Review – ACPD – Tunved and Ström

General comments:

This was a very thorough paper, and generally it looks like a strong study. The authors have obviously put a great deal of work into collecting and analyzing this unique dataset, and in my opinion it is worth publishing, pending that the below concerns are addressed (most of which are fairly minor). Future work could benefit from comparison with other datasets or models.

Specific comments (most important):

1. In section 4.5, the authors explore possible variations in transport patterns, but they did not mention how possible changes in wet deposition and cloud processing might also influence the aerosol concentrations, sources, and distributions. I understand that cloudiness and precipitation may concurrently be changing over the Arctic (e.g., see Morrison et al. (2018), Bintanja and Selten (2014)), although I am not sure about at or upwind of the Aspvreten site in particular. The authors might consider adding some information on this in the introduction, and discussing how changes in cloudiness and precipitation might impact the interpretation of their results.

2. I suggest the authors re-visit the text in the following places to make sure they are not over-emphasizing the certainty in how the results should be interpreted.

    - 2.3: Can the authors please address whether the clustering assumptions have the potential to have a major impact on the findings, and if so, whether any sensitivity analyses were conducted to make sure that the findings are robust?
    - P. 8, l. 32: "Absence of small particles signifies the lack of photochemistry." There can't be any other explanation?
    - Figure 5: Is it possible that strong differences in the amount of light at different hours, depending on season, would skew or bias the results presented in Figure 5? Are the results similar when separated be season?
    - The cluster history discussion in section 3.2: The authors make reasonable assumptions about the important atmospheric processes each cluster was exposed to, based on theoretical suppositions about the factors that drive aerosol size distributions. However, up to this point, the data presented are entirely based on size distributions alone with no other supporting data. I would suggest making it clearer in this section that the processes causing the aerosol size distributions are merely hypothesized (unless the authors can present or discuss other datasets supporting these suppositions further).
    - P12 l.1: "The presence of the two modes is suggestive of two different processes acting independent on the size distribution." This statement seems to imply to me that there are not other interpretations as well. Is that what was intended here, and if so, why?
    - Figure 14. I am wondering, and presumably other readers may also wonder, what the implications are for the authors assuming cloud droplet activation in warm clouds at such high latitudes, where ice-containing clouds are very common,

particularly during the winter. Can the authors comment on whether their results might be less applicable or have greater uncertainty during the winter periods? Can the authors provide any information on how common liquid-only clouds are in this region during the different periods of the year?

- Section 4.5 This part of the analysis assumes that the HYSLPIT back trajectories are correct, and equally correct, at each location and time. I am skeptical about whether this is a good assumption, as the Arctic is a generally poorly validated region. As such, it is unclear whether the results are meaningful, although the fact that signals are small might still be worth showing. I recommend either removing this section, moving it to a supplement, or adding substantially more discussion of the uncertainties.
- P. 17, l. 9: "The data has been analyzed from several different aspects in order to shed light on the degree, when and where changes in emissions (natural or anthropogenic) may have had an impact on the observed aerosol size distribution." The authors have only back trajectories to assess the sources of the aerosols in this study. To not oversell the study, I suggest removing the "(natural or anthropogenic)" part of this sentence.
- P. 13, l. 28: "This means a reduction of mass of roughly 52 % during the studied period." I recommend re-wording this sentence to reflect the uncertainties in this statement. One suggestion: "Based on this calculation, we estimate that mass was reduced by roughly 52 % during the studied period"
- P. 18, l. 1-8: How would changes in precipitation impact these findings, if at all? Please briefly discuss.

3. There were several places where the methods were insufficiently described. The authors should please provide the following information:

    - Methods: Readers are pointed to outside references for basic information about the sampling. To avoid readers having to go look up a different paper to interpret this one, it would be helpful to at least mention in the text whether these are ground-based data, at what altitude the data were collected, whether there were any potential upwind sources of contamination, and if so, what was done to avoid sampling these sources.
    - The trends the authors describe are based on a single instrument, if I understand the methods correctly. Can the authors please mention whether any measures were taken to rule out the possibility that the observed long-term trends are caused by errors in the instrument calibration? (Presumably the instrument was independently calibrated, but it may be useful to compare the data to other long-term satellite or ground-based remote sensing data in the region (e.g., MODIS or AERONET data) as well?)
    - Tables 1-2: Please define GSD and Dg in the caption and in the text as well. Also, how is modal diameter and concentration defined, specifically?
    - Fig. 7: The caption is confusing because there is only one panel in the figure; please rephrase. The figure is also confusing because the axis on the right labelled "Trend d(dN/dlogDp)" is in reverse order (5 to -50, bottom-to-top) compared to the axis on the left labelled "dN/dlogDp", which goes from 0 to 3250, bottom-to-

top). It would help readers if the plot were standardized so that both axes were going in the same direction. Please indicate in the text how the confidence intervals were calculated.

- Figure 8. Please indicate how the confidence interval for the trends were calculated, and which of the plotted trends are significantly different from zero. To address the significance, the authors might consider obtaining a confidence interval around the slope for each subset of the data, similar to what was done in Fig. 7 (preferably a bootstrapped confidence interval, as that would be valid even if the data don't meet all the assumptions of a normal linear regression analysis).
- P. 12, l. 33 and Table 3 – It may be helpful to describe (or at minimum, reference) the seasonal Kendall test used here, for those readers who are not familiar with it, and to explain why this test was chosen.
- Figure 9. Please explain in the text how the confidence intervals for the slope were calculated. Please indicate in the caption what the whiskers indicate (range of the data? SD? Etc.)
- Figure 10. Please indicate in the caption what the whiskers indicate.
- Figure 11: Please indicate in the caption what statistics are being presented in the boxes and whiskers. For example, that median, upper and lower quartiles, and some kind of confidence interval?

Specific comments (more minor):

In the abstract, the authors might consider more clearly stating the extent to which this work is new and different from previous published work on this dataset, and more general reasons why the work is important.

P.3 l. 23: “Clearly there is a lack of long-term aerosol microphysical data from which trend analysis has been reported or even can be reported because of too short measurement periods or interrupted data sets.” To clarify, can the authors please state whether there are similar observations in other locations, and/or what specific region the above statement is referring to?

Fig. 6: please add the location of the sampling site to this map.

P. 12, l. 2: “As sulfur is the particle precursor that has decreased the most …” please cite what information this statement is based on.

P. 16, l.2: “However, our analysis above also emphasize that the apparent trends are not the same over the whole year.” Apparent trends in what?

p. 17, l. 15: “The general trend found in this study is well in agreement with the findings presented by Asmi et al. (2013)….” Please mention here where the Asmi et al. study took place.

References

Bintanja, R. and Selten, F. M.: Future increases in Arctic precipitation linked to local evaporation and sea-ice retreat, Nature, 509(7501), 479–482, doi:10.1038/nature13259, 2014.

Morrison, A. L., Kay, J. E., Frey, W. R., Chepfer, H. and Guzman, R.: Cloud Response to Arctic Sea Ice Loss and Implications for Future Feedback in the CESM1 Climate Model, Journal of Geophysical Research: Atmospheres, 124(2), 1003–1020, doi:10.1029/2018JD029142, 2018.