Referee's Comment on "Characterizing observed aerosols and their impacts on shallow cumulus clouds during the TRACER field campaign"

Title	Characterizing observed aerosols and their impacts on shallow cumulus clouds during the TRACER field campaign
Author(s)	Azusa Takeishi et al.
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General Comments

This manuscript describes a study investigating aerosol-cloud interactions (ACI) in shallow cumulus and our ability to meaningfully represent them in LES. The authors conducted a "clustering analysis" to identify 4 distinct aerosol populations which were present during the TRACER field campaign, and then based on these populations they ran LES of a single shallow cumulus scene under various canonical aerosol conditions. Additionally, the study compared simulations of the control-case for aerosol conditions between LES using a bulk microphysics scheme and LES using the superdroplet method (SDM).

The study is clearly a substantial contribution to the atmospheric physics community, contributing to our understanding of how to characterise aerosols (in this particular case over Greater Houston), and how to interpret, through physical mechanisms, the results of ACI studies (here in the case of shallow cumulus). Their results are especially important for showing that the use of bulk schemes for studying ACI may be confounded by the unrealistic formation of precipitation as compared to SDM, which is somewhat corroborated by observations.

I found this manuscript quite exceptional both in terms of its scientific quality and presentation. The manuscript is well written and its arguments are compelling. As a result of thoroughly enjoying reading the manuscript I may have been a bit over-enthusiastic with my comments. This should be viewed as a sign of the manuscript's high quality, both in terms of readability and scientific content, which made it possible for me to engage with. Many of my comments could be viewed as suggestions / minor-revisions; the only points where I suggest substantial revision is in the description of the clustering method and the explanation of the clusters' relation to sea breezes, as well as in refining one of the key conclusions of the paper regarding the improvements required of bulk schemes. Notwithstanding all of my comments (somewhat over-eagerly given!), I recommend this paper for publication in ACP journal.

I look forward to the response of the authors to my comments, and also their forthcoming work on aerosol recirculation in developing cumulus.

Specific Comments

Major comments on the clustering method and its results:

- **Section 2.1:** The description of the aerosol clustering would greatly benefit from a short description and schematic of the Atwood et al. (2017, 2019) methodology. I get the impression that the current state of Section 2.1 is useful for readers already familiar with this analysis method and may be useful for researchers who wish to follow-up on the work presented here, however, it is extremely difficult for readers with no previous knowledge of this method to understand, even on a surface level. This also makes it hard as a reviewer to comment on what errors/uncertainties need to be considered when deeming the identification of 4 clusters as appropriate (see below). In order to cater to a wider audience than just those familiar with Atwood et al. (2017, 2019), it is necessary to add an overview/description of the methodology in this section, or in a short appendix.

- **L130-134:** As eluded to above, I cannot grasp what errors or uncertainties may be introduced by the remapping of the observational data to align more closely with Atwood et al. (2019). Superficially the coarsening of the data appears alarming and I question how the clusters would differ with different choices in this methodology, i.e. how different choices would translate into error bars/shading in Figure 2, and especially

Figure 2d (hence also Figure 5). Some tests of the clustering results to different choices in the clustering methodology would alleviate my concern that the identified clusters are not robust, or else an explanation of why this concern is not valid.

- **L131:** Why do the droplet size bins range only until 478.3 nm when, if I understand correctly, the original data ranges until 1 micron? Is there an appreciable mass (even if not number) of aerosol in higher droplet sizes and, if so, how would the results of this study differ if that had been accounted for in the simulations (i.e. in Figure 5), especially regarding precipitation? It would therefore be beneficial to have an explanation for the truncation of the DSDs and a conjecture of whether or not larger aerosols would have been significant.
- **L269-277:** Based on Figure 2e, I am not fully convinced by this explanation, please be more precise / clarify what is meant in the following:
  - How does Cluster 4 fit with the sea breeze argument? I interpret Cluster 4 in Figure 2e as a night-time phenomenon but this appears to contradict with the statements that "A continental air mass (i.e., Cluster 1) is more likely to reach the AMF1 site during nighttime/morning when a land breeze strengthens" and that "sea-breeze air mass (i.e., Clusters 2 and 4) in the afternoon/evening when a sea breeze prevails.". It is also unclear to me how the Park et al., 2020 relates to the frequency of occurrence of Clusters 2 and 4. Also later, in L280 and L461, Cluster 4 is referred to as occurring in morning and evening (as opposed to night-time).
  - It's ambiguous if statements in this paragraph are referring to more/less frequent as compared to each Cluster's own average daily occurrence, or as compared between clusters, e.g. "Cluster 2 is more prevalent in the afternoon, whereas Cluster 4 is more frequently observed in the evening and morning.". From Figure 2e, Cluster 4 is almost always more frequent than all the other clusters, so the reader can assume it is meant as more/less frequent relative to each Cluster's own occurrence, but this should be clarified. And perhaps comparing occurrence between Clusters makes no sense because there are observational biases in how often different clusters are detected??

Major comments on the improvements required of bulk schemes:

- **L426-429:** I find these statements quite important conclusions of this manuscript, however I think they could be more clearly differentiated / refined. To me points (1) and (2) are enompassing two fundamental problems of bulk schemes: (A) issues with DSD representation in bulk schemes (of aerosol and of droplets), and (B) issues with microphysical process representations in bulk schemes. However the current separation in (1) and (2) conflates them, making the separation instead between the activation and growth of so-called "cloud" droplets and then the growth of so-called "rain" droplets. This division may be how bulk schemes are constructed, but it is not really how the fundamental problem arises, nor how nature behaves (of course we cannot observe "autoconversion" — it is a modelling construct!). Perhaps this point is rather academic, but please consider re-dividing the problems contained in (1) and (2) along more natural/fundamental lines, as opposed to bulk scheme ones. For example, (1) "a deficiency in the representation of droplet activation rates" includes both the parametrisation of aerosol activation (point B) as well as the issue of bulk schemes prescribing constant aerosol distributions (point A). The same comment applies to L442-448 and in similar ways also to the conclusion in L468-471.

Minor Comments:

- **L75-78:** In discussing the advantages of SDM, it would be a good idea to mention SDM's explicit/fundamental representation of (de-/re-)activation, droplet evaporation and condensation (including droplet nucleation) according to Köhler theory, as opposed to "the growth process of wet aerosols into cloud droplets". I think the current phrasing neglects droplets shrinking and de-activation and re-activation, all of which are likely to be relevant to this study.
- **L77-78:** I find "the elimination of artificial size thresholds", a bit imprecise and "size thresholds" doesn't generalise well when we talk about the cold-phase too, or more complex aerosols and/or aerosol chemistry. Just focussing on the warm-phase it may be clearer to state explicitly that it's the elimination of categories which distinguish between cloud and rain droplets. In general it's the elimination of the artificial hydrometeor categorisation, which in the warm-phase distinguishes between cloud and rain droplets. (At least I think that's what you mean — please correct me if I misunderstood what was the intended meaning behind "size thresholds"!)
- **L100:** "LES scale" is too vague, please be more specific about the scale of interest, preferably in words not numbers.

- **L169-171:** This sentence is a bit hard to read and contains many points; consider separating the definition of multiplicity into a separate sentence and clarifying what is meant by "the Lagrangian framework" — such terminology for readers not familiar with SDM may invoke parcel trajectory modelling or something even further afield. Furthermore, clarifying "the Lagrangian framework" would also be a good opportunity to emphasise how well SDM's representation of cloud microphysics (that is, hydrometeors and cloud microphysical processes,) matches our fundamental particle-based theory of cloud microphysics — in several places this point could be "read between the lines", but this may be a good opportunity to make the point directly, especially for the benefit of readers less familiar with SDM.
- **L193:** from Figure 1 is it hard to understand why the simulation was cutoff after 8 hours; consider extending Figure 1 beyond (and possibly before) the LES simulation period to show how the cumuli continue to develop that day, and please also justify why a longer simulation was not necessary.
- **L196-197:** Why is the scaling of superdroplets per grid-box done in this way? Is it to keep their multiplicities similar? Some information on how superdroplets are initialised would also be beneficial, especially whether or not there are any super-droplet sources during the simulations (I assume there are none but I am not certain).
- **L193-206:** Some plots showing these comparisons between the LES thermodynamics and the observations would be insightful, even if only in the supplement.
- **Figure 2:** I would highly recommend also including a plot of the mass size distribution, either in Figure 2 or in Figure 5. Also what is meant here by "cluster-median"? If there is a spread in the distributions of the DSDs in each cluster then it is important to represent this in these results, or at least in the supplement if it makes the plot too chaotic here.
- **L312:** "cloud-base height increases with time from 500 m to 1.5-2 km" I dispute that the cloud base height in the observations reaches 1.5km, from Figure 1d it looks more like 1.25km. If so, then it should be pointed out that the cloud-base height is actually often overestimated, i.e. it rises too fast in the LES. Otherwise please explain how the cloud-base height in the observations is actually higher than it seems from Figure 1d.
- **L376-378:** Please be more specific about "macrophysical properties", this study seems to only look at cloud fraction. I would expect that individual cloud lifetimes (within the scene) are different depending on the microphysics, and their precipitation certainly is different, both of those things I am inclined to view as "macrophysical" too.
- **L379-382 and L385-387:** These lines suggest there is something "counterintuitive" in the results for cloud mass and CDNC, possibly attributable to numerical diffusion. To me these results seem to be explainable by the fact that the bulk schemes rain out, whereas SDM does not. Have I missed something here? If so, please explain. I also do not fully understand how the supply of aerosols could play a role here because I don't see signs of significant aerosol depletion in Figure 8e, i.e. it looks to me as though SDM doesn't suffer from an aerosol supply deficiency until maybe very late in the simulation (circa. 1pm) — which may perhaps be rationalised by the fact these are very polluted simulations. Although perhaps I have misunderstood something here too. If so, please explain this too.
- **L389-390:** Please rephrase "In the SDM run, non-zero rain mass is present in the atmosphere, ...", I think this is extremely misleading. From my understanding of Figure 8, the maximum rain mass in SDM is at least 8 orders of magnitude smaller than in the bulk schemes — if SDM was a person, a bulk scheme would be about 10x the population of Texas, or the entire population of Japan — I think it's safe to say the SDM rain mass is insignificant. Please also justify/explain or remove the following statement that "These bulk properties of the clouds in the SDM run still technically follow the pattern of "more aerosols, less rain" found in the bulk simulations ..."
- **L403:** Please elaborate on "either by evaporation or conversion into raindrops". From Fig. S4 the evaporation rate in SDM appears to be considerably larger and so I do not fully understand how evaporation can play a role here, perhaps it is really rather just raindrop conversion? Or maybe plotting evaporation rate also outside the cloud would help clarify this point.
- **L410:** "lower aerosol concentrations in CM1-SDM near the cloud base (¡ 1.5 km)" really only applies to Figure 10a. Consider alternatively that it is lower aerosol concentrations in the sub-cloud layer and particularly towards the surface. It would also help to plot in Fig. 10 the cloud base of the bulk and SDM schemes (as in Figure 6d-e) to show this.

- **L442-448:** I am of the opinion there is already quite an extensive literature on some of these subjects, and so these points need to be refined. As an example, some of the works Adele Igel has co-authored (e.g. https://doi.org/10.1029/2023MS004068, https://doi.org/10.1029/2022MS003039, https://doi.org/10.1175/JAS-D-15-0383.1, https://doi.org/10.5194/acp-17-4599-2017) evaluate the assumptions of bulk and bin models, albeit perhaps not in comparison with observations. Notwithstanding this, other comparisons of bulk microphysics schemes to observations are abundant.
- **Section 3.2 or Section 4** I think a discussion is required of what this study misses by keeping the LES setup entirely the same except for changing the aerosol distributions in order to investigate ACI. Consider that Cluster 4 and Cluster 3 occur at almost diametrically opposite times of the day, and the implications this has for the radiative forcing on clouds, and potentially even the cloud-type which the aerosols play a role in. How might this change the results? In other words, the setup of this study prevents an analysis of how concomitant aerosol and large-scale forcing cycles/changes could complement or negate one-another, and this should be discussed.

More Minor Comments / Suggestions:

- **L76:** "exclusion of assumptions for the hydrometeor DSDs" may be better phrased positively, e.g. the complete freedom of the evolution of hydrometeor DSDs. A more profound/fundamental description of this advantage of SDM would perhaps instead point out that DSDs in SDM are emergent/prognostic rather than diagnostic — SDM model particles and only for analysis purposes are particles then turned into DSDs.
- **L69-81 and L82-92:** For the flow of the introduction it could make more sense to swap the order of these two paragraphs, i.e. introduce SDM after introducing the need for modelling ACI of shallow cumulus.
- **L434-439:** This could be a good place to point out that historically we have attempted to make bulk schemes "bin-like" — perhaps the tuning we have done to bulk models to make them look like bin schemes can explain some of the discrepancy here between bulk and Lagrangian simulations??
- **L92:** I encourage the authors to make this statement bolder! Consider replacing "highly desired" with "required".
- **circa. L174-175:** it may also be helpful to state which terminal velocity parametrisation was used too.
- **L266-268:** The identification of four clusters is compelling, but their current presentation seems to under-appreciate the similarities between Clusters 2 and 4 and hinder readability. If Cluster 2 was relabelled as Cluster 3 (and vice versa) it would be easier to compare and contrast the clusters which have almost-purely land-aerosol sources (Clusters 1 and new-2) and those which have sea-breeze + another contribution (Clusters new-3 and 4). I hope this does not sound too pedantic, I think this simple re-labelling could really aid the comprehension of the results, especially when it comes to following the discussion from L279-286. Also adding labels Continental, Extreme Pollution, SB+Pollution, and SB+Rural to the sub-figures or figure caption would be good.
- **L347:** "all simulations (Fig. S4)" is misleading, please rephrase as "all bulk-model simulations" or alike — evaporation rates in the SDM simulations appear up to 100 times higher in some regions.
- **L430:** I think the phrase "we cannot obtain confirmation of (1), (2), or (3) from observations" needs tempering, perhaps we cannot *directly* obtain ... from *in-field* observations?

Technical Corrections

- **L12:** "such datasets" doesn't refer to any previous dataset mentioned, please reword.
- **L53:** "the Gulf" is potentially misleading out of context because the same terminology is used in the Middle East, please refer to "the Gulf of Mexico" as is internationally recognised.
- **L77:** "stochastic drop collision-coalescence based on the kernel function" is rather unusual terminology. A suggested rephrasing may perhaps be along the lines of e.g. stochastic droplet/particle collision-coalescence for a given collision kernel.
- **L93:** something doesn't read quite right about the numbered statement "In response to the above-mentioned points of (1) needs for ...", please reconsider the wording and/or grammar?

- **L529:** In the references, the date for the citation of Atwood et al., 2017 is 2019, please correct.
- **L170:** "representsin" is missing a space; also "location" invokes a fixed (often infinitesimal) point, consider instead "volume of space".
- **L178:** remove comma in "TRACER-IOP, based on our analysis".
- **L180:** is a word missing in "and showed macrophysical features ... of shallow cumulus clouds in the region.", perhaps "typical of shallow cumulus"?
- **L194:** for consistency, place UTC time in parenthesis and local time in main text.
- **L213-224:** consider moving this paragraph into the methods Section 2.1, structurally it seems out of place here.
- **L231:** "The second most "polluted" Cluster 1 has" is missing "cluster, " after the word polluted.
- **L272:** missing space in "labels.A".
- **Figure 5:** the faint lines in Figure 5 do not reproduce well, please make darker and/or thicker.
- **Figure 6, 7 and 8:** Consider swapping the order of some of the sub-panels to aid the reader to follow the comparisons made in the text. In particular Fig. 6(f-j) would be better placed with Fig. 7(a-e) and/or Fig.8 (a-d), and Figure 7(f-j) complements well with Figure 6(a-e).
- **Supplement:** please relabel figures with "Observed 7/20" to "Total observed (bulk)" to match the main text.