We would like to thank the reviewers for their very helpful and constructive feedbacks. Point-by-point responses to each of the reviewer's comments are provided below. Our replies are highlighted in bold while the original comments are italicized. All authors have read and agreed to the revised version.

Interactive comment on "Ice Particle Production in Mid-level Stratiform Mixed-phase Clouds Observed with Collocated A-Train Measurements" by Damao Zhang et al.

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The underlying manuscript of Zhang et al. is a follow-up publication in a series from the first author using combined CALIPSO and Cloudsat observations to investigate the structure of stratiform mixed-phase clouds on a global scale. The approach has been extended with respect to the previous studies by taking MODIS-retrieved LWP as additional constraint of microphysical properties of the observed clouds.

The presented results are of substantial value for the atmospheric science community, given that the study suggests the presence of a considerable hemispheric contrast in the ice formation efficiency in stratiform mixed-phase clouds. In principle, the dataset seems to be well characterized and the data analysis methods can be considered mature, considering that several related studies were published by Zhang et al. since 2010.

Consequently, there are only a few critics points with respect to the technical implementation of the study. Nevertheless, the implied relevance of the results and the conclusions drawn are way beyond the data basis provided by the authors. This is a major issue. Basically, the message of the dataset is clear -> There is much less ice observed in stratiform mixed-phase clouds in the southern hemisphere. However, the conclusions are too linearly pointing toward aerosol effects. More efforts should be put by the authors on either supporting their strong conclusion, or, on providing a more brought discussion that includes other effects besides the aerosols. More details will follow below in the itemized review comments.

Author Response: We thank the reviewer for the very helpful comments; we carefully revised the manuscript according to the reviewer's comments as presented below.

Major comments:

Technically:

1) What is the equation for the relationship between N_ice and Z? It should be presented because this relationship is discussed quite often. Which other parameters go into this equation? What is their role in the determination of Z? E.g., particle size. How much would particle size need to vary in order to explain the observed reflectivity differences? This is likely only a few percent due to the D⁶ relationship. May there be any ice growth processes that could explain

such a slight hemispheric difference in the crystal size? What if the cloud height and thus the pressure level of cloud formation varies regionally and seasonally? See, for example, Chapter 13.3. of the book of Pruppacher&Klett, 1997 (Fig. 13-29) that presents that the diffusional growth rate of ice crystals varies by up to 100% between pressures of 1000 mb and 500mb.

Author Response: We thank the reviewer for the very constructive comments. It is quite challenging to retrieve ice number concentration (N_{ice}) from radar reflectivity (Z_e) measurements. Zhang et al. (2014) developed a method to estimate N_{ice} in stratiform mixed-phase clouds by using combined Z_e measurements and 1-D ice-growth model simulations. Cloud top temperature, liquid water path (LWP), and vertical air motion are required as inputs in their algorithms and sensitivity tests show that they all have important impacts on N_{ice} estimations. Due to large uncertainties in the MODIS-derived LWP for mixed-phase clouds (Adhikari and Wang 2013), N_{ice} estimations in mixed-phase clouds using A-Train satellite measurements are not available at this stage. We added this discussion to page 6 lines 15-20 in the text.

To account for the observed Z_e differences of up to 8 dBZ in mid-level stratiform mixed-phase clouds and assuming N_{ice} does not change, ice particle size needs to vary approximately 35% for a given cloud top temperature and similar narrow LWP range. We thank the reviewer for pointing out that atmospheric pressure might have systematic impacts on the ice diffusional growth. Takahashi et al. (1991) show that the mass growth rate at 860 mb is approximately 30% larger than at 1010 mb due to the variation of diffusivity of water vapor in air with the pressure difference. However, from Figure 20 in their paper, the mass growth difference due to pressure difference is much smaller than that due to temperature deference. Within the Rayleigh scattering regime, radar reflectivity is proportional to the square of ice crystal mass. Therefore, the 30% difference in mass causes approximately a 2 dBZ difference in Z_e . As suggested by the reviewer, we investigated the regional and seasonal differences of atmospheric pressure at given subfreezing temperatures using four years of the ECMWF-AUX product between 2006 and 2010.



Atmospheric pressure profiles with subfreezing temperatures for the six latitude bands (a) and their seasonal variations (b).

From the above figure (a), hemispheric differences in atmospheric pressure profiles are negligible over mid- and low-latitude bands, and range from 40 mb to 140 mb over high latitudes. Therefore, pressure-level differences have negligible contribution to the hemispheric ZL differences over mid- and low latitude bands, and contribute less than 2 dBZ to the observed hemispheric ZL differences over high latitudes. Low-latitude bands have about 140 mb and 280 mb lower pressure than mid- and high-latitude bands, respectively, indicating the that ZL differences between low latitudes and mid- and high latitudes will be 1-2 dBZ larger after removing pressure difference effects. As shown in figure (b), northern mid- and high latitude atmospheric pressure profiles have 220 mb and 160 mb seasonal variations; while southern latitudes and northern low-latitudes have negligible seasonal variations. Overall, regional and seasonal variations of atmospheric pressure profiles cause less than 2 dBZ variations in ZL and have small impacts on our conclusions. We added the above figure in the revision (new Fig. 4) and added a discussion of the atmospheric pressure regional and seasonal differences and their impacts in the manuscript (pages 7-8).

References:

- Adhikari, L., and Wang, Z.: An A-train satellite based stratiform mixed-phase cloud retrieval algorithm by combining active and passive sensor measurements, British Journal of Environment and Climate Change, 3(4), 587-611, 2013.
- Zhang, D., Wang, Z., Heymsfield, A. J., Fan, J., and Luo, T.: Ice Concentration Retrieval in Stratiform Mixed-Phase Clouds Using Cloud Radar Reflectivity Measurements and 1D Ice Growth Model Simulations, J. Atmos. Sci., 71(10), 3613–3635, doi:10.1175/JAS-D-13-0354.1, 2014.

Takahashi, T., Endoh, T., Wakahama, G., and Fukuta, N.: Vapor diffusional growth of freefalling snow crystals between -3 and -23 °C, J. Meteorol. Soc. Jpn., 69, 15–30, 1991.

2) Why is layer maximum ZL used as reference value? Why not mean or, as done by Bühl et al., 2016, the value closest to the liquid layer?

Author Response: In page 4 line 7, we pointed out that 'the CPR has an effective vertical resolution of about 480 m and is oversampled at 240 m vertical resolution'. Therefore, to remove possible Z_e measurements above the top of liquid layer, we use the maximum Z_e within 500 m below the CALIOP-detected liquid-dominated layer top as the reference value. In this revision, we emphasized the impacts of the coarse CPR resolution and oversampling technique by adding 'oversampled at 240 m from the effective vertical range resolution of 480 m' to page 5 line 24. Bühl et al., (2016) use the Z_e value closest to the liquid layer base with ground-based high vertical resolution (30 m) radar measurements. However, this is difficult with A-Train satellite measurements as the CPR has a coarse vertical resolution and the liquid layer at the top quickly attenuates CALIOP signals, preventing reliable detection of the liquid layer base. We also added this discussion to page 5 lines 19-22 in the text.

3) Figure 5: When doing hemispheric studies it is not straightforward to use seasons. Better is to use month ranges and then refer to boreal and austral seasons in the text.

Author Response: We revised the figure as suggested and refer them to boreal and austral seasons in the text.

4) P 3, L 14: "usually decoupled" is a very vague statement. How often are clouds coupled to the surface or to the planetary boundary layer? This could be easily checked by using global model datasets such as GDAS1 which also provide an estimate of the mixing layer height. It would be a good test to investigate surface effects by excluding/including cloud layers touching the atmosphere-ground mixing layer from/into the statistics. Is there a hemispheric/seasonal variability of potential surface effects?

Author Response: We thank the reviewer for the suggestions. Ice processes in atmospheric clouds are very complicated. In this study, we aim to analyze primary heterogeneous ice formation characteristics in supercooled clouds with satellite remote sensing measurements by focusing on mid-level stratiform clouds. Turbulent vertical mixing within the boundary layer has significant impacts on ice particle growth and the lifecycle of mixed-phase clouds, making it very challenging to study primary ice formation with radar Z_e measurements. Therefore, we did not include boundary layer mixed-phase clouds in this study. To make it more clear, we deleted "usually" and added "Mid-level supercooled Cloud" to emphasize the targeted clouds page 3 line 24 in this revision.

Investigating surface effects on mixed-phase clouds is important but is out of the scope of this paper. Using ground-based remote sensing, including Doppler lidar and radar spectral measurements, turbulent vertical mixing impacts on polar boundary layer mixedphase cloud ice particle growth, secondary ice generation, supercooled liquid water fraction, and cloud lifecycle are under investigation in our group. 5) P3, L24-25: Sassen et al. 2012 showed strong effects of specular reflection on the CALIPSO measurements before it was tilted to 3° off-zenith. The authors argue on P4, L21ff that this does not affect the lidar-based liquid cloud determination. Was there an actual check performed to evaluate this assumption? The signal of CALIOP is known to attenuate quickly, also under compact cirrus conditions. Figure 3 in Sassen et al, 2012 shows a dramatic change in the relationship between LDR and temperature between nadir and off-nadir pointing, especially at $T>-30^{\circ}C$.

Author Response: We agree with the reviewer that horizontally oriented ice crystals present a challenge for lidar-based liquid cloud determination because they also cause large lidar backscattering and small depolarization ratio measurements. However, ground-based lidar measurements show that horizontally oriented ice crystals have a weak attenuation of lidar signals and lidar signals can penetrate ice cloud layer up to a few kilometers (Hogan et al., 2003; Balin et al., 2011). This provides an alternative way to effectively detect liquid layer using lidar signal profile. Wang (2013, Figure 10) shows that this approach correctly determines liquid clouds in terms of layer mean depolarization ratio and integrated backscattering coefficient. In addition, collocated MODIS cloud LWP greater than 10 g/m^2 is used to guarantee the detection of a liquid-dominated layer. We added this statement in page 5 lines 8-11 in the text.

References:

- Balin, Y. S., Kaul, B. V., Kokhanenko, G. P., & Penner, I. E. (2011). Observations of specular reflective particles and layers in crystal clouds. *Optics express*, 19(7), 6209-6214.
- Hogan, R. J., Illingworth, A. J., O'connor, E. J. and Baptista, J. P. V. P. (2003), Characteristics of mixed-phase clouds. II: A climatology from ground-based lidar. Q.J.R. Meteorol. Soc., 129: 2117–2134. doi:10.1256/qj.01.209.
- Wang, Z.: Level 2 combined radar and lidar cloud scenario classification product process description and interface control document. Jet Propulsion Laboratory Tech. Rep. D-xxxx, 61 pp.

Comments regarding the argumentation:

1) How do the different data analysis methods of Zhang 2010, 2012 and the current one differ? In the current version it is only argued that the current study differs from the 2010 study by considering only single-layer clouds. But can this explain why the results are so different? I would be happy to see some more text dealing with the cross-evaluation of the different studies. Perhaps a table would help to clarify methodological differences.

Author Response: The reviewer is right that this research is a follow-up study of Zhang et al. (2010) and Zhang et al. (2012). The data analysis methods in this study are similar to that in Zhang et al. (2010) and Zhang et al. (2012). Zhang et al. (2010) present for the first time the climatology of mid-level stratiform clouds and their macrophysical properties using A-Train satellite measurements. Considering that mid-level stratiform clouds are ideal targets for studying primary ice formation and aerosol impacts, Zhang et al. (2012) quantitatively estimated dust impacts on ice production in mixed-phase clouds over the

'dust belt', a region including the North Africa, the Arabian Peninsula and East Asia. Taking advantage of improved understanding of ice diffusional growth in mixed-phase clouds as shown in Zhang et al. (2014) and improved thin dust layer detection with CALIOP by Luo et al. (2014), this study further uses A-Train satellite measurements to provide a global statistical analysis of ice production in mid-level stratiform mixed-phase clouds. We introduced Zhang et al. (2010) study in line 33 and Zhang et al. (2012) study in line 20 on page 3.

2) It should be noted that strong hemispheric differences in het. ice formation efficiency were already presented by Kanitz et al., 2011. Since the way of data analysis is similar to the presented study it would be worth mentioning it.

Author Response: We thank the reviewer for the very helpful comment. We added discussions of Kanitz et al. (2011) study on page 8 lines 5-8.

3) P7, L4-13: There are a lot of statements given in this paragraph. "crystals reside longer", "grow larger by the WBF process", "larger ice growth rate by accretion". Are there references available supporting these statements?

Author Response: Direct observation of ice diffusional growth in clouds is difficult to obtain. Zhang et al., developed 1-D ice growth model to simulate ice diffusional growth in clouds and investigate LWP impacts. These statements are based on our fundamental understanding of the ice diffusional growth process and the 1-D ice growth model simulations. In this revision, we added the reference to Zhang et al. (2014) in the text.

Reference:

Zhang, D., Wang, Z., Heymsfield, A. J., Fan, J., and Luo, T.: Ice Concentration Retrieval in Stratiform Mixed-Phase Clouds Using Cloud Radar Reflectivity Measurements and 1D Ice Growth Model Simulations, J. Atmos. Sci., 71(10), 3613–3635, doi:10.1175/JAS-D-13-0354.1, 2014.

4) I personally strongly support the conclusion given on P8, L16ff. However, are there really no other effects beside aerosol properties that should be discussed? I strongly recommend to at least point to the possibility that the difference in the reflectivities can be either attributed to large changes in the number OR to very small changes in the size of the ice crystals. The evolution of ice crystals depends on a multitude of constraints. . . just take a look into Pruppacher&Klett, 1997 or into modeling studies of mixed-phase clouds, such as the ones of Ann Fridlind or Morrisson et a. 2012. Also the studies of Korolev and/or Field show that cloud dynamics can have a strong effect on the evolution of the ice crystals. Constraining LWP is already a great leap forward, but other environmental parameters such as cloud pressure or the relationship between the clouds and the planetary boundary layer are just a few examples of possible additional factors. Consider also, as another example: Average CCN concentrations in the atmosphere over the Southern-hemispheric (SH) Oceans are only one fifth of the northern-hemispheric values (Yum et al. 2004). Assuming constant cloud depth and liquid water path, much fewer but much larger droplets can be expected in the SH clouds. Heterogeneous freezing parameterizations (especially immersion freezing) rely mainly on temperature and aerosol

properties but not on droplet size (See, e.g., Demott et al., 2010). Thus, having much less droplets available for ice nucleation will result in correspondingly lower ice crystal concentrations, even in the absence of any aerosol effect. This pathway could also contribute to the apparently less efficient ice formation over the SH. There are still a lot of unknowns that need to be resolved before we can actually pin down the observations solely to aerosol effects. I'm looking forward to a lot of future studies dedicated to this key question of current atmospheric research.

Author Response: We thank the reviewer for the very constructive comments. We pointed out that "differences in Z_e can be either attributed to large changes in ice number concentration or small changes in ice crystal size" in page 6 lines 8-10. In this study we carefully isolate factors that impact ice diffusional growth and particle sizes by focusing on mid-level stratiform mixed-phase clouds with same CTT and similar LWPs. We also analyzed atmospheric pressure impacts as suggested in the Technical comment #1. The motivation for targeting mid-level stratiform mixed-phase clouds include that they are not affected by strong turbulent vertical mixing within the boundary layer and have less complex dynamic environment and straightforward ice growth trajectory. We are aware that cloud dynamics have a strong effect on the evolution of the ice crystals, making it very challenging to study primary ice formation with radar Z_e measurements. Therefore, we did not include boundary layer clouds in this study.

We agree with the reviewer that Southern-hemispheric Oceans have much lower CCN concentrations compared with low latitudes. However, the cloud droplet concentration is still several orders of magnitude higher than typical ice number concentrations and therefore the low CCN concentration condition is expected to have small impact on ice nucleation in cloud.

We agree with the reviewer that there are still a lot of unknowns related to ice processes in clouds and we toned down statements in the summary section.

Minor comments:

P2, *L* 16: An impressive demonstration of the lifetime effect as a function of temperature is also given by Bühl et al., 2016.

Author Response: We thank the reviewer for this helpful comment. We added discussion of the Bühl et al. (2016) study to page 2 lines 23-25 in the text.

Anonymous Referee #2

In their paper, Zhang et al. analyse collocated CloudSat and CALIPSO lidar measurements between 2006 and 2010 to study ice number concentration in stratiform mixed-phase clouds. They divide the global data set into six latitude bands (northern and southern tropical, mid- and high-latitudes) for their analysis. In general the paper is well written and the results are of interest to the community. However, the method needs further explanation and the analysis/interpretation should take into account differences in the macro- and microphysical properties of Arctic and mid-latitude clouds. The paper needs major revision before it can be published in ACP. Page and line number below refer to the document uploaded by the authors.

Author Response: We thank the reviewer for the helpful comments. We carefully revised the manuscript according to the reviewer's comments as presented below.

Comments:

In the introduction the authors should discuss the specific characteristic of Arctic mixed-phase clouds (e.g. observation, life time and limited CCN). For example the observed CCN concentrations in Arctic mixed phase clouds are usually of the order of 10 cm-3 (rarely as high as 100 cm-3) and sometimes less than 1 cm-3 (Birch et al. 2012). This is in contrast to lower latitudes where typical concentrations range from approximately 100 cm-3 to several 1000cm-3 in the marine environment (Raes et al., 2000). Such low CCN number concentrations affect cloud droplet size spectra, and hence, radiative properties of these clouds will differ from those at mid-latitudes. Mauritsen et al. (2011) argue that cloud formation is frequently limited by CCN availability in the central Arctic. They use the term "tenuous cloud regime" to describe situations in between an abundance of aerosol needed to form clouds and a hypothetical situation where aerosols are absent and cloud formation does only occur at very high supersaturation (a[']Lij400% relative humidity). Further, Arctic mixed-phase clouds are governed by a combination of local and large-scale processes (Morrison et al., 2012). At the small scale, the Wegener-Bergeron-Findeisen (WBF) process is one of the main mechanisms responsible for ice crystal growth at the expense of super-cooled water droplets (Bergeron, 1935; Findeisen, 1938; Wegener, 1911). Such a mechanism leads to a rapid glaciation of mixed-phase clouds. On the other hand, dynamical processes, such as turbulence or entrainment may facilitate the formation of new super-cooled water droplets. For example, resupply of water vapour from the surface or from entrainment of moisture from above the clouds may contribute to the continuous formation of liquid droplets. The coupling of various processes is, thus, necessary to maintain the unstable equilibrium between liquid droplets and ice crystals within Arctic mixed-phase clouds (Mioche et al 2015). This may explain the long lifetime of Arctic mixed-phase clouds, which can last up to several days or weeks. (Shupe, 2011; Verlinde et al., 2007; Morrison et al., 2012). Previous studies of Korolev et al. (2003), Korolev and Isaac, (2003) and Korolev (2007) also point out that the lifetime of Arctic mixed-phase depends on local thermodynamical conditions or is linked to cloud dynamics. Local and long-range dynamic processes (aerosol, heat and moisture transport) also have a significant impact on Arctic mixed-phase cloud formation and properties (Cesana et al. 2012, Morrison et al. 2012).

Author Response: We thank the reviewer for the very constructive comments. As suggested, we added a several sentences in the introduction part to discuss the specific

characteristics of polar mixed-phase clouds on page 2 lines 17-25.

Page 5, line 129-133. Why do you use max Ze and not mean Ze as Zhang et al. (2014)? Your results in Figure 3b (90-60 N) are similar to Figure 10 in Zhang et al. (2014) but shifted to larger values. What is the effect of using max or mean on the relationship between Ze and ice number concentration? Is Ze normally distributed to allow for a retrieval of a relationship to aerosol concentrations and the subsequent statistical analysis?

Author Response: We thank the reviewer for figuring out the differences. On page 4 line 7, we pointed out that 'the CPR has an effective vertical resolution of about 480 m and is oversampled at 240 m vertical resolution'. Therefore, to remove possible Z_e measurements above the top of liquid layer, we use the maximum Z_e within 500 m below the CALIOP-detected liquid-dominated layer top as the reference value. Zhang et al. (2014) use Z_e profiles from ARM ground-based radar measurements, which have a much higher vertical resolution of 45 m. Therefore, layer-mean Z_e is used in their study in order to be better compare with the 1-D ice growth model simulations. Another possible cause for mean Z_e profile differences is that the CPR has a sensitivity of ~ -29 dBZ, while the ARM ground-based millimeter-wavelength radar has a higher sensitivity of ~ -60 dBZ, as shown in the figure below.



Stratiform mixed-phase cloud distributions in terms of CTT and Z_e over the NSA Barrow site from: CloudSat CPR measurements (left) and ARM NSA KA-band cloud radar (KAZR) measurements (right). The distributions are normalized at each CTT.

The reviewer is right that the distribution in Figure 3 is normalized at each cloud top temperature bin. We pointed this out in the caption of Figure 3.

Page 6, line 156 to 160: Does the selected LWP range (20 to 70 gm-2) have an effect on the statistics for high-latitude clouds? Arctic mixed-phase clouds usually peak at lower LWP (Tjernström et al., 2012).

Author Response: We agree with the reviewer that Arctic mixed-phase clouds usually peak at low LWP compared with low latitudes. In this study, we chose the narrow LWP range centered on the LWP distribution peak to reduce the impacts of LWP variations on measured Z_e and also to ensure enough samples are obtained. As long as the LWP range is same for all latitude bands at a given cloud temperature range, ice crystal growth in midlevel stratiform mixed-phase clouds are similar. As for getting enough samples, although the selected LWP range (20 to 70 g/m²) is slightly off-peak of LWP distribution for Arctic mixed-phase clouds, we still have abundant samples because Arctic has high occurrence frequency of mixed-phase clouds. Therefore, the selected LWP range does not have an important impact on the statistics for high-latitude clouds.

Page 6 six latitudes bands: Can you please repeat your analysis for an Arctic latitude band (> 70°). Figure 1 shows the highest occurrence of mixed-phase clouds over the ocean in the Arctic and Antarctic (> 70° and <-70°) while Figure 7 shows aerosol occurrence beyond 70° that is lower than at latitudes below 70° in spring. Your results might be biased by your choice of latitude band, i.e. the results for the latitude bands 60-90° might be dominated by the signals from between 60 and 70°. In other words: I don't think that there is so much dust in the Arctic that it could have such a strong effect in the clouds (see comments regarding the Introduction).

Author Response: We thank the reviewer for the helpful comments. We repeated analyses for Arctic latitude band (> 70°) as shown below. Compared with Figure 7 in our manuscript, this higher Arctic latitude band (> 70°) shows similar seasonal variations with maxima ZLs in spring, and also a similar magnitude of seasonal mean ZL differences.



Mid-level stratiform mixed-phase cloud distributions in terms of CTT and Z_e for Arctic latitude band (>70^{θ}) and their seasonal variations (left), and mean ZL seasonal variations (right). Winter daytime

measurements are sparse due to polar night and therefore its mean ZL is not plotted.

Indeed, elevated dust layers have approximately a 14% occurrence frequency during MAM as observed with ground-based lidar depolarization measurements at the ARM NSA Barrow site (latitude: 71.25 N), as shown in Figure 3.11 in Zhao (2011).

Reference:

Zhao, M.: The Arctic clouds from model simulations and long-term observations at Barrow, Alaska, PhD thesis, Univ. of Wyo., Laramie, 2011. https://search.proquest.com/docview/1354441520

Page7, line 221-222, supercooled cloud fraction, [] lowest during springtime: Is the occurrence frequency of mixed-phase clouds according to Figure 5 not the highest in spring at high-latitudes?

Author Response: The occurrence frequency of mixed-phase clouds relative to all liquidcontaining supercooled clouds is the highest in spring at northern high-latitudes according to Figure 5 (now Figure 6). In Choi et al. (2010), supercooled cloud fraction (SCF) is derived as the ratio of liquid-containing supercooled clouds to all clouds at a given temperature range between -40 and 0 °C. Reductions in SCF indicate increases of glaciated cloud fractions. They therefore argue that strong correlations between mineral dust occurrence and reduction in SCF suggests that elevated mineral dust particles effectively glaciate supercooled clouds by providing abundant ice nucleation particles (INPs). Strictly, the definitions of SCF and occurrence frequency of mixed-phase clouds are not the same and should not be directly compared. We deleted "consistent with the results in Fig. 5 and 6 in our study" in the text.

Page 7, line 187, delete greater

Author Response: We deleted it as suggested.

Ice Particle Production in Mid-level Stratiform Mixed-phase Clouds

Observed with Collocated A-Train Measurements

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10 Abstract. Collocated <u>A-Train</u> CloudSat radar and CALIPSO lidar measurements between 2006 and 2010 are analyzed to study primary ice particle production characteristics in mid-level stratiform mixed-phase clouds on a global scale. For similar clouds in terms of cloud top temperature and liquid water path, Northern Hemisphere latitude bands have layer-maximum radar reflectivity (ZL) that is ~1 to 8 dBZ larger than their counterparts in the Southern Hemisphere. The systematically larger ZL under similar cloud conditions suggests larger ice number concentrations in mid-level stratiform

15 mixed-phase clouds over the Northern Hemisphere, which is possibly related to higher background aerosol loadings. Furthermore, we show that northern mid- and high Jatitude springtime has ZL that is larger by up to 6 dBZ (a factor of 4 higher ice number concentration) than other seasons, which might be related to more dust events that provide effective ice nucleating particles. Our study suggests that aerosol-dependent ice number concentration parameterizations are required in climate models to improve mixed-phase cloud simulations, especially over the Northern Hemisphere.

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1. Introduction

Ice particle production in a supercooled liquid cloud has dramatic impacts on the cloud's radiative properties, precipitation efficiency, and cloud lifetime due to distinct differences in particle sizes, shapes, fall velocities, and refractive indexes between liquid droplets and ice crystals (Sun and Shine, 1994; de Boer et al., 2011a). Such clouds significantly

- 5 impact global and regional radiation budgets (Matus and L'Ecuyer, 2017) having a global coverage of more than 34% and being particularly common at high latitudes (Shupe et al., 2011; Adhikari et al., 2012; Wang 2013; Scott and Lubin, 2016). In a mixed-phase cloud, once ice particles are formed, they grow through water vapor diffusion at the expense of liquid water because saturation vapor pressure is lower over ice than liquid. This process, known as the "Wegener–Bergeron– Findeisen (WBF) process" (Wegener, 1911; Bergeron, 1935; Findeisen, 1938), creates a thermodynamically unstable
- condition in mixed-phase clouds. In the absence of strong vertical air motions, the WBF process removes liquid droplets quickly, causing a mixed-phase cloud to glaciate completely (Korolev and Field, 2008; Fan et al., 2011). <u>Therefore, aerosol might impose a glaciation indirect effect on clouds by acting as effective ice nucleating particles (INPs) (Lohmann 2002)</u>. Global climate model (GCM) simulations show that the changing the glaciation temperature of supercooled clouds from 0 °C to -40 °C causes differences in the top-of-atmosphere longwave and shortwave cloud radiative forcing of ~ 4 W/m² and ~ 8 W/m², respectively (Fowler and Randall, 1996).

However, observations indicate that supercooled liquid water in mixed-phase clouds persists for tens of hours or even days and down to temperatures of as low as ~-36 °C (Seifert et al., 2010; Zhang et al., 2010; de Boer et al., 2011b). Over the

polar regions where mixed-phase clouds are commonly observed, cloud condensation nuclei (CCN) concentrations are usually low, on the order of 10 cm⁻³ and sometimes less than 1 cm⁻³ (Mauriten et al., 2011; Birth et al., 2012). An increase of

- 20 aerosol may enhance CCN concentrations, thereby increase cloud cover and reduce cloud droplet size. This aerosol indirect effect leads to a longer mixed-phase cloud lifetime, which is opposite to the aerosol glaciation indirect effect (Lance et al., 2011). Even more complicated, coupling of local thermodynamic conditions and large-scale dynamics contributes greatly to the long persistence of mixed-phase clouds (Korolev and Isaac 2003; Morrison et al., 2012). Buhl et al., (2016) estimated ice mass flux at mixed-phase cloud base using ground-based radar measurements and show that when temperatures are above -
- 25 <u>15 °C the water depletion due to ice formation is small and the cloud layer is very stable.</u> The WBF process in GCMs is typically too efficient, causing severe underestimations of supercooled liquid water fraction on a global scale (Cesana et al., 2015; McCoy et al., 2016). Tan et al. (2016) show that the equilibrium climate sensitivity (ECS) can be 1.3 °C higher in GCM simulations when supercooled liquid fractions (SLFs) in mixed-phase clouds are constrained by global satellite observations. Improved SLF parameterizations in GCMs requires better understanding of ice production processes in
- 30

) supercooled clouds under various dynamic environments and background aerosol conditions using extensive observations from cutting edge instruments.

Heterogeneous nucleation, which dominates ice formation in supercooled clouds at temperatures warmer than -36 °C (Pruppacher and Klett, 1997; Vali, 1996), is not well understood and parameterized in models because of the complicated

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three-phase interactions of water and the largely unknown properties of ice nucleating particles (Cantrell and Heymsfield, 2005; DeMott et al., 2011; Morrison et al., 2012). There are four well-recognized heterogeneous ice nucleation models: deposition nucleation, condensation freezing, immersion freezing, and contact freezing (Pruppacher and Klett, 1997). The immersion freezing mode, which refers to the process that an INP is immersed into a droplet at a relatively warm

- 5 temperature and freeze the droplet at a colder temperature, is suggested to be the dominant ice formation mechanism in stratiform mixed-phase stratiform clouds (de Boer et al. 2010). This mode provides a pathway for time-dependent ice production in clouds, which can be used to explain the long persistence of precipitating stratiform mixed-phase clouds (Westbrook and Illingworth, 2013). Of course, ice production in clouds also depends on the presence of INP aerosols and, for example, laboratory measurements of INP aerosol properties provide fundamental databases for developing and
- 10 improving ice nucleation parameterizations in models (DeMott et al., 2011; Hoose and Möhler 2012; Murray et al., 2012). While such databases are valuable, it is also important to observe ice nucleation processes in the real atmosphere to constrain and evaluate parameterizations on a global scale.

Observations of aerosol impacts on <u>ice production in supercooled clouds mainly come from ground-based and satellite</u> remote sensing measurements. Choi et al. (2010) and Tan et al. (2014) show that supercooled liquid cloud fraction is
 negatively correlated with aerosol occurrence (especially dust) using Cloud-Aerosol Lidar and Infrared Pathfinder Satellite
 Observation (CALIPSO) spaceborne lidar measurements. Unfortunately, because the lidar signal cannot penetrate the liquid-dominated layer at the top of mixed-phase clouds, aerosol impacts on ice production are not directly presented in their studies. Seifert et al. (2010) avoided this issue by using 11 years of grounded-based lidar depolarization measurements to study relationships between dust occurrence and ice-containing cloud fractions over central Europe. Also, Zhang et al. (2012)
 quantitatively estimated dust impacts on ice production in mixed-phase clouds using combined CALIPSO lidar and CloudSat

radar measurements over the 'dust belt', a region including the North Africa, the Arabian Peninsula and East Asia

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Our objective in this paper is to better characterize the primary heterogeneous ice production in clouds on a global scale. We focus on mid-level stratiform mixed-phase <u>clouds</u>, <u>which provide</u> a relatively simple target for studying ice generation for the following reasons. <u>Mid-level supercooled</u> clouds are, decoupled from Earth's surface and therefore are not affected by strong turbulent vertical mixing within the boundary layer. There is usually a liquid-dominated layer at the top of mid-level stratiform mixed-phase clouds (de Boer et al., 2011b; Riihimaki et al., 2012) and, when the temperature is low enough, ice particles form from liquid droplets, grow in the water-saturated environment, and fall out of the liquid-dominated layer (Fleishauer et al., 2002; Carrey et al., 2008; Zhang et al., 2014). Below the liquid-dominated cloud layer, ice crystals

continue to grow during the fall until they reach a level that is sub-saturated with respect to ice. The less complex dynamic
 environment and straightforward ice growth trajectory in mid-level stratiform mixed-phase clouds provides an ideal scenario for studying cloud thermodynamic phase partitioning and aerosol impacts on ice formation in clouds, as well as retrieving cloud microphysical properties with remote sensing measurements (Wang et al. 2004; Larson et al., 2006; Heymsfield et al., 2011; Zhang et al., 2012; 2014; Bühl et al., 2016). Zhang et al. (2010) present for the first time the climatology of mid-level stratiform clouds and their macrophysical properties using A-Train satellite remote sensing measurements. This study further

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uses four years of collocated CloudSat radar and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) lidar measurements together with other ancillary A-Train products between June 2006 and June 2010 to provide a global statistical analysis of ice production in mid-level stratiform mixed-phase clouds.

2. Dataset and Methodology

- 5 The description of the collocated A-Train measurements follows directly from Zhang et al. (2010). The main instrument on CloudSat satellite is a nadir-viewing 94 GHz Cloud **P**rofiling **R**adar (CPR)-the first spaceborne cloud radar. The sensitivity of CPR was approximately -30 dBZ during the period analyzed here. The CPR has an <u>effective vertical resolution</u> of about 480 m (oversampled at 240 m vertical resolution) and horizontal resolutions of between 1.3 and 1.4 km cross-track and between 1.7 and 1.8 km along-track (depending on latitude) (Stephens et al., 2008). The CPR can detect clouds with
- 10 large cloud droplets, or large ice crystals, or precipitating hydrometers, and provides the vertical structures of clouds (Stephens et al., 2002). The Cloud–Aerosol LIdar with Orthogonal Polarization (CALIOP) onboard the CALIPSO satellite is a near-nadir-viewing lidar with two wavelengths at 532 nm and 1064 nm with linear polarization measurements available at 532 nm (Winker et al., 2003). The CALIOP has vertical resolutions of 30 m below 8.2 km, and 60 m between 8.2 and 20.2 km. The horizontal resolutions of CALIOP are 333 m below 8.2 km, and 1000 m between 8.2 and 20.2 km. CALIOP is able
- 15 to provide global, high-resolution vertical profiles of aerosols and optically thin clouds (Winker et al., 2010). Due to differing wavelengths, the CPR and CALIOP measurements provide complementary capabilities that enable accurate detection of cloud boundaries and their vertical structures (Stephens et al., 2008). Their complementary nature is exemplified in the detection of supercooled liquid-dominated mid-level mixed-phase cloud layers, where the CPR is more sensitive to the large-sized ice crystals and the CALIOP is more sensitive to the higher number concentration of small liquid droplets (Zhang
- 20 et al., 2010). Because temperature is critical for ice formation in supercooled clouds, the European Center for Medium-Range Weather Forecast (ECMWF)-AUX product is collocated to provide temperature and pressure profiles with the same vertical resolution as CPR (Partain, 2007). In addition, MODIS on the Aqua satellite provides cloud liquid water path (LWP) determined from retrieved cloud droplet effect radius and cloud optical depth (Platnick et al., 2003). The ancillary CloudSat MODIS-AUX product that includes cloud LWP is collocated and employed in our analysis. This analysis is limited to
- 25 daytime hours since MODIS cloud property retrievals are only available when sunlit. Previous studies show that MODISretrieved LWP have a positive bias at high latitudes due to the solar zenith angle dependence in the retrieval algorithms (O'Dell et al., 2008). Through a comparison of MODIS retrievals with ground-based microwave radiometer (MWR) measurements at the Atmospheric Radiation Measurement (ARM) Facility's North Slope of Alaska (NSA) site, Adhikari and Wang (2013) show that MODIS overestimates LWP for stratiform mixed-phase clouds by 35% and 68% in the
- 30 temperature ranges of -5 to -10 °C and -10 to -20 °C respectively.

Algorithms using collocated CALIOP and CPR measurements to identify mid-level stratiform mixed-phase clouds were

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developed by Zhang et al. (2010). To summarize, candidate mid-level clouds are identified when the CALIOP-detected cloud top height is above 2.5 km from the ground level and cloud top temperature is greater than -40 °C. Of these clouds, many have a liquid-dominated layer at the top, which is detected by a strong peak in lidar total attenuated backscatter (TAB) near cloud top (i.e., layer maximum TAB greater than 0.06 sr⁻¹ km⁻¹) and a rapid attenuation of the lidar backscattering such

- 5 that the lidar-observed laver geometric depth is less than 500 m. We use the lidar TAB and rapid lidar signal attenuation to identify the presence of liquid layers, which is a method that has been widely used for liquid layer identifications from space-borne lidar measurements (e.g., Hogan et al., 2004; Zhang et al., 2010; Wang 2013). We note that horizontally oriented ice crystals can also have a large lidar TAB however they do not attenuate lidar backscattering significantly. Wang (2013, Figure 10) shows that this approach correctly determines liquid clouds in terms of layer mean depolarization ratio and
- 10 integrated backscattering coefficient. In addition, collocated MODIS cloud LWP greater than 10 g/m² is used to guarantee the detection of a liquid-dominated layer. The cloud system is identified as being stratiform when the cloud top height standard deviation is smaller than 300 m. To calculate the standard deviation, a cloud system is identified as containing at least 10 continuous cloudy profiles, which corresponds to a horizontal scale of approximately 11 km (the horizontal distance between two contiguous CPR profiles is 1.1 km). In addition, the CPR radar reflectivity factor Z_e must be smaller than 10 15 dBZ near the surface to exclude strongly precipitating mid-level stratiform clouds.

Radar measurements are used to detect the presence of ice particles in mid-level stratiform mixed-phase clouds. Cloud droplets and pristine ice crystals are much smaller than the radar wavelength so they fall within the Rayleigh scattering regime where Ze is proportional to the sixth power of the particle size. Ice crystals are typically larger than cloud droplets such that Ze is dominated by ice crystal scattering (Shupe et al., 2007). Bühl et al. (2016) use the Ze value closest to the

- 20 liquid layer base with ground-based high vertical resolution (30 m) radar measurements for studying ice particle properties. However, this is difficult with A-Train satellite measurements as the CPR has a coarse vertical resolution and the liquid layer at the top quickly attenuates CALIOP signals, preventing reliable detection of the liquid layer base. Given that the physical thickness of supercooled liquid layers at the top of mid-level stratiform mixed-phase clouds are generally smaller than 500 m and the vertical resolution of the CPR is oversampled to 240 m from the effective vertical range resolution of 480 m, we use
- 25 the maximum Ze (referred to as "ZL") within 500 m below the CALIOP-detected liquid-dominated layer top to ascertain the presence of ice particles for analysis. Using temperature-dependent ZL thresholds, Zhang et al. (2010) show that, at temperatures lower than -6 °C, approximately 83.3% of mid-level liquid-topped stratiform clouds are mixed-phased, revealing the importance of understanding their ice production. Furthermore, to exclude seeding from upper-level clouds and to enable use of MODIS column integrated LWP retrievals, only single-layer clouds detected with collocated CALIOP and CPR measurements are analyzed (Wang et al., 2012). Since we study ice production in stratiform clouds in this study, we
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focus on clouds with top temperatures within the -40 °C to 0 °C range.

To illustrate the importance of understanding ice production in these clouds, Fig. 1 shows the global distribution of single-layer mid-level stratiform cloud occurrence during daytime based on four years of collocated CALIOP and CPR

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measurements. The occurrences are smaller than what are presented in the Fig. 3 in Zhang et al. (2010) because we only focus on single-layer <u>supercooled stratiform</u> clouds here; while they include both single-layer and multiple-layer clouds <u>with</u> top temperatures <u>avarmer than -40 °C</u>. Single-layer mid-level <u>supercooled</u> stratiform clouds have an annual global mean occurrence of approximately 3.3% with occurrences greater than 6% over northeastern China and the northern polar regions, and greater that 10% over the southern polar regions.

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3. Results and Discussions

The straightforward ice crystal growth pattern in mid-level stratiform mixed-phase clouds as described above enables using Z_e magnitudes to quantitatively infer ice number concentration variation in stratiform mixed-phase clouds. It is noted that because Z_e is proportional to ice number concentration and also the sixth power of particle size, differences in Z_e can be

- 10 either attributed to large changes in ice number concentration or small changes in ice crystal size. Based on integrated in situ measurements and airborne remote sensing, Zhang et al. (2012) suggest that—for similar clouds in terms of cloud top temperature (CTT) and LWP—ice crystal growth in mid-level stratiform mixed-phase clouds are similar and that Ze differences reveal differences in ice number concentration. They compare ZL differences between dusty and non-dusty mid-level stratiform mixed-phase clouds and conclude that mineral dust statistically enhances ice number concentration by a
- 15 factor of 2 to 6, depending on CTT. It is quite challenging to retrieve ice number concentration (N_{ice}) from radar measurements. Zhang et al. (2014) developed a method to estimate N_{ice} in stratiform mixed-phase clouds by using combined Z_e measurements and 1-D ice-growth model simulations. Cloud top temperature, LWP, and vertical air motion are required as inputs in their algorithms and sensitivity tests show that they all have <u>large</u> impacts on N_{ice} estimations. Due to large uncertainties in the MODIS derived LWP for mixed-phase clouds (Adhikari and Wang 2013). N_{ice} estimations in mixed-phase
- 20 phase clouds using A-Train satellite measurements are not available at this stage. In order to use the ZL magnitude to infer ice number concentration variations in this study, a narrow LWP range is selected to remove the impacts of LWP variation on the measured ZL. Fig. 2 shows the probability distribution function (PDF) of LWP for single-layer mid-level stratiform clouds from MODIS retrievals. The global mean LWP for single-layer mid-level stratiform clouds is approximately 119 g/m² with a standard deviation of 101 g/m². The PDF of LWP has a peak at approximately 45 g/m² and values decrease
- 25 quickly away from the peak. For our statistical analyses, a narrow LWP range is selected from the third of the cumulative distribution centered on the LWP peak which is bounded by the values of 20 g/m² and 70 g/m².

Fig. 3 shows the global, annual-average, mid-level mixed-phase stratiform cloud ice production statistics. Fig. 3a shows the cloud distributions in terms of CTT and ZL for six latitude bands (northern and southern tropical, mid-, and high-latitudes) within the LWP range between 20 g/m² and 70 g/m². Local peaks are seen in the ZL distributions at \sim -15 °C,

which correspond to the <u>fast-planar</u> ice growth regimes, and troughs are seen at -10 °C and -20 °C, corresponding to the relatively slow isometric growth habit (Sulia and Harrington, 2011; Zhang et al., 2014). Below -20 °C, ZL increases steadily as CTT decreases, probably because of higher ice number concentrations at lower CTTs (Zhang et al., 2014). At a given

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CTT, the ZL distribution has approximately 10 dBZ variations, which might be related to different environmental aerosol loadings and/or cloud LWPs associated with each individual cloud. Nevertheless, comparing different latitude bands, the northern latitude bands statistically have larger ZL than their southern counterparts at a given CTT. The northern mid- and high Jatitudes have the largest ZL values.

- 5 A complementary way to view the latitudinal dependence of the cloud properties is given in Fig. 3b, which presents the mean ZL of mid-level stratiform mixed-phase clouds as a function of CTT for the narrow LWP range. Due to potential drizzle contributions to Z_e measurements at relatively warm CTTs, the mean ZL is only calculated for clouds with CTT lower than -10 °C (Rasmussen et al., 2002; Zhang et al., 2017). Using mean ZL differences, we can quantitatively estimate ice concentration variations in mid-level stratiform clouds under similar cloud conditions in terms of CTT and LWP, similar
- 10 to that presented in Zhang et al., (2012). From Fig. 3b, the northern mid- and high_Jatitudes have the largest mean ZL while the southern low latitude band, has the smallest values. Consistent with the cloud distribution statistics in Fig. 3a, northern hemisphere latitude bands have larger mean ZL at a given CTT than their counterparts in southern hemisphere. Depending on CTT range, the northern mid- and high_Jatitude bands are ~ 6and.8 dBZ larger than their southern counterparts; while the northern low latitude band, is only ~ 1 dBZ greater than southern low latitude band. These results are consistent with the
- 15 studies by Choi et al. (2010) and Tan et al. (2014) which show that the Northern Hemisphere has a smaller supercooled liquid fraction than the Southern Hemisphere for a given temperature range, and it is also consistent with Zhang et al. (2010) which shows that the Northern Hemisphere mixed-phase clouds have larger ice water paths (IWP).

Atmospheric pressure is another factor that could impact ice crystal diffusional growth and therefore the observed hemispheric and latitudinal ZL differences. The same subfreezing temperature at low latitudes corresponds to a higher height

- 20 above mean sea level and therefore a lower atmospheric pressure level than mid- and high latitudes. Takahashi et al. (1991) // show that the mass growth rate at 860 mb is approximately 30% larger than at 1010 mb due to the impact of pressure difference on the diffusivity of water vapor in air. It is also noted that, from Figure 20 in their paper, the mass growth difference due to pressure difference is much smaller than that due to temperature difference. Within the Rayleigh scattering regime, radar reflectivity is proportional to the square of ice crystal mass. Therefore, the 30% difference in mass causes
- 25 approximately a 2 dBZ difference in Z_e. We investigated hemispheric and latitudinal differences of atmospheric pressure at subfreezing temperatures using four years of ECMWF-AUX product between 2006 and 2010. As shown in Fig. 4, for a given temperature, hemispheric differences in atmospheric pressure profiles are negligible over mid- and low latitude bands, and range from 40 mb to 140 mb over the high latitude band. Therefore, pressure-level differences have a negligible contribution to the hemispheric ZL differences over mid- and low latitude bands, and contribute less than 2 dBZ to the
- 30 observed hemispheric ZL differences over the high latitude band. After removing the contributions from atmospheric pressure differences, mid-level stratiform mixed-phase clouds over northern mid- and high latitude bands still have ZL that are approximately 6 dBZ larger than their southern counterparts. By focusing on mid-level stratiform mixed-phase clouds and carefully isolating the impacts of CTT, LWP, and atmospheric pressure, the systematically larger ZLs suggest a factor of

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4, higher ice number concentrations over northern mid- and high latitudes than their southern counterparts.

The systematically larger ZL and higher ice number concentrations over the Northern Hemisphere for similar mid-level stratiform mixed clouds might be related to larger background aerosol loadings in the Northern Hemisphere. Using CALIOP measurements, Tan et al. (2104) show that the Northern Hemisphere has dramatically larger, frequencies of high aerosol

- 5 occurrence than the Southern Hemisphere at sub-freezing temperatures. Based on ground-based lidar and radar remote sensing measurements from sites in both Northern and Southern Hemispheres, Kanitz et al. (2011) found that layered supercooled clouds at northern mid-latitudes have significantly larger fractions of ice containing clouds compared with southern mid-latitudes, which is possibly related to the rather different aerosol conditions. In addition, larger mean ZL over the northern mid- and high latitude bands than the northern low latitude band can also be connected to larger aerosol
- 10 (especially dust) loadings at sub-freezing levels. Using multiple-years of ground-based Raman lidar measurements, Seifert et al. (2010) show that Leipzig, Germany (northern mid-latitude) has much higher ice-containing cloud fraction than Cape Verde (northern low latitude) at a given CTT below 0 °C, consistent with the results in Fig.3. They proposed that possible factors influencing the differences include different sources of INP, chemical aging, as well as removal of larger aerosol particles by washout in the tropics. Indeed, although the tropics and sub-tropics have extensive dust source regions, large 15 dust particles are not able to elevated to sub-freezing levels without strong convection (Luo et al., 2014).

We next investigate the global impact of LWP on ZL and its latitudinal variation. At a given CTT, cloud with a larger LWP has a geometrically thicker liquid water layer, which allows ice crystals to reside longer in the liquid-dominated layer and grow larger by the WBF process. In addition, cloud with a larger LWP also has a larger ice growth rate by accretion (Zhang et al., 2014). Fig. 5, shows the mean ZL of as a function of CTT and LWP for the six latitude bands. As expected, the 20 mean ZL increases gradually with LWP at a given CTT for all latitude bands. Mean ZL generally increases more than about 5 dBZ going from thin clouds, which are associated with small LWP, to very thick clouds, which are associated with large LWP. Therefore, observations show a dramatic impact of LWP on the measured ZL. However, within any given narrow LWP range, the mean ZL for northern latitude bands are still much greater than their southern counterparts, further supporting our conclusion that the systematic ZL differences between northern and southern latitude bands are related to aerosol activity.

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To further explore aerosol impacts on ice formation, Fig. 6 shows the seasonal variations of mid-level stratiform mixedphase cloud distributions in terms of CTT and ZL for the six latitude bands and Fig. 2, shows mean ZL seasonal variations as a function of CTT. As expected, northern latitude bands have greater ZL than their counterparts in the Southern Hemisphere at any season for similar clouds in terms of similar CTT and LWP, probably related to higher background aerosol loadings over the Northern Hemisphere. Comparing different seasons, southern latitude bands generally have little seasonal variation in ZL, as is evident in Fig. 7. In contrast, northern latitude bands have dramatic seasonal variations in ZL, with the largest ZL

occurring in MAM (boreal springtime) and smallest in DJF (boreal wintertime). The northern mid- and high Jatitude bands have the largest seasonal variations among all latitude bands. At CTTs warmer than -30 °C, ZLs over northern mid- and high

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latitude bands are larger in the boreal springtime than wintertime by approximately 4 dBZ and 6 dBZ for similar clouds in terms of CTT and LWP, respectively, From Fig. 4b, pressure profile differences between boreal springtime and wintertime are fairly small over northern mid- and high latitude bands. Therefore, the systematically larger ZLs of 4 dBZ and 6 dBZ during boreal springtime than wintertime suggests a factor of 2.5 and 4.0 higher ice number concentrations.

- 5 Dust particles are effective INPs and are recognized as one the dominant global INP sources (DeMott et al., 2010; Hoose and Möhler, 2012). Choi et al (2010) show the seasonal variation of global mineral dust occurrence at the -20 °C isotherm using the CALIOP level 2 vertical feature mask data. They observed a significant correspondence between mineral dust occurrence and <u>reduction in</u> supercooled cloud fraction, especially over the northern mid-latitudes, <u>suggesting that</u> elevated mineral dust particles effectively glaciate supercooled clouds by providing abundant INPs. In their study, the Arctic
- 10 regions have dramatic seasonal variations in supercooled cloud fractions, with the lowest during the springtime, However, no obvious dust activity over the Arctic regions is shown in their paper. Luo et al. (2014) point out that CALIOP level 2 data product often misses the detection of elevated thin dust layers. They presented improved algorithms to identify thin dust layers using CALIOP layer-mean particulate depolarization ratios and CPR measurements. Fig. & shows the distributions of global dust occurrence and their seasonal variations at different sub-freezing temperature ranges based on the dust dataset
- 15 developed by Luo et al. (2014). It is obvious that during March-April-May (MAM), the boreal springtime, northern mid- and high- latitude regions have significantly higher dust occurrences than other seasons at any given sub-freezing temperature range. Similarly, using multiple-years of ground-based remote sensing measurements at the ARM NSA Barrow site, Zhao (2011) shows that Arctic mixed-phase clouds in springtime have larger ice water paths (IWPs) and smaller supercooled liquid water fraction than the other three seasons, which might be related to there being more dust events observed with lidar
- 20 <u>depolarization measurements</u> during springtime that provide effective INPs for ice nucleation in clouds. The significant seasonal variations of ice production and their correspondence with dust occurrence in northern mid- and high-latitude mixed-phase clouds suggest that aerosol-dependent ice concentration parameterizations need to be used in GCMs and improved aerosol (especially dust) simulations are required in order to improve global mixed-phase cloud simulations, especially over the Northern Hemisphere.

25 4. Summary

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Four years of collocated CALIPSO lidar and CloudSat radar measurements together with other ancillary A-Train products during 2006-2010 are analyzed to study primary ice particle production characteristics in single-layer mid-level stratiform mixed-phase clouds on a global scale. Mid-level stratiform mixed-phase clouds have a simple dynamic environment and straightforward ice growth trajectory that enables using Z_e measurements to quantitatively infer ice number concentration variations. We carefully isolate factors that impact ice diffusional growth and measured cloud layer radar Z_g by focusing on mid-level stratiform mixed-phase clouds with same CTT and similar LWPs. We also analyzed atmospheric

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pressure impacts. Together with MODIS liquid water path retrievals and an improved thin dust layer detection algorithm, we connect the observed ZL differences and ice concentration variations to aerosol (especially dust) activities on a global scale.

Using the large dataset, we show that for similar clouds in terms of cloud top temperature and liquid water path, northern hemisphere latitude bands have ZL that are ~ 1 to 8 dBZ larger than their counterparts in southern hemisphere for a

- 5 given cloud top temperature, After removing contributions from atmospheric pressure differences, ZL is still 6 dBZ larger that suggests a factor of 4 higher ice number concentrations (on average) over northern mid- and high latitudes than their southern counterparts. The systematically larger ZL and higher ice number concentrations in mid-level stratiform mixedphase clouds over the Northern Hemisphere are possibly related to larger background aerosol loadings. LWP has a significant impact on measured ZL, but we show that within a given narrow LWP range, mean ZL over northern latitude
- 10 bands is always larger than their southern counterparts. Furthermore, we show that the northern mid- and high Jatitudes have dramatic seasonal variations in ZL, where ZL can be up to 6 dBZ larger in springtime than in wintertime. This might be related to more dust events during springtime that provide effective INPs for ice nucleation in clouds. Since mixed-phase cloud property evolution is strongly dependent on ice number concentration, our study suggests that aerosol-dependent ice concentration parameterizations are required in GCMs in order to improve global mixed-phase cloud simulations. The
- 15 results in this study can be used to evaluate global ice concentrations in mixed-phased clouds and aerosol impacts simulated by GCMs.

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5 from, respectively, <u>http://www.cloudsat.cira.colostate.edu</u> and <u>https://eosweb.larc.nasa.gov/order-data</u>.

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Figure 1 Global distribution of single-layer mid-level stratiform cloud occurrence frequency from four-years of collocated CALIOP and CPR measurements.



Figure 2. The probability distribution function (PDF) of LWP for single-layer mid-level stratiform clouds from MODIS retrievals. The red area indicates the third of the cumulative distribution that is centered on the peak of the LWP PDF. Given in the figure is LWP 1, the value for the lower third, and LWP 2, the value at the upper third.

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Figure 3. Global, annual-average, mid-level mixed-phase stratiform cloud ice production statistics. Results for six, 30°-latitude bands are shown covering the northern and southern tropical, mid-, and high-latitude regions. Cases are restricted so
that the supercooled liquid water path is within the range between 20 g/m² and 70 g/m². (a) Cloud distributions are in terms of cloud top temperature (CTT) and layer-maximum radar reflectivity (ZL), a proxy for ice production. (b) Mean ZL of clouds as a function of CTT. Distributions are normalized at each CTT bin. The bin sizes for CTT and ZL are 1°C and 1 dBZ, respectively.





Figure 4. Atmospheric pressure profiles with subfreezing temperatures for (a) the six latitude bands, and (b) their seasonal* variations. MAM stands for March-April-May, JJA for June-July-August, SON for September-October-November, DJF for December-January-February.

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Figure 5. Mean of the layer-maximum radar reflectivity (ZL) of mid-level stratiform mixed-phase clouds as a function of cloud top temperature and liquid layer path (LWP). Results shown for six latitude bands as in Fig. 1. The dashed lines are the narrow range of LWP between 20-70 g/m².

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Figure 6 Similar to Figure 3, except for the seasonal variation in stratiform mixed-phase cloud ice production statistics.

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Figure \S_{v} Distributions of global dust occurrences and their seasonal variations at different sub-freezing temperature ranges based on the dust dataset developed by Luo et al. (2014). Temperature ranges are given at the right. Each column is for a season, with the abbreviations described in Figure 4.

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Deleted: Each column is for a season: MAM for March-April-May, JJA for June-July-August, SON for September-October-November, and DJF for December-January-February.