Response to Anonymous Referee #2

Thank you, anonymous referee #2, for your insightful comments on the manuscript. The paper is more robust thanks to your input.

Our response is structured as follows: original comments from reviewer #2 are bolded, our responses are in italics, and the revised portions of the manuscript follow in quotation marks with specific changes/additions in red.

Comments

In the abstract and in the introduction the authors comment about the importance of studying the in-situ surface aerosol optical properties in the Arctic given the sensitivity of the Arctic climate to short-lived climate forcers.

In general, more speculations about the reasons explaining the observed differences among the stations are needed to improve the scientific significance of the presented work. In most cases the manuscript presents a list of extensive and intensive values/properties at each station but the reasons behind the observed aerosol optical properties is sometimes missing.

> *The authors respectfully disagree with this general comment, as we do not believe speculation improves the scientific significance of a paper. While speculation may guide research questions for future work, the scientific significance lies in the evidence-based aspects of this analysis. Much of the scientific value of this work lies in the finding that aerosol optical properties vary widely with season at each of the Arctic sites, and vary widely from station to station. Though this analysis alone can't explain this spatio-temporal variability, it is a robust springboard for future work exploring the reasons for this.*

The manuscript should be more focused on the Arctic haze phenomenon. For this, a reorganization of the manuscript is needed. Some suggestions are given below.

> *While the authors agree that there could be more discussion in the manuscript about the Arctic haze phenomenon in relation to the seasonality of optical properties presented here, we do not think a reorganization is necessary. The goal of this paper was not to focus on the Arctic haze phenomenon, but to present seasonality of aerosol optical properties throughout the entire year. Part of exploring the seasonality means comparing aerosols during the Arctic haze season to aerosols during other parts of the year; consequently, this comment has been incorporated and care has been taken to more specifically address the Arctic haze phenomenon throughout the manuscript, but a restructuring to analyze AH and non-AH time periods separately has not been done. See comments below.*

1) The six stations included in this work have two types of filter based absorption instruments: the "reference" instrument (CLAP, PSAP, or MAAP) and the Aethalometer model AE31. The AE31 attenuation data are corrected with the Arctic specific correction factor from Backman et al. (2017). The same Cf (= 3.20) is used to correct the AE31 data from the six observatories. Absorption data collected with the AE31 at 550 nm are presented in this manuscript. The absorption at 550 nm is calculated using the absorption Angstrom exponent (AAE) calculated from the 7-λ Aethalometer measurements. The authors show that the comparison between absorption from the "reference" instrument and the AE31 is "imperfect and variable among stations".

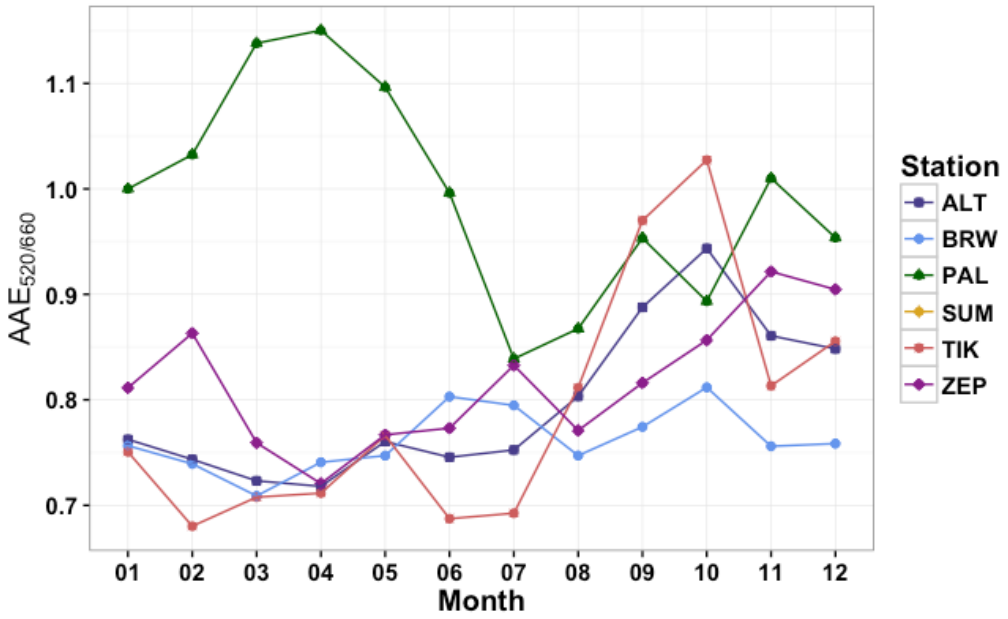
Why not present the absorption measurements from the "reference" instruments rescaled to 550 nm using the AAE from the AE31 instruments? If CLAP, PSAP or MAAP are considered as "reference" instruments, then data from these instruments should be presented in the manuscript. Moreover, the same C_f is applied to the seven absorption measurements from the AE31 instruments thus meaning that the AAE from uncorrected AE31 data and the AAE from corrected data should be approximately the same.

> *The reference absorption instruments at the stations were used to formulate an average best-guess correction factor for Arctic Aethalometers so that data from the same instrument could be used across the stations. Using homogeneously corrected Aethalometer data at all sites has the advantage of the data being more inter-comparable. From Backman et al. (2017): "The benefit of having the same type and model of instrument is that measurement artefacts for the same type of instrument would be expected to be more similar than between different types of instruments. The comparison of aerosol properties between different sites should be more robust when all sites have the same type of instrument than if the instruments would differ from site to site… For the sake of inter-comparability, a relative normalization factor is introduced to harmonize the determination of the absorption coefficient at the Arctic stations."*

2) Alternatively, if the authors think that the AE31 data are sufficiently robust to be presented in the manuscript (note that the supplemental material is not provided by the authors, so it is difficult to evaluate the goodness of the corrected AE31 data (and consequently SSA values)), then they should take more advantage of the multiwavelength absorption measurements from AE31 instruments.

Is there any specific/interesting feature in the AAE seasonality at the six stations? Why not study the spectral dependence of the single scattering albedo (SSA)? For example, presenting the SSA not only in the green, but also in the UV and near IR? The variability of these two quantities (AAE at least) should be discussed in the manuscript.

> *Our preference is to use Aethalometer data, as we feel these measurements are most robust and inter-comparable between stations. As for studying SSA in the UV and the near IR, this is not possible as our aerosol scattering coefficient measurements from the nephelometers are not made in the UV and the near IR.*
>
> *AAE values were indeed calculated for each site, and the data were originally included in the manuscript; however, there was not necessarily a specific or interesting feature in the AAE seasonality that was worth including. In order to shorten length of the manuscript, the AAE analysis was cut from the final version. There was little coherent seasonal signal, and differences between most sites was small (a spread of AAE values between 0.7 and 1.1 is actually quite small). For these reasons, the AAE analysis was not included in the manuscript. Included below, however, is the AAE seasonality plot and original description to further motivate this response.*



From a previous version of the manuscript in which AAE was included: "Absorption Ångström Exponent (AAE) climatologies have not previously been reported for stations in the Arctic. Statistics of AAE at the 520/660 nm wavelength pair, calculated using corrected Aethalometer absorption coefficients, are presented in Table 2. AAE values are not available at SUM, since SUM only has measurements from a 1 wavelength Aethalometer AE16. Two of the Arctic stations, PAL and TIK, have notable seasonality in AAE values. PAL has highest AAE values in the spring, and lowest AAE values in the fall. TIK, on the other hand, has lower AAE values in the spring and early summer, and higher values of AAE in the fall. These changes in AAE statistics throughout the year suggest that these sites might measure different aerosol compositions depending on the season; however, the range in AAE values is fairly minimal."

3) In the manuscript the Arctic Haze (AH) phenomenon is discussed together with the scattering and absorption measurements. ALT, BRW, TIK, and ZEP stations present an increase in both scattering and absorption in late winter/spring related to the AH phenomenon. However, there is no mention to the AH phenomenon in the sections presenting the intensive aerosol optical properties.

a) A table presenting the mean SAE, SSA, g (and possibly AAE) during AH period versus non-AH period should be presented and discussed. The spatial differences (from one site to another) in the intensive optical properties during AH period should be also discussed. For example, the seasonality of scattering and absorption at ALT, BRW, TIK and ZEP is very similar (and ascribed by the authors to the AH phenomenon) whereas the intensive properties are very different. For example, the SSA at ALT during AH is much higher (and different in term of seasonality) from the SSA observed at TIK during the AH period. The authors should comment/discuss the possible reasons explaining why the intensive properties change from one site to another during AH period.

We agree that more discussion of differences between intensive aerosol optical properties during the Arctic Haze season and the rest of the year is needed, and additions have been made to the manuscript. No additional table has been added to document the different

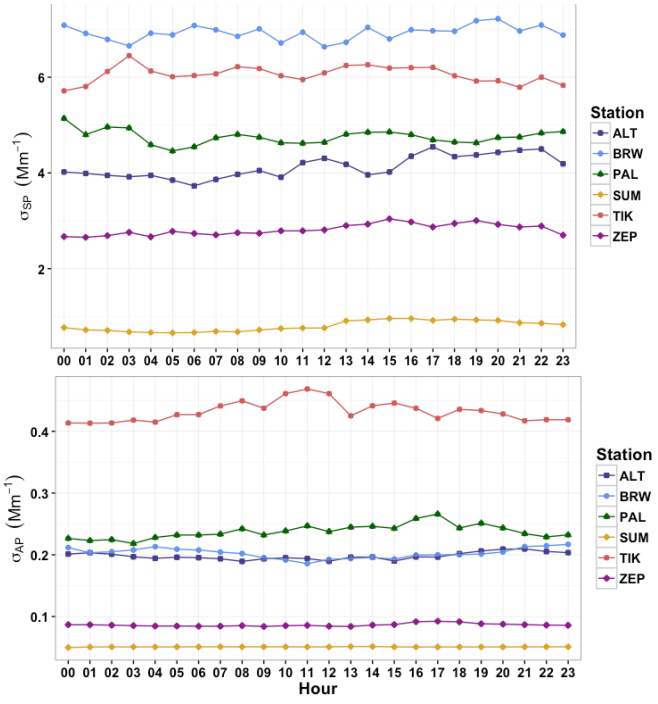
statistics between the Arctic Haze and non-Arctic Haze seasons, as the authors feel the differences between these time periods is easy to see on the monthly climatology plots in Fig. 3-7, and additionally, the focus of the manuscript is not to differentiate specifically between the AH and non-AH seasons but to document the seasonality throughout the entire year.

b) It is interesting the fact that the effect of AH on intensive properties is not observed at PAL and SUM which are located at higher altitude compared to the other stations. Is there any relationship between altitude of the station and AH phenomenon?

You are right that the fact that PAL and SUM do not show a large Arctic Haze season signal in their intensive aerosol optical properties is interesting; it might be worthwhile to dig deeper into the relationships between altitude of a measurement station and the AH phenomenon. However, there are not enough high elevation vs. low elevation observatories to say with much certainty whether or not differences between stations is due to elevation alone or other environmental differences. This may be a question more appropriately answered with a modeling study.

4) The authors say that "..surface Arctic aerosol optical properties in particular can help define and constrain inter-annual, seasonal and diurnal variability" (Pag. 2, Line 22-23). Why not present the diurnal cycles of both extensive and intensive aerosol particle optical properties? This can be done comparing AH period versus non-AH period.

We did perform a short analysis of diurnal variability during the preparation of the manuscript, though we felt the results were not worth including in this paper in particular. Since diurnal variability is not large at most stations, there was not much to comment on with regards to station to station differences in diurnal variability. We have addressed your comment by removing "diurnal variability" from page 22, line 22-23. See below for plots of hourly climatologies of aerosol absorption and scattering coefficients.



5) Improve the abstract. In the present form the abstract present a list of lowest/highest values of extensive and intensive properties at the six observatories, but the reasons/speculations behind the variability of the reported values is missing.

The authors do not feel comfortable including any speculations in the abstract, as we do not directly in this analysis provide evidence that explains most of the seasonality. The important finding of this paper is in the spatio-temporal variability of surface aerosol optical properties in the Arctic.

6) Pag. 8, Line 31. Figure 2 shows the time series of monthly median corrected AE31 data. Why not present the daily median? Note also that the supplemental material was not uploaded. Consequently, it is very difficult to evaluate the goodness of the comparisons using just monthly medians.

Our apologies that the supplemental material was not uploaded, it was made available on the ACPD portal soon after you noticed this. Comparing daily medians from the AE31 made for a noisy and difficult-to-read plot, whereas the plot of monthly medians conveyed the overall comparison in a more concise way, without losing the essential information. Furthermore, monthly medians have the distinct benefit of a much larger signal to noise ratio compared to daily medians, which as you point out in later comments, is especially important when making measurements in clean Arctic conditions.

7) Pag. 9, Line 33: The authors should explain where the data came from. For example, was it downloaded from EBAS. Or was it provided by data providers?

In the Data Availability section at the end of the manuscript, it is stated that all data used in the article are archived and accessible from the EBAS database. This sentence has also been added to the manuscript.

Page 9, line 33: "All data used in this analysis are archived and accessible from the EBAS database operated by the Norwegian Institute for Air Research (NILU)."

8) Pag. 10, Line 8: Add that also g was one of the variables considered in the manuscript.

Thank you for catching this. We have added g to the list of variables in this paragraph.

Page 10, lines7-8: The variables analyzed here include extensive aerosol optical properties that depend on aerosol amount, absorption (σ_{ap}) and scattering (σ_{sp}) coefficients and asymmetry parameter (g), ...

9) Pag. 10. How were the intensive properties calculated? Using all the scattering and absorption data or using only data above a given threshold (i.e. >1 Mm^{-1})?. Calculating the intensive properties using scattering or absorption data higher than a given threshold is important in order to remove undesired noise in the calculations.

Thank you for pointing this out. Given the incredibly small absorption and scattering coefficients measured at the Arctic sites, the typical thresholds of, say, >0.5 Mm^{-1} for absorption coefficients and >1 Mm^{-1} for scattering coefficients are not used in this analysis because it would eliminate a very large portion of the data measured at these sites. During some months (e.g., summer months when very low scattering coefficients are measured), that would exclude nearly all of the data.

Given the clean conditions in the Arctic, removing undesired noise in the data was accomplished through temporal averaging. The authors understand that computing intensive aerosol optical properties with low values of extensive aerosol optical properties can be problematic, and thus that is one reason why monthly medians are presented throughout the paper, as the authors have more confidence in a larger signal-to-noise ratio of monthly values compared to hourly or daily values. Furthermore, it was noted throughout the paper (page 14 line 31- page 15 line 1) that when intensive properties show large variability it is likely in part due to some noise from low scattering and/or absorption measurements.

For example: In Figure 2 the SSA at ALT and SUM in July and September, respectively, presents the lowest values when also scattering and absorption are low. The same is observed for the scattering Angstrom exponent at SUM in winter or the asymmetry parameter at SUM in January (for example).

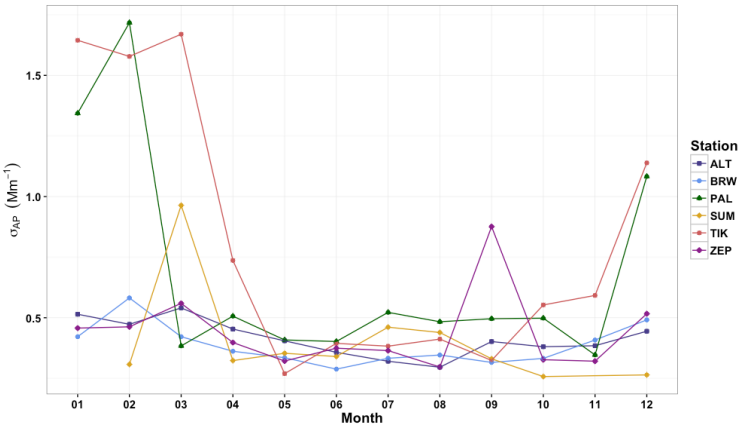
Furthermore, non-physical values of SSA (i.e., SSA<0 or SSA>1), for example, were removed from computations of monthly and annual statistics.

How do these figures (Figures 5, 6, 7) change if a threshold is applied before calculating the intensive properties? In the case of SSA at SUM in September the authors speculate that the low SSA is related to an increase in flights and transportation activity. However, for other stations/seasons no explanations are given to justify why the 5th and 95th percentiles are too low or high. It is important to demonstrate that these high deviations of intensive properties at some stations are not due to noise.

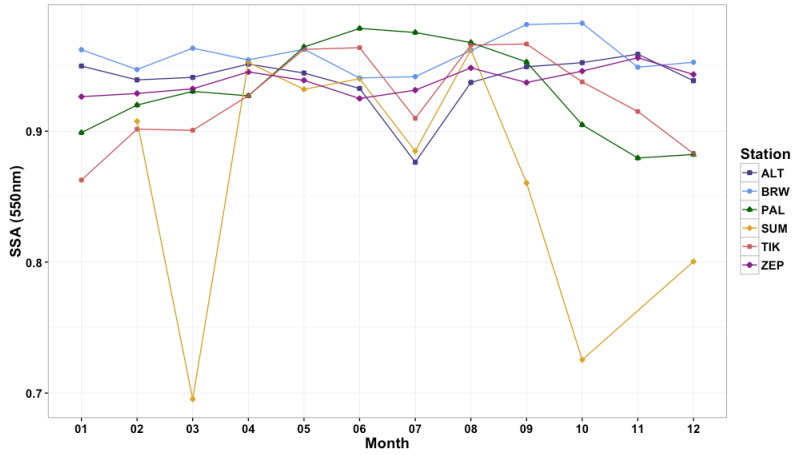
Figures 5, 6, and 7 would not necessarily be representative of typical Arctic conditions if a threshold was applied before calculating the intensive properties, as typical thresholds used in this type of aerosol optical property analysis (>0.5 Mm^{-1} for absorption coefficients and >1 Mm^{-1} for scattering coefficients), since median values of extensive aerosol optical properties at many of these Arctic stations are much below these thresholds. The authors discussed this problem in preparation of the manuscript, and decided it would not be prudent to remove data below these thresholds as it would eliminate so much of the dataset. Instead, noise was reduced by temporal averaging to monthly medians, then care was taken to remove intensive aerosol optical property values that were non-physical, for example, SSA values below 0 or above 1. The number of intensive aerosol optical property values removed with this 'threshold' was small.

For the purposes of considering this reviewer comment further, the authors administered slightly less strict thresholds (>0.25 Mm^{-1} for absorption coefficients and >0.5 Mm^{-1} for scattering coefficients) and remade the figures to see how they would change. With this threshold, much of the data points were removed. For ALT, this left only 31% of the original data points. The percentage of data points remaining after applying the thresholds at other stations was as follows: 19% (BRW), 24% (PAL), 4% (SUM), 9% (TIK) and 15% (ZEP).

Aerosol absorption coefficient medians with thresholds (see figure below), for example, do not look very different from those medians without thresholds. This finding is robust for Figures 4-7; though scattering medians do get higher when filtering out all scattering measurements below 0.5 Mm^{-1}.



Single scattering albedo medians computed after scattering and absorption coefficient thresholds are applied are shown in the figure below. At all stations with the exception of SUM, there is little change compared to medians without thresholds. The exception is SUM, which shows much lower SSA values than without the thresholds applied, particularly for the months of March, September, and October. Bear in mind that with the absorption and scattering thresholds applied, only 4% of the data points at SUM remained. In some months (January and November), this means there is no intensive data left at SUM at all. In the months that do have intensive optical property data available, the values are highly skewed towards what could be considered as 'polluted' events at this otherwise very clean site. This arguably skews SSA values to much lower than they 'normally' are at SUM, and therefore we argue that the seasonality shown below, when thresholds are applied, is not representative of typical conditions at these sites, especially SUM.



10) Pag. 10, Equation 2: Why not present the differences between the SAE calculated between 450 and 550 nm and the SAE calculated between 550 and 700 nm? Is there any interesting difference between the two SAE during the AH phenomenon versus periods without AH phenomenon?

This is an interesting question that could be worth exploring in another analysis, but due to time constraints, will not be investigated here.

11) Pag. 10. The AAE from AE31 was used to calculate the absorption at the same wavelength of the "reference" instrument. How was the AAE calculated? Were used all the wavelengths or only those close to the reference wavelength?

Thank you for asking us to clarify this. AAE was calculated with the following equation:

$$AAE = -\frac{\log(\sigma_{a1}) - \log(\sigma_{a2})}{\log(\lambda_1) - \log(\lambda_2)}$$

All of the wavelengths were not used. For this analysis, AAE values were calculated with the 520 nm and 660nm wavelength pair, the pair closes to the reference wavelength of 550 nm. This detail has been added to the manuscript.

Page 11, lines 9-10: "Absorption measurements were adjusted to the same wavelength with the AAE value calculated from the 520 nm/660 nm wavelength pair."

Moreover, (end of Pag. 10 – beginning of Pag. 11), the authors say that the SAE was also used for the wavelength adjustment of nephelometer data. However, the TSI nephelometer works at 550 nm which is the wavelength used to present the results. So, no adjustment of nephelometer data is in principle needed. Please, clarify.

Thank you for catching this. You are right, only data at 550 nm was in the end presented in the manuscript, therefore we have removed this sentence from the text.

12) Pag. 13, Line 6. PAL -> SUM

We still mean to say PAL here, but have added a clarifying term to indicate that we mean PAL has the highest absorption coefficients in the summer compared to the other Arctic stations.

Page 13, line 6: …PAL has the highest absorption coefficients during the summer compared to the other stations.

13) Pag. 13, Lines 16-18: Explain why at ALT the SSA values drop during July (any physical explanation or noise?)

*Upon further investigation of the ALT data in July, it is clear that there are many days of very low scattering measurements in that month that contribute to the low SSA values. This analysis alone cannot provide certainty about a physical explanation for why SSA could be so low. However, as mentioned on page 16 in lines 22-24 which comments on the systematic variability in Figure 8: "*The SSA vs. scattering relationship here suggests that whiter aerosols are preferentially scavenged such that darker aerosol remain at the lowest aerosol loadings (lowest scattering coefficients)".

For the purposes of addressing the reviewer comments, the threshold of $\sigma_{ap} > 0.25$ Mm^{-1} and $\sigma_{sp} > 0.5$ Mm^{-1} were applied to the ALT data. When the thresholds are applied, only 8,379 observations out of the total 26,304 aerosol observations made at ALT remain (only 89 observations in July). SSA was then calculated using only measurements that were greater than the thresholds, and values of SSA from filtered and non-filtered data were compared during July at ALT. For July in ALT with no thresholds applied to the data, the median SSA=0.90, while data with thresholds applied give a median SSA=0.87. The median of SSA value is slightly less with thresholds applied compared to data without thresholds.

14) Pag. 13, Line 18: Explain why the SSA values at BRW are the highest during September-October.

The high SSA average at BRW during this time is likely in part due to the minimum sea ice extent and thus increased open ocean and sea salt aerosol during September and October. Figure 10 lends some evidence towards this. Furthermore, at least one other publication supports this hypothesis (May et al., 2016), and thus a sentence speculating on this has been added to the manuscript.

Page 13, line 22-24: "SSA values at BRW could be highest in September and October due to low sea ice extent, more open ocean and thus the potential for more sea salt aerosol in the area (May et al., 2016). Figure 10 lends evidence for this, and is discussed later in the manuscript."

15) Pag. 13, Lines 22-24 ("This is explained by …….. is low and scattering is high"). Remove the sentence. This is obvious.

This sentence has been removed.

16) Pag. 13, Line 25 and Lines 27-28: The high scattering at PAL in summer is probably due to the enhanced formation of BSOA. This is probably consistent with the fact that absorption does not show the same increase in summer. Consequently, the SSA is the highest in summer (with quite low standard deviation of the data) and reflects the presence of very "white" particles. However, the authors say (Line 25) that there is an increased contribution from continental air masses in the summer at PAL. So, what is driving the evolution of the extensive and intensive properties at PAL in summer? The arriving of continental air masses (probably containing less "white" particles) or the BSOA formation (Lines 27-28)?

The seasonality of aerosol optical properties at PAL is very likely a combination of multiple factors, including an increase in continental air masses arriving at the station, a decrease in anthropogenic sources like wood burning (given higher summer temperatures, residential heating in Europe is no longer needed), and an increase in biogenic secondary organic aerosol formation. Chemical measurements and smaller scale trajectory analyses are needed to fully answer this question. This could be another manuscript in itself, and is likely worth exploring!

17) Pag. 14, Lines 24: Also here it is important to demonstrate that the large variability in SAE in July-September at TIK (when scattering and absorption are very low) is not due to noise. It is important to know if any threshold has been applied before calculating the intensive properties.

See also the answer to reviewer comment #13. In the manuscript, no thresholds were applied to extensive properties before calculating the intensive properties because this would eliminate so much of the available data. However, for the purposes of addressing the reviewer comments, the threshold of $\sigma_{ap} > 0.25\ Mm^{-1}$ and $\sigma_{sp} > 0.5\ Mm^{-1}$ were applied to the TIK data. When the thresholds are applied, only 2,444 observations out of the total 26,304 aerosol observations made at TIK remain. Intensive properties were then calculated using only data that were greater than the thresholds, and values of SAE, for example, were compared to filtered and non-filtered data during summer months at TIK. For July in TIK with no thresholds applied to the data, the median $SAE_{450/700nm} = 1.71$, while data with thresholds applied give a median $SAE_{450/700nm} = 1.74$. The medians of SAE values are nearly the same with or without thresholds, and this finding is robust across summer months at TIK and across intensive aerosol optical properties.

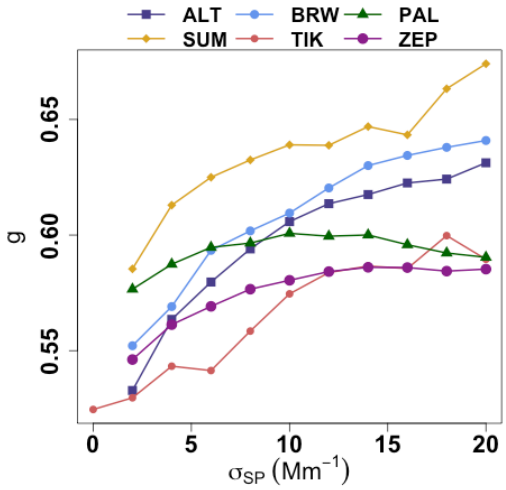
It is also worth mentioning that it has been acknowledged elsewhere in the paper (page 14 line 36 – page 15 line 1) that large variability in intensive properties tends to be concurrent with periods of low scattering and/or absorption measurements.

18) Pag. 14, Line 32: It seems that g also varies quite a lot from one station to another, whereas the authors say that "the asymmetry parameter, g, is similar for all sites except for SUM". Please, clarify/expand.

The authors stated that "the variability of the asymmetry parameter, g, is similar for all site except for SUM". This was meant to communicate that all stations except for SUM show the same general seasonality with larger values of g in the winter and smaller values of g in the summer.

19) Pag. 15, Line 7 and Figure 8: Why not show the g too?

Systematic variability of g with the other aerosol optical properties was explored (see one of the plots below), though nothing of particular interest was found in those plots, and thus they were not included in the manuscript.



20) Table 2: SUM station registers the highest SAE (small particles) and also the highest g (large particles). Any explanation for this?

The authors agree this is a bit of an enigma, but since g also depends on shape and composition in addition to size, this suggests shape and composition are likely playing a role here. We cannot with certainty from the analysis here what could explain this. Aerosol size distribution measurements and size-segregated chemical composition measurements could help with answering this question, but neither is available at SUM.

21) Section 4.3: Figure 10 is nice. It seems that there is a relationship between the time spent above open water and sea ice and Figures 3 and 4. For example, at TIK the scattering is the lowest when air masses spent more time over sea ice and open water (June to September). At ZEP the reduction of scattering between June and October reflects the relative increase of time spent over sea ice and open water (and less time spent over land). Can the authors say something more about Figure 10? Is it possible to relate the time spent over land with the Arctic haze phenomenon? The paragraph at Pag. 18, Lines 16-29 should be expanded.

We have taken your comments into consideration and have expanded the discussion throughout the paper to include more results from Figure 10.

Figure 9 seems less useful. The highest frequency is always observed for regions close to the stations. Why not use, i.e., the potential source contribution function or the concentration weighted trajectory? (Both are available for example in the OPENAIR r package). These plots could be colored by levels of scattering and absorption to get a clearer idea about source regions. The differentiation in terms of air masses between AH periods versus non-AH periods should be introduced and discussed.

We would argue that Figure 9 is useful in that it shows highest frequency is mostly symmetric around the station locations. For most stations, a preferential trajectory path does not jump out in either summer or winter. This result, although somewhat boring, is still valuable to know.

Your suggestion about concentration weighted trajectories is a good one, and concentration weighted trajectories were indeed already plotted during the process of this analysis. They showed interesting results, and are already being included in a forthcoming publication in preparation. Thus, those results are not shown here.

New References:

May, N. W., P. K. Quinn, S. M. McNamara, and K. A. Pratt (2016), Multiyear study of the dependence of sea salt aerosol on wind speed and sea ice conditions in the coastal Arctic, J. Geophys. Res. Doi.org/2016JD025273.