezing by atmospheric mineral dust  
414 (AMD), and therefore may be important for heterogeneous ice nucleation in atmospheric clouds. The submicron particles  
415 showed higher variability between events, possibly due to different composition of the particles or higher sensitivity to  
416 atmospheric processing during long-range transport. In general, supermicron particles contributed the most to the INP  
417 concentration, in agreement with other previous studies (Mason et al., 2016;Huffman et al., 2013;Ladino et al., 2019).

418 However, our current study is probably the only case where mineral dust dominated the samples. Nevertheless, all of these  
419 studies highlight the importance of the supermicron size class of AMD for atmospheric ice nucleation.

420 Mineral dust is important both on a regional scale, near its source region, and on a global scale, since it remains ice-active  
421 even after long transport in the atmosphere and thus over considerable distances (DeMott et al., 2003b;Chou et al., 2011). With  
422 the distance from the dust source, supermicron particles will settle, and submicron particles may then dominate ice nucleation  
423 on the global scale (Ryder et al., 2013;Murray et al., 2012). However, recent airborne measurements found coarse and giant  
424 particles in the vicinity and also far from source regions (Ryder et al., 2018). Therefore, including the particle size class in INP  
425 parameterizations can improve predictions of ice formation in clouds. Moreover, information on airborne INP size distributions  
426 may be helpful in identifying the dominant INP sources (Mason et al., 2016). The overprediction of AMD freezing ability  
427 demonstrated in this study, by the Atkinson et al. (2013), Niedermeier et al. (2015);Niemand et al. (2012) and Ullrich et al.  
428 (2017) parameterizations, especially for submicron particles, emphasizes the importance of future studies to better quantify  
429 the changes in the ice-nucleating properties of AMD by atmospheric processing.

430

431

432 **Data availability.** Data are available upon request to the first author.

433

434 **Author contributions.** NR and YR designed the experiments, carried out the field measurements, conducted freezing  
435 experiments in WISDOM, and wrote the paper. CB, LE and TK designed and performed freezing experiments in BINARY.  
436 SRR performed backtrajectory analyses. NR and IKA performed the chemical analyses of filters. All authors contributed to  
437 the discussion and analysis of data and the writing of the manuscript.

438

439 **Competing interests.** The authors declare that they have no conflict of interest.

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446 PM<sub>10</sub> data is available from the Israel Ministry of Environmental Protection website  
447 (<http://www.svivaqaqm.net/Default.rtl.aspx>). Other data used in this study can be retrieved from [osf.io/gpuqt](https://osf.io/gpuqt).

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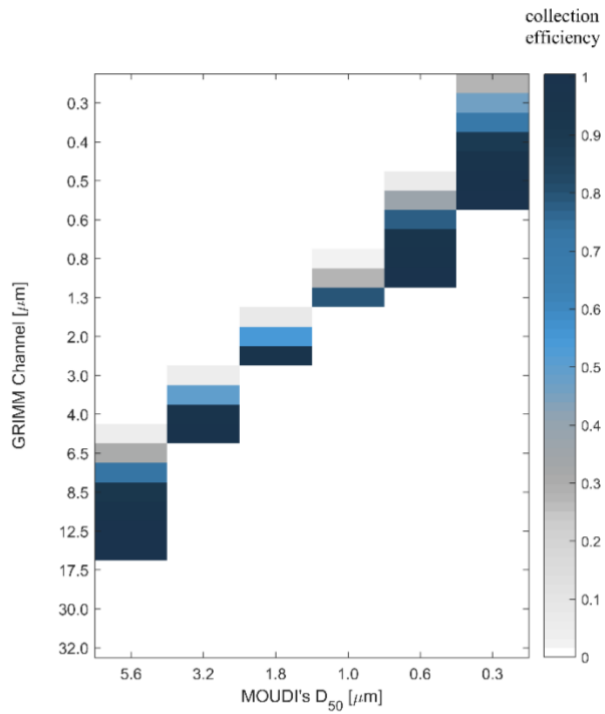
716 **Table 1: Summary of the investigated dust storm events. The events are denoted by their geographic origin: Saharan**  
717 **dust storms (SDS), Syrian dust storm (SyDS), mixed contribution of the two (MDS), and mix of dust event with a free-**  
718 **dust period (CSDS).**



Event	Start [UTC]	Sampling period [hour]	PM10 peak [ $\mu\text{g m}^{-3}$ ]	PM10 mean [ $\mu\text{g m}^{-3}$ ]	Freezing analysis technique
SyDS1 <b>19 April 2016</b>	07:30	25	132	76±20	WISDOM
CSDS <b>27 April 2016</b>	07:30	24	67	30±13	WISDOM, BINARY
SyDS2 <b>23 November 2016</b>	15:30	18	332	184±68	WISDOM
SDS1 <b>09 March 2017</b>	11:00	48	387	96±66	WISDOM
SDS2 <b>12 March 2017</b>	12:00	24	717	206±120	WISDOM
MDS <b>12 April 2017</b>	13:30	25	409	141±106	WISDOM

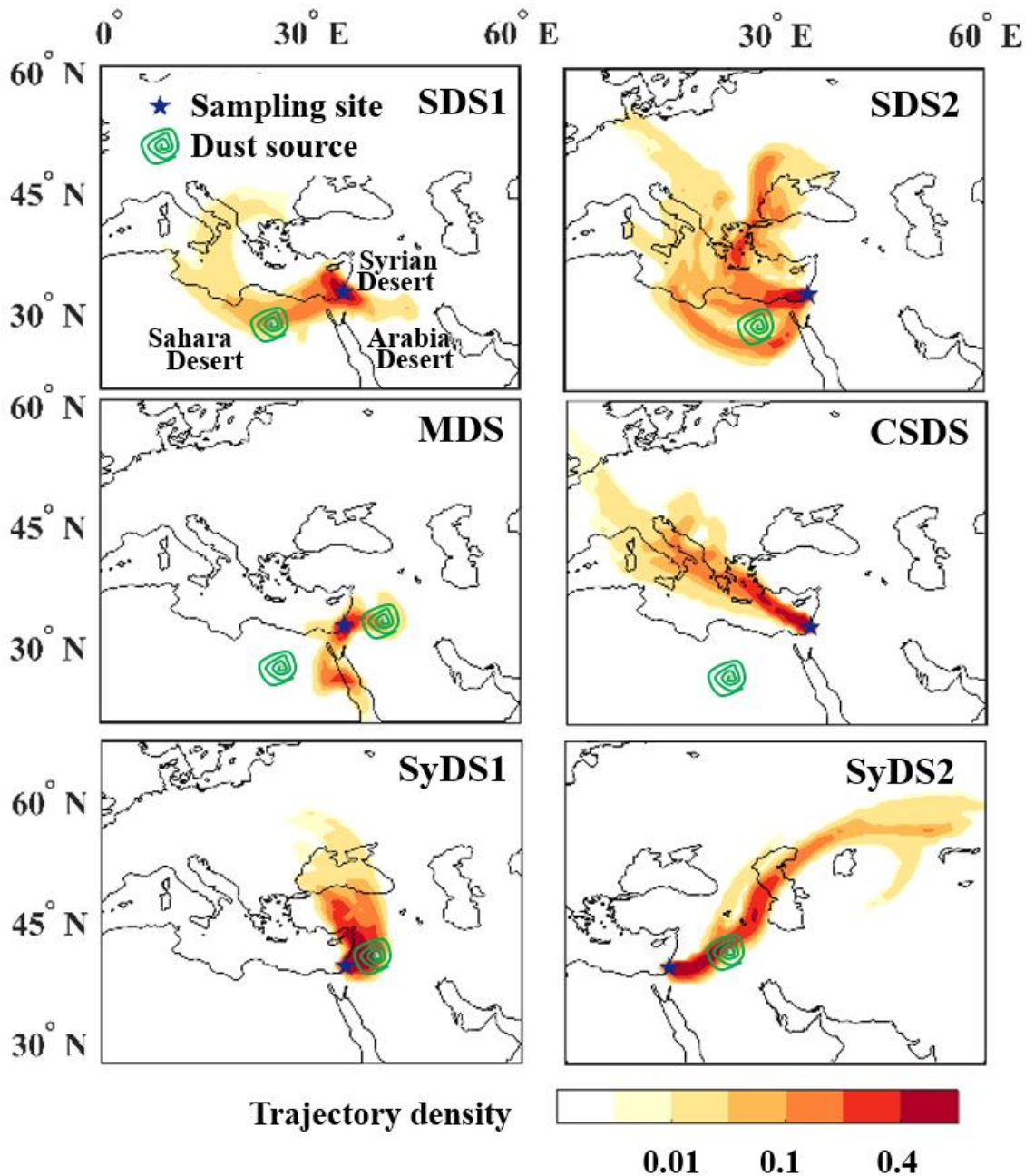
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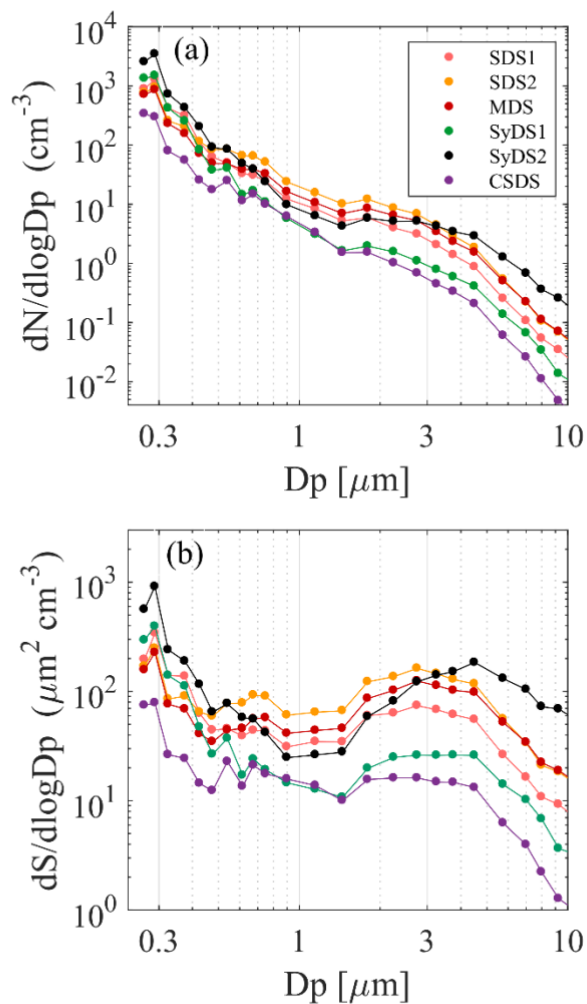
722 **Figure 1: A conversion matrix of GRIMM channels to MOUDI stages. The conversion was based on collection efficiency**  
 723 **curves from Marple et al. (1991). The color shades represent the fraction of particles of a specific GRIMM channel**  
 724 **to be impacted on a specific MOUDI stage.**



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726 **Figure 2: Air mass origin and atmospheric paths of the dust events. Colors represent the density of 72-h backward air**  
 727 **mass trajectories (normalized to the total trajectory counts). The green contours represent the geographic locations**  
 728 **where a high mass of the dust occurred during the air mass transition, which defined as the potential origin of the dust.**  
 729 **Abbreviations in the top right of each panel indicate the particular dust event.**

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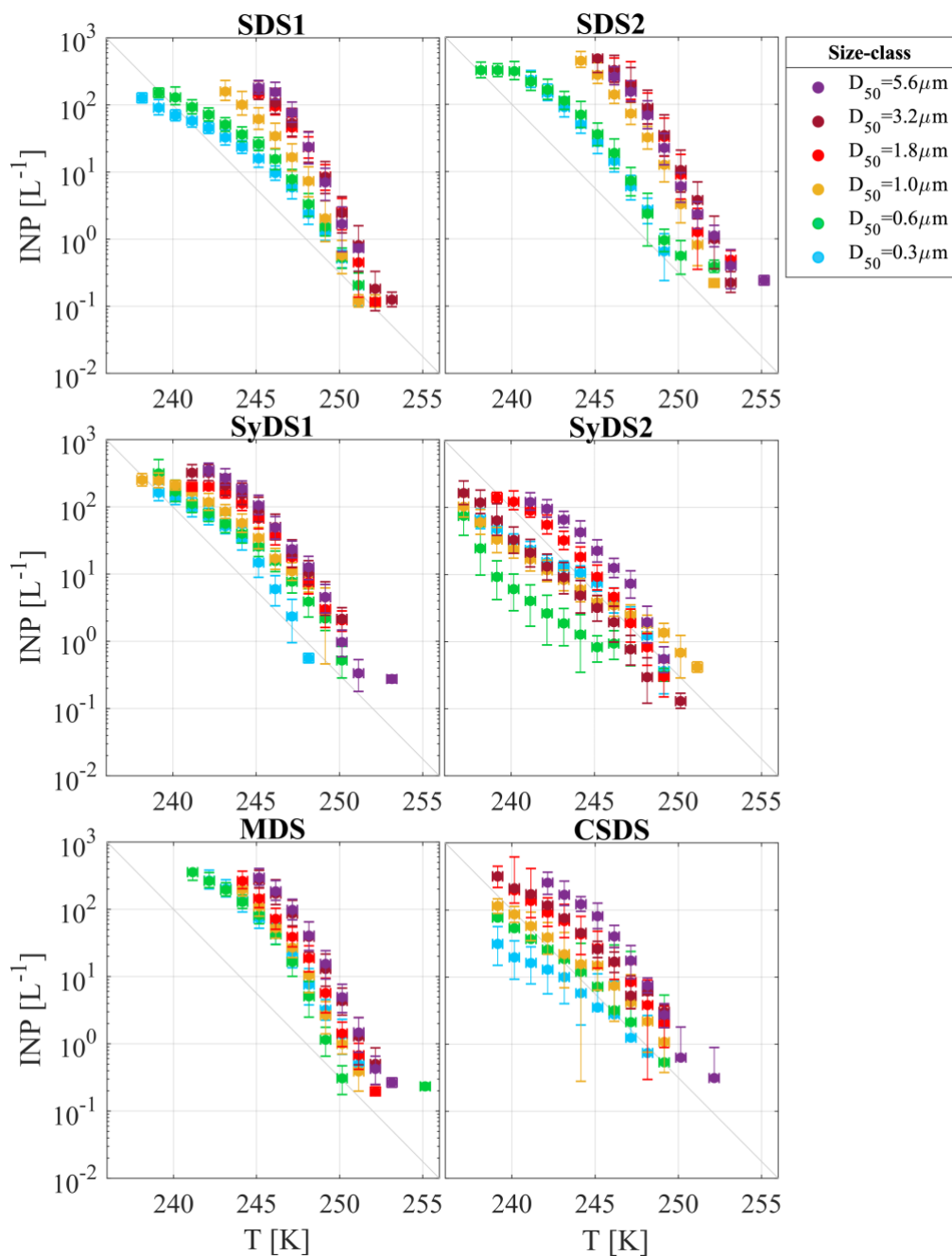
733 **Figure 3: Particle size distributions. Particle number size (a) and surface area size (b) distributions averaged over the**  
 734 **entire sampling periods of the events as monitored by GRIMM OPC during the studied events.  $D_p$  is the diameter of**  
 735 **the particles and set at the center of each GRIMM channel.**

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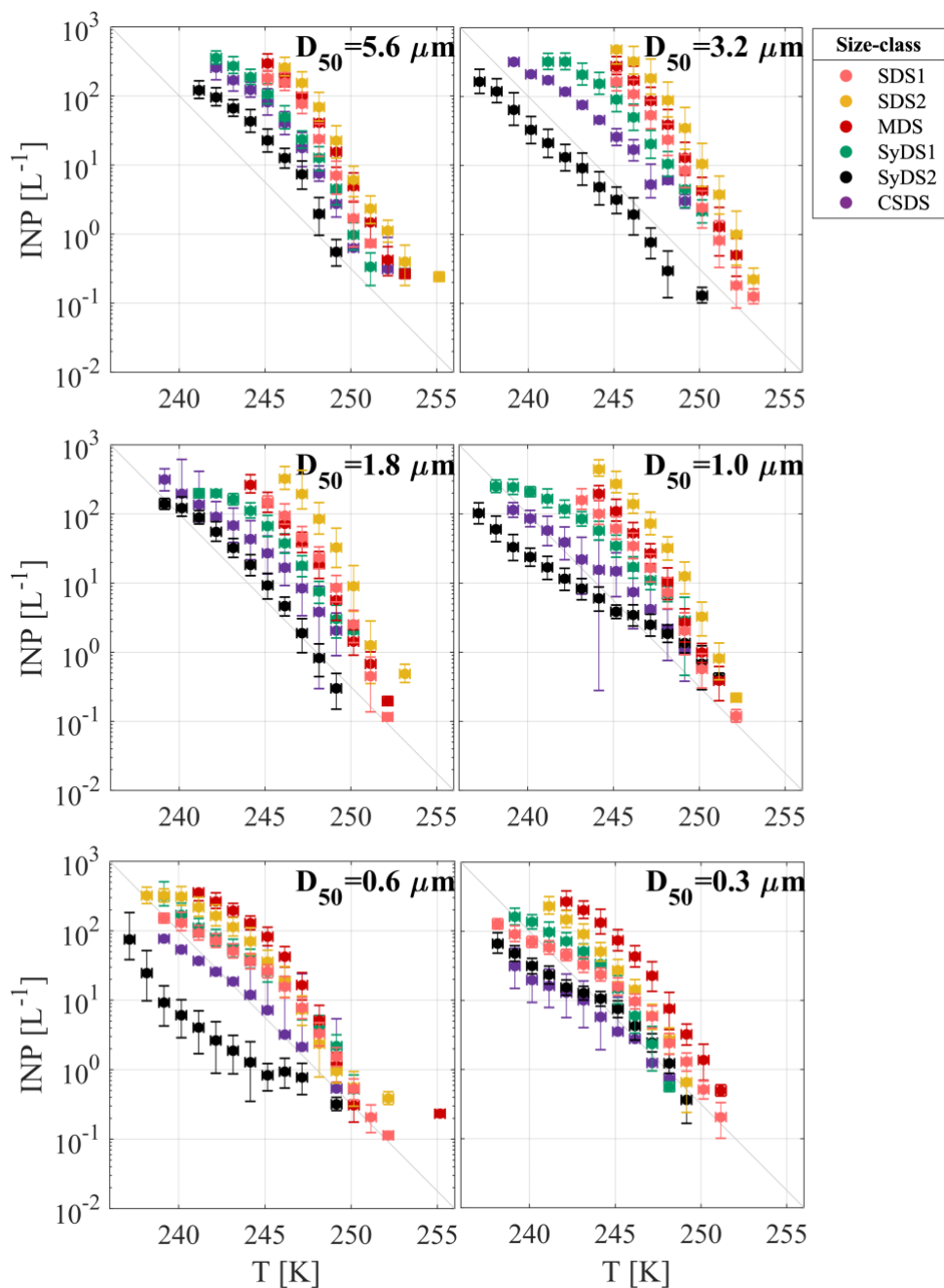
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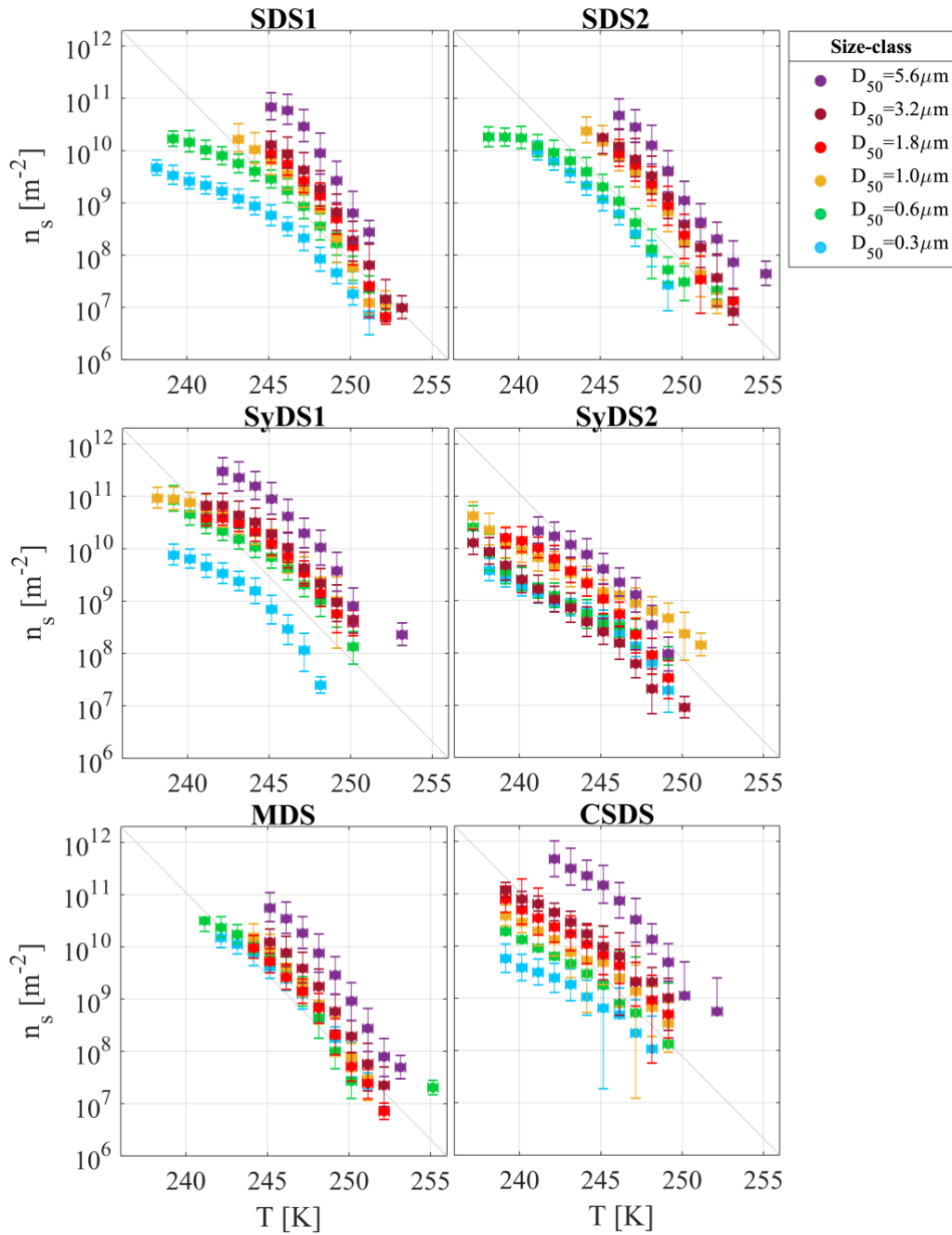
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742 **Figure 4: Airborne INP concentrations measured during dust events. INP concentrations per  $\text{L}^{-1}$  air as function of**  
 743 **temperature, presented in different colors for the different particle size-classes. Uncertainty in temperature is 0.3 K.**  
 744 **The grey diagonal line is presented for orientation only.**



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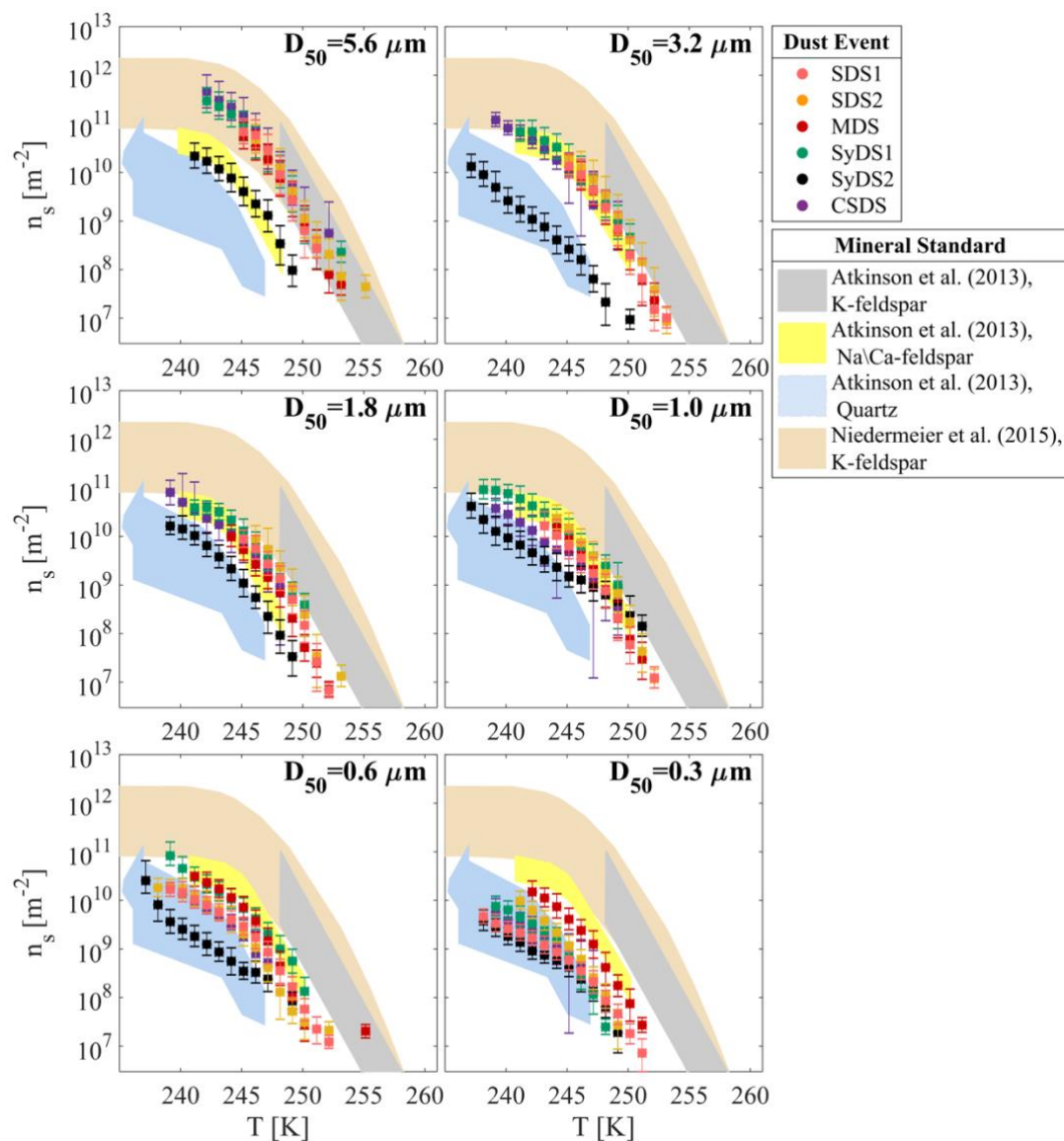
747 **Figure 5: Airborne INP concentrations for various size classes. INP concentrations per L<sup>-1</sup> air as function of**  
 748 **temperature, presented in different colors for the different dust events that were sampled. Uncertainty in temperature**  
 749 **is 0.3 K. The grey diagonal line is presented for orientation only.**



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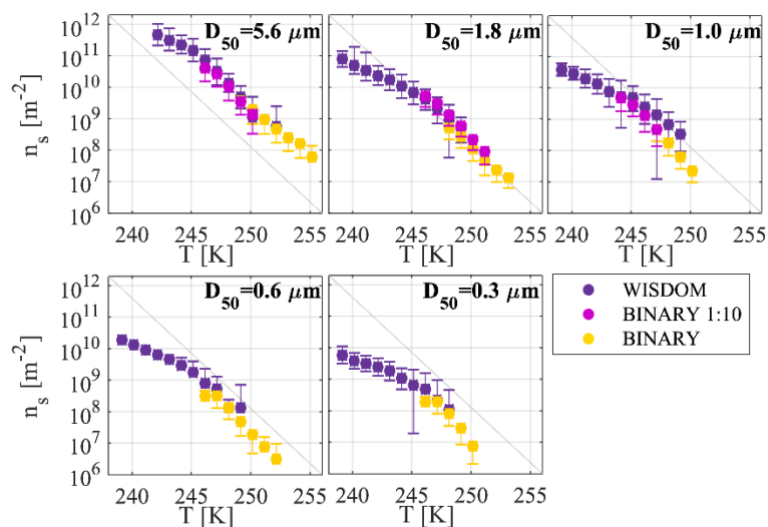
751 **Figure 6: Ice active site density as a function of temperature,  $n_s(T)$ , for airborne particles dominated by mineral dust**  
 752 **are presented individually for each dust event. The different colors represent the different size-classes that were**  
 753 **investigated. SDS, SyDS and MDS represent Saharan, Syrian, and mixed dust events, respectively (see text for more**  
 754 **details). The linear grey line is identical in each panel to facilitate comparison.**

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759 **Figure 7: Ice active site density during dust events in different particle size classes. Dust events from the Sahara Desert**  
 760 **(SDS), Syrian Desert (SyDS), or both (MDS) are marked by the different colors. Data for  $D_{50} = 3.2, 1.8$  and  $1.0 \mu\text{m}$  of**  
 761 **SDS#2 adopted from Reicher et al. (2018). Relevant standard minerals scaled to ambient values are shown: K-feldspar,**  
 762 **Na/Ca-feldspar, and quartz from Atkinson et al. (2013), and K-feldspar from Niedermeier et al. (2015).**





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766 **Figure 8: Complementary measurements of WISDOM and BINARY for CSDS. Analysis in the BINARY was**  
 767 **performed to increased detection sensitivity of ice active site densities. Two suspension with different dilution factors**  
 768 **were analysed by BINARY and are compared here to the WISDOM data for the different size-classes.**

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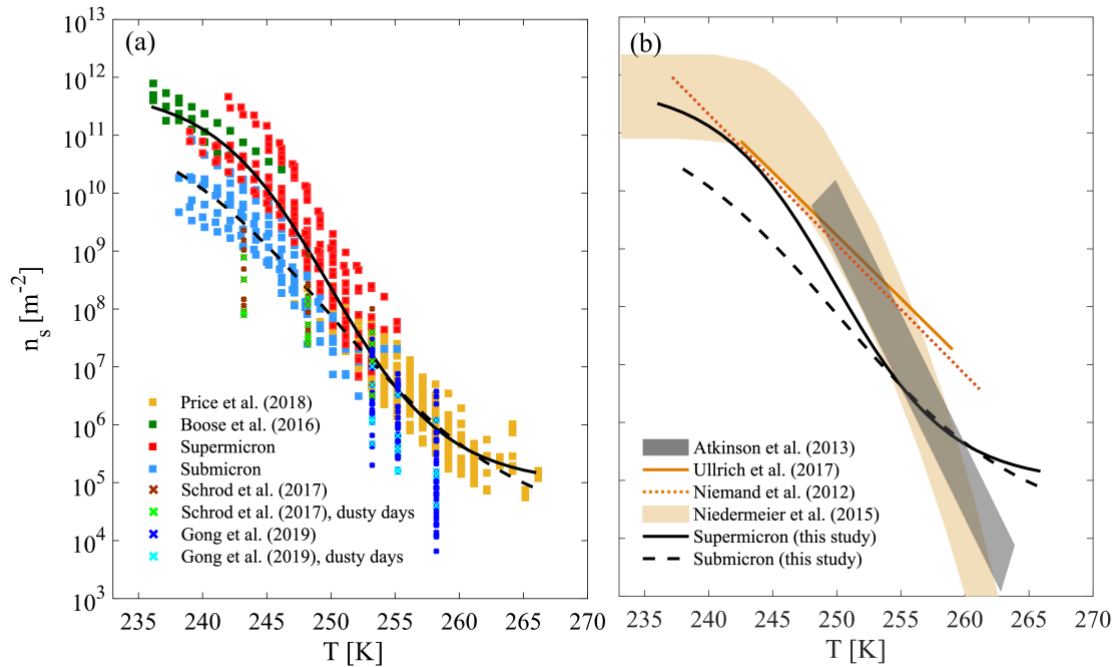
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 777 **Figure 9: Heterogeneous ice nucleation by airborne particles during dusty conditions. (a) Active site densities of**  
 778 **supermicron and submicron size-classes from this study are shown together with flight data (Schrod et al. (2017) and**  
 779 **Price et al. (2018)) and deposited or *in-situ* data (Boose et al. (2016b) and Gong et al. (2019)). New parameterizations,**  
 780 **which were derived in this work based on the combined AMD data of the different studies, are shown for supermicron**  
 781 **and submicron classes. (b) The new parameterizations derived in this study based on all AMD data, shown next to**  
 782 **recent parameterizations for desert dust (Ullrich et al. (2017) and Niemand et al. (2012)) and K-feldspar predictions**  
 783 **(Atkinson et al. (2013) and Niedermeier et al. (2015)).**  
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