Response to Reviewer #2

We appreciate your time for carefully reviewing our manuscript. We would like to thank you for the constructive comments and suggestions, which encourage and help us to improve the manuscript. The manuscript has been revised accordingly. In the response below, the reviewer's comments are provided in black text and our responses are provided in blue text.

Response:

General: Temporal resolution and number of data points While the temporal resolution of some of data products used to create the data set is given (I think everything is averaged to 5-minutes but it's not explicitly noted anywhere), nowhere is it stated the number of data points used to determine the ACIr index and other correlations. The 16 cloud cases and total time that the data set covers is provided in Table 1 but does not contain these statistics, which are important for interpreting results. If I do some math it seems that there are sufficient statistics but text needs to be added to fully and clearly describe the statistics of the data set for the reader.

Thanks for the comments. A total of 693 data points has been used in this study, and the detail of the number of data points used in every case was added in the revised Table 1. For clarification, the information of the number of data points has been added to the sentence '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' in section 2.5 in the revised manuscript.

In addition, to give the information of the number of data points that are categorized in two regimes, a sentence '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' has been added to the second paragraph of section 3.3.1 in the revised manuscript.

Furthermore, in the revised Figure 5d, the number of data points in every LWP bin is denoted by the numbers above every PDF bar for the two absorptive regimes. For clarification, the sentence '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' has been added in the first paragraph of section 3.3.2. in the revised manuscript.

Choice of aerosol data used - The authors choose to use the sub-micron aerosol optical properties from the available measurements rather than the sub-10 um. What is the motivation for this choice? The total aerosol number concentration Na and CCN are used which are not restricted to the sub-micron size cut. It's not fully consistent that the Na and CCN be sorted by high and low absorbing regime according to the sub-micron absorption – there could be a relationship between size and composition/absorption. Further, the sub-micron scattering fraction is presented alongside the scattering angstrom exponent for the sub-micron aerosol only. This lacks consistency and can make interpretation of the results difficult when reading through progressive steps in the analysis. An explanation (and implications) for how the sub-micron only properties relate to the others should be given if the choice of data is not changed. It probably won't change the overall picture given that the aerosol is largely sub-micron but why complicate the issue?

Thanks for the comments and suggestions. The original choice of submicron aerosol data was due to the consideration of fine-mode aerosol dominance over SGP. However, we totally agree that caused an inconsistency in aerosol number concentration and optical properties. Therefore, aerosol optical properties are based on measurements of the sub-10μm size-cut in the revised manuscript and the data were re-sorted by the (1-SSA) values of sub-10μm aerosols into the weakly and strongly absorptive regimes, accordingly. In general, the main results and conclusions did not change significantly given the fact that fine-mode aerosols dominated the aerosol plumes over SGP.

The Figures below show the corresponding changes after new aerosol results have been used in the revised version:

Figure 2: For the total dataset, the mean value of AE changes from 1.67 to 1.57; the mean value of SSA changes from 0.94 to 0.93, which results from the contribution of aerosols having a range of diameters from 1 – 10 microns.

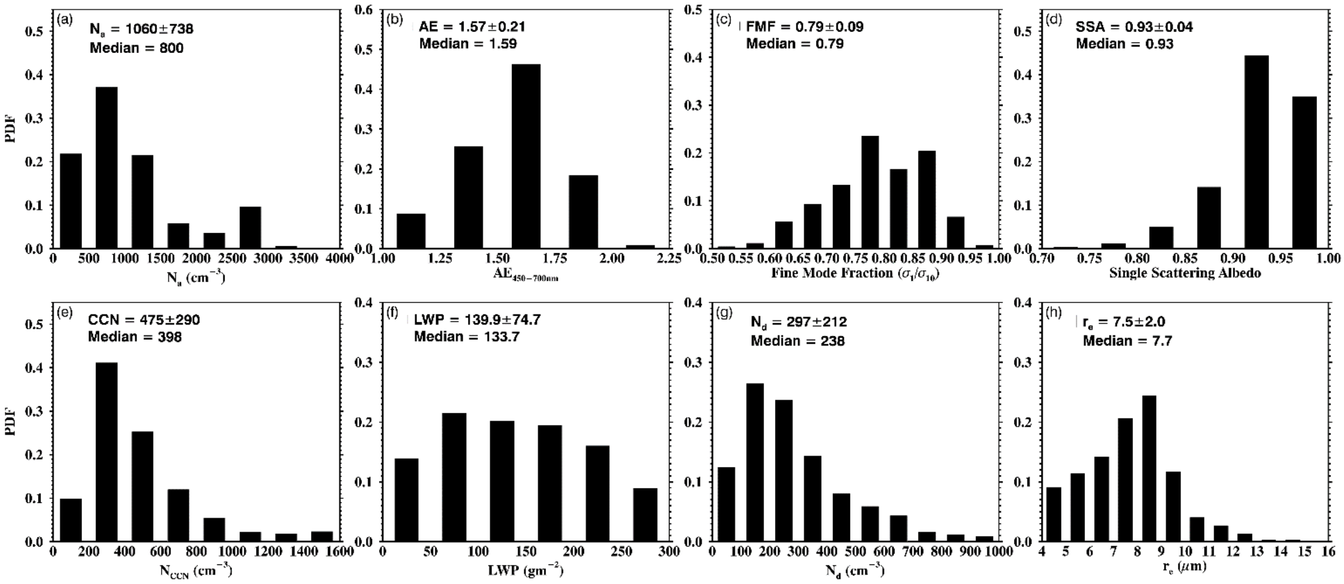


Figure 4: As indicated in the revised Figure 2, the values of (1-SSA) generally increase and AE values generally decrease owing to the inclusion of aerosols having diameters greater than 1 micron.

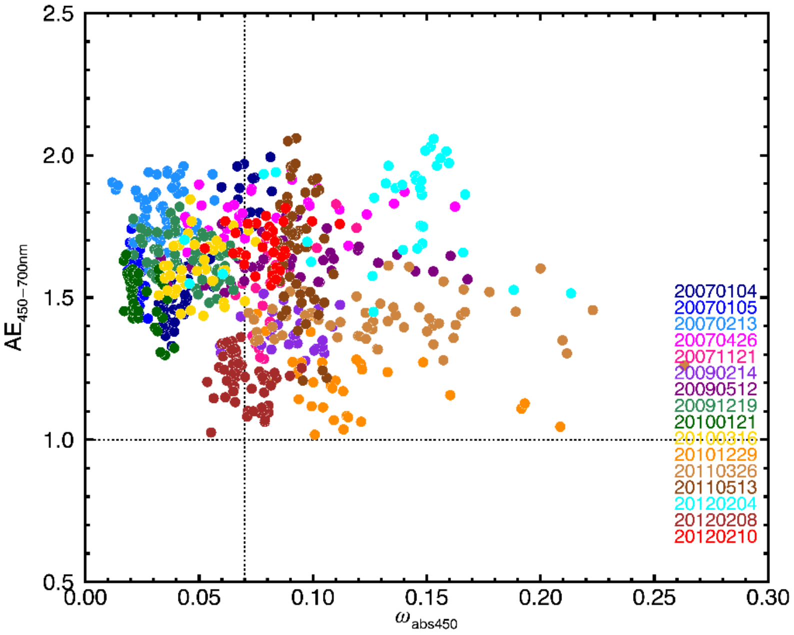


Figure 5: The mean values changed slightly due to the new categories of absorptive regimes, but the differences in distributions and mean values between the two regimes were preserved.

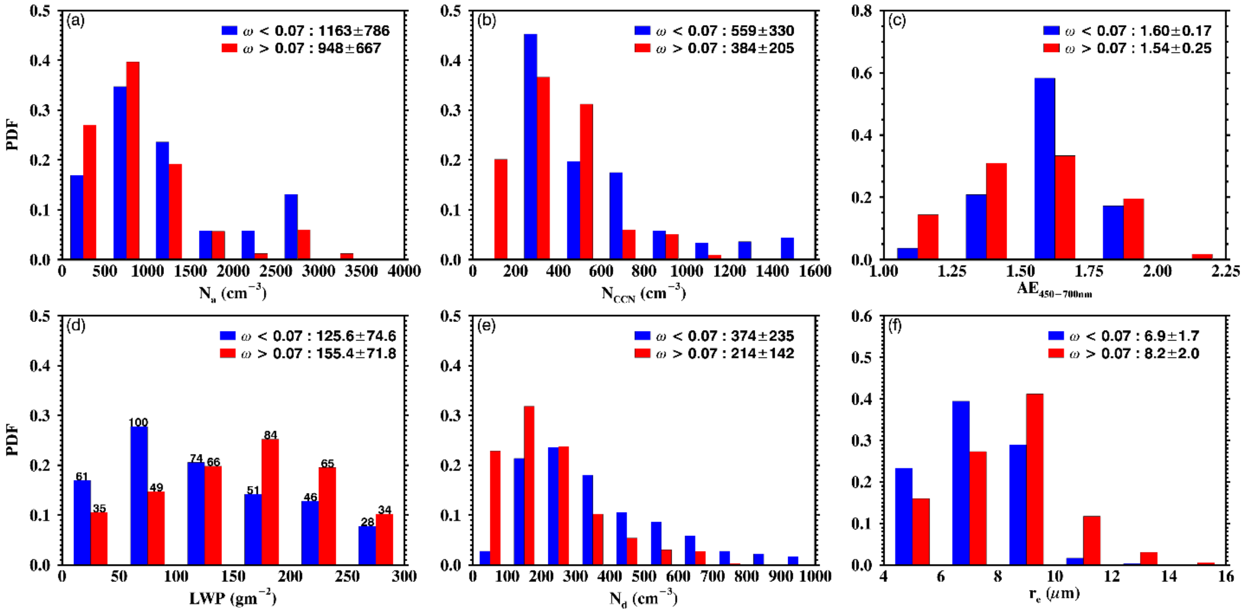


Figure 6: The overall activation rates did not change, with more data points from the strongly absorptive regime (red) located below the data points from weakly absorptive regime (blue).

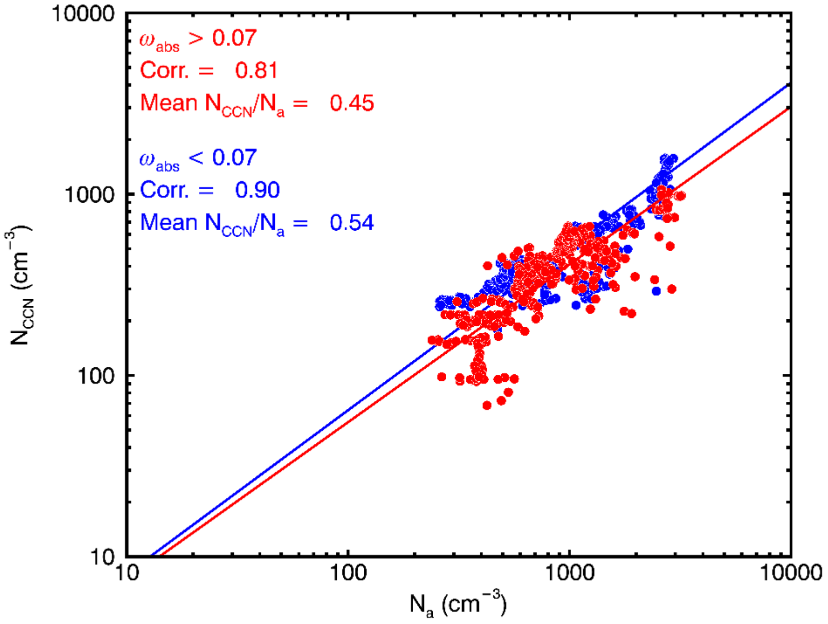


Figure 7: The standard deviations of the ratios were added as the dashed line. The differences in activation rates of N_{CCN}/N_a and N_d/ N_{CCN} changed slightly throughout the LWP range. The ratios of N_{CCN}/N_a range from 0.39 to 0.58 for the weakly absorptive regime and from 0.32 to 0.48 for the strongly absorptive regime. The ratios of N_d/N_{CCN} range from 0.58 to 0.86 for the weakly absorptive regime and from 0.47 to 0. 64 for the strongly absorptive regime.

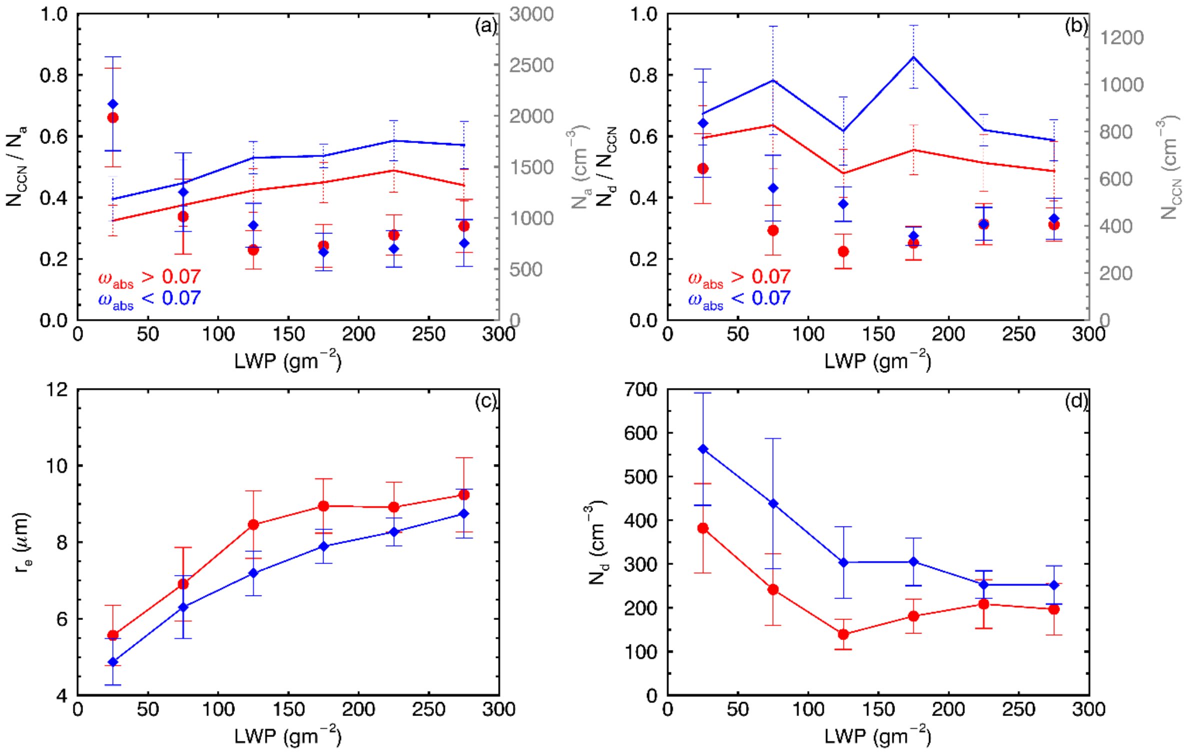


Figure 8: The top panel related to re as a function of N_a has been excluded in the revision following the suggestion of Reviewer #2. For the LWP bin of 0-50 gm^{-2}, the ACI_r values are 0.26 and 0.21 for the weakly and the strongly absorptive regimes, respectively. For the LWP bin of 200-250 gm^{-2}, the ACI_r values are 0.13 and 0.12 for the weakly and the strongly absorptive regimes, respectively. The differences in ACI_r between the two regimes and the damping of ACI_r with higher LWPs are still evident.

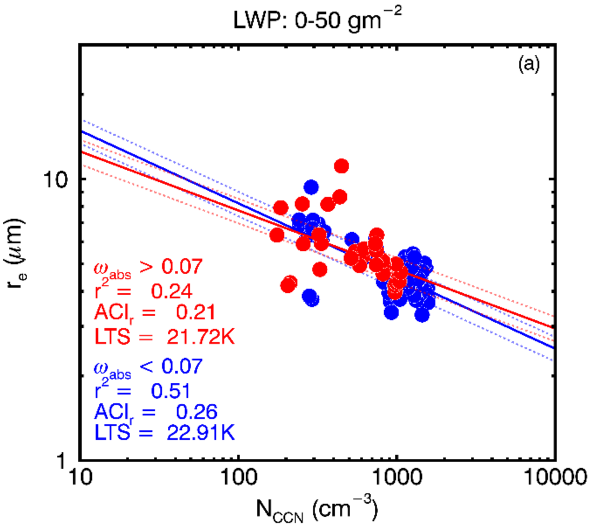
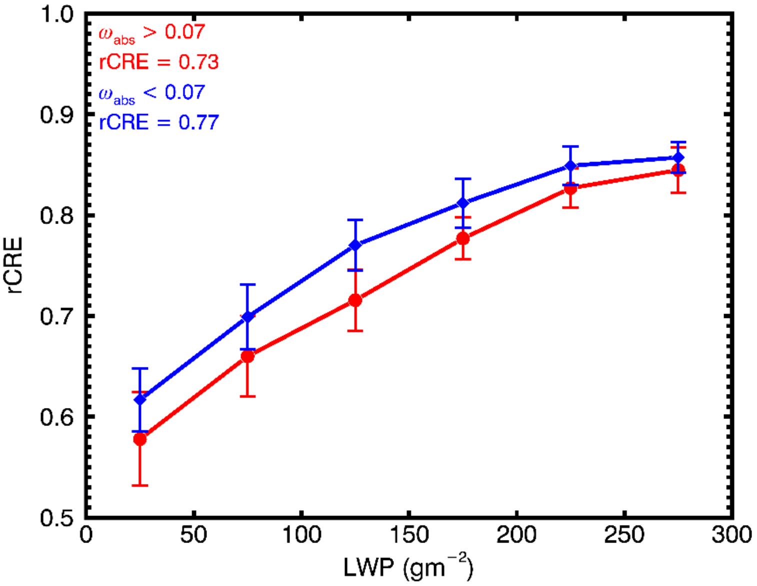


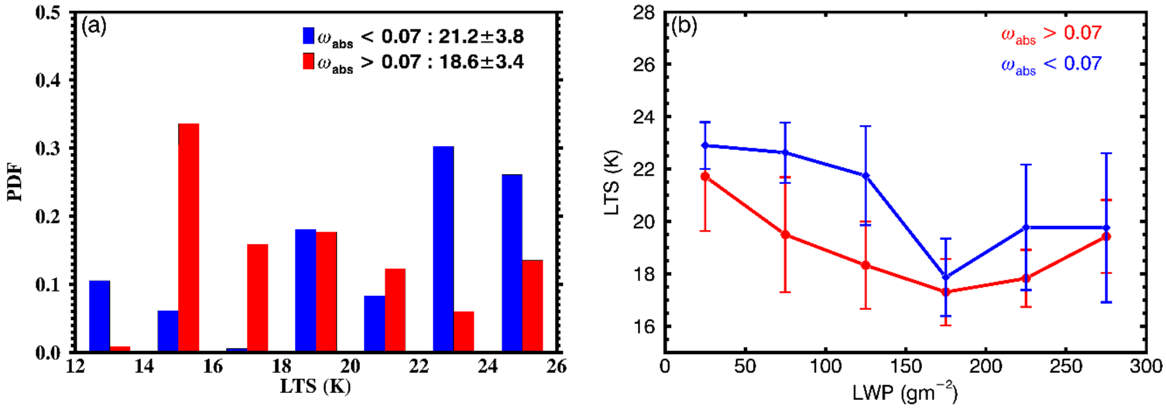
Figure 9: The mean rCRE for strongly absorptive regime changed from 0.72 to 0.73 while the mean value for weakly absorptive regime didn't change.



Lack of meteorological parameters or aerosol radiative effects in assessing covariances of aerosol and cloud properties - My greatest concern with this analysis relates to the association of aerosol absorption to cloud microphysics and cloud radiative effect without considering meteorological or systematic seasonal influences that may be affecting the co-variance of aerosol and cloud properties. The relationship of Na to CCN for high and low absorption regimes is compelling and it does seem that the difference in composition has an effect on the number of CCN. But then the examination of the relationship between CCN and drop number is presented without any discussion of controls by cloud dynamics or potential radiative effects of the absorbing aerosol on the environment of cloud dynamics.

Thanks for the comments and suggestions. We totally agree that both the meteorological factors and aerosol radiative effect could have a non-negligible influence on the aerosol-cloud interaction.

To examine the influence of meteorological factors, the Lower Tropospheric Stability (LTS), which is defined as the potential temperature difference between surface and 700hPa, is used to investigate the difference in 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. Accordingly, the above description of LTS dataset has been added to the revised section 2.3 - ‘Boundary Layer Condition and Lower Tropospheric Stability’ in the revised manuscript.



As shown in Figure (a), the weakly absorptive regime is generally observed in a high LTS environment, given by a higher mean value and the distribution of LTS for the weakly absorptive regime is more negatively skewed than for the strongly absorptive regime. 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). Furthermore, Figure (b) shows LTS values sorted by LWP for two regimes and attempts to rule out the LWP dependence on LTS. For each LWP bin, the weakly absorptive regime has a higher LTS value than the strongly absorptive regime. Such results indicate that even under similar available moisture conditions, 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.

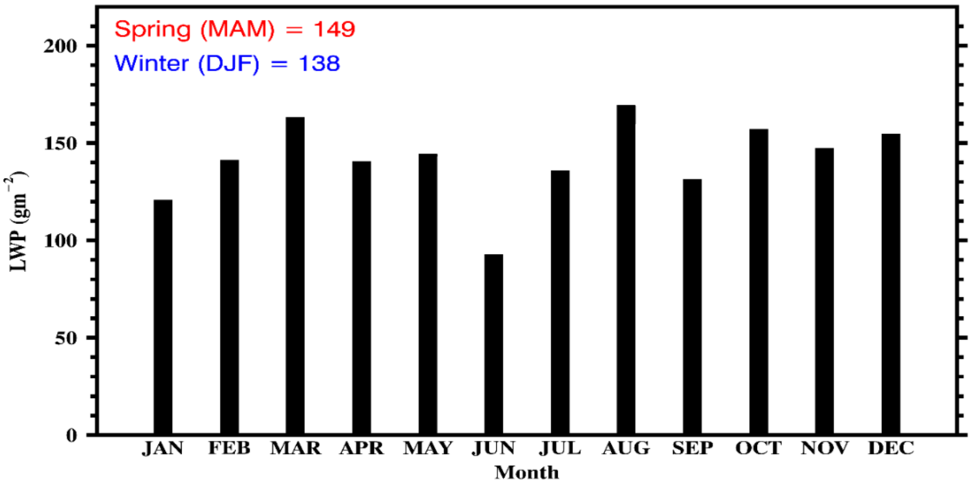
However, the LTS emphasizes a general thermodynamic condition in the lower troposphere with a wider domain as compared to the single-point measurement. The influence of cloud dynamics, presumably cloud-base updraft, is not negligible, since the sensitivity of cloud droplet to aerosol loading is enhanced 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 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 Nd/Nccn between the two regimes might be affected by the combined effects of LTS, updraft velocity, and aerosol absorption effect on the cloud environment.

Accordingly, the discussion above has been added to the last paragraph of revised section 3.3.4 in the revised manuscript.

Related is the fact that most cases occurred only during winter and spring and largely under northerly wind conditions. Also, the authors state that the high and low absorption cases split largely along the same lines with higher absorption occurring in spring, however the implications of the co-variability in aerosol and cloud properties is never discussed. You note that the LWP is larger under the high aerosol absorption regime – is this causal? A seasonal effect? You also note that higher absorption occurs in Spring. This fact is not revisited and explained in the discussion after all relationships have been analyzed. These two factors could be unrelated but both driven by seasonal effects on aerosol distributions and available moisture separately. What implications would this have for the relationships you present here?



Thanks for the comments and suggestions.
The figure above shows the seasonal variation of LWP for single-layered low clouds during the period 2007-2012. Note that the mean value of LWP in Spring (149 gm^{-2}) is slightly higher than that in Winter (138 gm^{-2}). Similar results (LWP=160 vs. 141.1 gm^{-2}) are found in Dong et al. (2005) who used the same dataset but for different period (1997-2002). In Spring, owing to the upper-level ridge centered over the western Atlantic, the SGP is located at the northwest edge of the Sub-tropical High. Therefore, the SGP during the spring months is under the influence of relatively frequent southerly transport, which is characterized by strongly absorbing carbonaceous aerosols produced from biomass burning from Central America, as well as the moisture transported from the Gulf of Mexico. While during Winter, the SGP site experiences airmasses from higher latitudes with less intrusion of airmasses from the south (Andrews et al., 2011; Parworth et al., 2015; Logan et al., 2018).

The seasonal differences in aerosol distributions and available moisture between the two absorptive regimes are largely due to the different airmass transport pathways induced by the seasonal synoptic patterns, and no clear causality is found between springtime higher LWP and absorbing aerosols. In addition, the analyses of aerosol-cloud interaction in the manuscript are performed by stratified LWP, which eliminates the effect of different LWPs on the aerosol and cloud properties.

Accordingly, the following discussion has been added to the last paragraph of section 3.3.1 in the revised manuscript:
‘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)’.

And the following statement has been added to section 3.3.2 in the revised manuscript:

‘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.’

In Fig 8, why look at drop effective radius as a function of Na if you have CCN measurements and have already established the Na to CCN relationship dependence on absorption? I feel like the effect of size and composition on drop activation are getting conflated here and in some other places in the manuscript. Given that the absorption dependence of Na to CCN is compelling, you might to better to simplify the paper by omitting some of these plots that don’t add to the message and can actually be confusing. On P16, the last paragraph of section 3.3.6 has a related discussion that is confusing. The relationship of Na to clouds and CCN to clouds is considered. This doesn’t make sense – the definition of CCN is the segment of the total aerosol population that will activate to form cloud drops (the statement ‘clouds are more sensitive to CCN than solely aerosol particles’ should be deleted.) In reality the number of cloud drops might not equal CCN, but if the measurements are good then that is due to some competing effect of cloud dynamics, available moisture, radiative effects, etc. (none of the latter are addressed here.) Other statements in the paper that follow this confusing logic are P18 L4-5 ‘. . .conversion rates of Nd/Nccn for weakly absorbing aerosols are higher than for strongly absorbing aerosols’ suggests that there is some other mechanism at play like a radiative effect – or the CCN measurement is not accurate. Also P18 L13-14 ‘..the mechanism from CCN to cloud droplet is more straightforward than from aerosol particle to cloud droplet.’ It ought to be.

Thanks for the comments and suggestions.
We totally agree that the current discussion about the relationship of CCN to cloud droplets conveys confusing messages. Therefore, we have deleted the related discussions in the revised manuscript.

More specifically, the last paragraph of section 3.3.6 in the revised manuscript has been modified to:
‘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, 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.’

Misc: The discussion at the top of P10 regarding results from other studies that calculated ACI should include the parameters and sampling used in those studies to provide some background on why values might differ. Some discussion of the results rather than a simple reporting of the numbers should be included. How the data is sorted and what dependencies are examined can have a large impact on this indexes due to the inherent sensitivity of cloud microphysics to a range of parameters.

Thanks for the comment and suggestion. The discussion about previous studies has been confined to the studies were carried out with respect to the low-level stratiform clouds over the SGP site only. The differences in the sampling of aerosol and cloud properties, as well as the conditional dependences of ACI_r, examined in every study were included in the revised discussion, to better understand the influence of different factors on the assessment of $ACI_{r.}$

Accordingly, this discussion in the last paragraph of section 3.2 in the revised manuscript has been changed to:
'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 Fig 7 are the differences in the ratios statistically significant? Where standard deviations are included it's easier to judge what might be significant, but there is a general lack of discussion of uncertainties and statistical significant throughout. This is further complicated by the lack of information on the number of data points used in each analysis (or each bin in the binned analyses) as commented above.

Thanks for the comments, a student's t-test was performed to test the ratio difference in every LWP bin at the 95% significance level. The standard deviations of the ratios were plotted as the dashed line in the revised Fig 7. For clarification, the sentence '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' has been added to the first paragraph of section 3.3.4 in the revised manuscript.

Figures: All of the labels need to be much bigger – many are very difficult to read. Figure 2 caption has the sub-figured listed out of order – should be ordered alphabetically from a-f Figure 3 red and orange colors are indistinguishable Figure 6 not points above the 1:1 line is curious – this almost always exists due to measurement error – were these removed?

Thanks for the comments. The labels of the revised figures have been enlarged for better viewing. The caption of Figure 2 has been corrected following the alphabetical order. The orange color (corresponding to date 20120204) in Figure 4 has been changed to a cyan color. And yes, we considered that the sample points with higher N_a value than N_{CCN} value were a result of instrument error of CPC or CCN counter, thus we removed those points for better data quality.

Specific: Page and Line P2 L23: ‘influence’ rather that ‘interact with’ (suggestion)

Thanks for the suggestion, the sentence has been changed to ‘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’ in the revised manuscript.

P2 L28: ‘inferred’ rather than ‘identified by’ (suggestion) – your explanation of the uses and limitations of inferring composition from optical properties is quite nice

Thanks for the suggestion, the sentence has been changed to ‘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’ in the revised manuscript.

P3 L6-9: may also not that measurements of absorption angstrom exponent typically carry large uncertainties

Thanks for the suggestion, the sentence has been changed to ‘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 measurement of this parameter typically carry large uncertainty, and has limited value when there are mixtures of different aerosol species that share similar spectral dependences’ in the revised manuscript.

P7 L25-27: and the restriction of LWP > 20 g m-3

Thanks for the suggestion, the sentence has been changed to ‘…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 $20\ \mathrm{gm}^{-2} < \mathrm{LWP} < 300\ \mathrm{gm}^{-2}$ and the coupled boundary layer conditions’ in the revised manuscript.

P7 L27: what is the reasoning behind the daytime only? Simply that the quantity is only available under sunlight conditions? Consider rewording

Thanks for the comments, the sentence has been changed to ‘Only daytime cloudy periods were considered in this study because the r_e retrieval required the information of solar transmission (Dong et al., 1998)’ in the revised manuscript.

P8 L17: ‘find’ should be ‘fine’

Thanks for pointing out, the correction has been made in the revised manuscript.

P8 L22-24: this sentence is confusing – maybe ‘. . .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.’

Thanks for the suggestion, the sentence has been changed accordingly in the revised manuscript.

P8 L25: ‘dominated’ should be ‘dominant’

Thanks for pointing out, the correction has been made in the revised manuscript.

P9 L16: note that ‘theoretical’ values of ACIr. . .

Thanks for the suggestion, the sentence has been changed to ‘Note that values of ACIr have theoretical boundaries of 0-0.33…’in the revised manuscript.

P11 L9: don’t think you can state that co-albedo provides information about composition, just more sensitive to the amount of absorption

Thanks for the comment, the sentence has been changed to ‘This parameter is more sensitive to the capabilities of aerosol light absorption (rather than scattering) in total aerosol light extinction and therefore can better infer the aerosol composition’ in the revised manuscript.

P11 L22-25: sentence needs rewriting – may just need a ‘For’ at the start and to remove ‘higher’ at the end

Thanks for the suggestion, the sentence has been changed to ‘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’ in the revised manuscript.

P15 L19-22: how much does the composition of CCN matter for growth once it’s already activated?

Thanks for the comments. We found that this statement cannot be fully supported by the current analysis. Therefore, the last part of section 3.3.5 has been modified to ‘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’ in the revised manuscript.

P15 L25: should be Fig 8

Thanks for pointing out, the correction has been made in the revised manuscript.

References

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