

## ***Interactive comment on “Superaggregates or instrument artifact?” by Ashley M. Pierce et al.***

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Anonymous Referee #1 Received and published: 19 December 2017 This work investigates the observations of superaggregates collected during a study at high and low elevation sites in Nevada in 2014. The authors investigated the potential influence of biomass smoke, a known source of superaggregates. The also investigated other possible sources and deduced that the source of superaggregates was likely related to the anodized Al tubing and fretting corrosion. This work is useful for others investigating observations of superaggregates and the potential role of sampling artifacts. Several fairly minor issues should be addressed before publication as noted in the following comments.

Response: Thank you for your thorough review of the manuscript.

Page 1, Line 17, Can the authors phrase this as 36 out of X sample days?

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Response: This has been changed to “36 out of 158 sample days”.

Page 1, Line 21, This sentence is a bit unclear because it groups the types of influences that could be the source of superaggregates with the types of analysis performed.

Response: This sentence has been changed to “To determine if the particles were superaggregates or an instrument artifact, the presence of certain elements, the occurrence of fires, high relative humidity and wind speeds, as well as the use of generators onsite were investigated.”

Page 1, Line 22, Change “samples with aggregates” to “samples with superaggregates”, or state that they are the same thing. I think “aggregates” is used interchangeably with “superaggregates” throughout the paper, so it might help to be clear about this initially.

Response: The term “aggregates” is used when discussing large aggregated particulates on the filter surfaces under discussion while “superaggregates” is specific to aggregates that have made it past the inlets and are similar in size and morphology to previous studies. This has been clarified throughout the abstract and introduction.

Page 1, Line 24, Change “high wind events were the probably reason” to “high wind events were probably the reason”

Response: “probably” has been changed to “probable”

Page 2, Line 2, Can the authors clarify what they mean by “can be trapped in a high particle volume fraction”?

Response: Large number of particles in a small area causing aggregation. This has not been changed as this is similar terminology to previous studies.

Page 2, Line 6, Can the authors define “length”

Response: Length has been removed.

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Page 2, Line 8, What are the “different behaviors” the authors are referring to here?

Response: This has been removed based on comments from referee 2.

Page 2, Line 10, Can the authors define “fractal dimensions”

Response: This has been removed based on comments from referee 2.

Page 2, Line 14, Chakrabarty (2014) should be Chakrabarty et al. (2014)

Response: This has been corrected and moved.

Page 2, Line 21, How abundant are superaggregates? Are they abundant enough to influence estimates of climate forcing?

Response: This has been removed based on comments from referee 2.

Page 2, Line 22, It would help to have a segue between this paragraph and the previous discussion.

Response: This section has changed based on comments from referee 2.

Page 2, Line 24, To help the reader, the authors could add “a high elevation site” before “Peavine Peak”, and a similar “at a lower elevation site \_12 km southeast in Reno”. The elevations may not be needed here since they are again reported in the site description section.

Response: This has been changed.

Page 2, Line 27. Can the authors add the total number of days here, so “36 out of X days”.

Response: This has been added.

Page 2, Line 31: Do the superaggregates have to conform to this particular description?

Response: Yes. If the aggregates are not “fluffy” or if the particles are much larger than

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they likely have different fractal dimensions and different behavior in regards to inlets. “(e.g. fractal dimensions and lengths)” has been added here.

Page 2, Line 34: Please define “SEM” and “EDS” at first usage.

Response: SEM and EDS were defined on Page 2 lines 12 and 13. However, this section has changed based on comments from referee 2.

Page 3, Line 19-21, What is the purpose of this study and how does it fit in with the 2014 study? Also, change “Statuses” to “Status”

Response: “The TAPI located at PEAV was then moved to GBNP and measurements were made at this high elevation site with impacts from wildfires.” was added here for clarification. Statuses has been changed to Status.

Page 3, Line 24, Please add “mass concentration” after PM2.5

Response: This has been added

Page 3, line 26, Should “<” actually be “>” (particles larger than 2.5 um are being prevented)

Response: Yes, thank you for catching that, it has been changed.

Page 4, line 32, Please add “Particles on the Teflon filters”

Response: This has been added.

Page 5, Line 9, How far away from the sample sites was the AERONET site?

Response: Distance has been added here.

Page 5, Line 12, Was AEE calculated using only 2 wavelengths or as a linear fit to several wavelengths?

Response: The wavelengths used for the AEE calculation were provided in the first sentence of this paragraph (440 and 870 nm).

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Page 5, Line 13, Given that the AERONET site is at the campus building, the impacts of urban pollution on AEE could be misinterpreted as biomass smoke if all AEE > 1.8 was flagged as fire. How did the authors separate urban influence from biomass smoke?

Response: Previous studies cited Loria-Salazar et al. 2014, 2016, and 2017 discuss the use of the fire flags. These studies conclude that local conditions of aerosol pollution in Reno, Nevada is most likely impacted by coarse mode particles when AEE is lower than 1.2. Urban emissions of aerosol pollution have an AEE that ranges from ~1.2 to ~1.7. Fine mode fraction coming from fires shows very high fine mode fraction and AEE larger than 1.8. "...and when fine mode fraction was > 0.6..." was added here for further clarity.

Page 5, Line 18, It is again unclear why data from the GBNP site are used here since it is a different time period. It would help to explain this earlier.

Response: A sentence was added to the site description section.

Page 6, Line 3, Add PM2.5 before "75th percentile". Does this paragraph only refer to 1 inlet?

Response: PM2.5 was added before concentration here.

Page 6, Line 12, Are "black aggregates" different from superaggregates? A similar comment for "black particles" on line 13. Are "black particles" just a general description or refer to "black aggregates"?

Response: Black aggregates refer to aggregates under investigation here and are not necessarily superaggregates as defined in previous work. "Black particles" has been changed to "black aggregates" here.

Page 6, Line 27: How did the aggregate analysis differ from the fluffy or compact aggregates shown in Figure 3 versus Figure 7? The visual appearance is quite different. Perhaps Figure 3 is actually a superaggregate?

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Response: Analysis for fluffy vs. compact aggregates was the same, SEM and EDS. The first SEM images were a cursory look at the aggregates; the second SEM analysis was more thorough. Yes, the goal of Fig. 3 and 7 is to illustrate that it is possible to find aggregates that look like superaggregates. Fluffy aggregates such as Fig. 3 lead to the hypothesis that superaggregates may be depositing past the inlets, however, these are not the particles that are visibly black and dominant on the filters as they are much smaller and not common. This is explained in the previous paragraph on this page and in section 4.2.

Page 6, Line 27: Did the elemental ratios differ between aggregates and non-aggregate PM2.5? For example, did the Al/O or the Al/Cu differ?

Response: The Al:O ratio differed from aggregates (0.40 to 0.91) to non-aggregates (0.06 to 0.61). Cu only occurred on one filter and only in an aggregate. Al:C and Al:F also differed between aggregate and non-aggregate. The rest of the elements were not consistent throughout the samples but differed when present on aggregates and non-aggregate samples. The authors think that this information is illustrated by the differences in the percentages of the elements.

Page 7, Line 7-12, This is somewhat confusing. All data had  $r^2 = 0.33$ , aggregates only was  $r^2 = 0.49$  and then no aggregates was  $r^2 = 0.58$ . If the correlation for aggregates only increased, then why did the correlation increase even higher for no aggregates?

Response: The aggregate only and the non-aggregate datasets are differently correlated, when combined into one dataset the correlation for all data decreased.

Page 7, Line 7-12, The offset in the regressions suggests that when AOD = 0, PM2.5 = 2 ug/m3 or 6 ug/m3 depending on all data or aggregates, respectively? Can the authors comment on this offset?

Response: AOD is a measure of the columnar aerosol loading and does not necessarily directly relate to the surface PM2.5 measurements. Both instruments also have

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different minimum detection limits that affect the offset.

Page 7, Line 10, A Theil regression would help with this in that it doesn't heavily weight outliers.

Response: Thank you for the suggestion. A Theil regression would be helpful if there were multiple outliers and if AOD was a probable cause of the aggregates. However, because there is only one major outlier and AOD is not the probable cause, we do not think it would add more insight to the paper.

Page 7, line 13, Figure 10(a) isn't referred to in the text. I am also concerned that the AEE used here to indicate smoke influence could also be indicating urban pollution. How did the authors account for this?

Response: 10a has been added here. Previous studies cited in section 3.3 Loría-Salazar et al. 2014, 2016, and 2017 discuss the use of the fire flags, clarification has been added in section 3.3.

Page 7, line 15-16, Change to "nor between AEE and PM2.5 on days with aggregates"

Response: This has been changed.

Page 7, line 19, Was RH averaged to 24 hours?

Response: Yes, "24 h" has been added to this line.

Page 7, line 24, If the authors are interested in decreasing the length of the article, I am not sure that Figure 11 is necessary. The discussion of results may be sufficient. Also, it would help to point out that the hypothesis with investigating RH is that hygroscopic effects could have resulted in larger particles. However, the elemental composition and SEM images don't suggest hygroscopic particles.

Response: Figure 11 has been removed. The last sentence of this paragraph was modified to indicate that hygroscopic growth could be causing larger particles.

## C7

Page 8, line 9, Why were multiple generators operated?

Response: This is explained in section 3.5.

Page 8, Line 15, Were generators not in use on the weekends?

Response: We do not know. Regular maintenance of anything at the station would occur during normal work hours unless there was an unexpected loss of power, which we are not aware of occurring during the sample period. This has been clarified.

Page 8, Line 23-25, High aerosol loading in the atmospheric column doesn't necessarily have to indicate the presence of superaggregates. It could just be related to urban pollution.

Response: This sentence points out that the correlation between high column loading and aggregates suggested that there could be a relationship between high column loading and aggregates, not that it indicates the presence of superaggregates.

Page 8, Line 29, The correlation of AOD and AEE may not be higher if there aren't enough superaggregates to affect the total column?

Response: This is based on findings in Loría-Salazar et al. 2017, which identifies the atmospheric physical conditions where AOD is associated with PM2.5.

Page 8, Line 30, It's not really clear why the authors included this paragraph? Did they perform some modeling of the hygroscopicity based on composition measurements? Can they tie this discussion back to their data?

Response: This paragraph explains what % RH is important for determining hygroscopic growth of different particulates. Some clarification has been added to this and the following paragraph.

Page 9, Line 30-35, Elements are defined here but not when listed on page 6 (24-29). I suggest being consistent or defining them once at first usage.

## C8

Response: This has been fixed.

Page 10, Line 12-13, "RH and AEE were not correlated with aggregate measurements". Can the authors be more specific about the measurements they are referring to? Presence of aggregates? Length? Composition?

Response: This has been changed to presence of aggregates.

Figure 2: The two blue colors were very similar in my version and so difficult to tell the difference.

Response: The blues have been changed to increase visibility.

Figure 3: The superaggregate shown in this figure is very different from Figure 7- which type was more typical?

Response: This is discussed in the manuscript in the introduction (section 1), section 4.2, and table 1.

Figure 9: Instead of "in the valley", can the authors provide a more defined location of the site?

Response: This has been changed to "near the low elevation site in Reno".

Figure 10: As mentioned earlier, this figure may not be necessary. But if the authors choose to keep it, the legend for "fire" was not pink although the data points were. Also, please provide the wavelengths over which AEE was calculated in the caption. And the same comment regarding "in the valley" also applies for this caption.

Response: This figure has been removed.

Figure 11: As also mentioned, this figure may not be necessary either.

Response: This figure has been removed.

Table 1: Please define "CEM", "SEM", and "EDS" in the caption. Also please include "PM2.5" in "PM2.5 mass concentration exceeded 75th percentile" in the caption.

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Response: This has been added.

Table 2: Please include site location in the table, define "both" and "Teflon". Ideally the reader could get the major points without having to read back through the text and "both" or "teflon" might be unclear.

Response: This has been added.

Table 3: Similar comments as for table 2.

Response: This has been added.

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