Response to Anonymous Referee #1

First of all, we want to thank the referee for submitting his/her helpful and productive annotations, which led to improvements and clarifications within the manuscript.

We have prepared a revised manuscript that addresses the questions and comments of the referees. Furthermore, below we explicitly respond to each of the items raised in the comments of anonymous referee #1. These comments are indicated by using *italics,* whereas the author's response is presented in blue. Changes in the manuscript are given in green; changes to the supplement are given in purple. The differences are also highlighted in separate PDFs using latexdiff. All line and page numbers refer to the ACPD manuscript version, not the revised manuscript.

Review of "Ice nucleating particles of the Eastern Mediterranean measured by unmanned aircraft systems" by Schrod et al.

In this paper, Schrod et al. leverage the use unmanned aircraft systems to measure the abundance of ice nucleating particles which are active in the immersion and condensation modes in the lower troposphere for the first time. The study is conducted in a region frequently influenced by Saharan dust emissions, making the results of particular importance to furthering our understandings of how desert dusts can impact upon clouds. During the study, a number of long-range transported dust events are captured, and the importance of dust as an INP in this environment is highlighted by correlation to PM10 mass, aerosol optical thickness and modelled dust concentrations.
The paper itself is well-written, and the work presented is both novel and likely to be of large interest to researchers interested in this topic. Even of the few remarks I have on the manuscript, most of these are relatively minor. As such, I recommend the paper for publication after consideration of the following:

Comments:

Pg. 1, ln. 5: It should be noted that here, and during all the other occurrences throughout the paper, that the plural of ice nucleating particle is "INPs" and not "INP"
We thank the referee for noticing and have implemented the plural as suggested.

Pg. 6, ln. 18: What is the efficiency of the sampling system (i.e. inlet + aerosol sampling unit) for different particle sizes?
We will answer this comment below, add this discussion to the supplement and add a remark to the manuscript.

> Pg.6, ln.18 now reads:
>
> Both UAS were equipped with a customized inlet ~~system~~ nozzle that was connected by tubing to ~~an~~ the aerosol sampling unit. The diameter D_s of the sample inlet nozzle was such that near isokinetic sampling was achieved at the average air speed U_0 of the UAS and sampling rate Q. The error due to anisokinetic sampling was estimated to be

typically less than 20% of particle number for particles up to 10 µm in diameter. A detailed discussion of sampling errors due to anisokinetic sampling is presented in the supplement.

The following paragraph was added to the supplement:

2 Estimate of errors from anisokinetic sampling

The diameter D_s of the sample inlet nozzle was such that near isokinetic sampling (i.e. the air sample inlet velocity U equals the air speed U_0 of the UAS) was achieved for the average operational air speed U_0 of the UAS and the mean aerosol sampling rate Q. The three quantities U_0, D_s and Q are related by Eq. 1:

$$D_s = 2\sqrt{\frac{Q}{\pi U_0}} \tag{1}$$

For the mean operational conditions these parameters are: a) for Cruiser: U_0 = 27.8 m s^{-1}, Q = 5 lpm, D_s = 1.95 mm; and b) for Skywalker: U_0 = 16.7 m s^{-1}, Q = 5 lpm, D_s = 2.52 mm.

In the following we estimate sampling errors due to anisokinetic conditions (i.e. $U \neq U_0$). The latter may arise when the UAS spirals in the wind field and air speed U_0 and pump rate Q deviate from conditions a) or b) due to tail wind or head wind. The range of U_0 and Q observed during the campaign is given in Tab. S1. All parameters vary typically by less than 20%. Our estimate follows the discussion of sampling errors in Hinds (1999), chapter 10. For simplicity's sake and for the lack of other measurements, we will use the assumption that the gas streamlines entering the sampling inlet show no misalignment whatsoever. For this idealized case the ratio of the aerosol number concentration C at $U \neq U_0$ to C_0 at isokinetic conditions is then given by Belyaev and Levin (1974):

$$\frac{C}{C_0} = 1 + \left(\frac{U_0}{U} - 1\right)\left(1 - \frac{1}{1 + (2 + 0.62\,U/U_0)\,Stk}\right), \tag{2}$$

with the Stokes number Stk being defined by

$$Stk = \frac{\tau\,U_0}{D_s}, \tag{3}$$

and the relaxation time τ being

$$\tau = \frac{\rho_p\,d^2\,C_C}{18\eta}. \tag{4}$$

Here ρ_p is the particle density (estimated for dust as 2.6 g cm^{-3}), d is the aerosol diameter, C_c is the Cunningham correction factor and η is the viscosity of the air.

Figure S2 and S3 present C/C_0 as calculated by Eq. 2 as function of particle size for the mean, maximum and minimum Q and U_0 occurring during flights of Cruiser (Fig. S2) and Skywalker (Fig. S3) at an altitude of 2000 m. The maximum error is negligible for particles below 1 µm, and grows with increasing particle size up to around ± 30% for particles of 10 µm in diameter for Cruiser and up to ± 60% for Skywalker.

Table S1: Variation of sample flow Q and airspeed U_0 during the campaign.

	Skywalker	Cruiser
Q_{mean} [lpm]	4.91	
Q_{min} [lpm]	4.37	
Q_{max} [lpm]	5.56	
$U_{0,mean}$ [m s^{-1}]	17.4	28.1
$U_{0,min}$ [m s^{-1}]	14.3	23.4
$U_{0,max}$ [m s^{-1}]	23.5	33.3

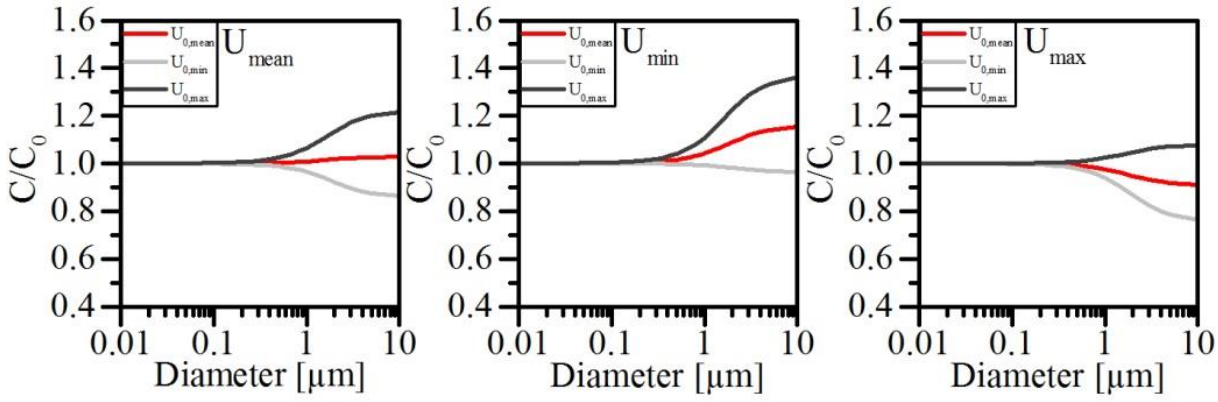

Figure S2: Aerosol number concentration ratio due to anisokinetic sampling effects as a function of particle diameter for Cruiser.

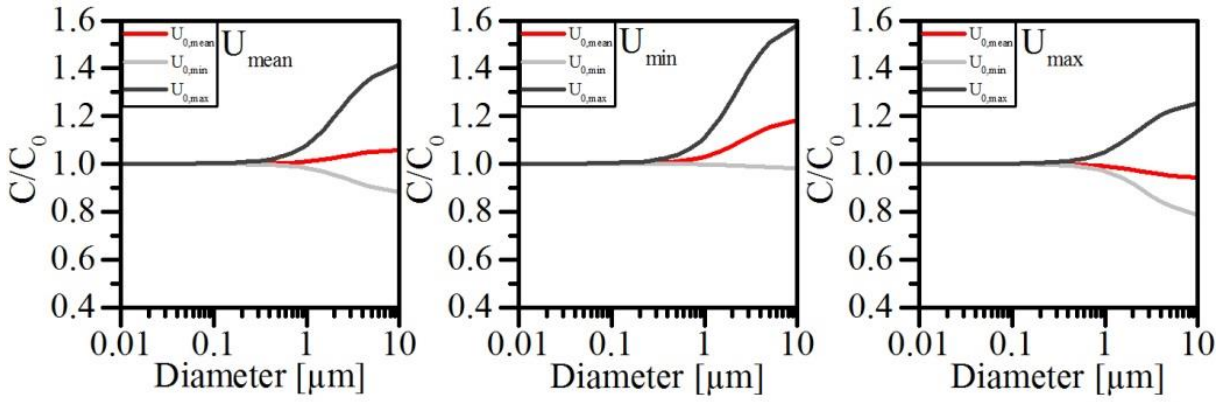

Figure S3: Aerosol number concentration ratio due to anisokinetic sampling effects as a function of particle diameter for Skywalker.

Pg. 7, ln. 8-9: I think a very brief discussion of the limitations and possible caveats of the FRIDGEs measurement principles is pertinent here. While these are listed in Schrod (2016), a brief summary here would also be useful, as these are of course also central to this work.
We added a brief summary of the limitations of the measurement principle at the end of section 2.4.

Pg. 7, ln. 8-9 now read:

For a detailed description of the sampling procedure and FRIDGE's measurement principle as well as its limitations and possible caveats, see Schrod et al. (2016). These limitations include for example a) the possible loss of volatile aerosol constituents due to the analysis under medium vacuum, b) the possibility of a transient depletion of water vapor above the nucleating particles due to the uptake of water occurring at very high numbers of particles on the substrate, and c) technical restrictions regarding the method's time resolution. Although our measurements can cover the freezing induced by nuclei that are immersed in droplets after condensation (i.e. condensation freezing), they do not involve freezing of macroscopic droplets with immersed INPs.

Pg.11, ln19: A brief mention as to what the physical meaning of the calibration factor, cf, is would be useful to the reader here.
The calibration factor *cf* introduced by DeMott et al. (2015) has no deeper underlying physical meaning, but is just a fit parameter that refers to special instrumental conditions applied in the CFDC, i.e. RH_w = 105%. In this manuscript, we use *cf* simply as a mathematical degree of freedom when fitting the observed measurements to the predicted INP concentrations.

We will add this paragraph to the manuscript.

Pg.11, ln.19 now reads:

The parameters α = 0, β = 1.25, γ = 0.46 and δ = −11.6 were empirically fitted by DeMott et al. (2015). ~~The calibration factor *cf* has been introduced to separately account for instrument-specific calibration and was set by default to *cf* = 1, but in special cases it shows a better fit when using cf = 3.~~ The calibration factor *cf* has no deeper underlying physical meaning. DeMott et al. (2015) state that the constants α, β, γ, and δ could have captured this coefficient, but they wanted to introduce it separately to account for instrument specific calibration of their CFDC. More precisely, they have found when measuring mineral dust aerosol in the AIDA cloud expansion chamber at RH_{water} = 105 % a factor of 3 lower INP concentrations than the maximum concentration shortly before droplet breakthrough was observed in the CFDC (usually at RH_{water} = 108 - 109 %). Therefore, they argue that a pre-factor of *cf* = 3 is needed to obtain the maximum immersion freezing INP concentration for mineral dust specific atmospheric data.
In this manuscript the calibration factor *cf* is handled completely independent of this definition. Instead, we use it simply as a mathematical degree of freedom when fitting the observed measurements to the predicted INP concentrations.

Pg. 12, ln. 26-29: Can you show this good agreement by using a plot?
See next answer.

Section 3.2.2.: As you have ns values, it might be interesting to compare your results to the lab-based parameterisation for the ice nucleating activity of mineral dusts developed by Niemand et al. (2012) and maybe even to that for feldspar by Atkinson et al. (2013), taking into account that only a fraction of the dusts are likely to be feldspar.

We will add a plot (Fig. S5) in the supplement that compares the measurements of this study to the data of Boose et al. (2016) and the parameterizations of Niemand et al. (2012) (hereafter: N12) and Atkinson et al. (2013) (hereafter: A13).

Our data agree well to the field data of Boose et al. (2016), but are lower than the lab data. The data agree acceptably with N12, where the highest measured n_s is usually within about one order of magnitude of N12 or better. However, the K-Feldspar parameterization A13 does not match the observed slope. Especially for cold temperatures the data diverge from A13 by several orders of magnitude. As the referee pointed out, probably only a fraction of atmospheric dust particles is composed of this highly ice active material, so we believe this to be a difficult comparison to make. It is noteworthy that these two parameterizations are purely dependent on temperature, whereas we find our data to be dependent on RH_{ice}.

We will add a text passage to the manuscript.

Pg.12, ln.24 and following now read:

The values of AF and n_s compare reasonably well with ~~published~~ measurements performed in ~~regions~~ atmospheric environments influenced by mineral dust. E.g. Boose et al. (2016) found the active site density ~~of~~ from two month measurement data ~~in the summers of 2013 and 2014~~ at Izaña in Tenerife to range between 7×10^7 – 3×10^8 m^{-2} at T = −25 °C, RH_{ice} = 130% (this study: 2×10^7 – 7×10^8 m^{-2} at T = −25 °C, RH_{ice} = 129%, Fig. 5a in the supplement) and between 2×10^8 – 2×10^9 m^{-2} at T = −33 °C, RH_{ice} = 135% (this study: 7×10^7 – 2×10^9 m^{-2} at T = −30 °C, RH_{ice} = 135%, Fig. S5b in the supplement). Figure S5 also compares the observed active site densities with the laboratory based mineral dust immersion freezing parameterizations of Niemand et al. (2012) (hereafter: N12) and Atkinson et al. (2013) (hereafter: A13). Both parameterizations predict higher active site densities than were found in this study. The data agree acceptably to N12, with the highest measured n_s being usually within the same order of magnitude as N12, or better. However, the K-Feldspar parameterization A13 does not match the observed slope. Especially for cold temperatures the data diverge from A13 by several orders of magnitude. As probably only a fraction of dust particles was composed of this highly ice active material, we did not expect a good agreement.

The following will be added to the supplement:

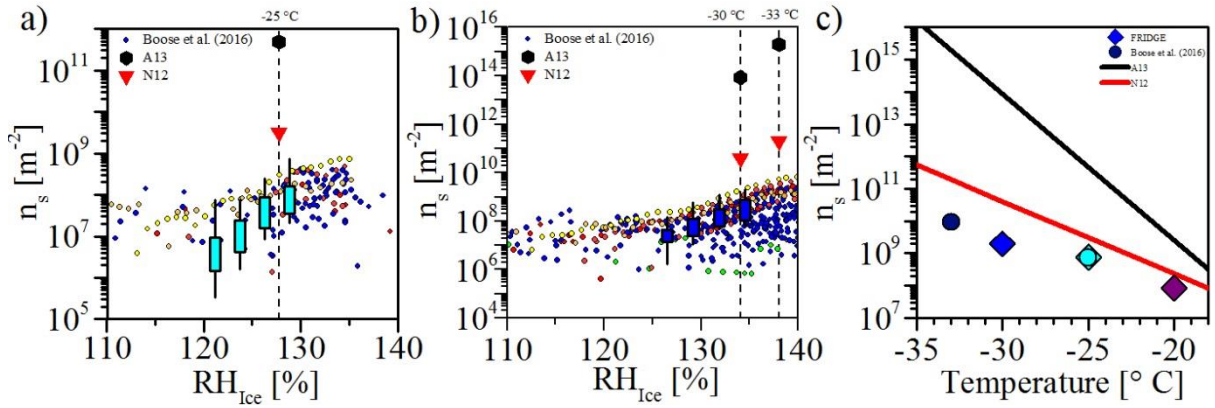

Figure S5: IN active site densities measured in the atmosphere and on mineral dust test aerosols. This study (boxes and diamonds); Boose et al. (2016) (circles); N12 (triangles and red line); A13 (hexagons and black line); dashed lines: water saturation at stated temperature.

Pg.15, ln. 1: It should be noted here that Conen et al. examined the ice nucleating activities of dusts at much warmer temperatures than were probed here. That being said, a similar point is made by Tobo et al. (2014), which might be good to reference here.

We added the temperature range in the sentence about Conen et al. (2011) and included a reference to Tobo et al. (2014).

Pg.15, ln. 1 now reads:

For example, Conen et al. (2011) ~~discussed~~ concluded from soil dust measurements in the range of −4 °C to −15 °C that the carbon content/biological residues within dust samples can define their ice nucleation properties. Similarly, Tobo et al. (2014) underlined the significance of organic matter in soil dusts as INPs in mixed-phase clouds at temperatures warmer than −36 °C.

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