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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Investigation of Aerosol-Cloud Interactions under Different Absorptive Aerosol Regimes using ARM SGP Ground-Based Measurements" *by* Xiaojian Zheng et al.

Anonymous Referee #2

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Overview: This study uses data from 16 low stratus cloud cases at the ARM SGP site in Oklahoma to determine the effect of aerosol absorption on commonly used aerosol-cloud interaction index, specifically the change in cloud drop effective radius with a change in aerosol (ACIr.) This approach is of interest as aerosol composition may impact cloud drop activation and ultimately aerosol indirect effects. The authors follow methods from existing literature very closely with the novel piece of adding information on aerosol absorption. It is understood that aerosol compositions that are more highly absorbing can be lass hygroscopic, thus affecting aerosol indirect effect so overall this

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Discussion paper

study is of interest. The data do indicate that aerosols that are less absorbing are more likely to be suitable as CCN that aerosols that are more absorbing. There are significant shortcomings however in the relationship from CCN to cloud properties as meteorological parameters that may be mediating cloud properties and may or may not be related to the aerosol absorption are not addressed. These topics as outlined below should at least be discussed and the potential implications included in the manuscript (if a further analysis of meteorology/radiative effects is not included) prior to publication. I've categorized these as minor revisions but could take some doing.

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.

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

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but why complicate the issue?

Lack of meteorological parameters or aerosol radiative effects in assessing co-variances 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.

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?

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

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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.

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.

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.

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?

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

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

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

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

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

P8 L17: ‘find’ should be ‘fine’

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.’

P8 L25: ‘dominated’ should be ‘dominant’

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

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

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

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

P15 L25: should be Fig 8

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