Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-745-RC1, 2017

Atmospheric Chemistry and Physics Discussions

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.

Anonymous Referee #2

Received and published: 21 September 2017

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) (Leaitch 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.

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 presented 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. 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.

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 bi-

modal 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.

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?

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?

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).

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.

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.

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.

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?

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.

References

Asmi, E., Kondratyev, V., Brus, D., Laurila, T., Lihavainen, H., Backman, J., Vakkari, V., Aurela, M., Hatakka, J., Viisanen, Y., Uttal, T., Ivakhov, V. and Makshtas, A.: Aerosol size distribution seasonal characteristics measured in Tiksi, Russian Arctic, Atmos. Chem. Phys., 16(3), 1271–1287, doi:10.5194/acp-16-1271-2016, 2016.

Browse, J., Carslaw, K. S., Arnold, S. R., Pringle, K. and Boucher, O.: The scavenging processes controlling the seasonal cycle in Arctic sulphate and black carbon aerosol, Atmos. Chem. Phys., 12, 6775–6798, doi:10.5194/acp-12-6775-2012, 2012.

Burkart, J., Willis, M. D., Bozem, H., Thomas, J. L., Law, K., Hoor, P., Aliabadi, A. A., Köllner, F., Schneider, J., Herber, A., Abbatt, J. P. D. and Leaitch, W. R.: Summertime observations of ultrafine particles and cloud condensation nuclei from the boundary layer to the free troposphere in the Arctic, Atmos. Chem. Phys., 17, 5515–5535, doi:10.5194/acp-17-5515-2017, 2017.

Collins, D. B., Burkart, J., Chang, R. Y.-W., Lizotte, M., Boivin-Rioux, A., Blais, M., Mungall, E. L., Boyer, M., Irish, V. E., Masse, G., Kunkel, D., Tremblay, J.-É., Papakyriakou, T., Bertram, A. K., Bozem, H., Gosselin, M., Levasseur, M. and Abbatt, J. P. D.: Frequent Ultrafine Particle Formation and Growth in the Canadian Arctic Marine Environment, Atmos. Chem. Phys. Discuss., 2017.

Croft, B., Martin, R. V., Leaitch, W. R., Tunved, P., Breider, T. J., D'Andrea, S. D. and Pierce, J. R.: Processes controlling the annual cycle of Arctic aerosol number and size distributions, Atmos. Chem. Phys., 16(6), 3665–3682, doi:10.5194/acp-16-3665-2016, 2016.

Gunsch, M. J., Kirpes, R. M., Kolesar, K. R., Barrett, T. E., China, S., Sheesley, R. J., Laskin, A., Wiedensohler, A., Tuch, T. and Pratt, K. A.: Contributions of transported Prudhoe Bay oil field emissions to the aerosol population in UtqiaÄąvik, Alaska, Atmos. Chem. Phys., 17, 10879–10892, doi:10.5194/acp-17-10879-2017, 2017.

Kolesar, K. R., Cellini, J., Peterson, P. K., Jefferson, A., Tuch, T., Birmili, W., Wiedensohler, A. and Pratt, K. A.: Effect of Prudhoe Bay emissions on atmospheric aerosol growth events observed in UtqiaÄąvik (Barrow), Alaska, Atmos. Environ., 152, 146–155, doi:10.1016/j.atmosenv.2016.12.019, 2017.

Leaitch, W. R., Sharma, S., Huang, L., Toom-Sauntry, D., Chivulescu, A., Macdonald, A. M., von Salzen, K., Pierce, J. R., Bertram, A. K., Schroder, J. C., Shantz, N. C., Chang, R. Y.-W. and Norman, A.-L.: Dimethyl sulfide control of the clean summertime Arctic aerosol and cloud, Elem. Sci. Anthr., 1(1), 17, doi:10.12952/journal.elementa.000017, 2013.

Leaitch, W. R., Korolev, A., Aliabadi, A. A., Burkart, J., Willis, M. D., Abbatt, J. P. D., Bozem, H., Hoor, P., Köllner, F., Schneider, J., Herber, A., Konrad, C. and Brauner, R.: Effects of 20–100 nm particles on liquid clouds in the clean summertime Arctic, Atmos. Chem. Phys, 16, 11107–11124, doi:10.5194/acp-16-11107-2016, 2016.

Moore, R. H., Bahreini, R., Brock, C. A., Froyd, K. D., Cozic, J., Holloway, J. S., Middlebrook, A. M., Murphy, D. M. and Nenes, A.: Hygroscopicity and composition of Alaskan Arctic CCN during April 2008, Atmos. Chem. Phys., 11(22), 11807–11825, doi:10.5194/acp-11-11807-2011, 2011.

Mungall, E. L., Croft, B., Lizotte, M., Thomas, J. L., Murphy, J. G., Levasseur, M., Martin, R. V., Wentzell, J. J. B., Liggio, J. and Abbatt, J. P. D.: Dimethyl sulfide in the summertime Arctic atmosphere: measurements and source sensitivity simulations, Atmos. Chem. Phys., 16(11), 6665–6680, doi:10.5194/acp-16-6665-2016, 2016.

Nguyen, Q. T., Glasius, M., Sørensen, L. L., Jensen, B., Skov, H., Birmili, W., Wiedensohler, A., Kristensson, A., Nøjgaard, J. K. and Massling, A.: Seasonal variation of atmospheric particle number concentrations, new particle formation and atmospheric oxidation capacity at the high Arctic site Villum Research Station, Station Nord, Atmos. Chem. Phys., 16(17), 11319–11336, doi:10.5194/acp-16-11319-2016, 2016.

Quinn, P. K., Shaw, G., Andrews, E., Dutton, E. G., Ruoho-Airola, T. and Gong, S. L.: Arctic haze: Current trends and knowledge gaps, Tellus, Ser. B Chem. Phys. Meteorol., 59(1), 99–114, doi:10.1111/j.1600-0889.2006.00238.x, 2007.

Sharma, S., Chan, E., Ishizawa, M., Toom-Sauntry, D., Gong, S. L., Li, S. M., Tarasick, D. W., Leaitch, W. R., Norman, A., Quinn, P. K., Bates, T. S., Levasseur, M., Barrie, L. A. and Maenhaut, W.: Influence of transport and ocean ice extent on biogenic aerosol sulfur in the Arctic atmosphere, J. Geophys. Res. Atmos., 117(D12), D12209, doi:10.1029/2011JD017074, 2012.

Tunved, P., Ström, J. and Krejci, R.: Arctic aerosol life cycle: linking aerosol size dis-

tributions observed between 2000 and 2010 with air mass transport and precipitation at Zeppelin station, Ny Alesund, Svalbard, Atmos. Chem. Phys, 13, 3643–3660, doi:10.5194/acp-13-3643-2013, 2013.

Wentworth, G. R., Murphy, J. G., Croft, B., Martin, R. V, Pierce, J. R., Côté, J.-S., Courchesne, I., Tremblay, J.-É., Gagnon, J., Thomas, J. L., Sharma, S., Toom-Sauntry, D., Chivulescu, A., Levasseur, M. and Abbatt, J. P. D.: Ammonia in the summertime Arctic marine boundary layer: sources, sinks, and implications, Atmos. Chem. Phys, 16, 1937–1953, doi:10.5194/acp-16-1937-2016, 2016.

Willis, M. D., Burkart, J., Thomas, J. L., Köllner, F., Schneider, J., Bozem, H., Hoor, P. M., Aliabadi, A. A., Schulz, H., Herber, A. B., Leaitch, W. R. and Abbatt, J. P. D.: Growth of nucleation mode particles in the summertime Arctic: a case study, Atmos. Chem. Phys., 16(12), 7663–7679, doi:10.5194/acp-16-7663-2016, 2016.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-745, 2017.