## **Response to Reviewer #1**

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 responses and changes in aerosol data:

Reviewer #2 suggested that the original use of submicron aerosol optical properties data could cause the inconsistency in aerosol number concentration and optical properties. Therefore, aerosol optical properties have been changed to the measurements of sub-10 $\mu$ m size-cut in the revised manuscript and the data was re-sorted by the (1-SSA) values of sub-10 $\mu$ m aerosols into high and low absorptive regime 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<sup>-2</sup>, the ACI<sub>r</sub> values are 0.26 and 0.21 for the weakly and the strongly absorptive regimes, respectively. For the LWP bin of 200-250 gm<sup>-2</sup>, the ACI<sub>r</sub> values are 0.13 and 0.12 for the weakly and the strongly absorptive regimes, respectively. The differences in ACI<sub>r</sub> between the two regimes and the damping of ACI<sub>r</sub> 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.



#### **General Comments:**

1. The uncertainties and hypotheses are mentioned in the article but a discussion about them is needed. For the hypotheses, it is referred that  $\sigma x$  is assumed constant at 0.38 (page 5 line 23): Is there a way to estimate the impact from a variation of this value according to a sensible range? The same comment goes for the supersaturation fixed at 0.2% (page 6 line 21), what are the impacts on the results if SS =

1.15%. The parameters are associated with uncertainties, as mentioned several times in the text: LWP (page 5 line 9), re and Nd (page 5 line 25), SW fluxes (page 7 line 17), the contamination by insect (page 4 line 27). Unfortunately, these uncertainties are not considered in the study when comparing the ACI parameters for the different regimes. What are the impacts of the uncertainties on the results? Can you estimate the impacts on ACI to ensure that the observed difference is real?

## Thanks for the comments and suggestions.

Dong et al. (1997) did a sensitivity study of retrieved Nd from different inputs, such as LWP, solar flux and  $\sigma_x$ . With a change of  $\sigma_x \pm 0.15$ , Nd values vary 15.7% and 30.4%, respectively. Considering the Nd retrieval depends on multiple variables, only counting the uncertainty of one parameter  $\sigma_x$  in retrieving N<sub>d</sub> cannot be representative to the actual N<sub>d</sub> uncertainty. Therefore, considering an average uncertainty of N<sub>d</sub> as 25% from the comparison with in-situ measurement (Dong et al., 1998) should be more appropriate. In that case the activation range for the weakly absorptive aerosol regime is 52% ~ 86%, while for the strongly absorptive aerosol regime is 41% ~ 67%. Therefore, based on the discussion above we decided to remove the sentence of 'Moreover, it is noteworthy that the uncertainty in deriving the CCN activation rate...'.

To test the uncertainty of CCN activation rate under different supersaturation levels, the CCN number concentration was interpolated for the 1.15% SS level using the same method described in the manuscript. As a result, with a range of SS values from 0.2% to 1.15%, the ratios of Nd/N<sub>CCN</sub> for weakly absorptive regime range from 0.54 to 0.38, while for strongly absorptive regime range from 0.45 to 0.25. In a given supersaturation level, the differences in CCN activation rate between two regimes do exist. In the continental boundary layer stratus, it is very hard to reach a supersaturation level of 1.15%. Therefore, we chose the SS level of 0.2% in this study to represent the most typical condition for this kind of cloud.

Based on the sensitivity study, the 10% changes 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<sub>r</sub>, 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<sub>r</sub>) is almost negligible, which indicates that the impact of  $r_e$  uncertainty on ACI<sub>r</sub> is minor and the observed differences do exist. Accordingly, the discussion above has been added to the second paragraph of section 3.3.6 in the revised manuscript.

2. Two "meteorological" parameters have been considered (section 2.3): the liquid water potential temperature and the total water mixing ratio. These parameters have been used to consider if the boundary layer is well-mixed or not. I think a deeper study on the impact of meteorological parameters on ACI would increase the impact of the paper. As shown in Table 1, the cases correspond to different seasons and airmass sources. The different ACI observed for absorptive or non-absorptive aerosols might be due to different meteorological parameters (as stated in page 11 line 16) and potentially not to the difference of aerosol optical properties. I think different regimes based on meteorology parameters (e.g., stability) should be considered to strengthen the results.

## Thanks for the comments and suggestions.

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 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, and thus enhance the sensitivity of cloud droplets to aerosol loading.

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 the solar radiation and heat the incloud 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 sensitivity of cloud droplets to strongly light-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 condition.

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

3. In the study, I do not understand if each day is taken separately to perform the analysis or if the study considers each measurements: For example, in Figure 4, we observe that some days have a large range of AE (2011/05/13), did the distribution of AE shown in Figure 2-b consider each point from Figure 4 or the average for each day (total of 16 points)? I assume that it is each point but the text needs to make it clear. Therefore, I do not understand why the cloud lifetime needs to be more than 3 hours (page 7 line 26) if each measurement is considered independently

# Thanks for the comments.

The analysis was performed considering each 5-min temporal resolution data point so that the AE distribution in Figure 2b includes every point from Figure 4. For clarification, a sentence '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.' was added to the first paragraph of section 3.1 in the revised manuscript.

The  $r_e$  retrieval involves the solar transmission (Dong et al., 1997 and 1998) so that an overcasting cloud condition is required to avoid the impact of broken clouds with leakage of direct solar radiation on the transmission calculation, which is reflected in the point-based cloud radar observation as a long-lasting continuous cloud layer. Therefore, the criterion of 3-hour is a good balance between the number of cloud cases and the feasibility and stability of the retrieval.

Specific Comment:

There is no indication of how many data points are considered in the analysis.

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.

Abstract: A sentence about the context, and why it is important to study the aerosol cloud interaction is missing.

Thanks for the comments, a sentence of 'Aerosol indirect effect on cloud microphysical and radiative properties is one of the largest uncertainties in climate simulations. In order to investigate the aerosolcloud interactions, a total of 16 low-level stratus cloud cases under daytime coupled boundary layer conditions are selected.', and a sentence of 'The impact of the aerosols with different light-absorbing abilities on the sensitivity of cloud microphysical responses is also investigated' have been added to the revised abstract.

page 3 line 10: I suggest to remove the "co-albedo" has it can confuse a reader which is not familiar with this term, or to specify that it is 1-SSA.

Thanks for the suggestion.

The sentence has been changed to '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' in the third paragraph of introduction in the revised manuscript.

Page 4 line 20: The study uses two different instruments with different spatial and temporal resolutions, how does it affect your results? Is the uncertainty from the KAZR lower?

Thanks for the comment.

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, thus it would not affect the results.

page 5 line 4: Why has the "cloud-top height lower than 3 km" limit been chosen?

This limit is chosen following the definition of single-layered low cloud in Dong et al. (2006), which characterized by clouds that have cloud top height less than 3km with no clouds above them. This definition is also consistent to the ISCCP defined low clouds (> 680 mb).

page 6 line 14: What is the initial temporal resolution?

The initial temporal resolution is 1 minute and then the data were averaged into 5 minutes to match other variables. For clarification, the sentence has been changed to 'In this study, the sub-10  $\mu$ m aerosol optical properties with original 1-min temporal resolution were averaged into 5-min bins to match the cloud microphysical properties' in the first paragraph of section 2.2 in the revised manuscript.

Is there a study comparing the measurements from SGP with in-situ data to evaluate the performance of the instruments? The results are provided for cloud microphysical properties (page 5 line 25), but is there something similar for the aerosol properties, cloud boundaries, and boundary layer conditions?

Yes, a study was conducted by Delle Monache et al., (2004) used in-situ aerosol measurements from 59 flights during March 2000 – March 2001 to compare with the surface aerosol measurements. Their results showed that the aerosol extensive properties measured within the boundary layer were well-correlated with surface measurements. Thus, under the well-mixed cloud-topped boundary layer condition, the surface measurements of aerosol properties are well representative of the boundary layer aerosols which actually influence the cloud microphysical properties.

page 7 line 6: 0.5 K and 0.5 g/kg: are these thresholds the same as in Dong et al. (2015)?

Yes, these thresholds are the same as in Dong et al., 2015, originally suggested by Jones et al. (2011).

page 8 line 24: Can you describe the difference between FMF and AE? I am not sure to understand why the study needs the two parameters.

The AE focuses on the relative difference of scattering abilities in a specific aerosol group (that belong to the size category of < 1µm or < 10µm) at two different wavelengths, which reveal the relative wavelength dependence of particle optical properties due to differences in particle sizes. But it can intrinsically carry uncertainty if the mixtures of different size aerosols share similar spectral dependences. While the FMF focuses on one single wavelength and describes the aerosol scattering ability at this wavelength, given by the ratio of the fine-mode (diameter < 1µm) aerosol scattering coefficient to the total (diameter <10µm) aerosol scattering coefficient ( $\sigma_{sp1}/\sigma_{sp10}$ ), which pertains to the relative contribution of fine-mode aerosol scattering in total scattering. The use of the FMF parameter along with AE can give a robust illustration of the fine-mode aerosol dominance in the selected cloud cases.

page 10 line 5: How is the uncertainty on ACI retrieved? Is it the 95% confidence interval of the fit?

## Yes, the uncertainty of ACI is retrieved from the 95% confidence interval of the fit

page 10 line 6-: The authors are comparing ACI values with previous studies. I am a bit skeptical about it: There is plenty of studies retrieving ACI values with different methods, datasets, geographical locations. ACI parameter depends on that. The authors only report ACI values which range with their study without a discussion on the differences. I think there is two different possibilities: Either you consider all the studies retrieving ACI and discuss about the potential differences or the comparison is limited to studies looking at the same region and/or same data.

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<sub>r</sub> 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<sub>e</sub>. The study conducted by Feingold et al. (2006) during an intensive operation period in May 2003 showed that the assessment of ACI<sub>r</sub> can be affected by the usage of different aerosol proxies and boundary layer conditions. Using surface measured N<sub>a</sub> to represent aerosol loading yielded unrealistic values of ACI<sub>r</sub> even after sorted by LWP, presumably owing to decoupled boundary layer conditions. However, if the surface aerosol scattering coefficient ( $\sigma_{sp}$ ) and aerosol extinction at an altitude of 350 m are used as CCN proxies, then similar ACI<sub>r</sub> values can be obtained with a range of 0.14-0.39. Under coupled conditions, the N<sub>a</sub> and  $\sigma_{sp}$  could serve as reliable CCN proxies. The  $\sigma_{sp}$  of accumulation-mode aerosols was used in Kim et al. (2008) to show that the ACI<sub>r</sub> 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.'

page 11 lines 23-26: I do not understand the sentence, can you rephrase it?

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<sup>-3</sup>) is larger than that from the strongly absorptive regime (384 cm<sup>-3</sup>), and the occurrence of high  $N_{CCN}$  values (larger than 1000 cm<sup>-3</sup>) is also higher in the weakly absorptive regime' in the first paragraph of section 3.3.2 in the revised manuscript.

page 11 line 26-27: Can it be quantified?

Thanks for the comment.

Unfortunately, since the value of AE is retrieved via a logarithmic slope, the AE is emphasizing the relative dominance of fine-mode or coarse-mode aerosols within an aerosol plume rather than the absolute amount of existence. Generally, the AE > 1 indicates the particle size distributions dominated by fine mode aerosols (submicron), and AE < 1 denotes the dominance of coarse mode aerosols. Thus, the dominance of fine-mode aerosol is hard be quantified based on the value of AE.

page 12 line16: The "majority", can it be quantified?

Thanks for the comment. The sentence has been changed to 'For a broad range of  $N_a$ , especially 200-700 cm<sup>-3</sup> and 1200-3500 cm<sup>-3</sup>, the majority (~74%) of sample points from the strongly absorbing regime are located below the samples from the weakly absorbing regime' in the first paragraph of section 3.3.3 in the revised manuscript.

Figure 2: Can the standard deviation be displayed with the mean? Also, considering that the distributions are not Gaussian, why did you consider the mean rather the median?

Thanks for the comments. We totally agree that the median value can better represent a non-normal distribution. The mean values were originally used considering some variables in Figure 2 were normally distributed. Therefore, in the revised Figure 2, the standard deviations are now displayed with the mean, and the median values of the variables are also displayed to better represent the data distributions.

Figure 7: Is there a reason why the standard deviation is not included for the ratios NCCN to Na and Nd to NCCN?

The standard deviations of the two ratios were originally not included because of the consideration of a better viewing of the figure.

The standard deviations of the ratios are now displayed as the dashed line in revised Figure 7.

Technical corrections:

page 5 line 18 "100" should not be here.

Thanks for the comment, the "100" here denote the value of LWP in a unit of  $gm^{-3}$  should be multiplied by 100 in the re retrieval algorithm (Dong et al., 1998).

page 8 line 17: find  $\rightarrow$  fine

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

page 15 line 25: Fig.  $10 \rightarrow$  Fig. 8

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

Figure2 caption: the order to describe the figures is: a-d-b-c-. . . instead of a-b-c-d-. . .

Thanks for pointing out, the order has been changed alphabetically in the revised Figure 2 caption.

Figure 4: The definitions of the dotted lines are missing in the caption?

Thanks for pointing out, a description 'Horizontal dotted line denotes the demarcation of  $AE_{450-700nm} = 1$ ; Vertical dotted line denote the demarcation of  $\omega_{abs450} = 0.07$ .' has been added to the revised Figure 4 caption.

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