Response to reviewer #1

General Comments:

This study investigates the relationship between number concentrations of ice-nucleating particles and ice crystals, based on which it proposed an identification method for two ice-nucleating regimes for dust-related cirrus clouds. The findings are interesting and worthy for publication after necessary revisions.

Response: We appreciate the reviewer's thoughtful review and constructive comments. All the comments have been addressed in the revised manuscript, and the responses to each comment are given below.

Specific comments:

Comments 1: *Line 40-41, considering the shattering of ice particles which can further serve as ice nuclei to help the heterogeneous formation of ice crystals, I am not sure if the description "and later homogeneous freezing is followed due to the depletion of ice-nucleating particles and the persistence of cooling" is accurate or not.*

Response 1: To our best knowledge, the shattering of ice particles indicates secondary ice production, including four types according to the current studies, i.e., rime splintering, collision fragmentation, droplet shattering, and sublimation fragmentation. These processes usually occur at modest supercooling temperature (>-10°C), as seen in a review from Field et al. (2017). However, cirrus clouds form at farther colder temperatures; thus, we consider the shattering of ice particles cannot serve as the additional ice nuclei in this 'second stage'. For in situ-origin cirrus formed in a slow updraft, Krämer et al. (2016) have stated that '***The formation mechanism of this cirrus type starts with HET freezing that is followed by a second, HOM ice nucleation event (since all IN are already consumed) if the cooling phase is long enough and temperature fluctuation do not cause a HOM freezing event earlier.***' (Please see section 5.1.1 therein). As a result, we would like to retain this sentence in the revised manuscript.

Reference:

Krämer, M., Rolf, C., Luebke, A., Afchine, A., Spelten, N., Costa, A., Meyer, J., Zöger, M., Smith, J., Herman, R. L., Buchholz, B., Ebert, V., Baumgardner, D., Borrmann, S., Klingebiel, M., and Avallone, L.: A microphysics guide to cirrus clouds – Part 1: Cirrus types, Atmos. Chem.

Phys., 16, 3463-3483, doi.org/10.5194/acp-16-3463-2016, 2016.

Field, P., Lawson, P., Brown, G., Lloyd, C., Westbrook, D., Moisseev, A., Miltenberger, A., Nenes, A., Blyth, A., Choularton, T., Connolly, P., Bühl, J., Crosier, J., Cui, Z., Dearden, C., DeMott, P., Flossmann, A., Heymsfield, A., Huang, Y., Kalesse, H., Kanji, Z., Korolev, A., Kirchgaessner, A., Lasher-Trapp, S., Leisner, T., McFarquhar, G., Phillips, V., Stith, J., and Sullivan, S.: Secondary ice production – current state of the science and recommendations for the future, Meteorol. Mon., 58, 7.1–7.20, doi.org/10.1175/AMSMONOGRAPHS-D-16-0014.1, 2017.

Comments 2: *Line 43-46, actually, at low temperature (such as below -25 degree C), even sulfate and nitrate particles can serve as IN as indicated by Che et al. (2019, doi: 10.1016/j.atmosres.2020.105196).*

Response 2: Thank you for providing this reference. We have added it to the revised manuscript.

Comments 3: *Line 55-57, this description is not correct in my opinion. The change of cirrus clouds here in principle reduces the outgoing longwave radiation to space while it increases the emission of cirrus, which is why it plays a more cooling effect.*

Response 3: For clarity, we have modified this sentence to **'These optically thinner cirrus clouds absorb outgoing long-wave (LW) radiation from the surface and allow more LW radiation to emit to space, contributing to a cooling effect (Gasparini and Lohmann, 2016). This cooling effect prevails over the warming effect caused by the increased incoming solar radiation (warming), resulting in a net-positive radiative effect (cooling) on the radiation budget of the Earth (Kuebbeler et al., 2014; Lohmann and Gasparini, 2017).'** (Please see lines 57-61)

References:

Gasparini, B., and Lohmann, U.: Why cirrus cloud seeding cannot substantially cool the planet, J. Geophys. Res.-Atmos., 121, 4877–4893, doi.org/10.1002/2015JD024666. 2016.

Lohmann. U., and Gasparini, B.: A cirrus cloud climate dial? Science, 357(6348), 248-249, doi.org/10.1126/science.aan3325, 2017.

Kuebbeler, M., Lohmann, U., Hendricks, J., and Kärcher, B.: Dust ice nuclei effects on cirrus clouds, Atmos. Chem. Phys., 14, 3027–3046, doi.org/10.5194/acp-14-3027-2014, 2014.

Comments 4: *Line 89-90, considering potential variation of cirrus properties, why do the authors only choose two cases, and why do they choose these two cases?*

Response 4: Our manuscript is a technical report to introduce a sole remote-sensing approach to separate ice-nucleation regimes for cirrus clouds. Compared with in-situ techniques, our approach can realize long-term observation over the globe using space-borne platforms, such as CALIPSO and EarthCARE (Illingworth et al., 2015) satellite, which is of high significance to quantify relative contributions of homogeneous/heterogeneous freezing to cirrus clouds formation and evaluate the effects of different parameterization schemes on global cloud simulations. However, many aspects need to be verified for this method (such as the applicability of regional conversion factors in INP retrieval in section 2.3, the selection of optimal INP parameterization scheme in section 2.3, the confirmation of dust-related cirrus clouds in section 3, and so on), which is thus better to start with some clear-cut case studies. Then, a robust long-term study on a global scale can be expected in the future. That's why we submit this manuscript as the type of '**technical report**'. Therefore, we have added the following sentence in the revised manuscript '**⋯, which are favorable for validating some aspects of this method (such as the applicability of regional conversion factors in INP retrieval, the selection of optimal INP parameterization scheme, the confirmation of dust-related cirrus clouds, and so on) and conducting a robust long-term study on a global scale subsequently.**' (Please see lines 94-97)

Reference:

Illingworth, A. J., Barker, H. W., Beljaars, A., Ceccaldi, M., Chepfer, H., Clerbaux, N., Cole, J., Delanoë, J., Domenech, C., Donovan, D. P., Fukuda, S., Hirakata, M., Hogan, R. J., Huenerbein, A., Kollias, P., Kubota, T., Nakajima, T., Nakajima, T. Y., Nishizawa, T., Ohno, Y., Okamoto, H., Oki, R., Sato, K., Satoh, M., Shephard, M., Velázquez-Blázquez, A., Wandinger, U.,Wehr, T., van Zadelhoff, G.-J.: The EarthCARE Satellite: The next step forward in global measurements of clouds, aerosols, precipitation and radiation, B. Am. Meteorol. Soc., 96, 1311–1332, doi.org/10.1175/BAMS-D-12-00227.1, 2015.

Comments 5: *Line 142-143, how could the authors make sure that the meteorology at the station is the same as that over the location CALIOP observes.*

Response 5: Generally, there are only two options for the meteorological parameters in INPC calculation, from either radiosonde measurements (He et al., 2021b) or the reanalysis data (ERA5 or GDAS) (Ansmann et al., 2019). We employ the radiosonde measurements since they have a better vertical resolution and accuracy for a fixed geo-location. As the reviewer mentioned, it contains an assumption that the atmosphere between the weather station and the location CALIPSO observes is horizontally homogeneous. This assumption is more valid for temperature and pressure (what we use here) than relative humidity. To be clear, we have added the following sentence '**It is assumed that the temperature and pressure profiles of the atmosphere between the weather station and the location CALIPSO observes are horizontally homogeneous.**' (Please see lines 153-154)

Comments 6: *Line 144-148, for ice forming, only these two mechanisms play the role?*

Response 6: These two (immersion freezing and deposition freezing) are considered as the primary mechanism playing the role here. Besides, condensation freezing can be considered a special type of immersion freezing. Contacting freezing, which needs an INP to collide with a supercooled droplet is ignored (the description can be found in the text regarding to figure 6). We have added the sentence below '**Besides, condensation freezing can be considered a special type of immersion freezing; contacting freezing, which needs an INP to collide with a supercooled droplet, was ignored.**' (Please see lines 160-162)

Comments 7: *Line 178, is there any method to confirm the interaction between dust and cirrus clouds?*

Response 7: The best method to confirm the dust-cirrus interaction is to sample the ice crystals inside cirrus clouds. Whether the residuals of sampled cloud particles contain dust particles can be a direct indicator of the interaction between dust and cirrus clouds (Cziczo et al., 2013). As for the sole employment of the remote sensing approach, it can be concluded the interaction between dust and cirrus clouds if cirrus clouds are observed to be **embedded** in the dust layer as also can be seen in figure 14 of Ansmann et al. (2019a). To better explain this issue, we have added the following statement '**The dust-cloud interaction is generally considered to take place if a cirrus cloud is embedded in a dust layer (Ansmann et al., 2019a; Marinou et al., 2019).**' (Please see lines 191-192)

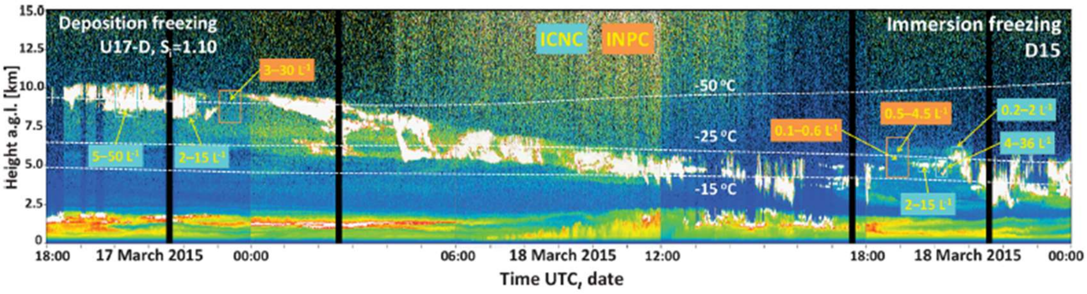

Figure 14. Continuous cirrus and mixed-phase cloud observations for 30 h over Nicosia on 17–18 March 2015 (also shown in Fig. 5e, g, and i). The air mass from 5 to 10 km height was replaced (starting at great heights) by dust-free, dry air advected from Turkey and southern Europe between 02:00 and 11:00 UTC on 18 March, leading to the impression of a descending dust and cirrus layer. Several INPC and ICNC values estimated from the lidar observations are given as numbers determined for the indicated orange (INPC) and blue (ICNC) boxes. The deposition nucleation U17-I(d) parameterization is used on 17 March (at 9–10 km height for $S_i = 1.1$) and the immersion freezing D15 parameterization is applied in the evening data analysis on 18 March (at 5–6 km height). Dashed white lines show the GDAS1 temperature isolines with a 3 h resolution.

References:

Ansmann, A., Mamouri, R.-E., Bühl, J., Seifert, P., Engelmann, R., Hofer, J., Nisantzi, A., Atkinson, J. D., Kanji, Z. A., Sierau, B., Vrekoussis, M., and Sciare, J.: Ice-nucleating particle versus ice crystal number concentration in altocumulus and cirrus embedded in Saharan dust: A closure study, Atmos. Chem. Phys., 19, 15087-15115. doi.org/10.5194/acp-19-15087-2019, 2019a.

Cziczo, D., Froyd, K., Hoose, C., Jensen, E., Diao, M., Zondlo, M., Smith, J., Twohy, C., and Murphy, D.: Clarifying the dominant sources and mechanisms of cirrus cloud formation, Science, 340, 1320-1324, doi.org/10.1126/science.1234145, 2013.

Krämer, M., Rolf, C., Luebke, A., Afchine, A., Spelten, N., Costa, A., Meyer, J., Zöger, M., Smith, J., Herman, R. L., Buchholz, B., Ebert, V., Baumgardner, D., Borrmann, S., Klingebiel, M., and Avallone, L.: A microphysics guide to cirrus clouds – Part 1: Cirrus types, Atmos. Chem. Phys., 16, 3463-3483, doi.org/10.5194/acp-16-3463-2016, 2016.

Marinou, E., Tesche, M., Nenes, A., Ansmann, A., Schrod, J., Mamali, D., Tsekeri, A., Pikridas, M., Baars, H., Engelmann, R., Voudouri, K.-A., Solomos, S., Sciare, J., Groß, S., Ewald, F., and Amiridis, V.: Retrieval of ice-nucleating particle concentrations from lidar observations and comparison with UAV in situ measurements, Atmos. Chem. Phys., 19, 11315-11342. doi.org/10.5194/acp-19-11315-2019, 2019.

Comments 8: *Line 205-209, it seems that there is a large fraction of ice particles with diameters between 5 and 25 μm (more than 50%). Is this reasonable, and why?*

Response 8: The large fraction of ice crystals with diameters of 5-25 μm is common as seen in the comparison with in-situs measurements (Sourdeval et al., 2018, see Figure 3 given as below and Appendix 3 therein). It is probably caused by the assumption of the ice-particle size spectrum in DARDAR N_{ice} retrieval. Sourdeval et al. (2018) have the following statements in section 3.1 of their paper: '***However, these numbers do not provide a complete estimation of the accuracy of N_i as DARDAR does not rigorously account for uncertainties related to assumptions on the PSD shape. A preliminary sensitivity study has shown that strong deviations from the assumed α and β parameters could reasonably lead to errors of up to 50% on N_i (see Sect. A3). Therefore, the overall uncertainties on Ni due to instrumental sensitivity and physical assumptions are difficult to quantify based on DARDAR products alone.***'

Therefore, we have also added the following sentences in the outlook part of last section '**In addition, the future launch of the EarthCARE satellite is more anticipated (Illingworth et al., 2015), since its 94.05-GHz cloud profiling radar can possess the capability of Doppler detection so that the in-cloud ICNC will be determined more accurately under the better constraint of the ice-particle size spectrum.**' (Please see lines 366-369)

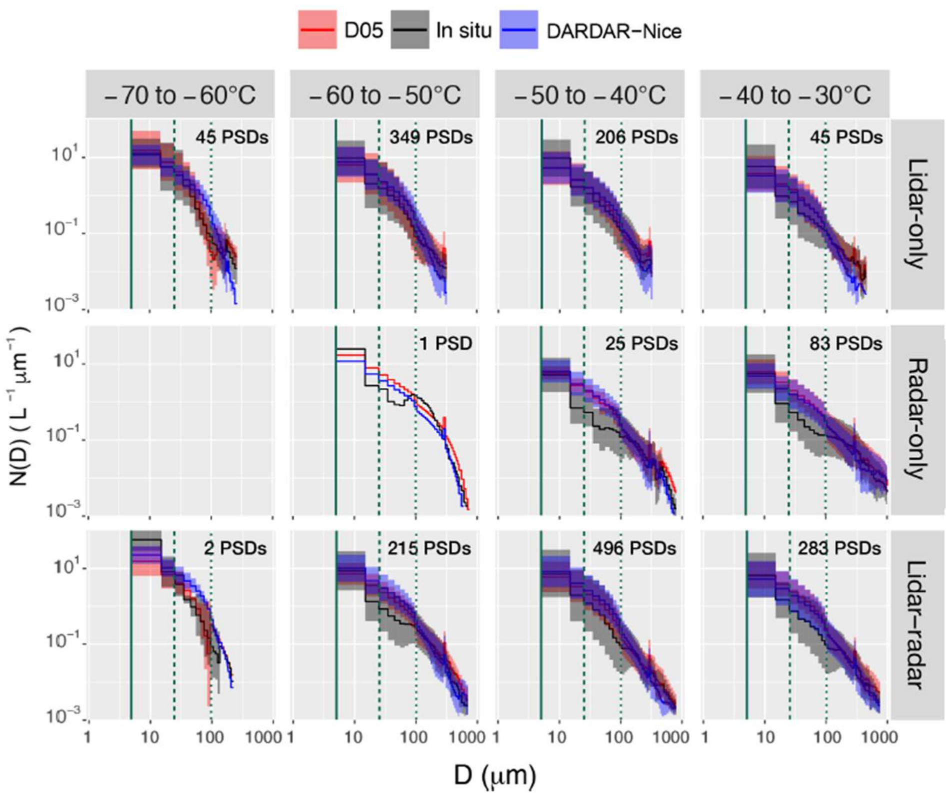

Figure 3. Similar to Fig. 1 but for SPARTICUS flights coincident with the A-Train overpass. PSDs estimated on the basis of DARDAR IWC and N_0^* retrievals (i.e., corresponding to the DARDAR-Nice N_i) are shown in blue. All PSDs are averaged per temperature bin (columns) and by instrumental conditions met during DARDAR retrievals (rows).

Reference:

Sourdeval, O., Gryspeerdt, E., Krämer, M., Goren, T., Delanoë, J., Afchine, A., Hemmer, F., and

Quaas, J.: Ice crystal number concentration estimates from lidar-radar satellite remote sensing. Part 1: Method and evaluation, Atmos. Chem. Phys., doi.org/10.5194/acp-2018-20, 2018.

Illingworth, A. J., Barker, H. W., Beljaars, A., Ceccaldi, M., Chepfer, H., Clerbaux, N., Cole, J., Delanoë, J., Domenech, C., Donovan, D. P., Fukuda, S., Hirakata, M., Hogan, R. J., Huenerbein, A., Kollias, P., Kubota, T., Nakajima, T., Nakajima, T. Y., Nishizawa, T., Ohno, Y., Okamoto, H., Oki, R., Sato, K., Satoh, M., Shephard, M., Velázquez-Blázquez, A., Wandinger, U.,Wehr, T., van Zadelhoff, G.-J.: The EarthCARE Satellite: The next step forward in global measurements of clouds, aerosols, precipitation and radiation, B. Am. Meteorol. Soc., 96, 1311–1332, doi.org/10.1175/BAMS-D-12-00227.1, 2015.

Comments 9: *Line 237, why is there a ")" here*

Response 9: ')' has been removed.

Comments 10: *Line 240-242, During the transport of dust, the concentration of dust aerosols could vary a lot with space. How do the authors make sure the dust obtained here can represent that contacting cirrus clouds?*

Response 10: Thank you for pointing out this. We took the closely linked cirrus and dust layers for ICNC and INPC retrievals, respectively, which makes dust sedimentation effects negligible within several kilometers in distance. On the other hand, it can be reasonably assumed that the dust number concentration contained in the cirrus cloud is comparable to that in the **adjacent dust layer**, which can be easily entrained into the cirrus from the top and each side of the cloud cell.

For clarity, we have added the following sentences '**Dust particles contained in the dust plume may undergo sedimentation (especially for coarse-mode dust particles) to a certain extent during the long-range transport, causing a variation in dust number concentration within the dust plume. The time scale for cirrus cloud formation is much smaller than that for dust transport (generally several tens of hours or a couple of days), suggesting that the removal of dust particles is negligible during cirrus formation. Therefore, it can be reasonably assumed that the dust number concentration contained in the cirrus cloud is comparable to that in the adjacent dust layer.**' (Please see lines 264-269)

Comments 11: *Line 305, how do the authors mean "emitting back to space"?*

Response 11: This sentence has been rewritten as **'…, reflecting more solar radiation to space as well as retaining more LW radiation in the atmosphere (i.e., emitting less LW radiation to space)'.** (Please see lines 335-336)