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Chemical Composition and Mixing State of Wintertime Aerosol from the European Arctic Site of Ny-Ålesund, Svalbard

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Abstract

The Arctic is rapidly warming, and aerosol plays an increasingly important role by scattering and absorbing sunlight and by participating in cloud formation. Their optical and cloud-forming properties depend on mixing state and chemical composition, but observations of these features remain limited. This study comprehensively characterizes 25,254 individual particles collected at Ny-Ålesund, Svalbard (November -December 2020), using micro-spectroscopy techniques to investigate their size, morphology, mixing state, and chemical composition. Fresh sea salt aerosols (SSA) were identified as the most abundant (~85%), of the total observed aerosol population, with potential sources from sea spray and blowing snow. Air masses originating from the Arctic Ocean surrounding Svalbard likely contribute to increased concentrations of submicron “Fresh SSA” particles. “Aged SSA” particles (7.4%) are enriched in sulfur and nitrogen, compared to “Fresh SSA”. These elevated ratios may result from various atmospheric aging processes, including the uptake of sulfuric and nitric acids. Our results suggest that aged SSA, with sizes larger than 300 nm, likely underwent chlorine depletion by sulfuric and nitric acids during transport. Additionally, elemental analysis reveals that both fresh and aged SSA can mix with dust particles, regardless of the SSA size (49.9% in submicron size and 50.1% in supermicron size, respectively). Dust particles are efficient ice-nucleating particles (INPs), and SSA is known to act as cloud condensation nuclei (CCN), therefore, their mixtures may inherit both properties. The non-negligible number (4.4%) of SSA-dust mixtures underscores the importance of these particles as potential sources of CCN and INP in the Arctic atmosphere.

Key words: Arctic, Arctic aerosol, single particle analysis, elemental composition, mixing state.

1 Introduction

The warming rate in the Arctic exceeds that of the global average by two to four times, with the Arctic environment experiencing dramatic changes.¹⁻³ This phenomenon, known as Arctic amplification, has gained significant attention in recent decades. Atmospheric modeling studies have highlighted tropospheric aerosols as significant contributors to the rapid warming observed in the Arctic.⁴⁻⁷ However, the existing knowledge gaps in aerosol sources, sinks, as well as uncertainties in aerosol-cloud interactions pose significant challenges in accurately simulating the current and future atmosphere of the Arctic.^{8,9}

Sea spray aerosol (SSA) is one of the dominant component of Arctic aerosols, primarily originating from the nearby Arctic Ocean.¹⁰⁻¹⁴ SSA can directly scatter solar radiation, act as cloud condensation nuclei (CCN) for the formation of cloud droplets, and is also a source of reactive halogens.¹⁵⁻²⁰ Traditionally, SSA production has been attributed to sea spray generated by wave breaking over open ocean waters, as well as from local sources, such as ice leads and frost flowers in the Arctic.^{21,22} Recent modeling studies and field observations also suggest blowing snow as a significant source of SSA in polar regions.^{14,23-25} SSA from blowing snow is normally generated from the sublimation of the snow grains blown into the atmosphere.^{14,25-27} During windy episodes, falling or previously fallen snowflakes can bounce and shatter into smaller snow crystals.^{28,29} During a blowing snow occurrence, snow grains can be lifted into the atmosphere by turbulent eddies.^{30,31} Blowing snow was observed more than 20% of the time from November 2019 to April 2020 in the central Arctic.²³ Although blowing snow frequently occurs in the Arctic, direct observational studies focusing on the size and chemical composition of blowing-snow-generated SSA remain very limited.^{12,32} Therefore, gaining a deeper understanding of the contributions from open ocean sources and blowing snow to the Arctic SSA burden is essential for advancing knowledge of SSA's impacts on the region's environment system.

Additionally, SSA can interact with other atmospheric particles like dust, leading to the formation of complex mixtures that influence cloud properties and climate dynamics.^{10,13,15,33} Recent studies have suggested the possible presence of mixtures of mineral dust and sea salt at Ny-Ålesund, Svalbard. For instance, Geng et al. (2010) observed 3 out of 61 particles as mixtures of aged SSA and dust in summer at Ny-Ålesund.³³ Weinbruch et al. (2012) found that mixtures of sea salt and silicates present in both fine and coarse mode with the mixed particles occurred predominantly in the size fraction > 0.5 μm aerodynamic diameter.¹⁰ Song et al.¹³ identified mixtures of mineral dust and sea salt occurring from June to October at Gruvebadet, Svalbard, with a peak in accumulation mode $\sim 0.7 \mu\text{m}$ and two peaks in coarse mode (3-4 μm and 12-14 μm). Adachi et al. (2022) presented an elemental map of a dust particle mixed with sea salt as small as 100 nm collected from a cloud residual sample in wintertime Zeppelin, Svalbard.¹⁵ Dust particles can act as ice-nucleating particles (INP), thereby altering cloud properties and lifetime.³⁴ Both high-latitude local dust emissions and long-range transport of windblown dust from low-to mid-latitudes can contribute to the Arctic dust population.³⁴⁻³⁷ A recent study found that simulated dust emission fluxes in the Arctic (above 60°N) increased by 20% from 1981–1990 to 2011–2020.³⁸ Given the increasing emissions of SSA and dust due to factors like sea ice retreat and enhanced windblown local dust events,³⁹⁻⁴² it is crucial to study SSA and dust in the Arctic to better understand their role in cloud microphysics and climate feedback mechanisms.

At Ny-Ålesund in Svalbard, sources and sinks of aerosol particles exhibit strong seasonality.⁴³⁻⁴⁶ Primary particles can originate from natural sources such as oceans, contributing to sea salt, organic aerosol, and primary biological particles, or from high-latitude glaciers and ice-free terrains, contributing to mineral dust emissions within the Arctic.^{10,34,47,48} The variety of processes in this region that govern the abundance of aerosols, as well as their chemical and physical properties, makes their role in the Arctic atmosphere particularly complex to understand.⁴⁹

In this study, microscopy analysis was conducted to improve the understanding of morphology, chemical composition, and sources of Arctic aerosols including SSA and dust. Observations were performed at the Gruvebadet atmospheric laboratory, situated in Ny-Ålesund, at the base of Zeppelin Mountain. Its

geographic location minimizes the influence of the town's activities due to the prevailing wind patterns, making Gruebadet an ideal site to represent remote marine Arctic conditions.⁵⁰ Through a comprehensive analysis of Arctic aerosol samples and integration of air mass back-trajectory analysis, this study aims to understand: (1) the chemical composition and relative abundance of Arctic aerosols, and (2) the sources of SSA and dust at the European Arctic site Ny-Ålesund. This study contributes to a better understanding of the morphologies, size distribution, and chemical composition of Arctic aerosols, with implications for their roles as CCN and INP in determining Arctic cloud properties.

2 Methods

2.1 Sampling site and particle sampling

Aerosol particles were collected at the Gruebadet Atmospheric Laboratory (GAL, 78.918° N, 11.895° E, 61 m asl), located about 1 km south of Ny-Ålesund village in the Kongsfjorden, Svalbard (Norway) (Figure S1). A comprehensive site description is available in Gilardoni et al (2023).⁵¹ The GAL site is part of the Ny-Ålesund Research Station and Svalbard Integrated Observing System network (SIOS). Meteorological data were collected at the Climate Change Tower (CCT) and the Automated Nivometric Station (ANS). CCT is located about 1 km away from GAL, and ANS is situated approximately 80 meters from GAL. The close distance of CCT and ANS to GAL makes these measurements highly representative of local conditions at GAL.⁵²

For offline microscopy and spectroscopy analysis, all samples were collected onto TEM B-film grids (300 mesh, Ted Pella, Inc.). Samples were obtained through the Total Aerosol Inlet at GAL, with no size cut. A four-stage Sioutas Personal Cascade Impactor (SKC, Inc.) was used for sample collection, operating at a flow rate of approximately 9 L/min. For each sample, particles were collected onto four different stages with 50% lower cut-off aerodynamic diameters of 2.5 µm, 1.0 µm, 0.5 µm, and 0.25 µm, respectively.⁵³ Nine sets of samples (sample ID: M2, M3, M4, M5, M6, M7, M8, M10, M11; 9 sets × 4 stages = 36 samples) were collected from Nov 5th to Dec 29th in 2020. The sampling period and meteorological conditions during each sampling period are shown in Table 1. The samples were subsequently stored under dry and dark conditions. However, transformations between sample collection and data analysis cannot be excluded. This limitation is inherent in conducting offline analysis on samples collected during field campaigns. Aerosol size distribution was monitored at a 10-min interval by a Scanning Mobility Particle Sizer (SMPS, TSI 3034) at GAL in the particle diameter range from 10 to 500 nm.

Table 1. Collection time, average, and standard deviation of ground-level (2 m) ambient temperature, RH, and wind speed during each sample collection period.

Sample ID	Start time (UTC)	End time (UTC)	Average T(K)	Average RH (%)	Wind Speed (m/s)
M2	11/5/20 10:12	11/7/20 12:08	274.0±0.9	75.3±4.5	1.2±0.5
M3	11/9/20 8:43	11/10/20 0:00	266.1±0.4	78.6±2.7	4.3±1.2
M4	11/12/20 12:38	11/13/20 15:19	273.9±1.2	78.2±7.2	6.3±3.0
M5	11/20/20 7:43	11/21/20 11:42	265.2±2.2	73.7±3.0	1.6±1.0
M6	11/24/20 12:55	11/25/20 9:19	267.3±0.5	76.2±3.6	3.5±2.2
M7	12/1/20 8:06	12/3/20 8:21	260.4±1.8	67.4±5.5	5.5±1.9
M8	12/7/20 8:43	12/8/20 8:30	265.9±1.7	70.9±9.2	5.3±2.0
M10	12/21/20 8:17	12/22/20 13:45	268.7±0.7	68.5±2.1	10.6±1.6
M11	12/28/20 14:49	12/29/20 13:14	271.1±1.7	69.2±3.6	9.6±2.3

2.2 Meteorological data and back trajectory analysis

Meteorological data (temperature, relative humidity (RH), wind direction, and wind speed) were continuously measured at the CCT at 1-minute time resolution, and hourly averages were calculated and used for analysis.⁵² Ambient temperature and RH were continuously monitored by a Vaisala thermo-hygrometer (model HMP45AC), installed within a suitable shield to prevent solar radiation-induced heating. A Young Marine wind sensor (model Wind Monitor 05106) was used to measure the wind speed and direction. Both instruments were deployed at four levels on the CCT (2, 5, 10, and 33 m above ground level (AGL)), facing north.⁵² Snow depth was measured by an ultrasonic snow-meter sensor (model NESA-LU06, resolution = 1mm, accuracy < 0.1% full-scale value at 20°C) at ANS. The snow data was downloaded from the online repositories provided by Scoto et al. (2023).⁵⁴

Figure 1 shows the meteorological conditions during the campaign. As shown in Figure 1a and Table 1, sampling took place under a range of meteorological conditions. Samples M2 and M4 were collected during warmer periods (274 ± 0.9 K), while M7 was collected at the coldest temperature (260.4 ± 1.8 K). As shown in Figure 1b, samples M2 through M6 were collected under higher RH (mean RH > 75% across these sampling periods) compared to the others. The shift in mode diameter from below 100 nm to 500 nm in Figure 1c indicates that M8 was sampled at the end of a new particle formation and growth event. Furthermore, M2, M3, and M4 exhibited mode diameters smaller than 100 nm, while M5, M6, M7, M8, M10, and M11 exhibited mode diameters larger than 100 nm. Figure 1d displays the snow depth measurements. The ANS was installed in the field on November 7, 2020, and the snowpack was 10-15 cm deep on that day. In contrast, snow depth was less than 0.01 cm during the collection period for M4. Images reported by Scoto et al. in 2023 confirm that intense rainfall (15.9 mm) on November 12, prior to M4 collection, melted the snow layer completely, leaving the tundra exposed.⁵⁵ Thus, except for M4, the ground was snow-covered during the sampling period, with snow starting to accumulate on November 16, 2020. Figure 1e shows the wind speed and wind direction. The color-coded line represents measured wind speed 10 meters above ground level, while the color denotes measured wind direction (0–359.9°), where 0° corresponds to north. The black line indicates the reference wind speed necessary for a blowing snow event (U_t) to occur, as determined by Eq. (1)

$$U_t \text{ (m/sec)} = 6.975 + 0.0033(T + 27.26)^2 \quad (1)$$

where T (°C) is the air temperature measured at 2 m.^{12, 56, 57} As described by Li and Pomeroy et al., blowing snow events are defined as periods when the wind speed measured at 10 meters above the ground (U_{10m}) surpasses U_t (i.e., $U_{10m} \geq U_t$).⁵⁷ In Figure 1e, the periods during which the 10 m wind speed (U_{10m}) exceeds the reference wind speed (U_t) coincide with the periods in Figure 1c where the mode diameter shifts toward smaller sizes. According to the blowing snow definition and snow depth measurement, two samples—M10 and M11—were collected during a blowing snow event. Wind patterns shown in the wind rose plot (Figure S2) reveal prevailing winds from the southwest and east throughout most sampling days. While southwest winds transport from the inland side of Svalbard, westward winds originate from the gulf on the right side of Ny-Ålesund, as indicated by the geographical location of the sampling site (Figure S1). Additionally, the wind rose plot in Figure S2 illustrates that M4 experienced predominant southwest winds, M11 encountered predominant east winds, and M10 experienced winds originating from both directions.

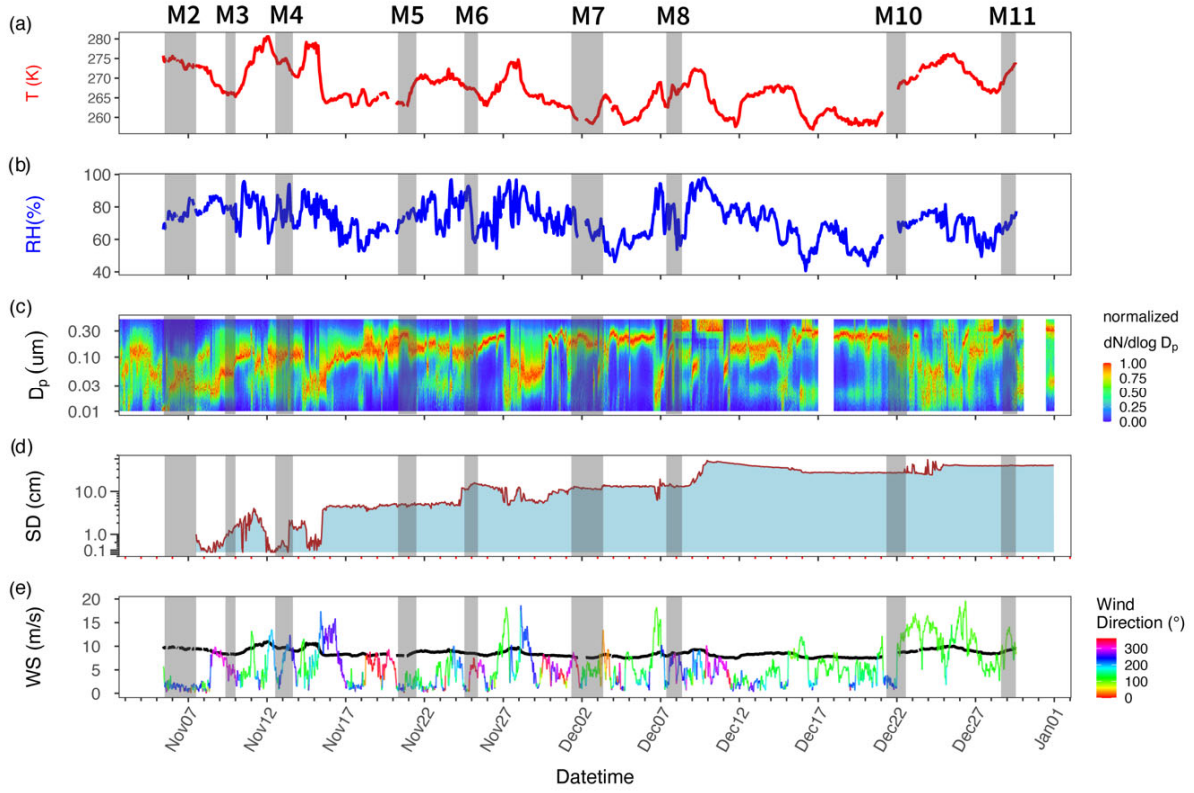


Figure 1. Ground-based measurements conducted from November 5 to December 29, 2020, at Gruedadet, Svalbard. The measurements include various parameters: (a) ambient temperature (red); (b) ambient relative humidity (blue); (c) normalized aerosol size distribution measured by SMPS. Color scale represents the values of normalized size distribution; (d) snow depth (SD, cm); and (e) wind speed and wind direction. The black line represents the reference wind speed U_t for blowing snow and the colored line denotes the measured wind speed at 10 m AGL with the color representing the wind direction. Shaded gray areas indicate the collection periods for each sample.

Air mass backward trajectories (48 hr duration) were simulated for each sample using the NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT) model with the global data assimilation system (GADS) $1^\circ \times 1^\circ$ meteorological data.⁵⁸⁻⁶⁰ The ending altitude for each trajectory is set to be the altitude of the GAL site, which is 4 m above the ground level (AGL) and 18 m above the sea level (ASL). The back trajectory of each sample is presented in Figure 2. Figures 2a–2i show the back trajectories for samples M2, M3, M4, M5, M6, M7, M8, M10, and M11, respectively. As depicted in Figure 2, GAL samples collected in this study were influenced by two transportation types: (1) air masses transporting from the Arctic Ocean (M4, M5, M6, M7, M10), and (2) air masses transporting from both inland and oceanic regions (M2, M3, M8, M11). Moreover, the transport of aerosols from land-based sources in M2 and M11 is predominantly from Eurasia and Russia, consistent with long-term observations at the Zeppelin station.^{44, 61}

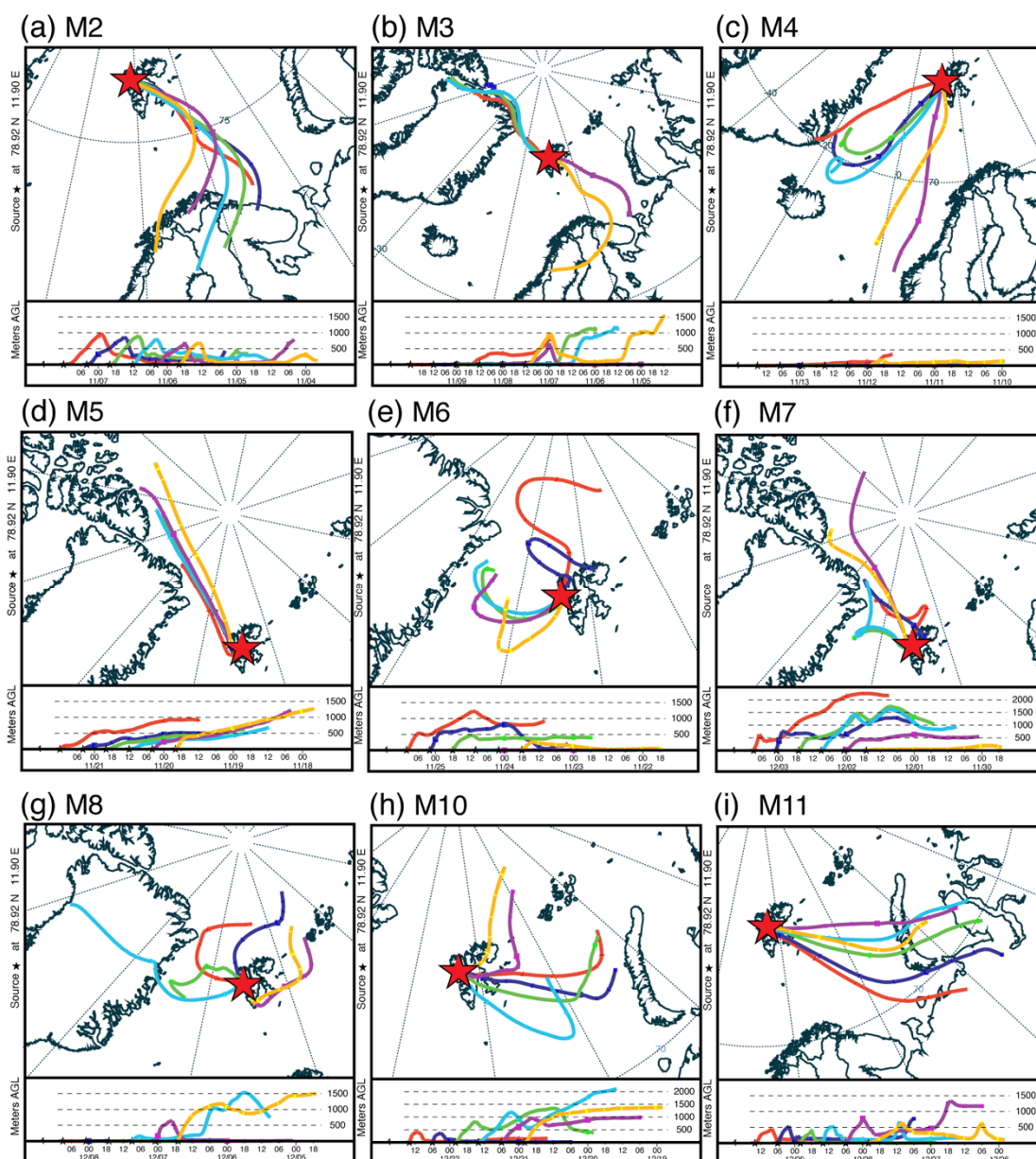


Figure 2. 48-hour NOAA HYSPLIT back trajectories for (a) M2, (b) M3, (c) M4, (d) M5, (e) M6, (f) M7, (g) M8, (h) M10 and (i) M11 sample. The red stars mark the location of GAL (latitude: 78.918, longitude: 11.895, height: 4 meters AGL/18 meters ASL). For each simulation, six separate back trajectories—each starting 8 hours apart from the end time of sample collection—are calculated in order to cover the entire sampling period (about 2 days). The 6 trajectories in each sample are shown in different colors. Each trajectory spans a 48-hour duration, and the height of the air parcel is reported with the corresponding date and time, shown in meters AGL.

2.3 Chemical imaging and single particle analysis

The particles collected from the GAL were analyzed with computer-controlled scanning electron microscopy, coupled with energy-dispersive X-ray spectrometer (CCSEM-EDX) and scanning transmission X-ray microscope (STXM) combined with Near Edge X-ray Absorption Fine Spectroscopy Structure (STXM-NEXAFS) to probe their physicochemical properties.^{62, 63} Microscopy images were obtained using an environmental SEM (ESEM, Quanta 3D, Thermo Fisher) coupled with a field emission gun operated at 20 kV and 480 pA. An EDX spectrometer (EDAX, Inc.) coupled with CCSEM was used to

determine the relative atomic percentages of 15 elements (C, N, O, Na, Mg, Al, Si, P, S, Cl, K, Ca, Mn, Fe, and Zn), and these elemental fractions were used for particle classification (for details see Section S1 in SI).

In this study, we focused on particles larger than 300 nm to avoid misrepresentation of particles due to size resolution and particle impaction artifact (See Section S1 and Figure S3 in SI). We adopted a hybrid approach combining rule-based classification and K-means clustering to classify particles (Figure S4 and Figure S5).⁶⁴⁻⁶⁶ The dataset utilized for classification encompassed data points derived from all 36 samples (9 samples each across 4 stages).

Overall, five classes were identified: “Carbonaceous”, “Sulfate”, “Fresh SSA”, “Aged SSA”, and “SSA and Dust mixture”. In this study, “Fresh SSA” refers to particles that have undergone minimal atmospheric aging as indicated by limited chlorine depletion (higher average Cl/Na ratio of 1.1 ± 0.3 observed in this group, in contrast to the significantly lower mean Cl/Na ratio of 0.1 ± 0.2 found in “Aged SSA” particles), regardless of whether they originate from seawater or blowing snow. It is important to note that “fresh” in this context pertains to the particle’s chemical composition rather than its actual atmospheric residence time or transport history. Indeed, some “Fresh SSA” particles observed in this study may have been transported over long distances but retained their chemically fresh signatures. “Aged SSA” particles show significant chlorine depletion (average Cl/Na of 0.1 ± 0.2), compared to the Cl/Na ratio of 1.2 in fresh seawater.⁶⁷ Previous study such as Chi et al.⁶⁸ have shown that morphology can provide valuable complementary information for distinguishing fresh and aged SSA, with fresh SSA typically exhibiting smooth, cubical shapes and aged SSA often displaying irregular structures due to atmospheric processing. While morphology was not used as a classification criterion in this work, visual inspection of representative SEM images suggests that particles with high Cl/Na ratios tend to exhibit smoother, more cubical morphologies, consistent with fresh SSA, whereas particles with depleted Cl often appear more irregular and internally mixed. Future studies combining chemical composition with morphological parameters could improve classification accuracy and reduce ambiguities. “SSA and Dust mixture” particles exhibit combined elemental compositions of sea spray aerosol and mineral dust. “Sulfate” particles are characterized by high sulfur content, often associated with long-range transport or regional pollution. “Carbonaceous” particles primarily consist of elemental or organic carbon, likely originating from combustion or natural biogenic sources. The number fraction and counts of each CCSEM-EDX particle class in each sample are provided in Table S1 and Table S2, respectively. Furthermore, on each day, the CCSEM-EDX data from stage A, B, C, and D were merged into one dataset for the calculation of size-resolved fraction of each class. The chemical composition of CCSEM-EDX in Figure 3a is plotted for particles collected on all four stages (See Section S1 in SI).

The STXM-NEXAFS was utilized to provide insight into the carbon speciation such as chemical bonding and functional group of carbonaceous components and to evaluate the mixing state of organic carbon, elemental carbon, and inorganic species.^{63, 69} STXM-NEXAFS experiments were conducted at beamline 5.3.2.2 of the Advanced Light Source (ALS) at Lawrence Berkeley National Laboratory at the carbon K-absorption edge (280–320 eV). STXM-NEXAFS data were only obtained for stage D samples to ensure X-ray transmission. A 25 nm Fresnel zone plate was used to focus a monochromated X-ray beam to a 30-35 nm spot size during raster scanning. The comprehensive description of the working principle of STXM-NEXAFS and the classification rules can be found in previous studies.⁷⁰⁻⁷³ Briefly, STXM classifies particle regions at four specific energies (pre-edge (278.0 eV), sp^2 (285.4 eV), -COOH (288.5 eV), and post-edge (320.0 eV)) into three main categories: OC ($OD_{288.5} - OD_{278.0} > 0$), IN ($OD_{278.0} / OD_{320.0} > 0.50$), or EC ($OD_{285.4} / (OD_{320.0} - OD_{278.0}) > 0.35$).⁷⁴ Then the identified three categories (OC, IN, EC) were grouped to yield six classes based on the internal mixing between OC, IN, and EC: (1) OC, (2) IN, (3) OCEC, (4) OCIN, (5) OCINEC. The number counts of each STXM-NEXAFS particle class in each sample are provided in Table S3. Additional details on particle and organic feature classification are provided in Section S2.

3 Results and Discussions

3.1 Chemical composition of Arctic aerosols

The normalized number fraction of the 5 particle classes derived from CCSEM-EDX and 5 STXM-NEXAFS based on mixing state of OC, EC, and IN for each sample is shown in Figure 3a and 3b, respectively. As shown in Figure 3a, "Fresh SSA" is the most common particle type, making up 84.8% of all particles analyzed by CCSEM-EDX in all nine sample sets (M2~M11). "Aged SSA" (7.4%) is the second most common type of wintertime Arctic aerosol at Ny-Ålesund in our samples. The "SSA and Dust mixture" class makes up 4.4% of the total particles analyzed and is found in all samples. "Carbonaceous" particles are also present in all samples, with a total fraction of 2.9%. "Sulfate" particles are detected in every sample but are less abundant, with a total fraction of 0.5%. Representative SEM images of each sample showing particle morphologies are shown in Figure S6.

Samples M3, M4, M5, M6, and M10 have high fractions of "Fresh SSA" particles, all above 80%. In contrast, sample M2 has the lowest "Fresh SSA" fraction at 40.2% and has the highest fractions of "Aged SSA" (33.2%) and "Carbonaceous" particles (16.1%). Sample M7 has the highest fraction of the "SSA and Dust mixture" at 12.0%. Finally, samples M8 and M11 show relatively high fractions of "Aged SSA" at 12.1% and 17.2%, respectively.

Figure 3b shows that 85% of particles analyzed by STXM-NEXAFS consist of "OCIN (organic carbon mixed with inorganics)", dominating in all sample sets. This finding is consistent with the SEM images (Figure S6) and STXM imaging results (Figure S7), which indicate that most particles have an organic coating on an inorganic core. Carbon maps (Figure S7) show relatively higher EC in the M11 sample. The air masses originated from inland regions (Eurasia and Russia), likely contributing anthropogenic sources and resulting in relatively higher EC content. Furthermore, Figure S7 shows that organic coating was commonly found in all samples, forming both partially-engulfed and core-shell morphologies with inorganic salts. Notably, both fresh and aged SSA can hold an organic coating, as shown in Figures S8 and S9. One possible explanation for the organic coating on "Fresh SSA" particles is that SSA particles are typically emitted from sea spray through wave breaking at the sea-air interface, which contains both organic compounds and salt.^{75, 76} Particles may acquire additional organic coatings during atmospheric transport. A comprehensive review by Li et al.⁷⁷ highlights that aqueous-phase secondary organic aerosols can form as coatings on dust and salt particles during atmospheric transport. The sea surface microlayer is enriched with organic compounds like amino acids, carbohydrates, fatty acids, and phenols. When bubbles burst at the ocean surface, these organic materials are incorporated into SSA particles, leading to organic coatings on fresh SSA.⁷⁸⁻⁸⁰ Thus, the "OCIN" particles found in this study can correspond to all SSA-related classes, including "Fresh SSA", "Aged SSA", and "SSA and Dust mixture". We further examined the organic coatings on particles by analyzing the organic volume fraction (OVF) derived from STXM measurements. As shown in Figure S10, particles across all samples are generally characterized by relatively moderate OVF (< 60%), with most falling within the 20–60% range. Organic contributions are most pronounced in the 300 nm–1 μ m size range, where 'Aged SSA' and 'Carbonaceous' particles dominate. In contrast, supermicron particles (>1 μ m) typically exhibit lower OVF (<60%). Notably, the M8, M10, and M11 samples display relatively lower organic content compared to the other samples.

Furthermore, Figures 4a–4i show the CCSEM-EDX derived size-resolved chemical composition of samples M2, M3, M4, M5, M6, M7, M8, M10, and M11, respectively. "Carbonaceous" and "Sulfate" particles are mostly found in sub-micron size ranges (< 1 μ m). "Fresh SSA" and "Aged SSA" particles are present in both fine and coarse modes across different samples, with sizes ranging from 300 nm to 8 μ m. As shown in Figure 4 and Figure S11, in M2 and M11, "Fresh SSA" is only observed in particles larger than 0.5 μ m, whereas in the other samples, "Fresh SSA" is present across all size ranges. It is interesting to note that the mixtures of SSA and dust dominate the submicron size range in the coarse-mode samples (stages A and B) of M7A and M7B (Figure S11), likely due to the higher density of dust or potential impaction artifacts. "Aged SSA" particles are present in both fine and coarse modes but are more concentrated in the

fine mode. "SSA and Dust mixture" particles are mainly found in the submicron size range (300 nm - 1 μ m).

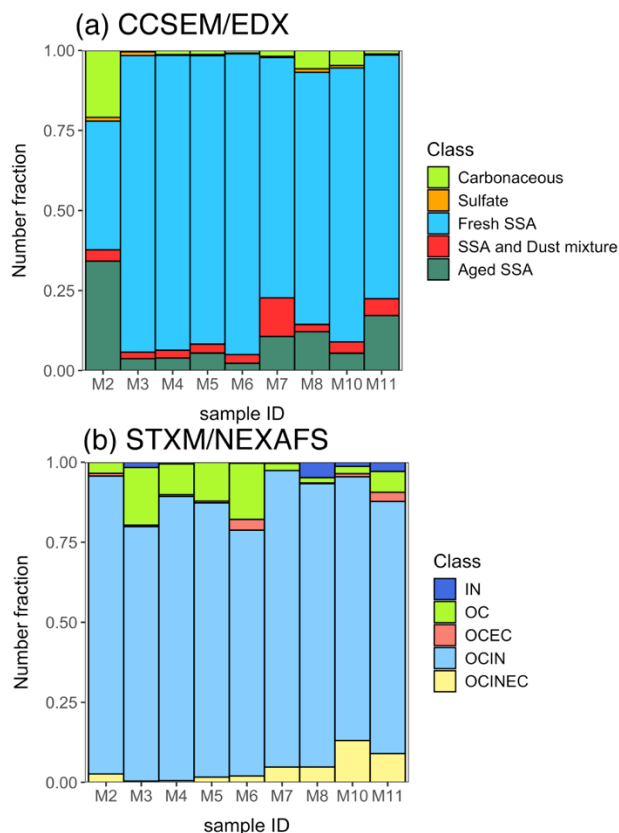


Figure 3: Summary of chemical composition of each sample from (1) CCSEM-EDX and (2) STXM-NEXAFS analysis. (a) Normalized number fractions of five CCSEM-EDX classes by combining measurements from all four stages (A, B, C, D) in each sample; (b) normalized number fraction of 5 STXM-NEXAFS classes from stage D sample.

While sea spray has long been studied as a source of SSA, recent studies have identified the sublimation of wind-blown saline snow as an additional source of SSA in polar regions.^{12, 14, 23, 26 26} We observed only minor contributions of Mg and Ca in SSA. Least squares linear regression of our dataset yielded weaker relationships due to other source or atmospheric processing, $[Mg^{2+}] = 0.029[Na^+] + 2.230$ ($R^2 = 0.033$) and $[Ca^{2+}] = -0.004[Na^+] + 0.954$ ($R^2 = 0.001$). This result indicates that elemental correlations in our samples are far less pronounced than those reported by Hara et al.²² In this study, we focus on samples M2 and M11 to explore the influence of blowing snow on Arctic aerosols because of their distinct wind speed (Table 1). Both samples were collected under similar temperatures, relative humidity, and broadly comparable transport pathways (based on HYSPLIT back trajectories). Figure 4a- 4i illustrates that in the supermicron ($> 1 \mu$ m) size range, M11 has a higher relative abundance of "Fresh SSA" and a lower relative abundance of "Aged SSA" compared to M2. The presence of supermicron SSA in M2, despite lower wind speeds, can indicate the contribution of transport from marine/sea ice regions. Table S5 shows that the Cl/Na atomic ratio in supermicron "Fresh and aged SSA" particles is about 1.0 ± 0.37 in M11 compared to 0.83 ± 0.44 in M2, suggesting minimal chlorine depletion of supermicron-sized SSA in M11 (seawater Cl/Na is 1.2).⁶⁷ This minor depletion in supermicron SSA implies that these larger SSA particles likely originate from local sources under blowing-snow wind speeds. The observed contribution of blowing snow to supermicron SSA particles with minor chlorine depletion is consistent

with recent findings that blowing snow is a significant source of supermicron SSA.¹² While blowing snow can be a potential source, it might not be the only source of these particles. Another recent study has shown that blowing snow sublimation produces high concentrations of fine-mode sea salt aerosols (aerodynamic diameters below 300 nm) compared to those generated by sea spray emissions.²³ While the observed strong wind episode in this study suggests blowing snow as a possible emission source of supermicron SSA, further investigation into the number and mass concentration of submicron and ultrafine SSA is necessary for a more comprehensive understanding of the role of blowing snow as a source of SSA.

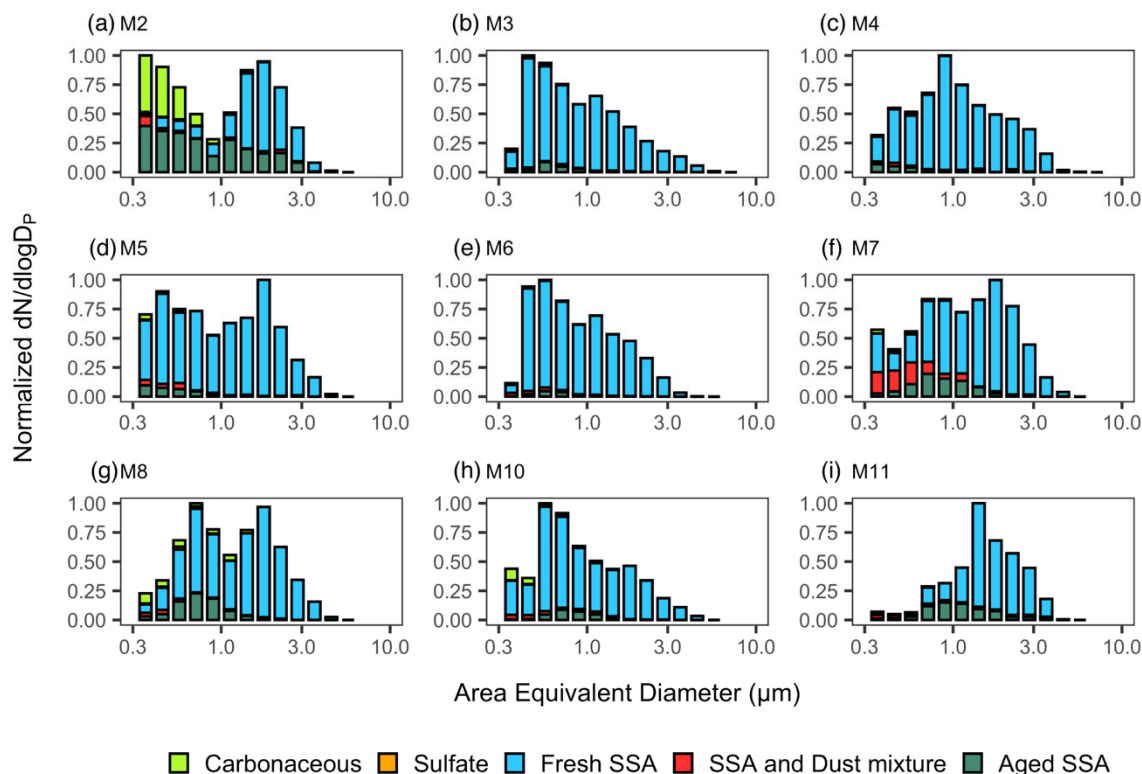


Figure 4: Size-resolved chemical composition of CCSEM-EDX analysis for (a) M2, (b) M3, (c) M4, (d) M5, (e) M6, (f) M7, (g) M8, (h) M10 and (i) M11 samples. For each sample, the particles' measurements of chemical composition from four stages are plotted against the area equivalent diameter. The particle counts of each class are normalized to the largest value among all size bins.

3.2 Fresh SSA

The predominance of "Fresh SSA" in wintertime Arctic aerosols underscores its significant role in the Arctic environment and regional energy balance. Gaining insight into the emission sources and formation mechanisms of "Fresh SSA" is essential for understanding interactions of these particles with the Arctic atmosphere. However, different transport patterns suggest that "Fresh SSA" in the Arctic may come from various locations of the sea such as open ocean or the ocean with covered ice. As shown in Figure S12, while most of the Arctic Ocean north of Svalbard was covered in ice, a small area surrounding the Svalbard islands remained unfrozen during November and December in the winter of 2020. Figure 2a-2i shows that the air masses came from various directions and locations but consistently passed over open

seawater before reaching the observatory on all sampling days, allowing for the transport of SSA emitted from sea spray in those open water regions.

According to Table 2, among transportation type (1) samples which were influenced by the Arctic Ocean emission, M4, M5 and M7 has similar distributions of “Fresh SSA” particles between the submicron (300 nm–1 μ m) and supermicron (>1 μ m) size ranges, with submicron fractions comprising 40%, 37%, and 35%, respectively. In contrast, M6 and M10 have higher proportions of submicron “Fresh SSA” particles (59% and 58%, respectively) compared to M4, M5, and M7. HYSPLIT back trajectory analysis reveals that the air masses associated with M6 and M10 originated from similar regions near Svalbard Island but traveled shorter distances compared to the air masses in M4, M5, and M7. For group (2) samples which were influenced by both continental and marine emissions, M2 and M11 have the lowest submicron “Fresh SSA” fractions of all samples, at 16% and 9%, respectively. In comparison, M3 and M8 have submicron fractions of 30% and 39%. HYSPLIT trajectory in Figure 2a-2i shows air masses during the M2 and M11 sampling period had a similar origin near northern Europe, while M3 and M8 were both influenced by transportation from Greenland.

In summary, the size distribution of “Fresh SSA” particles are likely influenced by the transport patterns of air mass. When the GAL site is mainly affected by air masses from the nearby Arctic Ocean surrounding Svalbard, the relative fraction of submicron (300 nm–1 μ m) “Fresh SSA” particles tend to be higher (>50%) than that of supermicron (>1 μ m) particles. In contrast, when the site is primarily influenced by transport from continental northern Europe, “Fresh SSA” is dominated by supermicron particles, with relative fractions exceeding 80%.

Table 2. Number of submicron (300 nm–1 μ m) and supermicron (> 1 μ m) “Fresh SSA” particles in each sample, the corresponding relative fraction concerning the total “Fresh SSA” particles in each sample is in parentheses.

Sample name	# of Submicron “Fresh SSA” (300 nm–1 μ m)	# of Supermicron “Fresh SSA” (> 1 μ m)
M2	60 (16%)	313 (84%)
M3	1123 (30%)	2725 (70%)
M4	611 (40%)	906 (60%)
M5	623 (37%)	1059 (63%)
M6	2683 (59%)	1852 (41%)
M7	770 (35%)	1463 (65%)
M8	1026 (39%)	1630 (61%)
M10	1627 (58%)	1163 (42%)
M11	164 (9%)	1606 (91%)

3.3 Aged SSA

“Aged SSA” accounts for 7.4% of the total analyzed particles in this study, making it the second most abundant particle type identified by CCSEM-EDX. Figure 5a shows the Cl/Na ratio for “Fresh SSA” and “Aged SSA”. “Fresh SSA” particles have a Cl/Na ratio close to 1, indicating minimal chlorine depletion, while “Aged SSA” exhibits a significantly lower ratio (0.1 ± 0.2), suggesting extensive chlorine depletion.

Figure 5b and 5c further illustrates that sulfur (S) and nitrogen (N) are enriched in "Aged SSA" compared to "Fresh SSA", indicating the incorporation of nitrate and non-sea-salt sulfates through chemical processing. The formation of "Aged SSA" is primarily driven by chlorine depletion, where sea salt particles react with $\text{H}_2\text{SO}_4/\text{SO}_2$ and HNO_3 during atmospheric processing. Acidic precursors like SO_2 , NO and NO_2 , originating from anthropogenic emissions in mid-latitudes, can be transported to the Arctic during wintertime and react with SSA particles.^{44, 81-83} These chemical reactions facilitate the conversion of $\text{H}_2\text{SO}_4/\text{HNO}_3$ to sulfate in deliquescent SSA through acid displacement, as widely proposed in previous studies.⁸⁴⁻⁸⁶ While this study primarily observed "Aged SSA" processed by sulfate and nitrate, it does not rule out the possibility of organic-driven aging. Since the analysis focused on particles >300 nm, smaller SSA particles that may have undergone organic processing could have been missed.

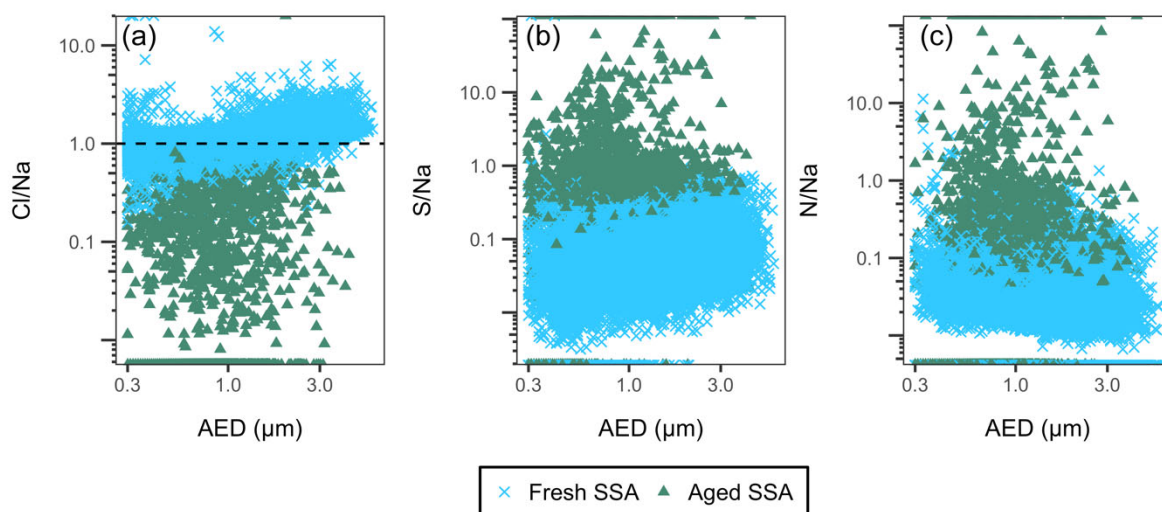


Figure 5. Elemental ratios measured by CCSEM/EDX of 21404 "Fresh SSA" particles and 1874 "Aged SSA" particles. (a) Ratio of chlorine to sodium Cl/Na, (b) ratio of sulfur to sodium S/Na, (c) ratio of nitrogen to sodium. The X-axis is area equivalent diameter (AED, μm). The black horizontal dashed line is at the Cl/Na ratio of 1, indicate chlorine is still largely preserved in the particle as NaCl.

M2, M7, M8, and M11 have a relatively higher fraction of "Aged SSA" than the average, with proportions of 34.3%, 10.7%, 12.1%, and 17.2%, respectively. Back trajectory analysis shows that air masses in these samples traveled longer distances and were influenced by both marine and inland sources. The longer travel distance allows SSA to undergo more atmospheric interactions, while air masses from inland sources may carry higher concentrations of atmospheric pollutants. As a result, samples influenced by both marine and inland air masses tend to have a higher "Aged SSA" fraction compared to those primarily influenced by marine sources. Moreover, the elemental ratios in Table S5 show that for "Aged SSA", submicron particles have a lower average Cl/Na ratio (0.08 in M2 and 0.03 in M11) compared to supermicron particles (0.11 in M2 and 0.12 in M11), indicating more extensive chlorine depletion in smaller particles. A two-sample t-test performed on pooled data across all samples confirms that this difference is statistically significant (mean Cl/Na = 0.15 for supermicron, 0.08 for submicron; $p < 0.001$; 95% Confidence interval for difference: 0.048–0.086), supporting the hypothesis that smaller particles experience greater chlorine depletion. This size-dependent trend is consistent with the increased aging of smaller SSA particles, as their limited chloride reservoir makes them more susceptible to depletion. Additionally, their higher surface-area-to-volume ratio enhances interactions with atmospheric acids and oxidants, accelerating chemical processing. Their longer atmospheric lifetime further prolongs exposure to aging mechanisms, allowing for more extensive modifications over time.⁸⁷⁻⁸⁹

3.4 SSA mixed with dust

SSA is found to be commonly mixed with dust, comprising 4.5% of the total analyzed particles. According to the elemental analysis shown in Figure 6, Cl/Na have values both greater and less than 1, indicating both fresh and aged SSA particles can mix with dust. In terms of size distribution, 79% of “SSA and Dust mixture” particles fall within the 300 nm–1 μm range, while 21% are larger than 1 μm . The size distribution observed here is notably smaller than that reported by Song et al.¹³ at the same site, where mixed particles exhibited a mode diameter of $\sim 3\text{--}4\ \mu\text{m}$. This discrepancy may reflect differences in particle sources, atmospheric processing, or measurement periods between the two studies. In the supermicron size range ($>1\ \mu\text{m}$), 70% of “SSA and Dust mixture” particles have Cl/Na >1 , suggesting dusts are primarily mixed with “Fresh SSA”, while the remaining 30% have Cl/Na <1 , indicating dusts mixing with “Aged SSA”. In contrast, for submicron particles ($<1\ \mu\text{m}$), the proportion is more balanced, with 51% mixed with “Aged SSA” and 49% mixed with “Fresh SSA”. This size-dependent trend is consistent with the overall chemical composition observed in the samples, where “Fresh SSA” dominates in the supermicron range, while “Aged SSA” becomes more prevalent in submicron sizes due to increased atmospheric aging and chlorine depletion in the submicron range. Representative SEM images and elemental maps of individual supermicron “SSA and Dust mixture” particles showing the morphology and mixing states can be found in Figure S13. We further examined particle processing by sulfur through analysis of the S/N ratios (Figure S14). A major fraction of particles exhibited an S/N ratio >0.5 , suggesting that a significant fraction of sulfur originates from non-nitrogenous sulfate species. A substantial fraction of particles in the SSA and Dust mixture class exhibit S/N <0.5 , suggesting that the excess nitrogen may originate from other inorganic or organic constituents.

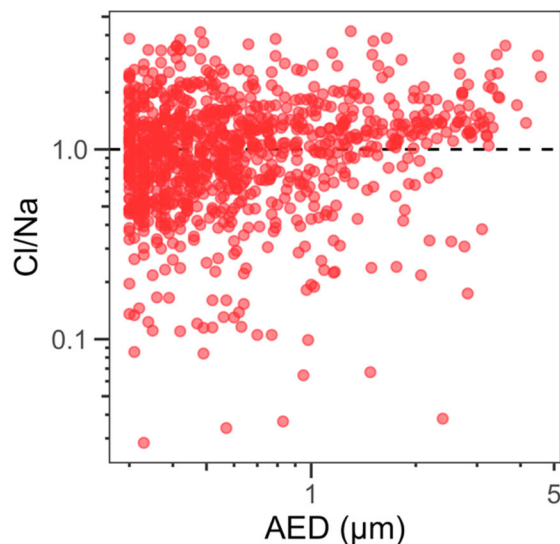


Figure 6. Elemental ratios measured by CCSEM/EDX of 1109 “SSA and Dust mixture” particles (red). X-axis is area equivalent diameter (AED, μm), y-axis is ratio of Cl/Na in log10 scale. The black horizontal line is at the ratio Cl/Na of 1, indicating minimal chlorine depletion.

We provide two potential formation pathways for the “SSA and Dust mixtures”: (1) these mixtures are formed during transport when SSA emissions interact with airborne dust, or (2) they may result from the uplift of snow containing previously deposited mineral dust in the snow surface. Dust particles can remain suspended in the atmosphere for several days, allowing long-range transport from source regions such as Asia and Europe.⁴² Model simulation also indicates that 3% of the global dust emissions originate from high-latitude dust sources within the Arctic.⁴⁷ In the Arctic, dust may originate from ice-free terrains or floodplains at high latitudes, where fine glaciofluvial sediment deposits can be exposed to wind, such as the western and northern coasts of Greenland, northern Eurasia, and coastal Alaska.^{34, 35, 90-95}

Since samples M10 and M11 were collected during blowing snow events, the "SSA and Dust mixture" particles in these samples are likely influenced by both pathways. In contrast, for the other samples, "SSA and Dust mixture" particles are more likely to be dominated by the first pathway. Among the samples not affected by blowing snow, M7 has the highest "SSA and Dust mixture" fraction (12%). M7 experienced the coldest temperature (260K) and lowest RH (67%) throughout the sampling period. And the combination of low humidity and cold air likely reduced dust removal processes such as wet deposition through precipitation or snow scavenging, allowing more dust to remain airborne and mix with SSA.^{96, 97} This suggests that under stable, dry atmospheric conditions, dust may persist longer in the Arctic atmosphere, increasing its likelihood of interacting with SSA.

4 Conclusions

This study presents a comprehensive analysis of the chemical composition and mixing state of individual Arctic aerosols collected in Ny-Ålesund, Svalbard. The dominance of SSA, particularly fresh SSA, highlights the significant impact of oceanic sources on Arctic aerosols. Backward trajectory analysis indicates marine emissions as a major source of SSA particles in the Arctic, and that transportation patterns can influence the "Fresh SSA" size distributions. Elemental analysis shows the potential of blowing snow as an SSA source in the wintertime Arctic by mobilizing and releasing the previously deposited SSA particles back into the atmosphere under high wind speeds. Moreover, a higher abundance of SSA particles with minimal chlorine depletion was observed during blowing snow events compared to low-wind periods. Thus, this study highlights the importance of incorporating blowing snow into atmospheric models as an SSA source to improve predictions of cloud dynamics, precipitation patterns, and the overall radiation budget in these sensitive environments.

Moreover, the common presence of organic coatings on both fresh and aged SSA particles, forming core-shell and partially-engulfed morphologies, underscores the significance of the organic-coated SSA morphology in the Arctic atmosphere. This organic-coated morphology can influence atmospheric aging processes and the particles' efficiency as CCN and INP.⁹⁸⁻¹⁰¹ The thickness and phase of the organic coating can significantly affect the diffusion timescale of gas-phase molecules (e.g., volatile organic compounds and ozone molecules), with thicker and viscous coatings significantly increasing the time required for diffusion into the particle, thus affecting the phase state of particles.^{102, 103} The particle coating with different phase states can significantly affect the heterogeneous chemistry by altering the surface properties of the particle, impacting the ability of gas-phase molecules to interact with and react on the particle's surface.¹⁰⁴⁻¹⁰⁷

The study also highlighted the common presence of "SSA and Dust mixtures" at all sizes with diverse morphologies and mixing patterns. Dust is considered an efficient INP and dominates the INP source in various regions of the world.¹⁰⁸⁻¹¹¹ Several studies have probed the INP composition in Arctic and identified mineral dust as a significant contributor to Arctic INP concentration.¹¹²⁻¹¹⁶ While SSA particles are occasionally found to act as INP, Si et al. compared the measurements of surface active sites density on SSA and mineral dust at three coastal marine sites including an Arctic site, and discovered that mineral dust might be the main source of INPs at all three sites at a freezing temperature of -25°C .^{99, 117, 118} However, the presence of "SSA and Dust mixture" might potentially alter the ice nucleation ability of SSA particles by providing more ice active sites and contributing significantly to the INP concentration in the Arctic leading to potential formation of ice clouds. Further investigations into these particles are warranted to systematically explore the mixing of dust and sea salt, thereby providing opportunities to quantify the effects of dust on SSA containing particles' ice nucleation ability. Overall, this study provides insights into the chemical composition of Arctic aerosols and their sources, contributing to our understanding of Arctic atmospheric processes.

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ASSOCIATED CONTENT

Supporting Information.

Detailed information on data quality control measures; chemical imaging using CCSEM-EDX; K-mean clustering and particle classification; methods to combine the CCSEM data from A, B, C, D stages; chemical imaging using STXM-NEXAFS; sea-ice edge data source.

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Notes

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