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Key Points:

- An extended analysis of marine biogenic new particle formation and growth dominating summer Arctic background conditions is presented
- Direct measurements of low aerosol hygroscopic growth factors (HGFs) dominate the aerosol ultrafine population— aerosols are organic rich
- Elevated cloud condensation nuclei concentrations occurred when Arctic background biogenic aerosols—secondary in origin—were detected

Supporting Information:

- Supporting Information S1

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Large Summer Contribution of Organic Biogenic Aerosols to Arctic Cloud Condensation Nuclei

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Abstract A k-means cluster analysis of a 5-year aerosol particle size distribution data set from northeast Greenland is combined with measurements of coincident shorter field studies of aerosol equivalent black carbon (eBC) content, hygroscopic growth factor (HGF), and cloud condensation nuclei (CCN) concentrations. This led to five clusters strongly controlled by natural emissions (eBC 8–15 ng/m³) and three anthropogenic clusters with larger particle concentrations in the accumulation mode (eBC 29–77 ng/m³). The HGF and CCN properties of the eight aerosol clusters differ drastically. Anthropogenic clusters feature high growth factors (1.62–1.81) and low CCN κ values (0.10–0.46), while natural clusters show lower HGF (1.38–1.70) but remarkably higher κ values (0.35–0.51). Extrapolating the CCN properties on the basis of the cluster analysis to annual timescales suggests that biogenic organic aerosol may drive Arctic aerosol production during summer.

Plain Language Summary The variability of the warming Arctic—particularly the decreasing sea ice extent—is likely to boost the control of the ocean on the atmospheric composition. Key factors controlling Arctic aerosol-climate interactions are sources, chemical transformations, and removal mechanisms of aerosols and their precursor gases. However, knowledge on the composition and sources of summer aerosols is insufficient. To tackle this problem, we use an approach where aerosol particle size distributions, aerosol equivalent black carbon, hygroscopic growth factor, and cloud condensation nuclei measurements at Villum Research Station within the period 2012–2016 are synergistically combined. Our results suggest that marine sources of new particle formation and growth dominate summer Arctic background conditions. These aerosols are organic rich in nature. Elevated cloud condensation nuclei concentrations occurred when secondary Arctic biogenic organic aerosols were detected.

1. Introduction

The impact of aerosol particles on the formation and microphysical properties of clouds is one of the largest remaining uncertainties in climate change projections (Carslaw et al., 2013). The cloud condensation nuclei (CCN) population depends on the ambient aerosol particle size distribution (PSD) and corresponding chemical composition (Farmer et al., 2015). Isolating and quantifying contributions from natural versus anthropogenic sources to the total aerosol is important to understand the climate system and to estimate human impacts on climate change (Hamilton et al., 2014). In the Arctic, aerosol-cloud-climate interactions depend on complicated combinations of high surface albedo and strong seasonal cycles in aerosol concentration, size, and composition. Winter and spring show accumulation mode-dominated aerosols (Lange et al., 2018; Tunved et al., 2013). By profound contrast, nucleation and Aitken mode particles dominate the number size distribution during summer. Natural boundary layer aerosols are emphasized to dominate the summer population relative to aerosols transported from continental sources (Dall'Osto et al., 2017; Heintzenberg et al., 2015; Leaitch et al., 2013).

One of the main gaps in our knowledge on the aerosol indirect effect in the Arctic is the origin of these natural Arctic aerosols, although there is some consent that marine and snow or ice-related sources are main candidates. For example, primary ultrafine aerosol particles—possibly related to marine polymeric gels produced by phytoplankton and sea ice algae biological secretions—can be transferred by bubble bursting from the sea-air interface into the polar atmosphere (Leck & Bigg, 2005; Orellana et al., 2011). However, a number of studies have also reported in situ formation of new aerosol particles via secondary processes in the Arctic. A main mechanism involves the formation of new particles from emissions of biogenic volatile

species (Burkart et al., 2017; Dall'Osto, Simo et al., 2018; Fu et al., 2013; Heintzenberg et al., 2015; Mungall et al., 2017; Tunved et al., 2013). Overall, the contribution of marine open ocean sources to aerosols—such as secondary sources related to under-ice algae communities—is expected to increase, given that the summer-ice coverage is decreasing due to Arctic warming. By contrast, primary blowing snow- and ice-related sources will decrease. Indeed, we recently reported a categorization of the aerosol population via cluster analysis of aerosol number size distributions taken at Villum Research Station (VRS; at Station Nord) in North Greenland during a 5-year record (2012–2016). We detected a number of new particle formation (NPF) events and those were associated with air masses that had previously traveled over open water and melting sea ice regions. We suggested that marine related biological activities within the open leads in the pack ice and/or along the melting marginal sea ice zone may be responsible for such events (Dall'Osto, Geels et al., 2018). Such results are fully supported by a similar study carried out from the period 2000–2010 at the Zeppelin Mountain Station (Dall'Osto et al., 2017). Despite the importance of the composition and sources of summer aerosols, there is currently also a lack of understanding, which unidentified aerosol growth precursors are present in this high-latitude marine environment. It should be kept in mind that—besides for emissions of NPF gas precursors associated with sympagic and pelagic biological communities (Dall'Osto, Beddows, et al., 2017; Dall'Osto, Ovadnevaite, et al., 2017; Lévassieur, 2013)—other biogenic sources may exist, including seabird colonies (Croft et al., 2016; Weber et al., 1998) and intertidal zones (Allan et al., 2015; Dall'Osto, Simo, et al., 2018). A comprehensive set of exciting new observations is being reported in the context of “NETwork on Climate and Aerosols: addressing key uncertainties in Remote Canadian Environments” (NETCARE), suggesting a key role for Arctic marine secondary organic aerosol in the Canadian Arctic Archipelago (e.g., Croft et al., 2016; Willis et al., 2016, 2017; Köllner et al., 2017; Leaitch et al., 2018, Croft et al., 2018, and reviewed in Abbatt et al., 2019). Recently, a new marine source of oxygenated volatile organic compounds was identified (Mungall et al., 2017), possibly related to heterogeneous air-sea reactions of biogenic organic matter enriched in the sea surface microlayer (Agogu et al., 2005; Wurl et al., 2017; Zhou et al., 2014). A study conducted in the Canadian Arctic suggested that the condensing organic vapors responsible for particle growth are more volatile in the Arctic, compared to lower latitudes (Burkart et al., 2017); observations that nucleation mode particles (~20 nm) showed lower growth rates than larger particles (~90 nm) were reported. Organic-rich particles contributed significantly to Arctic boundary layer aerosol mass, and correlations were found between such particles and elevated CCN concentrations (Bigg & Leck, 2001; Willis et al., 2016, 2017). Motivated by the need to further understand sources of Arctic aerosols, this work combines results of Dall'Osto, Geels, et al. (2018), which focused on sources of Arctic nucleation mode aerosol, and Lange et al. (2018), which characterized Arctic accumulation mode aerosol. The present work adds measurements of aerosol hygroscopic growth factor (HGF) and extends the previous analysis of equivalent black carbon (eBC) and of CCN. We show—in our study region—that it is very likely that during summertime, natural ultrafine organic aerosols, secondary in origin, play a major role in determining the ambient CCN population in the Arctic atmosphere.

2. Materials and Methods

All measurements in this study were carried out at the VRS at the northeastern coast of Greenland. Located at 81.6°N, 16.7°W, 24 m above sea level it is among the northernmost permanent research stations in the world. Submicrometer aerosol PSDs (9–915 nm; Freud et al., 2017; Nguyen et al., 2016) recorded in the period 2012–2016 at a resolution of 5 min were averaged to 33,678-hourly distributions. The PSDs were recorded by a custom-built scanning mobility particle sizer equipped with a Vienna-type medium column (Wiedensohler et al., 2012). A k-means cluster analysis was performed on the entire PSD data set, yielding eight representative PSD clusters. The choice of clustering method was justified by a Cluster Tendency test (Beddows et al., 2009). The choice of eight clusters represented a best compromise of similarity between the individual PSDs within each cluster (Silhouette width, 0.43) and cluster separation (Dunn index, 1.6×10^{-3}). Our instrument (Freud et al., 2017; Nguyen et al., 2016) and clustering procedure (Dall'Osto, Geels, et al., 2018; Lange et al., 2018) is described in further detail elsewhere.

eBC was measured in May 2011 to August 2013, by measurements of light absorption coefficient with a Multiangle Absorption Photometer (Model 5012 Thermo Scientific). A specific absorption coefficient of $6.6 \text{ m}^2/\text{g}$ was used to infer the eBC mass concentration. Here we carry out a reanalysis of the eBC

measurements, which were first published by Massling et al. (2015). The instrument is described in detail in Petzold and Schonlinner (2004).

Aerosol water uptake at subsaturated conditions was measured during two field studies in 2016 (April–May and August, total 23 days), with a humidified tandem differential mobility analyzer (HTDMA) Model 3100 from Brechtel (Lopez-Yglesias et al., 2014). These in situ measurements yielded particle HGF for particles with initial dry Dp of 30, 60, 120, and 240 nm at 90% relative humidity (RH). Data inversion was based on principles in Stolzenburg and McMurry (2008) and performed with an Igor Pro® (Wavemetrics, Inc.) algorithm provided by Brechtel. The HGF yields an indirect measurement of particle chemical composition for those particle diameters, which are probed. The accuracy of the set RH was frequently monitored (every ~3 hr) with monodisperse ammonium sulfate aerosol (Massling et al., 2011).

Particle CCN concentration was measured during two field studies in 2016 (April–June and August–September, total 95 days), with a CCN counter (100) by Droplet Measurement Technologies (DMT) (Roberts & Nenes, 2005). Ten different supersaturations (SSs) in the range of 0.1–1.0 were cycled. CCN concentrations at the maximum reachable instrument SS (~1.8% SS) served as a control metric against total particle number concentrations. The ratio CCN_{max}/N_{25} (CCN_{max} being the concentration at this high SS and N_{25} the concentration of particles with $Dp > 25$ nm) was on average 0.93 over both field studies. This deviation likely indicates the presence of particles that were smaller than the smallest particles that activated at this high SS. Instrument SS was calibrated with monodisperse ammonium sulfate aerosol before, during, and after the studies. Critical CCN activation Dp (Dp_{crit} ; Juranyi et al., 2010; Kristensen et al., 2016) and κ hygroscopicity parameter (Petters & Kreidenweis, 2007) was determined for all CCN measurements. The measurement procedure, calculations, and calibration are in detail described elsewhere (Lange et al., 2018).

3. Results

The eight clusters resulting from our previous aerosol number size distributions k-means analysis and their corresponding relative occurrence throughout the year are shown in Figure 1. They are divided into five ultrafine clusters (particles <100 nm, Dall'Osto, Geels, et al., 2018) and three accumulation clusters (particles >100 nm, Lange et al., 2018). Briefly, the *Acc. Haze* cluster, with a clear accumulation mode at Dp of 173 nm, has a dominant occurrence during the months January–April. Also, the *Acc. Bimodal* cluster—composed of two modes, with significant contribution of an Aitken mode at Dp of 38 nm—appears during these months. During the period May–September the contribution of the *Acc. Haze* cluster vanishes and the clusters characterized by ultrafine particles (*Nucleation*, *Bursting*, *Nascent*, and *Bimodal*) dominate the aerosol population. In Oct the *Pristine* cluster has its highest occurrence; low particle number concentrations are often found during this month (Nguyen et al., 2016). The *Acc. Aged* cluster with the highest mean Dp accumulation mode of all clusters (213 nm) is most frequently found during months October–January.

From the eBC measurements conducted in 2011–2013, we determine average aerosol eBC concentration of each aerosol cluster (Figure S1 in the supporting information). Pristine conditions with very minor anthropogenic influence occur for all ultrafine clusters: Pristine, Nucleation, and Bursting categories have average eBC concentrations of 11 ± 3 ng/m³. The Nascent and Bimodal clusters show higher, although still generally low concentrations of 15 and 14 ng/m³, respectively. By contrast, accumulation mode clusters are associated with high eBC concentrations of >20 ng/m³. The *Acc. Haze* cluster has an eBC content of 77 ng/m³, while the clusters *Acc. Aged* and *Acc. Bimodal* are associated with 53 and 29 ng/m³ eBC, respectively. The increased eBC concentration of accumulation mode clusters corresponds well with previously determined anthropogenic influences on Arctic aerosol during winter and spring (Barrie et al., 1981; Heintzenberg, 1982; Law & Stohl, 2007; Quinn et al., 2007; Shaw, 1995; Sirois & Barrie, 1999). Oppositely, clusters with little eBC occur when the probability of anthropogenic influence is low in summer and autumn. This is due to the fact that during winter and spring, the polar front expands southward, including regions with emissions of atmospheric pollution in North America, Europe, Asia, and Russia (Bishop et al., 2006), whereas this direct entrainment is largely absent during summer and autumn (Iziomon et al., 2006). We adapt a threshold for anthropogenic influence on the aerosol clusters of >20 ng/m³ (Cavalli et al., 2004; Gogoi et al., 2016). Linking the k-means statistical-cluster analysis of submicrometer aerosol number size distributions with eBC measurements allows us to term the Nucleation, Bursting, Nascent, *Ultrafine bimodal*, and Pristine as

Figure 1. (a) Cluster particle size distributions found by k-means analysis. The top panels are ultrafine aerosol clusters (as reported in Dall'Osto, Geels, et al., 2018), while the bottom panels show accumulation aerosol mode clusters (as reported in Lange et al., 2018). (b) Monthly frequency of occurrence of all eight ultrafine and accumulation aerosol clusters.

originating from natural sources within the Arctic, while the Acc. Haze, Acc. Bimodal, and Acc. Aged aerosol clusters are associated to anthropogenic sources.

Hygroscopic growth and water uptake modifies particle size and the optical properties of aerosols, impacting both direct and indirect climate effects. Larger particle sizes increase scattering efficiency (Tang, 1996). The CCN concentration at a given SS is a function of particle number concentration, particle size, and particle hygroscopicity. Particle chemical composition drives hygroscopicity; hence, measurements of aerosol hygroscopic properties may help elucidating the contribution of a specific aerosol chemical type to the CCN population. Given the recent studies pointing toward organic aerosol in clean Arctic air being efficient CCN (Willis et al., 2016, 2017), measurements of HGFs and κ may elucidate the relative contribution of a variety of different aerosol categories to the overall CCN population.

Figure 2. Hygroscopic growth factor as determined by humidified tandem differential mobility analyzer measurements at 90% relative humidity. Uncertainty bars are standard deviations within the clusters. Clusters not mentioned in this figure (*Nascent* and *Acc. Aged*) did not occur during the humidified tandem differential mobility analyzer field measurements.

Therefore, first, average HGF, derived from coincident HTDMA measurements, was assigned to those five of eight k-means cluster occurring within the 23 days of the HTDMA measurement periods. In other words, only five of the eight k-means aerosol clusters characterized in this study temporally appeared during the two HTDMA measurement periods in 2016. Marked differences between the different clusters were found, as shown in Figure 2. The natural clusters Nucleation, Bursting, and Bimodal have low HGF at dry D_p of 30 nm (1.40–1.50). When

considering 60- and 120-nm dry D_p , the HGF of Nucleation and Bursting increases to 1.59–1.70, which is slightly lower than the anthropogenic clusters. The Bimodal cluster clearly distinguishes itself by maintaining a low HGF (1.38) at 60 nm and increasing to 1.56 at 120-nm dry D_p . We associate the smaller mode of the natural Bimodal cluster (about 50–60 nm, Figure 1a) to the grown aerosols originating from Arctic NPF, as discussed in Dall'Osto, Geels, et al., 2018. Hence, it can be drawn from Figure 2 that natural secondary NPF growth events are driven by about 30- to 60-nm aerosols with an HGF of about 1.4 ± 0.1 . These in situ aerosol measurements point to the fact that low hygroscopicity chemical compounds such as organics may be an important chemical component. To reach such low HGF, organic mass fraction relative to sulfate would need to be larger than 0.5 (Gysel et al., 2007). The low HGF could also indicate that the organic compounds are not highly aged (Duplissy et al., 2011; Massoli et al., 2010). The higher HGF at 60 and 120-nm dry D_p show a shift to more hygroscopic organics and/or an increased sulfate fraction, which implies a varying chemical composition over the entire size range under investigation. Also, we did only observe very limited multiple mode HGF (<2% of measurements), hinting toward little contributions from sea spray particles, which is in line with generally low wind speeds prevailing during the measurements. But while it is important to stress that inorganic iodine oxides could also contribute to particle mass at 30–60 nm (Bialek et al., 2012), as the HGF of the Nucleation cluster at 30-nm dry D_p is similar to 1.34 reported by Allan et al. (2015), the summertime iodine concentrations are too low to solely explain the observed NPF in the study region (Dall'Osto, Simo, et al., 2018). A different scenario is observed for the aerosols related to anthropogenic emissions. Clusters Acc. Haze and Acc. bimodal show HGFs of about 1.62–1.81 depending on dry D_p (Figure 2). We expect sea salt to assert only minor influence during the spring months (Huang & Jaeglé, 2017; Nguyen et al., 2014), although sea salt concentrations remain significant at Alert (Canada) in April and diminish in May (Huang & Jaeglé, 2017). This suggests that the particles mainly contain partly neutralized sulfates, an expected compound in aged Arctic aerosol (Swietlicki, 2000; Zhou et al., 2001). In summary, our HTDMA measurements point toward organics contributing significantly to natural ultrafine Arctic aerosols.

Additionally to the HGF analysis, average CCN properties for each PSD cluster are determined from 95 days of coincident CCN measurements in 2016. Figure S2a shows that the natural Nascent and Bimodal clusters feature higher CCN concentrations than the anthropogenic clusters above 0.3% SS, corresponding to larger concentrations in the Aitken mode. CCN concentrations are $42\text{--}95\text{ cm}^{-3}$ for $\text{SS} \leq 0.3\%$ and $85\text{--}150\text{ cm}^{-3}$ for $\text{SS} > 0.3\%$. Also, the Bursting cluster surpasses anthropogenic cluster CCN levels from about 0.45% SS, with concentrations of $34\text{--}85\text{ cm}^{-3}$ for $\text{SS} \leq 0.4\%$ and $95\text{--}157\text{ cm}^{-3}$ for $\text{SS} \geq 0.5\%$. By contrast, the three anthropogenic clusters feature CCN concentrations of $54\text{--}102\text{ cm}^{-3}$ over the entire range of 0.1–1.0% SS, with only a small increase in concentration toward higher SSs. A larger fraction of the anthropogenic cluster PSDs are CCN active at any given SS (Figure S2b), simply resulting from the fact that these clusters feature higher particle numbers at higher D_p . All natural clusters have $D_{p,\text{crit}}$ at about $60 \pm 5\text{ nm}$ (Figure S2c) at SS of roughly 0.4 to 0.5, in line with previous other Arctic studies (Zábori et al., 2015; Leaitch et al., 2016; Burkart et al., 2017; Herenz et al., 2018). The clusters pristine and nucleation feature low CCN concentrations (about 50 cm^{-3}) due to their in general low particle number concentrations (Dall'Osto, Geels, et al., 2018). Overall, the observed CCN concentrations associated with the clusters predominant during summertime at VRS (Figure S2a) are higher than those observed in the Arctic Ocean (Martin et al., 2011), while lower than the concentrations found during aircraft measurements of Arctic-sourced air masses over the Canadian Arctic (Latham et al., 2013). The associated κ value (hygroscopicity parameter) for each PSD cluster is shown in Figure S2d. The natural clusters have κ values from roughly 0.25 to 0.4 over the entire range of SS, which suggests a similar chemical composition for all particles in the range of the examined $D_{p,\text{crit}}$, from 40 to 110 nm. The fact that there is a trend in HGF measurements, with larger HGF for larger particles, while this is not seen in the κ values, might originate in the different measurement periods, as CCN measurements were made during a longer period than HTDMA measurements. Generally, the κ values obtained in this study are larger than reported in Burkart et al. (2017), who determined κ values up to 0.15 at 0.5% SS during a marine particle growth event, and both Martin et al. (2011) and Lange et al. (2018) who obtained κ value of the organic fraction of Arctic aerosol of <0.2. Using instrumentation comparable to that used in the present study, also Herenz et al. (2018) reported κ values between 0.18 and 0.28 for SS from 0.1% to 0.7% for different types of Arctic aerosol. Based on an analysis of uncertainties originating from the state of the art use of the instrumentation, it is concluded in Herenz et al. (2018) that within the reported range, κ values are not

Figure 3. Extrapolated monthly contribution of natural and anthropogenic clusters to total ambient cloud condensation nuclei concentration at 0.4% supersaturation.

significantly different. Similar uncertainties should apply for the present study. So we found larger κ values compared to other studies and will come back to that while discussing κ values from HGF and CCN measurements below.

By contrast to the natural clusters, the κ values of the anthropogenically influenced clusters decrease notably with increasing SS (values). This observation is consistent with recent single particle and size-resolved aerosol mass spectrometry of springtime haze near Alert (Willis et al., 2019). For example, the κ value of the Acc. Haze cluster decreases from 0.3 to below 0.10 over a decrease in Dp_{crit} from 132 to 71 nm, suggesting a change in composition with a higher organic fraction for the smaller particles. We propose that the smaller particles have experienced less aging than those at higher Dp_{crit} and therefore contain less sulfates relative to organic com-

pounds. The responsible mechanism could be wet phase chemistry while particles may have been activated to cloud droplets (Pandis et al., 1992), particle-phase oxidation (Farmer et al., 2015), higher oxidation level of the organic fraction (Massoli et al., 2010), or progressing deposition of sulfates (Giamarelou et al., 2016). While overall the observed trend of hygroscopicity with particle size is consistent with the increase of HGF with increasing Dp , seen in Figure 2, it is still striking that for small particles the natural clusters show lower HGF but higher κ values than the Acc. Haze or in fact any of the anthropogenically influenced clusters. As already mentioned above, a direct comparison between κ values from HGF and CCN measurements will be done below. A similar tendency of κ values with particle size as observed for Acc. Haze is also observed for the Acc. Bimodal and the Acc. Aged clusters, which, within the typical uncertainty for κ values, cannot be distinguished. In principle, the behavior of these clusters follows that of the Acc. Haze cluster; thus, the same higher relative content of sulfates at larger Dp is hypothesized. Given that the eBC content is significantly lower and the κ value generally higher than that for the Acc. Haze cluster, it is proposed that the Acc. Bimodal and Acc. aged clusters contain a larger proportion of naturally derived organic compounds, which are more hygroscopic than the eBC contained in the Acc. Haze cluster (Lange et al., 2018).

An additional observation made for the correlation between HGF and κ values from CCN measurements is presented in Figure S3. For that, HGF measured at both 85% and 90% RH were converted to κ values (κ_{HGF}) for a comparison to κ values from CCN measurements (κ_{CCN} , done at SS of 0.3%). A different picture emerges for the two seasons. During both seasons, κ_{CCN} are below κ_{HTDMA} , but this deviation is larger during spring, when κ_{HTDMA} are comparably high while κ_{CCN} are low. Petters and Kreidenweis (2007) give κ values for a range of different substances, and while often κ_{CCN} are above κ_{HTDMA} , it is the opposite for sulfuric acid (which, however, has high κ values close to 1) and for some organic compounds such as malonic and glutaric acid. In general, it is known that particularly Arctic spring aerosol is acidic throughout the troposphere (Fisher et al., 2011). Specifically for the vicinity of VRS, it was recently described that acidic aerosol is present in spring, explained by high amounts of sulfuric acid (Nielsen et al.,). On the other hand, κ values in literature, such as those discussed above, indicate a rather high content of organic aerosol. While we cannot provide a chemical characterization of the aerosol examined in this study, we suggest that in spring the high acidity (likely from sulfuric acid and possibly other unknown chemical compounds) together with the presence of organic components is responsible for the observed large deviation between low κ_{CCN} and higher κ_{HTDMA} . This deviation clearly diminished in summer, where particle acidity is lower (albeit not neutral) and different organic components likely prevail, due to different sources of aerosol precursors from spring to summer (Quinn et al., 2007).

Our data shown in Figure S3 suggest that different unidentified organic compounds are responsible for the deviation between κ_{HTDMA} and κ_{CCN} during spring and summer.

We extrapolate the CCN concentration of each cluster with its respective occurrence throughout the year. This process is based on the assumption, that each PSD cluster can be robustly tied to its associated CCN properties. Hereby, we obtain the projected monthly contribution of natural and anthropogenic clusters to total CCN concentration, over the year, shown in Figure 3. There is variation of the CCN concentration within the clusters (relative standard deviation: 2–61%, mean 24%), and there could be additional variation outside of the CCN measurement period. Still, it can be seen that during winter and early spring months, the

CCN population is dominated by anthropogenic clusters, while natural clusters dominate the CCN population during summer and early autumn. In the months of transition May and October a mixed contribution is observed. Only little is known about cloud formation SS in the Arctic. Earle et al. (2011) determined a range of 0.1–0.4% SS, while Leaitch et al. (2016) estimate up to 0.6% SS for high-level clouds and 0.3% SS for low-level clouds. Different SSs only result in minute variations in the relative contribution of anthropogenic versus natural aerosol clusters to CCN concentration.

4. Conclusions

Five years of aerosol number size distributions is characterized in terms of k-means cluster analysis leading to eight classifications, as discussed in Dall'Osto, Geels, et al. (2018) and Lange et al. (2018). Here, we have used these results to classify data of eBC content, HGF, and CCN activities from the high Arctic VRS. We show that—over yearly timescales—natural aerosol particles of predominantly organic composition likely dominate the submicrometer number concentration and CCN population during summer. The contribution to aerosol formation and growth in the Arctic atmosphere likely depends upon a number of organic and inorganic chemical compounds. Sources responsible for such emissions are still poorly understood, and decreasing sea ice extent and thickness will affect them. Willis et al. (2017) presented evidence for a secondary source of marine biogenic organic aerosol in the Canadian High Arctic during a summertime period of pristine Arctic background conditions. Other results in the same study area also suggest Arctic marine secondary organic aerosol being the major ultrafine aerosol contributor (Croft et al., 2018). Some of the only in situ measurements of sub-100-nm particle composition—made in the Canadian Arctic archipelago during summer—also indicated that particles are composed largely of organics during growth and NPF events (Tremblay et al., 2018). Our in situ aerosol measurements are in line with previous studies, strongly supporting a major role for secondary organic aerosol formation dominating the aerosol population during summertime. In view of changing biogenic processes and corresponding source strengths of aerosol precursors in a changing Arctic climate with changing sea ice extent, these results are of particular importance with respect to possible climate feedback mechanisms, and the understanding of the ongoing Arctic temperature increase. Additional measurements of ultrafine particle chemical composition and gas phase species are needed in order to elucidate the interaction between aerosol, clouds and climate in the Arctic.

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