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 #1

We thank Referee 1 for taking the time to review our manuscript and for giving 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.

This paper presents aerosol size distribution and CCN data from a three week groundbased campaign in the Western Canadian Arctic. The campaign was conducted in the springtime during which it is known that there is a transition from springtime to summertime aerosol conditions. The findings of the paper are that both sets of aerosol distributions (i.e. summer and spring) can be inferred from the data, and the hygroscopicity parameter was observed to be about 0.2. The strength of the paper is that few aerosol measurements have been conducted in the Arctic and so this adds to the data base of such measurements. A weakness is that there is no new conceptual idea presented in the paper.

While currently there is a large amount of publications on Arctic aerosol, most of this (even publications from the last two years of which a number is now additionally included in the manuscript, see literature list below) deals with summer time aerosol and often focuses on new particle formation. Hence, while some data exists also on Arctic aerosol throughout the year, the herein presented data observed in spring, including also directly measured concentrations of CCN and the particle hygroscopicity parameter derived from measurements, adds new data to the still scarce database. Similarly, the comparison of simultaneously taken ground based and airborne measurements, presented for all overflights made during the campaign, adds new and valuable information to the discussion concerning the connection between ground based and airborne conditions. Also, the uncertainty analysis concerning the hygroscopicity parameters, which shows limitations of what can be learned by the respective kind of data, is new and valuable on its own, beyond its applicability to Arctic aerosol.

This paper would represent a more significant contribution if more vertically resolved measurements were presented, and so I am puzzled why the POLAR6 aircraft data are not presented more comprehensively given that the plane was probably flying more frequently that only during the overpass periods. I suggest that these data be added to the revised paper.

The work presented here is based on the measurements done by the main author who, together with some of the co-authors, is a member of the TROPOS cloud group. The TROPOS cloud groups' role during RACEPAC was to contribute ground based measurements, and these are analyzed herein, together with some aircraft data that others were willing to share with us and that were taken by these other groups during times of overflights.

The idea of the ground based measurements was to have a continuous characterization of the aerosol on ground, as such a continuous record naturally cannot be obtained by aircraft measurements. Additionally, in this study, all existing size distribution data taken during overflights were used to compare ground based data with aircraft data. Other aircraft data will be published by others and a further inclusion of them is beyond the scope of this work.

Also, the paper should be careful to not claim to have characterized the transition from spring to summer aerosol. That can only be done at a fixed location if a full annual cycle of aerosol parameters is observed, ideally over many years. During a short campaign, the best one can hope to observe are snapshots of different aerosol distributions from different sources. While the authors distinguish their air masses into spring and summer-types, I believe they could just have been easily characterized the periods as continental and marine. For publication, it has to be justified that the general trajectory pattern displayed in Period 2 is

indeed characteristic of springtime conditions at this location, i.e. do the size distributions during this period have the character they do because they are more like those in the spring or because they are of continental origin?

Thank you for pointing this out. We agree and have changed the naming of the two types of aerosol to "accumulation-type" and "Aitken-type" throughout the manuscript and also changed the wording in the text concerning the "transition" accordingly where needed.

The size distribution of Period 2, which is mono-modal and of "accumulation-type" is typical for the Arctic spring aerosol (shape and integrated number) and compares well with size distributions measured by others at different Arctic sites during this season e.g., Freud et al. (2017), Tunved et al. (2013). The trajectories of Period 2 show the advection of air masses with the origin in the central arctic rather than the North American continental region. These facts, i.e., shape and integrated number of the size distribution as well as the origin of the air masses, are clear indicators for the presence of air masses that are typical for the Arctic during spring. The main characteristic of the Arctic aerosol during spring is that most significant sources are inside the Arctic dome (e.g., biomass burning and industry at the Asian continent) and particles have a long residence time due to a lack of precipitation, and released aerosol particles are distributed widespread over the Arctic. To make this clearer in the manuscript, we have added this passage in the introduction:

"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."

Also we added a new section showing the results of the Potential Source Contribution Function (PSCF) to identify regions contributing to high N_{CN} measurements. Even though this analysis is not done specifically for Period 2, it shows that especially the former mentioned regions in Siberia ("Asian site") may significantly influence the aerosol at Tuktoyaktuk.

Lastly, although the paper is improved compared to the originally submitted version in how it references past work, it is still lacking references to past characterization of the CCN behavior of marine aerosol (currently, only one reference from the authors is presented) and Arctic aerosol. As well, there is a large suite of aerosol size distribution measurements from North American sites (e.g. work by Leaitch et al in Elementa, and Croft et al., Collins et al. and Burkart et al. in ACP) that is ignored and is arguably more relevant than the more geographically distant (but referenced) measurements at Svalbard. Lastly, there is the recent paper by Freud et al. in ACP that comprehensively describes aerosol character across the Arctic. Given that the merit of the current paper is that it adds new measurements to those already performed, it is necessary that the new measurements be presented alongside what has already been reported in the literature. The literature is more comprehensive that what is listed above; I only included recent publications.

We included an additional list of publications and hope this suffices your requirements. There is certainly more, however, as this work is not a review, adding more is beyond the scope of this work. To account for your suggestions we added the following paragraphs 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). 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 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."

To the size distribution section:

"*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). In general, 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 PNDSs 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."*

and to the CCN section:

"*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 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."

Here you find the list of references, that were added to the manuscript to respond of your suggestions:

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