Anonymous Referee #3

General comments: The manuscript presents an interesting study of mineral dust impacts on stratiform cloud glaciation occurrence using satellite remote sensing measurements and MACC aerosol reanalysis daily product. Mineral dust is one of the major atmospheric aerosols and are also efficient INPs. However, it is still challenging to quantify their impacts on ice nucleation in clouds and cloud thermodynamic phase partitioning. Their findings in this study confirm that mineral dust has positive correlations with ice cloud occurrence on a global scale even after limiting other impact factors. The approach presented in this study is very unique. Personally, I like the approach and also am very happy with their findings. However, many of their discussions are hard to explain the observed features (sometimes contradict with previous studies). Besides, the manuscript is not well organized and poorly written (especially the comparisons between FPRDARDAR and FPRGOCCP) and therefore is hard to read. Overall, I believe serious revisions are needed before the manuscript be accepted by ACP.

We thank Anonymous Referee #3 for his constructive comments. We realize that the discussion about our Figures was lacking some important information. We have addressed all points and believe that a significant improvement has been made.

Major comments:

Fig1.:
1) About the sample size, what causes the missing datapoints?

There are various sources of missing data:

a) The satellite swaths (orbits) produce different density of data at different latitudes.
b) Using only night-time data the sample size in the meteorological summer time (shorter nights) is lower.
c) The cloud phase data is (for obvious reasons) less frequently available in areas and heights (temperatures) of low cloud cover.
d) Certain temperature ranges near the poles can lay below the surface temperature and therefore no information is available for such temperatures (specially in the Antarctica).

These points have been now included in the Methods section.

See also Supplement S14 for an in-depth view of the distribution of the sample size across seasons, temperature and regions.

2) Why do the high-latitudes have less datapoints than the mid-latitudes?
See last question.

3) Is the sample size same or similar for DARDAR and MACC?
The MACC reanalysis dataset does not have missing values as it is produced from a model output. The DARDAR product uses only satellite swaths were both CALIPSO and CLOUDSAT satellite retrievals are available. Additionally, in both datasets we have omitted pixels where the CALIOP signal is fully attenuated. Therefore, the DARDAR sample size is equivalent to the GOCCP excepting downtimes of the CPR (the most relevant being 07/12/2009 – 16/01/2010) and other minor exceptions.

Fig 2. and related discussion:
1) According to line 143-144 and line 153, “lidar-not-fully-attenuated” and “only ice pixels are considered as ice clouds”, I would expect basically DARDAR and GOCCP have very similar FPR. For example, in Fig 2., DARDAR detected the ice virga below the liquid layer top. However, generally, the liquid layer at the top will fully attenuate the lidar signal (also as stated in line 119). Therefore, given the filters listed in line 143-144, I do not understand why DARDAR still show an ice layer below;

The clouds between -40°N and -45°N and between -6°C and -33°C are classified as “altostratus” by the 2B-CLDCLASS product. In most cases, such stratiform clouds are thin enough to be penetrated by the lidar.

FPR_GOCCP: The detected ice virgae below the liquid cloud top suggests the cloud top did not fully attenuate the lidar signal (cloud not thick enough). The number and/or size of the ice particles near the cloud top probably was not enough to increase the depolarization ratio above the threshold value for the GOCCP algorithm, therefore was classified as liquid.

FPR_DARDAR: In the decision tree of the DARDAR algorithm there are multiple alternatives for a mixture of cloud droplets and ice particles (e.g. at cloud top) to be classified as ice only (Mioche et al., 2014):

a) If the lidar backscatter signal (β) is lower than 2.10^{-5} m^{-1} sr^{-1}
b) If not “a)”: If it is “weekly attenuated” (less than 10 times) or “not rapidly attenuated” (at a depth larger than 480 m).
c) If not “b)”: If the vertical thickness of the cloud is larger than 300 m (equivalent to 5 pixels with a lidar resolution of 60 m).

Therefore, there are plenty cases where a mixed-phase cloud (and specially optically thin stratiform clouds) can be mis-classified as ice only in the DARDAR product and consequently in the FPR_DARDAR variable. In this specific case, we speculate that “c)” is the most probable cause because of the large vertical extension of the cloud around (1 to 3km using a lapse rate of -10°C/km for the estimation).

FPR_ALT_DARDAR: In the case of droplets and ice particles coexisting at cloud top, we expect that at some location of the swath, the cloud droplets will be enough in number to be classified as liquid (strong attenuation) by the DARDAR algorithm. If this is the case, the entire gridbox value of FPR_ALT_DARDAR will be LIQUID (interpreted as a non-completely glaciated cloud).

Further explanation: The GOCCP algorithm is unable to detect ice in mixed-phase clouds and the DARDAR algorithm tends to classify mixed-phase clouds as ice. Therefore, we avoid using the “frequency of cloud ice” (FPR) to compare the GOCCP and DARDAR products. Instead, we use a parameter which considers the limitations of both products. In FPR_ALT_{DARDAR}, mixed-phase clouds that would be otherwise classified as ice are now classified as liquid. This “recreates” the inability of the GOCCP algorithm to detect ice in mixed-phase clouds. Therefore, the “frequency of completely glaciated clouds” (FPR_ALT) allows a comparison between both algorithms (FPR_ALT_{DARDAR} and FPR_{GOCCP}).

We have added this explanation to the Sect. 4.1.

2) Comparing 2a, b, d, e, and f there are not clouds at latitudes between -40 and -45 and temperatures between -6 and 3, there shows no cloud in 2a, b, and e, but does have FPRDARDAR and FPRGOCCP in 2d and f, what's the reason for the differences?

In this gridbox, there is (erroneously) a small cloud fraction corresponding to a cumulus nimbus cloud (See figure #3.2).

The FPR_ALT$_{DARDAR}$ is 50% (50/50 ice and liquid) which is plotted white in 2.e. This is the result of the satellite orbit passing two adjacent gridboxes at the same latitude. One of this gridboxes was classified as liquid and the other as ice.

The FPR plots shown in Figure 2 were taken directly from the raw data and equivocally included the phase and cloud fraction of non-stratiform clouds (which were thought negligible for the case study).

We apologize for the mistake. This has been now corrected.

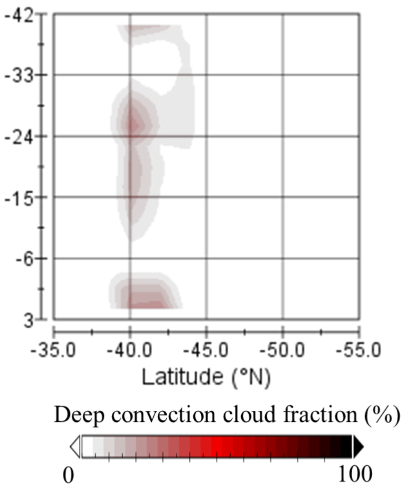


Figure #3. 2 Case study

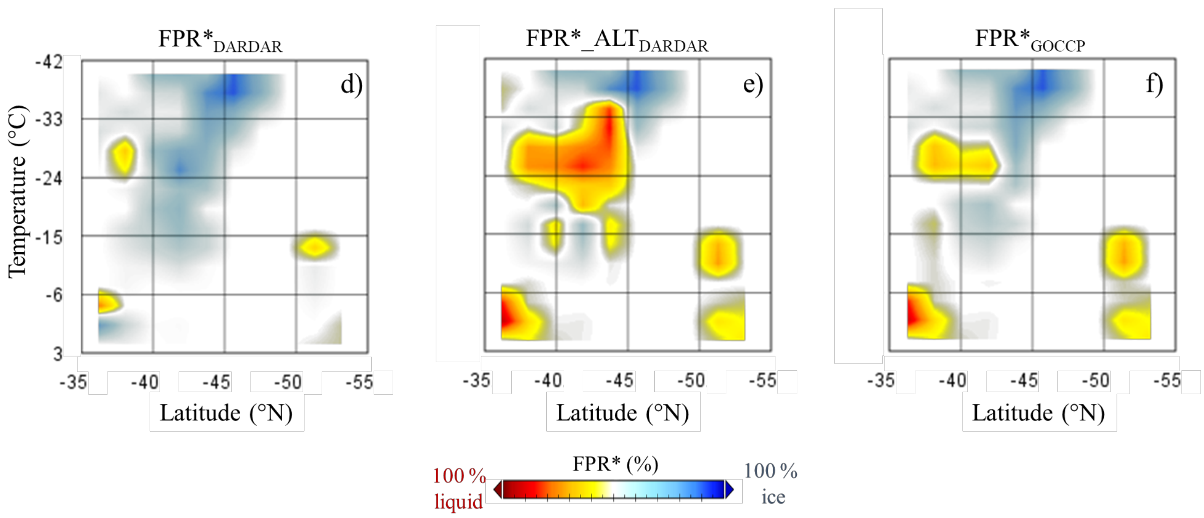


Figure 2 (Modified)

3) Comparing 2d and e, at latitudes between -40 and -45, the whole profiles shows ice cloud in 2d but it is liquid in figure 2e. Even with all the rules described in line 157-158, it is still difficult to understand why it is liquid in figure 2e (the grid box is still 1.875 x 1.875, right?)

The grid box is still 1.875 x 1.875. See the answer to question. The rules have been now clarified as discussed in the previous answer.

Fig.3:
1) I really object to put dust mixing ratio in this figure. There is no physical relationship between dust mixing-ratio and temperature. Dust mixing ratio decreases with height near the source region and temperature generally decrease with height in the atmosphere. But physically, there is no relationship between temperature and dust!

We agree that it may be disorienting to plot the dust mixing-ratio together with the ice occurrence.

We have changed the statement "Of course, this results alone from the relationship between temperature and dust, and it is therefore a good example of a correlation (between dust and FPR) without a direct causality." (lines 237-239), to: "The purpose of plotting both variables together is not to suggest a physical relationship, but to give an insight of the trend of both parameters along temperature."

2) In line 235, "mixing-ratios are higher near the surface than at high altitudes", this is not true for transported dust layer such as over the high latitude regions.

This is a valid exception. However, he intention of Fig. 3 is only to explain the mean vertical distribution of dust at different temperatures.

3) Overall, this study is trying to look at the relationship between pure ice cloud fractions and dust mixing ratios. Therefore, comparing FPRDARDAR and FPRGOCCP has little contribution to the main research topic in this study and the descriptions of converting different FPRs are quite confusing and distracting. In fact, in the proceeding sections, only FPRGOCCP is used most of the time. Therefore, I would suggest just use FPRGOCCP in the study.

We agree that this would simplify the study. However, we believed that the use of both products (FPR_ALT_{DARDAR} and FPR_{GOCCP}) adds confidence to the results shown in Table 1 and rules out any artefacts arising from different cloud phase classification algorithms.

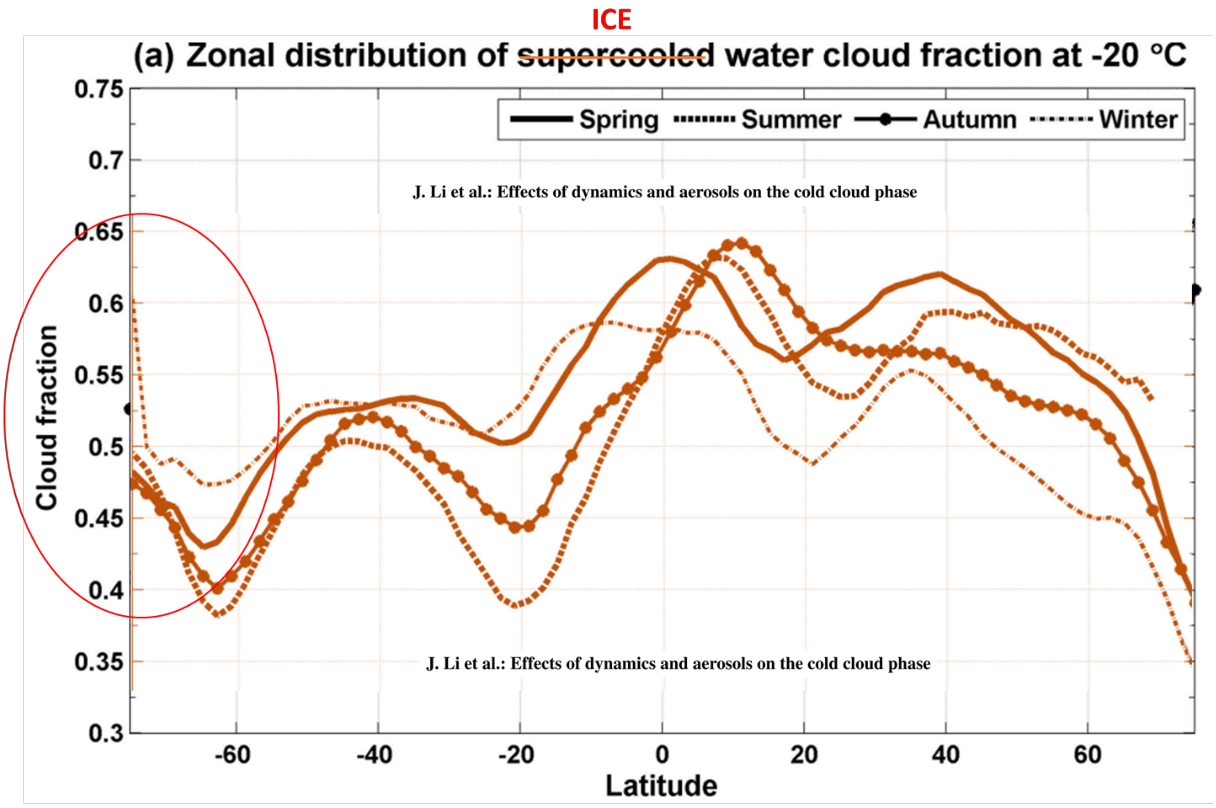
Fig. 4:
1) It is surprising that 'maximum of FPR is located near the Equator'. Kanitz et al. (2011) showed that Northern mid-latitudes have much higher ice-containing cloud fraction than the tropical regions. Do the authors have speculations why the results are so different?

This is a reflection of the larger fraction of cirrus clouds near the equator. Indeed, between 0 and -40°C cirrus clouds are almost exclusively found near the equator, as the polar cirrus clouds occur at a much lower temperature. The results from Kanitz et al. 2011, corresponds to a 2-month local measurement in Cape Verde (January- February 2008, see Ansmann et al., 2009) and are therefore not a suitable comparison to the climatological FPR presented here. These results are consistent with the ones reported in J. Li et al. 2017 (See #4. 1.c). Additionally, it was shown in Fig. 7.19 of the dissertation work of Seifert (2010), that the majority of the altocumulus layers observed over Cape Verde formed in the absence of mineral dust at height levels where westerly upper-tropospheric winds dominate.

References:

Ansmann, A., Tesche, M., Seifert, P., Althausen, D., Engelmann, R., Fruntke, J., … Müller, D. (2009). Evolution of the ice phase in tropical altocumulus: SAMUM lidar observations over Cape Verde. Journal of Geophysical Research Atmospheres. https://doi.org/10.1029/2008JD011659

http://nbn-resolving.de/urn:nbn:de:bsz:15-qucosa-71167



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

Line 251, what’s the reason for the steep increase of FPR at 84o?

The predominance of ice clouds in 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 Antarctica (Ardon-Dryer et al., 2011; Bromwich et al., 2012). Similarly, it has been shown that the orographic forcing in 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.

This has been now included in the manuscript after line 251.

2) Does this steep increase of FPR only occur at -15 C? How about at -20, -25 C?

This increase occurs also at other temperatures, seasons and different dust levels between 0 °C and -20 °C (See Figure #3. 4).

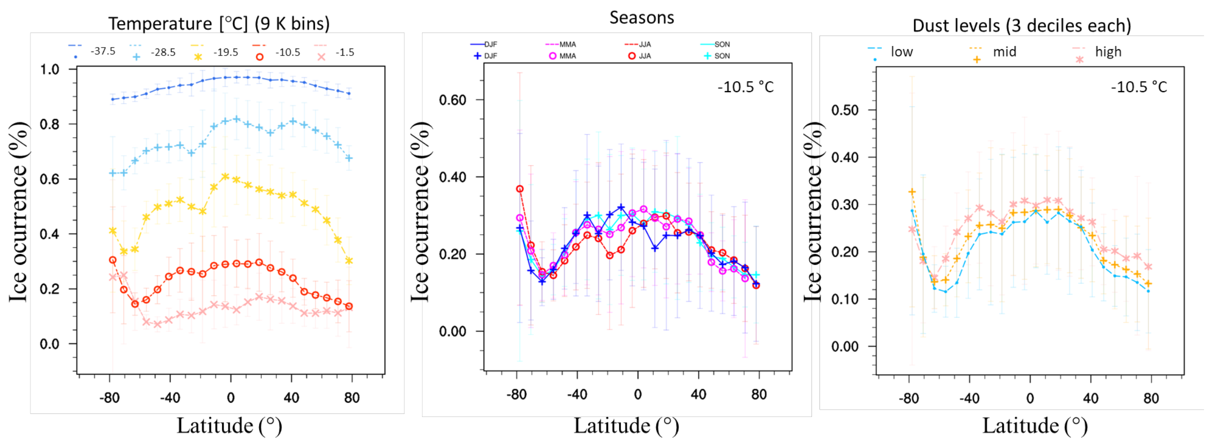


Figure #3. 4 : Zonal mean ice occurrence FPR_{GOCCP} for different temperature ranges, seasons and dust levels (latitude width = 7.5°).

3.a) Line 254, do the referred studies also comparing the fraction of ice clouds at the same temperature?

We have now found a more appropriate reference that reports the Supercooled water cloud fraction at -20 °C (J. Li et al. 2017). This steep increase can be also observed here.

Changed to: “However, a higher FPR in the southern than in the northern polar region is consistent with the fraction of ice clouds previously reported at -20 °C in the literature (Li et al., 2017)*”*

It is really hard to explain why southern polar regions have higher FPR than the northern at a given temperature.

Some studies have already pointed out the predominance of ice clouds in Antarctica, as well as an apparent unexpected source of INP in the southern high latitudes. Although this is an interesting question, further pursuing an explanation of this FPR increase would be out of the scope of the study.

3.b) Also, at -30 °C, the southern polar regions have much lower FPR than the northern, which contradicts with the two referred papers.

We realize that the referenced papers are not comparable to ours due to:

a) Mülmenstädt et al. 2015 uses the average cloud phase of the whole atmospheric column.
b) Huang et al. 2015a uses IWC and LWC, and cannot be directly compared to FPR.

Therefore, we have changed our reference to Li et al., 2017a, which does offer a comparable result at -20 °C. Nevertheles, the differences of FPR at the southern polar regions at -15 °C and -30 °C do rise an interesting question.

We apologize for the misleading citation. This is now corrected in the manuscript.

4) Line 255, I do not see any downdrafts (negative values I guess) in the figure.

The sentence is indeed misleading. The mean large-scale updraft values are only positive, because the average is weighted by cloud volume fraction, which tends to be higher for positive updrafts.

This sentence has been deleted and only the continuing sentence has been left: "*The pattern of the mean large-scale vertical velocity (MACC reanalysis) of the stratiform clouds studied is particularly similar to the FPR at -15°C*"

The purpose of this statement is to emphasize in the similar latitude dependence of updraft velocity and cloud ice occurrence.

5) Line 258, again, it is hard to understand why updraft positively correlated to the occurrence of ice clouds at -20 °C. The referred paper is looking at large scale motion impacts on supercooled liquid fraction. Generally strong updrafts have higher supersaturation and lead to the more liquid formation, and downdrafts cause liquid layer to evaporate, and thus impact the supercooled liquid fraction, which makes sense. I have difficulty to figure out the physical process that updraft velocity lead to the occurrence of ice cloud at -20 °C given that deposition ice nucleation is rare in the atmosphere.

We are not suggesting that updraft velocity lead to the occurrence of ice cloud at -15°C,-20 °C. We merely point out that the spatial correlation between updraft and ice occurrence is positive (as also found by Li et al., 2017a). The large-scale updraft velocity used in Li et al., 2017a is the same parameter used in our study.

Furthermore, there are some plausible (although speculative) explanations for this:

a) The spatial correlation points to a mere coincidence between the larger updraft velocities and higher ice cloud occurrences at lower latitudes.
b) The updrafts are associated to a higher availability of INP at the cloud level (from below the cloud) and the effect is high enough to mask the enhanced liquid formation.
c) The updrafts enhance a certain type of heterogeneous nucleation requiring supersaturation (e.g. immersion freezing). Updrafts generate a cooling rate, activating INPs that may have not been active before the cooling.

These points have been now added in the manuscript.

Figure 8 and Line 390:

1) RH has strong impacts on ice nucleation only for the deposition mode. While at the temperatures between -40 and 0 °C, ice particles are always formed from the freezing of liquid droplets in the atmosphere. Therefore, I do not think it is necessary to include the discussion of RH relationship with dust. In another word, large-scale motion has strong impacts on cloud thermodynamic phase partition, mainly on the formation and evaporation of liquid droplets, but it has little impacts on ice nucleation at the heterogeneous temperature range;

Liquid-dependent freezing only dominates above -25°C. Below this temperature, deposition mode or at least condensation mode plays an increasing role for decreasing temperatures (Hoose and Möhler 2012). We show RH in Figure 8 intending to rule out the possibility of large-scale motions masking the day-to-day relationship between dust and cloud ice/liquid partition. Low humidity may lead to an enhanced WBF mechanism. Additionally, some heterogeneous nucleation modes like immersion freezing require saturation over liquid water. If dust were strongly and positively correlated to RH, this could mean that the observed relationship between dust and ice occurrence could be due to RH. We believe both possibilities make the analysis of RH relevant.

2) Please explain why dust aerosol mixing-ratio increases with isotherm height;

Absorption of solar radiation by dust aerosol may lead to higher isotherms in the column. However, in this case we would expect a higher correlation in the northern mid-latitudes. The influence of dust-loaded air mases coming from warmer lower latitudes (where most dust sources are located) offers a more plausible explanation. This could also explain the larger correlation found in the southern high latitudes.

This has been now added to the discussion.

3) Why there are not downdraft?

As mentioned above, “The mean large-scale updraft values are positive, because they occur together with cloud cover.” Statistically, clouds form at higher saturations and updraft-dominated conditions.

Minor comments:

“CLOUDAT” -> CloudSat.
Corrected.

Line 120: “obtaining good results”. How good are the simulated dust mixing-ratios compared with observations?

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 from 0.6 over the Sahara and Sahel to 0.8 over dust transport regions.”

Line 125: Jan’0 -> Jan’09
Corrected.

Line 138 and 140: the word ‘pixel’ is confusing.
How does pixel be defined in this study?
Is this a layer bin in each lidar or radar profile?
Then how to know whether a pixel is precipitating or not?

Pixels are referred only to the points (x(ray), z(height)) in the raw data from the satellite strides of both the CALIOP(LIDAR) and the CPR(RADAR).

The pixels contain the information of whether the atmospheric column is precipitating.

See also response to Anonymous Referee #1: *"As briefly mentioned in lines 88-89, the 2B-CLDCLASS product uses mainly the radar reflectivity to classify clouds as "precipitating". The radar is sensitive to large particles (e.g. rain drops) and therefore clouds with a reflectivity larger than a temperature-dependent threshold are defined as "precipitating". The fifth range gate (~1.2 km above ground level) is used for the classification. The threshold is defined between -10 and 0 dBZ for temperatures between -10 °C and 0 °C and constant outside this temperature range.*

Hudak et al. 2009 offers a validation of the 2B-CLDCLASS precipitation product and a brief description (paragraphs 10-11) of the precipitation algorithm.

References: Hudak, D., Rodriguez, P. and Donaldson, N.: Validation of the CloudSat precipitation occurrence algorithm using the Canadian C band radar network, J. Geophys. Res. Atmos., doi:10.1029/2008JD009992, 2009.
These points have been added to the Data section."

Line 164: how is cloud volume fraction derived?
What's the benefit using this cloud volume fraction comparing FPR?

The cloud volume fraction is the number of cloudy pixel divided by the total number of pixels within a given length-height domain (satellite swath). The length is the segment of the satellite orbit crossing a given gridbox and the height interval in this study corresponds to each temperature bin (3 K). The main benefit of using the cloud volume fraction is that it is defined for each isotherm (3 K bin). Cloud cover is generally defined for the whole atmosphere or certain levels (low, mid or high). For a range containing multiple temperature bins, it allows the differentiation between vertically thick and shallow clouds. Therefore, *cvf* can be used as an area (horizontal-vertical) weight for the averaging of the data.

This has been now added to the methodology

Line 182 and equation 3.1: How is equation 3.1 is derived or why does this definition differentiate the clear sky and liquid phase condition?

The sole purpose of the definition of FPR* is to plot the cloud phase as a range between LIQUID and ICE while also blending out non-significant clouds (of *cvf~0*). Here the cloud volume fraction *cvf* is used merely as a filter to aid the visualization in Fig. 2.

Line 241: It is not a good starting sentence, and should be put behind Line 243.
Moved

Line 320: Any guess of the common mechanism?

Our leading hypothesis is that heterogeneous ice nucleation (contact and/or impact mode) of mineral dust results in a higher cloud ice occurrence.

This has been emphasised after line 320.

Figure 6: Given similar dust mixing-ratio at -15 °C, the ice occurrence is ~10% higher at mid-latitudes than polar regions, what's the reason?

We have added following explanation in Sect. 4.5:

Furthermore, given similar dust mixing-ratios at -15 °C, the ice occurrence is up to 10 % higher at the mid-latitudes than in the higher latitudes. At -15°C stratiform clouds in the lower- and mid-latitudes (at a lower pressure) are mostly cirrus clouds or altostratus (ice dominated). In contrast, at higher latitudes (at higher pressure) most stratiform clouds are stratocumulus at -15 °C. Stratocumulus are often liquid dominated clouds forming typically at the boundary layer. This results in a lower average occurrence of ice clouds in the higher latitudes compared to the mid-latitudes. Additionally, some studies have suggested that the lower occurrence of cloud ice in the higher latitudes may be associated with lower INP concentrations (Li et al., 2017; Tan et al., 2014; Zhang et al., 2012). This hypothesis has been supported mainly by the spatial correlation between relative aerosol frequency and the supercooled liquid fraction from satellite observations. However, the focus in this study is to assess the temporal (and not the spatial) co-variability between aerosol (mineral dust) and ice occurrence.

Figure 7: at -30 oC, ice occurrence shows little correlations with dust, but rather has dramatic latitude differences. Any speculations of the reason?

Additionally, large differences can be observed between northern and southern hemisphere for both high- and mid-latitudes. We speculate that INP differences between southern and northern hemisphere may cause this pattern. For example, black carbon is known to be active at this temperature and its atmospheric concentration is higher in the northern hemisphere. Other types of dust minerals may also become active at these temperatures (e.g., calcite, chlorite, mica, montmorillonite, Atkinson et al. 2013) and mineralogical differences between dust in the northern and southern hemisphere (Hoose et al., 2008) may also offer a plausible explanation.

We have now added this point in Sect. 4.6 manuscript.

I guess at such colder temperatures, other aerosol particles such as soot are also good INPs. It might be interesting to also look at the relationships between FRP and soot particles.

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 thorough comparison between all potential INP (organic, black carbon, sea spray, …) would greatly overextend the length of the manuscript and would require a regional scope.

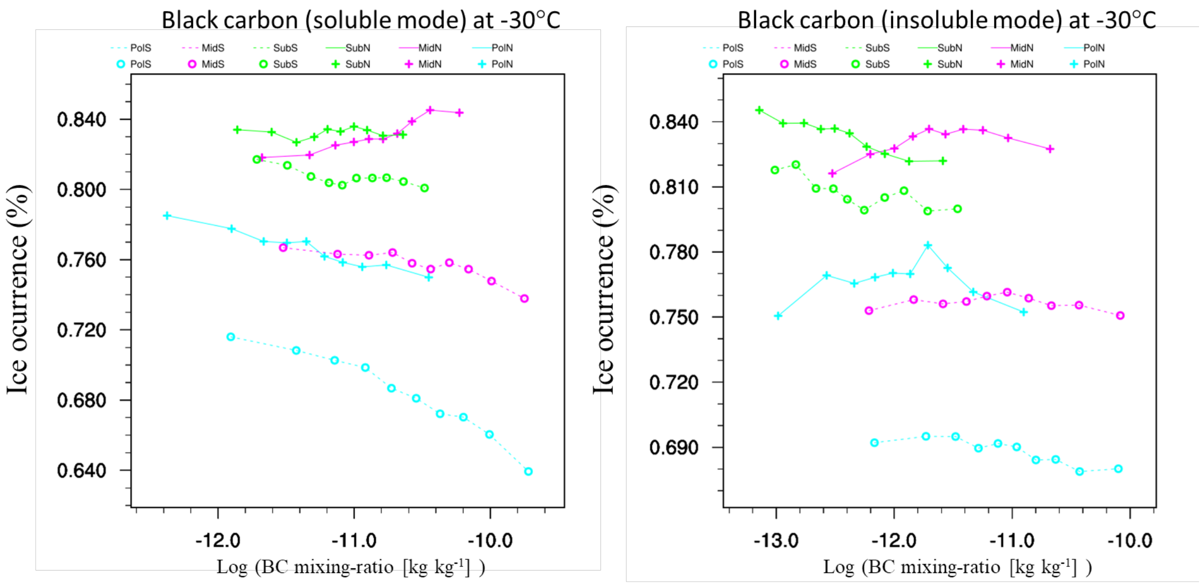


Figure #3. 5: FPR_{GOCCP} for different deciles of black carbon at -30°C

References: Kanitz, T., Seifert, P., Ansmann, A., Engelmann, R., Althausen, D., Casiccia, C. and Rohwer, E. G.: Contrasting the impact of aerosols at northern and southern midlatitudes on heterogeneous ice formation, Geophys. Res. Lett., 38(17), doi:10.1029/2011GL048532, 2011.