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Interactive comment on "Droplet Clustering in Shallow Cumuli: The Effects of In–Cloud Location and Aerosol Number Concentration" *by* Dillon S. Dodson and Jennifer D. Small Griswold

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REPLY: We would like to start by addressing concerns raised by both of the Referees. First, we apologize for the delayed response and posting of the revised manuscript. We put in a great deal of time and effort to do a rewrite of the paper, to shift the focus from the smaller scale clustering originally discussed to larger scale mixing as a result of entrainment. Both Referees raised concerns that the PCFs examined are more closely related to entrainment mixing as compared to preferential concentration, and we have also come to that conclusion. In particular, our PCF spatial scale does not extend into the Kolmogorov range, suggesting that inertial clustering (if present at all) isn't being

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measured. Our PCF curves also mirror those presented in Good et al. (2012) and Ireland and Collins (2012), which used the PCF for the purpose of analyzing larger-scale clustering due to entrainment mixing. Lastly, we have de-normalized all PCF curves displayed throughout the manuscript. In Saw et al. (2012) the PCF curves are normalized at the range the larger scale inhomogeneous mixing is occurring (i.e., the ‘shoulder’ region of the curve). Since the ‘shoulder’ region of the curve is what we are analyzing, normalizing it to a common value becomes unreasonable.

COMMENT: General evaluation: This paper reports analysis of observations applying a statistical technique that aims at documenting cloud droplet clustering. I found this subject interesting and fitting the ACP scope. However, I am confused by the specific detail of the analysis (PCF normalization) and I feel the way analysis is performed amounts to throwing the baby out with the bathwater. Specifically, forcing the PCF to approach zero at large scale is not appropriate as there are likely large-scale heterogeneities both at the cloud core (i.e., due to different updraft across the cloud base and thus fluctuations of cloud droplet concentrations above) and at cloud edges due to turbulent mixing and filamentation. I have to admit that I started to read the paper with large expectations, and my enthusiasm went down and down as I kept reading. I admit that I stopped reading at the end of section 4.1. I do feel that the analysis is flawed because of the normalization that forces the cloud to look homogeneous at large scales. There are plenty of cloud observations showing that such an assumption is simply not valid! Thus, I recommend the paper to be rejected and then resubmitted with the discussion based on PCFs without normalization. There are numerous other problems and their sheer number (see specific comments) also suggests the need for a significant rewriting.

REPLY: We have realized through your comment that a more clear discussion/explanation of the normalization process was needed. In responses to your other comments that deal with the normalization we will give a more detailed explanation. However, because we have de-normalized all PCF functions in the new manuscript, normalization no longer applies. To clarify a few things however, we were not attempting to say that there were not large-scale heterogeneities within the cloud. We were simply accounting for said heterogeneity by adjusting all PCF curves to a common value at larger scales (as is done in Shaw et al. (2002), with a much better explanation of the normalization coming in Saw et al. 2012)).

COMMENT: 1. This comment is arguably more to the ACP technical staff than to the authors. The collection of figures at the end of the manuscript is unacceptable: figures are way to small and the only way to review their details is to go to the electronic version of the paper and zoom in. I feel the journal staff should request the authors to revise the submission before publishing the paper online to have each figure legible (e.g., one figure per page).

REPLY: We used the Latex Template that is available through the ACP website. To account for the figures, we adjusted the input parameters to make the figures bigger in the new manuscript. Hopefully this will account for the problem.

COMMENT: 2. I do not feel the introduction is appropriate. First, it touches on some issues only remotely related to the specific focus of the paper (e.g., the indirect effects). The selection of references is incomplete or simply inappropriate. Second, the review of previous studies concerning clustering misses important publications as there are studies showing relatively small clustering (in contrast to the papers cited right now). Overall, I agree that there is likely some clustering at the cloud microscale, but its magnitude is relatively small and thus difficult to extract from observations.

REPLY: We have completely rewritten the introduction by shifting the focus from the small scale inertial clustering to clustering caused by entrainment mixing. We have made an attempt to include and remove references as you recommended in the following comments. We have mentioned that not all studies have measured clustering (see page 3 lines 14-17 in the new manuscript) by citing Chaumat and Brenguier (2001). Although we do not elaborate much, readers will at least come to the realization that not all studies have measured inhomogeneities.

COMMENT: 3. The discussion in the introduction and in section 2.1 excludes the impact of gravitational acceleration on droplet clustering. This is a serious omission as I would argue that droplet sedimentation is the mean reason for a relatively small droplet clustering. I provide detailed comments below, the Grabowski and Vaillancourt (1999) comment on Shaw et al. (1998) in particular.

REPLY: The original reason we left out a discussion on gravitational acceleration is due to the fact that there is no way to account for it in our observations/data. What we mean is, gravity is a constant in the atmosphere unlike in laboratory experiments where gravity can be accounted for by enhancing or reducing it, as was done in Ireland and Collins (2012). However, you are right that the impact that gravity has is significant, and it is still important for the readers to understand the impacts that gravity has. A discussion on the effects of gravity on droplets has been added on page 5 lines 14-22 of the new manuscript.

COMMENT: 4. The way PCF is calculated (section 2.3) need to be better explained. First, I am not sure why normalization is needed. Second, it is not explained how the normalization is performed (a simple shift?). I think there might be interesting differences between cloud cores and cloud edges due to entrainment and small-scale filamentation for the latter (this is what is illustrated in Fig. 1, correct?) In other words, cloud core and cloud edge may look different at different scales. For instance, near the edges, there may be differences at large scales (say tens of centimeters and meters, see Fig. 1), but similar clustering may take place at small-scales (the latter is the focus of the manuscript, correct?). Moreover, normalizing PCF does not allow estimating the magnitude of small-scale concentration fluctuations. The fact that Shaw et al. (2002) did the renormalization is not convincing (although I had just a quick look at that paper without getting into details). I feel this has a significant impact on the results and their interpretation.

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REPLY: You are right that a better job needed to be done to correctly explain the normalization process and why it was needed. I think Referee 2's initial comments do a good job of summarizing the findings in Saw et al. (2012). They found that when two clustering signatures (or inhomogeneities) are occurring and are uncorrelated to one another and have a large enough scale separation (such as inertial clustering on the Kolmogorov scale and entrainment induced clustering occurring on cm to tens of meters scale), the PCF equals the product of two PCFs, each results from the one of the two clustering phenomena acting alone, i.e., n(t) = n_1(t)*n_2(t).

The resulting PCF curve that is produces when both inertial clustering and larger scale inhomogeneities are present is described on page 6 lines 13-21 of the new manuscript. The curve has a power law region at the smallest scales where inertial clustering is present, followed by a 'shoulder' region that is due to the larger scale inhomogeneity. The curve then falls of towards zero at even larger scales (see Figure 4 in Saw et al. 2012). Figure 5 in Saw et al. 2012 goes on to show how the shoulder region of the curve can be 'normalized' to a common value (in this case a value of one for the Radial Distribution Function, in our case it was zero for the PCF). This then allows for analysis of the clustering at the smaller scales.

With that stated, we do believe that the process by which we did the original normalization was flawed. In particular, we were normalizing the PCF curves at the largest scales where the PCF continues to decrease sharply (the region directly after the shoulder region) in order to analyze the differences in the shoulder region. However, in Saw et al. (2012) the PCF is normalized at the shoulder region in order to compare differences in the inertial clustering. We are unaware of any work that has been conducted which shows PCF normalization is valid at the largest scales in order to analyze the shoulder region scale clustering. Although it has been shown that the PCF can be normalized in the shoulder region to analyze inertial clustering, normalizing the PCF at the largest scales to analyze the entrainment scale clustering may or may not be valid.

COMMENT: 1. P. 1, L. 20. I think it would be appropriate to cite Grabowski and Wang

(2013) that reviewed the progress in this area a decade after Shaw (2003).

REPLY: We have kept the original reference of Shaw (2003) but have also added Grabowski and Wang (2013). This section of text now occurs on page 1 line 25 to page 2 line 2.

COMMENT: 2. P. 1, bottom paragraph: This is a very pessimistic message. I think there are many realistic cloud simulations showing relatively fast rain formation (e.g., vanZanten et al. 2011, Seifert et al. 2010, Wyszogrodzki et al. 2013, Khain et al. 2013).

REPLY: The original idea with this was to motivate why droplet clustering is so important, since classical droplet growth theory does such a poor job of estimating the rain formation time. This discussion now appears on page 2 line 4-8. The references you mention have been added on page 2 line 8-11 to state that progress has been made in modeling precipitation. However, we also state that improvements still need to be made, as is mentioned in Wyszogrodzki et al. (2013).

COMMENT: 3. P. 2, L. 5: Earlier references than Khain and Lehman would be appropriate here. For instance, the exchange between Telford and Chai on one side and Jonas and Mason on the other in QJ in 1983 is worth referring to (the great phrase comparing the impacts of entrainment to turning down the gas to boil water faster!). Perhaps some papers of Charlie Knight at about the same time are also of relevance.

REPLY: Entrainment is first mentioned on page 2 lines 11-13 of the new manuscript, and includes the references of Telford and Chai and Jonas and Mason. Since the focus of the paper has been shifted from inertial clustering to clustering caused by entrainment, an extensive discussion on entrainment is given on page 2 lines 22-29 and page 4 lines 13-32 which includes references of some of the earlier work on entrainment, including Warner (1969) on page 2 line 12 and Baker (1980) on page 2 line 24.

COMMENT: 4. P. 2, L. 6: For GN and UGN, there is the Illingworth (1988) paper way before the Knight et al.

REPLY: The discussion on GN and UGN has been completely removed from the new manuscript.

COMMENT: 5. P. 2, L. 8: “no one theory” is simply incorrect, please see 2 above.

REPLY: We were not trying to imply that none of the theories are correct. We were implying that it is most likely a combination or all of the theories that account for the fast formation of rain that is observed. With that said however, this statement has been removed. And the discussion on the different theories on the fast formation of rain which originally occurred on page 2 lines 3-7 (such as turbulent deviations of supersaturation and GN and UGN) has also been removed to just focus on entrainment mixing and droplet clustering.

COMMENT: 6. P. 2, L. 9-18: this paragraph should be deleted as irrelevant to the results presented. Referring to Small et al. (2009) rather than to original papers (Twomey, Albrecht, etc.) is simply inappropriate. Also, Xue and Feingold (JAS 2006) is a more appropriate first reference to the impact of cloud-edge evaporation impact.

REPLY: This paragraph has been modified in accordance with your comments. We have included Xue and Feingold (2006) which is a modeling experiment, but have also kept Small et al. (2009) since this is an in-situ experiment. Any mention of direct and indirect effects have been removed from the paragraph (which we do agree is irrelevant, especially with how the manuscript has been refocused). However, the mention of the evaporation-entrainment feedback is important, as it explains why we would expect to see enhanced entrainment (and as a result enhanced clustering) for the high pollution clouds as compared to the low pollution clouds. This paragraph can now be found on page 2 lines 16-21 of the new manuscript.

COMMENT: 7*. P. 2, L. 19-26: Brenguier and Chaumat (2001) has to be cited here to show that not all studies indicate significant clustering at small scales! In fact, I would

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argue that the clustering is relatively small (e.g., see Fig. 3 in Kostinski and Shaw 2001 and Fig. 5 in Vaillancourt et al. 2002).

REPLY: A brief discussion on the findings of Brenguier and Chaumat (2001) are given on page 3 lines 14-16 of the new manuscript. We also report that not all of the clustering measured shows a statistical difference from a random distribution. In particular see page 9 line 29 to page 10 line 10 of the new manuscript. Also see page 14 lines 1-5, where we state that 31.7 percent of the data we measure (cloud traverses) are randomly distributed.

COMMENT: 8. P. 2, L. 34: there are many more references than Pinsky pointing out to the significance of droplet clustering for collision/coalescence. Perhaps as reference to a review by Grabowski and Wang (2013) would be appropriate here.

REPLY: The quantitative results that were previously discussed have been removed (page 2 lines 30-33 in the original manuscripts) since this is beyond the scope of the current manuscript. It is important that the readers understand how droplet clustering can effect collision coalescence in a conceptual manner however. Page 3 lines 10-13 of the new manuscript have added to Grabowski and Wang (2013) reference in relation to how droplet clustering effects collision-coalescence.

COMMENT: 9*. P. 2, L. 35. I do not think the impact of the Reynolds number (i.e., the range of spatial scales) is important for droplet clustering at the microscale. However, the difference in the eddy dissipation rate between laboratory experiments and natural clouds is the key. That was pointed out by Grabowski and Vaillancourt (1999) who also discuss the role of droplet sedimentation and referred to observations that were subsequently reported in Brenguier and Chaumat (2001). Discussion of the latter aspect, droplet sedimentation, is completely missing from the manuscript.

REPLY: We would agree that the dissipation rate is much more important than the Reynolds number (both for smaller and larger scale clustering), especially since the Stokes number depends on the dissipation rate. The original comment related to the

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Reynolds number has been removed. We mention the Reynolds number on page 5 lines 22-26 of the new manuscript, stating that it has been found that large scale clustering does not appear to depend on the Reynold's number as is stated in Ireland and Collins (2012). Note that we still mention the physical mechanisms that small scale clustering depends on. It is important to distinguish the physical differences between small scale inertial clustering and larger scale clustering as a result of entrainment mixing.

COMMENT: 10*. P. 3, last paragraph of the introduction. I think the introduction should lead to the questions posed in this paragraph. The way introduction is written right now does not do that. For instance, why one should expect clustering to depend on the aerosol concentration? Because clustering is expected to depend on the droplet size. Why it should be different between cloud edge and cloud center? Is that because of the droplet size (arguably smaller at the cloud edge) and intensity of the turbulence (larger at the edges)? Similar for the dependence of the distance from cloud base. Etc. Etc. I feel a complete rewrite of the introduction addressing all points above and leading to these questions is needed.

REPLY: We believe the points that you have made here have been met in rewriting the new manuscript. For example, after each of the questions proposed at the end of the introduction, a hypothesis is given which an explanation of why said hypothesis was formed. For example, question 3 on page 3 lines 31-32 states that "it is hypothesized that an increase in aerosol load leads to enhanced clustering due to the resulting increased entrainment from smaller droplet sizes and evaporation". The background information on the reasoning this hypothesis is formed is found on page 2 lines 16-21 of the new manuscript.

COMMENT: 11. P. 3, L. 23: Kolmogorov scale is around 1 mm in atmospheric turbulence.

REPLY: This has been corrected in the new manuscript, as seen on page 2 lines 31-32.

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COMMENT: 12. P. 4. Eq. (1) is valid only for the case without gravity, correct? If so, it is not appropriate for cloud droplets.

REPLY: Yes, Eq. (1) was valid only for the case without gravity. This equation has been removed from the revised manuscript.

COMMENT: 13. P. 4, center: the discussion here should include the effects of droplet sedimentation, see Grabowski and Vaillancourt (1999) and perhaps other papers (e.g., from Prof. Lian- Ping Wang?).

REPLY: A discussion on droplet sedimentation has been added as was discussed in the reply to your number 3 major comments, see page 5 lines 15-22.

COMMENT: 14. P. 4, L. 24: the reference to Shaw (2003) is not correct here. The shear near cloud edges comes from cloud dynamics, not evaporation, see original study of Grabowski and Clark (1993) and more recent support from Park et al. (2017).

REPLY: The statement from Shaw (2003) has been removed from the manuscript. Turbulent kinetic energy is no longer discussed in the new manuscript.

COMMENT: 15. P. 5, L. 1: Should Twomey's or Squiers' old classical papers on droplet activation and growth be referred to here instead of Small and Rosenfeld?

REPLY: The statement in which this comment is referring to has been removed from the new manuscript. However, we have cited Twomey (1997) in relation to aerosols and cloud droplet size on page 13 lines 11-12 of the new manuscript.

COMMENT: 16. P. 5, L. 9: Rather than Pinsky and Khain, the original inhomogeneous mixing papers (John Latham, Marcia Baker, etc.) should be brought here.

REPLY: The statement in which this comment is referring to has been removed from the new manuscript. However, since we have shifted the focus to entrainment mixing, we do include multiple citations from Baker, including Baker et al. (1980) on page 2 line 24 and Baker et al. (1984) on page 2 line 32.

COMMENT: 17*. P. 6, L. 23. Please explain how the normalization is done. Is the PCF simply shifted up or down to have zero at large scales? If this is an entrainment zone and droplets are clustered at large scales (as clearly illustrated in the left panel of Fig.1), the analysis should show that! This aspect is completely missed by the normalization. If the purpose of the analysis is to show small-scale clustering, then estimating the absolute magnitude of such small-scale clustering is impossible with the normalization.

REPLY: A detailed description of the normalization process has been provided in the reply to Major comment number 4. But in short, yes the PCF is simply shifted up or down to converge to a set value at larger values (in our case the set value is zero). Again, normalization is no longer done in the new manuscript.

COMMENT: 18*. P. 7, L. 17. Fig. 1 clearly shows that the patchiness is at large scales (meters), not at small scales. Renormalization takes this aspect away. Is the focus of the analysis on concentration fluctuations at large scale (meters) or at small scales (centimeters) scale? I would think the latter. To me this is the key flaw of the analysis.

REPLY: We believe you are correct (along with Referee 2's comments) in stating that the PCFs in this paper should be more appropriately associated with the inhomogeneity occurring at larger scales due to the entrainment of dry air, as opposed to inertial clustering. An explanation of the PCF curve and how it should look for inhomogeneous data is given in the new manuscript on page 6 lines 13-21, and this is also conveyed on page 7 lines 9-12 in describing the PCF for real data calculated in Figure 1. It is believed that the PCF scale we currently use does not extend to small enough scales to even measure inertial clustering (we would need to measure down to mm, whereas we currently only extend down to $\sim$ 3 cm). Lehmann et al. (2007) does not record an increase in the PCF due to inertial clustering for cumuli until length scales of approximately 1mm. We do not extend to the mm scale because we would need to reduce the dt used to calculate the PCF (see page 6 line 24-26), which would result in an even noisier PCF than what is already displayed.

COMMENT: 19*. Fig. 5. First, I am curious how the figure looks without the normalization (see major point 3 above). The specific discussion in lines 15-20 on p. 9 may change if no normalization is performed. For instance, I think the statement on line 21 (“droplet spacing shifts from non-homogeneous to homogeneous at larger spatial scales”) is incorrect as I expect the cloud edge to be quite heterogeneous at large scales (meters and up). The interpretation the authors provide comes from the normalization and it is counterintuitive.

REPLY: As can be seen in Figure 5 of the new manuscript, the overall PCF characteristics are unchanged. The main difference in the de-normalized curves is that they no longer converge to zero at the largest spatial lags on the x-axis (some PCF curves are greater than or less than zero). In relation to the discussion that was originally in lines 15-20 on page 9 (which can now be found on page 9 lines 16-22 in the new manuscript), you can see that the overall conclusions remain unchanged. L1, H1, and H2 still remain statistically significant from each other (when comparing center and edge data) while L2 is still statistically similar. In relation to the statement “droplet spacing shifts from non-homogeneous to homogenous at larger spatial scales”, this statement was made because we were forcing the PCF to zero (therefore, the statistical significance broke down at larger PCF values), not because homogenous conditions were physically present within the cloud at the time of measurement. Although this statement has been removed in the revised manuscript, this should have been made more clear originally.

COMMENT: 20. Fig. 6. I do not understand what the value of the PCF is shown. Is that the asymptotic values at small scales (i.e., the left edge in Fig. 1)?

REPLY: Yes, the PCF value is the average of the left-most edge of the PCF curve (the first 21 PCF values in the original manuscript). In the revised manuscript, the PCF value shown is an average of the first 60 PCF values that make up the x-axis. In other words, it is the average of the PCF values below $\sim$ 1 meter (or the PCF values that make up the ‘shoulder’ region of the PCF curve).

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