Characterizing the hygroscopicty of growing particles in the Canadian Arctic summer

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Abstract. The impact of aerosols on clouds is a well-studied, although still poorly constrained, part of the atmospheric system. New particle formation (NPF) is thought to contribute 40–80% of the global cloud droplet number concentration, although it is extremely difficult to observe an air mass from NPF to cloud formation. NPF and growth occurs frequently in the Canadian Arc-tic summer atmosphere, although only a few studies have characterized the source and properties of these aerosols. This study presents cloud condensation nuclei (CCN) concentrations measured on board the *CCGS Amundsen* in the eastern Canadian Arctic Archipelago from 23 July to 23 August 2016 as part of the Network on Climate and Aerosols: Addressing Uncertainties in Remote Canadian Environments (NETCARE). The study was dominated by frequent ultrafine particle and/or growth events, and particles smaller than 100 nm dominated the size distribution for 92% of the study period. Using κ-Köhler theory and aerosol size distributions, the mean hygroscopicity parameter (κ) calculated for the entire study was 0.12 (0.06–0.12, 25th–75th percentile), suggesting that the condensable vapours that led to particle growth were primarily non-hygroscopic, which we infer to be organic. Based on past measurement and modelling studies from NETCARE and the Canadian Arctic, it seems likely that the source of these non-hygroscopic, organic, vapours is the ocean. Examining specific growth events suggests that the mode diameter (D_{max}) had to exceed 40 nm before CCN concentrations at 0.99% SS started to increase, although a statistical analysis shows that CCN concentrations increased 13–274 cm^{-3} during all ultrafine particle and/or growth times (total particle concentrations > 500 cm^{-3}, D_{max} < 100 nm) compared to Background times (total concentrations < 500 cm^{-3}) at SS of 0.26–0.99%. This value increased to 25–425 cm^{-3} if the growth times were limited to times when D_{max} was also larger than 40 nm. These results support past results from NETCARE by showing that the frequently observed ultrafine particle and growth events are dominated by a highly non-hygroscopic fraction, which we interpret to be organic vapours originating from the ocean, and that these growing particles can increase the background CCN concentrations at SS as low as 0.26%, thus pointing to their potential contribution to cloud properties and thus climate through the radiation balance.

1 Introduction

The Arctic environment is changing at a rapid pace, driven by surface air temperatures that are increasing up to two or three times faster than the global average (Serreze and Barry, 2011) and summer sea ice extent that is steadily declining [ref]. Many of these changes are driven by factors that affect the summertime surface energy budget, when solar radiation is present and changes in surface albedo due to melting surface ice or snow can affect the balance in the surface energy. In addition, clouds and aerosols can influence the amount of incoming solar radiation that reaches the surface of the earth. The role of low-level clouds in the summer Arctic radiation budget is complex, as they can cause the surface to not only cool, as expected at lower latitudes, but also warm, depending on the surface albedo and solar zenith angle (Shupe and Intrieri, 2004). Liquid cloud droplets form on pre-existing aerosol particles suspended in the atmosphere. Particles that can activate into droplets are called cloud condensation nuclei (CCN), and whether a given particle will act as a CCN depends on its size, chemical composition and the supersaturation (SS) to which it is exposed. Therefore, it is important to characterize Arctic aerosol properties and their role in cloud formation processes to better understand the surface radiation budget in the changing Arctic atmosphere.

Aerosol concentrations in the Arctic experience a strong seasonal cycle, with long range transport from southerly latitudes bringing high mass concentrations during the winter and spring time, which is known as Arctic Haze. In contrast, the summer Arctic atmosphere is much cleaner with very low aerosol concentrations, providing conditions favourable for new particle formation (NPF). In the marine atmosphere at lower latitudes, NPF has been observed to occur in the free troposphere, but over open waters in the Arctic summer, frequent drizzle reduces the aerosol concentrations (condensation sink) to favour NPF in the boundary layer (Croft et al., 2016a; Browse et al., 2012). Modelling studies have estimated that NPF can contribute 40–80% of cloud droplets globally (Merikanto et al., 2009; Kuang et al., 2009; Pierce and Adams, 2006). However, most of those studies assume that all particles that reach a minimum size, typically 70–100 nm, will automatically contribute to cloud droplet number concentration (CDNC). In determining the contribution of NPF on cloud droplet formation, it is important to characterize: 1) the source of the condensing vapours that lead to NPF and subsequent growth, noting that these sources could be different; 2) the chemical composition, and therefore the hygroscopicity, of the growing particles to determine their ability to activate as CCN; 3) whether the particles grow large enough to activate at a SS that is relevant in the ambient atmosphere; and 4) whether the particles formed are exposed to a SS that allow them to activate. It is extremely rare to actually observe an entire event from NPF to cloud droplet formation since the temporal and spatial scales usually cannot be captured by a single measurement platform and most studies, including this one, can only contribute to investigating some of these aspects.

To understand the sources and chemical composition of summer Arctic aerosols, early studies at Alert, Canada identified large mass fractions from methane sulphonate (MSA) and sulphate, suggesting that secondary aerosol mass originated from the marine biological production of dimethyl sulphide (DMS) and its subsequent atmospheric oxidation products led to aerosol growth (Li et al., 1993). Observations from 2011–2012 at Alert (Leaitch et al., 2013) and during the International Polar Years (2007–2008) in the Canadian Arctic Archipelago (Rempillo et al., 2011; Chang et al., 2011) linked small and growing particles to sulphate and DMS through measurements and modelling, further supporting the role of marine biology in Arctic aerosol through DMS oxidation in NPF and growth. There is evidence that iodine can also contribute to nucleation, usually on the

55 coast of Greenland (Allan et al., 2015; Dall'Osto et al., 2018; Sipilä et al., 2016), as well as more recently in the Central Arctic Ocean (Baccarini et al., 2020). However, there is typically insufficient iodine to explain the observed growth in these studies. Similarly, NH_3 emitted from the guano of migratory birds is thought to significantly contribute to NPF in the Canadian Arctic (Croft et al., 2016b; Wentworth et al., 2016).

More recently, a growing body of work is providing evidence that organic compounds contribute to the growth of newly
60 formed particle at lower latitudes (Ehn et al., 2014), as well as in the Arctic. In the Canadian Arctic, direct measurements of chemical composition using aerosol mass spectrometry from aircraft (Willis et al., 2016) and at a ground site (Tremblay et al., 2019) showed significant organic contributions during three observed growth events. These growing particles have been linked to open waters at the local to regional scale through footprint sensitivity analyses, suggesting that the source of these organics was from the ocean. Indeed, oxygenated volatile organic compounds (OVOCs) have been observed in the Canadian
65 Arctic (Mungall et al., 2017), with their atmospheric concentrations correlated to sunlight and surface ocean coloured dissolved organic matter. These OVOCs would be less volatile and could, with short amounts of atmospheric ageing, potentially condense and contribute to aerosol growth if the conditions were favourable (Ehn et al., 2014). On the Fram strait, hygroscopicity measurements at super- and sub-saturated conditions have consistently shown that summer aerosols, which are dominated by NPF and growth (Tunved et al., 2013), are less hygroscopic than pure inorganics, with observed hygroscopicity parameters
70 (κ) ranging from 0.15–0.4 (Zábori et al., 2015; Kecorius et al., 2019; Lange et al., 2019). Since the expected κ values for ammonium sulphate and sulphuric acid range from 0.6–0.7 (Petters and Kreidenweis, 2007; Shantz et al., 2008), the observed κ values suggest that the aerosol was composed of a significant organic fraction. Similarly, aerosol hygroscopicity measured in the Pacific portion of the Arctic Ocean were 0.08–0.15 (Park et al., 2020; Herenz et al., 2018). A recent modelling study incorporating many of these results found that a large source of organic vapours originating from Arctic waters could explain
75 much of the growth observed after NPF in the Canadian Arctic summer (Croft et al., 2019).

An aspect to consider is whether particles formed from NPF also grow to sizes large enough to activate at atmospherically-relevant SS. Cloud studies often find CDNC correlating with the accumulation mode (e.g. Hegg et al., 2012; Jia et al., 2019). However, it has been hypothesized that the extremely low aerosol number concentrations sometimes found in the summer Arctic causes even the smallest particles to quickly activate in a CCN-limited regime (Mauritsen et al., 2011). In-cloud obser-
80 vations from aircraft in the Canadian Arctic (Leaitch et al., 2016) and a mountain site on Svalbard (Koike et al., 2019) have reported that particles as small as 30 nm could be activating when the concentrations were less than 30 cm^{-3}, suggesting that newly formed particles need only to grow to 30 nm before contributing to CDNC and potentially affecting climate. Although not direct measurements of cloud droplets, measured CCN concentrations have been observed to increase at SS > 0.1% during NPF and growth in Arctic (Willis et al., 2016; Kecorius et al., 2019; Lange et al., 2019) and Antarctic (Kim et al., 2019)
85 locations, further suggesting that these newly formed particles can activate and contribute to climate effects.

Although many of the above-mentioned studies have characterized the hygroscopicity of the summer Arctic aerosol, only four have focussed specifically on studying NPF and growth (Willis et al., 2016; Burkart et al., 2017; Kecorius et al., 2019; Lange et al., 2019), with the first three studies reporting results from five individual events. In this study we expand these findings to more events by presenting aerosol and CCN measurements sampled from the *CCGS Amundsen* in the Canadian

90   Arctic Archipelago during the summer of 2016 as a part of the Network on Climate and Aerosols: Addressing Uncertainties in
Remote Canadian Environments (NETCARE). While previous work by Collins et al. (2017) characterized the ultrafine particle
(UFP) and growth events observed during the cruise, this study focuses on understanding the hygroscopicity of the particles
during growth to determine if the particles grew sufficiently large to activate at a SS that is relevant for cloud formation.
Calculating the hygroscopicity parameter also allows us to characterize the chemical composition of the aerosols and thus infer

95   the sources of the condensing vapours that contributed to the growth of the UFP.

2   Methods

2.1   Sampling platform and instrumentation

This study uses data collected from the NETCARE ship-based observations on board *CCGS Amundsen* from 14 Jul to 25
Aug 2016, with the cruise track shown in Fig. 1. Based on the proximity to the open ocean, surrounding land mass, time, and

100   direction of the ship cruise, the study was divided into three legs: (i) Baffin Bay (23 Jul – 4 Aug), (ii) Nares Strait (5–17 Aug),
and (iii) Resolute Bay (17–23 Aug), shown in Fig. 1 as the red, blue, and green coloured boxes, respectively. While the ship
was in the Baffin Bay region, it was influenced more by the surrounding open waters. In contrast, the ship was closer to sea-ice
or land masses in Legs 2 and 3, respectively. Furthermore, the latitudinal change was greatest in the Nares strait region, while
the other two regions had greater variations in longitude.

105   Ambient measurements of CCN concentrations were made with a streamwise thermal gradient CCN counter (CCNC,
Droplet Measurement Technologies, CCN-100) (Roberts and Nenes, 2005). Ambient aerosols measured by the CCNC were
sampled from an inlet approximately 38 m from the bow on the port side, approximately 9 m above sea level. The CCNC was
operated at a total flow rate of 0.5 L min^{-1} and a sample flow rate of 0.05 L min^{-1}. The temperature gradient of the CCNC was
varied such that ambient aerosols were exposed to SS of 0.17, 0.26, 0.44, 0.63, 0.81 and 0.99%, where each SS was maintained

110   for 10 min at a time. The SS of the chamber were calibrated weekly during the observation period with ammonium sulphate
aerosols at 5 SS values between 0.1–1%, except on 18 August 2016 when it was only calibrated at 0.44 and 0.99% SS. The
SS was stable throughout the 4 weeks of observations, although the plateau of the calibration curves decreased throughout the
study. This could be interpreted as a reduction in the counting efficiency of the CCNC, although after extensive analysis, we
believe it is more likely due to issues with the calibration system. As such, we assume that the ambient CCN concentrations

115   were correct, and the analysis in this study uses the values directly reported by the CCNC without accounting for any additional
particle loss or counting efficiency corrections.

A scanning mobility particle sizer (SMPS, TSI Inc., Model 3080, 3787) measured the number size distributions of aerosol
between the diameter range of 10 and 430 nm, with a time resolution of 5 min, and sample and sheath flow rates of 0.6 and
6 L min^{-1}, respectively. This instrument was in the foredeck container, approximately 31 m from the CCNC. Further details

120   on the SMPS sampling methods, data accuracy and data filtering techniques for ship pollution are presented by Collins et al.
(2017). The results presented in this study use observations from 23 July to 23 August 2016 when measurements from both
instruments were available.
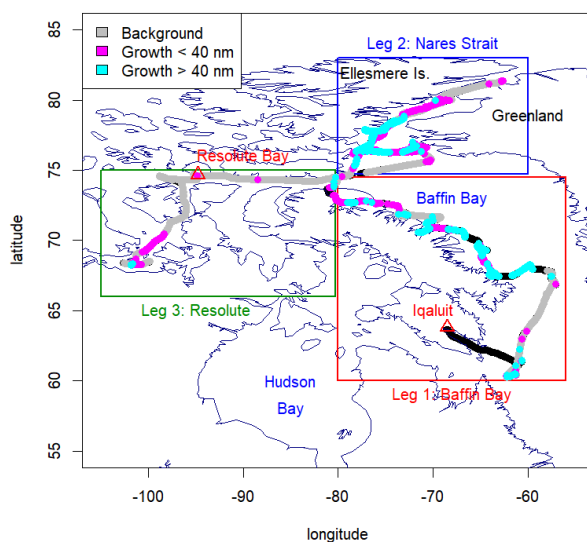
Figure 1. Track of the NETCARE ship cruise (black and coloured points). Rectangles show the three legs and red triangles denote communities. The colour of the points show times used in calculations of Background (grey) and Growth > 40 nm (light blue). Additional times included in the UFP and Growth calculations are shown in magenta.

2.2 CCN data analysis

The first 30 s of CCN data at every SS were excluded because the chamber temperatures require time to stabilize when changing
125 to a new SS. Only values with the temperature stabilized flag, as reported by the CCNC, were used in this analysis. Additionally,
the sample times that were previously identified to be contaminated by Collins et al. (2017) were excluded from the CCNC data.
It has been reported that the total uncertainty in the estimated CCN concentration above 100 cm^{-3} varies between 7 and 16%
due to factors such as temperature, pressure, flow, etc. (Moore et al., 2011). One second CCN concentrations were matched
to the 5 min SMPS sample times and the median concentrations calculated. The median was used to avoid the influence of
130 outliers that may have remained after filtering and are, on average, only 1.2% lower than mean concentrations. Of the 2061
concurrent SMPS and CCN observations, 69 were excluded due to high variability (standard deviation to median ratio was
greater than 0.5) or the concentrations exceeded 2000 cm^{-3}.

2.3 κ calculations

The hygroscopicity parameter (κ) was calculated using the aerosol size distribution, CCN concentrations and κ-K ohler theory
135 (Petters and Kreidenweis, 2007):

$$\kappa = \frac{4A^3}{27D_a^3(ln(S_c))^2},$$ (1)

where $A = (4M_w)/(RT\rho_w)$, M_w and ρ_w are the molar mass and density of water, respectively, while R and T are the ideal gas constant and the absolute temperature, respectively. D_a is the activation diameter and represents the diameter above which particles are large enough to activate as CCN at a saturation ratio of S_c. D_a was determined by integrating aerosol size distri-

140   butions downwards from the largest size bin until the cumulative particle number concentration was equal to the corresponding CCN concentration. The size bin at which this occurred was the D_a. This method has been previously used in studies where it is not possible to size select the particles before they are counted by the CCNC (e.g. Collins et al., 2013; Gao et al., 2020).

2.4 Defining growth and background times

To determine the overall contribution of growing UFP on the CCN concentrations throughout this study, UFP and/or growth

145   times were defined as times when the total number concentrations from the SMPS (N_T) > 500 cm^{-3} and $D_{max} \leq 100$ nm (magenta and light blue points in Fig. 1), where D_{max} is the statistical mode diameter in the size distribution (i.e. the diameter that had the maximum normalized concentration). An additional period when $N_T > 500$ cm^{-3} and $40nm < D_{max} < 100nm$ was defined as Growth > 40 nm (light blue points in Fig. 1), to examine CCN concentrations when the particles had grown larger than 40 nm but remained smaller than 100 nm. Both of these periods were compared to periods when minimal contributions

150   from UFP and growth were expected. These Background periods were defined as times when $N_T \leq 500$ cm^{-3} (grey points in Fig. 1). To retain clarity, periods when $N_T > 500$ cm^{-3} and $D_{max} \leq 40$ nm (magenta points in Fig. 1) will be referred to as Growth < 40 nm in the legends, although these events do not always show growth and are not considered separately.

3 Results and discussion

3.1 General overview

155   The aerosol size distributions measured by the SMPS throughout the study and D_{max} are shown in Fig. 2a, and the CCN concentrations and N_T are shown in Fig. 2b. The three legs are denoted by the shaded boxes above panel a. Summary statistics of CCN concentrations for the entire study and each leg are shown in Table 1. As seen in Fig. 2a, the study was characterized by frequent UFP and growth events, which caused N_T to vary by three orders of magnitude. Collins et al. (2017) identified 14 UFP and growth events during this cruise which accounted for 41% of the sample times. However, particles smaller than 100

160   nm dominated the SMPS number size distribution for 92% of the study, suggesting that most of the aerosol particles observed throughout the study had undergone secondary formation processes such as condensational growth. As such, even if particles were not actively growing at a given time, their CCN-activity and inferred chemical composition could still provide insight into the vapours that contributed to particle growth.

CCN concentrations at the higher SS usually varied according to N_T, showing evidence that UFP and growth can lead to

165   increased CCN concentrations. However, the median activation ratio (AR), defined as the CCN concentration / N_T, was only 0.38 at the highest SS of 0.99%, suggesting that most of the particles were either too small and/or non-hygroscopic to activate.
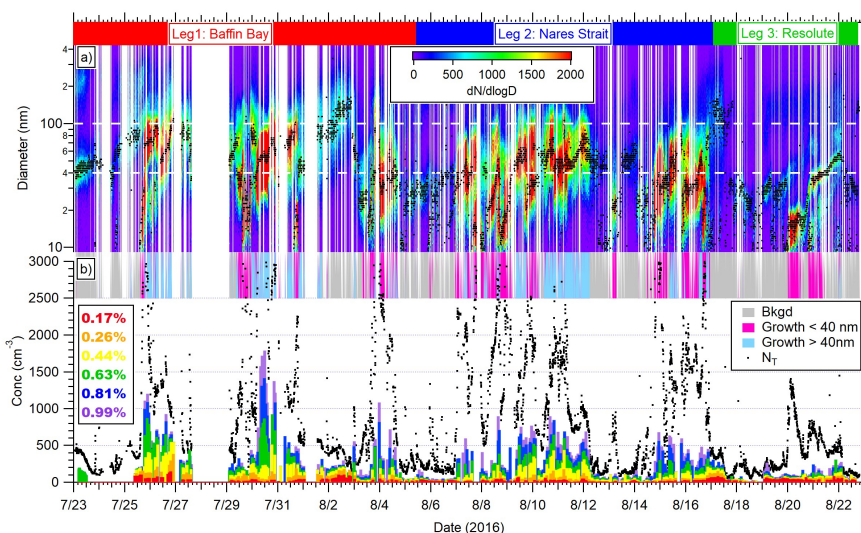
Figure 2. Time series of a) aerosol number size distribution and D_{max} (black points) and b) N_T (black points) and CCN concentrations. Dashed white horizontal lines show particle diameters of 40 and 100 nm. Times used in calculations of Background, Growth > 40 nm and Growth < 40 nm are shown in grey, light blue, and magenta, respectively.

Table 1. Summary of CCN concentrations and associated calculations, given as median (25th-75th percentile)

SS	0.17%	0.26%	0.44%	0.63%	0.81%	0.99%
			CCN [cm^{-3}]			
All	29 (20-45)	54 (35-92)	98 (48-171)	141 (80-211)	197 (89-353)	228 (99-456)
Baffin Bay	36 (20-57)	92 (48-124)	162 (98-234)	158 (127-221)	244 (153-441)	286 (184-549)
Nares Strait	22 (16-29)	40 (27-59)	89 (38-156)	140 (61-264)	209 (85-386)	280 (79-496)
Resolute Bay	41 (32-53)	56 (49-68)	70 (49-92)	87 (67-120)	118 (78-194)	136 (89-231)
			AR			
All	0.05 (0.02-0.13)	0.08 (0.04-0.18)	0.18 (0.09-0.27)	0.31 (0.16-0.39)	0.32 (0.2-0.48)	0.38 (0.24-0.52)
Baffin Bay	0.05 (0.02-0.09)	0.09 (0.05-0.2)	0.20 (0.1-0.33)	0.36 (0.24-0.4)	0.37 (0.21-0.5)	0.40 (0.3-0.48)
Nares Strait	0.02 (0.01-0.07)	0.05 (0.03-0.1)	0.13 (0.08-0.22)	0.21 (0.13-0.31)	0.27 (0.18-0.37)	0.34 (0.21-0.46)
Resolute Bay	0.16 (0.07-0.22)	0.24 (0.12-0.36)	0.27 (0.15-0.43)	0.33 (0.17-0.57)	0.47 (0.24-0.69)	0.54 (0.32-0.86)
			κ			
All	0.11 (0.09-0.17)	0.11 (0.09-0.14)	0.09 (0.06-0.11)	0.08 (0.03-0.11)	0.08 (0.05-0.11)	0.07 (0.04-0.11)
Baffin Bay	0.08 (0.06-0.11)	0.09 (0.07-0.11)	0.07 (0.05-0.1)	0.05 (0.02-0.08)	0.05 (0.03-0.08)	0.04 (0.03-0.07)
Nares Strait	0.12 (0.10-0.17)	0.11 (0.09-0.12)	0.10 (0.08-0.12)	0.10 (0.07-0.12)	0.09 (0.06-0.12)	0.08 (0.05-0.11)
Resolute Bay	0.21 (0.12-0.31)	0.14 (0.11-0.19)	0.08 (0.06-0.14)	0.10 (0.06-0.19)	0.09 (0.05-0.26)	0.11 (0.06-0.20)

Table 2. CCN and N_T concentrations from other studies

Season	Platform	CCN (cm^{-3})	SS (%)	N_T	Reference
High Arctic					
Summer	Ground	45–81	0.45		(Jung et al., 2018)
Summer	Ship	14 ±11	0.1		(Martin et al., 2011)
		47±31	0.73		
Summer	Aircraft	247	0.5	514	(Lathem et al., 2013)
Sub-Arctic					
Spring	Ground	10–250	0.1–0.7	20–500	(Herenz et al., 2018)
Antarctica					
Summer	Ground	10–1300	0.1–0.7	40–6700	(Herenz et al., 2019)
Summer	Ground	60–200	0.4	≈2000	(Yu and Luo, 2010)
High Altitude Regions					
Winter	Nainital	2180 ±16	0.46	2891±2020	(Gogoi et al., 2015)
Spring	Jungfraujoch	149±171	0.12	550	(Jurányi et al., 2010)
		568±401	1.18		

These low AR are consistent with observations from Ny Alesund (Jung et al., 2018), where summer time AR of 0.4 were reported, as well as other Arctic sites (Table 2).

Over the entire study, the median and interquartile range of CCN concentrations increased with increasing SS (see Table 1), with a median CCN concentration of 29 cm^{-3} at SS of 0.17% compared to 98 and 228 cm^{-3} at 0.44% and 0.99%, respectively. To put our observations into a global perspective, CCN and N_T concentrations observed at a select number of polar and remote sites are shown in Table 2. This list is by no means exhaustive, but shows that overall, our CCN concentrations are generally higher than other Arctic observations at Ny Alesund (Jung et al., 2018) and the Central Arctic Ocean (Martin et al., 2011). A notable exception is the aircraft observations over the Canadian Arctic Archipelago in 2008 which were influenced by biomass burning and transport of industrial emissions and resulted in higher aerosol concentrations (Lathem et al., 2013). Our observations are closer to those reported for a sub-Arctic Canadian site in the spring (Herenz et al., 2018) and Antarctica (Herenz et al., 2019; Yu and Luo, 2010). In addition, our observations are lower than measurements from high altitude locations at lower latitudes (Gogoi et al., 2015; Jurányi et al., 2010).

Calculated κ values and summary statistics are also presented in Table 1. Overall, the values were very low, with medians for the entire study ranging from 0.07–0.11 over the six SS. Values were highest at the lowest SS (0.08–0.21), suggesting that the larger particles were more hygroscopic, which is consistent with more processing in the larger aerosols or a different source resulting in a higher hygroscopicity, such as sea spray. The values also increased throughout the study, with the lowest values (0.04–0.08) corresponding to the first part of the study when the ship was in the warmer and more biologically-active waters of Baffin Bay where more UFP and growth events were observed, and the highest values (0.18–0.21) corresponding to

185 the shallower and more coastal waters during Leg 3 when only one UFP and growth event occurred. The κ values calculated

from this study are consistent with the NETCARE aircraft observations for one event (0.1) (Willis et al., 2016; Burkart et al.,

2017) as well as other mobile platforms over Arctic waters near Svalbard during four NPF and growth events (0.13 for 20 nm

particles) (Kecorius et al., 2019) and from air influenced by the Beaufort and Chukchi Seas (0.08) and the Pacific Ocean (0.03)

(calculated from Park et al., 2020). However, the observed particles appear less hygroscopic than those observed in the Central

190 Arctic Ocean (0.33±0.11) (Martin et al., 2011), as well as from land-based sites in spring at Tuktoyaktuk, Canada (0.23)

(Herenz et al., 2018); non-anthropogenically-influenced particles at the VRS site in northeast Greenland (0.25–0.4) (Lange

et al., 2019); and from Zeppelin (0.3–0.46) (Jung et al., 2018; Zábori et al., 2015). The lower values determined from the

mobile platforms likely reflect the more recently condensed vapours during individual growth events, whereas the land-based

sites generally reported values averaged over several months, causing them to be less sensitive to distinct UFP and growth

195 events.

Based on the median calculated κ values from the entire study, we infer that >80% of the aerosol volume fraction was

composed of a non-hygroscopic component, which we interpret as being organic. Previous modelling results have shown that

the air sampled during our study was influenced by source regions within the Arctic circle which is mostly marine (Collins

et al., 2017; Burkart et al., 2017), suggesting that these non-hygroscopic aerosols were influenced by the water. Other recent

200 studies in the Canadian Arctic have inferred that VOCs are emitted from the ocean (Mungall et al., 2017), that the source

of secondary aerosol mass for UFP and growth are driven by marine biological influences (Collins et al., 2017; Willis et al.,

2017), that particles < 100 nm during growth events are almost entirely organic and influenced by open waters (Tremblay et al.,

2019; Willis et al., 2016) and that including a significant source of Arctic marine secondary organic aerosol could explain the

frequently observed NPF and growth events (Croft et al., 2019). Together, these results suggest that the condensing material

205 contributing to particle growth is non-hygroscopic, likely organic originating from marine biological sources.

3.2 Influence of particle growth on CCN

To further explore the potential role of UFP and growth on climate, CCN concentrations and D_a were examined for specific

events when UFP and growth were observed. Figure 3 shows a period when particles smaller than 20 nm appeared on several

occasions and then grew to D_{max} of 40–60 nm, with the larger tail of the mode sometimes growing as large as 100 nm (upper

210 dashed white line in panel a). Since the D_a at 0.99% remained between 45 and 70 nm (purple triangles in panel a), CCN

concentrations remained low during the two UFP and growth events on 8 Aug, even as total concentrations exceeded 2000

cm^{-3} because the majority of the particles were too small and/or non-hygroscopic to activate. However, as the particle D_{max}

grew larger than 40 nm after 9 Aug 08:00 (light blue shading in panel b), CCN concentrations noticeably increased at all SS >

0.44%, suggesting that these growing particles became sufficiently large to activate. The κ values determined during this period

215 mostly remained between 0.05 and 0.18 at all SS, suggesting that the hygroscopicity, and therefore chemical composition, was

similar across the size range of the D_a determined at all SS (40–180 nm, pink circles and purple triangles in panel a). Similar

results are seen in Fig. 4 which shows three UFP and growth events between 29 Jul and 5 Aug. CCN concentrations most
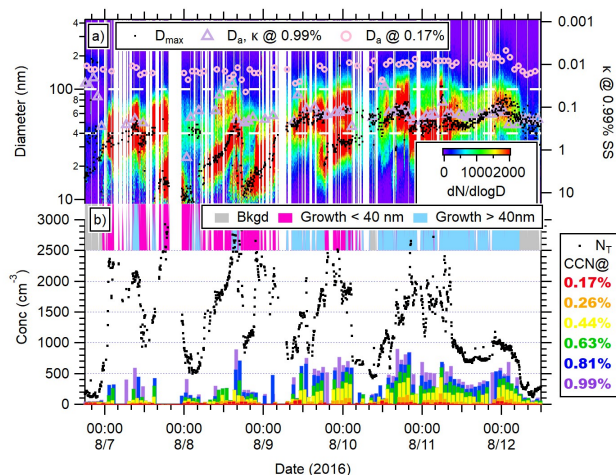
Figure 3. As in Fig. 2 but for 07–12 Aug 2016. D_a calculated at 0.17% and 0.99% SS are included in panel a) as well as the corresponding κ values calculated for D_a at 0.99% SS.
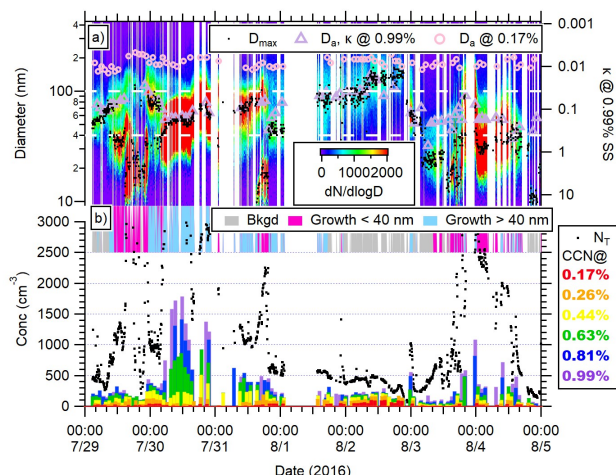
Figure 4. As in Fig. 2 but for 29 Jul–05 Aug 2016. D_a calculated at 0.17% and 0.99% SS are included in panel a) as well as the corresponding κ values calculated for D_a at 0.99% SS.

obviously increased after 30 July 00:00 when D_{max} exceeded 40 nm (light blue shading), and did not increase when bursts of smaller particles appear at 31 July 18:00 and 03 August 08:00 (magenta shading).

220   To determine the overall contribution of growing UFP on the CCN concentrations throughout this study, Fig. 5 shows the increase in the mean CCN concentration at each SS during all UFP and/or Growth times (i.e. Growth < 40 nm and Growth > 40 nm) compared to Background times (purple dashed line) as well as the increase during only Growth > 40 nm times compared to Background times (light blue line). The whiskers in Fig. 5 denote the 95% confidence interval calculated as 1.96 times the stan-
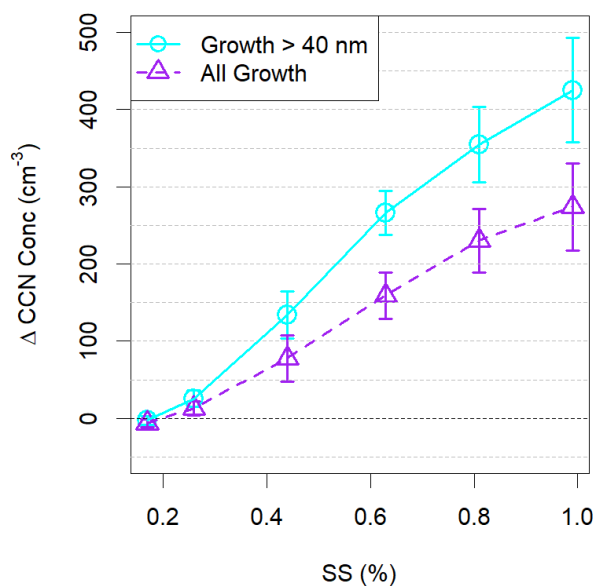
Figure 5. The increase in CCN concentration during Growth > 40 nm (cyan) and All Growth (purple) times compared to Background times at different SS. The error bars show the 95% confidence interval.

dard errors of the two means summed in quadrature. During all UFP and/or Growth times, the CCN concentrations increased
225 by 13–161 cm^{-3} at SS of 0.26%–0.99%, corresponding to a 22–167% increase in concentration compared to background,
demonstrating that particles formed from UFP and growth events in the Canadian Arctic can contribute to CDNC at a very
modest SS of 0.26%. This effect is even more pronounced if only Growth > 40 nm times are considered, when CCN concentrations increased by 25–425 cm^{-3} at SS of 0.26%–0.99%, corresponding to a 43–259% increase in concentration compared
to Background periods. These values are statistically significant ($p < 0.05$) when tested with a Mann Whitney test (*wilcox.test*
230 function, R version 4.0.3). Our findings are consistent with observations on northeast Greenland where CCN concentrations
were reported to increase by 42–95 cm^{-3} at SS of 0.3% and 85–150 cm^{-3} at higher SS during the Nascent and Bimodal clusters, which were characterized by NPF (Lange et al., 2019). Our findings also support NETCARE aircraft studies performed
during the summer of 2014 where CCN concentrations increased by ≈ 100 cm^{-3} at SS of 0.6% for one growth event (Willis
et al., 2016) and particles as small as 30 nm, presumably formed recently, were inferred to have activated at SS of 0.3% in
235 clouds (Leaitch et al., 2016). Similarly, particles as small as 30–50 nm were inferred to activate when aerosol concentrations
were less than 30 cm^{-3} at the mountain site Zeppelin on Svalbard (Koike et al., 2019). Together, these studies suggest that
small particles, influenced by condensational growth, can affect cloud radiative properties by contributing to CDNC.

Table 3. Calculated Twomey parameters

	All	Baffin Bay	Nares Strait	Resolute Bay
C	335	455	337	172
k	1.24	1.25	1.44	0.81

3.3 Twomey parameterization

Figure 6 shows the mean CCN concentrations at each SS for the full study period (solid black circles) and the three regions
considered in the study. The lines in Fig. 6 represent the best fit of the empirical Twomey parameterization often used in models
to relate CCN to SS using:

$$CCN = C \times SS^k, \tag{2}$$

where C represents the CCN concentration at 1% SS and k is the power law exponent (Twomey, 1959). The parameters C and
k are widely used in cloud microphysical models as they provide information about size and composition of the background
aerosol concentration (Seinfeld and Pandis, 1997), although more recent parameterizations have adopted Eq. 2 to include more
information about the aerosol microphysical parameters (Khvorostyanov and Curry, 2006). The Twomey parameters calculated
for our study are listed in Table 3. The C parameter for the full study period and the Baffin Bay and Nares strait regions are
very similar (between 335 to 455 cm^{-3}), whereas it was much lower (172 cm^{-3}) for the Resolute Bay region, reflecting the
overall lower N_T during Leg 3. With the exception of the last leg, these values are all significantly higher than the 100–140
cm^{-3} determined for the Zeppelin station at Ny Alesund for July and August (Jung et al., 2018), likely due to the greater
aerosol concentrations caused by UFP and growth during our study resulting in more CCN at 1% SS. Similarly, the maximum
k estimated at Ny Alesund was in July, with a value of 0.5606, which is lower than the values estimated for our study, especially
the Baffin Bay and Nares Strait regions (1.2–1.4). This can also be attributed to the persistent UFP and/or growth events during
our study since a greater number of small particles can activate as CCN at higher SS. In contrast, the Resolute Bay region,
which had less UFP and growth events, had a much lower k of 0.81, showing that the CCN spectrum was less sensitive to
changes in SS.

4 Conclusions

This study reports CCN concentrations measured in the eastern portion of the Canadian Arctic Archipelago during the NET-
CARE campaign onboard the *CCGS Amundsen*. The observations reported here took place from 23 Jul – 23 Aug 2016 when
UFP and/or growth events were highly prevalent, with particles smaller than 100 nm dominating the particle number size
distribution for 92% of the study. These UFP and/or growth events resulted in high particle concentrations which were also
reflected in increased CCN concentrations, suggesting that the frequently-observed small, growing particles have the ability
to contribute to CDNC and therefore the radiative budget in the Arctic. The mode diameter of the growing particles generally
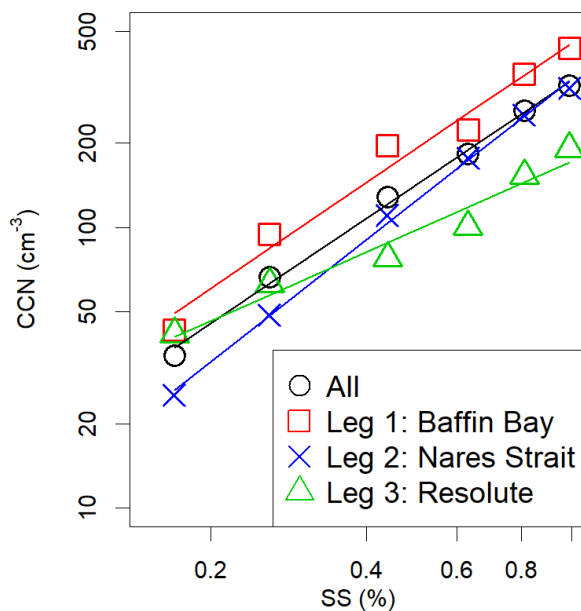
Figure 6. Twomey parameterization for the entire study (black) and the three legs (red, blue and green).

had to reach 40 nm before the particles became CCN-active at 0.99% SS, although CCN concentrations were shown to have
a statistically-significant increase of 13 and 78 cm^{-3} at even modest SS of 0.26 and 0.44% compared to Background times,
defined as when N_T was lower than 500 cm^{-3}. The influence of frequent UFP and/or growth events was also reflected in the
Twomey parameterization, which resulted in higher C and k values than previous studies.

The calculated κ values for the observed CCN provide insight on the hygroscopicity of the particles as well as their composition, and thus the source of the condensing vapour that contributed to the observed aerosol growth. Values were low, with
a mean of 0.12 (0.06–0.12, 25–75th percentile) for the entire study over all SS, suggesting that at least 80% of the volume
fraction of the aerosol was non-hygroscopic, likely organic. Since the history of the air mass was generally local or regional,
the condensing non-hygroscopic vapours most likely originated from the open waters within the Arctic. These κ values are
lower than many other Arctic observations on land, although these other studies reported seasonal means, which likely included
more aged aerosols. Our values are more consistent with other observations reported from mobile platforms in open Arctic waters, especially during four UFP and growth events near Svalbard (Kecorius et al., 2019) and one event in the Canadian Arctic
(Burkart et al., 2017; Willis et al., 2016), likely reflecting the hygroscopicity of the locally-produced condensing vapours before
significant chemical ageing could occur.

This work provides an important link between other publications from the Canadian Arctic, especially from the NETCARE
study (Willis et al., 2016, 2017; Burkart et al., 2017; Mungall et al., 2017; Collins et al., 2017), by confirming that the numerous
particle growth events observed in summer 2016 was indeed driven by non-hygroscopic vapours that are presumed to be
organic. The present study corroborates prior chemical composition measurements of a small number of events (Willis et al.,

2016; Tremblay et al., 2019) and expands their spatial and temporal scope, thus demonstrating that the phenomenon of non-hygroscopic, likely organic, vapours condensing on UFPs is likely prevalent throughout the region. The presence of an Arctic marine secondary organic aerosol was a fundamental assumption in the work of Croft et al. (2019) when simulating new particle formation and growth in the Canadian Arctic, and our results support this assumption.

Overall, these results are important because it is the only study besides the work of Kecorius et al. (2019), Burkart et al. (2017), and Willis et al. (2016) to characterize the hygroscopicity of specific UFP and growing particles in Arctic waters. Our study demonstrates that the frequently-observed UFP and/or growth in the Canadian Arctic summer increases CCN concentrations by 26% at a modest SS of 0.44%, although further work will be needed to determine the ultimate effect on cloud properties and the radiation budget. The chemical composition, as inferred from the hygroscopicity, of the growing particles also provides insight on the source of the condensing vapours and allows us to better understand how changing environmental conditions in the Arctic (e.g. increased temperatures, reduced sea ice extent, lower salinities) can alter future aerosol processes, and by extension, the radiation budget.

Data availability. The data is being added to a data respository and details will be made available when the paper is accepted

Author contributions. This study was conceived by RYWC and JPDA; the data were collected by RYWC, DBC and MB; the data were analyzed by RYWC and JPC; and the manuscript was primarily written by RYWC with contributions from all other authors.

Competing interests. The authors declare that they have no competing interests.

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