Reviewer 2

General Comments:

This referee agrees with the authors that this may be the first paper that studies the impact of aerosol concentration on cirrus cloud microphysics (through ice effective radius Rei in this case). The satellite observations appear valuable to our efforts to understand cirrus cloud-aerosol-radiation interactions, and should ultimately be published in ACP. However, the observations are interpreted narrowly, and much greater scope for interpretation should be provided. Alternate interpretations of the satellite retrievals have been provided under "Major Comments".

While the observations may contribute to our understanding of cirrus cloud-aerosol interactions, this may not be true for the cirrus cloud modeling work presented, as indicated below. It is recommended that Sect. 3.4 be dropped from the paper unless the concerns listed below can be adequately addressed. That is, the cloud model should predict cloud properties that are representative of in situ cirrus clouds, and the conditions assumed should also be representative. These modeling results are also irrelevant to anvil cirrus clouds, for the reasons stated in (14) below. The paper is well written and organized, with high quality figures. The observational methodology appears appropriate for this task; the other referee appears to be an expert in this area. The amount of supplementary material appears appropriate.

Response: We thank the Reviewer for constructive comments and suggestions. We have followed them carefully in revising the manuscript.

In particular, we have interpreted satellite observations more broadly (see our responses to the reviewer's 5th – 13rd major comments). We have also substantially improved the model simulations for application to in-situ ice clouds and clarified that the modeling results are not applicable to convection-generated ice clouds (see our responses to the reviewer's 14th – 16th major comments).

Below is a point-by-point response.

Major Comments:

1) Page 7, line 24: At what RHi do the deposition INP activate?

Response: Thank you. We have added the following sentence in the text. (Page 7, Line 19-22)

"The deposition nucleation on externally mixed dust (deposition INP) and immersion nucleation of coated dust (immersion INP) are parameterized following the work of Kuebbeler et al. (2014); the critical ice supersaturation ratios are 10% (T ≤ 220 K) or 20% (T > 220 K) for the former, and 30% for the latter."

2) Section 3: To gain confidence in the reported retrievals of Rei, these Rei retrievals could be compared against another Rei retrieval method reported in the literature. A global analysis of Rei is reported in Hong and Liu (J. Climate, 2015), based on CloudSat-CALIPSO measurements using the "DARDAR" method (a different method than used in this study). Although Hong and Liu do not relate Rei to aerosols, Rei is related to temperature, altitude and cloud optical depth, often as a function of latitude zone and season. Please make some comparisons, as direct as possible, between Hong and Liu Rei values and those reported in this paper.

Response: Thank you. Stein et al. (2011) has systematically compared the DARDAR R_{ei} retrievals with the MODIS data, as shown in Fig. R1. The default DARDAR retrievals of R_{ei} (denoted by VarCloud-OA, left panel) are mostly larger than MODIS's values. This discrepancy is partly induced by different assumptions of ice crystal habit (shape) in these two products. When the DARDAR retrievals are adjusted to mimic the MODIS assumption of ice crystal habit (VarCloud-BR, right panel), the joint distribution of individual R_{ei} retrievals has its peak close to the ratio of 1 between the two products, indicating a much better agreement. Nevertheless, the overall shape of the distributions indicates that the MODIS retrievals mostly lie between 10 and 50 μm, while both DARDAR products regularly retrieve R_{ei} above 60 μm. Hong and Liu (2015) reveals that the large R_{ei} values in DARDAR retrievals are predominantly associated with large cloud optical thickness (> 3.0, particularly > 20). In this study, however, we focus on ice-only clouds (mostly cirrus clouds), which typically have an optical thickness less than 5.0 (see Fig. 2 in the main text). For this reason, the agreement in R_{ei} between MODIS and DARDAR could be better for the type of cloud used in our analysis.

We have added the discussions above in the revised manuscript, citing Hong and Liu (2015) and Stein et al. (2011). (Page 4, Line 4-17)

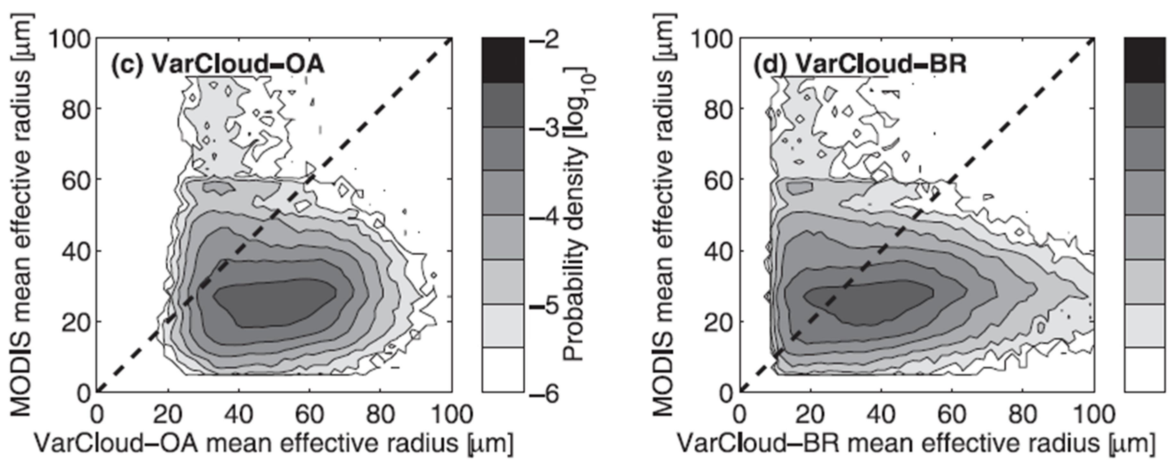

Figure R1. A comparison between the MODIS retrievals of R_{ei} and two DARDAR retrievals: left – the default DARDAR retrieval, denoted by VarCloud-OA; right – an adjusted DARDAR retrieval to mimic the MODIS assumption of ice crystal habit, denoted by VarCloud-BR. Data are from October 2008. Dashed lines in the figures indicate the 1:1 ratio. This figure is adapted from Stein et al. (2011). ©American Meteorological Society. Used with permission.

3) Section 3.1: The error bars in Fig. 1 and elsewhere denote standard errors ($\sigma/\sqrt{N}$) where σ is the standard deviation and N is the sample number. This makes the relationships difficult to interpret since we do not know what N is. Please use only σ for the error bars so the reader can better evaluate these relationships.

Response: Thank you. We submit that both standard error and standard deviation are widely used, but with different focuses. Standard deviation describes how spread out a set of measurements is, while standard error indicates how accurate our estimate of the mean is likely to be (McDonald, 2014). There is a probability of 68.3% that the population (true) mean would be within one standard error of the sample mean, and a probability of 95.4% to be within two standard errors (McDonald, 2014).

In this study, R_{ei} is affected not only by aerosol loading, but also a number of confounding factors such as meteorology, altitude, ice water content, etc. Some of these factors (e.g., meteorological conditions) may exert even larger effects on R_{ei} than aerosols. If standard deviations are plotted, we may not gain an idea whether changes in aerosol loading would induce significant changes in mean R_{ei}. However, the usage of standard errors could highlight the aerosol effects, because the population (true) mean for a given aerosol bin would very likely (with a 68.3% probability) fall within the error bars. Moreover, if the 95% confidence intervals (1.96 × standard error) of R_{ei} for two aerosol bins do not overlap, we would be sure that mean R_{ei} for these two aerosol bins are significantly different at the 0.05 level (McDonald, 2014). For these reasons, we submit that the standard error, which has been adopted by many observational studies on aerosol-cloud interactions (e.g., Jiang et al., 2011; Su et al., 2011; Koren et al., 2010; Li et al., 2011; Wang et al., 2015), appears to be suitable in our study. Additionally, we have specified the total number of samples used in each figure in the revised figure captions.

4) Page 9, line 14: Higher RH and CAPE imply that an air parcel will experience a longer time period exceeding ice saturation (i.e. longer time for supersaturation development, increasing the odds of exceeding the RHi threshold for homogeneous ice nucleation (henceforth hom)). This point could be made more clear.

Response: This suggestion is well taken. We have added this point in the revised manuscript:

"Under moist conditions (high RH, high CAPE, or negative U200), an air parcel could experience longer time for supersaturation development, increasing the odds of exceeding the supersaturation threshold for homogeneous ice nucleation." (Page 10, Line 29-32)

5) Page 10, lines 3-11 (1st paragraph): The similar dependence of Rei on column AOD (for all aerosol) and column AOD for dust aerosol only is critical to this study, and supports the assumption that ice nuclei (henceforth IN) concentration increases with increasing column AOD. However, this correspondence has only been demonstrated for column AOD and not for layer AOD (where layer AOD corresponds to cirrus cloud levels). Dust is often confined below cirrus cloud levels, and a column AOD-dust AOD relationship does not imply that one exists for layer AOD. Please make this point here.

Response: We have conducted a similar analysis for in-situ ice clouds and layer AOD for which the results are illustrated in Fig. R2 below (Fig. 3 in the revised manuscript). Similar to column AOD, the dependences of R_{ei} on layer AOD for all aerosols (Fig. R2a-c) and for dust only (Fig. R2d-f) are also similar. Since specific components of dust aerosols have been known as effective INPs, the similar R_{ei}-layer AOD relations imply that INP concentrations are also positively correlated with layer AOD, and that the proposed mechanisms for water vapor modulation is applicable to in-situ ice clouds and layer AOD.

We have supplemented this analysis in the revised manuscript. (Fig. 3; Page 11, Line 16-21)

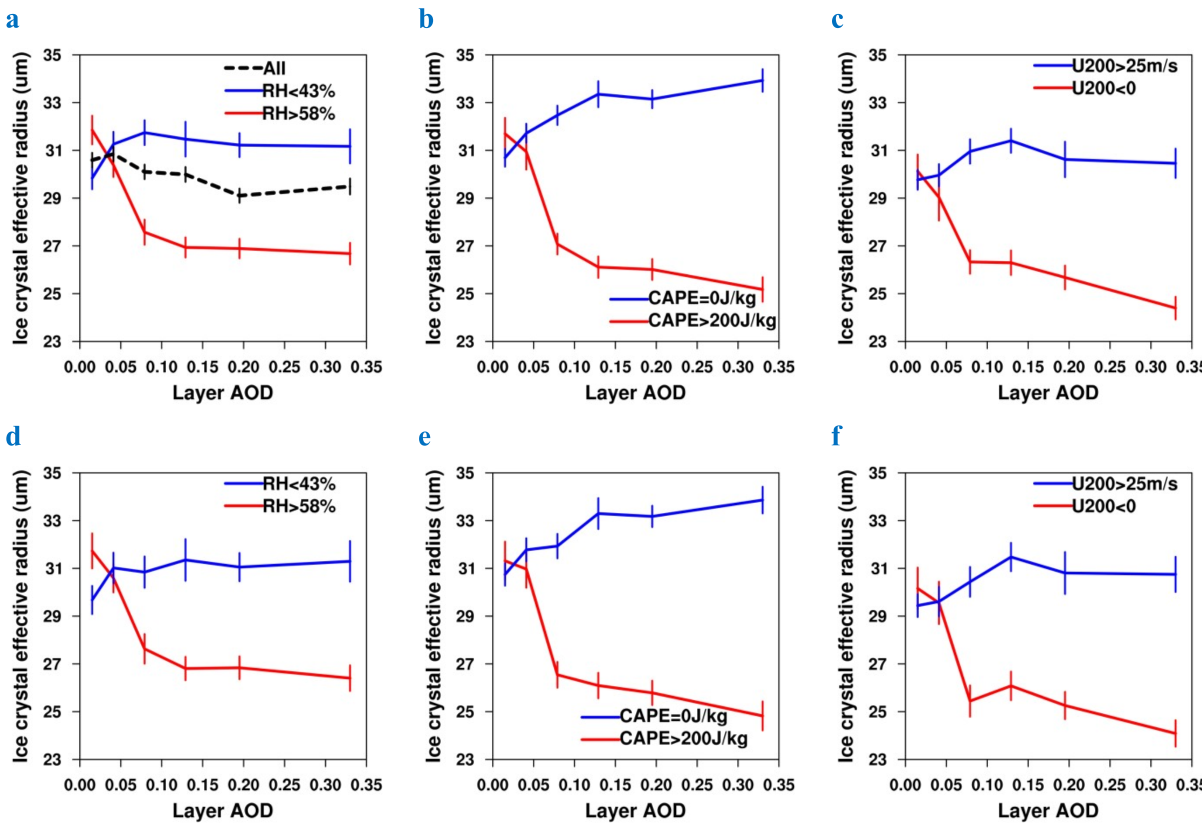

Figure R2. Changes in the R_{ei} of in-situ ice clouds with layer AOD for different ranges of (a) RH averaged between 100 hPa and 440 hPa, (b) CAPE, and (c) U200. (d-f) The same as (a-c) but for the profiles with dust aerosols only. The meteorological parameters are divided into 3 ranges containing similar numbers of data points, and the curves for the medium meteorological range are not shown. The error bars denote the standard errors ($\sigma/\sqrt{N}$) of the bin average, where σ is the standard deviation and N is the sample number. Note that we use AOD of the aerosol layers mixed with ice clouds rather than column AOD, since in-situ ice clouds are primarily affected by aerosols at the ice cloud height. The total number of samples used in this figure is 1.09×10^4.

6) Section 3.2: Rei is positively related to aerosol optical depth (AOD) under relatively dry conditions up to column AOD ~ 0.5 for convective ice clouds and up to ~ 0.13 AOD for in situ ice clouds. These Rei-AOD relationships in Fig. 1, 3 and 4 (for drier conditions) appear to result from competition effects between heterogeneous ice nucleation (henceforth het) and hom, where hom prevails at low AOD and het prevails at higher AOD. As het overtakes hom, Rei increases and ice crystal number concentration, Ni, decreases. This is known as the negative Twomey effect as first described by Kärcher and Lohmann (2003, JGR). Please explain this more thoroughly, citing this paper.

Response: Following the reviewer's comment, we have explained this process when describing both observational data and simulation results. Further, we have mentioned that this is known as the "negative Twomey effect" as first described by Karcher and Lohmann (2003). (Page 11, Line 1-7; Page 16, Line 9-19)

7) Section 3.2: Please state what percentage of the samples were convective vs. in situ.

Response: In response, the convective, in-situ, and other ice clouds account for 44.9%, 52.4%, and 2.7% for all ice cloud profiles. (Page 11, Line 25-27 in the revised manuscript)

8) Page 10, lines 32-33: For moist conditions in Fig. 3, this decrease in Rei with increasing AOD is no more than 2 microns, and the error bars show $\sigma/\sqrt{N}$, not σ (σ should be shown for meaningful interpretation). It is hard to argue that a significant decrease in Rei has occurred with increasing AOD.

Response: We have explained why we used standard error instead of standard deviation for the error bars in our response to the reviewer's 3rd comment. Based on the Student's t-test, the decreasing trends in R_{ei} under moist conditions (high RH, high CAPE, or negative U200) are all statistically significant at the 0.01 level. We have added the statistical test results in the revised manuscript. (Page 12, Line 21-24)

More discussion is needed here. As described in Kramer et al. (2016, ACP) and Luebke et al. (2017, ACP), anvil cirrus are a type of "liquid origin cirrus" where liquid cloud droplets contribute to the ice phase as they vertically advect into the cirrus zone (T < 235 K), freezing as they enter this zone. Ice particles from lower levels can also advect into the cirrus zone, especially for anvil cirrus. Cirrus ice from both sources can be viewed as "pre-existing ice" from a nucleation purview, which provides considerable ice surface area that suppresses the increase of ice supersaturation and prevents the RHi threshold for hom from being attained (Shi et al., 2015, ACP). For this reason, any new ice crystals formed in anvil cirrus are generally expected to form through het or ice crystal multiplication processes. This appears valid for both drier and moist conditions.

Lawson et al. (2015, JAS) combine laboratory measurements, in situ observations and modeling to show that Ni in tropical, convective cumulus clouds is dominated by ice multiplication, which may explain the relatively flat behavior of Rei for high CAPE, high RH and negative U. For such moist conditions, cloud droplets may grow to larger sizes required for ice multiplication. Ice crystals produced this way may be advected by the updraft into the anvil cirrus.

For zero CAPE, lower RH and positive U, ice multiplication may be less important (due to smaller droplet sizes), allowing Rei to increase with increasing AOD, characteristic of hom being overtaken by het (negative Twomey effect). For AOD > 0.4, Rei decreases in accord with het and increasing IN (positive Twomey effect expected when het prevails).

Please expand your discussion to include these points when discussing Fig. 3.

Response: The Reviewer's points are well-taken. The onset of ice multiplication may suppress or even prevent homogeneous nucleation to occur. In this situation, the rather weak decreasing trend in R_{ei} with increasing AOD can be explained, since ice multiplication is supposed to be stronger at the lower AOD that favors the formation of large cloud droplets required for ice multiplication (Lawson et al., 2015; Koenig, 1965, 1963).

We would like to submit that homogeneous nucleation might also play a role under moist conditions. To elucidate this point, we first need to clarify the meaning of "homogeneous nucleation" and "heterogeneous nucleation" with respect to convection-generated ice clouds in this study. The ice crystals in convection-generated ice clouds could be formed via several pathways. On one hand, ice crystals are produced by heterogeneous freezing of liquid droplets at temperatures larger than about –35 ^{o}C or possibly by homogeneous freezing of liquid droplets at about –35 ^{o}C (Kramer et al., 2016). The ice crystals are then lifted to the temperature range < –35 ^{o}C and are considered to be cirrus clouds (Kramer et al., 2016). On the other hand, an additional freezing of solution particles (in contrast to liquid droplets in the former case) may occur in the presence of "preexisting ice" if the updraft is sufficiently strong. The freezing mechanism is likely homogeneous nucleation, since INPs have already been consumed (Kramer et al., 2016). The reviewer points out that such additional freezing events are very difficult to occur and hence make less important contributions to ice crystal budget (Luebke et al., 2016), since the pre-existing ice suppresses supersaturation and prevents the threshold for homogeneous nucleation to take place (Shi et al., 2015). In this study, "homogeneous nucleation" refers to freezing of liquid droplets near the –35 ^{o}C isotherm as well as the freezing of solution particles below –35 ^{o}C. The former could be important for ice formation under moist conditions, because any liquid droplets would be homogeneously nucleated when they are lifted to the –35 ^{o}C isotherm. Evidence for homogeneous droplet freezing has been frequently observed in deep convective clouds and convection-generated cirrus clouds (Twohy and Poellot, 2005; Heymsfield et al., 2005; Rosenfeld and Woodley, 2000; Choi et al., 2010). In particular, liquid droplets are frequently observed to supercool to temperatures approaching –35 ^{o}C and even below, and at slightly colder temperature only ice is found, which serves as strong evidence for homogeneous droplet freezing (Rosenfeld and Woodley, 2000; Choi et al., 2010). Even if the occurrence frequency of homogeneous droplet freezing is low, its contribution to ice number concentration and R_{ei} may still be substantial in view of the fact that numerous ice crystals can be produced in a single homogeneous nucleation event. Under the situation dominated by homogeneous nucleation, the relatively flat response of R_{ei} to AOD (as compared to in-situ ice clouds) can also be explained as the mass fraction of homogeneously formed ice crystals is much smaller than that for in-situ ice clouds, as a result of substantial growth of heterogeneously formed ice crystals before reaching –35 ^{o}C isotherm. Whether the ice formation under moist conditions is dominated by homogeneous nucleation or ice multiplication is clearly dependent on environmental conditions such as updraft velocity, water vapor, cloud height and thickness, etc, a subject requiring further research.

We agree with the reviewer that, in dry conditions, ice multiplication may be less important due to smaller droplet sizes, therefore the increases in R_{ei} with increasing AOD are mainly attributable to the competition between heterogeneous and homogeneous nucleation in line with our original explanation. At a large AOD range (> 0.5), heterogeneous nucleation dominates and a further increase in aerosols would decrease R_{ei} due to the formation of more numerous and smaller ice crystals ("Twomey effect").

We have added the preceding discussions in the revised manuscript. (Page 12, Line 24-26; Page 12, Line 31 to Page 13, Line 22; Page 13, Line 32 to Page 14, Line 14)

9) Section 3.2, Fig. 4: For AOD < 0.10, the in situ cirrus Rei behavior for lower RH, zero CAPE and positive U could be interpreted as a negative Twomey effect with het overtaking hom due to increasing IN. For AOD > 0.10, if IN conc. is proportional to AOD, the trend should reverse with Rei decreasing with increasing AOD. This does not occur, and there is no evidence that the layer AOD is proportional to dust conc. As noted earlier. Thus it is possible that IN concentration is not tracking the layer AOD, and that IN conc. is relatively constant with AOD. This might explain the relatively flat Rei behavior for AOD > 0.10. Please point this out in the paper.

For the in situ cirrus Rei behavior for higher RH, higher CAPE and negative U (red curves), the interpretation given in this paper makes some sense. The freezing of solution droplets (i.e. hom) may be largely responsible for the decrease in Rei with increasing layer AOD.

Response: For AOD > 0.10 at lower RH, zero CAPE and positive U200, the trends in R_{ei} are not statistically significant judging from overlapping error bars. To evaluate the assumption that the INP concentration is not tracking layer AOD, which could explain these relative flat trends, we plot the relations between R_{ei} and layer AOD for dust only in Fig. R2d-f (shown in the response to the reviewer's 5th comment). We find that they are similar to the R_{ei}-layer AOD relations for all aerosol types (Fig. R2a-c). Since the INP concentrations certainly tracks AOD for dust aerosols, the above-mentioned assumption does not appear to account for the insignificant R_{ei} trend at AOD > 0.10. It appears likely that the layer AOD is not large enough, the environmental condition is not sufficiently dry, or the number of samples is not large enough to produce a significant decrease in R_{ei} with increasing layer AOD.

10) Page 11, lines 23-26: As stated in the paper, convective clouds vertically advect ice formed via het across the -35 C isotherm, but this "pre-existing ice" greatly suppresses supersaturations and generally prevents the RHi from reaching the RHi threshold for hom (Shi et al., 2015, ACP).

This may be true even for the "moist" convective conditions. Please include these points in the discussion (Sect. 3.2).

Response: The suggestion is well taken and we have included this point in the revised manuscript. (Page 13, Line 7-10)

11) Page 11, lines 29-32: Please state what percentage of sampled clouds were convective vs. in situ for each season. This is important for understanding the regional radiative implications of this work.

Response: The percentages of ice cloud profiles that are convection-generated type are 38.2%, 48.1%, 51.4%, and 39.1% in winter, spring, summer, and fall, respectively. The corresponding percentages for in-situ ice clouds are 57.0%, 49.6%, 47.0%, and 58.2%, respectively. We have included these descriptions in the revised manuscript. (Page 14, Line 32 to Page 15, Line 2)

12) Section 3.3, Fig. 5b: As noted under (8), ice multiplication can explain the relatively flat behavior of Rei during summer, and perhaps for spring and fall for AOD > 0.4. During winter, CAPE is much lower (see Fig. 5e), suggesting ice multiplication is less important here and Rei decreases for AOD > 0.4 in accord with het and increasing IN. For AOD < 0.4 during winter, spring and fall, Rei increases with increasing AOD, characteristic of hom being overtaken by het. (neg. Twomey effect). Please note this in the paper in regards to Fig. 5b.

Response: Following the reviewer's suggestion, we have included the following descriptions in the revised manuscript. (Page 15, Line 5-11)

"For convection-generated ice clouds, in winter, spring and fall, R_{ei} generally increases when AOD < 0.5, characteristic of homogeneous nucleation being overtaken by heterogeneous nucleation, while R_{ei} decreases slightly when AOD > 0.5 in accordance with heterogeneous nucleation and increasing INP concentrations. In summer, R_{ei} shows a weak decreasing trend with AOD, which could be explained by the domination of homogeneous nucleation or ice multiplication as described in Section 3.2."

13) Section 3.3, Fig. 5c: The summer in situ cirrus Rei behavior could be interpreted as a Twomey effect resulting from het and increasing IN, where deep convection injects more IN into the upper troposphere, thus promoting het. The deep convection during summer promotes tropospheric mixing, making it more likely that IN concentrations at cirrus levels track the layer AOD. It could also be argued that the flat in situ behavior during other seasons could be an indication that IN concentration is not tracking the layer AOD, and that IN concentration is

relatively constant with AOD (otherwise, an initial increase in Rei should be followed by a decrease in Rei as AOD increases). The different Rei values could then be explained in terms of seasonal differences in IN concentration, with lowest IN concentration in winter and highest in summer. Please discuss these points in the paper.

Response: Summer is characterized by relatively moist conditions (high RH, high CAPE, negative U200), in which homogeneous nucleation prevails for in-situ ice clouds as supported by both satellite data analysis and cloud parcel modeling. The "Twomey effect" resulting from heterogeneous nucleation and increasing INP should primarily occur at the dry condition with a large aerosol loading (see the reviewer's 8th and 12th comments and our replies). In other seasons, the weak correlation of INP concentration and layer AOD may partly explain the relative flat behavior of R_{ei}. Another probable reason for the weak R_{ei} trends is that each season consists of varying meteorological conditions (Fig. 4d-f). As shown in Fig. 3d-f, the decreasing trends in R_{ei} under moist conditions are strong, while the increasing trends under dry conditions are relatively weak. Even if the occurrence frequency of dry conditions is large in a season, say winter, the integration of all meteorological conditions may still yield a relative flat R_{ei}-aerosol relationship. We have described the seasonal variations in R_{ei}-layer AOD relations and the possible reasons in the revised manuscript. (Page 15, Line 11-20)

14) Page 13, lines 11-14: These modeling results may not apply to anvil cirrus for the reasons stated in (8) and (10). That is, Ni and Rei in anvil cirrus may be dominated by het and ice multiplication processes. Pre-existing ice should suppress RHi, suppressing hom, making the modeling results irrelevant to anvil cirrus.

Response: Thank you. We have clarified this point in the revised manuscript. (Page 16, Line 22-31)

"The current cloud parcel model simulates the environmental conditions and physical processes for in-situ ice clouds. For convection-generated ice clouds, the competition between homogeneous and heterogeneous nucleation may explain the observed R_{ei}-aerosol relations especially at dry conditions; however, the formation of this ice cloud type involves additional complex physical processes. As described in Section 3.2, ice multiplication together with heterogeneous nucleation may play an important role and dominate the ice formation in moist conditions. Furthermore, ice crystals in convection-generated ice clouds are formed primarily by freezing of liquid droplets rather than nucleation on solution particles."

15) Page 13, lines 23-28: The modeled Rei values for in situ cirrus clouds are ~ 1/3 those retrieved in this study for such clouds (and are typically ~ 1/3 or less of those from aircraft sampling of in situ cirrus clouds; e.g. Mishra et al., 2014, JGR). For a 30 minute simulation time,

the predicted values appear unrealistic. Isometric ice crystals grown at -22 ℃ reach ~ 100 microns in size after 10 minutes (Takahashi et al., 1991, J. Meteor. Soc. Japan), and would be much larger had the growth times been extended to 30 minutes. While growth rates will be lower at cirrus cloud temperatures, and vapor competition effects can limit growth rates, 30 minutes of growth time should still produce Rei values typical of cirrus clouds, which typically range from 10 and 45 μm at cirrus cloud temperatures based on aircraft measurements (Mishra et al., 2014, JGR).

The small Rei values imply very high Ni (assuming typical IWCs). Please also plot Ni vs. aerosol number conc. and comment on the realism of the Ni and IWC values.

The text here states variable updraft velocities as a possible reason for the small Rei predicted, but Sect. 2.3 states that a constant updraft velocity (w) of 0.5 m/s is applied throughout the 30 minute simulation time. The parcel model here is simulating in situ cirrus clouds, and w = 0.5 m/s is very high and not representative for in situ cirrus clouds. Hom is most sensitive to the cooling rate or w, and this simulation strongly favors hom due to the high w assumed. Hom can partly explain the small Rei values, but only when hom dominates. It cannot explain the black curve in Fig. 6a where het dominates for aerosol conc. above 200 cm-3; Rei should be ~ 3 times larger here. To summarize, the simulation here is not representative of in situ cirrus clouds and thus should not be used to interpret the satellite measurements.

Response: We have substantially improved the model simulations to make them representative of in-situ ice clouds including consideration of vertical velocity fluctuations and water vapor exchanges with other air parcels.

We conduct two groups of numerical experiments with different available water amount for ice formation, denoted by initial water vapor mass mixing ratios (pv). Each group is comprised of 100 sub-groups with initial sulfate number concentrations increasing linearly from 5 cm^{-3} to 500 cm^{-3}. The concentration ratios of externally mixed dust (deposition INP), coated dust (immersion INP), and sulfate (not INP) are prescribed with values of 0.75:0.25:10000 for all experiments, since INPs represent only 1 in 10^3 to 10^6 of ambient particles (Fan et al., 2016). In each sub-group, we conduct 100 one-hour experiments driven by different vertical velocity spectra following the approach described by Shi and Liu (2016). The vertical air motions at a 10 s resolution were retrieved from Millimeter Wave Cloud Radar (MMCR) observations at a site located in the Southern Great Plains (SGP; 36.6°N, 97.5°W) for a 6 h period (Shi and Liu, 2016). For each of the 100 experiments, we randomly sample a 1 h time windows from the 6 h vertical velocity retrievals, subtract the arithmetical mean, and adjust the standard deviation to 0.25 $m\ s^{-1}$. The sampled vertical velocity spectra are subsequently added a constant large-scale updraft velocity of 0.02 $m\ s^{-1}$ to drive the parcel model.

The model assumes that the air parcel has no mass or energy exchange with the environment except for sedimentation of ice crystals, which is not realistic. For example, the outburst of homogeneous nucleation in an air parcel can quickly exhaust supersaturation and take water vapor from surrounding parcels. To conceptually mimic this process, we have divided the 100 experiments within a sub-group into 10 combinations, each consisting of 10 experiments. It is assumed that the air parcels in the same combination can exchange water vapor and reach equilibrium. Consequently, the occurrence of homogeneous nucleation in one parcel will suppress the homogeneous nucleation in the connected parcels due to the depletion of water vapor.

Figure R3a,b shows the simulated changes in R_{ei}, ice crystal number concentration (N_i), and the fraction of ice crystal number produced by heterogeneous nucleation as a function of the total aerosol number concentration. The N_i for a given aerosol number concentration (i.e., a sub-group of experiments) is calculated using an arithmetical mean of the 100 experiments, while R_{ei} is calculated from mean N_i and mean ice volume. At moist condition (pv = 103 ppm), R_{ei} decreases with increasing aerosol concentration, attributed to the "Twomey effect" when homogeneous nucleation dominates. At dry condition (pv = 78 ppm), R_{ei} increases with small-to-moderate aerosol loading, indicative of homogeneous nucleation overtaken by heterogeneous nucleation, and decreases with further aerosol increase in accordance with heterogeneous nucleation and the "Twomey effect". More importantly, the simulated magnitude of R_{ei} has been close to satellite observations (Figs. 1 and 3 in the revised manuscript), mainly due to the consideration of variable vertical velocities and water vapor exchanges between parcels. The mean N_i (across 100 experiments in each sub-group) of 10 to 250 L^{-1} (Fig. R3b) and mean ice water content of 1 to 24 ppm (not shown) are within the range reported in Kramer et al. (2016) based on a series of aircraft measurements.

To evaluate the effect of the assumption concerning water vapor exchange, we have performed a group of similar sensitivity simulations, except that the water vapor exchanges between parcels are deactivated (Fig. R3c,d). When heterogeneous nucleation dominates (i.e., pv = 78 ppm, large aerosol loadings), the magnitude of R_{ei} is similar regardless of the treatment of water vapor exchange. However, when homogeneous nucleation plays an important role, the simulated R_{ei} without water vapor exchange (Fig. R3c) is much smaller than the baseline simulation (Fig. R3a). We also note that the simulated R_{ei} is larger than the value shown in our previous manuscript even if water vapor exchange is not accounted for (Fig. R3c), because fluctuated vertical velocity spectra are applied here, resulting in overall less frequent and weaker homogeneous nucleation as compared to a large constant vertical velocity of 0.5 m s^{-1} used in our original manuscript.

We have added the methods and results of the new model simulations into the revised manuscript. (Fig. 5; Page 8, Line 5-33; Page 15, Line 28 to Page 16, Line 21)

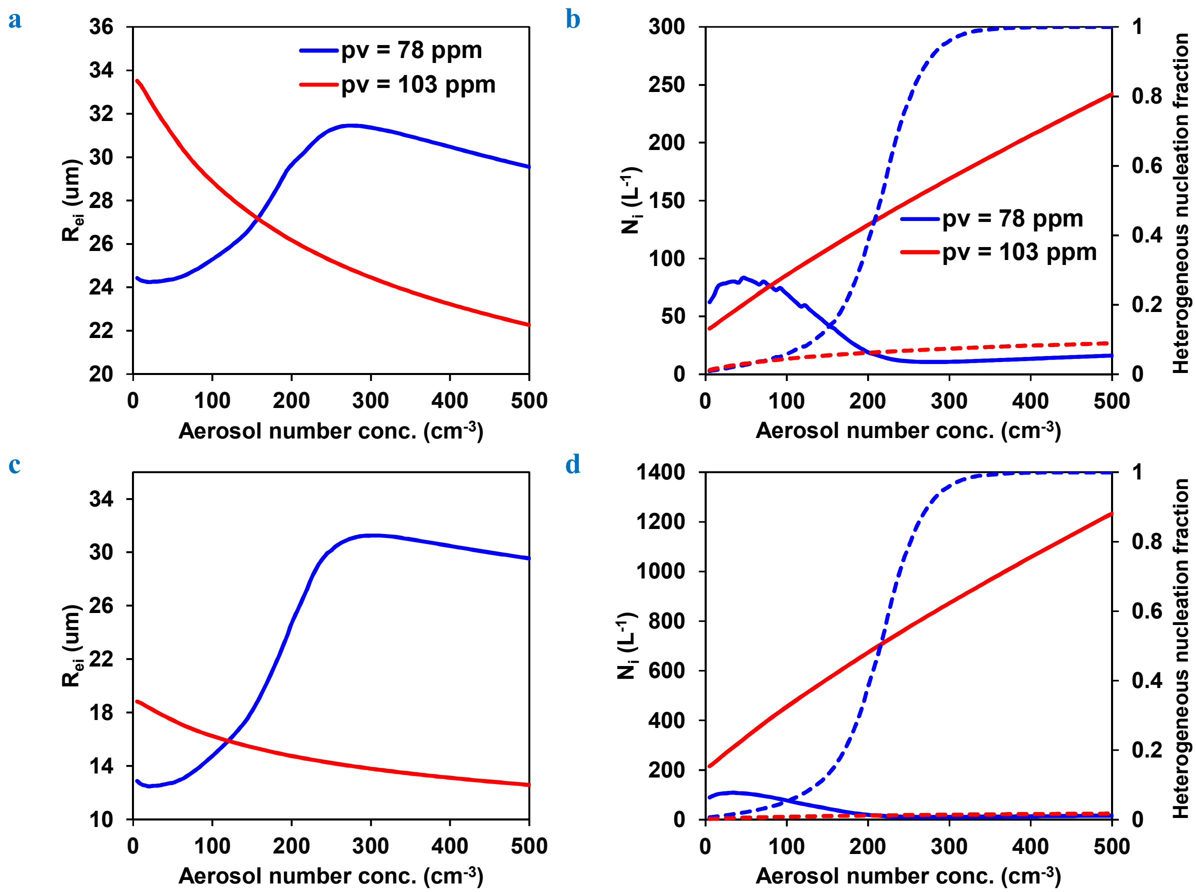

Figure R3. Simulated changes in (a) ice crystal effective radius (R_{ei}) and (b) ice crystal number concentration (N_i) and the fraction of ice crystal number produced by heterogeneous nucleation as a function of the total aerosol number concentration. (c-d) is the same as (a-b) except that the water vapor exchange between parcels is not accounted for. Simulations are conducted for two initial water vapor mass mixing ratios (pv) for ice formation. The ratios of externally mixed dust (deposition INP), coated dust (immersion INP), and sulfate (not INP) are prescribed with values of 0.75:0.25:10000 in all experiments.

16) Section 3.4, Fig. 6a: The beginning of the black curve is a manifestation of the "negative Twomy effect" (Karcher & Lohmann 2003, JGR) as hom is overtaken by het. The slope should become negative after aerosol conc. exceeds 200 cm-3 as increasing IN increases Ni, reducing Rei, but this does not happen. It is not clear why Rei does not decrease in this region.

Response: The decrease in R_{ei} in now shown in our new simulation results (blue line in Fig. R3a), as described in our response to the reviewer's 15th comment. This does not happen in the original simulations because the aerosol concentrations were not large enough to initiate a decline in R_{ei} in the context of a large constant vertical velocity of 0.5 m s^{-1}.

17) Section 3.4, Fig. 6b: As per my understanding, the initial water vapor mass mixing ratio (pv) determines the level of condensation and thus the portion of the 30 min. simulation time available for supersaturation development. In general, the INP concentrations assumed are sufficiently low to allow attainment of the hom RHi threshold, except for the pv = 38 ppm simulation which has less time for supersaturation development. If this is correct, then please make this clear in the text for greater clarity among the readership. In general, if this modeling section can be made relevant to in situ cirrus clouds, it needs to be expanded and explained better.

Response: We agree with the reviewer's explanation. For new simulations, the physical conditions are more complicated than the preceding description because the supersaturation is not always increasing under fluctuated vertical velocities. However, the general idea of "supersaturation development" is still applicable. We have revised the text as follows:

"With an adequate water vapor supply (pv = 103 ppm), the onset of deposition and immersion nucleation consumes only a small fraction of water vapor due to the small INP population. Considerable supersaturation remains. After further updraft movement, homogeneous nucleation is triggered and occurs spontaneously over a higher and narrow ice supersaturation range (140-160%). Therefore, homogeneous nucleation acts as the dominant ice formation pathway, as indicated by the very small number fraction (< 10%) of heterogeneously formed ice crystals, shown in Fig. 5b. With an inadequate water vapor supply (pv = 78 ppm), the occurrence of heterogeneous nucleation could consume a considerable fraction of water vapor such that the remaining supersaturation is quite low and would require extremely strong updraft to uphold the homogeneous nucleation threshold. When aerosol loading increases, homogeneous nucleation is gradually suppressed and reduced to a minimum." (Page 16, Line 1-16)

18) The following reference: "Ikawa, M., and Saito, K.: Description of a Non-hydrostatic Model Developed at the Forecast 38 Research Department of the MRI, Meteorological Research Institute, Tsukuba, Ibaraki, 39 Japan, 1991." is unconventional, and I wonder whether this is readily accessible. Can it be improved?

Response: We are sorry to report that this is the only reference we have found which describes the sedimentation scheme in detail. It is accessible to the public at http://www.mri-jma.go.jp/Publish/Technical/DATA/VOL_28/28_en.html. We have added the URL to the reference list.

Minor Comments:

1) Page 4, line 26: This might be a good place to state that your samples are strictly single-layer ice clouds, instead of at the end of this paragraph.

Response: Done, thank you. (Page 5, Line 16-18)

2) Page 5, line 1: Does cloud type assignment depend exclusively on the way it is flagged?

Response: The profiles of deep convective clouds are identified exclusively based on the "cloud type" flag in CALIPSO products, whereas the ice cloud profiles are identified following the standard that the "cloud type" flag is "cirrus" or its layer base temperature is colder than –35 ^{o}C. (Page 5, Line 5-6, Line 22-24)

3) Page 9, lines 15-16: Please indicate that U is the zonal wind as opposed to the meridional wind, and that positive U implies westerly winds; negative U implies easterly winds.

Response: Done, thank you. (Page 10, Line 18-20)

4) Page 12, line 32: Poor sentence; fix grammar. Should say something like "formation of more numerous and smaller ice crystals."

Response: Done, thank you. (Page 16, Line 7-9)

5) Page 13, line 15: Suggest replacing "discrepant" with "different" here and elsewhere throughout the paper.

Response: Done, thank you.

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