Anonymous Referee #1

Dall'Osto et al. present an interesting 3-year analysis of multi-site aerosol size distributions in the Atlantic Arctic and attempt to assess the sources based on spectral characteristics and linkages to collocated compositional information. This work is an extension of Freud et al. (2017), who presented a Pan-Arctic evaluation of aerosol size distributions but without chemistry linkages. Although the results are useful and warrant their place in the literature, there are several issues that need to be addressed prior to publication, as discussed in more detail below.

We thank reviewer 1 for finding the analysis interesting. Comments addressed below in italic.

General issues:

The authors allude to the fact that that the reason the 3 sites were chosen was because they surround the Fram Strait. There is nothing wrong with focusing on a specific region within the Arctic; however, the introduction describes Arctic processes in general and does not provide sufficient rationale for why this region in particular, other than it is warming the greatest during the last three decades. This reasoning is important but is only briefly mentioned. If the focus is sites surrounding the Fram, then more background and motivation is needed to clearly provide the link for why only these 3 sites are used and only for 3 years, as compared to the many sites presented by Freud et al.

Freud 2017 presents data from five Arctic stations, which was extraordinary and very valuable. However, it is important to stress that some of the data coverage (ie Barrow for example, 25%) was not good enough to compare stations. We therefore chose a different time period (2013-2015). For example, the VRS dataset provided here (2013-2015), was mostly not included in Freud et al., 2017 (2011-2013). The group of authors for the present paper is a consortium representing the three measurements sites, and those performing the mathematical analysis. Additional discussions are made in the introduction, including comparing stations nearby (ZEP, GRU) and compare a part of the Arctic that is changing rapidly (Svalbard region) and, finally, a discussion about different Arctic bioregion is also discussed.

We use data from the stations Gruvebadet (GRU), Zeppelin (ZEP) and Villum Research Station – Station Nord (VRS). The European Arctic is understood here as the part of the circumpolar Arctic located between Greenland and northwest Russia. Geographically, Greenland is part of the continent of North America. The Fram Stait, roughly between 77°N and 81°N latitude and centered on the prime meridian, is located between Greenland and Svalbard. The climate in the Northern hemisphere is centered in the Fram Strait. The golf Stream brings warm water to the eastern part of Fram Strait, where Svalbard is located creating a mild climate, whereas an ice stream is flowing out of the Arctic Ocean along the East Coast of Greenland with a strong

cooling effect. As a consequence is a large atmospheric temperature gradient exists across the Fram Strait of 16 ºC with an annual average temperature at Villum Research Station at Station Nord of -16 ºC and -2 ºC at Longyear byen, Svalbard (Last climate normal). This gradient has large consequences for the physical and chemical processes as well for the biological systems

On P6, I14, the authors even state, “However, to capture all scales of the variability of Arctic aerosols….” But that is not the goal of this work based on the information earlier in this paragraph. Along these lines, I get that the GRU site is the new addition from what Freud et al. presented, but other than that and a surface level utilization of the chemistry to infer the aerosol sources, I question what is new and novel about this work. Freud et al. presents all years possible at 5 sites and does a similar cluster analysis. Surely, the information is here, but the emphasis on how this study is a progression or even improvement from Freud et al. is not evident. The authors should take care to highlight the new findings and what is different from previous work. Also, in general, it would help if the introduction was rewritten to be more focused on the goals and motivation, with the appropriate background.

Freud et al. (ACP) presents a complete analysis focusing mainly on the Accumulation mode and its transport, leaving smaller ultrafine particles only partially presented. Additionally, the comparison between GRU and ZEP is not made, which is an important part of the discussion, as now stated in the paper. Finally, the cluster analysis reported here is a more in-depth analysis, not only because the number of clusters is higher and more accurate, but also because they are compared with a number of physical and chemical variables. In conclusion, whilst the Freud et al paper focuses more on Arctic particle number concentration and Accumulation mode anthropogenic transport, here we focus on a smaller dataset (3 stations) with higher time resolution (we report hourly and daily size distributions, not only monthly), discussing also chemical measurements reported across the stations, not discussed in Freud et al. (ACP).

Some of the categories need further explanation. First, it is not clear what the bursting category would originate from—any indication on the source? Why do they not grow to larger sizes? Is it possible this is local primary or secondary pollution particles, say, from generators?

The categories have been explained a number of times in a number of different papers (Dall´Osto et al., 2017,2018) and repeating the explanation would be redundant. We explain the possible sources without being too speculative. We exclude "generators" as data were checked for anthropogenic contamination, as also discussed in previous papers and in this current one.

Second, the definitions of Arctic haze and aged accumulation mode are solely based on a difference of 70 nm in the mode, but even with the chemical information presented, how can these sources be told apart? Where exactly does “aged accumulation mode” originate from? This distinction is not clear.

We would like to stress that one of the main difference - as discussed and emphasized in the manuscript - is also striking difference on the annual temporal trend shown in Figure 4. As discussed in the text, the Arctic haze shows a precise annual distribution, as reflected in the cluster name. Additional information on this specific accumulation clusters can also be found in a specific paper cited (Lange et al., 2018, Lange et al., Characterization of distinct Arctic aerosol accumulation modes and their sources. Atmospheric Environment 183 (2018) 1–10, 2018). This is now described in the text.

Along these lines, a couple sentences on the background of Arctic haze is warranted in the introduction.

Added.

Third, how can there be a coarse category when the coarse mode (> 1um) was not measured? Is this extrapolated from the tail of the spectra?

The author is correct in stating we did not measure particles >1µm, we stated coarse particles are the one detected in the highest detectable size of the SMPS, now edited in the paper. Main modes can be seen at 150 nm (category *accumulation_150*), at 220 nm (category *accumulation_220*) and in the largest detected SMPS modes at about 400-500 nm (category *coarse*).

Fourth, it is not clear what the nascent categories are—nascent as in sea spray? In general, more explanation and descriptions of the assumptions made are needed for the categories.

Again, we cannot speculate much about this source. Edited and added.

Provide uncertainties or standard deviations for any % values listed in the text and in a number of the figures (i.e., Figs 4, 5, 6, 7, 8, 9).

We added standard deviation where averages are reported. Edited.

It seems as if the chemistry is a crucial supporting aspect of the classifications. Perhaps these should be discussed first in the results and discussion, in tandem with the shapes of the spectra to justify the categories. I get lost in which sites had which chemical components measured. A table containing this information would be useful and would elucidate why most of the composition figures only have 1 site. In parallel, how can the limited information at each of the sites, with regard to the supporting chemical and CCN data, be used to glean information about the other sites? The sources/categories were not always the same as seen in Fig 4, so how can we expect to extrapolate the chemistry and CCN to all 3 sites?

We do not extrapolate any information about other sites, and each figure and legend has the names of the stations were the data were collected. This is a limitation of the study, and it shows to stress that very limited information are

collected simultaneously on different Arctic sites, as discussed in the conclusion, implication and recommendation.
Here we discussed the results of each station where data are available, in order to enhance - where possible - the SMPS cluster analysis. In other words - in addition to the aerosol size distribution clustering analysis - we report, where possible - chemical information for each station, where possible.

There is a bit of redundancy throughout. For example, the brief synopsis of Freud et al. is given twice with the same information in the introduction. Additionally, acronyms are defined but then the words are spelled out afterwards several times throughout. The manuscript could be streamlined by removing such redundancies.

The manuscript text was generally enhanced and streamlined, the mentioned redundancy was remedied. We thank for comments.

Specific comments:

Abstract: The importance of the ability to predict aerosol number oncentration is not clearly stated. Please provide the broader scope.

It was added that aerosols are, of course, an important part of the Arctic climate system.

P4, l15: Barrow has a long history of chemistry measurements there that support this generalized statement.

Added

P4, l21: “continental sources”, i.e., Arctic haze? Natural continental sources (e.g., mineral dust)? This needs to be made clearer.

Continental anthropogenic sources were meant. This has been rephrased.

P4, l24: What is “these frequent nucleation events” referring too? Which events?

The ambiguity has been clarified.

P5, l3 and 4: Provide appropriate references here.

The statement was rephrased to be slightly more hypothetical, and references were provided.

P6, l3-4: The fact that the Fram is the only deep-water connection between the oceans and the Arctic does not seem relevant.

While interesting, we agree that it might not be very relevant. The phrase was removed.

P7, l22 (and herein): The coordinates are all provided differently and are odd. Take care to fix throughout.

The way coordinates are displayed were harmonized.

P7, l24: By local sources, I assume the authors mean local anthropogenic sources. Surely, there are local emissions from natural processes.

Indeed the purpose is to state that the ZEP station is not influenced by local anthropogenic activity. This has been clarified.

P8, l20: The information about the hut name is unnecessary, since the coordinates are provided.

Historically a large number of important measurements have been made at Flygers Hut. It is a specific location of the Station Nord premises, where VRS is located. Since the establishment of VRS in 2015, the SMPS measurement setup has physically been moved to a different location, which is the "Air measurement hut". The two locations are very close, but the naming distinguishes these two locations. Flygers hut receives more anthropogenic influence, due to vehicle passages. It is an important information for readers who are familiar with the location and the record of measurements. Therefore, we prefer to keep the name.

P9, section 2.2.1: How does the overlapping sizes of the distributions compare?

The ZEP station is part of ACTRIS and aerosol size distributions are checked and validated. Overall, the overall size distributions overlap well. More information can be found in the ACTRIS web site, and in previous papers published in the ZEP site, including a long term analysis of Tunved et al., 2013.

Section 2.2: Some of the stations are missing the dates, time resolutions, or missing data. More details on the data are most certainly needed. Are there consistent specs between the measurements at the 3 sites?

Edited. Yes.

Section 2.3: Do these days overlap with the size distributions? I would assume so, but it is not clear. More details on the dates and where data were acquired are needed.

Yes, they do overlap. More information added. Simultaneously collected data are presented for the whole years (2013-2015).

P11, l5: Was this monthly average calculated from the daily averages or the highest time resolution of the data? Please be clear on how the averages were calculated, here and elsewhere.

Averages calculated from hourly data available, edited.

P15, l12-13: Or due to the large numbers of preexisting particles transported from midlatitudes.

Edited.

P29, l8: Methane sulphonic acid. Methane sulphonate is molecularly different.

Indeed, sulphonate is the corresponding base. Corrected to sulphonic acid

Figs 1 and 2: These are redundant. Figure 2 provides the information needed.

Figure 1 shows maps, while figure 2 shows size distributions. We assume that the reviewer refers to panel a and b of figure 1.While the impact of surface cover (Ice, water, snow or ground) was a part of the analysis, only fractional sea ice cover is included in this paper. Thus, the reviewer is correct in addressing the redundancy of the figure parts. However, they are provided with two different satellite measurements, and they are two different analysis. Whilst Fig 1a provide the total amount of sea ice, Fig 1b provides the amount of sea ice (open pack ice, consolidated pack ice) and these are important information given these marginal sea ice zones may be playing an important role in the aerosol size distributions, as discussed in the manuscript.

Fig 3: Redundant panels. Try combining or don't show each spectrum twice.

Agreed, since the y-axes are similar on each panel, the distributions don't need to be shown together all at once.

Figs 4, 5, and 6: I assume these are data from all 3 years? Provide more details in the captions when appropriate.

More explanatory figure captions have been provided.

Fig 5: Color for accumulation is not consistent between the graphs and legend.
Well spotted, this has been remedied.

Anonymous Referee #2
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The authors collected a unique data set of particle size distributions as well as chemical properties from three high Arctic sites during a three-year period and performed cluster analysis of the PSD to investigate the aerosol sources. Since there are few studies on Arctic aerosols, this paper certainly provides meaningful and valuable findings. It extends our knowledge of Arctic aerosols regarding particle size distribution, possible sources and chemical composition. I would recommend this paper be accepted for ACP, but major revision is needed.

We thank the reviewer for the appreciation of the paper, comments are found below

1. Is it possible to improve the title to a more precise one? Because the readers may expect to see the quantitative apportionment of natural and anthropogenic sources of Arctic aerosols. The sources are mentioned in the abstract, but there is no clear description or speculation later in results and conclusions about which category may link to natural/anthropogenic origin. I understand this apportionment could be very difficult. Perhaps authors can find a better way to summarize the main findings of the paper and show it in the title.

Edited. "Simultaneous European measurements of Arctic aerosol size distributions". Comment welcome, fair point.

2. The introduction looks quite long, so it could be shortened and the structure could be improved. For example, the third paragraph is very big (page 5 to page 7), mixing the research motivations of the three sites, the analysis method, and the need of international cooperation for future researches. Maybe authors can split the paragraph into two or more with a key sentence at the beginning or end of the paragraph, or remove some content.

The language of the introduction has been streamlined, and the overall text has been shortened a little. The paragraph was split up into three and topic sentences were added in the beginning of each new paragraph.

3. The method part actually does not contain the main method used in this paper, i.e. cluster analysis, which was found in the result part (Page 12, Lines 15-23). Any special reason for this arrangement? I would recommend authors to put the brief introduction of cluster analysis after the section 2.3. Also, the authors may provide more details of how to decide the cluster number and the reason of the current selection.

The section explaining the cluster analysis method, was moved to the method section.

4. The monthly average size distributions provide very interesting results but with limited interpretation. I would be interested to know the reason for the mode transition from June to August (single mode – bimode - single mode as shown in Fig.2). The size distributions in August and in October are very similar, anything wrong here? Should the diameter be shown as Dp to be consistent with previous text?

Many thanks for this comment, really well taken and much appreciated. The Figure of October was replaced with the correct one. All figures checked and edited and validated from original files.

I would be interested to know the reason for the mode transition from June to August (single mode – bimode - single mode as shown in Fig.2). We edited this in the text.

5. There are eight clusters of the aerosols in this paper, which are merged into three categories. The three categories were named quite early in section 3.2.1, but the following discussion was still pointing to eight clusters. I was lost in the middle of reading the paper. Maybe the authors can emphasize the three categories in the conclusion part rather than naming them early without discussion accordingly.

A clearer terminology was implemented. The cluster analysis resulted in eight size distribution categories, these categories are further assigned to three different classes. As shown here:

Class:	***Category:***
Nucleation mode dominated	*Pristine*
	Bursting
	Nucleation
Aitken mode dominated	*Nascent*
	Nascent broad
Accumulation mode dominated	*Accumulation_150*
	Accumulation_220
	Coarse (>300)

Table 1 edited

6. The authors are suggested to double check the seasons mentioned in the text with the months. For example, Page 15, Line 7: spring months are mentioned in the text while the peak of the curve was shown in June (summer?) at GRU site in Fig. 4a.

The description of the seasons and months where the categories appear was improved. Also, the caption of Figure 5 was made more precise.

7. Please double check the language to make sure the sentences are correct or precise enough on what you want to say. For example, Page 12, Lines 22-25, grammatical error? Page 18, Lines 12 - 14, it may be better to add a constraint of the sentence because the biogenic source could also be the major source for the nss-SO4 over the remote ocean. Page 19, Lines 11-13, does the "it" here refer to DMS or MSA? You may want to say DMS?

The text of the entire manuscript was generally streamlined and language precision improved.

We hope to have made a more correct statement about nss-SO_4

Note that DMS is released by the marine phytoplankton, MSA is formed later by atmospheric photooxidation. This was clarified in the text as well.

Other general comments:

1. Please use the uniform unit, e.g. degree for latitudes, Page 7 Line 22, Page 8 Line 11, Page 8 Line 16; for distance, km or Km; Fig. or Figure.

Use of units for coordinates, distance and figure references within the text have been harmonized. Please note that "Figure" should be spelled out, and not abbreviated "Fig." at the beginning of a sentence, according to the ACP author guidelines.

2. It seems there are many "double blank" between two words, which should be removed, e.g. Page 4, Line 20, between "they" and "have"; Page 4 Line 24, between "analysis" and "linked".

Double spaces have been removed.

3. Please correct the title of the Figure 1, should be Jan – Dec. (a – i), also Fig. 1a – i was covered by a shadow.

Caption of figure 1 has been corrected, please note that panel (a) was found to be obsolete at was removed. We could not identify any shadow, hopefully this was a temporary technical issue.