Anonymous Referee #4

This paper shows how satellite observations can be associated with aerosol model reanalysis to infer the impact of dust on cloud thermodynamic phase transition. The authors used LIDAR and RADAR measurements from the A-train to retrieve information on the cloud thermodynamic phase using different products (DARDAR, GOCCP) and the reanalysis MACC to co-locate dust mixing ratio and updraft. Therefore, the study retrieves the frequency phase ratio as a function of dust mixing ratio constrained for different regimes of latitude, temperature, season, etc. The aerosol-cloud interaction problem is a difficult subject to study with observations because aerosols and clouds properties cannot be spatially and temporally co-located. The use of satellite and reanalysis circumvents the problem. The results show that the cloud ice fraction increases with the concentration of dust, suggesting that dust plumes contain ice nuclei which is line with previous studies as mentioned by the authors. Each effects are quantified, the manuscript is well written and well structured in my opinion. The objectives of the study are clearly mentioned in the introduction and the results associated with their significances are stated in the conclusion. The topic of this study matches very well with the journal topics and is of great interest for the atmosphere community, in particular people studying aerosol-cloud interaction. However, I consider that the article needs important revision that I estimate necessary to be published: the discussion on meteorological parameter impacts is too short, the data section is too short... (see below for detailed descriptions).

We thank Anonymous Referee #4 for the new insights and ideas. We have carefully reviewed all concerns and made some new analysis of the data. We believe that the overall quality of the manuscript has been now greatly improved.

Major revisions:

1.a Meteorological parameters have a larger impact on cloud properties than aerosols (Gryspeerdt et al., 2016). Different meteorological regimes can change the aerosol-cloud interaction by an order of magnitude. Even if you mention in your paper the meteorological parameters (sections 4 and 5), it is missing in the paper. You refer to humidity, but the stability is also an important parameter in the aerosol-cloud interaction. The spatial resolution of ERA can seem coarse but it could constrain your situation and could avoid any correlation you are referring to (line. 400): You might not have the atmospheric state at the cloud but it refers to general atmospheric processes which are important as well.

We agree that stability is a useful parameter for separating between aerosol-cloud interaction regimes. However, Figure #4.1a shows that there is no significant difference in the day-to-day correlation between dust and ice occurrence for different stability regimes (defined as "unstable", "neutral" and "stable"). Nor were the day-to-day changes in stability associated with changes in ice occurrence. For the analysis we used the lower-tropospheric static stability (LTSS) defined in Klein and Hartmann, 1993 and following Li et al. 2017.

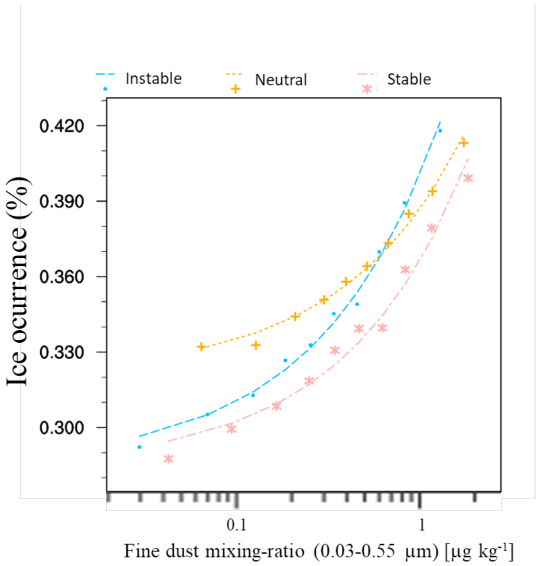
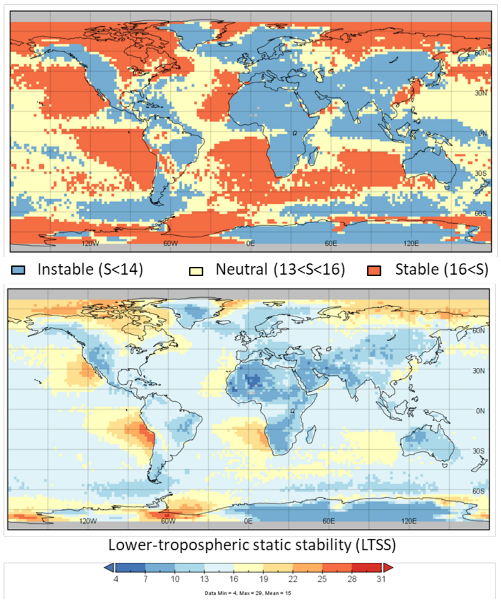
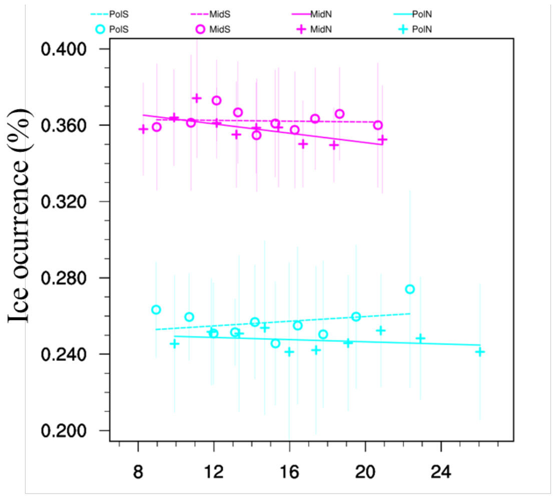


Figure #4. 1.a : FPR_{GOCCP} vs dust mixing-ratio for different stability regimes.

LTSS = T_{700}[1000/p700]^(R/C_p) – T_{sfc}[1000/p_{sfc}]^(R/C_p)





Lower-tropospheric static stability (LTSS)

1.b Also, the boxes you considered based on latitudes-longitudes contain both land and ocean which are in different regimes of aerosols and meteorological parameters, I would like to see a differentiation between land and ocean.

Figure #4.1b shows this differentiation. The shift between the curves results mostly from the differences in dust mixing-ratio between sea and land. We note that we find no significant changes in cloud occurrence between land and sea for temperatures ranging from -10 °C to -40 °C. This partly contradicts the results from Tan et al. 2014.

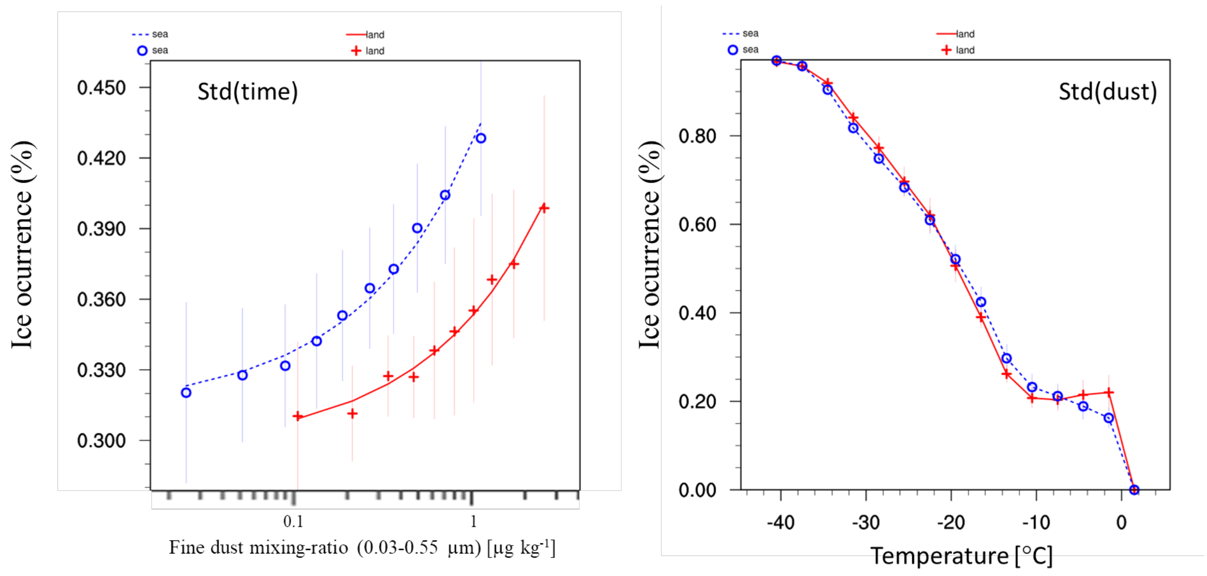


Figure #4. 1.b Sea-land

1.c Moreover, you based your study on dust aerosols, but other parameters can have an impact on the ice fraction (soot, sea salt, sulphate...), the low correlation you observe in the hemisphere north could also be due to the fact that there are more different aerosol types which can act as IN as well.

We agree that INP other than dust may have a significant impact on cloud ice occurrence. However, the impact of other type of aerosols including black carbon, organic matter, sulphate and sea salt was excluded in this paper to focus on the constrain of mineral dust.

See also answer to Anonymous Referee #3: *"Studying other INPs such as black carbon would be indeed interesting. Our first screening showed that the correlation is much lower for black carbon at -30°C (see Figure #3.5). Moreover, we believe that a thoroughly comparison between all potential INP (organic, black carbon, sea spray, ...) would greatly overextend the length of the manuscript and would require a regional scope. "*

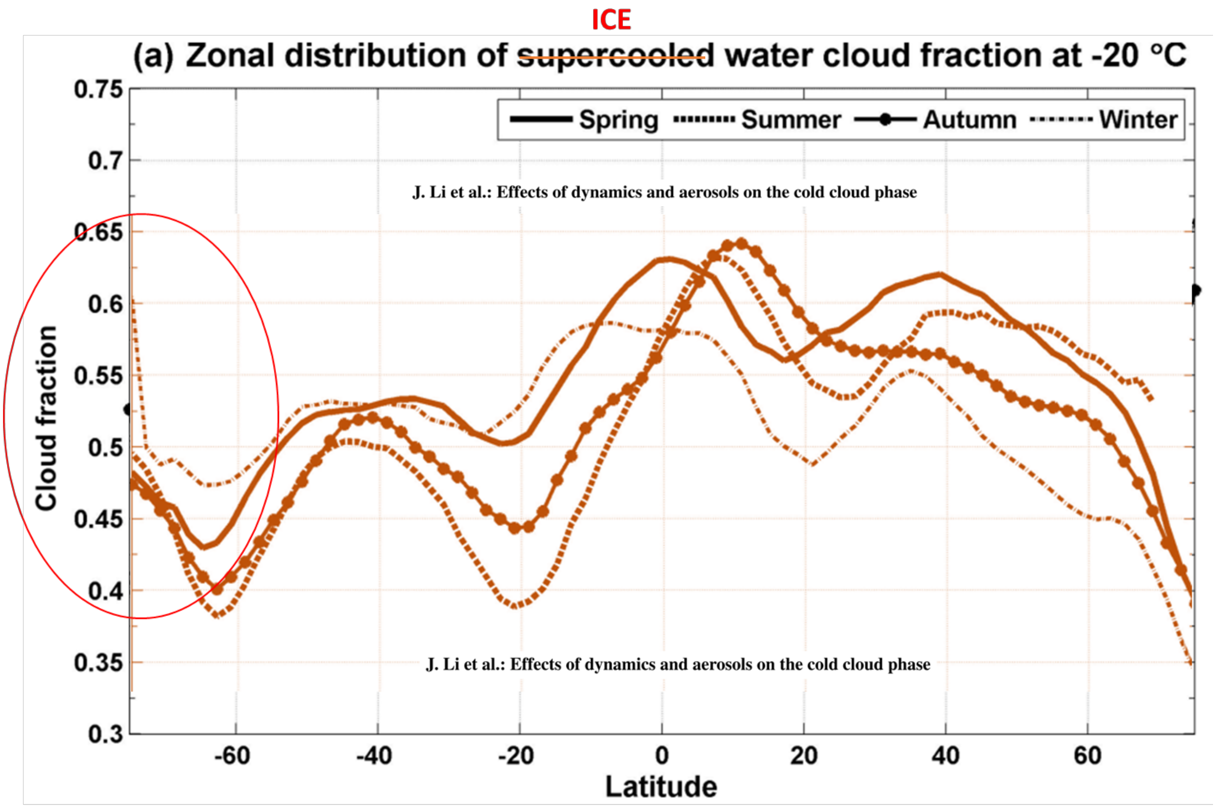
2.a You refer to the maximum number of points you can retrieve in line 126-127, but the actual number of points never appears in the article, I suggest to add the number of data points in the Figures 6, 7, 8 for each point. The work is based on statistical analysis so the number of points is important, especially if you compare different regimes.

We agree that sample size is a very important parameter in our statistical approach. The number of data points in Figures 6, 7 for each dust deciles have been already included in supplement S15. However, to also include this data in the manuscript would inevitably raise questions about the distribution of the sample size along the other dimension (season, latitude, longitude, temperature). We believe that focusing on the co-variability between dust and ice occurrence is fundamental for maintaining an adequate length of the paper. Nevertheless, figures S14, S15 and S16 in the supplement were intended to provide a complete view on the data sample size. Moreover, the contribution from each latitude to the sample size of the 30° latitude bands is also shown here.

2.b I am particularly concerned by Figure 4-b and the increase of ice occurrence for latitudes lower than 73S (maybe the results are not statistically significant because you do not have enough data points).

For temperatures between 0°C and -20°C, the gridboxes between 70°S and 80°S contain in average less than 25 days of data per gridbox (See supplement S14). This is indeed a small sample size and it results from both a lower cloud frequency in the Antarctica as well as surface temperatures below -15°C. As mentioned in the text, we suggest that this also results in the larger standard deviation below 73S in Figure 4.b. However, previous studies suggest that this result is significant despite the low sample size. Indeed, a comparable increase of ice occurrence for latitudes lower than 73S has been also reported (at -20°C) in the study of J. Li et al. 2017 using eight years of the night-time GOCCP dataset for all clouds. Furthermore, the predominance of ice clouds in the Antarctica has been already pointed out earlier in the literature (Ardon-Dryer et al., 2011; Bromwich et al., 2012). Additionally, this increase in ice occurrence can be observed at difference seasons and for different dust mixing-ratios (See attached figure #3.4). Therefore, we do not expect that the higher ice occurrence below 73S would result in a bias for the day-to-day variability analysis.

See also response to Anonymous Referee #3 *“The predominance of ice clouds in the Antarctica has been already pointed out earlier in the literature (Ardon-Dryer et al., 2011; Bromwich et al., 2012). Incoming air masses from the ocean may carry higher concentrations of INP like biogenic aerosol (Saxena, 1983), Patagonian soil dust or Australian black carbon (Bromwich et al., 2012). Specially immersion freezing INP have been shown to be significant in the Antarctica (Ardon-Dryer et al., 2011; Bromwich et al., 2012). Similarly, it has been shown that the orographic forcing in the Antarctica can lead to high ice water contents (IWC) for maritime air intrusions (Scott and Lubin, 2016). In other words, maritime air intrusions associated to higher temperatures, higher concentrations of INP and stronger vertical motions could explain the observed pattern in the southern polar regions. "*



Modified (inverted y-axis) from J. Li et al. 2017 (Fig 4)

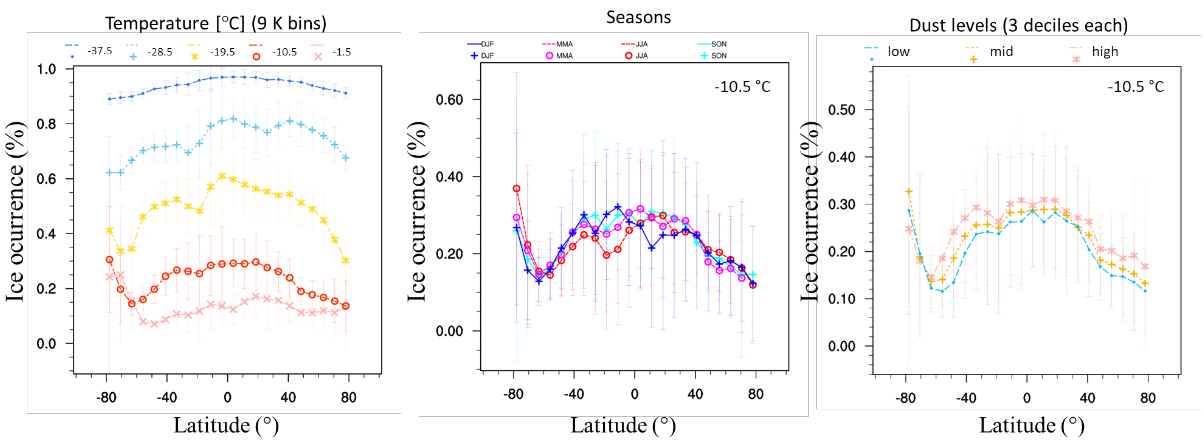


Figure #3. 4 : Zonal mean ice occurrence FPR_{GOCCP} for different temperature ranges, seasons and dust levels (latitude width = 7.5°).

2.c Also I am concern by Equation 3.6: If I understood, you average for each dust mixing ratio bin to have constant number of 10 data points. This method artificially increases the correlation coefficient.

The adjusted correlation coefficient is a way to account for small data samples (it is therefore always lower than the unadjusted correlation). We agree that the use of aggregated values increases the correlation with respect to the raw dataset. However, the intention of the correlation coefficient is not to study the co-variability between dust and ice occurrence (for this we use the normalized covariance in Sect. 4.2.). Instead, the sole purpose of equation 3.6 is to give a measure of the "goodness of the fit" as summarized in Table 1.

Could you measure the Pearson correlation based on the 2-D histograms. For example, in Figure 6-a, what would the correlation coefficient be if you consider all the couples (iceOccurence-FineDustMixingRation) without averaging for Fine dustmixing-ration bins first.

The variable iceOccurrence in the raw data is mostly binary (0 or 1, "U" shaped PDF, see FIG. 5-14 in Korolev et al. 2017). Only after aggregating the data does the variable behave close to normal. Furthermore, the aim of the section is to estimate the day-to-day correlation between dust and ice occurrence. Including all (iceOccurence-FineDustMixingRation) pairs would include spatial and seasonal variability and miss the goal of Section 4.4.

To circumvent the problem, averaging the ice occurrence trough one or more dimensions results in a (more or less) normal-distributed variable. The population Pearson correlation coefficient is defined as COV(m,FPR)/(STD(m)*STD(FPR)) which is equal to $\overline{COV}/STD(FPR)$ (see equation 3.2). Then, the result of calculating the Pearson correlation for all pairs FPR-dust in the space time-longitude (12*192 points) is equivalent to the values plotted on Fig 5.b divided by $STD(FPR)$.

We believe $\overline{COV}$ to be a much more adequate measure of the correlation between ice occurrence and dust mixing-ratio for this dataset. This is mainly because the limited satellite measurements result in binary-like values in regions of low cloud cover which in turn results in a much larger STD(FPR).

3. The data section lacks necessary information. The satellite needs to be described more precisely with information about the performance of the algorithms:
When they are compared to in-situ or ground-based measurements, how do they perform?

The authors are not aware of any proper evaluations of the cloud-phase algorithms with in-situ measurements. Only comparisons with other satellite retrieved products are available (Huang et al., 2012; 2015).

Nonetheless, some additional specifications of the algorithms have been now added in the manuscript.

What are the methods to derived cloud properties?

The cloud properties (RH and updraft velocity) in the MACC reanalysis are derived from the ECMWF Integrated forecast system (IFS Cycle 36r1 4D-Var). The atmospheric simulations and data assimilation are analogous to the ERA Interim reanalysis (Dee et al., 2011).

References: Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, L., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J.-J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N. and Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system, Q. J. R. Meteorol. Soc., 137(656), 553–597, doi:10.1002/qj.828, 2011.

This has been added to the data section.

The same goes for MACC, in line 120 you refer to "good results", can you develop and quantify.

Changed to*: "… showing a mean bias of 25% between MACC and LIVAS (dust product based on CALIPSO satellite) over Europe, northern Africa and Middle East. Additionally, the correlation between MACC and AERONET (network of ground-based remote sensing stations) was found in the range of 0.6 over the Sahara and Sahel to 0.8 over dust transport regions."*

4. You plotted the uncertainties in your figure but you do not refer to them in your text. For example in line 293, you use "notably higher", but if you consider the uncertainties in the figure, the difference is not that high.
Can you comment on that?

We do not find any clear pattern in the uncertainties worth mentioning other than in lines 353-354 referring to the day-to-day variability on different seasons.

We agree that the use of "notably higher" may be misleading. However, the difference between both points is higher than the sum of the respective standard deviations (We realize that the visualization of the error bars for this example is rather unclear). We have changed line 293 to:

"For the 60-90°S latitude band, the difference between the FPR_{GOCCP} at a dust mixing-ratio of 0.01 and 0.1 $\mu g\ kg^{-1}$ is almost 10 %."

We realize that the standard deviations are high relative to the changes in ice occurrence. However, this is to be expected given that the day-to-day changes in dust loading are not expected to constrain every aspect of the ice occurrence variability. Uncertainties in the measurements, retrieval algorithms and in the reanalysis are not the only source of error.

Furthermore, many factors may contribute to the high standard deviation for the ice occurrence assessed in Sect. 4.5 and 4.6, including:

a) Changes in dynamical forcing
b) Temperature changes post-glaciation
c) Ice sedimentation from above
d) Temperature of the cloud within the 12 K interval
e) Turbulence (favouring mixing and temperature fluctuations)
f) Differences in dust composition and INPs other than dust

We have now added this list in Sect. 5

Minor revisions:
- Introduction: There is plenty of different methods to study the aerosol-cloud interactions. The method you are using present fair advantages. A paragraph is needed to highlight this.
We have now further developed line 70:

"This method provides a new approach to study the link between dust and cloud phase variability. Its main advantage compared to previous studies is the ability to estimate aerosol concentrations at cloud level, which is otherwise a very difficult task using common remote sensing techniques. An additional advantage is the estimation of very low aerosol concentrations and its temporal variability, which may often lay below the lower detection limits of common remote sensing retrievals."

- The use of "e.g." needs a coma, example in line 28: (e.g., Patagonia, South Africa, and Australia)

Corrected

- Method section: Are clouds vertically co-located with the dust mixing ratio?

Both FPR and mixing-ratio are co-located to the same temperature bins.

- Figure 1 is not described, you need to introduce it and describe it.

Now properly introduced. "*Fig. 2 shows the zonal sum of the sample size for the FPRGOCCP at -15 °C and -30 °C. Each count corresponds to a month-decile pair.*"

- Figure 8-a and 8-b are not presented, can you describe them before referencing to them?

Now properly introduced. "*Fig. 8 shows the mean RH, cloud height and large-scale updraft at -15 °C for the different fine dust-mixing ratio deciles and for the four latitude bands studied in Sect. 4.5.* "

- Line 179: Why do you use 30 degree?

Is there any specific reason?

Latitude bands of 30° allow a direct comparison with previous studies (Zhang et al., 2018). Additionally, 30° is a natural choice for 1.875°x1.875° grids (16-fold).

Did you try with boxes of 20 degree for example?

We did try 15° degree. Smaller boxes still show the same trend but the standard deviation of the points is larger and the fit of the linear regression is worse.

You refer to "optimize the number of different satellite swaths", I do not understand, can you develop?

This max be misleading, we have changed it to "increase the sample size at each gridbox".

- Equation 3.1: Has it been used in a previous study?

No, it has been not.

See also the answer to Anonymous Referee #1:"… FPR* (Equation 3.1) is only used in Sect 4.1 as stated in text. FPR* is used exclusively to ease the visualization of the thermodynamic phase in the case study (to show only clouds with significant cover).

This has been now clarified in the methods section."

- Line 226: "2,5" should be "2.5"

Corrected

- Line 278: How do you explain that you have a larger correlation in the southern hemisphere compare to the northern hemisphere? Can you speculate?

We speculate in lines 382-284 that other type of INP like biogenic or background free-tropospheric aerosol may be correlated or internally mixed with low concentrations of mineral dust in the southern hemisphere. Differences in dust composition or different correlations with atmospheric dynamics may also play a role in the differences between northern and southern hemisphere.

These speculations are already mentioned in the manuscript, especially in Sect. 5.

- Line 309 - 315: This paragraph is not clear, can you reformulate?

Line 309 - 315: “The higher surface area concentration proportional to $m^{2/3}$ could then explain the higher occurrence of ice clouds (Atkinson et al., 2013; Niedermeier et al., 2015). We note, however, that more evidence is needed in order to support such a hypothesis. The regression coefficients shown in Table 1 can be interpreted as the sensibility to mineral dust surface area concentrations. For instance, although in Fig. 6a the slope at 30-60°S is similar to the slope at 30-60°N, the regression coefficient *A* in Table 1 is more than twice as high for 30-60°S. This is due to the lower dust mixing-ratio in 30-60°S. In comparison, the coefficients for 30-60°S and 60-90°N are much more similar. This in turn results from the similar dust mixing-ratio in these latitude bands.”

- Line 345: "In contrast ..." : For the other cases where you have a lower correlation, can it mean that the glaciation happened before, and therefore you do not find a good correlation but dust plumes still contain IN, can you comment on that?

If by “happened before” time is meant, then it is probably not the case. Most cases are either post-glaciated or liquid (because of the WBF process) at both temperature ranges.

However, if by “before” temperature is meant, this would be indeed plausible. Nonetheless, if the lower correlations are a result of already glaciated clouds, then we would expect the correlation to be lower for higher ice occurrences. In Fig. 7a the opposite is shown. The regions with the largest ice occurrence (northern hemisphere) have the higher correlation.

- Line 437: you mention in the paper that you are substituting the m2/3 to do a linear regression, but not in the conclusion, so it is confusing when you refer to m2/3 as linear. Can you clarify this in the conclusion?
Clarified.

- Figure 2 caption: "— are reclassified a ice", I think you mean liquid.
We thank Anonymous referee #4 for finding this mistake. Now corrected.

- Figure 2: You put arrows on the colorbars but it cannot be greater than 100% ice, or 100% liquid.
The arrows have been now omitted.

- Table1: It took me a while to understand Table 1, there is a lot of information, and some of them are never mentioned in the text. Can you simplify it? I have the feeling that it could actually be two different tables.
We have omitted the columns “FPR_0” and divided the table in two for simplicity. See figure #4.5.

Temperature: -15 °C Band	Variable	*FPR=A m^2/3+FPR_0 / m in [µg/kg]* Fine (0.03-0.55 µm) A	r^2_a	Coarse (0.9-20 µm) A	r^2_a
60-90S	FPR_ALT DARDAR	0.39	**0.93**	0.64	**0.87**
	FPR GOCCP	0.39	**0.99**	0.65	**0.93**
60-90N	FPR_ALT DARDAR	0.13	**0.98**	0.73	**0.94**
	FPR GOCCP	0.10	**0.93**	0.51	**0.91**
30-60S	FPR_ALT DARDAR	0.08	0.92	0.25	0.92
	FPR GOCCP	0.10	**0.99**	0.21	0.89
30-60N	FPR_ALT DARDAR	0.03	**0.91**	0.14	**0.93**
	FPR GOCCP	0.04	**0.96**	0.21	**0.95**
Temperature: -30 °C					
30-60N	FPR_ALT DARDAR	-	0.00	0.19	**0.95**
	FPR GOCCP	0.03	0.89	0.11	0.59

Table 1. Summary of the linear regression results for cloud ice occurrence as a function of dust mixing-ratio. A is the regression coefficient, FPR_0 is the y intercept and r^2_a is the adjusted square of the Pearson correlation coefficient. Bolded values of r^2_a represent the regressions for which r^2_a is higher for $x=m^{2/3}$ than for $x=m$,with m the dust mixing-ratio. For -30° C, only two regressions where significant ($r^2_a>0.8$). Underlined values represent the mean between the regression coefficients for each latitude band. The darker green colours correspond to the higher correlation coefficient and the darker blue colours correspond to the higher regression coefficient (A).

Band	$\frac{\overline{COV}_{fine,GOCCP}}{\overline{COV}_{coarse,GOCCP}}$
Temperature: -15 °C	
60-90S	1.4
60-90N	1.3
30-60S	2.1
30-60N	1.1
Temperature: -30 °C	
30-60N	0.9

Table 2. Ratio between the normalized covariance $\overline{COV}$ between FPR_{GOCCP} and the dust-mixing ratio from the MACC reanalysis m_y (with y=fine, coarse).

References: Gryspeerdt, E., Quaas, J., & Bellouin, N. (2016). Constraining the aerosol influence on cloud fraction. Journal of Geophysical Research: Atmospheres, 121(7), 3566-3583. Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1074, 2018.