

**Interactive comment on “Measurements of aerosol and CCN properties in the Mackenzie River delta (Canadian Arctic) during Spring-Summer transition in May 2014” by Paul Herenz et al.**

**Author answer to Anonymous Referee #2**

We thank Referee 2 for taking the time to review our manuscript and for giving valuable hints and suggestions. Below, comments from the referee are given in blue while our answers are given in black, with passages including new text given in italic. In the revised version of the text, new text is printed in bold, while text to be deleted is crossed out.

General Comments:

The manuscript by Herenz et al discusses an interpretation of data collected at a ground site in Tuktoyaktuk, Canada, in association with aircraft data during May 2014. This time period is a ‘shoulder season’ in the Arctic, where the spring ‘Arctic Haze’ influence is waning and the atmosphere is beginning to take on summer-like characteristics. The paper discusses two types of air mass influences observed at Tuktoyaktuk, which the authors have called ‘spring-type’ and ‘summer-type’. The spring-type air has characteristics similar to Arctic Haze and the summer-type air has an added influence from Aitken mode particles. The measurements and their analysis are technically sound, and data filtering has been presented appropriately. Some of the conclusions and interpretations of the data are confusing or incomplete in the opinion of this reviewer, and have been pointed out below. In addition, the labeling of ‘spring-type’ and ‘summer-type’ is perhaps generous (particularly ‘summer-type’) as it is not shown that the source region or processes shaping the size distribution during the study period are similar to those in the heart of the summer season (July-August). It is perhaps prudent to label the Aitken-mode containing PNSDs as simply ‘N. Pacific’, since the study does well to describe the highly time-resolved oscillations in air masses during near the spring-summer transition, and correlates this with one of two seasonal maxima in methanesulfonic acid (MSA) (Leaith et al., 2013; Quinn et al., 2007). As a curious point, the authors performed this research in the Canadian Arctic, yet have not cited much of the existing research that has been conducted in the region, so the suggested or argument-supporting citations provided in this review have been skewed slightly to correct for this fact.

We agree to your general comments, and will elaborate on that more below at your respective remarks. In short, what we did was to change the labeling of the air masses to “accumulation-type” and “Aitken-type” throughout the text. Also, several additional publications are now being cited.

This paper could be accepted to Atmospheric Chemistry and Physics pending major revisions to the discussion and interpretation of the data.

Specific comments:

The abstract of the paper highlights a ‘rapid transition from Arctic spring to summer that took place during the measurement period.’ This claim is difficult to support directly with the extent of analysis that took place in this study. Such a claim would be more amenable to inter-seasonal data that are analyzed at high temporal resolution – such a study does not exist to the knowledge of this reviewer – but the fact that two types of air masses influence this site is certainly supported within the manuscript. One longer period of focused analysis in the present manuscript indicates an oscillation between the so-called ‘spring’ and ‘summer’ influences on the scale of hours-to-days (Period 1, Figure 6b). Characterizing this oscillating influence as a rapid transition is perhaps technically accurate in some sense; it is also misleading, as the abstract leads the reader to believe that the site will change singularly from ‘spring’ to ‘summer’ like a step function. The presented data suggests this is not occurring, and rather the measurement site is alternately influenced by two different source regions (one of which is debatably ‘summer-like’), so it is recommended that the abstract be rephrased.

We agree with your comment here and deleted the remark concerning the “rapid transition from Arctic spring to summer”. This part of the abstract has been changed to:

*"Basic meteorological parameters and particle number size distributions (PNSDs) were observed and two distinct types of air masses were found."*

The introduction of the paper is quite well written and clear, with some very minor exceptions noted in the detail comments below.

Overall, the measurements presented in the study were analyzed systematically, clearly, and with substantial attention to quantitative detail, including a comprehensive error analysis. Both the size distribution data and the CCN data were used to appropriate lengths, considering uncertainties. The authors should be commended for working with this challenging data set collected in a difficult measurement environment.

We thank the reviewer for these positive and encouraging remarks.

The authors invoke cloud processing as the source of the bi-modal character of the PNSD\_C3 size distribution (Page 13, lines 20-24) and this is, indeed, one possibility; however, different explanations that are equally supported by the measurements (as presented) are possible. The interpretation of the factors driving the shape of PNSD\_C3 has broader consequences within this study, so this is quite an important point. In addition to the cloud processing argument presented by the authors for the bi-modal character of PNSD\_C3, possible explanations include:

1. The PNSD\_C3 average size distribution may be a mixture of two types of particles: newly formed particles in the Aitken mode and aged particles in the accumulation mode. Evidence for a mechanism to produce such a scenario has been provided by detailed analysis of size distributions from remote locations in the Arctic (e.g., Collins et al., 2017) wherein new particles may be forming on different days where growth of the existing population occurs simultaneously, causing particles to grow into the accumulation mode size range.

2. The PNSD\_C1 mode sizes are quite similar to those of PNSD\_C3, and it has been posited that Period 1 has a mixed character of spring-like and summer-like influences. Could there be additional source variability within Period 3 that gives rise to the bimodal character? Indeed on page 17-18, the authors discuss the vertical structure of air masses, suggesting that during Overflight 3, spring-like Arctic air resides above a surface inversion and could mix down into the surface layer. This mechanism would produce a size distribution with characteristics similar to those observed in PNSD\_C3.

3. Recent studies at Utqiagvik, AK have shown the strong influence that fossil fuel extraction and processing facilities can have on the aerosol in this region (Gunsch et al., 2017; Kolesar et al., 2017) and could contribute to the shape of PNSD\_C3 since the Period 3 trajectories pass over Northern Alaska.

Based on your suggestions we discussed more carefully. However, the bottom line for the observed minimum to occur is that there are at least two types of aerosol with different sources, where accumulation mode particles might usually be assumed to be the more aged ones. For all of the cases you listed, it could still be that particles with larger sizes got a large fraction of their particulate matter by cloud processing, followed by mixing the air mass with a second air mass that contained newly formed particles (as suggested in your point 2). Collins et al. (2017) unfortunately only show one case where there was a particle growth event in parallel to new particle formation and only mention that there were more similar events, while it is also mentioned that the conditions during the measurements often were foggy, so that cloud processing could have occurred in the fog. But still, summarizing your and our arguments, we added to following:

*"While cloud processing is a well known process for gaining particulate matter and growing particles to larger sizes, particles can also grow by generation of particulate matter directly from the gas phase as described recently for Arctic conditions in e.g., Willis et al. (2016), Burkart et al. (2017b) and Collins et al. (2017). The observed minimum in the PNSD occurs when new particle formation takes place, either by adding small particles to an already aged air mass or by mixing of different air masses with one air mass containing aged and the other one newly formed particles, where one could come from aloft. It should also be mentioned that it was recently described in Gunsch et al. (2017) and Kolesar et al. (2017), that emissions from Prudhoe Bay oil field, which is located at the northern shore of Alaska roughly 700 km west of our measurement location, influenced Arctic PNDSSs by adding both high concentrations of small particles and*

*particulate mass to larger particles. Summarizing there is a number of reasons that can add to the observed bi-modality of the size distribution, but small, comparably newly formed particles will make up the observed Aitken mode in all cases.”*

If uncertainties in the CCN data were smaller, the kappa values of each mode may have provided useful in constraining these four options (one provided by the authors plus three above). Can the authors constrain the system further using any supplementary data or analysis that is not already provided?

We tried all that was possible with the data we have available to get better constrained values, and what is presented here is what possibly can be done. We don't have any supplementary data. Indeed, we regard it as an important result to show the uncertainties in kappa values based on measurement uncertainties. The uncertainty in the CCN measurements and inferred kappa values are based on uncertainties in counting and sizing (size distribution) as well as supersaturation (CCNc). As we used state of the art measurement technology, it can be expected that similar measurement uncertainties will always influence kappa retrievals.

The authors conclude in Section 3.5 that the shape of the PNSD is driven by transport and that local sources are not active in shaping the distribution. This is somewhat antithetic to the behaviour of canonical summertime Arctic aerosol which are thought to have a substantial contribution from new particle formation (Asmi et al., 2016; Croft et al., 2016; Tunved et al., 2013) based on sources within the polar dome via isentropic transport, which the authors discuss. Wet deposition processes (e.g., nucleation and precipitation scavenging) play a major role in shaping the size distribution in summer as well (Browse et al., 2012; Croft et al., 2016), which is not discussed or considered in the manuscript. Recent studies from the Canadian Arctic suggest that transport in heart of the summer is either from the higher Arctic latitudes or from the Canadian boreal forest (Collins et al., 2017; Mungall et al., 2016; Sharma et al., 2012; Wentworth et al., 2016), rather than from the North Pacific.

Critical question: Can the authors provide a stronger connection between the characteristics of the summer Arctic atmosphere and the Period 3-type air masses outside of the presence of the Aitken mode? Should the 'summer-type' (Period 3) air masses be thought of as truly characteristic of summer, or are these specific to a transient seasonal transport pathway from the N. Pacific?

We agree with your argument that Arctic aerosol in summer might differ from that we observed, although the size distributions are similar. Therefore, throughout the whole text, we renamed the types of the air masses, now focusing on what we observe, i.e., they are called accumulation-type and Aitken-type now.

The following was also added to the introduction: *“Indeed, these precipitation related scavenging processes, which are effective from late spring throughout the summer, were shown to be one of the drivers of the yearly cycle in Arctic PNSDs (Browse et al., 2012; Croft et al., 2016a). Resulting low number concentrations of particles in the accumulation mode size range enable new particle formation (NPF).”*

It is also stressed stronger in the text in Section 3.3, that: *“Sources for the precursor gases forming these particles will differ from spring to summer, as mentioned above (Li et al., 1993).”* – where Li et al. (1993) has been cited in the previous version of our manuscript (and still is) as: *“The earlier maximum [of MSA] that occurs around the time of our measurements, can be associated with long-range transport from marine source regions from the North Pacific (Li et al., 1993).”*

Page 3, lines 14-15: The authors claim a lack of in-situ measurements of CCN in the Arctic region. Recently, CCN and cloud property studies from the NETCARE campaigns have documented aspects of aerosol-cloud interactions, including airborne estimations of activation diameters and chemical effects on CCN activity, specifically in the Canadian Arctic (Burkart et al., 2017; Leaitch et al., 2016; Willis et al., 2016). Other studies, including those referenced in the manuscript, have studied CCN concentrations and properties in other regions (e.g., Moore et al., 2011).

Thank you for pointing out these recent publications to us. We removed the sentence claiming a lack of in-situ measurements and added the following to our text in the paragraph on CCN:

*“Within the NETCARE project based on summer time measurements in the Canadian Arctic Archipelago, high concentrations of newly formed particles were observed particularly in the marine boundary layer and*

above clouds (Burkart et al., 2017a). One particle growth event measured during NETCARE was described in Willis et al. (2016), showing newly formed particles growing to sizes above 50 nm, subsequently being able to activate to cloud droplets at 0.6% supersaturation. For the same project, Leaitch et al. (2016) examined cloud droplet number concentrations for 62 cloud samples and reported that particles with comparably small diameters, below 50 nm, activated to cloud droplets in 40% of all cases.”

Page 8, lines 11-13: Since this study was conducted in the Canadian Arctic, it is worthwhile to compare with seasonal aerosol concentrations at Alert, Canada and Utqiagvik (Barrow), AK. If the authors desired to make a more pan-Arctic comparison, inclusion of observations made at Station Nord, Greenland (Nguyen et al., 2016) and Tiksi, Russia (Asmi et al., 2016) should also be discussed.

It is correct that we measured in the Canadian Arctic while the work by Tunved et al. (2013) to which we refer in details was done on Svalbard. We therefore now also added a comparison with literature done in other parts of the Arctic, where also number concentrations and size distribution similar to those observed by us are described. The following two larger paragraphs were added to the introduction, together with a few minor additions:

*“Indeed, these precipitation related scavenging processes, which are effective from late spring throughout the summer, were shown to be one of the drivers of the yearly cycle in Arctic PNSDs (Browse et al., 2012; Croft et al., 2016a). Resulting low number concentrations of particles in the accumulation mode size range enable new particle formation (NPF). The latter is also based on the presence of MSA (methane sulfonic acid), an oxidation product of DMS (dimethyl sulfide) that is emitted by the oceans (Quinn et al., 2007; Leaitch et al., 2013), with increasing emissions related to the decline of the Arctic sea ice cover (Sharma et al., 2012). Additionally, ammonia, also a contributor to NPF, was described to be connected to seabird colonies by Croft et al. (2016b) and Wentworth et al. (2016) and was discussed to have a far ranging influence on the Arctic aerosol.”*

*“Croft et al. (2016a) reported data collected in the years 2011 to 2013 from Mt. Zeppelin, i.e., examining different years than Tunved et al. (2013), together with additional data from Alert, Canada. Both yearly cycles of NCN and PNSDs were similar at Alert and Mt. Zeppelin, and also similar to those discussed in Tunved et al. (2013). Croft et al. (2016a) suggest that the observed similarities at these two stations, which are 1000 km apart, and between the different years examined at Mt. Zeppelin indicate the existence of an annual cycle that spans the high Arctic. This assumption is strengthened by Nguyen et al. (2016), reporting again comparable yearly cycles of number concentrations and PNSDs for Villum Research Station in northern Greenland, only differing in more pronounced Aitken modes in the summer month. The shape of the yearly cycle of NCN and the most often occurring PNSDs observed at Tiksi, Russia, described in Asmi et al. (2016), were again similar to those observed at Mt. Zeppelin and Alert. However, number concentrations were higher in general in Tiksi, and NPF events occurred more readily, which is discussed to be related to regional continental sources of nucleating and condensing vapors. Generally, a comparison of PNSDs presented in Freud et al. (2017) from Alert, Villum, Mt. Zeppelin, Tiksi and Barrow (Alaska) shows some differences between Arctic sites due to local effects, but concludes that on a large scale there is a pronounced annual cycle in PNSDs with common features, with all Arctic sites sharing the Asian side as the main large-scale source region of accumulation mode aerosols.”*

*“Within the NETCARE project based on summer time measurements in the Canadian Arctic Archipelago, high concentrations of newly formed particles were observed particularly in the marine boundary layer and above clouds (Burkart et al., 2017a). One particle growth event measured during NETCARE was described in Willis et al. (2016), showing newly formed particles growing to sizes above 50 nm, subsequently being able to activate to cloud droplets at 0.6% supersaturation. For the same project, Leaitch et al. (2016) examined cloud droplet number concentrations for 62 cloud samples and reported that particles with comparably small diameters, below 50 nm, activated to cloud droplets in 40% of all cases.”*

Page 10, lines 9-10: It is difficult to see the size distribution results as a corroboration of the prior discussion of N\_CNraw, since this is the same data shown in a different manner. On page 9, the N\_CCN and N\_CNraw>150nm were shown and noted as being correlated since the smaller particles are due to pollution.

Figure 5 is simply a re-statement of the same information since the accumulation mode size is 150 nm. It is likely that the choice of 150 nm as a cutoff for the metric in Figure 4 was deliberate, and the data do certainly tell a convincing story about the difference between polluted and 'filtered' data, but these figures represent one piece of evidence, rather than two separate, corroborating pieces of evidence. If other types of techniques distinguished polluted from non-polluted data (chemical or physicochemical observations), then these would corroborate the number size distribution differences between polluted and filtered cases as presented.

We apologize as there is a misunderstanding in what we had intended to say. The corroboration we refer to here was connected to the literature we cited before, i.e., referring to Wegener et al. (2012), who also had seen modes between 10nm and 40nm in median urban PNSD caused by traffic, domestic and district heating, comparable to our results. We rephrased this part to avoid this misunderstanding. It now is:

*"The observations by Wegener et al. (2012) support our assumption ... "*

Page 11, lines 19-20: When discussing the Period 3 air masses, no discussion on similarity to prior studies is provided to support the notion that these air masses are indicative of what might be found in the actual summer months at Tuktoyaktuk. On page 13, lines 17-20, a statement about corroborating evidence for N. Pacific sources of air is made in reference to trends in MSA – but it is not shown that this type of air mass is truly typical of Arctic 'summer' as labeled.

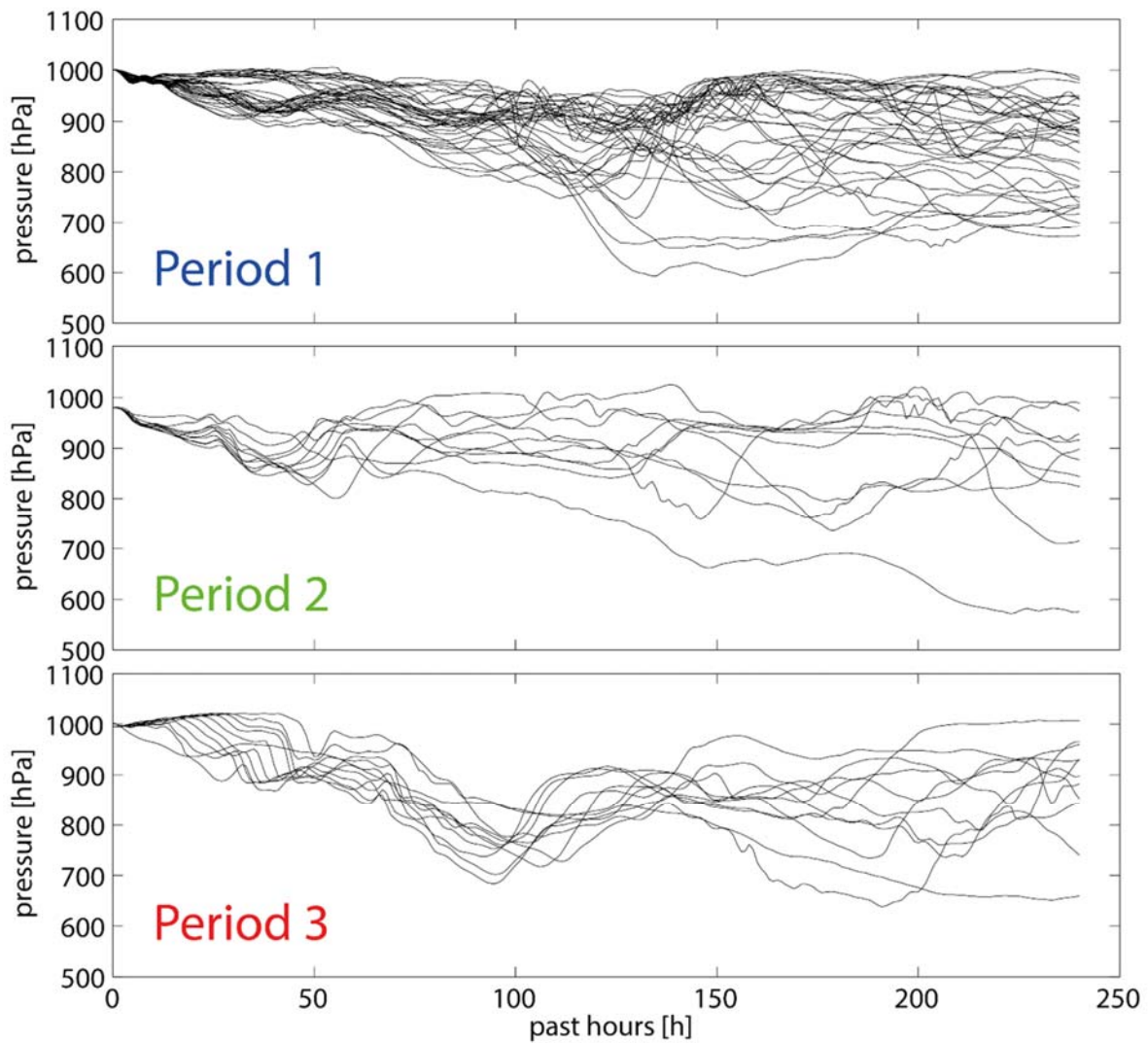
We agree, and as mentioned above, we renamed the air masses to accumulation-type and Aitken-type and stress stronger that precursor gasses have different sources and source regions in spring and in summer.

Figure 6: It would be helpful to understand the behavior of the ensemble of back trajectories in the vertical dimension to some degree, so that impacts of surface sources along the trajectory could be understood. Is it possible to include some information on the vertical position of the trajectory or each ensemble of trajectories as a function of time or position?

We agree, that information about the vertical distribution of the air masses could provide further information. To get an idea about the vertical behavior of the back trajectories of the three periods we plotted the pressure along the trajectory in the figure below. For a better overview we only used the trajectories that started right at the location of Tuktoyaktuk (we also started 12 additional trajectories in a close proximity of Tuktoyaktuk per time step to account for uncertainties). It is obvious that the trajectories span a wide vertical range, especially for periods extending further back in time. For Period 1 and 2 one can not really obtain any conclusive information used for further interpretation. Only for period 3 it can be argued that the air masses are in the free troposphere at 100 h back in time, while some of the trajectories got back to the boundary layer (roughly between 150 and 240 h back in time). From this it could be assumed, that the air masses of period three have the closest touch to the ground in the area of the North-West Pacific and Siberia.

We decided not to include this plot in the manuscript as we consider the level of additional information low and inconclusive.





Technical Corrections:

Page 1, line 4: “: : were indicative for the rapid: :” change to “: : were indicative of the rapid: :”

Page 8, line 11: change to “time periods where the measurements”

Page 9, lines 8-10: These two sentences are hard to follow. It is recommended that the sentences be re-phrased, especially when referring to “it is associated with lower sampling statistics”, but the prior sentence refers to two different metrics.

Thank you! The suggested corrections have been made to the manuscript.

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