Overall Comments: The authors use multiple measures to investigate the physical properties of non-sea salt aerosols collected in the Arctic. They qualitatively describe the mixing state of organic, soot, and sulfate aerosols. Although the core data set of this manuscript is informative, as is, the presentation and discussion are confusing and possibly misleading. No large scale descriptive statistics are provided, the methods and presentation are often unclear and/or redundant, the environmental context is lacking, and the optical conclusions are not well explained. This manuscript would benefit from additional analysis, clarification of methods, adding proper context to results, and careful grammar review.

General Comments:

1 ***Unclear Sample Selection*:** In multiple sections of this paper the authors discuss a subset of their samples without explaining why they were chosen. This detracts from their discussion and leaves the reader questioning what is not being discussed. Anytime a subset of samples is chosen or used, an explanation needs to be provided explaining why these are the best samples for that specific analysis or investigation. A few example occurrences can be found in lines 275, 281, 118, 190.

2 ***Environmental Context:*** The only environmental context explored in this text is the back trajectories provided for 4 of the sampling days. Whenever field data is discussed, external variables such as temperature, humidity, and back trajectories can help explain variability observed in the data. To improve this work, the context of the sampling should be explored as a driving factor between differences in particle characteristics.

2.1 I am also not convinced that the daily averaged back trajectory calculations are meaningful for these samples. (I'm also not sure the trajectories were daily averaged as the methods section just said they were calculated for a given day, not the frequency of times or given time on the day.) Because the sample length was between 20 minutes and 2 hours, a back trajectory occurring during the actual sampling time for all samples would be more appropriate.

3 ***Optical Property Calculations:*** The authors attempt to estimate the potential radiative forcing implications of their results using a core-shell model and Mie theory. Although this is a potentially meaningful result, the methodology needs to be discussed further and improved.

3.1 When performing Mie theory calculations the input size distribution characteristics and wavelength dependence (angstrom exponent) have dramatic effects on the results. These data need to be reported. The authors state that a single size distribution is used. How was this size distribution established? Could you run your analysis over all of the size distributions you observed to estimate the variability?

3.2 The way the authors include soot in their calculations is not appropriate, as soot is treated as a core with a mixed sulfate-OM shell. This is in direct conflict with their observations that

soot is only observed to be associated with the OM shell and sulfate is always the core. It would be better not to include a soot calculation than include a misleading one.

3.3 The relevancy of the refractive index (RI) choice needs to be discussed further. The authors use an RI from biomass burning brown carbon as their slightly absorbing case but frequently state that they believe the OM to be secondary. If that is the case, their estimations are likely too high for all cases but the non-absorbing case. The authors should include refractive indexes from secondary organic aerosol brown carbon to get a more realistic answer. These are generally much lower than the RI values that they used. Additionally, the scattering component chosen (1.65) is relatively high and not explained. Please explain why this value was chosen as it is especially important in core-shell cases.

3.4 Stemming from the above comment, the relevancy of each refractive index case needs to be discussed in the context of the Arctic. Which case seems the most likely?

3.5 The authors use m=1.55 for sulfate. RI values have wavelength dependence, was this included in any way?

3.6 The authors average the absorption cross section on a per particle basis. This isn't meaningful since observed absorption cross sections will depend on the whole aerosol population (and the size of the particle). The authors should calculate an absorption cross-section for the ambient aerosol concentration during their sampling and compare it to other absorption observations.

4 ***Redundancy and Clarity****:* The writing of this paper needs improvement. There are multiple times when the authors restate the same point twice or fail to introduce a topic before discussing their results. An example of redundancy can be observed when Copper TEM grids are introduced in lines 120, 140, and 156. An example of an improper discussion occurs in the discussion of satellite particles, which are introduced in line 280 with no context or explanation. This forces the reader to infer what the authors mean by satellite particles, possibly leading them to wrong conclusions. To improve the manuscript, I recommend careful reconsideration of the presentation of the data, with special consideration to avoiding redundancy and ensuring the appropriate context is present.

4.1 There are multiple points in the methods, a few of which I've included in the specific comments, that are unclear or confusing. The reader needs to be able to understand exactly which analysis was performed on which filters for how many particles if they are to believe your result.

5 ***Sample Information*****:** The only summary of the total data set is provided in figure S6 and this figure states that only 3 samples (of 46 collected and 21 analyzed). No explanation is given as to why is summary is so limited given the authors have EDS data (which they used for classification) of 20-30 particles over 21 samples (at least 400 data points). All the of following discussions only make sense if they are provided in the context of overall sample composition.

Specific Comments:

1 There are many grammatical errors and redundancies that I have not addressed below.

2 The phrase internal mixing is used throughout the manuscript without an explicit definition. An explanation of what exactly you mean when you say something is "internally mixed" would improve the manuscript

3 **Line 55:** Change "Artic" to "the Artic"

4 **Line 60:** Change "nature" to "natural"

5 **Line 64-66**: Restructure this sentence for clarity. Treat the percentages in a consistent way as to not confuse the reader. For example, this sentence could be changed to:" For example, Winger et al.(2017) showed most Arctic BC is sourced from domestic activities (35%) and transportation (38%), with only minor contributions from gas flaring (6%), power plants (9%), and open fires (12%)"

6 **Line 72:** I'm not sure exactly how this sentence fits in with the brown carbon theme of this paragraph. Are these compounds commonly found in brown carbon or organic aerosols in general? Please add some context.

7 **Line 84:** replace "were" with "have been"

8 **Line 96-68:** This sentence is confusing. Please rewrite it more concisely and clearly.

9 **Line 99-100:** Change "collected on 7 to 23 August, 2012 in the Arctic." to "collected in the Artic between August 7th and 23rd, 2012."

10 **Line 104:** replace "on substrate" with "on a substrate"

11 **Line 118:** Change "samples between 7 and 23 August, 2012." to "samples collected between August 7th and 23rd, 2012."

12 **Line 119:** Replace "analyzed for TEM analysis" with "analyzed with TEM"

13 **114-131:** Restructure your sampling section. As written the reader may think that you sampled with 2 separate samplers an individual particle sampler and cascade impactor. After reading the paper, there is only one sampler. This confusion can be remedied by introducing the cascade impactor earlier.

14 **122-123:** Add the top size cutoff for this sampler.

15 **138:** replace "within a" with "for a"

16 **148:** 2002 particles examined over all the samples, or in a specific filter?

17 **151:** Clarify this sentence. Do you mean to say you ", we only checked elemental compositions of 20-30 particles" in each sample?

18 **155-156:** The statement about Cu is redundant. This has already been stated in line 120 and 140.

19 **156-157:** What is the difference between what is stated here and what is stated in lines 147-149?

20 **162-163:** Replace "is **the** image analysis platform…" with "is **an** image analysis platform".

21 **177:** Replace "Organic Matters" with "Organic Matter"

22 **Line** 184: Replace "TEM grids was" with either "The TEM grid was" or "TEM grids were"

23 **Line 192-194:** Incomplete sentence

24 **Line 194:** I stopped making basic grammar and structure critiques at this point.

25 **Line 222**: Does treating this as a core-shell system with BC in the middle and sulfate and OM mixed on the outside have any basis? You've indicated that you have soot inclusions on the outside of predominantly sulfate particles, so why would soot be on the inside?

26 **Line 224:** This sentence reads as if you've calculated the refractive index of the particles. Did you measure the optical properties of these particles?

27 **Line228:** "In this study". It is unclear whether this is referring to your previous work or this manuscript.

28 **Line 253:** Is it possible that coagulation of primary organic particles and S-rich particles could have led to the formation of organic coatings? Are you sure assumption that all organic coatings are secondary valid?

29 **Line 272:** Can you say a percentage of NSS particles that are S-rich with an OM coating? Or a percentage of S-rich particles that have a coating? This would strengthen the paper if an actual number was given.

30 **Line 274-276**: Are these specific samples special or is there something that you think may have caused the low frequency of soot inclusions? If so, please explain why.

31 **Line 277:** This statement needs to be better supported. Just because a site is remote does not mean particles are local in origin. If this is supported by your trajectory calculations, mention them here. Also, don't the soot inclusions also imply that perhaps the OM is not secondary in nature? Soot is 100% primary and often co-emitted with primary OM, so if there is soot associated with OM coatings the soot itself is primary and so possibly some of the OM is primary as well.

32 **Line 280:** You need to define what satellite particles are; you have not discussed or defined them previously. Are they simply splatter of liquid portions of the particle when the particle is collected?

33 **Line 280-281:** is there a reason why satellite particles would have been observed on these days but not other days?

34 **Line 281:** This is misleading and implies you performed the NanoSIMS analysis on 11 samples. In the methods section, it says only two samples were analyzed with NanoSIMS.

35 **Line 287-289:** This is misleading. It reads as if you have done molecular characterization of the organic matter.

36 **Line 314**: You back trajectories are only for specific days, be transparent about this in the discussion

37 **Line 317-318:** This conflicts with your earlier comment that most BC should be local.

38 **Line 326-342**: This discussion reads like a list of facts, but why they are all relevant is not always stated. Explain why each observation is important and how it adds context to your results.

39 **Line 343:** Why is dry included here? Are there also wet particles that you have not discussed?

40 **Line 344-347:** Explain LLPS in simpler terms and why it's important.

41 **Line 346:** It's unclear what 90% is referring to in this statement.

42 **Line 348:** There's likely a complex relationship between phase state, oxidation state, and humidity. This needs to be investigated and explained further if statements about aerosol age are going to be made. Additionally, shouldn't you see a variety of ages of aerosol in your samples? Showing contrast between aged and unaged particles would be interesting and convincing.

43 **Line 376-378:** This is circular reasoning because $^{12}C^{14}N^-$ was what you used to identify OM so of course it was observed in the OM coatings. I don't think this data set is appropriate to make conclusions about the N content of OM coatings. That said, if you were able to calculate the mass concentration of N in the coatings with NanoSIMS that might give you a better indication of the BrC potential of the OM.

44 **Line 390-393:** The average absorption cross-section is reported on a particle basis. This would be much more meaningful if it was extrapolated to environmental conditions. Because you've sampled from the atmosphere, you should be able to approximate particle concentrations, correct? Is so, you could back calculate this to an actual atmospheric absorption contribution and compare it to expected absorption from other species and measurements. This would be significantly more meaningful.

45 **Line 413, 260:** 29% number should include a standard deviation.

46 **Line 410:** The last section shouldn't simply repeat what was stated in the above sections, but instead present the data in additional context and discuss the implications.