Interactive comment on “Is Positive Correlation between Cloud Droplet Effective Radius and Aerosol Index over Land Due to Retrieval Artifacts or Real Physical Processes?” by Hailing Jia et al.

Anonymous Referee #1

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General comments:

The observed aerosol-cloud relationship from space remains controversial due to the wide range of influential factors, including the artifact retrieval of aerosol and cloud properties (large biases), and the confounding meteorological variables that simultaneously govern the aerosol and cloud systems. The manuscript is an useful attempt to address the controversial phenomenon frequently observed over high-polluted land—positive correlation between cloud droplet effective radius (CER) and aerosol loading. This study proposal new physical explanation (i.e., positive feedback caused by increased CER that in turn initialize the collision-coalescence processes) for this positive
correlation from satellite observations. This manuscript is logically organized, the analysis methods are technically sound but not novel, and the results are very interesting albeit some points not adequately illustrated. I have some comments on interpretation of the major results. Arguably, this topic is worth of further investigation. As such, I recommend its publication pending the following concerns satisfactorily addressed.

Major comments:

1. L29-38: The descriptions of aerosol climate effect (direct, indirect, ACI effects e.t.c.) are duplicated in the 1st and 2nd paragraph. Thus, the authors can consider to combine them into one paragraph. 2. L50-69: these are about why you choose the proxy of aerosol index for CCN, and CER in the present study, which could be moved to section 2 and replaced with literature reviews of the role of vertical observations in the cloud-aerosol-precipitation interaction studies, which are omitted, including the measurements provided by CALIOP (Costantino and Bréon, ACP 2013; Zuidema, et al. BAMS 2016), Cloudsat (Christensen et al. JGR, 2016; Chen et al., JAS 2016; Peng J. et al., JAS), and TRMM (Wall et. al JAS 2014; Li et al., Rev Geophys, 2016; Guo et al., ACP 2018). Besides, the preference for use of aerosol index rather than AOD should be clarified in a more straightforward way, since there exist large uncertainties in the retrieval of Ångström exponent over land.

3. L75-81: The fourth factor can be added here impairing the quantification of aerosol-cloud interaction from observations: the vertical overlapping status of aerosol and cloud layers (e.g., Costantino and Bréon, ACP 2013, doi:10.5194/acp-13-69-2013; Huang et al. JGR 2015, doi:10.1002/2014JD022898)

4. Section 2.2: The LTS is a proxy for the magnitude of the inversion strength in the lower troposphere. The readers are curious for the meaning with regard to the various LTS values. Large LTS means unstable conditions? Please clarify it. 5. L200-202: Any references to support the argument “Under cloudy sky, the response of ... have larger retrieval biases”? 6. In Fig. 4a, the samples of positive TOA albedo difference
is almost equal to the samples with negative albedo difference. Besides, Fig. 4b-c has a large fraction of positive albedo difference (more than 20%). How the authors claim that “implying that as AI increases, the reflected solar shortwave radiation at TOA will reduce over land while increase over ocean.” More importantly, necessary discussion is warranted for the difference of TOA albedo response to aerosol between over land and ocean. 7. Extensive previous studies have pointed to the saturation effect as the aerosol keeps rising. e.g, Breon et al. (Science 2002) argued that as the AI is greater than 0.15, the CER will keep constant. From Fig. 5 in this manuscript, most of the AI values in three regions over land are greater than 0.15. I wonder whether there exists such saturation effect when the samples are divided into two parts taking the threshold of AI=0.15? Or at the very least, the authors make sure to clarify the number of samples (the ratio) with the AI values less than 0.15 over each region of interest in this investigation. Also, the potential inference induced by saturation effect should be taken into account in the future submission of revision. 8. In section 3.3.2.2: Can “the degree of entrainment mixing” be represented by LTS? LTS is an indicator of temperature inversion proposed for stratocumulus over ocean. Further justification is needed. 9. Figure 9: why not show the The slopes of CER versus AI for the adjacent ocean areas? The readers are curious to know the difference of this slope between over land and ocean.

Minor comments: 1. L50: cloud droplet number -> cloud droplet number concentration, and make sure to correct for all instances in the manuscript. 2. L310-317: It is well known that much lower LTS (more unstable) and lower RH CT over land compared with over ocean, which is not probably concerned about any aerosol effect, but about the effect induced by the difference of underlying surface properties. Actually, even over ocean, positive correlation between CER and AI is observed over south Atlantic (e.g., in Fig. 2 of Nakajima et al. JGR 2001), and land (e.g., West US, West Africa in Fig. 2 of Breon et al. Science 2002), we can see negative slope between CER versus AI. It suggests that the relationship varies greatly by regions, and is still extremely challenging to be interpreted as a causal connection. I suggest the authors rephrase
the section more accurately. 3. What is the actual number of samples for each bin in Figures 4-7? Clarification in the figure caption will help the readers to better follow. 4. What does it mean for the labelled number in different color in each panel in Fig. 5? The slope? It is better to clarify in figure caption.

Interactive comment on “Is Positive Correlation between Cloud Droplet Effective Radius and Aerosol Index over Land Due to Retrieval Artifacts or Real Physical Processes?” by Hailing Jia et al.

Anonymous Referee #2

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Before getting to the review of the manuscript, I want to say... why do people accept papers to review and then not review them (and don’t let the editor know this quickly)? It seems like this happens way too often, as it did here.

Ok, now to the manuscript...

This manuscript explores the differences between land and ocean regions in the correlation of near-cloud aerosol index (Al) to cloud effective radius (CER) and liquid water path (LWP). The paper looks various potential reasons (real and artificial) for the positive correlations between Al and CER are over land, as opposed to the more-expected negative correlation over oceans. While no precise reasons are found, several hypothe-
ses were able to be eliminated. I feel that the paper may be useful, though I have one major concern and several minor comments that I feel need to be addressed before publication.

Major comment:

The Angstrom exponent, used for deriving the aerosol index, is notoriously bad over land for MODIS Dark Target land retrieval (not sure about Deep Blue retrievals, and I'm unsure about which retrieval products were used here, though I believe that the 3 land regions used in this paper will mostly be Dark Target rather than Deep Blue). In the Levy et al. 2013 paper describing collection 6: “On a global basis, we and others have found little quantitative skill in MODIS-retrieved aerosol size parameters over land (e.g., Levy et al., 2010; Mielonen et al., 2011). We have decided to discontinue further attempts at validating Ångström Exponent (AE) and fine-AOD. A user can still choose to derive AE (from spectral AOD) or fine-AOD (from product of $\tau \eta$) and evaluate the results themselves.” In Levy et al. 2010, they show that the R^2 of the Angstrom exponent over land is only 0.3 with AERONET.

Levy et al. 2013: https://www.atmos-meas-tech.net/6/2989/2013/amt-6-2989-2013.pdf
Levy et al. 2010: https://www.atmos-chem-phys.net/10/10399/2010/acp-10-10399-2010.pdf

Related, for research on estimating surface-level PM2.5 from AOD, it would be useful to use the Angstrom exponent or fine-mode AOD products to help filter coarse PM from the PM2.5 estimate. However, due to the poor ability for MODIS to retrieve these products over land, the AOD-PM2.5 community uses just AOD as using the Angstrom exponent or fine-mode products adds noise to evaluations. It is unclear why the aerosol-cloud interactions community should have more confidence using the MODIS Angstrom exponent over land.

It seems that the aerosol index over land used here may be very poor (due to the Angstrom exponent uncertainty), and this could be contributing to counterintuitive re-
results over land. There is no mention of this in the paper. I feel that the authors should evaluate their aerosol index data in their 3 regions against AERONET to show if their aerosol index product has any skill in their land regions, and discussion needs to be added.

Minor comments

L29: “Earth” should be “Earth’s”

L31-33: It is more precise to say that ACIs are the largest forcing uncertainty to present climate change. I’m nearly certain that the uncertainty in what humans will do (e.g. in terms of how much CO2 we will emit and other changes) is the biggest uncertainty in the anthropogenic contribution to *future* climate change.

L50-51: Kohler theory only describes the relationship between a particle and its critical supersaturation for activation. Cloud droplet number at the cloud base depends on the number distribution of critical supersaturations and the updraft velocity. The second part of the sentence discusses these effects, but I feel that this sentence can be rewritten to be more precise.

L64: “Loosely related”. Is it possible to be more quantitative here (e.g. using numbers from the Stier reference)?

L68: Same, can you be quantitative?

L125: Which MODIS AOD retrieval algorithms are you using (dark target, deep blue, MAIAC)?

L268: “Become*s*”

L269: Either “*the* collision-coalescence process” or “collision-coalescence process*es*”.

Figure 7 and associated discussion: It is unsurprising that the slope of CER-AI is stronger when including all data vs. when binning to >14um and <14um. If the slope
is positive, more observations will move from the <14um to >14um with increasing AI. With increasing AI, a CER of 13.9 um (which would be at the large end of the <14 um bin) would shift to 14.1 um (which would be on the low end of the >14 um bin), so increasing AI removes some of the higher <14um cases and creates new lower >14 um cases... the all prevents the CER for the two size bins from change much with AI. Hence, the slopes for the binned sizes are buffered from changes with AI. A similar phenomena will occur of a negative slope. Thus, the discussion in lines 274-278 seems unnecessary.