Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-548-RC2, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "A water vapor modulated aerosol impact on ice crystal size" *by* Bin Zhao et al.

Anonymous Referee #2

Received and published: 28 August 2017

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.

Major Comments:

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

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.

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.

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.

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.

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.

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

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.

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.

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.

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).

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.

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.

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.

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.

15) Page 13, lines 23-28: The modeled Rei values for in situ cirrus clouds are $\sim$ 1/3 those retrieved in this study for such clouds (and are typically $\sim$ 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 °C reach $\sim$ 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 $\sim$ 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.

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.

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.

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?

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.

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

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.

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

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

throughout the paper.

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