#### acp-2019-661

The impact of mineral dust on the day-to-day variability of stratiform cloud glaciation occurrence

Dear Dr. Tesche,

Thank you very much for gathering the referee comments for our paper. Both reviewers expressed their interest in the results and contributed useful concerns about the uncertainty arising from the statistical method. However, we regret that the opinion of the two referees is so critical compared to the opinion of the original four referees from the previous submission of the paper (acp-2018-1074), where already two of the referees had explicitly recommended acceptance after minor/major revisions. We also regret that the uniqueness and advantages of the approach didn't receive much attention from the Referees on this occasion. We hope to have correctly interpreted the comments of Referee #1, despite several erroneous line references — some numbers corresponding to the old version of the manuscript.

Sincerely Diego Villanueva For clarity, referees' comments are written in black, our comments to each concern are written in blue and extractions from the original paper are written in green.

#### Anonymous Referee #1

#### Received and published: 27 September 2019

The manuscript by Villanueva et al. aims at contributing to the interpretation of the role of mineral dust in the formation of ice clouds. In particular, the authors want to demonstrate that the day-to-day the co-variability between mineral dust concentration and cloud glaciation can be used in future as a proxy to better understand heterogeneous freezing mechanism.

First of all I must admit that the manuscript is hard to read in several parts, and it takes more than one reading to make sure that the reported content is fully understood. The assessment of the most suitable datasets utilized for the study of heterogeneous freezing as well as the statistics related to the relationship between ice occurrence, updrafts and dust concentration are very interesting. Nevertheless I am not fully sure that this manuscript demonstrates the value of the day-to-day co-variability as a proxy of the effect on cloud glaciation.

We thank Referee 1 for thoroughly reading the manuscript and for his/her detailed comments. We believe that the manuscript is now easier to read, after the changes we made in response to the Referee's comments. We also thank the reviewer for his/her encouraging comments about the statistics related to the ice occurrence and dust concentrations, which are indeed the main new idea that we want to present in the manuscript. We regret that the referee is still not convinced of the day-to-day statistical approach, but we hope that our response may satisfy the reviewer's concerns.

Below I report my general comments.

• First of all, the authors have selected data from many difference sources, MACC and ERA interim meteorological reanalysis CALIPSO-GOCCP, DARDAR products from the A-Train satellite constellation. What are the effects on the final results presented in the manuscript of combining these datasource with different speculation (resolutions, sampling, uncertainties, ..)?

Concerning the different product resolutions, the choice of 3 K bins is based mainly on the resolution of the CPR (480 m) — used in the DARDAR product — and the original 3 K bins of the CALIPSO-GOCCP product.

In our methodology, a higher resolution (e.g., 1 K bins) would result in a more heterogeneous distribution of the sample size — for any given temperature and day, a larger fraction of gridboxes would contain no clouds — and therefore the statistical uncertainty in the day-to-day statistics would increase. Although the instantaneous satellite products could be resampled to 1 K bins, for the reanalysis the vertical levels would need to be interpolated. Furthermore, some additional technical disadvantages would be associated with a higher resolution. In general, increasing the resolution of the dataset combination would require a new methodology including interpolation of some products. To test if smaller bins could increase the confidence in the results by better constraining the temperature effect on cloud phase, we have performed a quick sensitivity study using the whole range 2007-2010 of the DARDAR-MASK product. In this quick study, we used 3 K bins and 1 K bins to quantify how the vertical resolution affects the variance of the ice-to-liquid ratio. This sensitivity study shows that at -30°C the temporal standard deviation of the FPR decreases by less than 10% in the 1 K bins, despite a decrease of almost 50% in the standard deviation of the temperature inside the temperature bin. Although this represents only a small subset of the data, it shows that a higher vertical resolution not necessarily leads to a decrease in the uncertainty.

Table R1. Temporal standard deviation (STD) of the ice to liquid ratio from the DARDAR-MASK product by different resolutions (RES) for the temperature bins. The daily values for the whole period 2007-2010 were used. Only global means are shown (96 lat×12 lon)

INTERVAL	RES	STD	VARIABLE
-30°C TO -33°C	3K	0.17	FPR_DARDAR
-30°C TO -31°C	1K	0.15	FPR_DARDAR
-31°C TO -32°C	1K	0.16	FPR_DARDAR
-30°C TO -33°C	3K	0.047	Temperature
-30°C TO -31°C	1K	0.028	Temperature
-31°C TO -32°C	1K	0.028	Temperature

Analogously, using a coarser vertical resolution (e.g., 6 K bins) would allow larger temperature variations. For example, a decrease of 3 K is roughly equivalent to a fivefold increase in INP concentrations (e.g., Niemand et al., 2012). However, because the typical range of the day-to-day variations of dust mixing-ratio is also large — about 1 order of magnitude — we expect that the variability of dust loading should dominate over temperature variations, given a constraint of at least 3 K.

The statistical distribution of the ice ratio also limits the resolution options. Single pixels values of cloud phase from the DARDAR and GOCCP products are binary (1 or 0). Therefore, a minimal sample size is required for the averaged cloud phase —within a certain temperature range, gridbox and percentile of dust— to achieve a normal distribution, which allows interpreting the correlation with dust loading directly.

We have now included this matter in the discussion section (without the table):

+(new Lines 517-528) "Concerning the vertical resolutions of the different products, the choice of 3 K bins is based on the resolution of the CPR (480 m) — used in the DARDAR-MASK product — and the original 3 K bins of the CALIPSO-GOCCP product. Using a coarser vertical resolution (e.g., 6 K bins) would hinder the assessment of the role of dust as INP. For example, a decrease of 3 K in temperature is roughly equivalent to a fivefold increase in INP concentrations (e.g., Niemand et al., 2012). Because at the mid- and high-latitudes the typical standard deviation of the day-to-day dust mixing-ratio corresponds to roughly a fourfold increase from the mean (See supplement figure S.5), we expect that the variability of dust loading should dominate over temperature variations, given a temperature constraint of 3 K or less. The statistical distribution of the phase ratio also limits the resolution options. The cloud phase values for single pixels in the DARDAR and GOCCP products are binary (1 or 0). Therefore, a minimal sample size is required for the averaged cloud phase ratio within a certain temperature range, gridbox and percentile of dust — to achieve a normal distribution along time and space, which allows interpreting the correlation with dust loading directly. For this reason, temperature bins smaller than 3 K result in a less normally-distributed cloud phase ratio."

• The authors often do assumptions and simplifications (e.g., use of night time measurements only, neglecting of ice in the mixed phased clouds, . . .) which can strongly increase the uncertainty of the final results and limit the value of the data interpretation. Cloud phase is mainly regulated by temperature and so it is not clear to me why the authors considered only nighttime measurements. What's the effect of this data selection on the final results?

As mentioned in section 3.1, we exclude the daytime retrievals from CALIOP to avoid the influence of noise from sunlight scattering on the retrievals (Li et al., 2017). This is a known issue present in the daytime CALIOP retrievals and this data selection is a necessary step to prepare a consistent dataset (e.g., due to sunlight scattering day and night-time CALIOP retrievals are not directly comparable for our purposes). Nevertheless, the first experiments with the original data showed that the inclusion of daytime retrievals lead to lower FPR-dust correlations (not shown).

As for the neglection of ice in mixed-phase clouds, this was only done in the FPR\_DARDAR\_ALT variable to study the differences between the CALIPSO-GOCCP and DARDAR products. The final results using the GOCCP product are not affected by this simplification.

• The scope of the manuscript is to demonstrate that the day-to-day co-variability of dust concentration ice cloud glaciation may be used to quantify the role of dust aerosol on the cloud thermodynamic phase around the globe. I understand that, in order to use the day-to-day variability, the authors removed the seasonal component subtracting the monthly means. I am not sure this is sufficient to remove all the possible variabilities which can affect the selected data. Weather variability for example occurs on a scale of 5-6 days. Can the authors explain how they can assure the data are not affected by any other relevant variability cycle?

As correctly pointed out by the reviewer, the day-to-day variability may still include the effects of weather variability. To disentangle the weather variability from the day-to-day variability is indeed an interesting idea. Nevertheless, the number of data samples available is not enough to additionally subclassify the retrievals according to local weather states and, at the same time, to also assess the effect of day-to-day dust variability.

We are convinced that it is still possible to study the relationship between dust and cloud phase even without excluding cycles shorter than one month. Although cloud phase may be affected by such cycles (e.g., more liquid clouds at convective fronts and more cirrus clouds at the detrainment regions), it is still possible to distinguish between dusty and non-dusty conditions at each point of the weather cycle. Therefore, once we average over the weather cycle — using monthly means inside each dust percentile — it is still possible to observe the dust-cloud-phase relationship.

These points have been added to the discussion section:

+(new Lines 529-535): "As mentioned in section 3.5, we excluded the seasonal component of the dust-cloudphase correlation by calculating the deciles independently for each month of the year. However, shorter cycles (e.g., weather variability) may still have an influence in the variability of dust and cloud phase. Although the cloud phase may be affected by such cycles (e.g., more liquid clouds at convective fronts and more cirrus clouds at the detrainment regions), it is still possible to distinguish between dusty and non-dusty conditions at each point of the weather cycle. Therefore, once we average over the weather cycle – using monthly means inside each dust percentile – we expect the dust-cloud-phase relationship to be dominated by the microphysical effect of dust on cloud phase."

• In addition, it is not clear to me from the reported description whether the 3K binning can smooth the day-to-day variability (though the binning was needed with respect to the considered dataset)

The 3 K can be understood as averaging the satellite pixels between the height of two different isotherms. These isotherms change on a day-to-day basis as estimated by the reanalysis. Therefore, even small day-to-day changes in temperature (e.g., 1 K) are traduced in slight changes in the isotherm heights and in the interval for which the 3 K bins are calculated. Therefore, we do not expect the effect of day-to-day changes in temperature to be an important source of uncertainty in our analysis.

• ... or anyhow mix different observation scenarios, i.e. high dust content and low content but at the same temperature.

This is an interesting concern. However, the 3K binning is unlikely to produce a significant mixing between different dust scenarios. Even though aerosol plumes can absorb short and longwave radiation, local differences are usually less than 3 K. Even Saharan dust layers has been found to produce temperature differences of only about +2 K in the North Atlantic (Wang and Liu, 2014).

Although cases of high dust concentration over land could produce higher temperature differences — and therefore a temperature inversion —, we do not expect such rare cases to introduce a significant systematic bias in the results. Neither do we expect temperature inversions to play a significant role at temperatures below  $-15^{\circ}$ C, except perhaps near the poles.

#### • I think the description in section 3.2 must be clearer.

-"3.2 Regridding and rebinning: Temperature levels and 1.875°×30° gridboxes

It will become clear in Sect. 4.2. That the cloud thermodynamic phase is mainly a function of temperature. Anticipating this, temperature bins of 3 K each were used as vertical coordinate throughout the study. The temperature profiles were obtained from the ECMWF-AUX reanalysis for the DARDAR and CLDCLASS products and from the MERRA reanalysis for the GOCCP product.

To fill the horizontal gaps between the satellite orbits, we regridded the dataset into a Gaussian T63 grid, aggregating 16 gridboxes along the longitude (1.875°×30°; lat×lon). The Gaussian T63 grid is commonly used in Global Climate Models (Randall et al., 2007) and facilitates future comparisons with global simulations of cloud thermodynamic phase. In section 4.4 and onwards, latitude bands of 30° are used to allow a direct comparison with previous studies (Zhang et al., 2018)."

We have changed the description to:

+(new Lines 178-190) "3.2 Regridding and rebinning: Temperature levels and 1.875°×30° gridboxes

It will become clear in Sect. 4.2. that the cloud thermodynamic phase is mainly a function of temperature. Anticipating this, temperature bins of 3 K each were used as a vertical coordinate throughout the study. The temperature profiles were obtained from the ECMWF-AUX reanalysis for the DARDAR and CLDCLASS products and from the MERRA reanalysis for the GOCCP product. Thus, the same temperature information was used for each product algorithm and for its postprocessing. The temperature at each profile pixel is interpolated using the information from the reanalyses and between -42°C and +3°C, the pixels are averaged into 3 K intervals.

For each of these 3 K intervals, the pixels are then averaged horizontally in a process known as regridding. We regridded the dataset into a Gaussian T63 grid, aggregating 16 gridboxes along the longitude (1.875°×30°; lat×lon) to better fill the horizontal gaps between the satellite orbits. The Gaussian T63 grid is commonly used in Global Climate Models (Randall et al., 2007) and facilitates comparisons with global simulations of the cloud thermodynamic phase. In section 4.4 and onwards, latitude bands of 30° are used to allow a direct comparison with previous studies (Zhang et al., 2018)."

• Many time the authors state that the presented results can be affected by assumptions or effects not considered but also that several properties, which at regional scale may have significant difference, should be reconciled by the fact that the static stability is calculated at the global scale. This is not true for all the variables, RH is an example. There might be strong variations of RH at regional scale in one hemisphere only, which can affect the value of the interpretation of results. Sampling uncertainties are mentioned a few times by the authors themselves, though these are never quantified.

The constraints on statistic stability and RH were intended to constrain the temporal and not the regional variability. Therefore, intervals used as constraints are based on the day-to-day meteorological variations and do not consider the regional variability. We agree with the referee that the uncertainty from the regional variability may be a relevant source of uncertainty. However, such regional variations are at least partially accounted by the standard deviation (zonal) shown in Figures 8-11 (error bars). The focus of the study is not the local regional differences but rather the potential hemispheric differences. Therefore, a deeper investigation of smaller-scale regional correlations is out of the scope of this study.

• For example, if the regions where the data sample is larger and more complete is resampled to reduce the amount and obtain a dataset of homogenous size across the zonal regions what the effect would be?

The suggestion is to change the gridbox size ("resample") at each latitude to obtain the same number of data point inside each gridbox. The result would be a highly irregular grid, however, as the reviewer suggests, the data distribution should be much more homogeneous between such gridboxes. Indeed, there would

be some statistical advantages of this approach (e.g., the zonal averaging would be associated with a lower variance). Nevertheless, new problems would arise, as for example a bias towards regions with higher cloud cover — and therefore a larger sample size. Moreover, our analysis is based on the assumption of a regular grid and we consider the implementation of such a challenging dynamic grid structure out of the scope of our study.

Below I report some detailed comments sometimes still of general breath.

• Line 177-180: Re-gridding operated here generates also a degradation of the horizontal resolution whose effect on the provided analysis is not quantified. It is not clear to me if this is a real advantage or not.

-(Lines 183-186) "To fill the horizontal gaps between the satellite orbits, we regridded the dataset into a Gaussian T63 grid, aggregating 16 gridboxes along the longitude (1.875°×30°; lat×lon). The Gaussian T63 grid is commonly used in Global Climate Models (Randall et al., 2007) and facilitates future comparisons with global simulations of cloud thermodynamic phase. In section 4.4 and onwards, latitude bands of 30° are used to allow a direct comparison with previous studies (Zhang et al., 2018). "

• As stated in the text, the purpose of this regridding is to ensure enough datapoints in each gridbox. It is correct that this degrades the horizontal resolution. However, the analysis is focused on the latitudinal differences — like the North-South hemispheric contrast — rather than on regional differences.

In general, due to the explorative nature of the approach, we did not pursue a detailed sensitivity study of the dataset configuration (e.g., temporal, vertical and horizontal resolution). However, during the initial screening, higher horizontal resolutions (e.g.  $2x2^{\circ}$ ) resulted in a too-large zonal variance, due to the binary-nature of the ice-to-liquid ratio. In other words, gridboxes with only few retrievals per day were strongly biased towards single clouds —liquid(0) or ice(1)—, so that the zonal standard deviation was higher than the changes due to dust loading variability.

- Line 185: replace "in the study" with the "in the study by Huang et al"

   Replaced (Line 194)
- Line2 190-192: please clarify that the choice to ignore ice in mixed phase clouds is an advantage according to the approach you are adopting but also that the authors cannot be sure this has not an impact on the final results.

—(Lines 196–198) "In this alternative definition, which we will call ALT- DARDAR, only gridboxes  $(1.875^{\circ} \times 30^{\circ} \times 3 \text{ K})$  filled with ice pixels are considered as ice (fully glaciated), so that just a single liquid pixel is enough to define a gridbox as liquid (not fully glaciated). One advantage of this marginal definition is that it ignores cloud ice in mixed-phase clouds, which is mostly only detected as such by the DARDAR-MASK product and neglected by the CALIPSO-GOCCP product."

We have now clarified this statement by adding the following:

+(new Lines 203-204)"*However, this neglection of ice in mixed-phase clouds is only carried out to clarify the differences between the products.*"

• Lines 264-266: the authors limit their investigation to the altocumulus clouds: can they quantify the impact of this choice on the final results?

-(Lines 261-266) "Fig. 4a-b show for the same segment the cloud volume cover (CALIPSO-GOCCP) of clouds classified (2B-CLDCLASS) as cirrus or altocumulus (Fig. 4a) and as altostratus or stratocumulus (Fig. 4b). These cloud types are frequently thin enough to be penetrated by lidar and radar systems and are therefore a good target to study cloud glaciation processes (Bühl et al., 2016; D.Zhang et al., 2010b). "

• We are a bit confused by this remark. We are afraid we didn't clarify this point enough. We do not limit the analysis to altocumulus only. Furthermore, these lines refer only to the case study. We have added an introductory statement to improve clarity.

+(new Lines 273-280) "To better understand the differences between the ice-to-liquid ratio retrieved in the DARDAR-MASK and CALIPSO-GOCCP product, this section provides a detailed case study of a stratiform cloud scenario, in which four stratiform cloud types from the CloudSat classification are included — stratocumulus(low-level clouds), altostratus and altocumulus (mid-level clouds), and cirrus (high-level clouds). Although not present in the case study, Nimbostratus are included in the analysis of cloud phase as well and are particularly important in the high latitudes. Stratus clouds are defined for temperatures above 0°C; therefore, they are not relevant for this study. Finally, the horizontal extension of Cumulus and Deep Convection clouds is very low compared to the stratiform clouds and can be therefore ignored in our study, especially outside the tropics (Sassen and Wang, 2008)."

#### • Line 322: I think this simplification can create confusion only.

— "Additionally, in the DARDAR algorithm water can be still classified as ice at +1.5 °C due to the melting layer being set to a wet-bulb temperature (Tw) of 0 °C. This allows the detection of ice at temperatures slightly above 0 °C dry-bulb temperatures (<u>named simply temperature in this work</u>). "

We understand this concern, which was already pointed out in the quick reports — "Line 315: please use Tw, remove the sentence in brackets and do not adopt the proposed simplification throughout the text". We agree that the role of Tw (wet-bulb) and Td (wet-dry) in the DARDAR-MASK product is somewhat confusing. However, the use of Tw throughout the analysis of the DARDAR product would aggravate the problem. Tw is only used in the classification algorithm of the DARDAR-MASK product, and its only purpose here is to define the height below which ice is not acceptable anymore. In other words, Tw has only an influence in the Td range +3°C to 0°C, where it can extend the temperature interval where ice is allowed in the classification. This temperature bin is only briefly mentioned to explain the high temperature end in Figure. 5 and is not further considered afterwards."

• Lines 323-325: though concentration of dust is lower at high altitudes, this does not necessarily indicates that this is due to lower temperatures; this sentence create confusion and solve in a few words a more complicated issue which involves also many other factors, such as atmospheric dynamics and radiative budget. For example, there might be a feedback mechanism influencing the top altitude of aerosols. I think the sentence must be rephrased or otherwise removed.

— (Lines 323-325) "Additionally, the average fine-mode dust mixing-ratio is also shown in Fig. 5. At 0°C the mixing-ratio is five times higher than at -42°C (note the logarithmic right y-axis). This reflects the fact that dust mixing-ratios tend to be lower at higher altitudes where temperatures are lower. However, there are important exceptions to this, such as in the long-range transport of dust layers over the ocean."

#### We have rephrased as follows:

+(new Lines 344-348)" Additionally, the average fine-mode dust mixing-ratio is also shown in Fig. 5. At the height of the 0°C isotherm, the mixing-ratio is on average higher than at the -42°C isotherm (note the logarithmic right y-axis). This reflects the fact that, on average, dust mixing-ratios tend to be higher near the dust sources at the surface. However, this does not imply any general relationship between dust and temperature. Moreover, instant vertical profiles of dust loading and temperature may differ greatly from this average, especially in the long-range transport of dust plumes."

• Line 330: the authors should clarify the reason for the supposed correlation between the maxima observed in the NH and at the tropics, and how the transport of ice clouds downward may occur. Can this be related to any wave activity at the synoptic scale?

—"These maxima are probably associated with the enhanced homogeneous freezing in the tropics at temperatures below  $-40^{\circ}$ C and the resulting downward transport of cloud ice."

First of all, we must clarify that is not the scope of the paper to clarify the mechanisms of ice production in the tropics. Our assumption is based on previous global climate simulations studies, where the main source of cloud ice below the -42°C isotherm is the ice detrained from convective outflows (Gasparini and Lohmann, 2016). These ice particles may be produced by the rapid injection of cloud droplets to temperatures lower than -42°C. Ice particles tend to grow and sediment faster than cloud droplets, and the associated downdrafts may enhance the downward transport of the detrained cloud ice. Because convection in the mid-latitudes is closely associated with cyclone activities, the synoptic-scale may indeed play an important role in the large-scale correlation between updrafts and cloud ice. We encourage future studies seeking to investigate this possibility.

• Lines 342-343: I do not see the steep increase at the Northern Pole. I ask the authors to clarify.

We are a bit puzzled by this remark. The steep increase described in the text is refers to the Southern Pole. We have clarified this as following.

—(Lines <u>348-349</u>)"For both variables, a local minimum near 73°S is followed by a steep increase at 84°S"

This was changed to

+(new Lines 363-365) "Moreover, the FPRGOCCP at  $-15^{\circ}$ C is lower than the FPR\_ALTDARDAR at the southern mid-latitudes and northern high-latitudes. In the southern high latitudes, for both variables, a local minimum near 73°S is followed by a steep increase at 84°S."



• Lines 345-347: I think the authors may remove this lines, too conjectural; digressions are not needed in that part of the manuscript.

(Lines <u>351-353</u>)—"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). "

We have removed these lines.

• Lines 348-350: how much does the number of data influence your conclusions at the South Pole? The authors should discuss this aspect in the paper.

(Lines <u>355-358</u>)— "Similarly, it has been shown that the orographic forcing in Antarctica can lead to high ice water contents for maritime air intrusions (Scott and Lubin, 2016). In other words, maritime air intrusions associated with higher temperatures, higher concentrations of INP and stronger vertical motions could explain the observed pattern in the southern polar regions."

We recognize that these lines are somewhat speculative. We agree with the referee, that the low sample size near the South Pole (Fig. 3 and supplement material s14.b) together with the low altitude of the -15°C isotherm (s12.b) hinders more robust statistics. For example, at -15°C, the zonal standard deviation of the FPR significantly increases from 60°S towards the South Pole — from about ±0.08 to ±0.16 in Fig.6a — at the same time that the sample size decreases from 2200 to 300 (Fig.3).

We have now included this issue in the text.

+ (new Lines 370-374) "However, the low sample size near the South Pole (**Fig. 3** and supplement material **S.14.b**) and the low altitude of the -15°C isotherm (**S.12.b**) result in a lower confidence in the results for this region. For example, at -15°C, the zonal standard deviation of the FPR significantly increases from 60°S towards the South Pole — from about ±0.08 to ±0.16 in Fig.6a — at the same time that the sample size decreases from 2200 to 300 (**Fig.3**)."

• Lines 352 - 355: the correlation mentioned here between the updraft and the FPR looks not so strong, can the authors provide numbers (i.e. regression coefficient or any other statistical tests)?

—"The pattern of the mean large-scale vertical velocity (MACC reanalysis) of the clouds studied is particularly similar to the FPR at  $-15^{\circ}$ C. Moreover, the spatial correlation between large-scale updraft velocity at 500 hPa is positively correlated to the occurrence-frequency of ice clouds at  $-20^{\circ}$ C (Li et al., 2017a). In other words, both the dust mixing-ratio and the large-scale vertical velocity seem to be positively correlated (spatially) to FPR. There are some plausible explanations for this: "

We have calculated the regression coefficient associated with the zonal averages and the  $30^{\circ} \times 1.875^{\circ}$  gridbox averages and we have also emphasized that we suggest only a large-scale correlation with the average updraft velocity. We have changed these lines as follows:

+(new Lines 375-380)"The time-averaged large-scale vertical velocity (MACC reanalysis) of the clouds studied is regionally correlated with the FPR at  $-15^{\circ}$ C — with a pearson correlation coefficient of 0.47 using zonal averages and of 0.31 using the  $30^{\circ} \times 1.875^{\circ}$  gridbox averages. Moreover, in another study, the spatial correlation between large-scale updraft velocity at 500 hPa was also found to be positively correlated (spatially) to the occurrence-frequency of ice clouds at  $-20^{\circ}$ C (Li et al., 2017a). In other words, both the dust mixing-ratio and the large-scale vertical velocity appear to be to some extent correlated (spatially) to the FPR. There are some plausible explanations for this: "

## • Given that a correlation with two different parameters of the FPR is studied, is it the case to carry out a partial correlation analysis?

The vertical velocity from the reanalysis is only a large-scale estimation and it may not coincide with the instant position of the clouds retrieved from the satellite products. For the same reason, LTSS and RH are better parameters to evaluate the possible influence of convection and constrain the influence of dynamics in the dust-cloud-phase correlation. Within this perspective, when we study the response of cloud-phase to different aerosol concentrations at constant LTSS and RH we fulfil the same objective that of a partial correlation analysis. As for the effect of dynamics (i.e., effect of updraft at a constant aerosol loading), this analysis has been already carried out in previous studies (e.g., Li et al., 2017) and is not the focus of this study.

• Lines 423 - 427: I agree with the statement provided by the authors though they should acknowledge that in the SH with low UTSS and high RH the positive correlation is much lower than in other conditions.



(Lines 423-428)—" For dust mixing ratios between 0.1 and 1.5  $\mu$ g kg<sup>-1</sup>, the cloud ice occurrence-frequency at -30°C increase by about +5%. The highest increase is found for the northern latitudes. However, the results from the southern mid-latitudes contradict the notion that the INP activity of mineral dust is of secondary importance in the Southern Hemisphere due to low dust aerosol concentrations (Burrows et al., 2013; Kanitz et al., 2011). Nevertheless, recent studies have acknowledged that the importance of mineral dust in the southern latitudes still cannot be ruled out (Vergara-Temprado et al., 2017)"

• Can the authors comment a bit more on this aspect?

We agree with the referee. However, because the difference is only evident for the southern mid-latitudes, it is very difficult to speculate about the reason behind it. Because this difference is not found in the NH, it is difficult to attribute the effect to the stability and humidity conditions. However, the correlation seems to vary little between the regimes, and therefore, it suggests that the positive correlation is consistent for the different cloud-forming conditions.

Do the authors envisage a larger contribution in the SH of the homogenous nucleation than in other regions?
 We consider that still much investigation is needed before taking a stand in this question. The relative contribution of homogeneous and heterogeneous freezing — and of the different INP types — is still a matter of debate (Barahona et al., 2017; Dietlicher et al., 2018), especially in the mixed-phase regime (temperature range 0°C to -42°C). Furthermore, even if mineral dust was not a dominant INP in the SH, other particles like marine organic aerosols could still represent important INP and influence cloud ice formation.

• Lines 435-439: these lines are to speculative, I'd honestly remove them.

(Lines 435-439)—"In general, for temperatures between  $-36^{\circ}$ C and  $-9^{\circ}$ C, higher fine-mode dust mixingratios are associated with an increasing cloud ice occurrence-frequency. The results suggest that only the lower static stability at  $-15^{\circ}$ C has a strong influence on the relationship between mineral dust and cloud ice. This is may be a consequence of the dynamic component of the atmospheric stability at lower temperatures (e.g., gravity waves), which is not included in the static stability parameter. "

Removed.

• Lines 441-443: Can these results be due to the purer nature of the dust in the SH compared to the NH, where it is often mixed to other aerosol types? In the discussion following to these lines, the authors mention the aged aerosol but never the effect of the aerosol mixing.

(Lines 441-443)—"However, against our expectations, for similar dust loadings the cloud ice occurrence-frequency at -15°C was higher in the Southern than in the Northern Hemisphere."

• This is a very interesting point. Indeed, the higher concentrations of sulphate in the NH are believed to produce coating in dust aerosol and deactivate its freezing potential. It is therefore not difficult to imagine that mixing with other types of aerosols may cause a similar effect. We do mention in the text the potential role of biogenic aerosol mixed with dust aerosol, which would have an enhancing effect in the freezing potential. When we mentioned aged aerosol we refer mostly to the internal mixing (coating) of sulphate with dust aerosol. We have mention this explicitly.

+(new Line 509) "The ageing (e.g., internal mixing with sulfate or "coating") of dust particles may also reduce the freezing efficiency of dust aerosol during the transport from low to high latitudes."

• Line 502: among the significant number of factors contributing the uncertainty affecting the presented analysis I'd add the limitation to consider only a specific type of cloud type, and only night time observations, as well as the effect of the electric charge of mineral dust particles.

• We have added these limitation to the list:

+(new Lines 542-549)"

- Changes in dynamical forcing (e.g., updrafts) and cloud regimes
- *Temperature changes after cloud glaciation (e.g., latent heat release)*
- Ice sedimentation from above (cloud seeding), and INPs other than dust

- Cloud vertical distribution within the studied temperature ranges
- Turbulence favouring aerosol mixing and sub-grid temperature fluctuations
- Differences in dust mineral composition, electric charge and/or size
- Coatings (e.g. Sulfate) affecting aerosol solubility and freezing efficiency
- Subsetting of the data (e.g., only night-time retrievals)"
- Line 535 and following: in this section there are few sentences which are very speculative and though these are able put on the table the pletora of different interpretations to the presented data, at the same time, may be not always helpful to the users, also considering that this is not a research article. I suggest to shorten it or arrange in clearer way.
  - To improve the clarity, we have shortened these paragraphs as follows:

+(new Lines 583-601)" In general, meteorological parameters have a larger impact on cloud properties than aerosols do (Gryspeerdt et al., 2016). For example, different updraft regimes can change the aerosol-cloud interactions in warm clouds by an order of magnitude. Therefore, it is important to study how such meteorological parameters relate to the dust aerosol loading. With this purpose, Fig. 11 shows the mean relative humidity, cloud height and large-scale updraft at  $-15^{\circ}$ C for the different fine-mode dust mixing-ratio deciles and for the four latitude bands studied in Sect. 4.4. Firstly, the correlation between fine-mode dust mixing-ratio from the MACC reanalysis and the RH from the ERA-Interim reanalysis – weighted by cloud volume fraction – was found to be negative (Fig. 11a). We note that the RH from the ERA-Interim reanalysis represents the conditions at a large-scale and not the conditions at a specific location and the moment of the interaction between dust aerosol and supercooled cloud droplets. Still, this relationship is consistent with the intuition that dust is mostly associated with drier air masses. Second, The significant positive correlation found between dust aerosol mixing-ratio and the height of the isotherms (weighted by cloud volume fraction) points to an important source of uncertainty (Fig. 11b). This could be due to clouds being detected in a higher temperature bin after being glaciated at lower temperatures, thus erroneously suggesting an enhanced glaciation occurrence frequency at higher temperatures. Therefore, it is crucial for future studies to take into account this possibility when studying the occurrence of ice clouds at a certain isotherm. More details on the spatiotemporal variability of the cloud height can be found in the supplement (S12) to this article. Lastly, Fig. 11c shows a positive correlation between the fine-mode dust and the large-scale vertical velocity from the MACC reanalysis at  $-15^{\circ}$ C. Updrafts favour saturation over liquid water and therefore CCN activation, droplet growth and inhibition of the WBF (Wegener–Bergeron–Findeisen) process. Therefore, a positive dust-updraft correlation could lead to an underestimation of the dust-cloud-phase relationship."

#### Line 547: something missing in this sentence.

--- "It is possible to find cases where the reanalysis and the detected have different temperatures."

#### Meant was:

+"It is possible to find cases where the reanalysis and the detected **clouds** have different temperatures."

However, we have removed this line after the previous suggestion to shorten this section.

For clarity, referees' comments are written in black, our comments to each concern are written in blue and extractions from the original paper are written in green.

#### Anonymous Referee #2

#### Received and published: 25 September 2019

This study is aimed to estimate the role of dust aerosol on the cloud thermodynamic phase using CALIPSO-GOCCP and DARDAR products for cloud phase and the MACC reanalysis for dust mixing ratio. There are some interesting results regarding the relationship between dust and cloud ice. However, I personally found the manuscript to be difficult to follow, which makes it difficult for me to evaluate the scientific merit of this study.

We thank Referee 2 for the useful comments. We believe that the revised version of the manuscript is now easier to follow.

I have the following major concerns for the authors to considered and clarified:

1. Since this paper talks about the role of dust aerosol on the cloud thermodynamic phase, I thought the analyses would focus on mixed-phase clouds. However, mixed-phase clouds are categorized into liquid clouds in this manuscript (Section 2.3).

- Does the analysis focus on pure ice clouds?
  - If yes, what about the effect of dust on cloud phase?

Line 131: —" In this case, the pixel is categorized as supercooled or mixed-phase depending on the radar signal, which is assumed a priori to indicate the presence of ice particles. Otherwise, the pixel is categorized as ice (Delanoë et al., 2013; Mioche et al., 2014). For reasons that will become clear later, we will coerce the mixed-phase category into the liquid category."—

This may not have been clear enough in the text. We have now emphasized that we do not mean mixed-phase *clouds* but mixed-phase *pixels*. Coercing mixed-phase pixels (Supercooled droplets and ice particles) into supercooled liquid is a common simplification even when studying mixed-phase clouds — in which the pixels at cloud top are classified as liquid, while the pixels below are classified as ice. In other words, the analysis does include mixed-phase clouds, although they are only detected as such in the DARDAR product.

Nevertheless, the analysis is rather focused on the occurrence ratio between ice and supercooled liquid clouds in the CALIOP-GOCCP product at different temperatures and dust conditions. In this product, mixed-phase clouds are mostly detected as supercooled liquid cloud tops.

#### 2. Why only stratiform clouds are considered in this study?

Firstly, including convective clouds from the study does not change the results significantly (not shown). This is because the phase ratio is calculated as an area ratio between ice and liquid pixels at each temperature. Therefore, the phase ratio is strongly dominated by the phase of stratiform clouds.

Moreover, stratiform clouds are a perfect target to study cloud glaciation thanks to the simpler microphysics compared to convective clouds. Because stratiform clouds are thinner than convective clouds, a larger fraction of the vertical structure can be penetrated by CALIOP.

We have now extended this explanation in the methodology section.

+(new Lines 282-287): "These cloud types are frequently thin enough to be penetrated by lidar and radar systems and are therefore a good target to study cloud glaciation processes (Bühl et al., 2016; D.Zhang et al., 2010b). Moreover, stratiform clouds have also simpler microphysics compared to convective clouds, where the dynamical forcing is usually stronger."

3. How would the uncertainties in MACC data, such as the significant overestimate of the fine-dust fraction, affect the analysis results?

We are aware that a significant overestimation exists on the fine-to-coarse dust ratio in the MACC reanalysis (lines 156-157). However, we focused on the relative variations on the dust loading. Therefore, we expect that such overestimations would cause merely a shift in the x-axis with respect to the true values (Fig 8-11), assuming that this overestimation is homogeneous along the dust loading spectrum, which of course would cause some uncertainty. We have now included this issue in the discussion section:

+(new Lines 579-583): "Furthermore, biases such as the overestimation of the fine-mode dust aerosol in the MACC reanalysis (Ansmann et al., 2017; Kok, 2011) may shift the mixing-ratios shown in Sect. 4.4. However, as long as such biases are not limited to certain meteorological conditions, the cloud phase averaged inside each dust decile should remain unaffected."

### 4. How would the authors ensure the consistency among the different datasets, i.e., satellite products and reanalysis data?

As discussed in the text, we expect that the assimilation of the total AOD from MODIS in the MACC reanalysis produce a fair estimation of the large-scale aerosol conditions. At least for the Northern Hemisphere, this has been validated with in situ measurements.

As for the different reanalyses, both the ERA-Interim and the MACC reanalysis are based on the IFS model and use a similar assimilation algorithm.

Among the different satellite products, both rely on CALIOP to determine the presence of clouds. Nevertheless, we are aware that several uncertainties remain, for example, between the meteorology in the reanalysis and in the real atmosphere, a topic that is also discussed in the manuscript.

#### • And how would the inconsistency affect the analyses and conclusions?

In the worst-case that the reanalysis is entirely inconsistent with the retrievals of cloud phase, we expect the result would be the lack of correlation between dust and the ice occurrence (Fig 8-10). In other words, given the large dataset included in the study, we expect that mismatches between reanalysis and cloud retrievals would cause an underestimation of the dust-cloud-phase correlation.

We have included these points in the discussion section:

+(new Lines 560-570): "In general, we expect that the assimilation of the total AOD from MODIS in the MACC reanalysis produce a fair estimation of the large-scale aerosol conditions on a day-to-day basis. At least for the Northern Hemisphere, this has been already validated with in situ measurements (Cuevas et al., 2015). As for the consistency among the different reanalyses, both the ERA-Interim and the MACC reanalysis are based on the IFS model and use a similar assimilation algorithm. Among the different satellite products, both CALIPSO-GOCCP and DARDAR-MASK rely on CALIOP to determine the presence of clouds. Nevertheless, the reader should be aware that several uncertainties remain, for example, between the meteorology in the reanalysis and in the real atmosphere, particularly on the sub-grid scale. In the worst-case that the reanalyses are entirely inconsistent with the retrievals of cloud phase, we expect the result would be the lack of correlation between dust and the ice occurrence (Fig 8-10). In other words, given the large dataset included in the study, we expect that mismatches between reanalysis and cloud retrievals would cause an underestimation — and not an overestimation — of the dust-cloud-phase correlation."

## Relevant changes: acp-2019-661

+(new Lines 178-190)"3.2 Regridding and rebinning: Temperature levels and 1.875°×30° gridboxes

It will become clear in Sect. 4.2. that the cloud thermodynamic phase is mainly a function of temperature. Anticipating this, temperature bins of 3 K each were used as a vertical coordinate throughout the study. The temperature profiles were obtained from the ECMWF-AUX reanalysis for the DARDAR and CLDCLASS products and from the MERRA reanalysis for the GOCCP product. Thus, the same temperature information was used for each product algorithm and for its postprocessing. The temperature at each profile pixel is interpolated using the information from the reanalyses and between  $-42^{\circ}$ C and  $+3^{\circ}$ C, the pixels are averaged into 3 K intervals.

For each of these 3 K intervals, the pixels are then averaged horizontally in a process known as regridding. We regridded the dataset into a Gaussian T63 grid, aggregating 16 gridboxes along the longitude (1.875°×30°; lat×lon) to better fill the horizontal gaps between the satellite orbits. The Gaussian T63 grid is commonly used in Global Climate Models (Randall et al., 2007) and facilitates comparisons with global simulations of the cloud thermodynamic phase. In section 4.4 and onwards, latitude bands of 30° are used to allow a direct comparison with previous studies (Zhang et al., 2018)."

+(new Lines 273-280) "To better understand the differences between the ice-to-liquid ratio retrieved in the DARDAR-MASK and CALIPSO-GOCCP product, this section provides a detailed case study of a stratiform cloud scenario, in which four stratiform cloud types from the CloudSat classification are included — stratocumulus(low-level clouds), altostratus and altocumulus (mid-level clouds), and cirrus (high-level clouds). Although not present in the case study, Nimbostratus are included in the analysis of cloud phase as well and are particularly important in the high latitudes. Stratus clouds are defined for temperatures above 0°C; therefore, they are not relevant for this study. Finally, the horizontal extension of Cumulus and Deep Convection clouds is very low compared to the stratiform clouds and can be therefore ignored in our study, especially outside the tropics (Sassen and Wang, 2008)."

+(new Lines 282-287): "These cloud types are frequently thin enough to be penetrated by lidar and radar systems and are therefore a good target to study cloud glaciation processes (Bühl et al., 2016; D.Zhang et al., 2010b). Moreover, stratiform clouds have also simpler microphysics compared to convective clouds, where the dynamical forcing is usually stronger."

+(new Lines 344-348)" Additionally, the average fine-mode dust mixing-ratio is also shown in Fig. 5. At the height of the 0°C isotherm, the mixing-ratio is on average higher than at the -42°C isotherm (note the logarithmic right y-axis). This reflects the fact that, on average, dust mixing-ratios tend to be higher near the dust sources at the surface. However, this does not imply any general relationship between dust and temperature. Moreover, instant vertical profiles of dust loading and temperature may differ greatly from this average, especially in the long-range transport of dust plumes."

+ (new Lines 370-374) "However, the low sample size near the South Pole (**Fig. 3** and supplement material **S.14.b**) and the low altitude of the -15°C isotherm (**S.12.b**) result in a lower confidence in the results for this region. For example, at -15°C, the zonal standard deviation of the FPR significantly increases from 60°S towards the South Pole — from about ±0.08 to ±0.16 in Fig.6a — at the same time that the sample size decreases from 2200 to 300 (**Fig.3**)."

+(new Lines 375-380)"The time-averaged large-scale vertical velocity (MACC reanalysis) of the clouds studied is regionally correlated with the FPR at  $-15^{\circ}$ C — with a pearson correlation coefficient of 0.47 using zonal averages and of 0.31 using the 30°×1.875° gridbox averages. Moreover, in another study, the spatial correlation between large-scale updraft velocity at 500 hPa was also found to be positively correlated (spatially) to the occurrence-frequency of ice clouds at  $-20^{\circ}$ C (Li et al., 2017a). In other words, both the dust mixing-ratio and the large-scale vertical velocity appear to be to some extent correlated (spatially) to the FPR. There are some plausible explanations for this: "

+(new Lines 517-528) "Concerning the vertical resolutions of the different products, the choice of 3 K bins is based on the resolution of the CPR (480 m) — used in the DARDAR-MASK product — and the original 3 K

bins of the CALIPSO-GOCCP product. Using a coarser vertical resolution (e.g., 6 K bins) would hinder the assessment of the role of dust as INP. For example, a decrease of 3 K in temperature is roughly equivalent to a fivefold increase in INP concentrations (e.g., Niemand et al., 2012). Because at the mid- and high-latitudes the typical standard deviation of the day-to-day dust mixing-ratio corresponds to roughly a fourfold increase from the mean (See supplement figure S.5), we expect that the variability of dust loading should dominate over temperature variations, given a temperature constraint of 3 K or less. The statistical distribution of the phase ratio also limits the resolution options. The cloud phase values for single pixels in the DARDAR and GOCCP products are binary (1 or 0). Therefore, a minimal sample size is required for the averaged cloud phase ratio — within a certain temperature range, gridbox and percentile of dust — to achieve a normal distribution along time and space, which allows interpreting the correlation with dust loading directly. For this reason, temperature bins smaller than 3 K result in a less normally-distributed cloud phase ratio."

+(new Lines 529-535): "As mentioned in section 3.5, we excluded the seasonal component of the dust-cloudphase correlation by calculating the deciles independently for each month of the year. However, shorter cycles (e.g., weather variability) may still have an influence in the variability of dust and cloud phase. Although the cloud phase may be affected by such cycles (e.g., more liquid clouds at convective fronts and more cirrus clouds at the detrainment regions), it is still possible to distinguish between dusty and non-dusty conditions at each point of the weather cycle. Therefore, once we average over the weather cycle — using monthly means inside each dust percentile — we expect the dust-cloud-phase relationship to be dominated by the microphysical effect of dust on cloud phase."

+(new Lines 560-570): "In general, we expect that the assimilation of the total AOD from MODIS in the MACC reanalysis produce a fair estimation of the large-scale aerosol conditions on a day-to-day basis. At least for the Northern Hemisphere, this has been already validated with in situ measurements (Cuevas et al., 2015). As for the consistency among the different reanalyses, both the ERA-Interim and the MACC reanalysis are based on the IFS model and use a similar assimilation algorithm. Among the different satellite products, both CALIPSO-GOCCP and DARDAR-MASK rely on CALIOP to determine the presence of clouds. Nevertheless, the reader should be aware that several uncertainties remain, for example, between the meteorology in the reanalysis and in the real atmosphere, particularly on the sub-grid scale. In the worst-case that the reanalyses are entirely inconsistent with the retrievals of cloud phase, we expect the result would be the lack of correlation between dust and the ice occurrence (Fig 8-10). In other words, given the large dataset included in the study, we expect that mismatches between reanalysis and cloud retrievals would cause an underestimation — and not an overestimation — of the dust-cloud-phase correlation."

+(new Lines 579-583):"Furthermore, biases such as the overestimation of the fine-mode dust aerosol in the MACC reanalysis (Ansmann et al., 2017; Kok, 2011) may shift the mixing-ratios shown in Sect. 4.4. However, as long as such biases are not limited to certain meteorological conditions, the cloud phase averaged inside each dust decile should remain unaffected."

+(new Lines 583-601)" In general, meteorological parameters have a larger impact on cloud properties than aerosols do (Gryspeerdt et al., 2016). For example, different updraft regimes can change the aerosol-cloud interactions in warm clouds by an order of magnitude. Therefore, it is important to study how such meteorological parameters relate to the dust aerosol loading. With this purpose, Fig. 11 shows the mean relative humidity, cloud height and large-scale updraft at -15°C for the different fine-mode dust mixing-ratio deciles and for the four latitude bands studied in Sect. 4.4. Firstly, the correlation between fine-mode dust mixing-ratio from the MACC reanalysis and the RH from the ERA-Interim reanalysis - weighted by cloud volume fraction was found to be negative (Fig. 11a). We note that the RH from the ERA-Interim reanalysis represents the conditions at a large-scale and not the conditions at a specific location and the moment of the interaction between dust aerosol and supercooled cloud droplets. Still, this relationship is consistent with the intuition that dust is mostly associated with drier air masses. Second, The significant positive correlation found between dust aerosol mixing-ratio and the height of the isotherms (weighted by cloud volume fraction) points to an important source of uncertainty (Fig. 11b). This could be due to clouds being detected in a higher temperature bin after being glaciated at lower temperatures, thus erroneously suggesting an enhanced glaciation occurrence frequency at higher temperatures. Therefore, it is crucial for future studies to take into account this possibility when studying the occurrence of ice clouds at a certain isotherm. More details on the spatiotemporal variability of the cloud height can be found in the supplement (S12) to this article. Lastly, Fig. 11c shows a positive correlation between the fine-mode dust and the large-scale vertical velocity from the MACC reanalysis at -15°C. Updrafts favour saturation over liquid water and therefore CCN activation, droplet growth and inhibition of the WBF (Wegener-Bergeron-Findeisen) process. Therefore, a positive dust-updraft correlation could lead to an underestimation of the dust-cloud-phase relationship."

# The day-to-day co-variability between mineral dust and cloud glaciation: A proxy for heterogeneous freezing.

Diego Villanueva<sup>1</sup>, Bernd Heinold<sup>1</sup>, Patric Seifert<sup>1</sup>, Hartwig Deneke<sup>1</sup>, Martin Radenz<sup>1</sup>, Ina Tegen<sup>1</sup>

5 <sup>1</sup>Leibniz Institute for Tropospheric Research, Leipzig, 04318, Germany

Correspondence to: Diego Villanueva (ortiz@tropos.de)

Abstract. To estimate the global co-variability between mineral dust aerosol and cloud glaciation, an aerosol model reanalysis was combined with satellite retrievals of cloud thermodynamic phase. We used the CALIPSO-GOCCP and DARDAR products from the A-Train satellite constellation to assess whether clouds are composed of liquid or ice and the

- 10 MACC reanalysis to estimate the dust mixing-ratio in the atmosphere. Night-time retrievals within a temperature range from +3°C to -42°C for the period 2007-2010 were included. The results confirm that the cloud thermodynamic phase is highly dependent on temperature and latitude. However, at mid- and high latitudes, at equal temperature and within narrow constraints for humidity and static stability the average frequency of fully glaciated clouds increases by +5 to +10% for higher mineral dust mixing-ratios. The differentiation between humidity-stability regimes reduced the confounding influence
- 15 of meteorology on the observed relationship between dust and cloud ice. Furthermore, for similar mixing-ratios of mineral dust, the cloud ice occurrence-frequency in the Northern Hemisphere was found to be higher than in the Southern Hemisphere at -30°C but lower at -15°C. This may suggest a difference in the susceptibility of cloud glaciation to the presence of dust. Based on previous studies, the differences at -15°C could be explained by higher feldspar fractions in the Southern Hemisphere, while the differences at -30°C may be explained by the higher freezing efficiency of clay minerals in
- 20 the Northern Hemisphere.

#### 1 Introduction

Aerosol-cloud interactions affect the Earth's climate through different mechanisms. These include impacts of aerosol particles on cloud glaciation that subsequently influence the clouds' thermodynamic phase, albedo, lifetime and precipitation. Specifically, there is growing evidence for a role of mineral dust aerosol (or of ice nucleating particles correlated to dust aerosol) in influencing heterogeneous cloud ice formation on a global scale (Boose et al., 2016; Kanitz et al., 2011; Seifert et al., 2010; Tan et al., 2014; Vergara-Temprado et al., 2017; Zhang et al., 2018). Cloud droplets can freeze heterogeneously between 0°C and -42°C after interacting with Ice Nucleating Particles (INP) or already existing ice particles (Hoose and Möhler, 2012). It has been shown that specific aerosol types such as mineral dust and biogenic particles can act efficiently as INP already at temperatures between -10 and -20°C (Atkinson et al., 2013). Mineral dust aerosol is emitted from arid regions, mainly from the Saharan and Asian deserts. Despite this, several dust sources exist at the Southern

mid-latitudes (e.g., Patagonia, South Africa, and Australia) and simulations show that long-range transport of dust, although sporadic, can result in considerable dust concentrations even in remote areas (Albani et al., 2012; Johnson et al., 2011; Li et al., 2008; Vergara-Temprado et al., 2017). Mineral dust aerosol is therefore suspected to be a main contributor to the atmospheric INP reservoir, especially in the Northern Hemisphere, where the mixing-ratio of dust aerosol is typically one to two orders of magnitude larger than in the Southern Hemisphere (Vergara-Temprado et al., 2018).

The dust occurrence-frequency retrieved from spaceborne instruments like the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP, Wu et al., 2014) has been previously used to assess the spatial correlation between dust and cloud thermodynamic phase (Choi et al., 2010; Li et al., 2017; Tan et al., 2014). Two main problems arise from this approach. First, aerosol within and below thick clouds cannot be detected by lidar. Second, low dust concentrations usually fall below

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- 40 the lower detection limit of CALIOP. The Aerosol Robotic Network (AERONET, Dubovik et al., 2000), a network of ground-based remote sensing stations, has been used to evaluate and validate the dust retrievals from CALIOP. The stations from the AERONET mission use sun photometers to measure the spectrum of the solar irradiance and sky radiance to determine the atmospheric Aerosol Optical Thickness (AOT). It has been shown that the CALIOP level 2 data misses about half of the dust aerosol events detected by AERONET when the AOT is less than 0.05 (Toth et al., 2018). In contrast, dust
- 45 loadings simulated by state-of-the-art models show that most of the regions in the Southern Hemisphere have an annual mean AOT lower than 0.01 (Ridley et al., 2016).

Ice particles and cloud droplets may coexist in a so-called mixed phase state (Korolev et al., 2017). Shallow mixed-phase clouds with a liquid-dominated cloud top and ice virgae beneath are very frequent (Zhang et al., 2010), whereas cloud tops classified as mixed-phase are much rarer (Huang et al., 2015; Mülmenstädt et al., 2015). Indeed, supercooled liquid layers at

- 50 cloud top are generally observed down to temperatures of -25°C (Ansmann et al., 2008; De Boer et al., 2011; Westbrook and Illingworth, 2011). Ground-based and satellite retrievals are not yet able to accurately estimate the mass ratio of the cloud liquid and ice phase, which is especially the case in mixed-phase layers. Therefore, the Frequency Phase Ratio (FPR) is often used instead (Cesana et al., 2015; Cesana and Chepfer, 2013; Hu et al., 2010). For satellite retrievals, this is defined as the ratio of ice pixels to total cloudy pixels for a region of interest. Because most retrievals classify the cloud
- 55 thermodynamic phase either as pure ice or pure supercooled liquid, the average of the FPR represents the ratio of glaciated clouds with respect to total cloud occurrence. Therefore, the FPR should not be confused with the ice-to-liquid ratio within a single cloud. Cloud phase in the Northern and Southern Hemispheres has been studied in terms of FPR both by ground-based lidar (Kanitz et al., 2011) and by different spaceborne instruments (Choi et al., 2010; Morrison et al., 2011; Tan et al., 2014; Zhang et al., 2018). These studies found significant differences between the two hemispheres. In these studies, it has been
- 60 suggested that such differences are related to differences in aerosol and INP concentrations. Moreover, the local FPR measured at various temperatures between 3°C and -42°C by a lidar in Central Europe over a time span of 11 years has been shown to increase for higher dust loadings (Seifert et al., 2010). Furthermore, spaceborne lidar measurements of cloud thermodynamic phase and aerosol occurrence-frequency show a significant positive spatial correlation between FPR and the

frequency of detectable dust, especially at temperatures of around -20°C (Choi et al., 2010; Tan et al., 2014; Zhang et al.,

- 65 2012, 2015). This spatial correlation has been found under different atmospheric conditions including variations in humidity, surface temperature, vertical velocity, thermal stability and zonal wind speed (Li et al., 2017a). However, the analysis of the day-to-day variability of cloud thermodynamic phase has received less attention, especially in remote areas like the Southern Ocean (Vergara-Temprado et al., 2017). Additionally, a more comprehensive and quantitative assessment of the potential effect of mineral dust on cloud glaciation is currently lacking.
- 70 In this work, the role of dust aerosol on the cloud thermodynamic phase will be assessed based on the daily occurrencefrequency of cloud glaciation around the globe between +3°C and -42°C. For this purpose, the MACC global aerosol reanalysis will be used together with the cloud thermodynamic phase retrievals of the CALIPSO-GOCCP (Global Climate Model Oriented Cloud Calipso Product; Cesana and Chepfer, 2013) and DARDAR-MASK (raDAR-liDAR product; Delanoë and Hogan, 2008, 2010). The FPR obtained from these satellite products is ranked on a day-to-day basis according
- to the dust mixing-ratio from the reanalysis at the moment of the retrieval, considering available observations for the period 2007-2010. Separating the retrievals in different humidity-stability regimes was crucial for assessing the real impact of dust aerosol on cloud glaciation. This work provides a new approach to study the link between dust and cloud thermodynamic phase variability. Its main advantage compared to previous studies is the ability to estimate aerosol mixing-ratios at cloud level and at very low concentrations due to the use of a reanalysis dataset, which is impossible using common remote sensing techniques, due to the detection limits of such retrievals.
- In Sect. 2, the datasets used for the study and the corresponding retrieval algorithms will be presented. In Sect. 3, our processing of the datasets will be described, including the data structure used, the different filters applied to the data and the methodology used to assess the day-to-day correlation between dust and cloud thermodynamic phase. In Sect. 4.1, a case study will be presented to compare the different products of cloud thermodynamic phase used in the study. The variability of
- 85 cloud thermodynamic phase with respect to latitude and temperature will be briefly assessed in Sect. 4.2 and 4.3. In Sect. 4.4, the main results will be presented showing the day-to-day correlation between dust and clouds for different latitudes, temperature ranges, and humidity/stability regimes. In Sect. 5.1, the differences in ice occurrence-frequency between different latitudes will be interpreted based on previous studies of the mineralogical composition of mineral dust. Finally, in Sect. 5.2 the main assumptions, limitations and sources of uncertainty of the approach will be discussed, while conclusions
- 90 will be drawn in Sect. 6.

#### 2 Data

This section presents an overview of the A-Train satellite products and the aerosol reanalysis dataset used in <u>thethis</u> study. To focus the study of cloud phase on stratiform clouds, we will use a cloud classification product (2B-CLDCLASS) to filter out convective clouds in Sect. 4.1-4.3. The cloud phase information will be obtained from two different products: The

95 CALIPSO-GOCCP product will be the focus of the study, while the DARDAR-MASK product will serve to evaluate the possible limitations of the GOCCP product. The aerosol information for the study will be obtained from the MACC reanalysis dataset and will be used in Sect. 4.4 to study the cloud phase at different mixing-ratios of mineral dust. Additionally, the ERA-Interim reanalysis will be used to obtain meteorological information and the MERRA and ECMWF-AUX reanalysis will provide the temperature profiles to rebin the satellite profiles into temperature levels.

#### 100 2.1 2B-CLDCLASS

Different algorithms exist to classify clouds in the observations of spaceborne active instruments (Li et al., 2015). The CloudSat cloud scenario classification (2B-CLDCLASS, Sassen and Wang, 2008) mainly uses the radar reflectivity observed by the Cloud Profiling Radar (CPR) onboard CloudSat together with the attenuated backscatter signal from CALIOP to classify clouds into 8 different types (Sassen and Wang, 2008). These are: low-level (stratocumulus and stratus), mid-level

- 105 (altostratus and altocumulus) and high-level clouds (cirrus), and clouds with vertical development (deep convection clouds, cumulus, and nimbostratus). The main criteria for the classification of non-precipitating clouds are the radar reflectivity and temperature obtained from the ECMWF-AUX product. The CPR is mainly sensitive to large particles (e.g., raindrops) and therefore clouds with a reflectivity larger than a given temperature-dependent threshold can be defined as precipitating. The fifth range gate of the CPR (~1.2 km above ground level) is used for this classification. The threshold is a function of
- 110 temperature, and ranges from -10 to 0 dBZ (Hudak et al., 2009). The standard error of the ECMWF-AUX temperature, which is based on the Integrated Forecast System of the European Center for Medium Range Weather Forecasting (ECMWF), has been estimated to be around 0.6 K in the troposphere (Benedetti, 2005).

#### 2.2 CALIPSO-GOCCP

The CALIPSO-GOCCP v.3.0 product (Cesana and Chepfer, 2013) uses the Attenuated Total Backscatter (ATB), the 115 molecular ATB (ATB<sub>mol</sub>) and the cross-polarized ATB (ATB<sub>1</sub>) from CALIOP at 532 nm wavelength to detect cloudy pixels. The nadir angle of CALIOP was increased from  $0.3^{\circ}$  to  $3^{\circ}$  in November 2007 to reduce specular returns from horizontally oriented ice crystals. The lidar has a horizontal resolution of 333 m and a vertical resolution of 30 m. Cloudy pixels are defined as pixels with a scattering ratio higher than 5 (SR = ATB/ATB<sub>mol</sub> > 5). The cloud volume fraction is defined as the ratio of cloudy to total pixels within a gridbox. The product also uses the depolarization ratio of the retrieved signal

120 components to make a decision on cloud-phase (ICE or LIQUID). The decision is based on an empirical threshold for the depolarization ratio of ice particles and is made for each pixel, with a a vertical resolution of 480 m. From this information, the FPR is calculated for each 2°×2° gridbox. These gridboxes are then regridded into 3 K bins using the temperature from the Modern Era Retrospective-analysis for Research and Applications (MERRA, Bosilovich et al., 2011) reanalysis.

#### 2.3 DARDAR-MASK

- 125 The DARDAR-MASK v1.1.4 product (Delanoë and Hogan, 2008, 2010) available at the ICARE data center combines the attenuated backscatter from CALIOP (at 532 nm; sensible to small droplets), the reflectivity from the CPR (at 94 GHz; sensible to larger particles) and the temperature from the ECMWF-AUX product to assess cloud thermodynamic phase. The radar pixels have a horizontal resolution of 1.4 km (cross-track) × 3.5 km (along-track) and a vertical resolution of 500 m, with a nadir angle of 0.16° of the radar beam. A decision is made for each pixel with a 60 m vertical resolution to take
- 130 advantage of the lidar resolution. These pixels are collocated with the CloudSat footprints (1.1 km horizontal resolution). If the backscatter lidar signal is high (>2·10<sup>-5</sup> m<sup>-1</sup> sr<sup>-1</sup>), strongly attenuated (down to at least 10% in the next 480 m) and penetrates less than 300 m into the cloud, it is assumed that supercooled droplets are present. In this case, the pixel is categorized as supercooled or mixed-phase depending on the radar signal, which is assumed a priori to indicate the presence of ice particles. Otherwise, the pixel is categorized as ice (Delanoë et al., 2013; Mioche et al., 2014). For reasons that will become clear later, we will coerce the mixed-phase category into the liquid category.

#### 2.4 MACC and ERA-Interim

The Monitoring Atmospheric Composition and Climate reanalysis (MACC, Eskes et al., 2015) is based on ECMWF's Integrated Forecasting System (IFS), and simulates the emission, transport and deposition of various aerosol species and trace gases with an output resolution of  $1.125^{\circ} \times 1.125^{\circ}$  and 60 vertical levels. In this study, we use the dust mixing-ratio

- 140 and large-scale vertical velocity from the daily MACC reanalysis product on model levels provided by the ECMWF. Additionally, the Relative Humidity (RH) from the ERA-Interim reanalysis daily product (Dee et al., 2011) will be used in Sect. 5. The cloud properties in the MACC reanalysis are derived from the ECMWF Integrated forecast system (IFS Cycle 36r1 4D-Var). This atmospheric model is analogous to the one used in the ERA-Interim reanalysis (IFS Cycle 31r2 4D-Var). At the time of this study, the new generation of reanalysis based on IFS Cycle 41r was not yet publicly available. However,
- 145 it is expected that future studies will use the new CAMS (Copernicus Atmosphere Monitoring Service) and ERA5 reanalysis instead of the MACC and Era-Interim reanalysis.

Dust emission in the MACC model is parameterized as a function of the 10 m wind, vegetation, soil moisture and surface albedo. The dust loadings are corrected by the assimilation of the total column AOT at 550 nm retrieved from the MODIS instrument on board NASA's Aqua and Terra satellites. Dust sinks are simulated including dry and wet deposition, as well as

150 in-cloud and below-cloud removal.

The freezing efficiency of INPs depends mainly on their surface area concentration (Atkinson et al., 2013; Hartmann et al., 2016; Murray et al., 2011; Niedermeier et al., 2011, 2015; Price et al., 2018). While the number concentration of dust aerosol is generally dominated by fine-mode dust (particle diameter <  $0.5 \mu m$ ), the surface area concentration is often determined by both fine and coarse (particle diameter > 1  $\mu m$ ) dust particles (Mahowald et al., 2014). Moreover, the atmospheric lifetime of

- 155 fine-mode dust is longer than that of coarse-mode dust due to the lower dry deposition rates of finer particles (Mahowald et al., 2014; Seinfeld and Pandis, 1998). Additionally, it has been shown that the MACC model underestimates the coarsemode dust fraction in relation to fine-mode dust (Ansmann et al., 2017; Kok, 2011). In the MACC reanalysis, dust aerosols are represented by three size bins, with size limits of 0.03, 0.55, 0.9 and 20 µm diameter. In this work, we will define the size bin between 0.03 and 0.55 µm as *fine-mode dust*. Because the fine mode contributes to both the number and surface area
- 160 concentration it will be used as a proxy for the concentration of dust INP. Although mostly focused on the Northern Hemisphere, several studies have evaluated the simulated dust mixing-ratios from the MACC reanalysis with observations. A mean bias of 25% was found between MACC and LIVAS, a dust product based on CALIPSO observations over Europe, northern Africa and Middle East (Georgoulias et al., 2018). Additionally, the correlation between MACC and AERONET was found to range from 0.6 over the Sahara and Sahel to 0.8 over typical regions of dust transport (Cuevas et al., 2015).
- 165 Finally, using shipborne measurements of long-range dust transport, it was found that the MACC model significantly overestimates the fine-dust fraction compared to observations (Ansmann et al., 2017).

#### 3 Methods

175

In this section, the different steps in our processing of the datasets presented in Sect. 2 will be described. Fig. 1 presents a flow chart of the data processing and a roadmap for the following subsections.

#### 170 **3.1** Selection of cloud profiles

To exclude the effects of the scattering of sunlight on the detection of the CALIOP lidar signal, only night-time retrievals were used. Additionally, to avoid biases in the radar retrievals at pixels where the lidar is fully attenuated only pixels where the CALIOP retrieval was classified as cloudy (SR > 5) were used. This warranted a dataset free of lidar-attenuated pixels. To avoid biases on the radar reflectivity due to rain droplets, only non-precipitating clouds were included in Sect. 4.1-4.3. Using the 2B-CLDCLASS classification, we defined non-precipitating gridboxes (1.875x1.875) as containing less than 10 % precipitating pixels compared to the total number of cloudy pixels. The three filters (night-time, lidar-not-fully-attenuated and non-precipitating) were applied to both the CALIPSO-GOCCP and DARDAR-MASK cloud products.

#### 3.2 Regridding and rebinning: <u>3K</u> temperature levels and 1.875°×30° gridboxes

HtAs will become clearbe discussed in Sect. 4.2. That, the cloud thermodynamic phase is mainly a function of temperature. Anticipating this, temperature bins of 3 K each were used as <u>a</u> vertical coordinate throughout the study. The temperature profiles were obtained from the ECMWF-AUX reanalysis for the DARDAR and CLDCLASS products and from the MERRA reanalysis for the GOCCP product. Thus, the same temperature information was used for each product algorithm and for its postprocessing. In the products algorithms, the temperature at each profile pixel is interpolated using the information from the reanalyses and between -42°C and +3°C. Then, using this information, we agreggate the pixels into 3 K

185 intervals.

To fill the horizontal gaps between the satellite orbits, For each of these 3 K intervals, the data is then averaged horizontally using nearest-neighbor regridding. We regridded the dataset first into a Gaussian T63 grid, aggregating then aggregated every 16 gridboxes along the longitude  $(1.875^{\circ} \times 30^{\circ}; lat \times lon)$ , gridboxes) to better fill the horizontal gaps between the satellite orbits. The Gaussian T63 grid is commonly used in Global Climate Models ((Randall et al., 2007) and facilitates future comparisons with global simulations of the cloud thermodynamic phase. In section 4.4 and onwards, latitude bands of  $30^{\circ} \times 360^{\circ}$  are used to allow a direct comparison with previous studies (Zhang et al., 2018).

190 c

#### 3.3 Frequency Phase Ratio and cloud volume weighting

Huang et al. (2015) compared the cloud thermodynamic phase retrieved by the DARDAR-MASK and the CALIOP level 2 cloud layer product (Hu et al., 2009) for clouds over the Southern Ocean (at 40-60°S, 125-145°W) and over the North
Atlantic (45°-65°N, 13°-35°W). In thisthe study, by Huang et al., at a cloud top temperature of -10°C almost all cloud tops are classified as liquid by the CALIOP product, whereas most clouds are classified as ice or mixed-phase by the DARDAR-MASK. This discrepancy is attributed partly to the instrumental differences and partly to differences in the classification algorithms.

To assess the differences between the cloud phase from the DARDAR-MASK and CALIPSO-GOCCP products, we defined

- 200 a new phase ratio based on the DARDAR-MASK classification. In this alternative definition, which we will call ALT-DARDAR, only gridboxes (1.875°×30°×3 K) *filled* with ice pixels are considered as ice (*fully* glaciated), so that just a single liquid pixel is enough to define a gridbox as liquid (not *fully* glaciated). One advantage of this marginal definition is that it ignores cloud ice in mixed-phase clouds, which is mostly only detected as such by the DARDAR-MASK product and neglected by the CALIPSO-GOCCP product. However, this neglection of ice in mixed-phase clouds is only carried out to clarify the differences between the meduate.
- 205 <u>clarify the differences between the products.</u>

For FPR<sub>GOCCP</sub> and FPR<sub>DARDAR</sub>, the FPR is calculated as the ratio of ice pixels to the total number of pixels within each gridbox. The FPR\_ALT<sub>DARDAR</sub> uses gridboxes instead: The gridbox phase is set to one (*fully* glaciated cloud) if all cloud pixels in the gridbox are classified as ice and zero (not *fully* glaciated cloud) otherwise. The differences in FPR<sub>GOCCP</sub>, FPR<sub>DARDAR</sub> and FPR\_ALT<sub>DARDAR</sub> will be studied in detail in Sect. 4.1-4.2. Here it will be shown that the FPR\_ALT<sub>DARDAR</sub> definition does well in mimicking the limitations of the CALIPSO-GOCCP product.

- 210 definition does well in mimicking the limitations of the CALIPSO-GOCCP product. We expect cloudy overcast gridboxes to be statistically more robust than partly cloudy gridboxes. Cloud cover is generally defined for the whole atmosphere or certain levels (low, mid or high). Therefore, we use instead the cloud volume fraction (also known as three-dimensional or 3-D cloud fraction; Chepfer et al., 2010, 2013; Li et al., 2017a; Yin et al., 2015) retrieved by the CALIPSO-GOCCP product as a weight for the averages used in Sect. 4.1-4.3. The cloud volume fraction is
- 215 defined as the number of cloudy pixels divided by the total number of pixels within a given length-height domain along the

satellite swath. The length is the segment of the satellite swath crossing a given gridbox, and the height interval corresponds to each temperature bin (3 K) in this study. The main benefit from using the cloud volume fraction instead of the cloud cover is that the former is defined for each temperature bin. This allows to differentiate between vertically thick and shallow clouds. Using the cloud volume fraction as a weight results in a higher representation of clouds with larger spatial extension

220 (vertical as well as horizontal). It also introduces a bias towards the cloud tops for thick clouds because the lidar signal is attenuated at higher cloud depths. More details on the spatiotemporal variability of the cloud volume fraction can be found on the supplement (S8) to this article.

In Sect. 4.1, the adjusted ice volume fraction

$$FPR^* = (2 \cdot FPR - 1) \cdot cvf \tag{3.1}$$

is used instead of the traditional *FPR*, with *cvf* the cloud volume fraction obtained from the GOCCP product. The adjusted FPR\* helps to visualize the cloud thermodynamic phase of the significant (high *cvf*) clouds in the retrieval. In Sect 4.2-4.3, the FPR averages (for each dimension) were calculated as

$$FPR_{avg} = \frac{\sum (cvf_i \cdot FPR_i)}{\sum cvf_i}$$
(3.2)

with cvfi the stratiform cloud volume fraction in each gridbox, defined using the 2B-CLDCLASS classification as

230  $cvf_i = cvf_{i,altostratus} + cvf_{i,cirrus} + cvf_{i,altocumulus} + cvf_{i,stratocumulus}$ (3.3)

#### 3.4 Meteorological regimes

235

Dust aerosol can produce or be accompanied by changes in atmospheric stability and relative humidity. To disentangle such effects, we constrain the cloud environment in Sect. 4.4 using the air relative humidity, the lower troposphere static stability (LTSS) and the upper tropospheric static stability (UTSS). The latter two are defined as:

$$LTSS = T_{700} \left[ \frac{1000}{700} \right]^{\frac{R}{Cp}} - T_{sfc} \left[ \frac{1000}{p_{sfc}} \right]^{\frac{R}{Cp}}$$
(3.4)

$$UTSS = T_{350} \left[ \frac{1000}{350} \right]^{\frac{R}{Cp}} - T_{500} \left[ \frac{1000}{500} \right]^{\frac{R}{Cp}}$$
(3.5)

With  $T_x$  and  $P_x$  the temperature and pressure at the surface or at *x* hPa using the pressure levels of the ERA-Interim reanalysis. *R* is the gas constant and  $C_p$  the specific heat capacity of air (Klein and Hartmann, 1993). The relative humidity is obtained directly from the ECMWF-AUX reanalysis.

240 To increase the sample size prior to the regime classification, we included back gridboxes containing precipitating or convective clouds back into the dataset. However, most of such clouds are expected to fall into high RH and low LTSS regimes and, therefore, could still be excluded later on.

#### 3.5 Classification of dust loads and day-to-day correlation

In contrast to previous studies, in this work we want to isolate the day-to-day correlation between dust aerosol and cloud

- 245 phase. To exclude the spatial component of the correlation, the complete time span 2007-2010 is used to determine the timedeciles of dust mixing-ratio using the MACC reanalysis for each volume gridbox (latitude, longitude, temperature). These deciles are used to sort the daily data depending on the daily dust mixing-ratio into 10 different decile ranks. These ranks can be also understood as dust mixing-ratio bins (from now on simply *deciles*).
- Next, we also need to exclude the seasonal component of the temporal correlation. With this purpose, for each 3 K temperature bin and each gridbox the daily data is averaged within each dust decile and each month of the year. This is done as a multiyear average (e.g., January containing Jan'07, Jan'08, Jan'09 and Jan'10). The resulting field contains one extra dimension for each gridbox (month, *dust decile*, temperature, latitude, longitude). Fig. 2 present a visualization of this process.

#### 3.6 Data availability and averaging order

- 255 Fig. 3 shows the zonal sum of the sample size for the FPR<sub>GOCCP</sub> at −15°C and −30°C. Each count corresponds to a monthdecile pair. Within a 12 K range, each latitude bin (1.875°) contains about 1500 to 2000 observational datapoints in the midlatitudes and about 500 to 1500 datapoints in the high-latitudes, with the lowest sample size found for the high southern latitudes. Potential reasons for missing data are:
  - The satellite swaths (orbits) produce a different density of retrieved profiles at different latitudes.
  - Using only night-time data, the sample size in the meteorological summer time (shorter nights) is lower.
    - The cloud phase retrievals are less frequent for seasons, regions and heights with low cloud cover.
    - At high latitudes, relatively warm temperatures (e.g., -15°C) exceeding the surface temperature can be found, and therefore no information is available for such temperatures (e.g., over Antarctica in winter).
- To avoid artefacts arising from the averaging of dimensions containing missing values, the averaging order of dimensions 265 was defined (going from the first to the last dimension to be averaged) as: longitude, month, decile, latitude, temperature. Latitude and temperature are averaged last because of the higher associated correlations with cloud phase (Sect. 4.2 - 4.3 of this study; Choi et al., 2010; Tan et al., 2014). Each 1.875°×30° gridbox contains on average 100 to 200 datapoints at -15°C (within a 12 K range) in the mid-latitudes. Meanwhile, in the subtropics and in the high latitudes, the sample size is much more heterogeneously distributed and can drop below 50 datapoints near the poles and in subsidence regions. A detailed
- 270 view of the spatiotemporal distribution of the sample size for stratiform clouds can be found in the supplement (S14) to this article.

260

#### 4 Results

#### 4.1 Case study

To better understand the differences between the ice-to-liquid ratio retrieved in the DARDAR-MASK and CALIPSO-

- 275 GOCCP product, this section provides a detailed case study of a stratiform cloud scenario, in which four stratiform cloud types from the CloudSat classification are included stratocumulus(low-level clouds), altostratus and altocumulus (mid-level clouds), and cirrus (high-level clouds). Although not present in the case study, Nimbostratus are included in the analysis of cloud phase as well and are particularly important in the high latitudes. Stratus clouds are defined for temperatures above 0°C; therefore, they are not relevant for this study. Finally, the horizontal extension of Cumulus and
- 280 <u>Deep Convection clouds is very low compared to the stratiform clouds and can be therefore ignored in our study, especially outside the tropics (Sassen and Wang, 2008).</u>

**Fig. 4** shows a case study at 9:50 UTC on Dec 14, 2010 over the Southern Ocean for temperatures between -42°C and +3°C. This A-train segment has been already chosen for a previous case study by Huang et al. (2015) due to the variety of cloud types it contains. **Fig. 4a-b** show for the same segment the cloud volume cover (CALIPSO-GOCCP) of clouds classified

- (2B-CLDCLASS) as cirrus or altocumulus (Fig. 4a) and as altostratus or stratocumulus (Fig. 4b). These cloud types are frequently thin enough to be penetrated by lidar and radar systems and are therefore a good target to study cloud glaciation processes (Bühl et al., 2016; D.Zhang et al., 2010b). Moreover, stratiform clouds have also simpler microphysics compared to convective clouds, where the dynamical forcing is usually stronger. Fig. 4c shows the mixing-ratio of fine (0.03µm-0.55µm) dust aerosol (MACC reanalysis) for the same vertical plane. Fig. 4d-f shows the FPR\* (see Sect. 3) which is
- 290 weighted by cloud volume fraction to highlight the phase of extensive clouds. Some major differences can be observed between the three FPR\* variables in Fig. 4d-f. For the altocumulus cloud at 35-40°S and +3°C to −6°C, the ice virgae falling from the cloud (FPR<sub>DARDAR</sub>) are missed in the FPR<sub>GOCCP</sub>. Because this study aims at assessing the occurrence-frequency of fully glaciated clouds, such mixed-phase clouds are then reclassified in FPR ALT<sub>DARDAR</sub> as liquid clouds. A similar case is observed for the stratocumulus clouds at 50-55°S and +3°C to −6°C, and
- 295 for the altostratus at 35-45°S below the -20°C isotherm (at higher temperatures). Finally, the cirrus clouds above -33°C remain nearly unaffected by the reclassification in FPR\_ALT<sub>DARDAR</sub> as it is classified as fully glaciated. Clouds between 38°N and -44°N, ranging from -6°C to -33°C in temperature, are classified mostly as *altostratus* by the 2B-CLDCLASS product. These altostratus clouds offer a good opportunity to compare the three FPR variables in more detail.

 $FPR_{GOCCP}$ : The detected ice virgae below the liquid cloud top suggest that the cloud top did not fully attenuate the lidar 300 signal (not optically 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 and was therefore classified as liquid.

*FPR*<sub>DARDAR</sub>: 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 ( $\beta$ ) is lower than 2.10<sup>-5</sup> m<sup>-1</sup> sr<sup>-1</sup>
- b) If not *a*): If it is *weakly attenuated* (less than 10 times) or *not rapidly attenuated* (at a depth larger than 480 m).
  - c) If not *b*): If the *layer thickness* of the cloud is larger than 300 m. This is equivalent to 5 pixels with a lidar vertical resolution of 60 m.

Therefore, there are many cases where a mixed-phase cloud (and especially an optically thin stratiform cloud) can be missclassified as ice only in the DARDAR product and consequently in the FPR\_DARDAR variable. In this specific case, we

310 speculate that *c*) is the most probable cause because of the large vertical extent of the clouds around 1 to 5 km using a moist adiabatic lapse rate of -6 K/km for the estimation).

*FPR\_ALT<sub>DARDAR</sub>*: In the case of droplets and ice particles coexisting at cloud top, we expect that at some location the cloud droplets will be enough in number for the pixel to be classified as liquid (strong attenuation) in the DARDAR-MASK algorithm. If this is the case, the entire gridbox value of FPR\_ALT\_DARDAR will be LIQUID. This should be interpreted as a

315 *non-completely glaciated* cloud.

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In summary, 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 introduced FPR\_ALT<sub>DARDAR</sub>, which has been defined to address the limitations of both products. In FPR\_ALT<sub>DARDAR</sub>, a significant portion of mixed-phase clouds that would otherwise be classified as ICE are now

320 classified as LIQUID. This however partly reintroduces the inability of the GOCCP algorithm to detect ice in mixed-phase clouds. Therefore, the *frequency of completely glaciated clouds*, which is represented by FPR\_ALT<sub>DARDAR</sub> and FPR<sub>GOCCP</sub>, allows a better comparison of both algorithms, mostly by ignoring ice virgae in FPR\_ALT<sub>DARDAR</sub> when cloud droplets are also present in the same gridbox. This idea is summarized in Table 1.

#### 4.2 Temperature dependence

- 325 Mixed-phase clouds between 0°C and -25°C are usually topped by a liquid layer (Ansmann et al., 2008; De Boer et al., 2011; Westbrook and Heymsfield, 2011). Below this layer, there is often a thicker layer containing ice particles. Because the CPR is more sensitive to larger particles, this results in a large fraction of the cloud classified as ICE in the DARDAR-MASK. In contrast, the CALIOP backscatter signal is usually already strongly attenuated at such depths and often cannot detect large ice particles. Therefore, the CALIPSO-GOCCP algorithm usually classifies the whole cloud layer as liquid 330 (Huang et al., 2012; 2015). As a result, FPR<sub>DARDAR</sub> tends to be higher than FPR<sub>GOCCP</sub>.
- Fig. 5 shows that the global average FPR<sub>GOCCP</sub> as a function of temperature decreases roughly from 100 % at  $-40.5^{\circ}$ C to about 20 % at  $-1.5^{\circ}$ C and down to 0 % at  $+1.5^{\circ}$ C. This temperature dependence between  $-42^{\circ}$ C and 0°C is also observed for a wide range of parameterizations in global climate models (Cesana et al., 2015), in ground-based measurements (Kanitz et al., 2011), in spaceborne lidar measurements (Tan et al., 2014) and in aircraft measurements (McCoy et al., 2016). However,
- 335 for the same temperature range, the FPRDARDAR only decreases down to 60 % at 1.5°C. This is partly due to the higher

sensitivity of the radar to ice particles, especially falling ice. Additionally, in the DARDAR algorithm water can be still classified as ice at  $+1.5^{\circ}$ C due to the melting layer being set to a wet-bulb temperature (Tw) of 0°C. This allows the detection of ice at temperatures slightly above 0°C dry-bulb temperatures (named simply temperature in this work). For instance, at a relative humidity of 50%, a temperature of about  $+2.5^{\circ}$ C would correspond to a Tw of  $-2.5^{\circ}$ C. Nevertheless,

- 340 this last effect is not relevant for temperatures below freezing. In contrast, FPR\_ALT<sub>DARDAR</sub> follows very closely the pattern of the FPR<sub>GOCCP</sub> down to -1.5°C. The absolute differences of the global averaged FPR\_ALT<sub>DARDAR</sub> and FPR<sub>GOCCP</sub> are less than 10 % between -42°C and 0°C. This shows that the temperature dependence of the alternative phase ratio FPR\_ALT<sub>DARDAR</sub> and FPR<sub>GOCCP</sub> agree better than for FPR<sub>DARDAR</sub>. Therefore, for the rest of the study, only FPR\_ALT<sub>DARDAR</sub> and FPR<sub>GOCCP</sub> will be considered.
- Additionally, the average fine-mode dust mixing-ratio is also shown in Fig. 5. At <u>the height of the 0°C isotherm</u>, the mixingratio is <u>five timeson average</u> higher than at <u>the</u>-42°C <u>isotherm</u> (note the logarithmic right y-axis). This reflects the fact that, <u>on average</u>, dust mixing-ratios tend to be <del>lower at</del> higher <del>altitudes</del> where temperatures are lower.near the dust sources at the <u>surface</u>. However, there are important exceptions to this, such as does not imply any general relationship between dust and temperature. Moreover, instant vertical profiles of dust loading and temperature may differ greatly from this average,
- 350 <u>especially</u> in the long-range transport of dust layers over the ocean. plumes.

#### 4.3 Latitude dependence

Fig. 6 shows the latitudinal dependence of dust and cloud thermodynamic phase at -30°C (averaged from -36°C to -24°C; Fig5a) and at -15°C (averaged from -21°C to -9°C; Fig 5b). For both temperature ranges shown in Fig. 6 the absolute maximum of FPR is located near the Equator. At -30°C, the maximum is 85% for FPR<sub>GOCCP</sub> and 78% for FPR\_ALT<sub>DARDAR</sub>
and at -15°C the FPR from both products peak at 44%. These maxima are probably associated with the enhanced homogeneous freezing in the tropics at temperatures below -40°C and the resulting downward transport of cloud ice. Similarly, minima are observed towards the high latitudes. At -30°C, the FPR<sub>GOCCP</sub> has two local maxima with values of 76% and 84 % near 39°S and 39°N, respectively. Similar local maxima are observed for the FPR\_ALT<sub>DARDAR</sub> but at higher latitudes, at 61°S and 61°N with values 69 % and 74 %. At -30°C, both products show a higher FPR in the Northern Hemisphere, in particular for the high latitudes. This higher FPR coincides with the higher average dust mixing-ratio in the Northern Hemisphere. Such positive spatial correlations between FPR and dust aerosol have been already pointed out using the dust occurrence-frequency derived from CALIOP (Choi et al., 2010; Tan et al., 2014;

Zhang et al., 2012).

In comparison, the differences between FPR<sub>GOCCP</sub> and FPR ALT<sub>DARDAR</sub> at -15°C are much lower than at -30°C as shown in

365 **Fig. 6b**. Moreover, the FPR<sub>GOCCP</sub> at -15°C is lower than the FPR\_ALT<sub>DARDAR</sub> at the southern mid-latitudes and northern high-latitudes. In the southern high latitudes, for both variables, a local minimum near 73°S is followed by a steep increase at 84°S. The larger standard deviation in these latitudes is possibly a result of the low sample size in this the region, as

mentioned in Sect. 2. However, the higher FPR in the southern than in the northern polar region is consistent with the fraction of ice clouds reported previously in the literature at  $-20^{\circ}$ C (Li et al., 2017). The predominance of ice clouds in

- 370 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 earbon (Bromwich et al., 2012). Particularly, immersion freezing INP are thought to be significant in Antarctica (Ardon Dryer et al., 2011; Bromwich et al., 2012). SimilarlyOn the other hand, it has been shown that the orographic forcing in Antarctica can lead to high ice water contents for maritime air intrusions (Scott and Lubin, 2016). In
- 375 other words, maritime air intrusions associated with higher temperatures, higher concentrations of INP and stronger vertical motions could explain the observed pattern in the southern polar regions. However, the low sample size near the South Pole (Fig. 3 and supplement material S.14.b) and the low altitude of the -15°C isotherm (S.12.b) result in a lower confidence in the results for this region. For example, at -15°C, the zonal standard deviation of the FPR significantly increases from 60°S towards the South Pole from about ±0.08 to ±0.16 in Fig.6a at the same time that the sample size decreases from 2200

#### 380 to 300 (Fig.3).

385

390

The pattern of the meanFor the clouds studied, the time-averaged *large-scale* vertical velocity (from the MACC reanalysis) of the clouds studied is particularly similar togomewhat regionally correlated with the FPR at  $-15^{\circ}$ Cr — with a pearson correlation coefficient of 0.47 using zonal averages and of 0.31 using the 30°×1.875° gridbox averages. Moreover, in another study, the spatial correlation between large-scale updraft velocity at 500 hPa is also found to be positively correlated (spatially) to the occurrence-frequency of ice clouds at  $-20^{\circ}$ C -(Li et al., 2017a). In other words, both the dust mixing-ratio and the large-scale vertical velocity seemappear to be positively to some extent correlated (spatially) to the FPR. There are

some plausible explanations for this:

- The spatial correlation can be a result of an enhanced transport of water vapor to higher levels at temperatures below -40°C and the subsequent sedimentation of ice crystals from the homogeneous regime (cloud seeding).
- The updrafts are associated with higher availability of INP at the cloud level (from below the cloud) and the effect is large enough to mask the enhanced droplet growth typically associated with updrafts.
  - The updrafts enhance a certain type of heterogeneous nucleation requiring saturation over liquid water (e.g., immersion freezing). Updrafts generate a local adiabatic cooling, possibly activating INPs that may have not been active before at higher temperatures.
- 395 However, to understand which (if any) of these explanations influence the freezing processes inside the cloud remains a complex challenge forof ongoing debate (Sullivan et al., 2016).

#### 4.4 Constraining the influence of static stability and humidity on the dust-cloud-phase relationship

To study the temporal correlation between mineral dust mixing-ratio and cloud ice occurrence-frequency (from now on referred to as *dust-cloud-phase relationship*) it is crucial to systematically classify weather regimes to constrain the

- 400 meteorological influence. By doing so, the resulting dust-cloud-phase relationship may offer a good insight into how heterogeneous nucleation of mineral dust may affect the day-to-day average cloud thermodynamic phase. In other words, to extract the specific influence of mineral dust on cloud glaciation, it is necessary to identify and constrain relevant meteorological confounding factors (Gryspeerdt et al., 2016). The atmospheric relative humidity and static stability are good candidates for such a confounding factor (Zamora et al., 2018). Both are correlated with the transport of mineral dust and
- 405 vary between different cloud regimes. Additionally, relative humidity is, next to the temperature, one of the main factors in the initiation of ice nucleation in laboratory studies (Hoose and Möhler, 2012; Welti et al., 2009). The static stability of the atmosphere (see equations 3.4 and 3.5) represents the gravitational resistance of an atmospheric column to vertical motions and is defined as the difference of potential temperature between two pressure levels (Klein and Hartmann, 1993). Because such vertical motions are traduced in a temperature change rate within the air parcel, the static stability can have an
- 410 important impact on the heterogeneous freezing rates, especially on immersion freezing. We note that the dynamic component of the atmospheric stability is not included in the static stability. Especially in the upper troposphere, atmospheric gravity waves occurring during stable thermal conditions may also result in vertical motions affecting ice production. In general, moist and unstable conditions are associated with enhanced lifting of air that likely causes nucleation of
- hydrometeors. The effect of humidity and static stability on ice production is not straightforward. In moist convective conditions (high humidity and low static stability) between 0°C and -40°C, the supersaturation of water vapor over liquid increases the liquid formation because ice growing (deposition nucleation) is rather inefficient at this conditions. However, at temperatures below -40°C, ice production by deposition and homogeneous nucleation are favoured, which can result in a higher occurrence of cloud ice in the mixed-phase regime below due to ice sedimentation. To constrain both the atmospheric stability and humidity, a subset of the data must be found within a narrow range of these variables. At the same time, enough
- data points must still be available to assess the dust-cloud-phase relationship. For this purpose, we use a probability histogram to define the regime bounds such that at least 10% of the data is included in each regime.
  Fig. 7 shows the probability density function of the dataset against the relative humidity from the ECMWF-AUX dataset and the static stability from the ERA-Interim reanalysis at -22°C. The bounds for each regime are shown with boxes. For the

relative humidity, the bounds are defined at 60, 70 and 80%, for the LTSS at 10, 15 and 20 K, and for the UTSS at 4, 6 and

- 425 8 K. The fraction of data inside each regime correspond to the integral of the probability density within the regime bounds. For example, if the probability density between 4–6 K and 70–80% is 0.01, then 20% of the data is contained between these bounds. The magenta boxes represent the different stability-humidity regimes used for the lower and upper troposphere. To maximize the sample size, for this classification and for the following results precipitating and convective clouds are also included.
- 430 Fig. 8 shows the dust-cloud-phase relationship for the mid- and high-latitudes separated by humidity and LTSS at  $-15^{\circ}$ C using the FPR<sub>GOCCP</sub> product and MACC reanalysis. For dust mixing-ratios between 0.1 and 2.0 µg kg<sup>-1</sup>, the cloud-phase curve in the mid-latitudes follows a similar logarithmic increase of cloud ice occurrence-frequency of about +6% for low-

LTSS and +4% for high-LTSS conditions. After analysing 11 years of ground-based lidar measurements in Leipzig, Seifert et al. (2010) reported a slightly higher increase by about +10% between  $-10^{\circ}$ C and  $-20^{\circ}$ C for dust concentrations

- 435 between 0.001 to 2 μg m<sup>-3</sup> (note the different units). In our results at -15°C, the cloud ice occurrence-frequency tends to be higher for higher relative humidity and the LTSS seems to have a major effect on the dust-cloud-phase relationship. For high-LTSS conditions (**Fig. 8a-b**), a positive dust-cloud-phase correlation can be observed at all four latitude bands. The slope is similar for the Northern and Southern Hemisphere and for mid- and high latitudes. Because the horizontal axis is logarithmic, this means that for the high-latitudes small increases in dust mixing-ratio are associated with a high increase in
- 440 cloud ice occurrence-frequency. However, the range of ice occurrence-frequency values is higher for the high latitudes. Particularly for the low-RH regime, the ice occurrence-frequency in the southern high latitudes increases by +8%, and at the mid-latitudes, the increase is only about +4%. In both mid- and high latitudes, the cloud ice occurrence-frequency for the same dust mixing-ratio is about +2% to +8% higher in the Southern than in the Northern Hemisphere. This could point to a factor other than dust aerosol causing an increased ice occurrence-frequency in the Southern Hemisphere. It could also suggest a potential difference in the sensitivity of cloud glaciation to mineral dust between hemispheres. The difference
- between the Northern and Southern Hemisphere is reduced in the high-RH regime, as well as the standard deviation of the ice occurrence-frequency, possibly due to the higher sample size density in the high-RH regime. For the low-LTSS regime (Fig. 8c-d), the cloud thermodynamic phase in the high-latitudes remains mostly constant for increasing dust mixing-ratios, and the dust-cloud-phase curves for the mid-latitudes coincide so that the maximum cloud ice occurrence-frequency in the southern mid-latitudes is similar to the minimum in the northern mid-latitudes.
- Fig. 9 show a similar constraint on humidity and UTSS at -30°C. For all regimes, the cloud ice occurrence-frequency in the southern high latitudes remains almost constant for increasing dust mixing-ratios. For the high-RH regime, the cloud ice occurrence-frequency tends to be higher, especially for the southern high latitudes for which the cloud ice occurrence-frequency is about +4% higher at the high-RH regime. For dust mixing ratios between 0.1 and 1.5 µg kg<sup>-1</sup>, the cloud ice occurrence-frequency at -30°C increase by about +5%. The highest increase is found for the northern latitudes. However, the results from the southern mid-latitudes contradict the notion that the INP activity of mineral dust is of secondary importance in the Southern Hemisphere due to low dust aerosol concentrations (Burrows et al., 2013; Kanitz et al., 2011). Nevertheless, recent studies have acknowledged that the importance of mineral dust in the southern latitudes still cannot be ruled out (Vergara-Temprado et al., 2017). Fig. 10 shows the dust-cloud-phase relationship for different UTSS and relative
- 460 humidity at -22°C. Similar to the results at -15°C and -30°C, the cloud ice occurrence-frequency is higher in the high-RH regime. For high-UTSS conditions, the dust-cloud-phase curves are in closer agreement between the Northern and Southern Hemisphere. Overall, at -22°C the four latitude bands show the best agreement between Northern/Southern Hemisphere and mid-/high- latitudes. Combining the results from all mid- and high latitudes, the ice occurrence-frequency increases by about 25% at high-UTSS conditions and by about 20% at low-UTSS conditions for mixing ratios between 0.01 and 1.0 μg kg<sup>-1</sup> at

465 -22°C. This suggests that the dust mixing-ratio may explain both the day-to-day differences in cloud ice occurrencefrequency and the differences between latitudes.

In general, for temperatures between -36°C and -9°C, higher fine-mode dust mixing ratios are associated with an increasing eloud ice occurrence-frequency. The results suggest that only the lower static stability at -15°C has a strong influence on the relationship between mineral dust and cloud ice. This is may be a consequence of the dynamic component of the atmospheric

470 stability at lower temperatures (e.g., gravity waves), which is not included in the static stability parameter. At all temperatures studied, higher humidity values were associated with a higher cloud ice occurrence-frequency. For similar dust loadings, the cloud ice occurrence-frequency was found to be higher at the mid-latitudes than at the high-latitudes. However, against our expectations, for similar dust loadings the cloud ice occurrence-frequency at −15°C was higher in the Southern than in the Northern Hemisphere.

#### 475 5 Discussion

Some studies have already suggested that the lower occurrence-frequency of cloud ice in the higher latitudes may be associated with lower INP concentrations (Li et al., 2017a; Tan et al., 2014; Zhang et al., 2012). This hypothesis has been supported mainly by the spatial correlation between the dust relative aerosol frequency and the occurrence-frequency of ice clouds retrieved from satellite observations. However, evidence of the global temporal co-variability between INP and ice

480 occurrence-frequency on a day-to-day basis was lacking up to now. Furthermore, by studying the temporal correlation between mineral dust and cloud ice occurrence-frequency it is possible to extract new information about the differences in cloud glaciation at different latitudes and to connect these differences to previous studies of heterogeneous freezing. Particularly, our results may be used to evaluate our current knowledge of the global differences in the mineralogy of dust aerosol and its freezing efficiency.

#### 485 5.1 North-South contrast

We have found that the ice occurrence-frequency can vary at different latitudes even for similar mixing-ratios of mineral dust. This could be explained by differences in the mineralogical composition of the mineral dust aerosol at the Southern and Northern Hemisphere. It has been suggested that the freezing efficiency of clay minerals from the Northern Hemispheres (composed mostly from Illite and Smectite minerals) can be well represented by the mineral Montmorillonite while the Southern clay minerals are better represented by the mineral Kaolinite (Claquin et al., 1999; Hoose et al., 2008). The freezing efficiency of Kaolinite and Montmorillonite are known for the immersion and contact freezing mode (Diehl et al., 2006; Diehl & Wurzler, 2004). Following this simplification, the immersion freezing rates at -30°C would be 300 times higher in the Northern than in the Southern Hemisphere. This could explain the higher ice occurrence-frequency in the Northern Hemisphere relative to the Southern Hemisphere for similar dust mixing-ratios at -30°C. Below -25°C, the contact

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- 495 freezing is expected to dominate over immersion freezing. However, for contact freezing between -25°C and -16°C the freezing rate is similar for Kaolinite and Montmorillonite. This again may explain why the ice occurrence-frequency in the Northern Hemisphere is only slightly higher for similar dust mixing-ratios at -22°C. Finally, between -15°C and -4°C, the contact freezing efficiency of Montmorillonite is slightly higher than for Kaolinite. However, this fails to explain the higher ice occurrence-frequency found in the Southern Hemispheres at -15°C. Nevertheless, at such high temperatures, other dust
- 500 minerals like feldspar mineral are much more efficient as ice nucleating particles than clay minerals (Atkinson et al., 2013). Moreover, it could be that the effect of such feldspar minerals dominates over the effect of clay minerals at high temperatures. Indeed, such efficient minerals are believed to be quickly depleted trough heterogeneous freezing, so that only few would reach lower temperatures. Therefore, they are likely more relevant at temperatures above -20°C, where the immersion efficiency of clay minerals quickly decay (Boose et al., 2016; Broadley et al., 2012; Murray et al., 2011). If
- 505 feldspar minerals do dominate the heterogeneous freezing due to mineral dust above -20°C, then the higher cloud ice occurrence-frequency in the Southern Hemisphere may be due to a higher fraction (or higher efficiency) of feldspar minerals in the southern dust particles. Some evidence for this has been already found by comparing the immersion freezing efficiency of dust particles from different deserts worldwide (Boose et al., 2016). In these results, the immersion efficiency of dust particles lays mostly between Kaolinite and K-feldspar. The dust samples from sources in the Southern Hemisphere
- 510 (Australia, Etosha and Atacama milled) have a higher freezing efficiency than most of the samples from the Northern Hemisphere sources including Saharan sources for temperatures below -24°C. Although only four of these samples were studied for higher temperatures, between -23°C and -11°C it was again a sample from the Southern Hemisphere (Atacama milled) which exhibited the highest freezing efficiency. Assuming that the higher freezing efficiency of the southern dust sources may be extrapolated to temperatures above -20°C, the higher immersion efficiency of southern mineral dust,
- 515 possibly due to higher feldspar fractions, may explain the higher ice occurrence-frequency in the Southern Hemisphere at  $-15^{\circ}$ C. The highly efficient particles, most likely feldspar minerals, would be quickly depleted at temperatures around  $-15^{\circ}$ C and would therefore not interfere with the Kaolinite-Illite(Montmorillonite) differences at  $-30^{\circ}$ C. Furthermore, such a depletion of highly efficient INP during the transport of dust aerosol may also explain the higher ice occurrence-frequency at the mid-latitudes compared to the high-latitudes for similar mixing-ratios of mineral dust, especially at higher temperatures.
- 520 The ageing (e.g., internal mixing with sulfate or "coating") of dust particles may also reduce the freezing efficiency of dust aerosol during the transport from low to high latitudes. The hypotheses explaining the differences in the freezing behaviour of dust between the Northern and Southern Hemisphere are summarized in Table 2.

#### 5.2 Assumptions and uncertainties

525 In the analysis presented above, certain assumptions were made to assess the potential effect of mineral dust on cloud thermodynamic phase. In this section, these assumptions and the uncertainties that arise from them, as well as the subsequent limitations of the resulting interpretation will be discussed.

Concerning the vertical resolutions of the different products, the choice of 3 K bins is based on the resolution of the CPR (480 m) — used in the DARDAR-MASK product — and the original 3 K bins of the CALIPSO-GOCCP product. Using a

- 530 coarser vertical resolution (e.g., 6 K bins) would hinder the assessment of the role of dust as INP. For example, a decrease of <u>3 K in temperature is roughly equivalent to a fivefold increase in INP concentrations (e.g., Niemand et al., 2012). Because at</u> the mid- and high-latitudes the typical standard deviation of the day-to-day dust mixing-ratio corresponds to roughly a fourfold increase from the mean (See supplement figure S.5), we expect that the variability of dust loading should dominate over temperature variations, given a temperature constraint of 3 K or less. The statistical distribution of the phase ratio also
- 535 limits the resolution options. The cloud phase values for single pixels in the DARDAR and GOCCP products are binary (1 or 0). Therefore, a minimal sample size is required for the averaged cloud phase ratio within a certain temperature range, gridbox and percentile of dust to achieve a normal distribution along time and space, which allows interpreting the correlation with dust loading directly. For this reason, temperature bins smaller than 3 K result in a less normally-distributed cloud phase ratio.
- 540 As mentioned in section 3.5, we excluded the seasonal component of the dust-cloud-phase correlation by calculating the deciles independently for each month of the year. However, shorter cycles (e.g., weather variability) may still have an influence in the variability of dust and cloud phase. Although the cloud phase may be affected by such cycles (e.g., more liquid clouds at convective fronts and more cirrus clouds at the detrainment regions), it is still possible to distinguish between dusty and non-dusty conditions at each point of the weather cycle. Therefore, once we average over the weather
- 545 <u>cycle using monthly means inside each dust percentile we expect the dust-cloud-phase relationship to be dominated by</u> <u>the microphysical effect of dust on cloud phase.</u>

Despite the long period (2007-2010) used in the study, a significant fraction of the 5-dimensional space used for our analysis (10 dust deciles, 12 months, 15 temperature bins, 96 latitudes, and 12 longitudes) is sparsely sampled or even contains missing values. In the high-latitudes, a sampling bias exists towards the respective winter seasons because very few night-

- 550 time retrievals are available in summer. However, the seasonal variability was not found to be a dominating factor in the day-to-day impact of dust mixing-ratio on the FPR (See S.19 in the Supplement to this article). Furthermore, many factors may contribute to higher standard deviations for the ice occurrence, including:
  - Changes in dynamical forcing (e.g., updrafts) and cloud regimes
  - Temperature changes after cloud glaciation (e.g., latent heat release)
  - Ice sedimentation from above (cloud seeding), and INPs other than dust
    - Cloud vertical distribution within the studied temperature ranges

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- Turbulence favouring aerosol mixing and sub-grid temperature fluctuations
- Differences in dust mineral composition, electric charge and/or size
- Coatings (e.g. Sulfate) affecting aerosol solubility and freezing efficiency
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#### • Impact of INPs other than dust

• Subsetting of the data (e.g., only night-time retrievals)

Additionally, some issues arise from the coarse spatial resolution used in our study. A high dust mixing-ratio simulated in a gridbox indicated as *cloudy* by the satellite observations does not ensure that the dust is actually mixed with the cloud. The subgrid-distribution of dust relative to the exact cloud position remains unresolved. Higher dust mixing-ratios should be interpreted as <u>an</u> indicator foror a higher probability that a significant amount of dust was mixed with a collocated cloud. This mixing may have happened during or before the observation by the satellite. The latter <del>assumes that is only true if</del> both the cloud and the dust followed the same trajectory <u>up to the moment of the observation</u>. Overall, at coarse resolutions, the combination of modelled dust concentrations with satellite-retrieved cloud properties cannot guarantee the mixture of aerosol and clouds (R. Li et al., 2017b). Similarly, the atmospheric parameters obtained from the reanalysis may not match the conditions for the exact position of the clouds in the satellite retrievals. However, the atmospheric parameters are expected to

- match inon average the large-scale conditions influencing the aerosol-cloud interactions. In general, we expect that the assimilation of the total AOD from MODIS in the MACC reanalysis produce a fair estimation of the large-scale aerosol conditions on a day-to-day basis. At least for the Northern Hemisphere, this has been already validated with in situ measurements (Cuevas et al., 2015). As for the consistency among the different reanalyses, both the
- 575 <u>ERA-Interim and the MACC reanalysis are based on the IFS model and use a similar assimilation algorithm. Among the</u> different satellite products, both CALIPSO-GOCCP and DARDAR-MASK rely on CALIOP to determine the presence of clouds. Nevertheless, the reader should be aware that several uncertainties remain, for example, between the meteorology in the reanalysis and in the real atmosphere, particularly on the sub-grid scale. In the worst-case that the reanalyses are entirely inconsistent with the retrievals of cloud phase, we expect the result would be the lack of correlation between dust and the ice
- 580 <u>occurrence (Fig 8-10). In other words, given the large dataset included in the study, we expect that mismatches between</u> reanalysis and cloud retrievals would cause an underestimation — and not an overestimation — of the dust-cloud-phase <u>correlation</u>.

<u>Concerning the interpretation of our results</u>, it cannot be ruled out that the increase in ice cloud occurrence in the Southern Hemisphere for higher dust loading arises from other types of INP such as biogenic aerosol (Burrows et al., 2013;

585 O'Sullivan et al., 2018; Petters and Wright, 2015) or background free-tropospheric aerosol (Lacher et al., 2018), which could be misclassified as mineral dust in the reanalysis. Similarly, a possible correlation between ice cloud occurrence and the atmospheric conditions leading to the emission and transport of mineral dust should be further investigated (e.g., dusty air masses from land are usually warmer and drier). Another interesting explanation of the results presented in this study could involve the mixing of mineral dust particles with ice nucleation active macromolecules (Augustin-Bauditz et al.,

- 590 2016). Such particles are in the size of few 10 nm (Fröhlich-Nowoisky et al., 2015) and would therefore not be detected if mixed with dust aerosol. Furthermore, biases such as the overestimation of the fine-mode dust aerosol in the MACC reanalysis (Ansmann et al., 2017; Kok, 2011) may shift the mixing-ratios shown in Sect. 4.4. However, as long as such biases are not limited to certain meteorological conditions, the cloud phase averaged inside each dust decile should remain unaffected.
- 595 <u>In general</u>, meteorological parameters have a larger impact on cloud properties than aerosols do <u>Different</u> meteorological(Gryspeerdt et al., 2016). For example, different updraft regimes can change the aerosol-cloud interactioninteractions in warm clouds by an order of magnitude. <u>Therefore, it is important to study how such meteorological</u> parameters relate to the dust aerosol loading. With this purpose, Fig. 11 shows the mean relative humidity, cloud height and large-scale updraft at -15°C for the different fine-mode dust mixing-ratio deciles and <u>for</u> the four latitude bands studied in
- 600 Sect. 4.4. <u>Firstly</u>, the correlation between fine-mode dust mixing-ratio from the MACC reanalysis and the RH from the ERA-Interim reanalysis (<u></u>weighted by cloud volume fraction) <u></u>was found to be negative (Fig. 11a). The decrease in RH at higher dust mixing ratios could be associated with drier air masses coming from land. If higher dust mixing ratios are indeed associated with ice production, then the depletion of water vapor due to the growth of ice particles could also explain the correlation. We expect, however, that the impact of the decreasing RH would rather lead to an underestimation of the role of
- 605 dust mixing-ratio in eloud glaciation. We note that the RH from the ERA-Interim reanalysis represents the conditions at a large-scale and not the conditions at a specific location and the moment of the interaction between dust aerosol and supercooled cloud droplets. Because of this issue, the role of subgrid RH variability for the results cannot be assessed. Still, this relationship is consistent with the intuition that dust is mostly associated with drier air masses. Second, The
- significant positive correlation found between dust aerosol mixing-ratio and the height of the isotherms (weighted by cloud volume fraction) is alsopoints to an important source of uncertainty (Fig. 11b). Shifts in the isotherm height, either due to warm dry continental air masses carrying high dust loadings, or the heating effect of absorbing dust aerosol, may indeed eause a dynamic response of the atmosphere leading to higher isotherms. This could eausebe due to clouds to bebeing detected in a higher temperature bin after being glaciated at lower temperatures, thus erroneously suggesting an enhanced glaciation occurrence frequency at higher temperatures. Therefore, it is crucial for future studies to take into account this possibility when studying the occurrence of ice clouds at a certain isotherm.
- It is possible to find cases where the reanalysis and the detected clouds have different temperatures. Nevertheless, a systematic bias towards lower or higher temperatures would rather have an influence on the average FPR and not on the day-to-day variability of FPR. Furthermore, if absorption of solar radiation by dust acrosol were to cause the observed increase in isotherm height, we would expect a greater correlation in the northern mid-latitudes. The influence of dust-loaded air masses
- 620 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. More details on the spatiotemporal variability of the cloud height can be found in the supplement (S12) to this article.

Perhaps the largest difficulty of attributing the observed changes in cloud glaciation to variability in dust concentration is the positive correlation found between dust and the vertical velocity. **Fig. 11e** shows the large-scale vertical velocity from the

- 625 MACC reanalysis against the fine-mode dust deciles at -15°C. Stronger updrafts are often associated with higher relative humidity. Saturation over liquid water is a necessary condition for the immersion freezing mode, which is believed to be the most important freezing mode in cloud glaciation, especially at temperatures above -25°C. showed that using the monthly values of the CALIPSO GOCCP product, the FPR at -10, -20 and -30°C is positively (spatially) correlated with the large-scale vertical velocity (at 500 hPa) from the ERA Interim reanalysis product. The same study showed by using the CALIOP
- 630 level 2 aerosol layer product that this correlation is similar for different relative aerosol frequencies (dust, polluted dust and smoke combined). However, in the same study, the increase in FPR for higher vertical velocities (stronger updrafts) was found to be higher for lower relative aerosol frequencies. Furthermore, stronger Lastly, Fig. 11c shows a positive correlation between the fine-mode dust and the large-scale vertical velocity from the MACC reanalysis at -15°C. Updrafts favour saturation over liquid water and therefore CCN activation, droplet growth and inhibition of the WBF (Wegener–Bergeron–
- 635 Findeisen) process. These processes would cause Therefore, a positive dust-updraft correlation could lead to an underestimation of the dust-cloud-phase relationship.

In summary, much of the co-variability between dust, humidity, updrafts, temperature and cloud ice occurrence-frequency is still poorly understood. However, we expect that the constrains on humidity and static stability minimized most of the biases discussed in this section.

#### 640 6 Conclusions

For the first time, the MACC reanalysis was combined with satellite-retrieved cloud thermodynamic phase to investigate the potential effect of mineral dust as INP on cloud glaciation on a day-to-day basis at a global scale. Satellite products of cloud thermodynamic phase for the period 2007-2010 were included. We focused on clouds observed at night-time in the mid- and high latitudes. Our main findings can be summarized as follows:

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- Between -36°C and -9°C, day-to-day increases in fine-mode dust mixing-ratio (from lowest to highest decile) were mostly associated with increases in the day-to-day cloud ice occurrence-frequency (FPR) of about 5% to 10% in the mid- and high- latitudes.
  - 2. The response of cloud ice occurrence-frequency to variations in the fine-mode dust mixing-ratio was similar between the mid- and high- latitudes and between Southern and Northern Hemispheres. Moreover, increases in FPR from first to last dust decile were also present in the northern and southern high-latitudes, even though dust aerosol is believed to play a minor role in cloud glaciation in the Antarctic region.
    - 3. Using constraints on atmospheric humidity and static stability we could partly remove the confounding effects due to meteorological changes associated with dust aerosol.

- 4. The results also suggest the existence of different sensitivities to mineral dust for different latitude bands. The north-south differences in ice occurrence-frequency for similar mineral dust mixing-ratios agree with previous studies on the mineralogical differences between Southern and Northern Hemisphere. A larger fraction of feldspar in the Southern Hemisphere could explain the differences at -15°C, and the higher freezing efficiency of Illite and Smectite (more abundant in the Northern Hemisphere) over Kaolinite (more abundant in the Southern Hemisphere) could explain the differences at -30°C.
- We believe these new findings may have an important influence on improving the understanding of heterogeneous freezing and the indirect radiative impact of aerosol-cloud interactions. The authors hope that the results of this work will also motivate further research, including field campaigns in remote regions to study the day-to-day variability of cloud thermodynamic phase and the role of mineral dust in ice formation, satellite-based studies of associated changes in the radiative fluxes, and modelling studies to test the representation and relevance of specific processes involved in ice
- 665 formation and mineral dust transport. Such studies could help to further improve our understanding of the influence of mineral dust on cloud glaciation and the climate system.

#### 7 Author contribution

DV, IT, BH and PS contributed to the design of the study. DV processed the datasets, performed the analysis, designed the figures and drafted the manuscript. All authors contributed valuable feedback throughout the process. All authors helped with the discussion of the results and contributed to the final manuscript.

#### 8 Competing interests

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The authors declare that they have no conflict of interest.

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