Response to Reviewer #1

We appreciate your time for thoroughly reviewing our manuscript. We would like to thank you for the constructive comments and suggestions that help to improve the manuscript. The manuscript has been revised accordingly. The reviewer's comments are provided in black text and our responses are provided in blue text.

Response:

I would like to thank the authors for the different answers to the comments and the changes made in the article. The quality of the paper has increased and the results are now more robust and clear. Nevertheless, some of my comments have not been answered adequately or are not mentioned in the final article. I would like the authors to comment on few points I am referring to here.

From the first general comment answer:
The uncertainty for Nd of 25 % is considered by the author, I find this value excessively low, especially when compared with Grovesnor et al. (2017) values: Their review ranges the uncertainty of Nd from 20 % to 75% from ground based radar measurements, please refer to Section 5 from their paper. The uncertainty of LWP leads to an uncertainty on Nd much greater than 25 % (especially for LWP in the range considered in the study). Also in Dong et al. (1997), they conclude to an uncertainty on Nd of 36 %. I would like to see a deeper analysis on the uncertainty of Nd for the different parameters. Also, I think the paragraph on the uncertainty from the answer should appear in the article and not only the last part about re.

Thanks for the comments.

As reviewed in Grovesnor et al. (2017), Schmidt et al., (2014) used a dual-Field-of-View Raman lidar technique to retrieve r_e and further computed N_d following the equation $N_d = \frac{\alpha}{2\pi l r_e^2}$. They found that the large errors in r_e (25% - 40%) contribute to the N_d relative uncertainty of 50% - 80%. Determination of cloud microphysical properties relies on measurements and calculation methods.

In Dong et al. (1997), the r_e was retrieved based on the method involving spectroradiometer transmittance and LWP. The 10% change in cloud LWP and downward SW at the surface would result in the 10% uncertainty in r_e retrieval.

For the original N_d retrieval in Dong et al. (1997), the uncertainties of cloud droplet concentrations were up to 36%, which were estimated by exerting random perturbations onto the input variables in the retrieval, and then compared the ratios of the standard deviation to the mean. This value of 36% rather represents the sensitivity of N_d to random errors in the retrieval inputs than the uncertainty compared to in-situ measurements.

In this study, the 25% uncertainty of N_d is adapted from the previous experiments compared with the aircraft in situ measurements at the Penn State surface site during the Fall 1996 (Dong et al. 1998) and at the ARM SGP site during March 2000 Cloud Intensive Observational Period (IOP) (Dong et al. 2002; Dong and Mace, 2003). The 25% uncertainty in N_d is statistically estimated based on 5 hours of aircraft in situ measurements during the Fall 1996 and 10 hours of aircraft data during the March 2000 IOP.

Regarding the expression used in this study to compute N_d ($N_d = \left(\frac{3LWP}{4\pi\rho_w r_e^3 \Delta Z}\right)\exp(3\sigma_x^2)$), the uncertainties of input parameters are as follows: 0.15 for logarithmic width of the droplet size distribution (Miles et al., 2000); 20 gm^{-2} for LWP (Liljegren et al. 2001); 60 m for cloud thickness (Widener et al., 2012); and 10% for r_e (Dong et al., 2002).

To assess the contributions of different parameters' uncertainties to N_d retrieval, every input parameter was perturbed by its uncertainty with other parameters held fixed, and then we recalculated N_d for all the samples in this study. The results are presented in the Table below.

	N_d (cm^{-3})
Baseline	297
$\sigma_x + 0.15$	448 (50.8%)
$\sigma_x - 0.15$	226 (23.9%)
$LWP + 20$ (gm^{-2})	380 (27.9%)
$LWP - 20$ (gm^{-2})	215 (27.6%)
$r_e + 10\%$ (μm)	224 (24.6%)
$r_e - 10\%$ (μm)	408 (37.4%)
$\Delta Z + 60$ (m)	254 (14.5%)
$\Delta Z - 60$ (m)	366 (23.2%)

The baseline value is the mean value of N_d from all the samples (same as in Fig. 2). The values in the right column denote the retrieved N_d after perturbing the input variables by their estimated uncertainties. The values in the parentheses are the percentage changes with respect to the baseline value. As shown in the Table, the percentage changes in N_d are due to different uncertainty inputs which range from 14.5% to 50.8%, with the majority falling between 20% and 30%. Note that the largest uncertainty of N_d happens when σ_x is increased by 0.15. However, considering that continental stratocumulus generally contain smaller droplets, one might expect their distribution width to be smaller than 0.38 as well (Dong et al., 1997). Therefore, considering the combined contribution of input uncertainties to the N_d retrieval, the overall uncertainty of 25% compared to the aircraft in-situ measurement should be a reasonable estimation. In this case, the mean ratio of N_d/N_{CCN} for the weakly absorptive aerosol regime range from 52% to 86%, while the mean ratio of N_d/N_{CCN} for the strongly absorptive aerosol regime range from 41% ~ 67%.

Accordingly, the statement '*As the results, the 10% change in cloud LWP and downward SW at the surface would result in the 10% uncertainty in r_e retrieval. And the N_d uncertainty is*

statistically estimated to be 25%, compared with the aircraft in situ measurements at the Penn State surface site during the Fall 1996 (Dong et al. 1998) and at the ARM SGP site during the March 2000 Cloud Intensive Observational Period (IOP) (Dong et al. 2002; Dong and Mace, 2003)' has been added to Section 2.1.2 in the revised manuscript.

Moreover, the following discussion below has been added to the third paragraph of Section 3.3.4 in the revised manuscript:
'*In addition, the sensitivity and uncertainty of N_d is examined in order to estimate the impact of N_d uncertainty on the assessment of CCN activation rate. To assess the contributions of different input parameter uncertainties to N_d retrieval, every input parameter was perturbed by its own uncertainty with other parameters held fixed. The results are as follows: (a) an increase (decrease) of LWP by 20 gm^{-2} leads to 27.9% (27.6%) change in N_d while an increase (decrease) σ_x by 0.15 leads to a 50.8% (23.9%) change in N_d; (b) an increase (decrease) cloud thickness by 0.15 leads to a 14.5% (23.2%) change in N_d; and (c) an increase (decrease) in r_e by 10% leads to 14.5% (23.2%) change in N_d. The percentage changes in N_d due to different input uncertainties range from 14.5% to 50.8%, with the majority falling between 20% and 30%. Note that the largest uncertainty of N_d happens when increasing σ_x by 0.15. However, when considering that continental stratocumulus generally contains smaller droplets, one might expect their distribution width to be smaller than 0.38 (Dong et al., 1997). Therefore, the overall uncertainty of 25% compared to the aircraft in-situ measurement should be a reasonable estimation. In this case, the mean ratio of N_d/N_{CCN} for the weakly absorptive aerosol regime range from 52% to 86%, while the mean ratio of N_d/N_{CCN} for the strongly absorptive aerosol regime range from 41% ~ 67%.*'

Also, can you make explicit the formula you use to propagate the uncertainty of re on ACI? With a simple equation, I find an uncertainty on ACI between 0.01 and 0.01 for a change in re of 10% and Nccn of 0 % (optimistic), which is not negligible considering the value from figure 8-b and the conclusion in paragraph 3.3.6. I would like to see some clarifications here.

In the previously revised manuscript, we added 10% of r_e on their original values and deducted 10% of r_e on their original values, then re-do the regression to obtain the upper and lower bounds of the ACIs respectively, which are denoted by the dash lines in the original Fig. 8. The results differ from the original ACIs by only ~0.0003. However, this method treated the r_e uncertainty as uniform and did not consider any different circumstances.

In the newly revised manuscript, we use the Monte Carlo method to propagate the r_e uncertainty on ACI. The procedure is given as follows:
1. For each data point, the r_e value is randomly perturbed to be increased or decreased by 10%.
2. The corresponding ACI is re-calculated based on the perturbed set of r_e.
3. Repeat Step 1 & 2 100,000 times.
4. After 100,000 iterations, we obtain a distribution of ACIs
5. The uncertainty of ACI is given by one standard deviation of 100,000 values of ACIs.

Since the distribution of ACIs follows a normal distribution with a narrow peak, this uncertainty value represents the uncertainty in the computed ACI due to error in r_e retrieval.

The uncertainties of ACI for both regimes under the two LWP ranges are denoted in the revised Fig. 8 below:

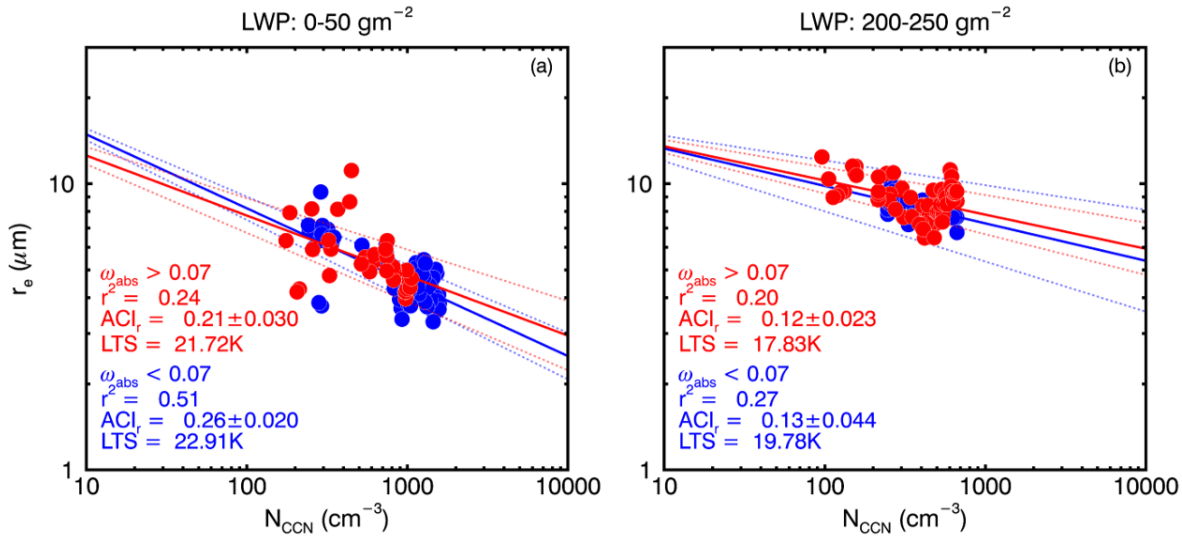

Figure 8. r_e as a function of N_{CCN} and the values of ACI_r under the strongly absorptive (in red) and weakly absorptive (in blue) aerosol regimes at two LWP bins: 0-50 g m^{-2} (a) and 200-250 g m^{-2} (b). Note that the dashed lines denote the uncertainties of ACI_r due to 10 % error in r_e retrieval.

In the lower LWP range (Fig. 8a), the ACI uncertainty is 0.02 (0.03) for weakly (strongly) regime, account for the uncertainties, the difference in ACI between two absorptive regimes is preserved. In the higher LWP range (Fig. 8b), the ACI uncertainty is 0.04 (0.02) for the weakly (strongly) regime, which is non-negligible. Taking the uncertainties of ACI into account, the ACI in the two absorptive regimes cannot be well separated, owing to the enhanced collision-coalescence process accompanied by higher LWP and the diminished cloud response to aerosol associated with different ω_{abs} values. In general, the 10% uncertainty in r_e retrieval contributes to 0.02 ~ 0.04 in ACI uncertainties.

Accordingly, the following discussion has been added to the second paragraph of Section 3.3.6 in the revised manuscript.

'*In order to assess the impact of r_e uncertainty on ACI_r, we use the Monte Carlo method to propagate the r_e uncertainty on ACI_r, with the procedure given as follows: For each data point, the r_e value is randomly perturbed to be increased or decreased by 10%, and thus the corresponding ACI_r can be re-calculated based on the perturbed set of r_e. After 100,000 iterations, we obtain a distribution of ACI_rs. The uncertainty of ACI_r is given by one standard deviation of those 100,000 values of ACI_rs, since the distribution of ACI_rs follows a normal distribution with a narrow peak, this uncertainty value represents the uncertainty in the computed ACI_r due to errors in the r_e retrieval. The uncertainties of ACI_r for the two absorptive regimes are denoted as the dashed line in Fig. 8. In the lower LWP range (Fig. 8a), the ACI_r uncertainty is 0.020 (0.030) for the weakly (strongly) regime, account for the uncertainties, the difference in ACI_r between the two absorptive regimes is preserved. In the higher LWP range (Fig. 8b), the ACI_r uncertainty is 0.044 (0.023) for the weakly (strongly) regime, which is non-*

negligible. Taking the uncertainties of ACI_r into account, the ACI_r in the two absorptive regimes cannot be well separated, owing to the enhanced collision-coalescence process accompanied by higher LWP and the diminished cloud response to aerosols associated with different ω_{abs} values. In general, the 10% uncertainty in r_e retrieval contributes to 0.02 ~ 0.04 in ACI_r uncertainties.'

From the second general comment answer:
In the article (p. 17, l. 3 of the new document), the lack of vertical velocity measurements is mentioned. But, it can be retrieved from ECMWF (with LTS), it would not be directly at cloud base and will concern a large-scale value but did you look at this parameter? I would also be curious to see the effect of humidity on the results. Did you look at other meteorological parameters than LTS which can impact the ACI?

Thanks for the comments.

Yes, we have checked the vertical velocity in pressure coordinate (Omega) values at the 925 hPa level for all sample points, which represent the large-scale forcing on the vertical motion between surface and cloud-layer over the SGP site. The results are shown in Fig. R1 below:

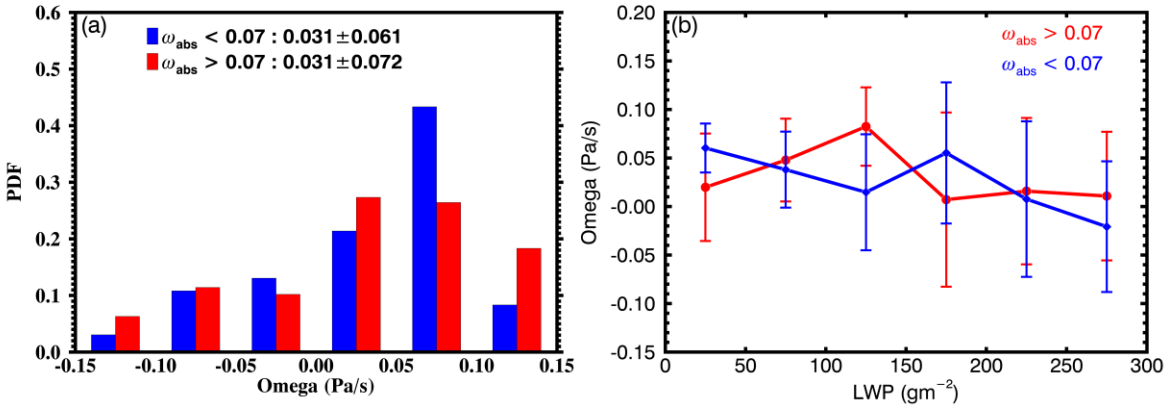

Figure R1

As shown in Fig. R1a, both absorptive regimes shared the same mean value of 0.031 Pa/s with a slightly different distribution. In addition, the majority of Omega values in both regimes are greater than 0, which means the large-scale environments over the SGP are generally dominated by sinking motion. Fig. R1b shows the Omega values sorted by LWP. The vertical velocities are not correlated with ω_{abs} and show no dependence on LWP.

Given that all the cases happened during the Spring and Winter seasons, the distributions of Omega are greatly influenced by large-scale synoptic patterns. To examine the synoptic patterns associated with them, we obtained the geopotential height data from ERA5 reanalysis and applied the composite analysis on the cases that in weakly/strongly absorptive regime. Results for 925 hPa level are shown in the figure below:

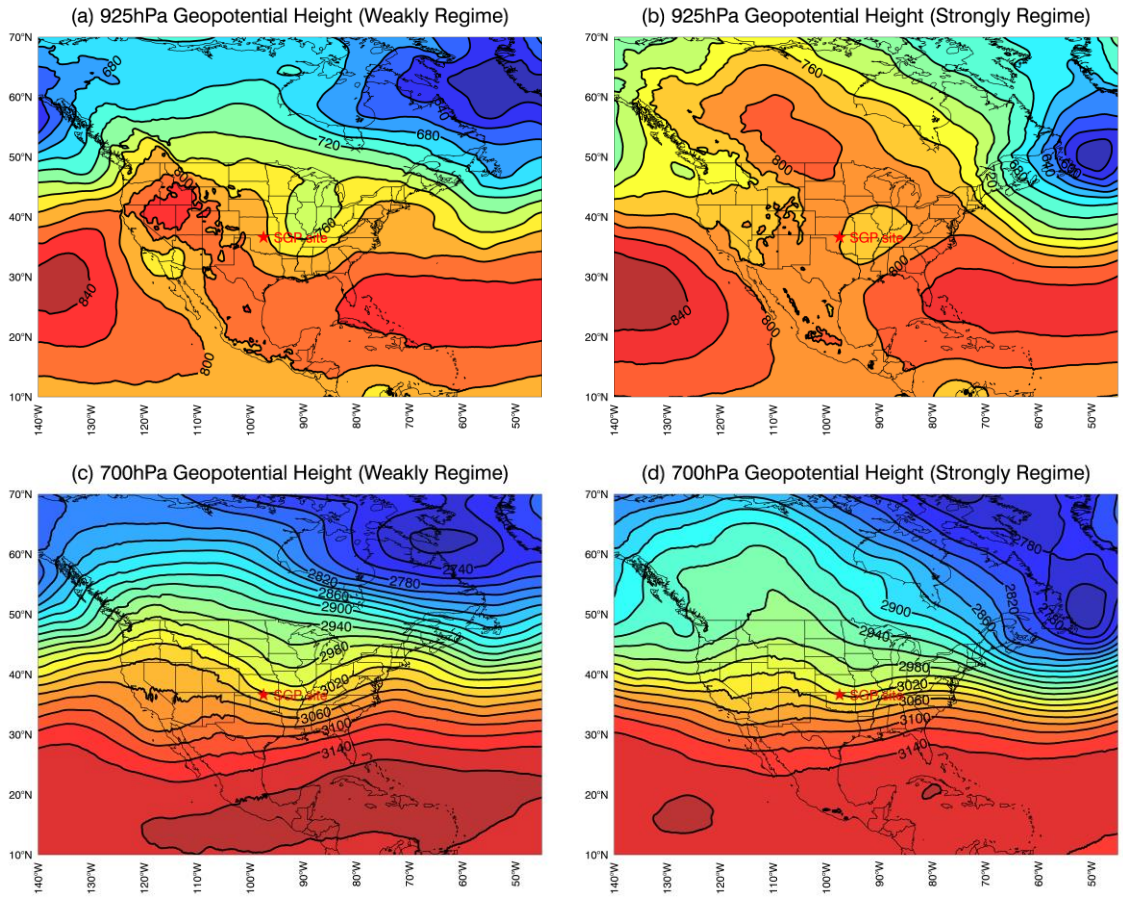

Figure R2

Fig. R2 (a) 925 hPa composite geopotential height in the weakly absorptive regime with the SGP site located ahead of the ridge. (b) 925 hPa composite in the strongly absorptive regime with the SGP site within a large domain of relatively high pressure. As for higher pressure level up to 700 hPa (Fig. R2c & d), the SGP site located ahead of the ridge for both regimes. Therefore, the synoptic patterns are favorable for generation of downward motion at the lower troposphere, and the sinking motion induces relatively stable environments in the lower troposphere which is consistent with the LTS measurements.

Since the Omega value is obtained from a relatively larger domain surrounding SGP, it is hard to reflect the true cloud-scale dynamics, especially the vertical velocity or turbulence strength at the cloud base.

Accordingly, the following discussion has been added to the fifth paragraph of Section 3.3.4 in the revised manuscript.

'In addition, the vertical velocity in pressure coordinate (Omega) values at the 925 hPa level, which represent the large-scale forcing on the vertical motion between surface and cloud-layer, are also sorted by LWP for the two absorptive regimes in order to check the potential influence of the environmental dynamic state (figure not shown). However, the Omega for both absorptive regimes share the same mean value of 0.031 Pa/s and show no dependence on LWP, indicate that the large-scale environments over the SGP are generally dominated by sinking

motion. The synoptic patterns of composite geopotential height for the two absorptive regimes show that the SGP site is located ahead of the 700 hPa ridge and is located within the 925 hPa high. The meteorological pattern is favorable for generation of downward motion at the lower troposphere, and the sinking motion induces relatively stable environments in the lower troposphere which is consistent with the LTS measurements. Considering the fact that the Omega value is obtained from a relatively larger domain surrounding the SGP, it is difficult to reflect the true cloud-scale dynamics, especially the vertical velocity or turbulence strength at the cloud base. Therefore, the influence of cloud-scale dynamics, presumably cloud-base updraft, is not negligible since the sensitivity of cloud droplet to aerosol loading is enhanced with increasing updraft velocity as reported in previous studies (e.g., Feingold et al., 2003; McComiskey et al., 2009).'

Additionally, we also checked the relative humidity (RH) in both regimes to examine whether there is any causality between RH and ACI. The RH is obtained from the Merged-Sounding product at SGP, then calculated as the mean value within the cloud-layer at every sample point, the RH distributions and dependences on LWP in two regimes are presented in the figure below:

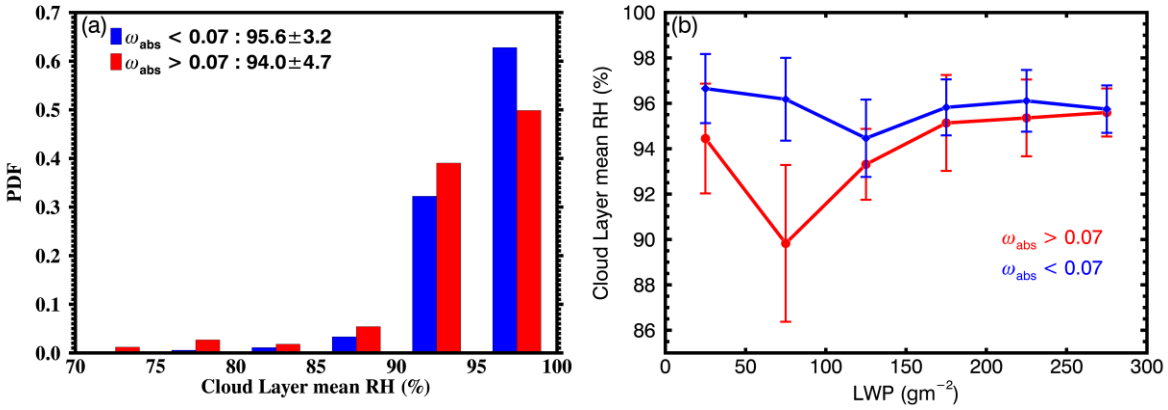

Figure R3

As shown in Fig. R3(a), the cloud-layer mean RH in the weakly absorptive regime share a similar distribution as in the strongly absorptive regime, with a 1.6% difference in the mean value. More samples with relatively lower RH were found in the strongly absorptive regime. When the RH data are sorted by LWP (Fig. R3b), the values in the weakly absorptive regime are also higher than those in the strongly absorptive regime. This discrepancy is in accord with the discussion of the absorptive effect of aerosol on the environment, with the presence of strongly light-absorbing aerosols, the cloud layer heating induced by the aerosol absorptive effect can result in the reduction of in-cloud relative humidity (Wang et al., 2013).

Accordingly, the statement *'Furthermore, the radiative effect of light-absorbing aerosols on the cloud environment cannot be neglected, since the strongly light-absorbing aerosols can absorb solar radiation and heat the in-cloud atmosphere by emission, which results in the reduction of relative humidity (or supersaturation) in the cloud layer (Bond et al., 2013; Wang et al., 2013). This effect is evident by the observation as the values of in-cloud relative humidity*

in the strongly absorptive regime are slightly lower than those in the weakly absorptive regime.' has been added to the sixth paragraph of Section 3.3.4 in the revised manuscript.

The stratus formation is enhanced by high LTS (Klein et al. 1993) and, therefore, more prone to high ACI, which is in line with the results presented. The results show that ACI is a function of LTS and a function of wabs. Moreover, from the Figures (a) and (b) from the answer of the second general comment, LTS and wabs seem correlated with each other. The effect of LTS on ACI does not seem negligible, maybe even larger than the effect of wabs on ACI. I think a discussion is needed about the conclusions of the article and the correlations from observation that are not necessarily causality, and that the observed effect can be inhibited or enhanced (Grysperdt, 2016). You mention it but I think it would help to see the ACI for the two regimes of wabs and constrained for two regimes of LTS to ensure that the results described by Figure 8 are not an artifact.

Thanks for the comments.

In order to constrain the ACI for the two regimes by their LTS values, we adapted the criteria described in Grysperdt et al. (2016) that the LTS value of 18K denotes the demarcation line between high and low LTS regimes. However, the sample points that fall into the low LTS category are highly limited, largely owing to the selected cases. In 0 - 50 gm^{-2} LWP range, only 3(6) samples in weakly (strongly) regime have LTS values that are lower than 18K; in 200 – 250 gm^{-2} LWP range, only 11 (12) samples in weakly (strongly) regime are lower than 18K. Since the calculation constrained in the low LTS regime cannot pass the 95% significant test, the figure below shows the ACI constrained in high LTS regime only.

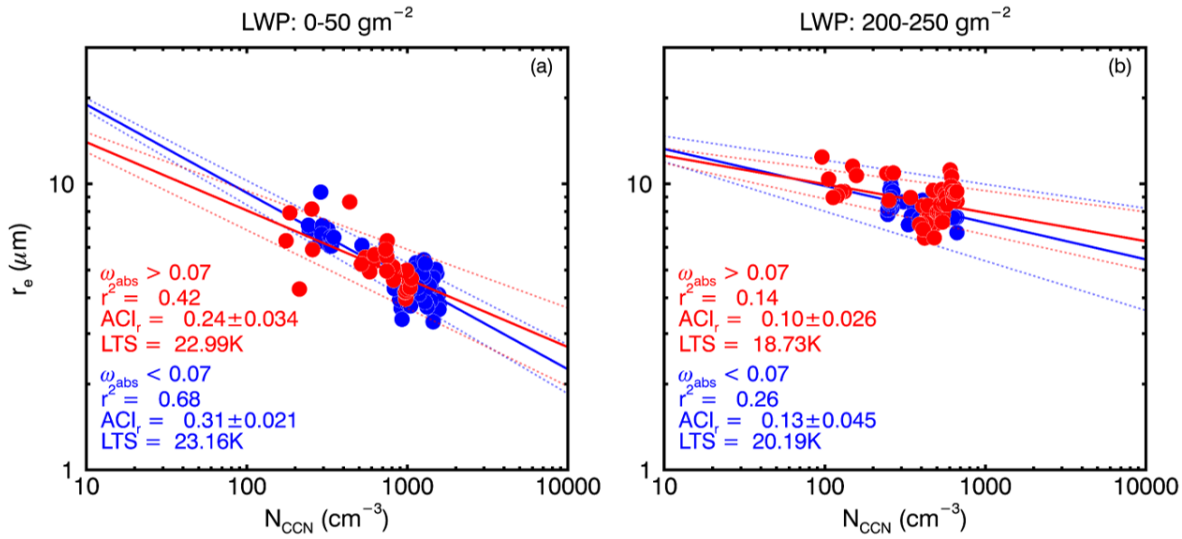

Figure R4

In 0 - 50 gm^{-2} LWP range (Fig. R4a), after considering only the sample points with LTS higher than 18K, the mean values of LTS in the two regimes are closer. The ACI for the weakly absorptive regime is enhanced from 0.26 to 0.31, and ACI for the strongly absorptive regime

is increased from 0.21 to 0.24. The enhancement effect of LTS on the ACI is noticeable, which is in accordance with the previous discussion that a high LTS environment is associated with sufficient turbulence in the boundary layer and a closer connection between surface and cloud layer, and thus enhance the cloud microphysical responses to the CCN below. Note that though ACIs are increased for both regimes, the difference between them becomes larger. Owing to the low-level stratus is more susceptible to weakly absorptive aerosol, the enhancement of ACI induced under a high LTS environment is better manifested in the weakly absorptive regime. As for 200 – 250 gm^{-2} LWP range (Fig. R4b), the LTS effects on ACI are less significant compared to low LWP range. No significant change in weakly absorptive regime and the ACI in strongly absorptive regime decreased from 0.12 to 0.10, partly owing to the enhanced collision-coalescence process accompanied by higher LWP, and thus inhibit the impact of LTS on ACI.

Overall, ACIs are enhanced under the high LTS, but the difference between two regimes do exist, indicating that both ω_{abs} and LTS can be the impact factor of the ACI but they are not necessarily having causality between them.

Accordingly, the following discussion has been added to the third paragraph of Section 3.3.6 in the revised manuscript.

'In order to quantify the impact of LTS on ACI_r, we adapted the criteria described in Grysperdt et al. (2016) that the LTS value of 18K denotes the demarcation line between high and low LTS regimes, and constrain the ACI_r for the two regimes by their LTS values accordingly. Owing to the highly limited sample points that fall into the low LTS category, the ACI_c can only be constrained in the high LTS condition. For the 0 - 50 gm^{-2} LWP range, the ACI_r for the weakly absorptive regime increases from 0.26 to 0.31, and the ACI_r for the strongly absorptive regime increases from 0.21 to 0.24. The enhancement effect of LTS on the ACI_r is noticeable, which in accordance with the previous discussion that high LTS environment is associated with (a) sufficient turbulence in the boundary layer and (b) a closer connection between the surface and cloud layer, which enhances the cloud microphysical responses to the CCN. Note that though ACI_rs are increased for both regimes, the difference between them becomes larger (from 0.05 to 0.07) because low-level stratus clouds are more susceptible to weakly absorptive aerosol. Furthermore, the enhancement of ACI_r in the high LTS environment is more evident in the weakly absorptive regime. In the case of the 200 – 250 gm^{-2} LWP range, the LTS effects on ACI_r are less significant compared to the lower LWP range. No significant change in weakly absorptive regime is evident and the ACI_r in the strongly absorptive regime decreased from 0.12 to 0.10, partly owing to the enhanced collision-coalescence process accompanied by higher LWP, and thus inhibits the impact of LTS on ACI_r. Overall, ACI_rs are enhanced under high LTS conditions, but the difference between two regimes indicates that both ω_{abs} and LTS can be the impact factor of the ACI_r but they are not necessarily having causality between them.'

From specific comment:

I suggest to put the location SGP at the end of the second sentence: "… are selected over the Southern Great Plains region of the United States (SGP). The physicochemical…" Otherwise, the logic of the abstract is difficult to follow.
Thanks for the suggestion.
The sentence has been changed to '*a total of 16 low-level stratus cloud cases under daytime coupled boundary layer conditions are selected over the Southern Great Plains region of the United States (SGP)*' in the revised manuscript.

The answers of the following comment should appear in the text:
- the different resolution and uncertainty between KAZR and MMCR

The following statement '*Although the uncertainty of KAZR (~30m) is lower than MMCR (~45m), the difference of 15m between these two cloud radars would not cause a significant difference in detecting the cloud boundaries*' has been added to section 2.1.1 in the revised manuscript.

- The comparison with aircraft measurements from Delle Monache et al. (2004) with a quantification of their results to assess the reliability of the measurements.

The following statement '*A study was conducted by Delle Monache et al., (2004) used in-situ aerosol measurements from 59 flights from March 2000 – March 2001 to compare with the surface aerosol measurements. Their results showed that the aerosol extensive properties such as the total extinction by particles measured within the well-mixed boundary layer were well-correlated with surface measurements with the* R^2 *value of 0.88.* ' has been added to section 2.3 in the revised manuscript.

- The threshold you are using are from Dong et al. (2015), originally suggested by Jones et al. (2011)

The following statement '*These thresholds are the same as in Dong et al. (2015), originally suggested by Jones et al. (2011)*' has been added to section 2.3 in the revised manuscript.

- The uncertainty of ACI corresponds to the 95 % confidence interval.

The following statement '*The* ACI_r *values range from 0.09 – 0.24 with a mean value of 0.145 ± 0.05, the uncertainty of* ACI_r *corresponds to the 95 % confidence interval.*' has been added to section 3.2 in the revised manuscript.

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Response to Reviewer #2

We appreciate your time for thoroughly reviewing our manuscript. We would like to thank you for the constructive comments and suggestions that help to improve the manuscript. The manuscript has been revised accordingly. The reviewer's comments are provided in black text and our responses are provided in blue text.

Response:

The authors have revised the manuscript in meaningful ways in response to the first reviews but there are still some passages that give a mixed impression of their understanding of the concerns raised in the reviews. The comments here fall somewhere between suggesting minor and major revisions.

One of my reservations about the first version of this manuscript was the way in which processes for cloud drop (Nd) activation were described relative to measurements of total aerosol number concentrations (Na) and cloud condensation nuclei (CCN.) The analysis of the observations showing the difference in Na conversion to CCN in weakly and strongly absorptive regimes is interesting and fairly straightforward (with the exception noted in the paragraph below.) This analysis is done for an observed %S of 0.2. While there is some discussion of how different %S might affect the results it should be clearly stated in the conclusions that they rest on this chosen value of %S.

Thanks for the comments.

The following discussion about how different %S might affect the results has been added to the first paragraph of Section 3.3.3 in the revised manuscript:

'Note that those ratios are computed for an observed supersaturation level of 0.2%. The fraction of aerosols that can activate as CCN increases with an increase in supersaturation level, under the same aerosol size and composition condition (Dusek et al., 2006). A sensitivity test of how the aerosol activation rate varies with different supersaturation levels is done by first interpolating the N_{CCN} from 0.2 % to 1.15 % and then calculating the N_{CCN}/N_a. As a result, the ratios of N_{CCN}/N_a for the weakly absorptive regime range from 0.54 to 0.38, while the ratios for the strongly absorptive regime range from 0.45 to 0.25. Considering that the supersaturation level in the continental boundary layer stratus is nearly impossible to reach the level of 1.15%, the supersaturation level of 0.2% used in this study, which represents the most typical condition for continental low stratus, yields reasonable results.'

With respect to the effect of aerosol composition/absorption on the efficacy of particles to serve as CCN, on P13 L23-25 you interpret the data from Fig 6. You note that there are ranges of Na for which the more absorbing aerosol are below the weakly absorbing aerosol. Are the ranges of number concentration meaningful in some way? What I see are two slopes that are very much alike except for defined excursions that you call out in these ranges. What's happening for those particular observations? What makes them different? It raises the question of why some highly absorbing aerosol fall within the same relationship of the weakly absorbing aerosol

and some do not. Is there another or additional property of this latter set of aerosol has that make them poor CCN?

Thanks for the comments.

The different relationships between the two absorptive regimes shown in Fig 6 are the result of the mixed effect of ω_{abs}, water availability and aerosol size on the aerosol activation.

Looking closely to Fig 6, we defined three clusters of different N_a range: 200-500 cm^{-3}, 500 – 1100 cm^{-3} and 1100-3500 cm^{-3}. The mean values of LWP, AE and ratio of N_{CCN} to N_a in each range are shown in the Table below:

N_a Range (cm^{-3})	LWP (gm^{-2})		AE		N_{CCN}/N_a	
	Weakly	Strongly	Weakly	Strongly	Weakly	Strongly
(a)200-500	158	162	1.59	1.73	0.77	0.35
(b)500-1100	138	167	1.53	1.40	0.58	0.51
(c)1100-3500	95	127	1.67	1.57	0.42	0.32

In 200-500 cm^{-3} N_a range, the samples from the two absorptive regimes are the most separated. The mean values of LWP indicate relatively sufficient water availability with less aerosol concentration, accompanied by the fact that the weakly absorbing aerosol sizes are relatively larger indicated by smaller AE. It is known that larger aerosol particles easily activate under same composition (Dusek et al., 2006), considering the weakly absorbing aerosol is more hydrophilic, the largest conversion rate difference among these three ranges are to be expected.

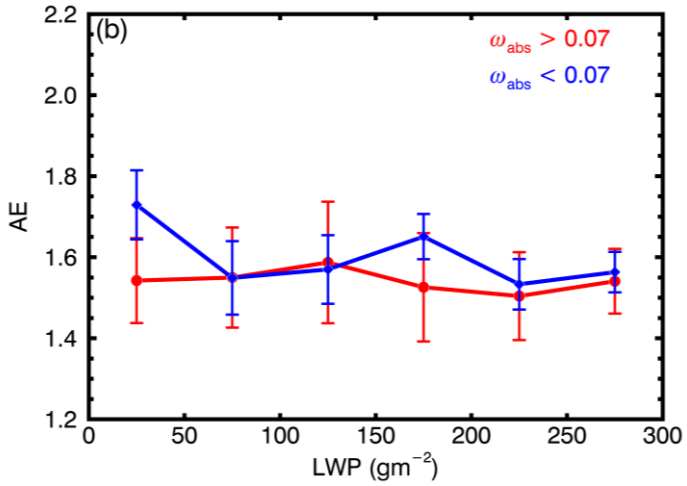
In 500-1100 cm^{-3} N_a range, the AE (LWP) value for strongly absorbing aerosols is noticeably smaller (higher) than those in the weakly absorptive regime. Therefore, the combined effect of more large particles and more water in the strongly absorptive regime lead to a N_{CCN}/N_a ratio close to the N_{CCN}/N_a ratio in the weakly absorptive regime.

In 1100-3500 cm^{-3} N_a range, there are more smaller aerosol particles and less water availability which leads to a greater aerosol competing effect with respect to water among both absorptive regimes, resulting in the lowest activation rate among the three ranges in both regimes.

Accordingly, the discussion above has been added to the third paragraph of Section 3.3.3 in the revised manuscript.

Therefore, to better examine the difference in aerosol activation capacity, Fig. 7 shows the N_{CCN}/N_a sorted by LWP for the two absorptive regimes. Moreover, the AE values are also sorted by LWP, thereby ruling out the influence of LWP and AE on aerosol activation. As shown in the Figure below, in every LWP bin, the AE for the weakly absorptive regime is either higher than or very close to the AE for the strongly absorptive regime. Even with relatively smaller particle sizes, under similar water availability, the weakly absorbing aerosol

can better activate as CCN. In conclusion, the significant effect of aerosol composition, which is inferred by aerosol absorbing ability, on the aerosol activation capacity does exist.



Accordingly, the following discussion has been added to the fourth paragraph of Section 3.3.3 in the revised manuscript.

'*As shown in Fig. 6, for three N_a ranges (200 - 500; 500 - 1100 and 1100 - 3500 cm^{-3}), the strongly absorbing aerosols show different relationships compared to weakly absorbing aerosols. The mean N_{CCN}/N_a values for those three N_a ranges for weakly absorptive regimes are 0.77, 0.58, and 0.42, respectively, while the mean N_{CCN}/N_a values for the strongly absorptive regimes are 0.35, 0.51, and 0.32, respectively. This phenomenon is due to the mixed effect of aerosol composition (inferred by absorbing ability), aerosol size, and water availability on the aerosol activation. In the 200 - 500 cm^{-3} N_a range, where the samples from the two absorptive regimes are most separated. The mean values of LWP (158 gm^{-2}/162 gm^{-2} for weakly/strongly absorptive regimes) indicate relatively sufficient water availability with less aerosol concentration. In addition, the weakly absorbing aerosol sizes are larger (AE = 1.59) than the strongly absorbing aerosol (AE = 1.73). It is known that larger aerosol particles easily activate under same composition (Dusek et al., 2006), considering the weakly absorbing aerosol is more hydrophilic, the largest conversion rate difference among these three ranges are to be expected. The samples in the 500 - 1100 cm^{-3} N_a range, have AE value (1.40) for strongly absorbing aerosols are noticeably smaller than those in the weakly absorptive regime (1.53), and the LWP in strongly absorptive regime (167 gm^{-2}) is much higher than in weakly absorptive regime (138 gm^{-2}). Therefore, the combined effect of larger particles and more water in strongly absorptive regime lead to the N_{CCN}/N_a is close to the N_{CCN}/N_a ratio in weakly absorptive regime. The samples in the 1100 - 3500 cm^{-3} N_a range exhibit smaller (AE = 1.67/1.57 for weakly/strongly absorptive regimes) aerosol particle size and less water availability (LWP = 95 gm^{-2}/127 gm^{-2} for weakly/strongly absorptive regimes) which results in the lowest activation rate (N_{CCN}/N_a ratio = 0.42/0.32 for weakly/strongly absorptive regimes) among the three ranges for both regimes.*'

The statement '*Furthermore, in the following section, the values of N_{CCN}/N_a and AE are sorted by LWP for the two absorptive regimes, in order to rule out the influence of LWP and AE on aerosol activation to the utmost extent*' has been added to last paragraph of Section 3.3.3 in the revised manuscript.

The statement *'Moreover, in every LWP bin, the AE value for the weakly absorptive regime is either higher than or very close to the AE value for the strongly absorptive regime (Figure not shown). Even with relatively smaller particle sizes, under similar water availability, the weakly absorbing aerosol can better activate as CCN. In conclusion, the significant effect of aerosol composition, which inferred by aerosol absorbing ability, on the aerosol activation capacity does exist*' has been added to second paragraph of Section 3.3.4 in the revised manuscript.

In the previous version of the manuscript there was not a clear message that the conversion of Na to CCN was dependent on aerosol properties and the conversion of CCN to Nd primarily dependent on cloud state and dynamics. In the discussions in Section 3.3. and later this has been largely rectified. The authors provide an explanation that the CCN to Nd relationship is impacted by environmental heating of absorbing aerosol and the resulting effects on cloud microphysics (rather than differences in cloud microphysics being due to the activation process associated with strongly and weekly absorbing aerosol.) The details of this dynamic are not explored due to a lack of relevant observation.
Given that the latter is not an aerosol-cloud microphysical effect there are passages earlier in the paper that are still confusing. On P2 L5-8 the implication is still that there is some microphysical effect for CCN to Nd that is different for absorbing and non-absorbing aerosol.

We have revised this statement to emphasize that the difference in CCN to N_d conversions is the result of different thermodynamic states and the environmental heating effect of strongly absorbing aerosol.

Accordingly, the corresponding part of the abstract has been changed to '*In terms of the sensitivity of cloud droplet number concentration (Nd) to CCN, the conversion ratio of N_d/N_{CCN} for weakly (strongly) absorptive aerosols is 0.69 (0.54), owing to the combined effect of different cloud dynamic, thermodynamic states and cloud-layer heating effect of the strongly light-absorbing aerosol. The measured ACI_r values in the weakly absorptive regime are relatively higher, indicating that clouds have greater microphysical responses to aerosols in weakly absorptive regime than in strongly absorptive regime. Consequently, we expect larger shortwave radiative cooling effect from clouds in the weakly absorptive regime than those in the strongly absorptive regime.*'

Some of the problem may just be the use of terminology. For Example, on P3 L1 "the efficacy of the activation of CCN" would be clearer as "the efficacy of the activation of aerosol." For the CCN measurements as they are made here, at a given supersaturation, the number concentration is assumed to be 100% efficacious for cloud droplet activation. Size and composition, however, will affect the activation rate of aerosol.

Thanks for the suggestion.

The sentence has been changed to '*The efficacy of the activation of aerosol has been widely known to be influenced by aerosol size distribution and chemical composition which are the primary sources of uncertainty in assessing ACI*' in the revised manuscript.

On P10 L28-29 to P11 first paragraph there is a statement that Na does not serve as a realistic CCN proxy for calculating ACIr but attributes the poor result on decoupled boundary layer condition. Is this the author's assumption or the assumption made by Feingold et al. 2003? What about the fact that Na can include large numbers of very small particles that will not activate, especially at these supersaturations? The difference between Na and scattering as a CCN proxy and coupling of the BL are mixed up in this discussion. Scattering (or aerosol index) have long been used as a more reliable CCN proxy due to their sensitivity to larger particle sizes. It just contributes to the confusion with how the manuscript characterizes total aerosol and CCN with respect to droplet activation.

Thanks for the comments.

The statements in P10 L28-29 to P11 L5 are the assumptions and findings in Feingold et al. (2006). To avoid further confusion, the discussion about the role of boundary layer coupling is omitted in the revised paragraph, since the usefulness of using surface aerosol measurement to represent aerosol aloft under coupled boundary layer condition has been discussed in Section 2.3.

Therefore, the discussion in Section 3.2 of the revised manuscript has been changed as follows: '*Previous studies have focused on the aerosol-cloud interaction in stratocumulus clouds at the ARM SGP site. Based on the analysis of seven selected stratocumulus cases during the period 1998 - 2000, Feingold et al. (2003) reported the first ground-based measured* ACI_r *values of 0.02 to 0.16 using the lidar measured aerosol extinction at a wavelength of 355 nm as the proxy for aerosol loading. A later study conducted by Feingold et al. (2006) assessed the* ACI_r *using different aerosol measurements as CCN proxies, in three selected stratus cases during the intensive operation period in May 2003. They found that the* ACI_r *values were unrealistic when using* N_a *to represent CCN loading while using the surface aerosol scattering coefficient (*σ_{sp}*) and aerosol extinction at an altitude of 350 m as CCN proxies yield similar* ACI_r *values ranging from 0.14 to 0.39 (Feingold et al. 2006). The assessment of* ACI_r *can be largely affected by the usage of different aerosol measurements that served as CCN proxies due to their own characteristics. Aerosol scattering and extinction coefficient are known to be relatively reliable CCN proxies since they are more sensitive to aerosols that have larger particle sizes. As for* N_a*, which represents the concentration of aerosol particles with diameters larger than 10 nm, it is likely to pick up the very small aerosols generated by new particle formation events. This proportion of aerosols is presumably hard to activate as CCN so that would not be counted in* N_{CCN}*, especially under the 0.2% supersaturation used in this study. Hence, it is less representative to use* N_a *to accurately represent* N_{CCN} *without the prior knowledge of the aerosol capacity to activate as CCN. Therefore, the usage of* N_{CCN} *in this study is favorable to*

yield a more straightforward assessment of ACI_r, since the CCN measurement directly represents the amount of aerosol droplets that already activated and have the potential of further growth.'

This continues in the next paragraph, P11 L10. Is there an assumption that a constant fraction of aerosol effective activates as you state? At what %S? You've shown yourself this relationship can depend on composition and it certainly will depend on size and that is known to be variable at SGP. New particle formation events can push aerosol concentrations up with a large number concentration at very small sizes that you would not see in the CCN or scattering measurements. I don't think the paragraph is the right way to lead into your results.

Thanks for the comments.

The discussion in this paragraph was meant to suggest that the statement 'a nearly constant fraction of aerosol effectively activates as CCN' and 'aerosol loading is more important than the aerosol size and composition' are not true in this study, and therefore the role of aerosol species is necessary to be examined. We agree that the way of presentation here conveys a confusing message. In order to avoid further confusion, this part of the discussion has been changed as follows:
'*In order to better understand the aerosol particle activation process in typical continental low-level stratus clouds, in the latter part of this study, the ratios between N_{CCN} and N_a are examined and used to represent the aerosol activating capacities. Since the aerosol activating capacities greatly depend on the aerosol sizes and compositions, in order to further examine the role of aerosol species in aerosol activation process and the potential impact on ACI_r, the samples from the 16 selected cases are divided into two groups according to their absorptive regime which is discussed in the following section.*'

Ultimately I would suggest leaving the Na measurements out of the discussion when covering CCN to Nd or ACI. Na should be the focus when discussing how absorption impacts how well aerosol serve as CCN. Because you have CCN measurements and can use these directly to reference ACI in terms of drop numbers or effective radius just leave it at that. You can still go back and show that ACI differs in high and low absorption regimes by using the co-SSA quantity and leave total number concentration out of it. I don't see the need for referring to a CCN proxy when you have CCN. It's really confusing the discussion and leaves the reader wondering how the authors are considering the cloud drop activation process. Related, P20 L29-P21 L1 - not sure this statement is worth making.

Thanks for the suggestions.

The relevant discussions have been modified accordingly in the revised manuscript.

The revised Fig. 8 below now showing only the ACI_r with respect to CCN.

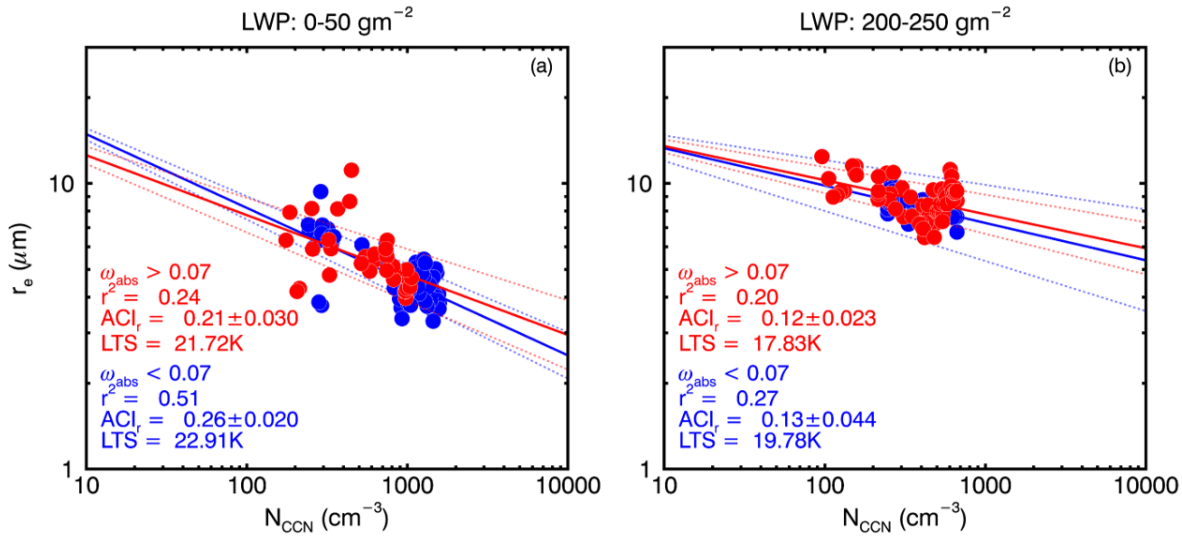


Figure 8. r_e as a function of N_{CCN} and the values of ACI_r under the strongly absorptive (in red) and weakly absorptive (in blue) aerosol regimes at two LWP bins: 0-50 g m^{-2} (a) and 200-250 g m^{-2} (b). Note that the dashed lines denote the uncertainties of ACI_r due to 10 % error in r_e retrieval.

The statement at P20 L29-P21 L1 has been deleted, and the last two paragraphs of Section 4 has been changed as follow:

'*The conversion rates of N_d/N_{CCN} for the weakly absorbing aerosols (mean ratio of 0.67) are higher than for the strongly absorbing aerosols (mean ratio of 0.54). Partly owing to the higher LTS environment for the weakly absorptive regime which enhance the connection between cloud and the below-cloud moisture and CCN. Also the cloud layer heating effect induced by the strongly light-absorbing aerosols results in the reduction of in-cloud supersaturation and leads to the damping of CCN activation process for the strongly absorptive regime. As a result, cloud droplets that form from weakly absorbing aerosols tend to have smaller sizes and higher concentrations than cloud droplets forming from strongly absorbing aerosols. Furthermore, the cloud droplets in the weakly absorptive regime exhibit a greater growing ability, as given by larger r_e values that increase with LWP under similar N_d. The differences in cloud droplet development between the two regimes is a likely result of the combination of thermodynamics, dynamics, and aerosol radiative effects.*

Under low LWP conditions, the measured ACI_r values in the weakly absorptive regime are relatively higher, indicating that clouds have greater microphysical responses to aerosols in weakly absorptive regime than in strongly absorptive regime, owing to favorable LTS condition in the weakly absorptive regime, and the cloud-layer heating effect of light-absorbing aerosol in the strongly absorptive regime. The observed ACI_r is enhanced after constrained by high LTS. Under higher LWP conditions, the enhanced collision-coalescence process diminish the LTS impact on ACI_r, and the damping of ACI_r is more evident, which is consistent with the results from all the cases. In general, the 10% uncertainty in r_e retrieval contribute to ACI_r uncertainties range from 0.02 to 0.04 for the two absorptive regime, with the ACI_r difference between the two absorptive regimes still well-preserved. As a result, clouds that develop from

weakly absorbing aerosols exhibit a stronger shortwave cloud radiative effect than clouds originating from strongly absorbing aerosols. Additional future work will focus on investigating detail composition of different aerosol plumes, with respect to their physicochemical properties. The aerosol-cloud-interaction processes under the influence of different aerosol types associated with airmasses and the sensitivity to dynamic and thermodynamic factors will be further examined.'

In addition, some discussion of the heating effect of absorbing aerosol on the environment leading to differences in stability and cloud state/dynamics (updraft speeds and moisture availability) early in the manuscript might help to clarify the different effects of aerosol absorption on the cloud properties that you are postulating.

Thanks for the suggestion.

The further discussion of the heating effect of absorbing aerosol on the environment leading to differences in stability and cloud state/dynamics has been added to the last paragraph of Section 3.3.4 in the revised manuscript, which is as follows:

'*Furthermore, the radiative effect of light-absorbing aerosols on the cloud environment also cannot be neglected, since the strongly light-absorbing aerosols can absorb solar radiation and heat the in-cloud atmosphere by emission, which results in the reduction of relative humidity or supersaturation in the cloud layer (Bond et al., 2013; Wang et al., 2013). Additionally, this aerosol heating effect disrupts the boundary layer temperature structure by enhanced warming aloft, and consequently, inhibits the vertical transport of sensible and latent heat between surface and cloud layer. The impacts of light-absorbing aerosol on cloud-scale thermodynamics and dynamics state might eventually dampen the conversion process from CCN to cloud droplet.*'

I think this manuscript still need work before being considered for publication.

Thanks again for the thorough review of this manuscript.

Specific:
P1 L 26: I think this is supposed to read "…suggests a diminished cloud microphysical response to aerosol loading"

The sentence has been changed to 'The magnitude of ACI_r decreases with increasing LWP which suggests a diminished cloud microphysical response to aerosol loading presumably due to enhanced collision-coalescence processes and enlarged particle size.'.

P6 L8: Aerosol Observing System

The correction has been made.

P7 L5 “question of whether surface aerosols can be linked to what actually happens in clouds aloft.”

The correction has been made.

P15 L15 – switch position of Nccn and Na

The correction has been made.

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Mark-up Manuscript Version:

Investigation of Aerosol-Cloud Interactions under Different Absorptive Aerosol Regimes using ARM SGP Ground-Based Measurements

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Abstract

Aerosol indirect effect on cloud microphysical and radiative properties is one of the largest uncertainties in climate simulations. In order to investigate the aerosol-cloud interactions, a total of 16 low-level stratus cloud cases under daytime coupled boundary layer conditions are selected over the Southern Great Plains region of the United States (SGP). The physicochemical properties of aerosols and their impacts on cloud microphysical properties are examined using data collected from the Department of Energy Atmospheric Radiation Measurement (ARM) facility – at SGP site ~~over the Southern Great Plains region of the United States~~ (ARM-SGP). The aerosol-cloud interaction index (ACI_r) is used to quantify the aerosol impacts with respect to cloud-droplet effective radius. The mean value of ACI_r calculated from all selected samples is 0.145 ± 0.05 and ranges from 0.09 to 0.24 at a range of cloud liquid water paths (LWP=20-300 g m^{-2}). The magnitude of ACI_r decreases with increasing LWP which suggests a diminished cloud microphysical response to ~~diminished~~ aerosol loading presumably due to enhanced collision-coalescence processes and enlarged particle size. The impact of the aerosols with different light-absorbing abilities on the sensitivity of cloud

microphysical responses is also investigated. In the presence of weak light-absorbing aerosols, the low-level clouds feature a higher number concentration of cloud condensation nuclei (N_{CCN}) and smaller effective radii (r_e) while the opposite is true for strong light-absorbing aerosols. Furthermore, the mean activation ratio of aerosols to CCN (N_{CCN}/N_a) for weakly (strongly) absorbing aerosols is 0.54 (0.45), owing to the different hygroscopic abilities associated with the dominant aerosol species. In terms of the sensitivity of cloud droplet number concentration (N_d) to ~~aerosol loading~~CCN, the conversion ratio of N_d/N_{CCN} for weakly (strongly) absorptive aerosols is 0.69 (0.54), owing to the combined effect of different cloud dynamic, thermodynamic states and cloud-layer heating effect of the strongly light-absorbing aerosol. The measured ACI_r values in the weakly absorptive regime are relatively higher, indicating that clouds have greater microphysical responses to aerosols in weakly absorptive regime than in strongly absorptive regime. Consequently, we expect larger shortwave radiative cooling effect from clouds ~~influenced by~~in the weakly absorptive regime~~bing aerosols~~ than those in the strongly absorptive regime.~~strongly absorbing aerosols.~~

1. Introduction

Clouds play a critical role in the Earth's climate by acting as the dominant modulator of radiative transfer in the atmosphere and have substantial impacts on the global climate. The radiative effect of clouds contributes to one of the largest uncertainties in climate modeling (IPCC, 2013), and has been well known to be influenced by aerosol loading. An increase in aerosol concentration can lead to the enhancement of cloud droplet number concentration (N_d) and the reduction of cloud droplet effective radii (r_e), which results in an increase of cloud albedo. This phenomenon is defined as the aerosol first indirect effect (Twomey, 1977), and it is denoted as a general cooling effect in terms of global radiation balance. More fundamentally, the aerosol effects on cloud reflectance result from the cloud microphysical response to aerosol concentration (e.g., aerosol-cloud interaction, ACI).

The magnitude and sensitivity of ACIs in low-level clouds have been investigated by numerous studies, using various observational datasets such as ground-based measurements (Garrett et al., 2004; Feingold et al., 2006; Kim et al., 2008; McComiskey et al., 2009; Wang

et al., 2013, 2018a), satellite retrieved products (Sekiguchi et al., 2003; Su et al., 2010) and airborne in situ measurements (Twohy et al., 2013; Painemal and Zuidema, 2013; Zhao et al., 2018). However, large variations exist among various assessments, because of intrinsic instrument uncertainty, differing analysis methods, and more physically, the inherent variation in aerosol properties. The physical mechanism underlying the aerosol effect on clouds is that aerosols activate as cloud condensation nuclei (CCN) and then influence the cloud microphysical features. The efficacy of the activation of aerosol~~The efficacy of the activation of CCN~~ has been widely known to be influenced by aerosol size distribution and chemical composition which are the primary sources of uncertainty in assessing ACI (Dusek et al., 2006; McFiggans et al., 2006; Liu and Li, 2014; Che et al., 2016).

Previous studies have suggested that the composition of aerosols can be inferred by their optical properties such as aerosol optical depth, single scattering albedo, and Ångström exponent (Clarke et al., 2004; Bergstrom et al., 2007; Clark et al., 2007; Russell et al., 2010; Cappa et al., 2016). For instance, fine mode carbonaceous particles (e.g., black and organic carbon) have strong light-absorbing abilities in the ultraviolet and visible spectra (Logan et al., 2013). On the other hand, urban pollution aerosols associated with sulfate and nitrate particles are considered as weakly absorbing aerosols (Eck et al., 1999, 2005; Bergstrom et al., 2007; Chin et al., 2009). Although studies have been done to classify aerosol types using the absorption Ångström exponent, which is associated with the absorptive spectral dependence of particles, the measurements of this parameter typically carry large uncertainty, and can provide limited information when there are mixtures of different aerosol species that share similar spectral dependences (Bergstrom et al., 2007; Lack and Cappa, 2010). Alternatively, the single scattering albedo (SSA) and co-albedo (1-SSA) can be used to better separate the aerosol types because they focus on the relative absorbing ability of aerosols at specific wavelengths (Logan et al., 2013; Tian et al., 2017). Given the wide availability of aerosol optical property measurements, the feasibility of inferring aerosol species from their optical properties is useful particularly in areas with no direct measurements of aerosol chemical composition (Logan et al., 2013; Schmeisser et al., 2017).

The Atmospheric Radiation Measurement (ARM) program initiated by the U. S. Department of Energy (DOE) aims to improve the parameterization of clouds in global climate models (Stokes and Schwartz, 1994). Thus far, the ARM program has established over 20 years of long-term ground-based measurements of cloud properties and surface measured aerosol properties at the Southern Great Plain (SGP) site which represents typical continental conditions (Ackerman and Stokes, 2003; Dong et al., 2006). The size and composition of aerosols have been found to have a considerable seasonal and regional dependence, and their impacts on clouds also vary with different aerosol regimes (Sorooshian et al., 2010; Logan et al., 2018). The prevailing fine mode aerosols at ARM-SGP site typically contain organic and black carbon associated with biomass burning and inorganic aerosols composed of sulfate and nitrate species (Parworth et al., 2015; Logan et al., 2018). The differences in intrinsic hygroscopicity among those aerosol species play various roles in aerosol activation processes and consequently lead to various interactions with clouds. Thus, it is necessary to investigate the aerosol and cloud properties as well as the magnitude of the ACI index at the ARM-SGP site, in order to (a) enhance the understanding of ACI and (b) reduce the uncertainty in quantifying the ACI and associated radiative effects when modeling aerosol influences on low level continental clouds.

In this study, the aerosol and cloud properties at the ARM-SGP site from 16 selected non-precipitating low-level stratiform cloud cases during the 2007-2012 period are examined. Details of the observational measurement platforms and methods are introduced in section 2. The development and analysis of the ACI for the 16 selected cases, the aerosol activation and cloud microphysical responses, as well as consequent cloud radiative effects under different aerosol absorptive regimes, are investigated in section 3. Lastly, a summary of our findings and future work is presented in section 4.

2 Data and methods

2.1 Cloud Properties

2.1.1 Cloud Boundaries

The cloud boundaries at the ARM-SGP site were primarily determined by the ARM Active Remotely-Sensed Cloud Locations (ARSCL) product, which is a combination of data detected by multiple active remote-sensing instruments, in particular, the Millimeter-wavelength Cloud Radar (MMCR). The MMCR operates at a frequency of 35 GHz (and wavelength of 8.7 mm) with a zenith pointing beam width of 0.2° and provides a continuous time-height profile of radar reflectivity with temporal and spatial resolutions of 10 seconds and 45 m, respectively (Clothiaux et al., 2000). After 2011, the MMCR was replaced by the Ka-band ARM Zenith Radar (KAZR) which has the same operating frequency and shares similar capabilities as the MMCR, but with the major improvement of a new receiver that allows for more sensitivity in cloud detection (Widener et al., 2012). The temporal and vertical resolutions of KAZR-detected reflectivity are 4 seconds and 30 m, respectively. The cloudy condition as well as cloud top height is identified via cloud radar reflectivity. Although the uncertainty of KAZR (~30m) is lower than MMCR (~45m), the difference of 15m between these two cloud radars would not cause a significant difference in detecting the cloud boundaries.

The cloud radar is sensitive to the sixth moment of droplet size distribution and can be contaminated by insects below the cloud base (Dong et al., 2006). The ceilometer and Micropulse Lidar (MPL) measurements, which are sensitive to the second moment, are used to provide an accurate cloud base estimation. Hence, the lidar-radar pair provides the most precise determination of cloud boundaries from a point-based perspective. In this study, the cloud base and top heights were averaged into 5-min bins where the low-level stratus cloud is defined as a cloud-top height lower than 3 km with no overlying cloud layer (Xi et al., 2010).

2.1.2 Cloud Microphysical Properties

The cloud liquid water path (LWP), defined as the column-integrated cloud liquid water, was retrieved based on the measured brightness temperatures from the Microwave Radiometer (MWR) at 23.8 and 31.4 GHz, using the statistical method described in Liljegren et al. (2001). The uncertainty of LWP retrieval is 20 $\mathrm{g\,m^{-2}}$ for LWP less than 200 $\mathrm{g\,m^{-2}}$ and around 10% for LWP higher than 200 $\mathrm{g\,m^{-2}}$. In this study, we exclude the data points with LWPs less than 20 $\mathrm{g\,m^{-2}}$ to eliminate optically thin clouds, as well as exclude the samples with LWPs greater than 300 $\mathrm{g\,m^{-2}}$ to prevent potential precipitation contamination issues (Dong et al., 2008).

For microphysical properties of low-level stratus, following the methods developed by Dong et al. (1998), the daytime layer-mean cloud-droplet effective radius (r_e) can be calculated by:

$$r_e = -2.07 + 2.49\mathrm{LWP} + 10.25\gamma - 0.25\mu_0 + 20.28\mathrm{LWP}\gamma - 3.14\mathrm{LWP}\mu_0, \quad (1)$$

where γ is the solar transmission, μ_0 is the cosine of solar zenith angle, and the units of r_e and LWP are μm and 100 $g\,m^{-2}$, respectively. N_d is obtained after r_e is known, by the following calculation:

$$N_d = \left(\frac{3\mathrm{LWP}}{4\pi\ {}_w r_e^3 \Delta Z}\right) \exp(3\sigma_x^2), \quad (2)$$

where N_d has units of cm^{-3}, ΔZ is cloud thickness determined from cloud boundaries with units of m, and σ_x is the width of the lognormal size distribution of cloud droplet, which is assumed to be a constant value of 0.38 (Miles et al., 200~~02~~). The sensitivities of retrieved r_e and N_d to the uncertainties of cloud LWP, σ_x and γ have been investigated in Dong et al. (1997 and 1998). The uncertainties of retrieved r_e and N_d have been estimated against aircraft in situ measurements over the ARM-SGP site (Dong et al., 2002, 2003) and other regions (Dong et al. (1998). ~~In general, the uncertainties of retrieved daytime r_e and N_d are ~10% and ~20-30%, respectively.~~ As the results, the 10% change in cloud LWP and downward SW at the surface would result in the 10% uncertainty in r_e retrieval. And the N_d uncertainty is statistically estimated to be 25%, compared with the aircraft in situ measurements at the Penn State surface site during the Fall 1996 (Dong et al. 1998) and at the ARM SGP site during March 2000 Cloud Intensive Observational Period (IOP) (Dong et al. 2002; Dong and Mace, 2003).

2.2 Aerosol Properties

Surface aerosol properties were collected from the Aerosol ~~Observation~~ Observing System (AOS), a platform consisting of an array of instruments to monitor real-time aerosol information. The total condensation nuclei number concentration (N_a) represents the overall loading of aerosol particles with diameters larger than 10 nm and was obtained by the TSI model 3010 condensation particle counter. The aerosol scattering coefficient (σ_{sp}) was measured by the TSI model 3653 nephelometer at three wavelengths: 450, 500, and 700 nm. The relative humidity inside the nephelometer was set to 40% to maintain a dry condition and

prevent potential aerosol hygroscopic effects (Jefferson, 2011), and the quality of retrievals has been assured using the Anderson and Ogren (1998) method. The absorption coefficient (σ_{ap}) was measured by the Radiance Research particle soot absorption photometer (PSAP) at three slightly different wavelengths (470, 528 and 660 nm), with the calibration and quality control process done by the method developed in Anderson et al. (1999). Note that both the nephelometer and PSAP employ two impactors with size cuts of 1 μm and 10 μm. The measurements switch between total aerosol (<10 μm) and submicron aerosol (<1 μm) every hour. In this study, the sub-10 μm aerosol optical properties with original 1-min temporal resolution were averaged into 5-min bins to match the cloud microphysical properties.

The optical particle counter developed by Droplet Measurement Technologies is used to measure the CCN number concentration (N_{CCN}). The supersaturation (SS) level inside the instrument cycles between 0.15% and 1.15% every hour. The CCN activity can be presented as a function of SS: $N_{CCN} = cSS^k$ (Twomey, 1959), where c and k are calculated by using a power law fit for each hour. In this study, 0.2% is used as this represents typical supersaturation conditions of low-level stratus clouds (Hudson and Noble, 2013; Logan et al., 2014; Logan et al., 2018).

2.3 Boundary Layer Condition and Lower Tropospheric Stability

Given the fact that the aerosol properties were measured at the surface, there is a question of whether surface aerosols can be linked to what actually happens in clouds aloft.~~of how to link surface aerosols to what actually happens in clouds aloft.~~ This study adopts the method presented in Dong et al. (2015), which defined the boundary layer condition into two categories: coupled and decoupled. The vertical sounding profiles at a 1-min temporal resolution were collected from the ARM Merged Sounding product with a vertical resolution of 20 m below 3 km (Mace et al., 2006; Troyan, 2012). The vertical profiles of liquid water potential temperature (θ_L) and total water mixing ratio (q_t) for coupled and decoupled boundary layer conditions, as well as the criteria to differentiate between them, are illustrated in Fig. 1. The coupled condition was identified by the change of θ_L and q_t from surface layer to cloud base of less than 0.5 K and 0.5 g/kg, respectively. These thresholds are the same as in Dong et al. (2015), originally suggested by Jones et al. (2011). In that case, the boundary layer is considered to be well-mixed

and suggests that the surface aerosols are comparable to in-cloud aerosols. ~~However~~In the decoupled condition, the θ_L and q_t vary more drastically from surface to cloud base under decoupled conditions, which denotes a stratification of the sub-cloud layer thereby disconnecting the surface aerosols from the ones aloft.

A study was conducted by Delle Monache et al., (2004) used in-situ aerosol measurements from 59 flights from March 2000 – March 2001 to compare with the surface aerosol measurements. Their results showed that the aerosol extensive properties such as the total extinction by particles measured within the well-mixed boundary layer were well-correlated with surface measurements with the R^2 value of 0.88. Therefore, selecting cloud cases under coupled conditions can better constrain the thermodynamic condition since the measured surface aerosols are representative in terms of aerosol-cloud interaction.

The Lower Tropospheric Stability (LTS), which is defined as the potential temperature difference between surface and 700hPa, is used to represent the large-scale thermodynamic condition. The LTS is obtained from the ECMWF model output which specifically provides for analysis at the ARM SGP site. The value is obtained by averaging over a grid box of 0.56*0.56° which is centered at SGP. The original temporal resolution of LTS is 1-hour and is then interpolated to 5-min to match the other variables, assuming the large-scale forcing would not have significant changes during every 1-hour window.

2.4 Shortwave radiation fluxes at the Surface

The surface measured broadband downwelling shortwave (SW) radiation fluxes and estimated clear-sky SW fluxes were collected from Radiative Flux Analysis Value Added Products (Long and Ackerman, 2000; Long and Turner, 2008), with an uncertainty of 10 W m^{-2}. The combination of cloudy and clear-sky SW fluxes was used to calculate the cloud radiative effect. In order to minimize the influence of non-cloud factors, such as solar zenith angle and surface albedo, a representation of the relative cloud radiative effect (rCRE) is defined as

$$\mathrm{rCRE} = 1 - \mathrm{SW}_{\mathrm{cld}}^{dn}/\mathrm{SW}_{\mathrm{clr}}^{\mathrm{dn}}, \quad (3)$$

where SW_{cld}^{dn} and SW_{clr}^{dn} are cloudy and clear-sky downwelling shortwave radiation fluxes, respectively (Betts and Viterbo, 2005; Vavrus, 2006; Liu et al., 2011).

2.5 Selection of low-level stratus cloud cases

As previously discussed, the selection of cloud cases is limited by the following criteria: non-precipitating and cloud-top height less than 3 km with lifetime more than 3 hours under the limitation of 20 gm^{-2} < LWP < 300 gm^{-2} and the coupled boundary layer conditions. Only daytime cloudy periods were considered in this study because the r_e retrieval required the information of solar transmission (Dong et al., 1998). Note that all the variables used in the study are averaged in 5-min temporal resolution bins. A total of 16 cases were selected during the 6-year period from 2007 to 2012, which represents a total of 693 samples (~ 58 hours) in this study, the detailed time period and the number of sample points of each case are listed in Table 1. Most cases occurred during the winter and spring months since low-level cloud occurrences are higher during those seasons (Dong et al., 2006). The 72-hour NOAA HYSPLIT backward trajectories (Stein et al., 2015) for sub-cloud air parcels that advected over the ARM-SGP site are used to identify the aerosol source regions (Logan et al., 2018). Aerosol plumes consisting of different species from local sources and long-range transport can impact the ARM SGP site because of different transport pathways and can induce different cloud responses which are further investigated in this study.

3 Result and Discussion

3.1 Aerosol and cloud properties of selected cases

The probability density functions (PDFs) of aerosol and cloud properties from all 16 cases are shown in Fig. 2, note that the distributions include each of the 5-min data points. For the aerosol properties shown in the top panel, the Ångström Exponent (AE) was calculated based on the nephelometer observed spectral scattering coefficient (σ_{sp}) at 450 nm and 700 nm, using the equation of $AE_{450-700nm} = -\log(\sigma_{sp450}/\sigma_{sp700})/\log(450/700)$. The negative log-log slope denotes the relative wavelength dependence of particle optical properties due to differences in particle sizes (Schuster et al., 2006). Therefore, AE can be a good indicator of

aerosol particle sizes since AE > 1 indicates the particle size distributions dominated by fine mode aerosols (submicron), while AE < 1 denotes the dominance of coarse mode aerosols (Gobbi et al., 2007; Logan et al., 2010). The aerosol Fine Mode Fraction (FMF) is given by the ratio, $\sigma_{sp1}/\sigma_{sp10}$, where σ_{sp1} and σ_{sp10} are the nephelometer measured scattering coefficients at 550 nm for fine mode aerosols (1 μm size cut) and total aerosols (10 μm size cut), respectively. This ratio indicates the dominant influence of fine mode aerosols owing to the physical properties of the entire aerosol plume. For example, FMF values greater than 0.6 represent the dominance of fine mode aerosol in the total population and values less than 0.2 represent the dominance of coarse mode aerosols in the total population (Anderson et al., 2003). As illustrated in Figs. 2b and 2c, fine mode aerosols are dominant in the 16 selected cases. All AE values are higher than 1, with most of the values ranging from 1.5 to 2. In addition, the majority of the FMF values are greater than 0.6 and range from 0.7 to 0.9.

The variation in aerosol single scattering albedo (SSA) suggests different roles of the fine mode aerosol absorptive properties that influence total light extinction which in turn is a result of different aerosol species in the plume. This is further explained in section 3.3. The distributions of N_a, N_{CCN}, and N_d represent typical continental aerosol conditions with mean values of 1060 cm^{-3}, 475 cm^{-3}, and 297 cm^{-3}, respectively, and r_e values are more normally distributed with the majority of values between 7-9 μm. Note that the variation in the PDF of LWP is relatively small, which allows for a better investigation of the LWP dependence of cloud microphysical properties.

3.2 Measured Aerosol-Cloud-Interaction

To examine the microphysical response of cloud to aerosol loading, the quantitative Aerosol-Cloud-Interaction (ACI) term can be expressed as

$$ACI_r = -\frac{\partial \ln(r_e)}{\partial \ln(\alpha)}\Big|_{LWP}, \quad (4)$$

where α denotes aerosol loading. ACI_r represents the relative change of layer mean r_e with respect to the relative change of aerosol loading thereby emphasizing the sensitivity of the cloud microphysical response (Feingold et al., 2003; Garrett et al., 2004). Note that values of ACI_r have theoretical boundaries of 0-0.33, where the lower bound means no change of cloud

microphysical properties with aerosol loading and the upper bound indicates a linear relationship.

As suggested by previous studies, the ACI_r should be calculated and compared at constant LWP owing to the dependence of r_e on LWP (Twomey et al., 1977; Feingold et al., 2003). Therefore, in this study we use six LWP bins ranging from 0-300 $g\ m^{-2}$ with bin size of 50 $g\ m^{-2}$ and then group the sample data accordingly. Note that the first bin is actually 20-50 $g\ m^{-2}$ due to the elimination of LWP less than 20 $g\ m^{-2}$. The r_e-N_{CCN} relationship is presented in Fig. 3a where only the samples from three LWP bins are used to illustrate the r_e-N_{CCN} response. In general, r_e decreases with increasing CCN number concentration as expected. The ACI_r values range from –0.09 – 0.24 with a mean value of 0.145 ± 0.05, the uncertainty of ACI_r corresponds to the 95 % confidence interval. Note that t~~T~~he ACI_r values from six LWP bins show a generally decreasing trend of ACI_r with increasing LWP (Fig. 3b). Particularly, this decreasing trend is more obvious in a range of LWPs that are less than 150 $g\ m^{-2}$. The higher values of ACI_r at lower LWPs indicate that the clouds are more susceptible to aerosol loading under lower liquid water availability. When LWP increases, there is increased collision-coalescence activity within the cloud which results in the reduction of N_d as shown in Fig. 3b (blue diamonds). This partly leads to the damping of cloud microphysical sensitivity as evidenced by decreased ACI_r (Kim et al., 2008; McComiskey et al., 2009).

Previous studies have focused on the aerosol-cloud interaction in stratocumulus clouds at the ARM SGP site. Based on the analysis of seven selected stratocumulus cases during the period 1998 - 2000, Feingold et al. (2003) reported the first ground-based measured ACI_r values of 0.02 to 0.16 using the lidar measured aerosol extinction at a wavelength of 355 nm as the proxy for aerosol loading. A later study conducted by Feingold et al. (2006) assessed the ACI_r using different aerosol measurements as CCN proxies, in three selected stratus cases during the intensive operation period in May 2003. They found that the ACI_r values were unrealistic when using N_a to represent CCN loading while using the surface aerosol scattering coefficient (σ_{sp}) and aerosol extinction at an altitude of 350 m as CCN proxies yield similar ACI_r values ranging from 0.14 to 0.39 (Feingold et al. 2006). The assessment of ACI_r can be largely affected by the usage of different aerosol measurements that served as CCN proxies due to their own

characteristics. Aerosol scattering and extinction coefficient are known to be relatively reliable CCN proxies since they are more sensitive to aerosols that have larger particle sizes. As for N_a, which represents the concentration of aerosol particles with diameters larger than 10 nm, it is likely to pick up the very small aerosols generated by new particle formation events. This proportion of aerosols is presumably hard to activate as CCN so that would not be counted in N_{CCN}, especially under the 0.2% supersaturation used in this study. Hence, it is less representative to use N_a to accurately represent N_{CCN} without the prior knowledge of the aerosol capacity to activate as CCN. Therefore, the usage of N_{CCN} in this study is favorable to yield a more straightforward assessment of ACI_r, since the CCN measurement directly represents the amount of aerosol droplets that already activated and have the potential of further growth. ~~At the ARM SGP site, based on the analysis on seven selected stratocumulus cases during the period 1998 - 2000, Feingold et al. (2003) reported the first ground-based measured ACI_r values of 0.02 to 0.16 using the lidar measured aerosol extinction at a wavelength of 355 nm as the proxy for aerosol loading. The data were stratified in similar LWP bins to eliminate the LWP effect on r_e. The study conducted by Feingold et al. (2006) during an intensive operation period in May 2003 showed that the assessment of ACI_r can be affected by the usage of different aerosol proxies and boundary layer conditions. Using surface measured N_a to represent aerosol loading yielded unrealistic values of ACI_r even after sorted by LWP, presumably owing to decoupled boundary layer conditions. However, if the surface aerosol scattering coefficient (σ_{sp}) and aerosol extinction at an altitude of 350 m are used as CCN proxies, then similar ACI_r values can be obtained with a range of 0.14-0.39. Under coupled conditions, the N_a and σ_{sp} could serve as reliable CCN proxies. The σ_{sp} of accumulation-mode aerosols was used in Kim et al. (2008) to show that the ACI_r can be better manifested in the adiabatic cloud than in sub-adiabatic environment, despite the relatively lower values (0.04 – 0.17) retrieved in stratus cloud cases during the period 1999 - 2001. Moreover, this influence of thermodynamic condition on ACI_r was further documented in Kim et al. (2012) where the aerosol-cloud interaction found to be enhanced under the condition of strong inversion above the stratus layer.~~

In order to better understand the aerosol particles activation process in typical continental low-level stratus clouds, in the latter part of this study, the ratios between N_{CCN} and N_a are

examined and used to represent the aerosol activating capacities. Since the aerosol activating capacities are greatly depend on the aerosol sizes and compositions, in order to further examine the role of aerosol species in aerosol activation process and the potential impact on ACI_r, the samples from the 16 selected cases are divided into two groups according to their absorptive regime which is discussed in the following section. ~~The assumption when using ACI_r is that there exists a significant relationship between aerosol loading and CCN, thus a nearly constant fraction of aerosol effectively activates as CCN. In essence, aerosol loading is more important than the aerosol size and composition. However, the ACI_r values from all samples should be interpreted with caution since this assumption may not always be valid and is conditional. In order to further examine the role of aerosol species in ACI_r, the samples from the 16 selected cases are divided into two groups according to their absorptive regime which is discussed in the following section.~~

3.3 Relationship between aerosol absorptive properties and ACI

3.3.1 Aerosol absorptive properties of the 16 selected cases

The measured absorptive properties of aerosols can aid in inferring the general information of different aerosol species since different types of aerosols can demonstrate different absorptive behaviors at certain wavelengths. Aerosol plumes dominated by organic carbonaceous particles tend to represent strong absorptive capabilities in the visible spectrum but weakly absorb in near-infrared (Dubovik et al., 2002; Lewis et al., 2008) while black carbon particles (e.g., soot) absorb across the entire solar spectrum with a weak dependence on wavelength (Schuster et al., 2005; Lack and Cappa, 2010). However, when the aerosol plume is dominated by anthropogenic inorganic pollution, the absorbing ability becomes even weaker (Clark et al., 2007), partly due to sulfate chemical species (Chin et al., 2009). Therefore, the general existence of carbonaceous and pollution particles can be inferred via absorptive properties.

In this study, we adopt the classification method involving AE and the ratio of aerosol absorption coefficient to total extinction coefficient or single scattering co-albedo, ($\omega_{abs} = \sigma_{abs}/(\sigma_{abs} + \sigma_{scat})$) (Logan et al., 2013; Logan et al., 2014). This parameter is more sensitive

to the capabilities of aerosol light absorption (rather than scattering) to total aerosol light extinction and therefore can better infer the aerosol composition (Logan et al., 2013). The ω_{abs} values at a wavelength of 450 nm along with the $AE_{450\text{-}700nm}$ of all the samples are shown in Fig. 4. A ω_{abs} value of 0.07 is used as a demarcation line of aerosols that are weakly and strongly absorbing. This value was determined using a frequency analysis performed at four AERONET sites that are dominated by single aerosol modes (Logan et al., 2013). Of the 16 cases, six cases are dominated by strongly absorbing aerosols, six cases are dominated by weakly absorbing aerosols, and four cases have samples which broadly scatter across the ω_{abs} domain which denotes a mixture of different absorbing aerosol species.

Within the 693 selected samples, 360 data points are classified in the weakly absorptive aerosol regime, while the remaining data points are in the strongly absorptive aerosol regime. It is interesting to note that the majority of the winter cases are dominated by weakly absorbing aerosols while most of the spring cases exhibit a strongly absorbing aerosol dominance which suggests that the aerosol plumes over the SGP site also have a seasonal dependence. In spring, owing to the upper-level ridge centered over the western Atlantic, the SGP is located at the northwestern edge of the sub-tropical high. Under this synoptic pattern, the SGP is under the influence of relatively frequent southerly transport of the airmasses from Central America, which is characterized by strongly absorbing carbonaceous aerosols produced from biomass burning, as well as the moisture transported from the Gulf of Mexico. During the winter, the SGP site experiences the transported airmasses from higher latitudes with less intrusion of airmasses from the south (Andrews et al., 2011; Parworth et al., 2015; Logan et al., 2018).

3.3.2 Aerosol and cloud properties under different absorptive regimes

Figures 5a-5c show the PDFs of total N_a, N_{CCN}, and AE for the two absorptive regimes classified by ω_{oabs}. The distributions of N_a from the two absorptive regimes is comparable to one another. The mean N_{CCN} for the weakly absorptive regime (559 cm^{-3}) is larger than that from the strongly absorptive regime (384 cm^{-3}), and the occurrence of high N_{CCN} values (larger than 1000 cm^{-3}) is also higher in the weakly absorptive regime. This suggests different responses of CCN concentration to aerosols that have similar magnitudes but different absorptive properties. The AE distributions suggest dominant fine mode aerosol contributions

for both regimes. As for the cloud microphysical property distributions, cloud samples between the two regimes exhibit different characteristics (Fig. 5d-5f). The numbers above the bars in LWP distribution (Fig. 5d) for the two absorptive regimes denote the number of data points which will be used in the analysis with binned LWP in the later sections. Cloud LWPs and r_e values under the strongly absorptive regime have larger values which contrasts with those under the weakly absorptive regime. On average, the weakly absorbing regime has higher N_d and smaller r_e (374 cm^{-3} and 6.9 μm, respectively) compared to the strongly absorbing regime (214 cm^{-3} and 8.2 μm). Note that the LWPs under the strongly absorptive regime are generally higher those under the weakly absorptive regime. This LWP difference might be associated with the seasonality of airmass transport over the SGP as discussed in section 3.3.1. Although the seasonality of aerosol distribution and LWP have similar trends, no clear causality has been found between them. Thus, the question behind these results is whether the differences in cloud microphysical properties between the two regimes are due to the difference in LWP. As previously stated by Dong et al. (2015), cloud droplets generally grow larger at higher LWPs, which eventually leads to lower droplet number concentration.

3.3.3 Relationship of aerosol activating as CCN under different absorptive regimes

The measured N_a and N_{CCN} under the strongly and weakly absorbing aerosol regimes are plotted in Fig. 6. Note that N_a samples from both regimes cover a broad range of values from 200-3500 cm^{-3}, suggesting a wide variety of aerosol loading conditions. These highly overlapping distributions allow a quantitative comparison between the ratios of N_{CCN} to N_a. For a broad range of N_a, especially 200-5700 cm^{-3} and 11200-3500 cm^{-3}, the majority (~74%) of sample points from the strongly absorbing regime are located below the samples from the weakly absorbing regime. The linear regressions (95% confidence level) between N_{CCN} and N_a for two regimes demonstrate the sensitivity of $CCN_{0.2\%SS}$ to total aerosol loading. Note that the slope derived from the weak regime is slightly steeper than the strong regime, indicating that the N_{CCN} values in the weakly absorptive regime increase faster than in the strongly absorptive regime with same amount of aerosol increment. On average, 54% of weakly absorbing aerosols can effectively activate as CCN compared to 45% of the strongly absorbing aerosols. Note that those ratios are computed for an observed supersaturation level of 0.2%.

The fraction of aerosols that can activate as CCN increases with an increase in supersaturation level, under the same aerosol size and composition condition (Dusek et al., 2006). A sensitivity test of how the aerosol activation rate varies with different supersaturation levels is done by first interpolating the N_{CCN} from 0.2 % to 1.15 % and then calculating the N_{CCN}/N_a. As a result, the ratios of N_{CCN}/N_a for the weakly absorptive regime range from 0.54 to 0.38, while the ratios for the strongly absorptive regime range from 0.45 to 0.25. Considering that the supersaturation level in the continental boundary layer stratus is nearly impossible to reach the level of 1.15%, the supersaturation level of 0.2% used in this study, which represents the most typical condition for continental low stratus, yields reasonable results.

The aerosol capacity to activate as CCN is substantially associated with size and chemical composition (Seinfeld and Pandis, 2006). Although it is generally considered that the role of aerosol particle size distribution is more important than the chemical component in terms of becoming CCN (Dusek et al., 2006), many studies have found that aerosol chemical composition can also have a non-negligible impact on the aerosol activating ability under different polluted conditions (Rose et al., 2011; Che et al., 2016), especially under low supersaturation conditions. According to Kohler theory, the critical level of supersaturation for aerosol activation depends on the aerosol solubility which decreases with increasing soluble particle number concentration. Hence, the role of aerosol chemical composition is more important at lower supersaturation and diminishes with increasing supersaturation level (Zhang et al., 2012).

As discussed in section 3.3.1, both weakly and strongly absorptive regimes are linked to aerosol plumes that are dominated by pollution and carbonaceous aerosols, respectively. Therefore, the difference in the ability of aerosol activation between the two regimes can be explained by the different hygroscopicity factors of the particle types. For example, anthropogenic pollution is associated with inorganic particles that are highly hygroscopic and have great ability in taking up water (Hersey et al., 2009; Massling et al., 2009; Liu et al., 2014), while carbonaceous species (e.g., black and organic carbon) exhibit varying degrees of hygroscopicity with species dominated by hydrophobic soot and black carbon being the least hygroscopic (Shinozuka et al., 2009; Rose et al., 2010). Thus, for the given amount of aerosol

loading, aerosols in the weakly absorptive regime can better attract water vapor molecules and result in more aerosol particles activating as CCN.

As shown in Fig. 6, for three N_a ranges (200 - 500; 500 - 1100 and 1100 - 3500 cm^{-3}), the strongly absorbing aerosols show different relationships compared to weakly absorbing aerosols. The mean N_{CCN}/N_a values for those three N_a ranges for weakly absorptive regimes are 0.77, 0.58, and 0.42, respectively, while the mean N_{CCN}/N_a values for the strongly absorptive regimes are 0.35, 0.51, and 0.32, respectively. This phenomenon is due to the mixed effect of aerosol composition (inferred by absorbing ability), aerosol size, and water availability on the aerosol activation. In the 200 - 500 cm^{-3} N_a range, where the samples from the two absorptive regimes are most separated. The mean values of LWP (158 gm^{-2}/162 gm^{-2} for weakly/strongly absorptive regimes) indicate relatively sufficient water availability with less aerosol concentration. In addition, the weakly absorbing aerosol sizes are larger (AE = 1.59) than the strongly absorbing aerosol (AE = 1.73). It is known that larger aerosol particles easily activate under same composition (Dusek et al., 2006), considering the weakly absorbing aerosol is more hydrophilic, the largest conversion rate difference among these three ranges are to be expected. The samples in the 500 - 1100 cm^{-3} N_a range, have AE value (1.40) for strongly absorbing aerosols are noticeably smaller than those in the weakly absorptive regime (1.53), and the LWP in strongly absorptive regime (167 gm^{-2}) is much higher than in weakly absorptive regime (138 gm^{-2}). Therefore, the combined effect of larger particles and more water in strongly absorptive regime lead to the N_{CCN}/N_a is close to the N_{CCN}/N_a ratio in weakly absorptive regime. The samples in the 1100 - 3500 cm^{-3} N_a range exhibit smaller (AE = 1.67/1.57 for weakly/strongly absorptive regimes) aerosol particle size and less water availability (LWP = 95 gm^{-2}/127 gm^{-2} for weakly/strongly absorptive regimes) which results in the lowest activation rate (N_{CCN}/N_a ratio = 0.42/0.32 for weakly/strongly absorptive regimes) among the three ranges for both regimes.

Due to the lack of detailed chemical observations for all the cloud sample periods, as well as the uncertainties among aerosol optical and microphysical properties induced by aerosol transformation processes such as aging and mixing (Wang et al., 2018b), the bulk activation rates revealed from this study cannot be significantly distinguished from each other. However,

the effect of different aerosol species inferred by the absorptive properties with respect to aerosol activation is evident, especially at the 0.2% supersaturation level. Furthermore, in the following section, the values of N_{CCN}/N_a and AE are sorted by LWP for the two absorptive regimes, in order to rule out the influence of LWP and AE on aerosol activation to the utmost extent.

3.3.4 LWP dependence of aerosol and CCN activation under different absorptive regimes

In order to better understand the role of aerosol activation ability in the microphysical process from aerosol to CCN and then to cloud droplet, comparisons must be considered under similar available moisture conditions due to the discrepancy of LWP between the two regimes. Accordingly, the sorted N_a values by stratified LWP are presented in Fig. 7a, along with the conversion ratios of N_{CCN}/N_a which are denoted by solid lines. For a range of LWPs from 20-300 $\mathrm{g\,m^{-2}}$, the ratios of N_{CCN}/N_a under both regimes increase slightly with increased LWP. In addition, all binned N_{CCN}/N_a values from the weakly absorptive regime (ranging from 0.4 to 0.6) are higher than those from the strongly absorptive regime (ranging from 0.3 to 0.5). A student's t-test is performed to test the ratio difference in each LWP bin at the 95% significance level. The results indicate the ratio differences between two absorptive regimes are statistically significant.

Taking the variation of N_{CCN} into account, the conversion rates of ~~N_{CCN}~~-N_a to ~~N_a~~-N_{CCN} under low LWP conditions (<50 $\mathrm{g\,m^{-2}}$) in both regimes could be simply due to the linear combination of high aerosol concentration and insufficient moisture supply, such that aerosols are competing against each other thus resulting in a low conversion rate. However, as LWP increases, the activation rates tend to increase as well, especially at LWP values higher than 100 $\mathrm{g\,m^{-2}}$. In fact, the values of N_a in both regimes are relatively small with little variation for LWP > 100 $\mathrm{g\,m^{-2}}$, while the N_{CCN}/N_a ratio demonstrates a more noticeable increasing trend in the weakly absorptive regime. Despite a higher aerosol loading in the strongly absorptive regime at large LWPs, there are still more weakly absorbing aerosols being activated, which corresponds to greater water uptake ability. Moreover, in every LWP bin, the AE value for the weakly absorptive regime is either higher than or very close to the AE value for the

strongly absorptive regime (Figure not shown). Even with relatively smaller particle sizes, under similar water availability, the weakly absorbing aerosol can better activate as CCN. In conclusion, the significant effect of aerosol composition, which inferred by aerosol absorbing ability, on the aerosol activation capacity does exist.

As for the process from CCN to cloud droplet, a similar assessment is presented in Fig. 7b, which illustrates the N_{CCN} values and conversion rates of N_d to N_{CCN} in relation to LWP. The conversion rates of N_d/N_{CCN} in the weakly absorptive regime range from 0.58 to 0.86 with a mean value of 0.69, and highly fluctuates with LWP. In contrast, the conversion rates in the strongly absorptive regime show lower values and less variability (from 0.47 to 0.64) with a mean value of 0.54. It is interesting to note that the variation of N_d/N_{CCN} in the strongly absorptive regime mimics the variation in N_{CCN} with LWP, indicating a relatively lower aerosol to CCN activating capacity. Therefore, the conversion rate for CCN to cloud droplet shows no significant dependence on LWP, which is consistent with previous studies which suggest the response of N_d to the change in N_{CCN} has no fundamental relationship with LWP (e.g., McComiskey et al., 2009). In addition, the sensitivity and uncertainty of N_d is examined in order to estimate the impact of N_d uncertainty on the assessment of CCN activation rate. To assess the contributions of different input parameter uncertainties to N_d retrieval, every input parameter was perturbed by its own uncertainty with other parameters held fixed. The results are as follows: (a) an increase (decrease) of LWP by 20 gm^{-2} leads to 27.9% (27.6%) change in N_d while an increase (decrease) σ_x by 0.15 leads to a 50.8% (23.9%) change in N_d; (b) an increase (decrease) cloud thickness by 0.15 leads to a 14.5% (23.2%) change in N_d; and (c) an increase (decrease) in r_e by 10% leads to 14.5% (23.2%) change in N_d. The percentage changes in N_d due to different input uncertainties range from 14.5% to 50.8%, with the majority falling between 20% and 30%. Note that the largest uncertainty of N_d happens when increasing σ_x by 0.15. However, when considering that continental stratocumulus generally contains smaller droplets, one might expect their distribution width to be smaller than 0.38 (Dong et al., 1997). Therefore, the overall uncertainty of 25% compared to the aircraft in-situ measurement should be a reasonable estimation. In this case, the mean ratio of N_d/N_{CCN} for the weakly absorptive

aerosol regime range from 52% to 86%, while the mean ratio of N_d/N_{CCN} for the strongly absorptive aerosol regime range from 41% ~ 67%.

The overall differences in CCN conversion rate are likely a result of the combined effects of meteorological factors and aerosol radiative effect on the cloud environment. To examine the meteorological influence on cloud droplet activation, the LTS parameter is used to investigate the difference in the large-scale thermodynamic condition. By sorting the LTS by LWP for the two absorptive regimes, the LWP dependence on LTS can be ruled out, which can provide a better understanding of the potential role of LTS in cloud droplet development. For each given LWP bin, the weakly absorptive regime has higher LTS values than the strongly absorptive regime (figure not shown). The LTS is largely impacted by the potential temperature difference throughout the mixed layer and if a strong temperature inversion that caps the boundary layer is present, it will result in high LTS values and in turn, a well-mixed boundary layer (Wood et al., 2006). Such results indicate that even under similar available moisture condition, the more sufficient turbulence can transport the below-cloud moisture as well as the CCN that activated from weakly absorbing aerosols into the cloud more efficiently, contributing to a higher conversion rate of N_d/N_{CCN} in the weakly absorptive regime. However, the LTS emphasizes a general thermodynamic condition in the lower troposphere with a wider domain as compared to the single-point measurement.

In addition, the vertical velocity in pressure coordinate (Omega) values at the 925 hPa level, which represent the large-scale forcing on the vertical motion between surface and cloud-layer, are also sorted by LWP for the two absorptive regimes in order to check the potential influence of the environmental dynamic state (figure not shown). However, the Omega for both absorptive regimes share the same mean value of 0.031 Pa/s and show no dependence on LWP, indicate that the large-scale environments over the SGP are generally dominated by sinking motion. The synoptic patterns of composite geopotential height for the two absorptive regimes show that the SGP site is located ahead of the 700 hPa ridge and is located within the 925 hPa high. The meteorological pattern is favorable for generation of downward motion at the lower troposphere, and the sinking motion induces relatively stable environments in the lower troposphere which is consistent with the LTS measurements. Considering the fact that the

Omega value is obtained from a relatively larger domain surrounding the SGP, it is difficult to reflect the true cloud-scale dynamics, especially the vertical velocity or turbulence strength at the cloud base. Therefore, the influence of cloud-scale dynamics, presumably cloud-base updraft, is not negligible since the sensitivity of cloud droplet to aerosol loading is enhanced with increasing updraft velocity as reported in previous studies (e.g., Feingold et al., 2003; McComiskey et al., 2009).~~The influence of cloud dynamics, presumably cloud-base updraft, is not negligible since the sensitivity of cloud droplet to aerosol loading is enhanced with increasing updraft velocity as reported in previous studies (e.g., Feingold et al., 2003; McComiskey et al., 2009).~~

Furthermore, the radiative effect of light-absorbing aerosols on the cloud environment cannot be neglected, since the strongly light-absorbing aerosols can absorb solar radiation and heat the in-cloud atmosphere by emission, which results in the reduction of relative humidity (or supersaturation) in the cloud layer (Bond et al., 2013; Wang et al., 2013). This effect is evident by the observation as the values of in-cloud relative humidity in the strongly absorptive regime are slightly lower than those in the weakly absorptive regime. Additionally, this aerosol heating effect disrupt the boundary layer temperature structure by enhanced warming aloft, and consequently, inhibit the vertical transport of sensible and latent heat between surface and cloud layer. The impacts of light-absorbing aerosol on cloud-scale thermodynamics and dynamics state might eventually dampens the conversion process from CCN to cloud droplet.~~Furthermore, the radiative effect of light-absorbing aerosols on the cloud environment also cannot be neglected, since the strongly light-absorbing aerosols can absorb solar radiation and heat the in-cloud atmosphere by emission. This perturbation of temperature structure results in the reduction of supersaturation in the cloud layer (Bond et al., 2013; Wang et al., 2013), and eventually dampens the conversion process from CCN to cloud droplet.~~

Unfortunately, due to the lack of measurement of cloud-base vertical velocity throughout the studying period, this competing effect of cloud thermodynamic and dynamic cannot be fully untangled from the aerosol effect given the currently available dataset. The differences in conversion rates of N_d/N_{CCN} between the two regimes might be affected by the combined effects of LTS, updraft velocity, and aerosol absorption effect on the cloud environment.

3.3.5 r_e and N_d dependence of LWP under different absorptive regimes

In the previous section, we examined the activation rates of aerosol to CCN and then from CCN to cloud droplet between the two regimes as well as their dependences on LWP, that eventually led to the cloud droplet variation for a given LWP range. Figures 7c-7d demonstrate that r_e increases while N_d decreases with increased LWP up to roughly 150 g m^{-2} in both regimes. Note that as LWPs greater than 150 g m^{-2}, N_d values in both regimes show less variation with LWP while r_e values in the strongly absorptive regime also show little variation which implies limited growth even with increasing water availability. However, the r_e values in the weakly absorptive regime increase from 7.8 to 8.8 μm, which suggests that under a given number concentration, the cloud droplet can grow by continuing to collect moisture. As shown in each LWP bin, the r_e values in the weakly absorptive regime are smaller than those in the strongly absorptive regime, while the N_d values in the strongly absorptive regime are much lower than those in the weakly absorptive regime.

The combination of cloud thermodynamic, dynamic, and aerosol radiative effects impact the conversion process from CCN to cloud droplet. Under a given moisture availability, a greater number of CCN in the weakly absorptive regime can be converted to cloud droplets. This results in higher number concentrations of smaller cloud droplets, while the lower CCN activating rate in the strongly absorptive regime leads to fewer and larger cloud droplets at a fixed LWP.

3.3.6 Aerosol-cloud-interaction under different absorptive regimes

To examine the sensitivity of clouds to both weakly and strongly absorbing aerosol loading, the relationships between cloud r_e and N_{CCN} are shown in Fig. 8. Two LWP ranges (0-50 g m^{-2} and 200-250 g m^{-2}) are selected in order to better represent ACI_r at low and high LWP conditions. For the examination of r_e as a function of N_{CCN} (Fig. 8a for low LWP range), the ACI_r values in the weakly absorptive regime are higher than those in the strongly absorptive regime. This suggests that the cloud droplets are more sensitive to weakly absorbing aerosols than to strongly absorbing aerosols in clouds with low LWPs. In other words, if there is some increment in aerosol particles, clouds influenced by weakly absorbing aerosols will respond to this increment more effectively and decrease faster in droplet sizes relatively. Under high LWP

conditions (Fig. 8b), the ACI_r values are lower and show less difference between the two regimes, which is in agreement with previous discussions on the sensitivity of cloud microphysical properties to aerosol loading.

Based on the sensitivity study, the 10% change of cloud LWP and downward SW at the surface would result in the 10% uncertainty in r_e retrieval (Dong et al., 1997). When compared with aircraft in situ measurements, the differences between retrievals and in situ measurements are around 10% (Dong et al. 1998 and 2002). ~~Therefore, to assess the impact of r_e uncertainty on ACI_r, we placed the anthropogenic perturbations within the corresponding uncertainty (±10%) range onto r_e and recalculated the additional regression fits (dotted lines) for each regime in Figure 8. As a result, for that 10% change in r_e, the change in the logarithmic slopes (ACI_r) is almost negligible, which indicates that the impact of r_e uncertainty on ACI_r is minor and the observed differences do exist.~~ In order to assess the impact of r_e uncertainty on ACI_r, we use the Monte Carlo method to propagate the r_e uncertainty on ACI_r, with the procedure given as follows: For each data point, the r_e value is randomly perturbed to be increased or decreased by 10%, and thus the corresponding ACI_r can be re-calculated based on the perturbed set of r_e. After 100,000 iterations, we obtain a distribution of ACI_rs. The uncertainty of ACI_r is given by one standard deviation of those 100,000 values of ACI_rs, since the distribution of ACI_rs follows a normal distribution with a narrow peak, this uncertainty value represents the uncertainty in the computed ACI_r due to errors in the r_e retrieval. The uncertainties of ACI_r for the two absorptive regimes are denoted as the dashed line in Fig. 8. In the lower LWP range (Fig. 8a), the ACI_r uncertainty is 0.020 (0.030) for the weakly (strongly) regime, account for the uncertainties, the difference in ACI_r between the two absorptive regimes is preserved. In the higher LWP range (Fig. 8b), the ACI_r uncertainty is 0.044 (0.023) for the weakly (strongly) regime, which is non-negligible. Taking the uncertainties of ACI_r into account, the ACI_r in the two absorptive regimes cannot be well separated, owing to the enhanced collision-coalescence process accompanied by higher LWP and the diminished cloud response to aerosols associated with different ω_{abs} values. In general, the 10% uncertainty in r_e retrieval contributes to 0.02 ~ 0.04 in ACI_r uncertainties.

Note that the LTS values from the weakly absorptive regime (22.91K and 19.78K) are higher than those from the strongly absorptive regime (21.72K and 17.83K) for the selected two LWP bins. As discussed in the previous section, on the one hand, owing to the stronger temperature inversion indicated by the higher LTS values, low clouds are more closely connected to weakly absorbing aerosols and moisture below cloud by efficient turbulence. ~~On the other hand~~ In order to quantify the impact of LTS on ACI_r, we adapted the criteria described in Grysperdt et al. (2016) that the LTS value of 18K denotes the demarcation line between high and low LTS regimes, and constrain the ACI_r for the two regimes by their LTS values accordingly. Owing to the highly limited sample points that fall into the low LTS category, the ACI_c can only be constrained in the high LTS condition. For the 0 - 50 gm^{-2} LWP range, the ACI_r for the weakly absorptive regime increases from 0.26 to 0.31, and the ACI_r for the strongly absorptive regime increases from 0.21 to 0.24. The enhancement effect of LTS on the ACI_r is noticeable, which in accordance with the previous discussion that high LTS environment is associated with (a) sufficient turbulence in the boundary layer and (b) a closer connection between the surface and cloud layer, which enhances the cloud microphysical responses to the CCN. Note that though ACI_rs are increased for both regimes, the difference between them becomes larger (from 0.05 to 0.07) because low-level stratus clouds are more susceptible to weakly absorptive aerosol. Furthermore, the enhancement of ACI_r in the high LTS environment is more evident in the weakly absorptive regime. In the case of the 200 – 250 gm^{-2} LWP range, the LTS effects on ACI_r are less significant compared to the lower LWP range. No significant change in weakly absorptive regime is evident and the ACI_r in the strongly absorptive regime decreased from 0.12 to 0.10, partly owing to the enhanced collision-coalescence process accompanied by higher LWP, and thus inhibits the impact of LTS on ACI_r. Overall, ACI_rs are enhanced under high LTS conditions, but the difference between two regimes indicates that both ω_{abs} and LTS can be the impact factor of the ACI_r but they are not necessarily having causality between them.

Furthermore, with the presence of strongly light-absorbing aerosols, the cloud layer heating induced by the aerosol absorptive effect can result in the reduction of in-cloud supersaturation and leads to the damping of cloud microphysical sensitivity to strongly

absorbing aerosols. In general, the results indicate that the ACI_r can be counteracted by the absorbing aerosol radiative effect and be enhanced under a thermodynamic environment of high static stability, especially under lower LWP conditions.

3.4 Cloud shortwave radiative effects under different absorptive regimes

Aerosols with different absorptive properties can alter the ability of clouds to reflect incoming shortwave radiation. Accordingly, cloud radiative effects on shortwave radiation for the two absorptive regimes are investigated. Both cloudy and clear-sky downwelling shortwave fluxes for samples in the weakly absorptive regimes are generally higher than those in the strongly absorptive regime (not shown in here), largely owing to the discrepancies in solar zenith angle, seasonal variation of insolation, and surface albedo. Therefore, to ensure the comparison is under minimum influence of non-cloud factors, the shortwave relative Cloud Radiative Effects (rCREs) are introduced and their dependencies on LWP between the two regimes are examined. With all else being equal, as shown in Fig. 9, rCREs in both regimes noticeably increase with LWP, especially for LWPs less than 150 $\mathrm{g\,m^{-2}}$. Using fixed LWP, rCREs in the weakly absorptive regime are always higher than those in the strongly absorptive regime, because the greater activating ability of the weakly absorbing aerosols leads to higher N_d and smaller r_e as opposed to the strongly absorbing aerosols. Thus, clouds with a larger amount of small cloud droplets contribute more to the extinction of incident solar radiation. The difference in mean rCRE between two regimes is small but non-negligible (~0.04). Quantitatively speaking, taking the climatological downwelling solar flux of the winter season (~150 $\mathrm{W\,m^{-2}}$, Dong et al., 2006) as an example, the extinction of incident solar radiation by clouds that develop from weakly absorbing aerosols is $6.0\ \mathrm{W\,m^{-2}}$ more than those by clouds from strongly absorbing aerosols. From independent radiative measurements, the phenomenon that clouds are more susceptible to weakly absorbing aerosols is further evident.

4 Conclusions

A total of 16 non-precipitating overcast low-level stratiform cloud cases under daytime coupled boundary layer conditions were selected in order to investigate the sensitivity of cloud microphysical properties to aerosol physicochemical properties. The Ångström exponent and fine mode fraction distributions indicate that the aerosol plumes that advected to the SGP site

during all the selected cases were dominated by fine mode particles, while the variation in aerosol single scattering albedo suggests different characteristics of optical properties among the aerosol plumes. In terms of the sensitivity of cloud droplets to aerosol number concentration, the values of ACI_r range from 0.09 to 0.24 with the mean of 0.145 ± 0.05, which supports the finding of previous studies using ground-based measurements. The magnitude of ACI_r shows a decreasing trend with increasing LWP, partly owing to the enhanced collision-coalescence process accompanied by higher LWP. However, clouds that develop under lower LWP conditions are more susceptible to aerosol loading, owing to the enhanced competition between aerosols to activate as cloud droplets with a limited supply of moisture.

The analysis of the N_{CCN}/Na ratio under the two regimes further demonstrates that weakly absorbing aerosols have statistically significant higher activation rates (mean ratio of 0.54) than the strongly absorbing aerosols (mean ratio of 0.45). The fraction of weakly absorbing aerosols that activate as CCN show a noticeable increase with increased LWP, while the activation rates for strongly absorbing aerosols tend to slightly increase with LWP under comparable aerosol loading conditions. This is likely related to the hygroscopicity associated with the aerosol species. For example, weakly absorbing aerosols are typically dominated by pollution aerosols that have greater water uptake ability, while strongly absorbing aerosols are generally hydrophobic, such as freshly emitted black and organic carbon (Wang et al., 2018b).

T~~Consequently, t~~he conversion rates of N_d/N_{CCN} for the weakly absorbing aerosols (mean ratio of 0.67) are higher than for the strongly absorbing aerosols (mean ratio of 0.54). Partly owing to the higher LTS environment for the weakly absorptive regime which enhance the connection between cloud and the below-cloud moisture and CCN. Also the cloud layer heating effect induced by the strongly light-absorbing aerosols results in the reduction of in-cloud supersaturation and leads to the damping of CCN activation process for the strongly absorptive regime. As a result, cloud droplets that form from weakly absorbing aerosols tend to have smaller sizes and higher concentrations than cloud droplets forming from strongly absorbing aerosols. Furthermore, the cloud droplets in the weakly absorptive regime exhibit a greater growing ability, as given by larger r_e values that increase with LWP under similar N_d. The

differences in cloud droplet development between the two regimes is a likely result of the combination of thermodynamics, dynamics, and aerosol radiative effects.

Under low LWP conditions, the measured ACI_r values in the weakly absorptive regime are relatively higher, indicating that clouds have greater microphysical responses to aerosols in weakly absorptive regime than in strongly absorptive regime, owing to favorable LTS condition in the weakly absorptive regime, and the cloud-layer heating effect of light-absorbing aerosol in the strongly absorptive regime. The observed ACI_r is enhanced after constrained by high LTS. Under higher LWP conditions, the enhanced collision-coalescence process diminish the LTS impact on ACI_r, and the damping of ACI_r is more evident, which is consistent with the results from all the cases. In general, the 10% uncertainty in r_e retrieval contribute to ACI_r uncertainties range from 0.02 to 0.04 for the two absorptive regime, with the ACI_r difference between the two absorptive regimes still well-preserved. As a result, clouds that develop from weakly absorbing aerosols exhibit a stronger shortwave cloud radiative effect than clouds originating from strongly absorbing aerosols. Additional future work will focus on investigating detail composition of different aerosol plumes, with respect to their physicochemical properties. The aerosol-cloud-interaction processes under the influence of different aerosol types associated with airmasses and the sensitivity to dynamic and thermodynamic factors will be further examined.~~Under low LWP conditions (<100 g m^{-2}), the measured ACI_r values in the weakly absorptive regime are relatively higher, indicating that clouds have greater microphysical responses to weakly absorbing aerosols than to strongly absorbing aerosols. Also, the observed ACI_r with respect to N_{CCN} is generally higher than N_a, which demonstrates that the mechanism from CCN to cloud droplet is more straightforward than from aerosol particle to cloud droplet. Under higher LWP conditions, the damping of ACI_r is more evident, which is consistent with the results from all the cases. As a result, clouds that develop from weakly absorbing aerosols serving as CCN exhibit a stronger shortwave cloud radiative influence than clouds originating from strongly absorbing aerosols. Additional future work will focus on investigating the seasonal dependence of aerosol sources, with respect to their physicochemical properties. The aerosol-cloud-interaction processes under the influence~~

~~of different aerosol types associated with airmasses and the sensitivity to dynamic and thermodynamic factors will be further examined.~~

Author contributions. The original idea of this study has discussed by XZ, BX, and XD. XZ performed the analyses and wrote the manuscript. XD, TL, YW, and PW participated in further scientific discussion and provided substantial comments and edits on the paper.

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Table 1. Dates and time periods of selected low-level stratus cloud cases and their airmass source.[a]

Date	Start Time (UTC)	End Time (UTC)	Airmass Source	Number of Data Points
4 Jan 2007	15:00	22:30	S	58
5 Jan 2007	14:00	18:10	S	40
13 Feb 2007	17:00	22:30	N	60
26 Apr 2007	14:00	17:30	NE	31
21 Nov 2007	13:20	18:15	N	24
14 Feb 2009	15:15	17:35	NW	29
12 May 2009	16:55	20:05	SE	37
19 Dec 2009	14:40	19:35	NW	58
21 Jan 2010	15:25	22:30	N	44
16 Mar 2010	15:00	20:00	N	41
29 Dec 2010	16:00	18:35	SE	32
26 Mar 2011	16:35	23:55	NE	59
13 May 2011	12:25	18:20	N	59
4 Feb 2012	16:40	21:10	NE	37
8 Feb 2012	14:30	19:45	N	54
10 Feb 2012	17:15	19:50	NW	30

[a]Airmass sources denote the relative directions from where the airmasses advected to the ARM-SGP site.

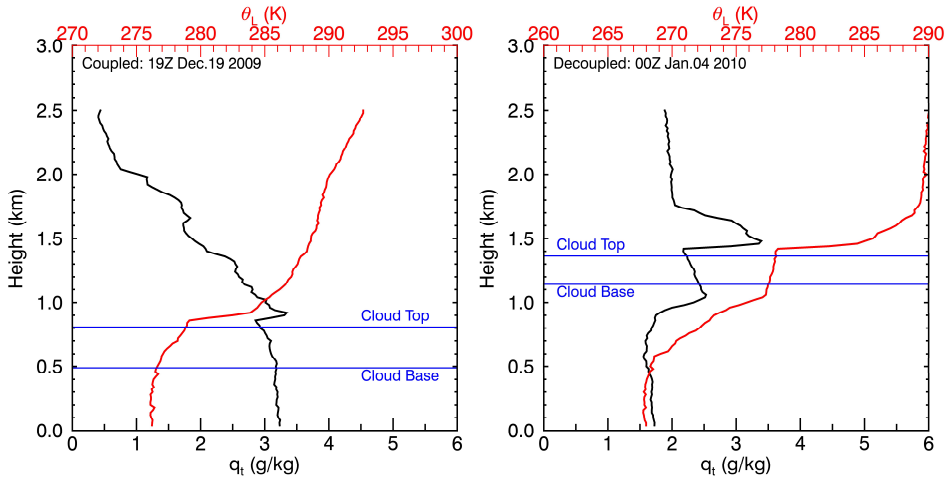

Figure 1. Vertical profiles of liquid water potential temperature (θ_L) and total water mixing ratio (q_t) for coupled (a) and decoupled (b) boundary layer conditions. Blue lines denote cloud top and base heights, respectively.

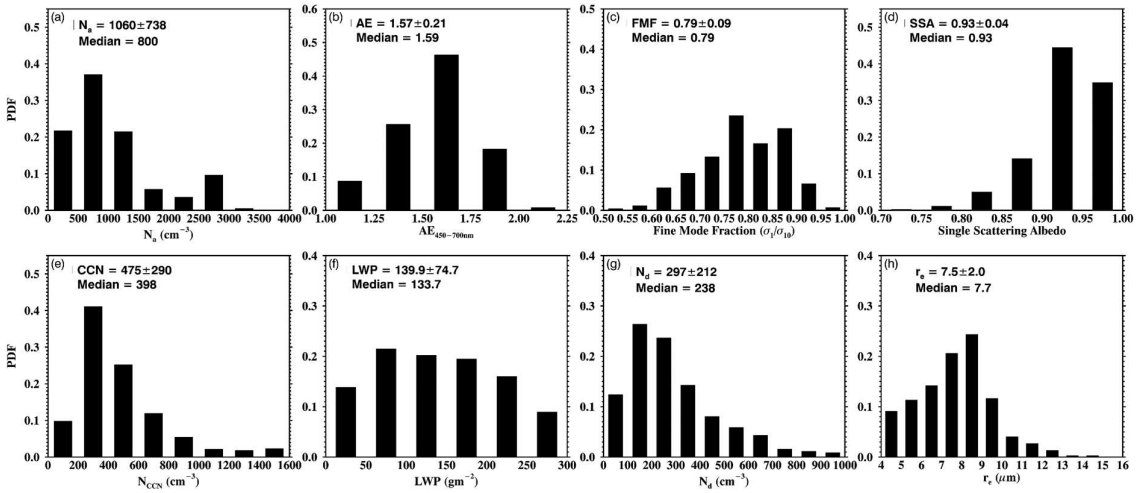

Figure 2. Probability distribution functions (PDFs) and mean values of low-level stratus cloud and aerosol properties for all cases: (a) total aerosol number concentration (Na); (b) Ångström Exponent (AE) derived from nephelometer measurements; (c) fine mode fraction at 550 nm; (d) single scattering albedo at 450 nm (SSA); (e) cloud condensation nuclei number concentration (N_{CCN}); (f) liquid water path (LWP); (g) cloud droplet number concentration (N_d); (h) cloud droplet effective radius (r_e).

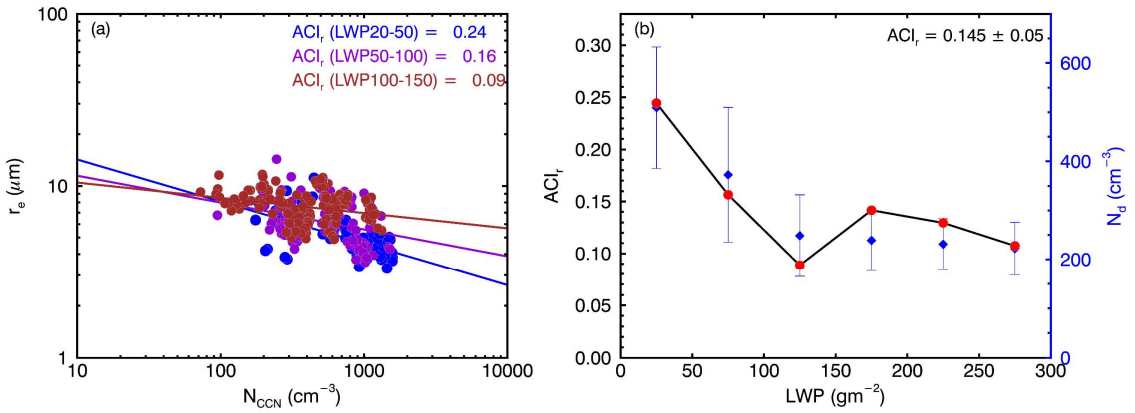

Figure 3. ACI_r derived from (a) r_e to N_{CCN} in following three LWP bins: 20-50 gm^{-2} (blue), 50-100 gm^{-2} (purple), 100-150 gm^{-2} (dark red) and (b) Relationship of ACI_r (red dot, left ordinate) and N_d (blue diamond, right ordinate) to binned LWP. Blue whiskers denote one standard deviation for each bin.

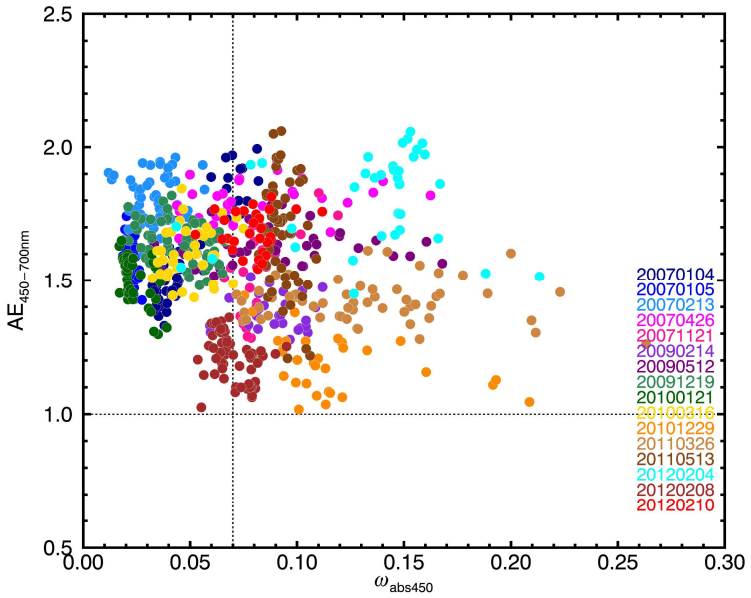

Figure 4. Angstrom Exponent ($AE_{450-700nm}$) and single scattering co-albedo ω_{abs450} of all samples (color coded by case). Horizontal dotted line denotes the demarcation of $AE_{450-700nm}$ = 1. Vertical dotted line denotes the demarcation of ω_{abs450} = 0.07.

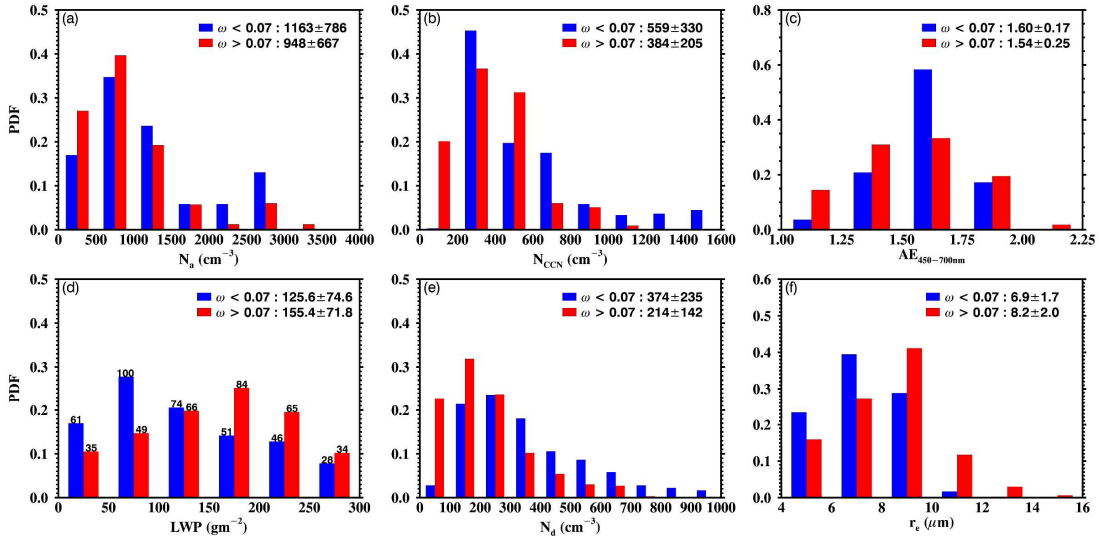

Figure 5. Aerosol and cloud properties under the strongly absorptive (in red) and weakly absorptive (in blue) aerosol regimes. PDFs, mean values and standard deviations of (a) N_a; (b) N_{CCN}; (c) $AE_{450-700nm}$; (d) LWP; (e) N_d; (f) r_e.

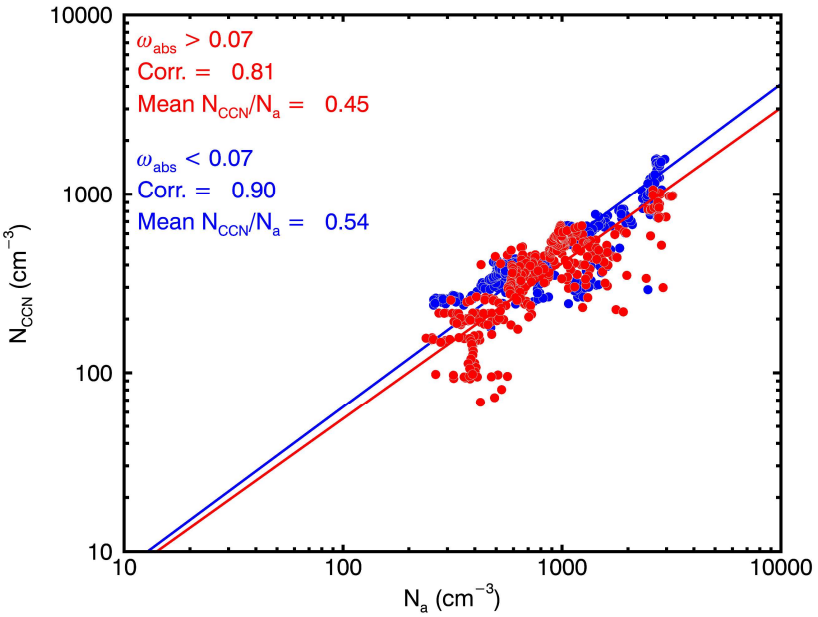

Figure 6. Relationship between N_{CCN} and N_a under the strongly absorptive aerosol regime (in red) and weakly absorptive aerosol regime (in blue).

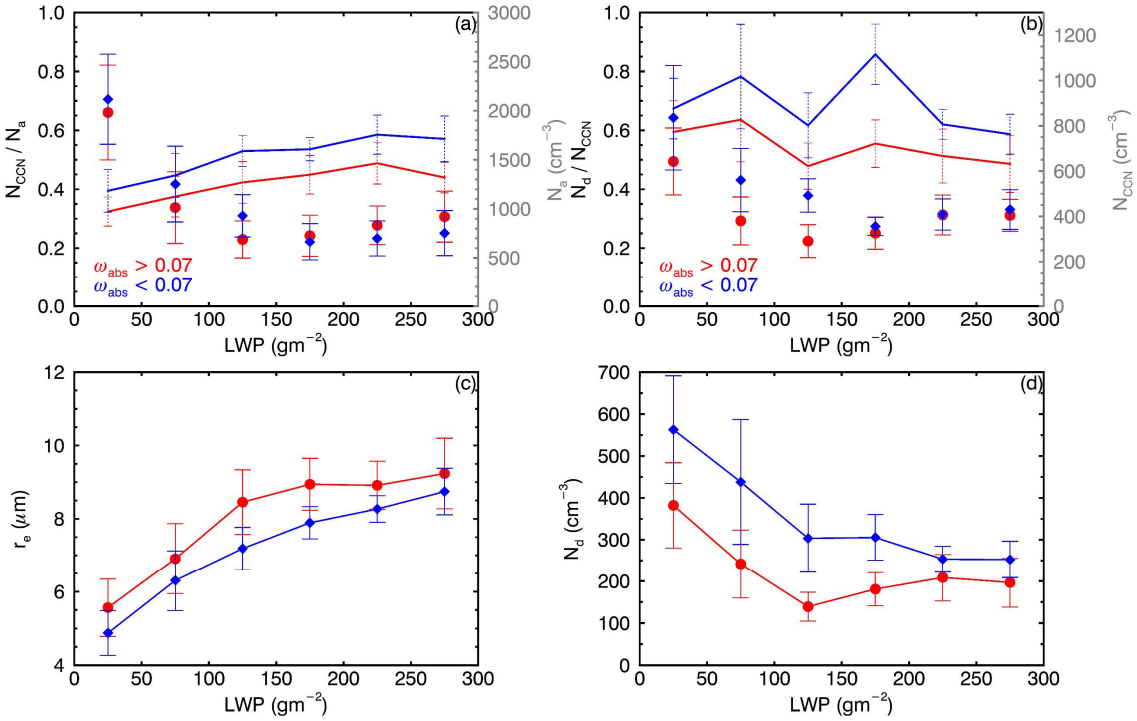

Figure 7. (a) N_a (dot) and the ratio of N_{CCN} to N_a (line); (b) N_{CCN} (dot) and the ratio of N_d to N_{CCN} (line); (c) r_e; and (d) N_d as a function of LWP under strongly absorptive (in red) and weakly absorptive (in blue) aerosol regimes. Whiskers denote one standard deviation for each bin.

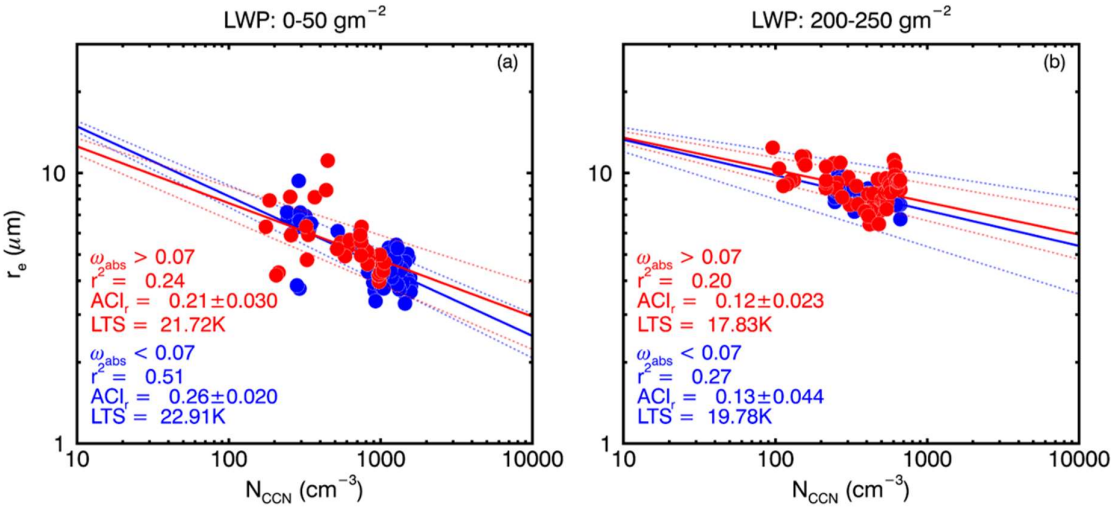

Figure 8. r_e as a function of N_{CCN} and the values of ACI_r under the strongly absorptive (in red) and weakly absorptive (in blue) aerosol regimes at two LWP bins: 0-50 g m^{-2} (a) and 200-250 g m^{-2} (b). Note that the dashed lines denote the uncertainties of ACI_r due to 10 % error in r_e retrieval.~~regarding the 10% uncertainty in r_e.~~

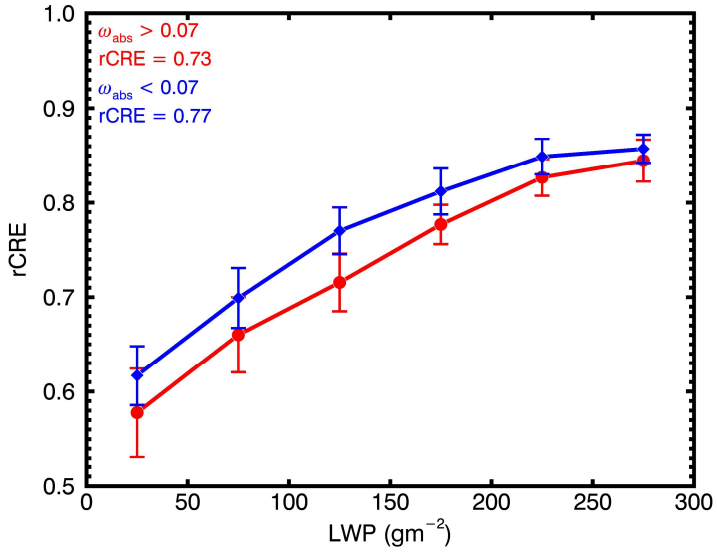

Figure 9. Relative Cloud Radiative Effect (rCRE) as a function of liquid water path (LWP) under the strongly absorptive (in red) and weakly absorptive (in blue) aerosol regimes. Whiskers denote one standard deviation for each bin.