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Key Points:

- Arctic new particle formation (NPF) involves a large variety of chemical species varying by location and season
- NPF and growth are observed throughout all seasons with enhancement when solar radiation is stronger
- A molecular-level understanding of nucleation across the Arctic is still missing, and future studies should focus on filling this gap

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Progress in Unraveling Atmospheric New Particle Formation and Growth Across the Arctic

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Abstract New particle formation (NPF) and growth can be an important source of cloud condensation nuclei for the Arctic atmosphere, where cloud formation is sensitive to their availability. Low-level clouds influence the Arctic energy budget, and likely contribute to amplified Arctic warming. Molecular information of NPF is rarely reported, despite its importance to determine sources of condensing vapors and nucleation mechanisms. Beck et al. (2020, https://doi.org/10.1029/2020gl091334) shed light on the complexity of NPF and growth at two Arctic locations. They reveal that chemical drivers and sources are diverse across the Arctic. This advance in knowledge calls for similar studies throughout all seasons and in various Arctic environments to obtain a more systematic understanding of NPF and growth.

Plain Language Summary Atmospheric particles can be formed from gaseous vapors, a process called new particle formation (NPF). These and other vapors further condense on the newly formed particles, growing them large enough to become cloud condensation nuclei (CCN), which are essential for cloud formation. The addition of CCN from NPF is particularly important in the summertime Arctic, where the nuclei numbers are generally low, even so low that cloud formation can be inhibited. Low elevation atmospheric clouds in the Arctic are important because they control the surface energy budget. The study of Beck et al. (2020, https://doi.org/10.1029/2020gl091334) investigates in detail the sources and chemical mechanisms of NPF on Svalbard and in northern Greenland. The results show that there are many different gases, which contribute to NPF. The specific contribution and processes are a question of season and environmental properties such as phytoplankton, sea ice, and snow presence or absence. These results, together with other recent studies, now provide a more complete picture of the complexity and diversity of Arctic NPF. However, despite the new insights into the chemistry, the contribution of newly formed particles to cloud formation remains poorly quantified.

1. Background

The Arctic is warming about twice as fast as the global average (Pörtner et al., 2019). The acceleration is responsible for critical changes of the Arctic environment (Schmale et al., 2021) including sea ice retreat (Serreze & Stroeve, 2015), increased emissions of dimethyl sulfide (DMS) from phytoplankton (Galí et al., 2019) and of volatile organic compounds (VOCs) from boreal forest and tundra (Lindwall et al., 2016). DMS and VOCs are precursors to low-volatility vapors such as sulfuric acid (SA), methanesulfonic acid (MSA) (Barnes et al., 2006; Hoffmann et al., 2016) and highly oxygenated molecules (HOMs) (Bianchi et al., 2019), which are fundamental for new particle formation (NPF) and growth. Global modeling studies suggest that NPF contributes the majority of the Arctic cloud condensation nuclei (CCN) population (Gordon et al., 2017). Arctic low-level clouds have a strong influence on the surface energy budget (Shupe & Intrieri, 2004) and depend among other processes on the availability of CCN (Mauritsen et al., 2011).

The Arctic features a strong seasonal cycle: Haze conditions, with accumulation mode particles larger than about 100 nm in diameter, dominate between fall and spring (Quinn et al., 2007). The summer, with efficient wet removal of anthropogenic emissions, is a natural-aerosol-dominated environment with low concentrations (Freud et al., 2017). Consequently, the concentration of vapors with the ability to form new particles can be higher because the loss to pre-existing aerosols (the condensation sink, CS) is reduced. In addition, the higher solar insolation promotes photochemical reactions (Tunved et al., 2013), while low-level inversions can concentrate surface emissions (Burkart et al., 2017). Microbiologically active waters contribute to NPF relevant gases, for example, SA and MSA (Levasseur, 2013), while animal colonies emit ammonia

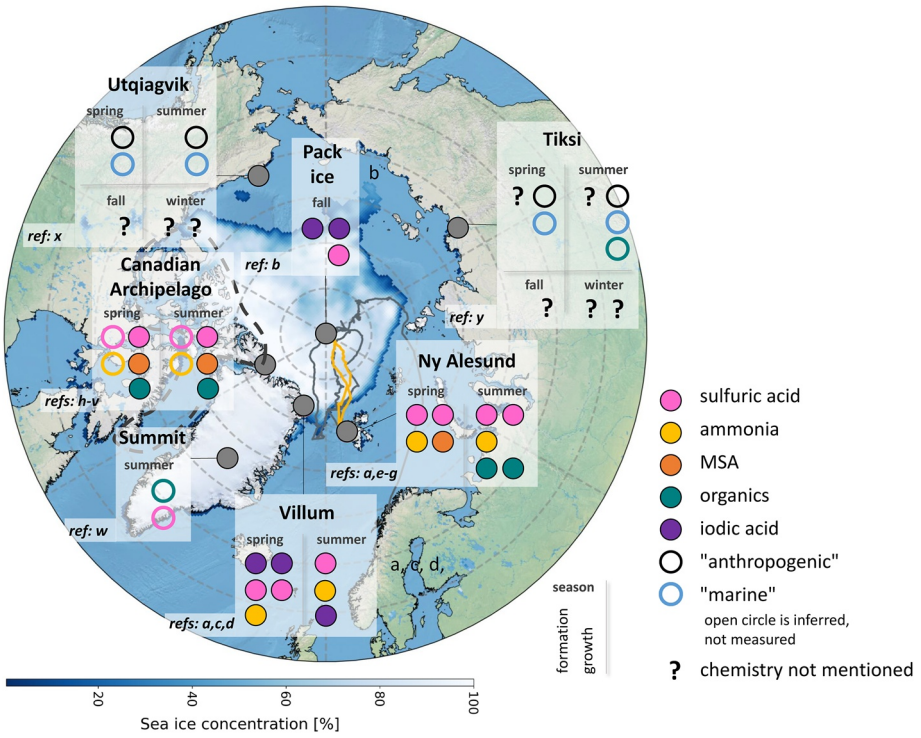

Figure 1. Compounds contributing to new particle formation (NPF) and growth across the Arctic. Gray dots and the gray dashed line represent locations with observations described in the text. Ship tracks are from the Arctic Ocean 2018 (yellow) and MOSAiC (gray) cruises (Baccarini et al., 2020; Shupe et al., 2020). The panels show for each season, which compounds were observed in NPF (left colum) and new particle growth (right). Full colors represent measured compounds, open circles refer to inferred compounds. Question marks mean growth was reported but without chemical information. References ("ref" are indicated with letters, which are attributed to studies mentioned in the text). The blue shading represents sea ice concentration (September 15, 2018), reported as a fraction of covered surface (Maslanik & Stroeve, 1999). The base map is Natural Earth II (https://www.naturalearthdata.com/).

(Croft et al., 2016), and yet unresolved processes lead to iodic acid (IA) production (Baccarini et al., 2020; Beck et al., 2020).

Globally, the main NPF agent is SA (Gordon et al., 2017). However, under typical boundary layer conditions, a base like ammonia is required to stabilize the nucleating clusters (Kirkby et al., 2011; Kürten et al., 2014). Ions have a similar effect and may enhance NPF rates, known as ion-induced nucleation. HOMs can form clusters with SA (Lehtipalo et al., 2018) or nucleate alone (Kirkby et al., 2016). Low-volatility organic vapors are fundamental for the growth of nucleated particles while SA is usually too scarce to explain most of the growth (Tröstl et al., 2016). IA and other iodine oxoacids can also drive NPF in coastal environments and over the high Arctic pack ice (Baccarini et al., 2020; He et al., 2021; Sipilä et al., 2016).

2. Recent Advances in Understanding Arctic NPF

Beck et al. (2020, "a" in Figure 1) reveal nucleation mechanisms for two Arctic sites, that is, Ny Alesund (NA) on Svalbard and Villum Research Station (VRS) in northern Greenland. They used a nitrate chemical ionization atmospheric pressure interface time-of-flight mass spectrometer (CI-APi-ToF-MS) (Jokinen et al., 2012) to measure low-volatility vapors. Together with two other studies (Baccarini et al., 2020, b in Figure 1; Sipilä et al., 2016, c), this work significantly advances our comprehension of Arctic NPF providing molecular level information.

At VRS, where sea ice is present most of the year, they find that IA drives NPF and early growth in spring, with concentrations up to 5×10^7 molecules cm^{-3}. In summer, fewer events were detected, driven mainly by SA and ammonia. Particles did not grow beyond 10 nm. Also at NA, separate spring and summer

Table 1
Comparison of Condensation Sink (CS), Formation and Growth Rates of New Particles Across the Arctic

Location, season	CS (s^{-1})	Formation rate (cm^{-3} s^{-1})	Growth rate (nm h^{-1})	Reference
Ny Alesund, Svalbard, summer	2×10^{-4}–10^{-2}			Giamarelou et al. (2016)
Zeppelin, Ny Alesund, summer	3×10^{-4}–5×10^{-4}	0.7 (11 nm) 0.3 (22 nm)	0.4 (11 nm) 1.4 (22 nm)	Heintzenberg et al. (2017)
Zeppelin, Ny Alesund, all but winter	3.8×10^{-4} (nucleation cluster)			Dall´Osto et al. (2017)
Zeppelin, all but winter		0.08 (spring) 0.032 (summer) 0.0066 (fall)	1.4, (spring) 1.2, (summer) 1.6 (fall)	Nieminen et al. (2018)
Ny Alesund, spring–summer	$\sim 4 \times 10^{-4}$	0.27 median (1 × 10e−3–6 × 10e−1 total range) (1.5 nm)	0.71–1.04	Beck et al. (2020)
Lancaster Sound, Canadian Arctic, summer	3.33×10^{-4} (2016) 6.11×10^{-4} (2014) 5.56×10^{-5}–2.78×10^{-3} (total range)		0.2–15.3 4.3 ± 4.1 (average)	Collins et al. (2017)
Alert, spring and summer		0.042 (spring) 0.0081 (summer)	0.8 (spring) 1.1 (summer)	Nieminen et al. (2018)
Summit Greenland, summer			0.09–0.3	Ziemba et al. (2010)
Villum, Greenland, spring–summer	$\sim 3 \times 10^{-4}$		0.25	Beck et al. (2020)
Central Arctic Ocean	8×10^{-5} median (4–14×10^{-5}, interquartile range)		0.2–1.2 (full range)	Baccarini et al. (2020)
Central Arctic Ocean, summer			0.8–3.6	Karl et al. (2012)
Utqiagvik, Alaska, summer, spring	2.4×10^{-3} (all) $1.7 \times 10^{-3} \pm 1.6 \times 10^{-3}$ (summer) $3.5 \times 10^{-3} \pm 1.9 \times 10^{-3}$ (spring)		2.2 ± 1.3, 3.6 ± 4.3, 5.0 ± 3.1 (June, July, and August) 1.8 ± 0.8 (February–May)	Kolesar et al. (2017)
Tiksi, Russia		0.01–0.41 (7 nm)	2.4 ± 2.0, 3.6 ± 2.5, 2.6 ± 1.7 (June, July, and August) 0.1–3.6 (all year average)	Asmi et al. (2016)

Note. Diameters in parentheses indicate the size of particles for which the formation or growth rate was calculated.

mechanisms were identified. In spring, with the increase in radiation intensity and coincidental with phytoplankton blooms, NPF occurred through ion-induced nucleation of SA (>10^6 molecules cm^{-3}) with ammonia, and growth by SA and MSA (>10^7 molecules cm^{-3}). In summer, high concentration of HOMs was measured (several 10^7 molecules cm^{-3}). HOMs were associated to snowmelt and potentially a terrestrial source but their actual composition could not be determined. The work from Beck et al. is the first reporting HOMs' contribution to NPF and growth in the Arctic. They also demonstrated that newly formed particles at NA can grow above 20 nm.

Here, we discuss how the most recent studies on Arctic NPF have contributed to our knowledge and summarize the information in a geographical overview in Figure 1. Table 1 compiles microphysical observations such as the CS, formation and growth rates. Several studies have investigated nucleation around Svalbard prior to Beck et al. (2020). For example, based on volatility measurements, Giamarelou et al. (2016, e) found that NPF involves SA and ammonia, in line with Beck et al. (2020). Long-term size distribution data from the Zeppelin Observatory highlighted that NPF is rare in winter and occurs mainly in summer, linked to peak solar radiation over the year and day (early afternoon) (Dall'Osto et al., 2017, g; Heintzenberg

et al., 2017, f). These studies suggested that NPF is linked to photo-oxidation of marine organic vapors and ammonia, and that the main source regions are the marginal ice zone and open waters between Svalbard and Greenland. The long-term data implied that smaller sea ice extent has a positive effect on NPF.

Alert and the archipelago in the Canadian Arctic are further well-studied locations (Abbatt et al., 2019, h). Multiple studies (Chang et al., 2011, i; Ghahremaninezhad et al., 2016, j; Leaitch et al., 2013, k; Sharma et al., 2012, l; Tremblay et al., 2019, m) report that the presence of nucleation mode particles and growth was linked to biogenic sulfur compounds (DMS, MSA, and SA). Back trajectory analyses showed that no particular surface type preferentially led to NPF, while significant growth of these particles was through organics (Burkart et al., 2017, n; Leaitch et al., 2018, o; Tremblay et al., 2019, p; Willis et al., 2016). Willis et al. (2017, r) identified organic alkyl groups, which exist in the sea surface microlayer, contributing to particle growth, in line with Mungall et al. (2017, s) who found that the microlayer is an important source of oxygenated VOCs. A modeling study confirms that a steady flux of organic vapors from the ocean is needed to explain Canadian Arctic aerosol size distributions (Croft et al., 2019, t). Ammonia emissions from bird colonies were identified as important ingredients to NPF as well (Croft et al., 2016, u).

On the Greenland ice sheet, growth of nucleation mode particles was observed, potentially dominated by organics emitted from the snow with only a minor role of SA (Ziemba et al., 2010, w). Near the North Pole on dense pack ice, Baccarini et al. (2020) found IA to drive formation and growth. These findings are in line with Beck et al. (2020) who also pointed out the role of IA for NPF and growth near sea ice. However, the iodine source may be different considering that at VRS, IA takes a prominent role in spring, while in the central Arctic Ocean, IA increases during the freeze-up in fall.

To our knowledge, there is no evidence for the contribution of anthropogenic precursors to NPF in any of the discussed locations. Kolesar et al. (2017, x) report for Utqiagvik, Alaska, that semi-volatile gases from Prudhoe Bay oil field appear to drive particle growth. At Utqiagvik, NPF was observed also in the absence of sunlight similar to Tiksi, Russia. There, Asmi et al. (2016, y) found that most NPF happens in spring, likely associated to the oxidation of anthropogenic emissions accumulated during the haze season. In summer, biogenic emissions from the tundra seem to play an important role together with marine air masses.

The microphysical observations reported in the literature (Table 1) are difficult to compare as for example, the formation and growth rates refer to different particle sizes. However, a large degree of variability is evident, an indication for different sources and processes responsible for NPF across the Arctic. Comparing these values across the globe (Kerminen et al., 2018) shows that processes in the Arctic are significantly slower.

The observations presented here are based on a selection of studies, which may not be complete, that is, other precursors may also contribute to NPF and growth. For example, iodine has been detected in particles at Alert and during a growth event close to the Eastern coast of Greenland (Allan et al., 2015; Sirois & Barrie, 1999) but its role for NPF and/or growth remains unclear.

3. Conclusions and Outlook

These recent observations reveal that Arctic NPF and growth involve a complex mixture of chemical compounds (SA, NH_3, IA, MSA, organics), and their contributions vary with location and season (Figure 1). While precursors mostly originate from natural sources, anthropogenic contributions exist in specific locations. The exact mechanisms of NPF and early growth have been explored only in three locations, that is, NA (Beck et al., 2020), VRS (Beck et al., 2020; Sipilä et al., 2016), and in the central Arctic Ocean (Baccarini et al., 2020). Everywhere else, information on chemical compounds was inferred from indirect measurements and modeling. A key question is the source of organics, which can come from the sea surface microlayer (Mungall et al., 2017), anthropogenic sources (Kolesar et al., 2017), and terrestrial vegetation (Beck et al., 2020). BVOC emissions from tundra-type vegetation have only been scarcely investigated and deserve dedicated studies. Similarly, the iodine precursor sources remain unknown as well as the location-specific sources of ammonia. Additionally, many locations in the Arctic have not been studied: further important sources and compounds could be revealed. Investigations have mainly focused on spring and summer, but

NPF and growth are also observed in the absence of sunlight. For the central Arctic Ocean, the 13-months drift of the MOSAiC campaign between 2019 and 2020 (Figure 1), with a CI-APi-ToF, will shed light on the seasonal cycle.

The relevance of NPF and growth for the Arctic CCN budget remain to be assessed. While individual in situ observations demonstrate the growth of nucleation mode particles into CCN size (Abbatt et al., 2019; Baccarini et al., 2020; Beck et al., 2020) a systematic evaluation is complicated. Models are required to quantify the contribution of NPF to the Arctic CCN budget and estimate the sensitivity of low-level clouds to NPF-formed CCN. However, not all relevant nucleation mechanisms are implemented in relevant models (e.g., IA nucleation) and fluxes of precursors are highly uncertain. Equipping models with these processes would result in a better quantification of their effects on the Arctic surface energy budget. With enhanced models, the effect of a warming Arctic on NPF processes could be investigated, contributing to better predictions of future climate scenarios.

Data Availability Statement

Data were not used, nor created for this research.

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