

Seasonal and interannual variability on the chemical composition of the Svalbard surface snowpack

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31 **Abstract**

32 Arctic Amplification (AA) is driving long-term changes in the Arctic climate system, including
33 glacier ice melt, rapid sea ice decline, and alterations in atmospheric and geochemical processes, with
34 consequences on the formation, transport, and chemical composition of aerosols and seasonal
35 snowpack. Svalbard, particularly vulnerable to AA, provides a valuable site to assess changes in local
36 environmental processes contributing to the seasonal snow chemical composition. From 2018 to
37 2021, sampling campaigns at the Gruvebadet Snow Research Site in Ny-Ålesund, in the North-West
38 of the Svalbard Archipelago, captured the interannual variability in ionic and elemental impurities
39 within surface snow, reflecting seasonal differences in atmospheric and oceanic conditions. Notably,
40 warmer conditions prevailed in 2018-19 and 2020-21, contrasting with the relatively colder season
41 of 2019-20. Our findings suggest that impurity concentrations in the 2019-20 colder season are
42 impacted by enhanced sea spray aerosol production, likely driven by a larger extent of sea ice, and
43 drier, windy conditions. This phenomenon was particularly evident in March 2020, when extensive
44 sea ice was present in Kongsfjorden and around Spitsbergen due to an exceptionally strong, cold
45 stratospheric polar vortex and unusual Arctic Oscillation (AO) index positive phase. By comparing
46 the snow chemical composition of the 2019-20 season with 2018-19 and 2020-21, we provide insights
47 into the interplay between short-term meteorological variability and the long-term climatic impacts
48 of AA in Svalbard, as well as associated shifts in aerosol production process.

49 **1. Introduction**

50 Chemical analysis of surface Arctic snow and ice can provide valuable comprehension of the
51 composition of Arctic aerosols, its deposition, and exchange processes (Lai et al., 2017). These
52 dynamics are influenced by a range of factors, including Arctic Amplification (AA) – a pronounced,
53 long-term increase in near-surface air temperature observed since 1975 (Chylek et al., 2022). AA is
54 recognized as an inherent characteristic of the global climate system, with multiple intertwined causes
55 operating on a spectrum of spatial and temporal scales. These include, but are not limited to, changes
56 in sea ice extent that impact heat fluxes between the ocean and the atmosphere, and water vapour that
57 alters longwave radiation (Serreze and Barry, 2011). The Svalbard Archipelago is particularly
58 sensitive to these effects due to the relatively low altitude of its main ice fields and its geographical
59 location in the higher North Atlantic, where the impact of AA is especially pronounced (Spolaor et
60 al., 2024). Therefore, in the 21st century, predicting and characterising climate change in Svalbard is
61 particularly crucial, as changes in near-surface air temperature, precipitation, and sea ice extent occur
62 at an extremely high pace (Gjermundsen et al., 2020; Rantanen et al., 2022). The Svalbard region,

63 located at the southern edge of the seasonal Arctic sea-ice zone, is characterised by a maritime climate
64 with strong temperature variations during winter (Hansen et al., 2014; Barbaro et al., 2021). In the
65 Arctic winter, the stratospheric polar jet fosters a high-atmospheric vorticity zone. This winter vortex
66 typically acts as a strong barrier for the long-range transport of pollutants from mid-latitudes
67 (Lawrence et al., 2020). However, it occasionally allows warm southern air to penetrate the region
68 (Schoeberl and Newman, 2015). Additionally, Svalbard frequently experiences intense cyclonic
69 storms in autumn and winter, which bring both heat and moisture from lower latitudes (Rinke et al.,
70 2017). These intense meteorological variations, generally linked with a weaker polar vortex (Sobota
71 et al., 2020; Salzano et al., 2023), favour long-range transport of aerosols to the archipelago, including
72 pollutants from continental sources (Stohl et al., 2006b; Yttri et al., 2014a; Vecchiato et al., 2024;
73 D’Amico et al., 2024).

74 Arctic snow captures dry and wet deposition and forms an archive that includes a range of seasonal
75 chemical species such as major ions and trace elements, as well as human-made pollutants emitted
76 into the Arctic atmosphere (Koziol et al., 2021). Ny-Ålesund is a well-monitored area and a natural
77 laboratory for complex system observations, ideal for exploring both long-range contaminants from
78 mid- to high-latitude regions of Eurasia and Canada (Nawrot et al., 2016; Song et al., 2020; Vecchiato
79 et al., 2024; D’Amico et al., 2024), and local inputs from both natural processes and human settlement
80 (Vecchiato et al., 2018). Previous research has extensively investigated the chemistry of Arctic snow
81 and the exchange of inorganic species between the cryosphere and the atmosphere across multiple
82 sites, including Barrow, Summit Greenland, Alert, Sodankylä, and over the Arctic Ocean during the
83 MOSAiC expedition (e.g., Beine et al., 2003; Björkman et al., 2013; Jacobi et al., 2019). Specific
84 studies in Ny-Ålesund and surrounding areas have explored the temporal and compositional aspects
85 of the lower atmosphere (Stohl et al., 2006a; Eleftheriadis et al., 2009; Geng et al., 2010; Zhan et al.,
86 2014; Feltracco et al., 2020, 2021; Turetta et al., 2021), though relatively few have addressed the
87 detailed seasonal dynamics of snow-atmosphere interactions in this region. Building on this existing
88 research, our study aims to enhance the understanding of these interactions, particularly in the context
89 of recent climatic changes.

90 In this study, we evaluate the surface snow concentration of ionic (Cl^- , Br^- , NO_3^- , SO_4^{2-} , MSA, Na^+ ,
91 NH_4^+ , K^+ , Ca^{2+}) and elemental impurities (Li, Be, Mg, Al, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se,
92 Rb, Sr, Ag, Cd, Sb, Cs, Ba, Tl, Pb, Bi, U) for the snow seasons between 2018-2021, at the Gruvebadet
93 Snow Research Site (GSRS) location, 1 km south of Ny-Ålesund, where clean and undisturbed snow
94 conditions are guaranteed throughout the whole sampling season. The differences in average
95 meteorological and climatological conditions across the studied seasons are analysed to assess how

96 sea ice extent, polar vortex, and Arctic Oscillation (AO) conditions influence the composition of
97 surface snow in connection with the aerosol-producing and deposition processes in Kongsfjorden.
98 Additionally, a Kruskal-Wallis's test, followed by Dunn's post-hoc analysis, has been employed to
99 identify species most affected by inter-annual and seasonal variations, with the goal of uncovering
100 potential correlations between meteorological-climatic conditions and chemical concentrations.

101 **2. Methodology**

102 *2.1 Sampling and processing*

103 Three sampling campaigns were conducted in Svalbard between 2018 and 2021, covering the period
104 from October to May according to the onset of the snowpack formation and melting. Sample
105 collection followed the protocol presented in Spolaor et al. (2019), further adopted by Bertò et al.
106 (2021), where consecutive and adjacent sampling was carried in a 3x3 meters snow sampling area,
107 within a clean sampling snowfield of 100 m wide. Each sample was collected 10 cm apart from the
108 previous one, along a precise path. This method was designed to minimise the temporal variability
109 between consecutive samples and reduce the impact of potential spatial variability (within the 5-15%
110 range, according to Spolaor et al., 2019).

111 During the first sampling campaign, carried out from October 4th, 2018 to May 10th, 2019, 133 snow
112 samples were collected at the Gruebadet Snow Research Site (GSRS), a clean-area located about 1
113 km south of Ny-Ålesund, nearby the Gruebadet Atmospheric Laboratory (GAL), dedicated to the
114 chemical and physical monitoring of the seasonal snowpack (Scoto et al., 2023; Fig. 1).

115 The surface snow was sampled within the upper 3 cm, as this is the snow layer most impacted by
116 aerosol deposition and exchange processes at the snow-atmosphere interface (Spolaor et al., 2018,
117 2021b). This choice also minimised the effect of different physical snow conditions (density, crystal
118 shape, and size).

119 Throughout the season, the sampling resolution varied based on light conditions. During the polar
120 night (from October to early March), snow sampling was carried out every 3-5 days. With the
121 beginning of the polar day, daily sampling was conducted until the end of the snow season.

122 The second campaign was carried out from October 26th, 2019 to May 25th, 2020, with a total of 107
123 samples collected. Consecutive samples represent the same snow layer in the absence of snowfall or
124 wind drift/erosion. However, factors such as snow aging, potential element re-emission,
125 transformation, and dry deposition can introduce variability, making continuous monitoring essential.
126 Finally, during the third snow sampling campaign, lasting from October 27th, 2020 to June 15th, 2021,
127 a weekly sampling was conducted at GSRS, with a total of 32 samples collected.

128 During snow sampling, the temperature and density of surface snow were measured, and the density
129 of snow was calculated based on weighting a 100 cc cylinder. After collection, snow samples were
130 melted, and two different aliquots were obtained and stored in separate vials. In a 1.5 mL
131 polypropylene (PP) vial, 1 mL of sample was stored for ionic species, while another aliquot was
132 stored in a 5 mL LDPE vials for trace elements analysis. PP vials designated to ionic species analysis
133 were previously sonicated for 30 min in UltraPure Water (UPW) ($18 \text{ M}\Omega \text{ cm}^{-1}$ at 25°C) for
134 decontamination. LDPE vial used for trace elements analysis were instead conditioned with HNO_3
135 2% and sonicated for 30 min. All sample aliquots were stored at -20°C in dark conditions and
136 transported to the Venice ISP-CNR laboratories.

137 To assess potential contamination during sampling, handling, and transport, field blanks were
138 collected during each campaign. Metal-free vials (Avantor, VWR Centrifuge Tubes, CHN) were
139 opened to ambient air at the sampling sites for a few minutes without collecting snow, then sealed
140 and transported to the Ny-Ålesund laboratory. There, they were filled with 2% HNO_3 and stored under
141 the same conditions as the snow samples. In parallel, analytical blanks were prepared by opening
142 vials to air, sealing them, and transporting them directly to Venice, where they were filled with 2%
143 HNO_3 and ultrapure water from the laboratory. Both field and analytical blanks were analysed
144 alongside the snow samples, confirming that background contamination levels were consistently
145 below LODs or one order of magnitude lower than the lowest concentration detected for all analytes.
146

147 Furthermore, seawater temperatures and salinity at 10 m depth were also monitored in Kongsfjorden
148 (Kb3; $78^\circ57.228'\text{N}$, $11^\circ57.192'\text{E}$) during 2019-2021 spring seasons, with data collected every 3-6
149 days (Assmy et al. 2023). Data was derived from Conductivity Temperature Depth (CTD) casts with
150 either a MiniSTD model SD-204 (SAIV A/S, Bergen, Norway) or a XR-620 CTD (RBR Ltd, Ottawa,
151 Canada). Combined casts of both instruments conducted in May 2020 and 2021 did not reveal
152 differences in temperature or salinity in the reported accuracy (two post comma digits).

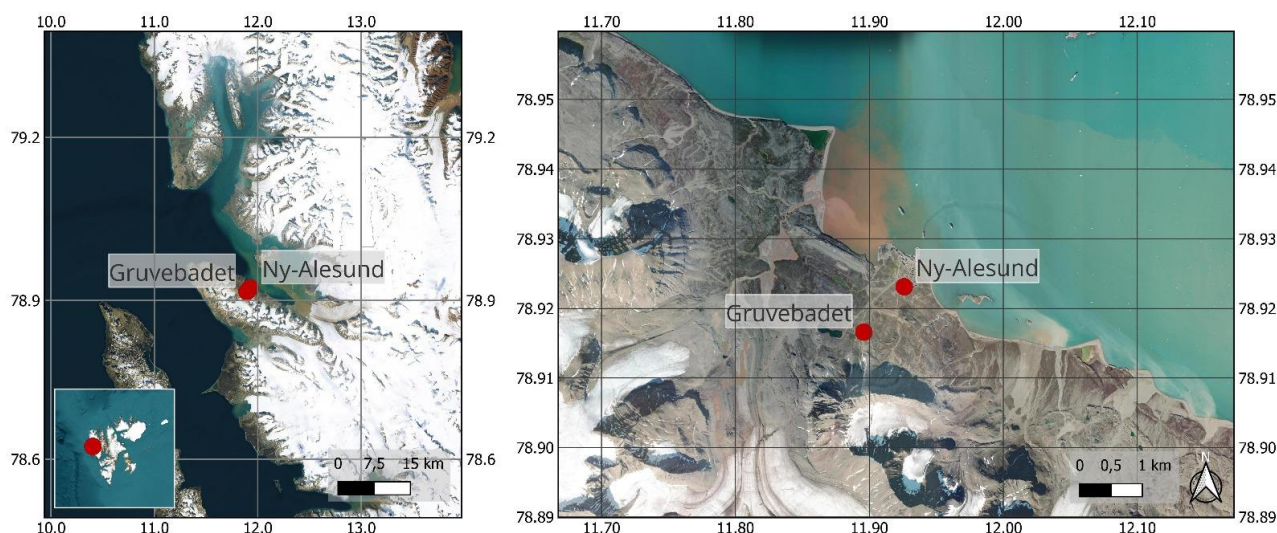


Fig. 1. Gruvebadet Snow Research Site (GSRS) location with respect to Ny-Ålesund Research Station, in the Brøgger peninsula (Svalbard Islands).

2.2 Analysis of ionic species

The analysis of anionic species (Cl^- , Br^- , NO_3^- , SO_4^{2-} , MSA) was carried out using an ion chromatograph (IC, Thermo Scientific Dionex™ ICS-5000, Waltham, MA, USA) coupled with a single quadrupole mass spectrometer (MS, MSQ Plus™, Thermo Scientific, Bremen, Germany). The separation was performed using an anionic exchange column (Dionex Ion Pac AS 19 2 mm ID $\times$ 250 mm length) equipped with a guard column (Dionex Ion Pac AG19 2 mm ID $\times$ 50 mm length). Sodium hydroxide (NaOH), used as mobile phase, was produced by an eluent generator (Dionex ICS 5000EG, Thermo Scientific). The NaOH gradient with a 0.25 mL min^{-1} flow rate was: 0–6 min at 15 mM; 6–15 min gradient from 15 to 45 mM; 15–23 min column cleaning with 45 mM; 23–33 min equilibration at 15 mM. The injection volume was 100 μL . A suppressor (ASRS 500, 2 mm, Thermo Scientific) removed NaOH before entering the MS source. The IC-MS operated with a negative electrospray source (ESI) with a temperature of 500°C and a needle voltage of 3 kV. The other MS parameters are reported by Barbaro et al. (2017). The same IC system was simultaneously used to determine cationic species (Na^+ , K^+ , Ca^{2+} and NH_4^+). However, Ca^{2+} was not measured within the samples collected during the second campaign due to instrumental limitations.

The separation occurred with a capillary cation-exchange column (Dionex Ion Pac CS19–4 mm 0.4 mm ID $\times$ 250 mm length), equipped with a guard column (Dionex Ion Pac CG19–4, 0.4 mm ID $\times$ 50 mm length), and the species were determined using a conductivity detector. Analytical blanks of ultrapure water ($> 18 \text{ M}\Omega \text{ cm}$) were included in the analysis, and the Method Detection Limit (MDL) was set to 3 times the standard deviation of the blank values. Checks for accuracy were made using

176 certified multi-element standard solutions for anions (Cl^- , Br^- , NO_3^- , SO_4^{2-} , no. 89886-50ML-F, Sigma
177 Aldrich) and cations (Na^+ , K^+ , Ca^{2+} , no. 89316-50ML-F, Sigma Aldrich) at a concentration of 10 mg
178 $\text{L}^{-1} \pm 0.2\%$. The analytical precision was quantified as the relative standard deviation (RSD) for
179 replicates ($n > 3$) of standard solutions and was always $< 10\%$ for each ion.

180 *2.3 Trace Elements analysis*

181 Twenty-six elements (Li, Be, Mg, Al, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Ag, Cd, Sb,
182 Cs, Ba, Tl, Pb, Bi and U) were analysed on samples previously melted and acidified to 2% v/v with
183 HNO_3 (UpA grade, Romil, UK) for 24 hours before analysis (Spolaor et al., 2018; Spolaor et al.,
184 2021a).

185 The analysis was performed using Inductively Coupled Plasma Mass Spectrometry (ICP-MS, iCAP
186 RQ, Thermo Scientific, US). The ICP-MS was equipped with an ASX-560 autosampler (Teledyne
187 Cetac Technologies), PolyPro PFE nebulizer, PFE cyclonic spray chamber thermostated at 2.7°C ,
188 sapphire injector, quartz torch and Ni cones. The acquisition was performed at 1550 W of plasma RF
189 power in Kinetic Energy Discrimination (KED) – high matrix mode, using He as collision gas (4.3
190 mL min^{-1}). Instrument parameters were optimised for best sensitivity in the whole mass range,
191 minimum oxides ($< 1\%$) and double charges ($< 3\%$). Quantification was obtained by external
192 calibration with multi-elemental standards prepared in ultrapure water ($18 \text{ M}\Omega \text{ cm}^{-1}$ at 25°C) with
193 2% v/v ultrapure grade HNO_3 (UpA grade, Romil, UK), with a combination of certified level multi-
194 elemental solutions IMS-102 and IMS-104 from UltraScientific. Analytical quality control was
195 performed by memory test blank (repeated analysis of ultrapure grade HNO_3 2% v/v blank solution)
196 after each sample and calibration verification (repeated analysis of reference standards) every 11
197 samples. More details are found in Spolaor et al., 2021a.

198

199 *2.4 Transport modelling, sea ice, Kongsfjorden condition, and polar vortex*

200

201 The Lagrangian particle dispersion model HYSPLIT (Draxler, 1998; Stein et al., 2015) was used to
202 determine the source region of air masses over Ny-Ålesund. This model has previously been shown
203 to be an effective tool for the prediction of transport pathways into and within the Arctic and Antarctic
204 regions (Barbaro et al., 2015; Feltracco et al., 2021). The simulations were driven using
205 meteorological data from the Global Data Assimilation System (GDAS) one-degree archive, set the
206 top of the model at 10000 m and the height source equal to the GSRS altitude. Back-trajectories were
207 calculated every 6 h, with a propagation time of 120 h for each sampling period. The choice of a 6-
208 hour interval for the calculation of back-trajectories allows for the capture of temporal variability in

209 air mass origins over the day, which is particularly important in polar regions where atmospheric
210 circulation patterns can change rapidly. This temporal resolution strikes a balance between
211 computational efficiency and the need for sufficient detail to characterise the variability in source
212 regions during each sampling period. The propagation time of 120 hours was selected to provide an
213 adequate temporal window to trace long-range transport pathways that influence air mass
214 composition at Ny-Ålesund. This configuration is consistent with previous studies on atmospheric
215 circulation in the same site (Feltracco et al., 2021).

216 This approach was used to ensure an envelope working for all investigated tracers. The resulting
217 multiple trajectories were based on the screen-plot analyses of total spatial variance.

218 The Ice Service provided by the Norwegian Meteorological Institute (NIS) was employed to analyse
219 the weather conditions via remotely sensed data and to generate ice charts of Svalbard, ice-edge
220 information, and sea surface temperatures trends. Sea ice extent variability in Kongsfjorden was
221 evaluated based on dataset made available by Gerland et al. (2022).

222 Differences between the sampling campaigns were evaluated through the National Centers for
223 Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) Reanalysis
224 data from NOAA Physical Sciences Lab's daily composites tool, used to calculate the near-surface
225 air temperatures across the Northern Hemisphere from October to May.

226

227 *2.5 Statistical procedures and Enrichment Factors (EFs) Analysis*

228

229 Results below the limit of detection were assumed to be equal to $\frac{1}{2}$ of Method Determination Limit
230 (MDL) prior to perform statistical analysis, to approximate their likely level based on the data
231 distribution curve (best approximated as log-normal for most of the studied variables) (George et al.,
232 2021).

233 Enrichment Factors (EFs) using Ba as a crustal element of reference (Widepohl, 1995; Spolaor et al.,
234 2021a; Ruppel et al., 2023), were calculated to explore the mixed sources of the investigated elements.
235 To complement the EFs analysis, a Hierarchical Cluster Analysis (HCA) was performed using Ward's
236 algorithm and Euclidean distances as clustering criteria, to determine the presence of some clusters
237 and simplify the interpretation of the dataset.

238 Additionally, to examine the inter-annual and seasonal variations in chemical's concentrations and to
239 identify the species most affected by these changes, a Kruskal-Wallis' test was employed, followed
240 by Dunn's post-hoc test for pairwise comparisons. The Kruskal-Wallis' test, a nonparametric
241 alternative to Anova, which is typically applied when samples do not follow a normal distribution
242 (Van Hecke, 2013), was chosen to assess differences in chemical concentrations across the years of

sampling campaigns and multiple seasons. This allowed for an objective evaluation of temporal differences in concentration levels, while Dunn's post-hoc analysis, which uses the same shared rankings and pooled variance assumed under the Kruskal-Wallis null hypothesis, helped identify which specific seasons showed statistically significant differences from each other. The Bonferroni correction was used for the Dunn's test to control for false positives when performing multiple pairwise comparisons (Dunn, 1961).

The ultimate goal of this analysis was to uncover potential correlations between meteorological-climatic conditions and chemical concentrations in surface snow, helping to better understand how environmental factors contribute to contamination trends.

These combined statistical approaches provided a comprehensive framework for analysing the complex dataset.

3. Results

3.1 Interannual trends of chemical species on the surface snow

Three consecutive snow seasons were evaluated to define the chemical composition of the surface snow in the Arctic site of GSRS. The sea salt ions Cl^- (50 %), and Na^+ (23%) represent the most abundant species, followed by SO_4^{2-} (11 %), Mg (7 %), Ca (2%), Fe (1%) and Al (1%). Similar relative concentration abundances were also found in previous studies on the snow of the Svalbard Archipelago (Beaudon and Moore, 2009; Vega et al., 2015; Barbaro et al., 2021; Spolaor et al., 2021b).

Table 1 reports the average ionic loads of the most abundant ($> 1\%$) species in the surface snow, considering three different seasons: autumn is defined until December 21st, winter until March 21st, and spring from then to the melt onset, identified by 5-6 consecutive days of negative snow accumulation. The average ionic loads of the less abundant ($< 1\%$) species are reported instead in Table S1.

Table 1. Average ionic loads of the most abundant ($>1\%$) ionic and elemental species in the surface snow during each season of the three consecutive sampling campaigns. The standard deviation is shown in brackets, while in the case of nss-SO_4^{2-} the brackets represent the percentage of nss-SO_4^{2-} compared to the total SO_4^{2-} . “n” indicates the number of samples considered for the calculation of the average. Annual averages are reported at the end of each sampling year.

mg m ⁻²	Total	Cl ⁻	Na ⁺	SO ₄ ²⁻	nss-SO ₄ ²⁻	Mg	Fe	Ca	NO ₃ ⁻	K ⁺	NH ₄ ⁺
autumn 2018 (n=22)	32 (25)	15 (21)	7 (11)	3 (4)	1 (36%)	3 (3)	2 (5)	0.3 (0.3)	0.5 (0.5)	0.3 (0.4)	0.04 (0.03)
winter 2018-19 (n=41)	115 (80)	55 (68)	31 (39)	16 (16)	8 (51%)	8 (8)	0.4 (0.3)	0.4 (0.3)	2 (2)	2 (2)	0.3 (0.3)
spring 2019 (n=51)	75 (50)	36 (43)	19 (24)	9 (9)	4 (48%)	6 (5)	2 (3)	0.6 (0.4)	1 (1)	1 (1)	0.5 (0.4)
Year 2018-19	222 (25)	106 (20)	58 (12)	28 (7)	13 (48%)	17 (3)	4 (1)	1 (0.1)	4 (0.8)	3 (0.6)	0.8 (0.2)
autumn 2019 (n=15)	212 (98)	101 (71)	40 (53)	26 (20)	16 (61%)	10 (6)	1 (1)	9 (12)	5 (4)	2 (3)	4 (5)
winter 2019-20 (n=43)	335 (120)	159 (88)	79 (73)	41 (20)	21 (52%)	16 (8)	2 (5)	9 (10)	3 (2)	3 (4)	7 (8)
spring 2020 (n=49)	264 (131)	110 (91)	52 (77)	28 (25)	15 (53%)	21 (21)	6 (9)	13 (16)	4 (2)	2 (4)	5 (8)
Year 2019-20	811 (39)	370 (31)	171 (20)	95 (8)	52 (55%)	47 (6)	9 (3)	31 (6)	12 (1)	7 (1)	16 (2)
autumn 2020 (n=6)	796 (542)	435 (466)	191 (205)	66 (83)	18 (27%)	84 (165)	1 (2)	2 (5)	6 (11)	9 (10)	2 (2)
winter 2020-21 (n=13)	327 (203)	207 (194)	64 (48)	41 (36)	24 (60%)	6 (5)	0.2 (0.4)	0.2 (0.1)	5 (3)	3 (2)	1 (1)
spring 2021 (n=13)	178 (92)	107 (86)	36 (26)	16 (12)	7 (43%)	9 (10)	4 (7)	1 (2)	3 (2)	2 (2)	1 (1)
Year 2020-21	1300 (194)	749 (168)	291 (83)	123 (25)	49 (40%)	99 (44)	5 (2)	3 (1)	14 (2)	14 (4)	4 (0.5)

276

277 The average loads of the first sampling year were lower compared to the other campaigns (Fig. S1;
278 Table 1).

279 High average loads were observed instead for Cl⁻ and Na⁺ in the top snowpack layer during the 2019-
280 20 and 2020-21 winter seasons. The highest concentrations of sea salt species were found in autumn
281 2020, despite low snow accumulation (17.25 cm) in a period of relatively high precipitation (4.91
282 mm) (Fig. 2, Table S3). This apparent contradiction can likely be attributed to the increased wind
283 speeds during that time, which may have caused snow redistribution, thereby limiting accumulation
284 despite the considerable precipitation.

285 The non-sea-salt sulphate (nss-SO₄²⁻), calculated using a seawater SO₄²⁻/Na⁺ mass ratio of 0.252
286 (Millero et al., 2008), was the most abundant fraction of the total sulphate in autumn 2019 and winter
287 2020-21, while in autumn 2018 and 2020 sea salt sulphate (ss-SO₄²⁻) was the dominant fraction. No
288 clear predominance between the two fractions was achieved during the other investigated seasons,
289 which may reflect a more mixed influence of marine and continental sources (Table 1).

290 The abundance of all chemical species investigated is quite similar for all years (Fig. S1), although
291 the sampling campaign 2019-20 showed higher percentage of calcium ranging between 3% and 5%,
292 in contrast to the typical concentrations < 1% found in the other two campaigns.

293 To further examine differences in species concentrations across seasons and years, a Kruskal-Wallis
294 test was performed. The test indicates significant differences ($\chi^2 > 15.51$, df = 8, p-value = 0.05) for
295 several species across the nine seasons of the three sampling campaigns. These species include Br⁻

296 ($\chi^2 = 16.9$), MSA ($\chi^2 = 81.3$), SO_4^{2-} ($\chi^2 = 21.3$), nss- SO_4^{2-} ($\chi^2 = 23.4$), Bi ($\chi^2 = 19.2$), Cd ($\chi^2 = 31.9$),
 297 Cu ($\chi^2 = 44.5$), Fe ($\chi^2 = 21.9$), Pb ($\chi^2 = 45.1$), Ni ($\chi^2 = 20.6$), Ag ($\chi^2 = 24.5$), U ($\chi^2 = 22.1$), V ($\chi^2 =$
 298 40.9), Zn ($\chi^2 = 29.9$). To identify which seasons accounted for these differences, a Dunn's test with
 299 Bonferroni correction was applied as a post-hoc test, revealing significant seasonal differences for
 300 multiple elements (Fig. S2), with pronounced differences particularly between autumn and winter,
 301 and between spring and winter ($P_{\text{adj}} < 0.05$; Table S5).
 302 Finally, correlations between meteo-climatic variables (air temperature, wind speed, wind direction,
 303 precipitation, AO index) and the concentrations of the selected species were computed to assess
 304 potential meteorological influences. The results indicated generally weak positive correlations ($\rho <$
 305 0.5, p-values < 0.05) for the variables considered. Notably, MSA showed a correlation with air
 306 temperature ($\rho = 0.2$, p-values < 0.05), as did Bi ($\rho = 0.2$, p-values < 0.05), Cu ($\rho = 0.2$, p-values $<$
 307 0.05), Fe ($\rho = 0.2$, p-values < 0.05), Pb (0.1, p-values < 0.05), Ni, ($\rho = 0.2$, p-values < 0.05), Ag ($\rho =$
 308 0.2, p-values < 0.05), U ($\rho = 0.3$, p-values < 0.05), V ($\rho = 0.3$, p-values < 0.05), and Zn ($\rho = 0.3$, p-
 309 values < 0.05).

310 *3.2 Polar vortex and Arctic Sea ice extent in 2019-20*

311 According to the 2023 survey conducted by the National Snow and Ice Data Center (NSIDC), the
 312 maximum extent of Arctic Sea ice since 2014 has been recorded in March 2020, with 14.73 million
 313 square kilometres of the Arctic Ocean surface, in a decadal trend characterised by a -2.53% of decline,
 314 due to the Arctic Amplification. Considering the Kongsfjorden area, the total sea ice extent varied
 315 from 63.94 km² in March 2019 to 129.81 km² in March 2020, and was with 46.26 km² lowest in
 316 March 2021 (Gerland et al., 2022). Specifications on Drift Ice (DI), Fast Ice (FI), and Open Water
 317 (OW) extent are reported in Table S2. The 2020 maximum sea ice extent followed the exceptionally
 318 strong and cold stratospheric polar vortex that took place in the Northern Hemisphere (NH) during
 319 the 2019-20 polar winter, together with low wave activity from the troposphere, which allowed the
 320 polar vortex to remain relatively undisturbed (Lawrence et al., 2020). Notably, the 2020 Arctic Sea
 321 ice extent is 16% and 9% higher than previous (2018-19) and following (2020-21) records (dataset
 322 NSIDC, NOAA). Lower surface mean air temperatures (-14.7°C, compared to the -11.4°C recorded
 323 in 2018-19, and the -8.0°C recorded in the 2020-21), average reduced precipitations (2.1 mm
 324 compared to 2.6 mm in 2018-19, and 3.2 mm in 2020-21), higher maximum mean wind speed (17.7
 325 ms⁻¹ for 6 days, compared to 15.4 ms⁻¹ for 3 days in 2018-19, and 13.8 ms⁻¹ for 5 days in 2020-21),
 326 and higher mean snow height (62.63 cm compared to 58.06 cm in 2018-19, and 57.77 cm in 2020-
 327 21) were observed during the 2019-20 winter season, attributed to the influence of a strong polar
 328 vortex triggered by a net positive Arctic Oscillation (AO) phase. The 2020 anomalous AO index is

displayed in Fig. S3. Seasonal values of mean air temperatures ($^{\circ}\text{C}$), mean precipitation (mm), maximum mean wind speed (m sec^{-1}) and mean snow depth (cm) during the three consecutive sampling campaigns are reported in Table S3. Temperature data were provided by the Norwegian Centre for Climate Services (NCCS), while sea ice extent data were supplied by National Snow and Ice Data Center (NSIDC). Seawater temperature data collected at 10 m depth at a mid-fjord station near Ny-Ålesund (Kb3) was found to be colder during 2020 compared to 2019 and 2021 spring seasons (Table S4). Although these temperatures were still above the freezing point for the observed salinity levels, they contributed to colder overall conditions in Kongsfjorden, supporting the formation and prolonged presence of sea ice when combined with cold atmospheric conditions.

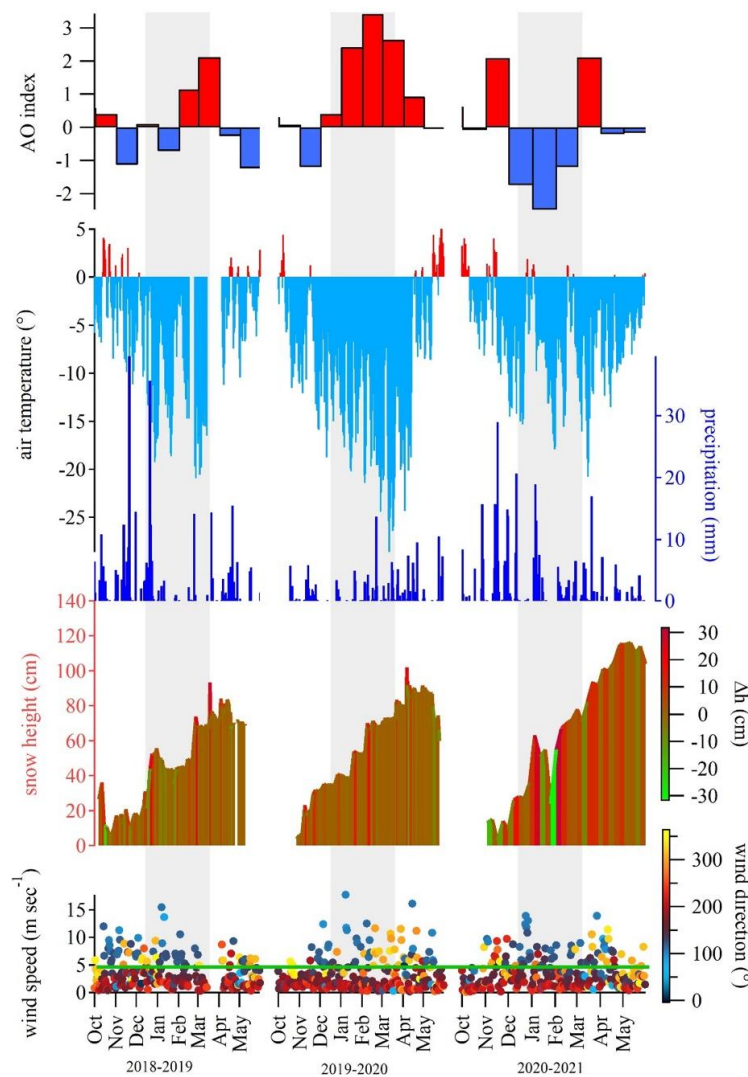
4. Discussion

4.1 Ny-Ålesund seasonal and interannual trends variability in surface snow

The three consecutive sampling campaigns conducted from 2018 to 2021 confirmed the dominance of sea salt input in the surface snow of Svalbard, due to the proximity of the sampling site to the coastline (Barbaro et al., 2021). The dominant ions are Na^+ , Cl^- , and SO_4^{2-} , associated with the scavenging precipitation of marine aerosol (Hodgkins and Tranter, 1998). The observed mean seasonal trends (Fig. S1) display the highest concentrations of marine species in autumn 2020, followed by 2020-21 and 2019-20 winter seasons. However, wintry concentrations are presumably linked to weakened (2019-20) or destroyed (2020-21) polar vortex (Fig. 2) and intense cyclonic storms, associated with anomalous warming events capable of transporting both heat and moisture from lower latitudes to Svalbard (Rinke et al., 2017). In addition to these factors, it is important to account for snow ablation or erosion by wind. The change in snow height (Δh) between consecutive sampling events provides valuable insights into such processes. A decrease in snow height signals ablation can indeed result from snow melting (under positive temperatures), snow aging, sublimation, or snow drift. In particular, when wind speeds exceed 5 m s^{-1} , the threshold commonly accepted for initiating snow drift and ablation episodes (Pomeroy, 1989), we can infer the snow erosion due to wind drift likely occurred (Fig. 2). This effect may explain some of the variability in snow ion concentrations, especially during intense storms or strong winds.

Autumn 2020 represents most likely an outlier, due to scarce precipitations (Fig. 3) that led to more concentrated impurities in the surface snow. In contrast, late spring 2020 saw a notable increase in typical marine ions (Na^+ , Cl^- , Br^- , MSA , SO_4^{2-}) and geogenic elements (Al, Ca, Mn, Fe, Sr), compared to spring 2019 (Fig. 3). This increase may be attributed to the very close drift Arctic Sea ice presence in Kongsfjorden (Table S2), which reached its maximum extent in March 2020 due to low-

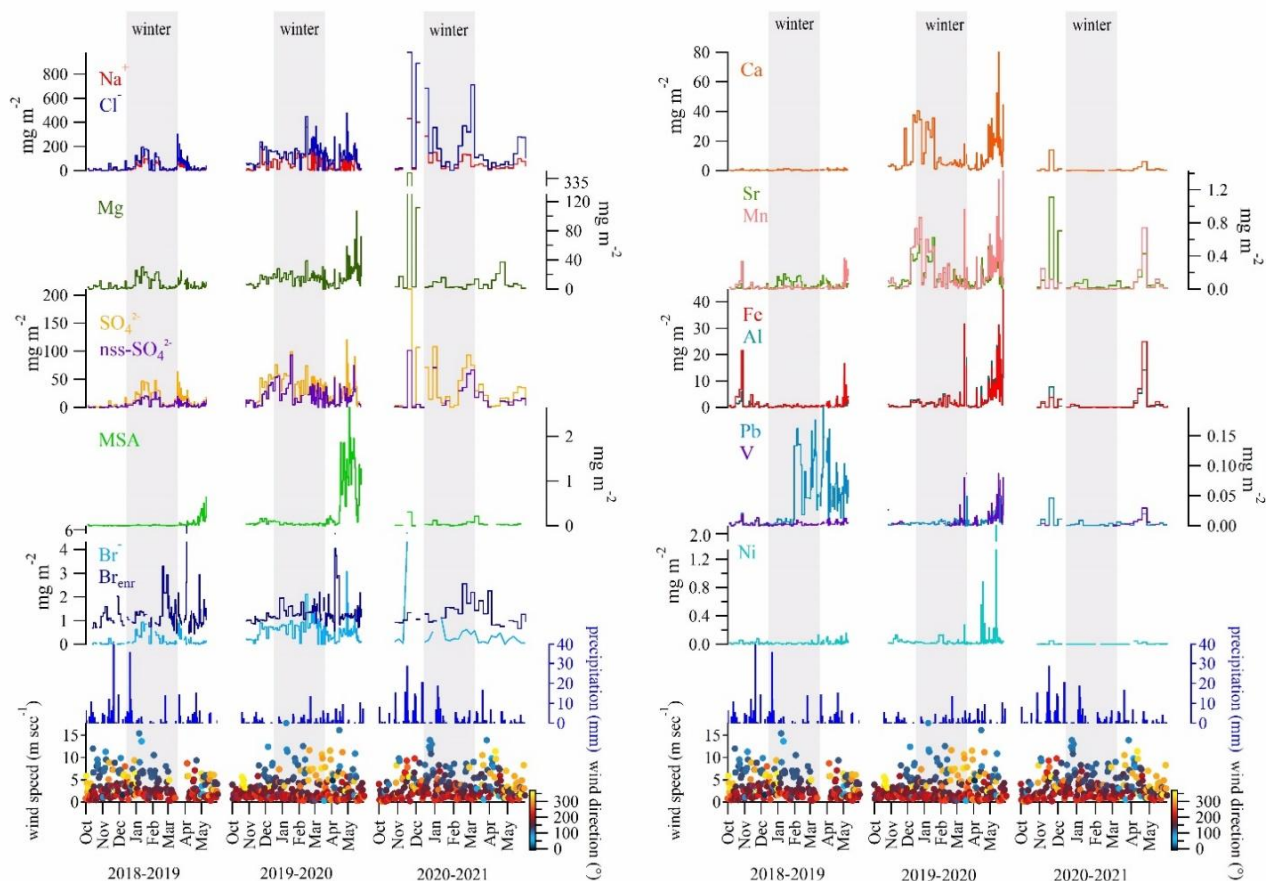
361 temperature anomalies and intensified atmospherically driven sea ice transport and deformation,
 362 caused by higher than normal winter wind speeds (Fig. S4). These conditions likely enhanced the
 363 production of sea spray aerosols, which, when carried by winds, may have increased the deposition
 364 of marine species onto the snowpack. The increased deposition of geogenic elements might also have
 365 been influenced by low temperature anomalies (as seen with Fe), and/or by stronger wind speeds,
 366 although significant correlations have not emerged in this preliminary statistical analysis.
 367 Additionally, an outstanding positive phase of the Arctic Oscillation (AO) in the troposphere (Fig. 2)
 368 was recorded in January-March 2020 (Lawrence et al., 2020; Dethloff et al., 2022). Although the
 369 entire period was remarkable, January and February featured as clear outliers in the historical
 370 timeseries (1950–2023) reported by the NOAA service, while March 2020 exhibited significantly
 371 elevated values but did not exceed the statistical threshold for outliers (Fig. S3).



372
 373 **Fig. 2.** AO Index, air temperature (°C), precipitation (mm), snow height (cm), Δh snow height (cm), wind speed (m sec⁻¹), and wind direction (°) from the NCEP/NCAR Reanalysis data. The green horizontal line above the wind speed graph
 374

375 indicates the 5 m sec⁻¹ threshold, above which wind drift may occur on surface snow layers. The colour of the line refers
376 to the Δh color scale, which indicates negative values of Δh , NOAA Physical Sciences Lab's daily composites tool was
377 used to calculate the near-surface air temperatures across the Northern Hemisphere from October to May. Grey bands
378 indicate the winter periods.

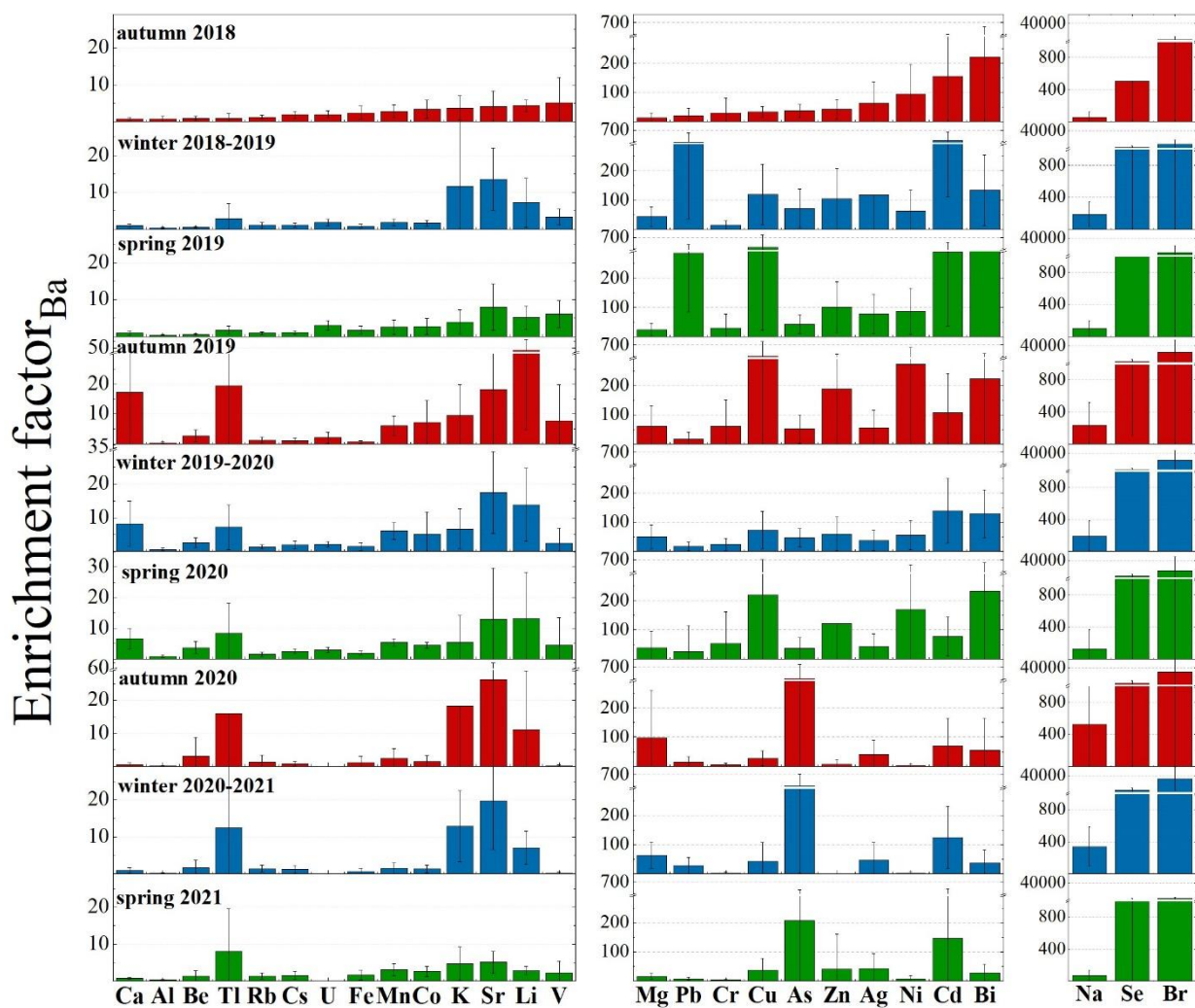
379 A 2021 spring peak of marine species was also recorded, although more attenuated than in spring
380 2020 (Fig. 3, Fig. S1). This variation is likely attributable to different extents of sea ice in the fjord.
381 Nonetheless, seawater temperatures in 2021, similar to those in 2020 and 2.3°C colder than in 2019
382 (Table S4), along with comparable wind speed conditions (Fig. S4), may also have contributed to the
383 observed trends in marine species concentrations. Similarly, the spring peak of Mg, Sr, Mn, Fe, Al,
384 and V in 2021 seems to reflect the high wind speed and positive AO index recorded from March to
385 April 2021. Positive anomalies for air temperatures (A) and wind speed (W) conditions, together with
386 negative anomalies in seawater (O) conditions were observed during the 2020-21 campaign, while
387 negative A and O conditions were accompanied to positive W during 2019-20. On the contrary, 2018-
388 19 diverges from the other campaigns for positive O condition associated to negative W condition
389 anomalies. These observations provide valuable insights into how shifts in atmospheric and oceanic
390 conditions impact the concentrations of ionic and elemental species in surface snow, enhancing our
391 understanding of the underlying mechanisms that govern these changes in the context of AA
392 conditions.



393

394 **Fig. 3.** Wind direction ($^{\circ}$), wind speed (m sec^{-1}), and ionic loads (mg m^{-2}) of Na^{+} , Cl^{-} , Mg , SO_4^{2-} , nss-SO_4^{2-} , MSA , Br^{-} ,
 395 Ca , Sr , Mn , Fe , Al , Pb , V , Ni in the surface snow for the three sampling campaigns: 2018-19, 2019-20, 2020-21. Seasonal
 396 trends are here presented for specific elements to provide a detailed view of how concentrations vary across distinct
 397 sampling periods.

398 A singular case is represented by Pb , which showed a 12.5-fold increase during winter-spring 2019
 399 period compared to autumn 2018. The EF for Pb during these seasons exceeded 100, indicating a
 400 strong anthropogenic contribution. In contrast, the peak observed in 2020 no longer shows such
 401 elevated EF values (i.e., $\text{EF} = 26$), suggesting a mixed source (Fig. 4). This implies that the April-
 402 May 2020 peak is largely of crustal origin, as the overlap with V ($\text{EF} < 10$) also suggests, possibly
 403 due to local dust events driven by strong winds exceeding the 5 m sec^{-1} threshold (Fig. 3; Table S3).



404

405 **Fig. 4.** Enrichment Factors (EFs) calculated on all the presented trace elements for the three sampling campaigns: 2018-
 406 19, 2019-20, 2020-21. Calculating EFs for the full dataset offers a more robust assessment of potential sources and
 407 enrichment patterns, minimizing the variability inherent in individual seasons and allowing for a clearer distinction
 408 between crustal and non-crustal contributions.

409 The calculated EFs (Fig. 4) for Be, Al, V, Mn, Fe, Co, Rb, Cs, and U were consistently below 10,
 410 indicating a predominantly crustal (geogenic) origin for these elements (Wedepohl, 1995; Gabrieli et
 411 al., 2011). In contrast, Ni, and Sr displayed EFs greater than 10, suggesting contributions from mixed
 412 sources. Notably, Ni exhibited exceptionally high EF values – above 100 – during autumn 2019 and
 413 spring 2020 (Ni). Mixed sources were also recognised for Li, K, Cr, Cu, Zn, As, Ag, Cd, and Tl with
 414 occasional EF values over 100. This suggests significant anthropogenic contributions, especially for
 415 Cu (spring and autumn 2019; spring 2020), As (from autumn 2020 to spring 2021), Zn (autumn 2019)
 416 and Cd (from autumn 2018 to spring 2019; winter 2019-20).

417 The 2019 springtime Pb concentration maxima are typically consistent with a mixture of eastern and
418 western European sources (Sherrell et al., 2000; Bazzano et al., 2015, 2021). In this study, cluster
419 mean trajectories obtained for winter 2018-2019 highlighted a 25% of air mass provenance from
420 Russian Arctic and a 13% from eastern Siberia (Fig. S5), possibly explaining the higher
421 concentrations of Pb revealed in spring 2019, following a reduced precipitation regime that occurred
422 in January 2019. A local anthropogenic origin can be excluded though, since no activities (ordinary-
423 extraordinary maintenance or particular events) were recorded in the vicinity of the sampling site in
424 the 2019 sampling season. However, at present, the long-range transport of Pb remains a hypothesis,
425 likely supported by the breakdown of the wintry polar vortex (Fig. 2).

426 Backward trajectories (Fig. S5) for Ny-Ålesund area (78.92° N, 11.89° E) appear mostly in line with
427 literature findings (Platt et al., 2022; Vecchiato et al., 2024), showing three main seasonal characters:
428 a prevalent mass movement from ice-covered Central Arctic Ocean, Kara Sea, and Greenland Sea
429 during autumn, a main provenance from Central Arctic Ocean and Kara Sea during winter, and a
430 predominant trajectory from Northern Canada in addition to air masses arriving from Arctic Ocean
431 and Kara seas during spring.

432 *4.2 The main ion sources in the seasonal snow of Ny-Ålesund*

433 Examining the dominant ions associated with marine aerosol, we found Cl^-/Na^+ median ratios ranging
434 from 1.3 to 1.5 w w^{-1} , slightly lower than the expected value of 1.8 w w^{-1} in the pure seawater (Zhuang
435 et al., 1999), pointing the occurrence of a minimum Cl^- depletion in aerosol, quantified as 14% for
436 the 2018-19 and 2019-20 campaigns, and as just 2% for the 2020-21 campaign. A possible
437 explanation for this phenomenon could be the de-chlorination of sea-spray aerosol during transport.
438 This reaction occurs when sea-salt particles containing NaCl interact with acids such as HNO_3 ,
439 H_2SO_4 , or organic acids, leading to the release of gaseous HCl (Zhuang et al., 1999, and reference
440 therein). Although less likely, this process could also occur at the snow-atmosphere interface. On the
441 contrary, a possible influence of biomass burning on Cl^- depletion process has been excluded by the
442 very low correlation (0.18, $p\text{-value} < 0.05$) found between Cl^- depletion values and $\text{nss-K}^+/\text{K}^+$ ratios,
443 which is a tracer of relative contribution of biomass burning (Song et al., 2018).

444 Positive correlations between Mg, Ca, and K^+ with Na^+ and Cl^- (Fig. 5, $p\text{-value} < 0.05$) suggest a
445 common sea-spray source. However, the concentrations of Mg are also positively correlated with nss-
446 Ca ($\rho_{\text{load}} = 0.55$, $p\text{-value} < 0.05$), calculated according to Morales et al. (1998), indicating a shared
447 non-marine source. This suggests that in addition to their marine origin, these ions (Mg, Ca) also have

448 contributions from a non-marine, crustal source. Further evidence comes from the Ca/Mg ratios in
449 surface snow samples collected during the three campaigns, which were higher than those found in
450 seawater (0.32, Millero et al., 2008). This excess indicates the likely presence of mineral particles
451 (i.e., calcite and dolomite), potentially originating from local rock or soil dust (e.g., limestone,
452 dolostone, and marble, which are abundant in Svalbard), as previously observed by Barbaro et al.
453 (2021). To further explore the mixed sources of these elements, we calculated the Enrichment Factors
454 (EFs) of Mg and Ca. For Mg, the EF values were consistently above 10 in all the seasons analysed,
455 indicating a significant non-crustal source (e.g., marine). In contrast, Ca displayed EF values greater
456 than 10 only during autumn 2019, suggesting that its non-crustal contribution (likely from sea spray)
457 was more pronounced during that specific season. Based on these findings, we conclude that Mg and
458 Ca effectively share a common sea spray origin, but the sea-salt contribution of Ca was mainly
459 significant in autumn 2019, while the excess of Ca in other seasons likely reflects inputs from crustal
460 source, such as local mineral dust.

461 Similarly, sulphate (SO_4^{2-}) is highly and significantly correlated (p -values < 0.05) with both Na^+ (ρ_{load}
462 $= 0.80$) and Cl^- ($\rho_{\text{load}} = 0.91$), indicating that sea-spray is its main source. Nonetheless, $\text{Na}^+/\text{SO}_4^{2-}$ and
463 $\text{Cl}^-/\text{SO}_4^{2-}$ ratios are significantly lower than typical seawater values (3.97 and 7.13, respectively,
464 according to Millero et al., 2008) for the former two campaigns (2018-19, 2019-20). The elevated
465 sulphate concentrations compared to sodium, also in winter snow, suggest the absence of a strong
466 frost flower signature. Additionally, the lack of significant depletion in nss-SO_4^{2-} further supports the
467 minimal role of frost flowers in contributing to the snow composition. Therefore, while frost flowers
468 are known to impact snow chemistry in Svalbard (Rankin et al., 2002; Beaudon and Moore, 2009;
469 Roscoe et al., 2011), our analysis indicates that their contribution to the observed sea salt peaks in
470 Kongsfjorden during the 2018-19, 2019-20 campaigns was likely limited. This analysis highlights
471 that, while sea ice extent supports higher sea-salt concentrations in snow, the specific sulphate and
472 ion ratios observed point to sea spray as the main source, with only a minor role for frost flower-
473 related inputs.

474 Instead, the presence of nss-SO_4^{2-} suggests potential inputs from other sources, such as crustal
475 material, anthropogenic emissions (e.g., fossil fuel combustion), or the oxidation of dimethylsulfide
476 (DMS) released from marine biological activities.

477 To estimate the crustal fraction of sulphate (cr-SO_4^{2-}), the nss-Ca (as crustal marker) content was
478 multiplied by 0.59 ($\text{SO}_4^{2-}/\text{Ca}$ w/w ratio in the uppermost Earth crust - Wagenbach et al. 1996),
479 obtaining variable contributions for the three sampling campaigns, ranging from 2.45% up to 12.94%.

Conversely, the anthropogenic contribution to nss-SO_4^{2-} concentrations was investigated by the application of the $[\text{ex-SO}_4^{2-}]$ concentration formula (Schwikowski et al., 1999), considering the average concentration of $[\text{Ca}]$ instead of the average ionic concentration $[\text{Ca}^{2+}]$ because Ca^{2+} concentrations were not measured in the samples collected during the second campaign due to instrumental limitations:

$$[\text{ex-SO}_4^{2-}] = [\text{SO}_4^{2-}] - (0.12 [\text{Na}^+]) - (0.175 [\text{Ca}^{2+}])$$

The obtained results showed a 50 up to 60% of anthropogenic contribution for the nss-SO_4^{2-} input. This finding corroborates previous results from Amore et al. (2022), who noted that anthropogenic sulphate was the most abundant apportioned component at Grubebadet, accounting for at least 50% all over the year during the 2010-2019 investigated period. The plausible source of the anthropogenic fraction is the atmospheric transport of secondary aerosols containing SO_4^{2-} , and ammonium sulphate. This sulphate can be formed by SO_x emitted from coal combustion throughout the winter and biomass burning in the spring (Barbaro et al., 2021 and reference therein). The nss-SO_4^{2-} does not correlate significantly with other ionic species (except for Mg), thus suggesting a separate origin (Fig. 5).

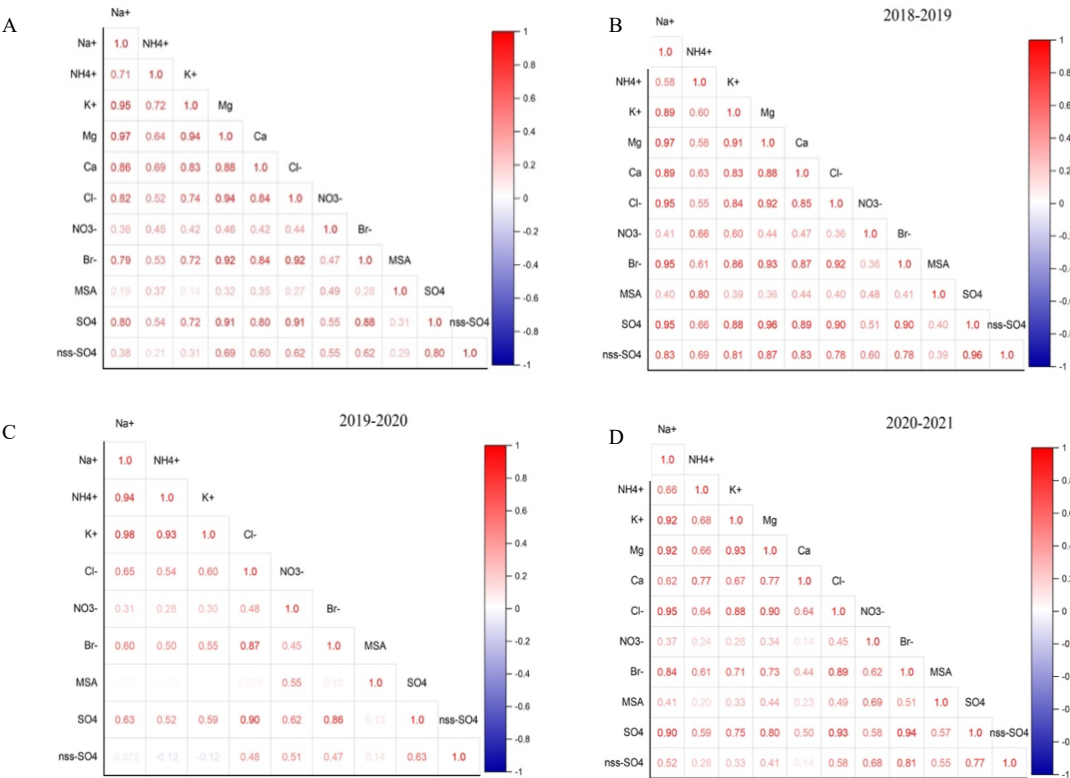


Fig. 5. Spearman correlation plots. A) Correlation plot for the three sampling seasons; B) Correlation plot for the 2018-2019 season; C) Correlation plot for the 2019-2020 season; D) Correlation plot for the 2020-2021 season.

497 To quantify the biogenic nss-SO_4^{2-} contribution, the methanesulfonic acid (MSA) loads - the final
498 product of DMS oxidation - were multiplied by 3.0 (Udisti et al., 2016), revealing biogenic SO_4^{2-}
499 contributions ranging from 0.15% (2018-19, 2020-21) up to 0.38% (2019-20). Furthermore, the
500 MSA/ nss-SO_4^{2-} ratio was inspected, revealing a mean value of 0.02 ± 0.03 during the first (2018-19)
501 and the third (2020-21) sampling campaigns, and a maximum ratio equal to 0.06 ± 0.18 reached
502 during the second campaign (2019-20), similar to the subarctic western North Pacific ratio found by
503 Jung et al. (2014). However, several factors can influence MSA formation, a univocal marker of
504 biogenic emissions, including higher biological productivity related to higher nutrient input; the
505 concentrations of NO_3 radicals as key oxidants for DMS decomposition (higher NO_3 gives higher
506 MSA); and lower air temperatures, which tend to yield higher MSA levels (Andreae et al., 1985;
507 Udisti et al., 2020).

508 For the 2019-20 campaign, it seems likely that a combination of these three factors, together with the
509 positive expansion of sea ice and the very close drift ice presence in March 2020, as revealed from
510 satellite reconstructions (Fig. S6), contributed to the increased release of MSA in aerosol, and its
511 consistent deposition in surface snow (Fig. 3). Indeed, DMS was likely accumulated under the sea
512 ice cover in the fjord and surrounding areas, and then being released and oxidised in the atmosphere
513 when the ice broke off and melted (April-May). Furthermore, lower temperatures (-17.3°C in March
514 2020, compared to -16.1°C in March 2019, and -9.8°C in March 2021), positive correlation between
515 MSA and NO_3^- ($\rho_{\text{load}} = 0.49$, $p\text{-value} < 0.05$), and high-speed short-range transport (wind directions
516 between 0° - 60° and speeds $> 5 \text{ m sec}^{-1}$) from the source to the near-coast sink site (GSRS) would
517 have aided elevated concentrations of MSA in atmospheric depositions. However, the dominant south
518 and southwest winds (180° - 240°) during the major MSA peak in April 2020 (Fig. 3) likely transported
519 marine aerosols and DMS from open ocean regions, further facilitating the increased MSA
520 concentrations observed.

521 Contrarily, in the 2018-19 season, sea ice lasted only until April and was restricted to the inner,
522 shallower parts of Kongsfjorden (Assmy et al., 2023), possibly not allowing enough time with
523 adequate sunlight for substantial biological activity to accumulate beneath or within it. This occurred
524 despite the dominance in 2019, unlike the following year, of *Phaeocystis pouchetii*, a phytoplankton
525 species known for its capacity to generate DMS in significant quantities, according to Assmy et al.
526 (2023).

527 Finally, the ammonium (NH_4^+) load showed positive correlations (Fig. 5) with Na^+ ($\rho_{\text{load}} = 0.71$, $p\text{-value} < 0.05$),
528 Cl^- ($\rho_{\text{load}} = 0.52$, $p\text{-value} < 0.05$) and K^+ ($\rho_{\text{load}} = 0.72$, $p\text{-value} < 0.05$), as well as with

529 SO_4^{2-} ($\rho_{\text{load}} = 0.54$, $p\text{-value} < 0.05$), NO_3^- ($\rho_{\text{load}} = 0.45$, $p\text{-value} < 0.05$), MSA ($\rho_{\text{load}} = 0.37$, $p\text{-value} <$
530 0.05) and Br^- ($\rho_{\text{load}} = 0.53$, $p\text{-value} < 0.05$), suggesting a close link with sea-salt ions and biogenic
531 emissions. However, some contribution from biomass burning events and potential influence from
532 anthropogenic activities cannot be excluded.

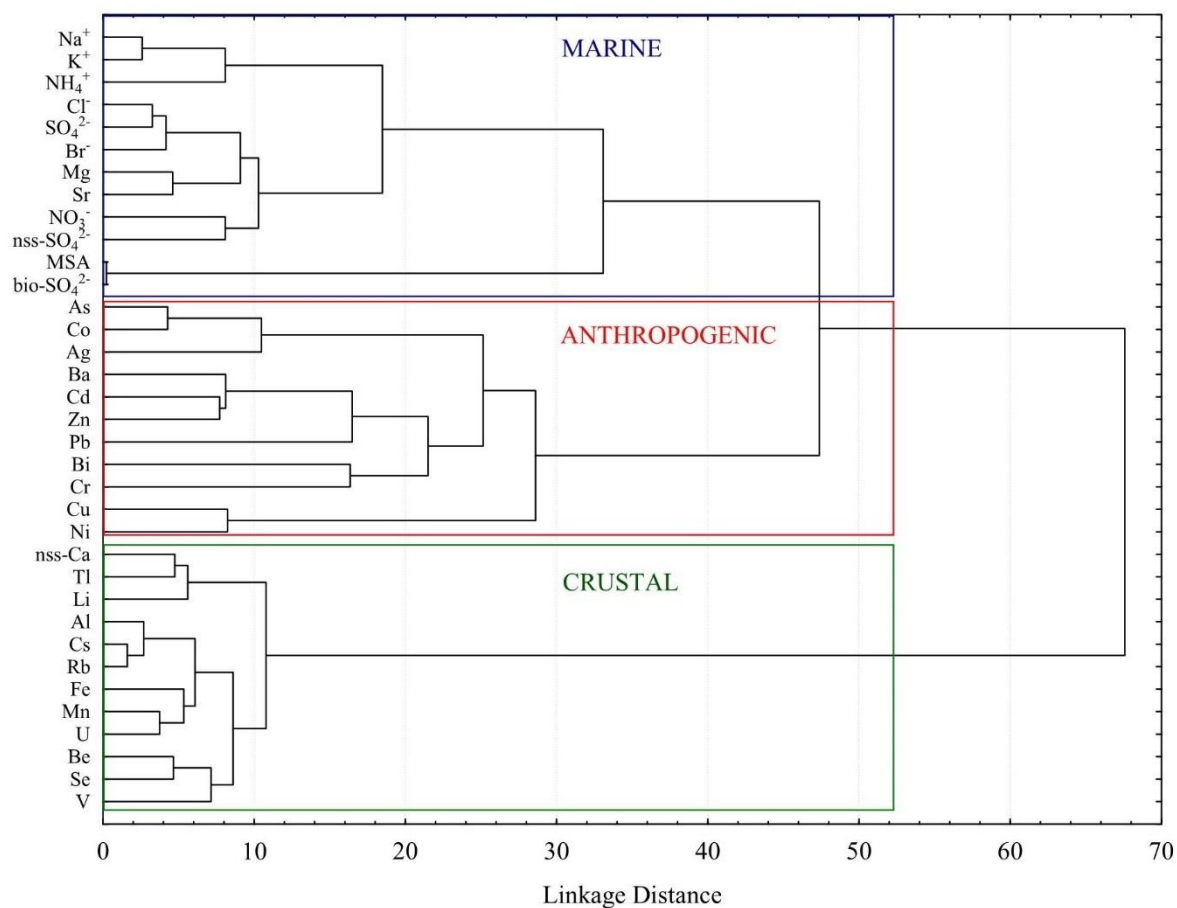
533 *4.3 Bromine enrichment*

534 The bromine enrichment factor (Br_{enr}) is well known to reflect specific processes (i.e., sea ice gas
535 phase Br emission) that affect the Br concentration and load in the snowpack (Spolaor et al., 2014).
536 Therefore, calculating the relative enrichment over the Br/Na ratio in sea water can offer crucial
537 insights on sea ice variability for the investigated Arctic areas (Barbaro et al., 2021). As reported in
538 previous studies (Maffezzoli et al., 2017; Barbaro et al., 2021), the Br enrichment factor (Br_{enr}) can
539 be calculated as $\text{Br}_{\text{enr}} = \text{Br}^- / (0.0065 \text{ Na}^+)$, where 0.0065 represents the Br^-/Na^+ seawater mass ratio.
540 Contrarily to what observed in a former study (Barbaro et al., 2021) for the Hornsund area and north-
541 western Spitsbergen, where the Br_{enr} mean values were often < 1 , indicating some Br^- depletion, in
542 this study we observed Br_{enr} mean values ranging from 1.5 up to 17.7, with the highest value
543 associated to the second sampling campaign conducted in 2019-20, which showed the most extensive
544 sea ice coverage. These results support the impact of the sea ice expansion and the close drift ice in
545 the Kongsfjorden on the snow chemical composition. Indeed, newly formed sea ice releases gas-
546 phase Br into the polar atmosphere, thus supplying an extra Br^- source in addition to sea spray
547 (Spolaor et al., 2016).

548 *4.4 Anthropogenic and natural sources of ions and particulate trace elements*

549 To complement the EFs analysis and further distinguish possible anthropogenic contributions from
550 natural ones (marine and geogenic) for ions and particulate trace elements, a Hierarchical Cluster
551 Analysis (HCA) method was carried out. Results of clustering (Fig. 6) clearly disentangle marine
552 (Na^+ , Cl^- , K^+ , NH_4^+ , SO_4^{2-} , NO_3^- , Br^- , Mg, Sr, bio- SO_4^{2-} , MSA), anthropogenic (As, Co, Ag, Ba, Cd,
553 Zn, Pb, Bi, Cr, Cu, Ni), and geogenic (nss-Ca, Tl, Li, Al, Cs, Rb, Fe, Mn, U, Be, Se, V) sources of
554 ionic and elemental species, considering the whole sampling campaign period (2018-2021).
555 Interestingly, nss- SO_4^{2-} is brought together with the marine cluster, suggesting that its presence is
556 largely influenced by marine biogenic sources, alongside contributions from secondary sulphate
557 formation in the atmosphere. This indicates that nss- SO_4^{2-} , despite having a variety of sources such
558 as human contribution or dust, is closely linked to the marine environment. One important reason for
559 this is the emission of DMS by phytoplankton. Additionally, secondary sulphate formation may have

560 further contributed to the nss-SO_4^{2-} . Cobalt was grouped within the anthropogenic cluster in HCA, in
 561 contrast with EFs that suggested a crustal origin. This apparent discrepancy may reflect the relatively
 562 larger uncertainties typically associated with EF calculations, which can inherit errors from the choice
 563 of reference element, assumptions about crustal composition, and variability in background
 564 concentrations, compared to the more integrative approach of HCA (e.g., Reimann and de Caritat,
 565 2000). Moreover, the trend for Co closely aligns with anthropogenic ones (e.g., As), while distinct
 566 trends are evident for crustal elements. Therefore, while the HCA results offer a reliable perspective
 567 on source differentiation and clustering patterns, they are best interpreted as complementary to the
 568 EF calculations rather than directly integrated with them. Incorporating EFs directly into the HCA
 569 could introduce significant inaccuracies, as EFs rely on reference element ratios that may vary and
 570 thus add complexity to the clustering process, potentially skewing the results. Consequently, HCA
 571 independently provides robust insights in this context, enhancing our understanding without the
 572 additional uncertainties that EF-based clustering might introduce.



573
 574 **Fig. 6.** Hierarchical cluster analysis applied to further disentangle the particulate trace element non-crustal sources.
 575

576 **5. Summary and Conclusion**

577 In this study, trace elements and major ions were investigated in surface snow samples collected in
578 Ny-Ålesund between October 2018 to June 2021. Seasonal and interannual variations of impurities
579 have been observed, with general higher concentrations of marine species revealed in late spring
580 2020, associated to more extensive sea ice in Kongsfjorden in March 2020, promoted by negative
581 temperature anomalies in both atmosphere and ocean and likely related to higher air mass recycle
582 within the Arctic. These results provide direct evidence of how sea ice extent modulates the storage,
583 release, and transport of marine-derived impurities, thereby influencing snow-atmosphere chemical
584 exchange processes under varying climatic conditions. Higher concentrations in spring 2020 for
585 geogenic and anthropogenic species are consistent with the influence of higher wind speeds, low
586 atmospheric temperature anomalies, and generally drier conditions resulting from the exceptional
587 occurrence of a strong and cold stratospheric polar vortex, accompanied by an unprecedentedly positive
588 phase of the Arctic Oscillation in the troposphere during January-March 2020. While correlations
589 between meteorological variables and concentrations were generally weak, the overall meteorological
590 context suggests that these unusual conditions likely contributed to the observed patterns. This is
591 particularly evident especially in cold years, when the production of sea spray related aerosol likely
592 derives by a larger extension of sea ice and stronger local Arctic circulation. The identification of
593 geogenic, marine, and anthropogenic sources in the snowpack was allowed by a chemometric
594 approach (HCA), which clarified the EFs results. The use of chemometric techniques (HCA) and
595 back-trajectory analysis enabled a clearer attribution of sources and transport pathways, improving
596 the interpretation of snow composition in relation to meteorological drivers. Specifically, the distinct
597 seasonal air mass patterns revealed, characterised by dominant Arctic-origin air masses in fall and
598 winter and a lack of expected mid-latitude inputs in spring, underscore the changing dynamics of
599 snow-atmosphere interactions in a warming Arctic. Overall, these insights advance our understanding
600 of how recent climatic anomalies, such as altered sea ice extent, shifts in Arctic Oscillation phases,
601 and stronger polar vortices, modulate the chemical composition of the snowpack in Svalbard. Our
602 findings highlight the sensitivity of snow-atmosphere exchanges to both local and large-scale climatic
603 processes, offering important context for interpreting snow chemistry trends in a rapidly changing
604 Arctic environment. Further statistical investigations, including multivariate and longer-term
605 analyses, would be valuable for better quantifying these correlations.

606 **Data availability**

607 The data supporting the findings of this study are available within the article and its supplementary
608 materials. Other data that support the findings of this study are available from the corresponding
609 author upon request.

610 **Author contribution**

611 AS: Conceptualization, Data curation, Investigation, Writing-original draft, Writing-review and
612 editing. EB: Conceptualization, Field work, Data curation, Formal Analysis, Writing-original draft,
613 Writing-review and editing, Funding acquisition. MF: Formal Analysis, Field work, Data curation,
614 Writing-review and editing. FS: Field work, Formal analysis, Investigation, Writing-review and
615 editing. MV: Writing-review and editing. MV: Field work, Writing-review and editing. MM:
616 Investigation. FB: Investigation, Writing-review and editing. FB: Investigation. Field work. CJMH:
617 Investigation, Data curation, Writing-review and editing. AB: Field work, Data curation, Writing-
618 review and editing. AG: Resources, Supervision, Validation, Writing-review and editing, Funding
619 acquisition. CB: Resources, Supervision, Validation, Writing-review and editing, Funding
620 acquisition. AS: Funding acquisition, Supervision, Validation, Writing-review and editing.

621 **Competing interests**

622 The authors declare that they have no conflict of interest.

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635 **References**

636 Amore, A., Giardi, F., Becagli, S., Caiazzo, L., Mazzola, M., Severi, M., Traversi, R.: Source
637 apportionment of sulphate in the High Arctic by a 10 yr-long record from Gruebadet Observatory
638 (Ny-Ålesund, Svalbard Islands). *Atmos. Environ.*, 270, 118890, 1-10,
639 <https://doi.org/10.1016/j.atmosenv.2021.118890>, 2022

640

641 Andreae, M.O., Ferek, R.J., Bermond, F., Byrd, K.P., Engstrom, R.T., Hardin, S., Houmère,
 642 P.D., LeMarrec, F., Raemdonck, H., Chatfield, R.B.: Dimethyl sulfide in the marine atmosphere,
 643 *J. Geophys. Res.*, 90, D7, 12891-12900, <https://doi.org/10.1029/JD090iD07p12891>, 1985

644 Assmy, P., Kvernvik, A.C., Hop, H., Hoppe, C.J.M., Chierici, M., David, D., Duarte, P.,
 645 Fransson, A., García, L.M., Patuła, W., Kwaśniewski, S., Maturilli, M., Pavlova, O., Tatarek, A.,
 646 Wiktor, J.M., Wold, A., Wolf, K.K.E., Bailey, A.: Seasonal plankton dynamics in Kongsfjorden
 647 during two years of contrasting environmental conditions, *Prog. Oceanogr.*, 213, 102996,
 648 <https://doi.org/10.1016/j.pocean.2023.102996>, 2023

649 Barbaro, E., Koziol, K., Björkman, M.P., Vega, C.P., Zdanowicz, C., Martma, T., Gallet, J.C.,
 650 Kępski, D., Larose, C., Luks, B., Tolle, F., Schuler, T. V., Uszczyk, A., Spolaor, A.: Measurement
 651 report: Spatial variations in ionic chemistry and water-stable isotopes in the snowpack on glaciers
 652 across Svalbard during the 2015-2016 snow accumulation season, *Atmos. Chem. Phys.*, 21, 3163–
 653 3180, <https://doi.org/10.5194/acp-21-3163-2021>, 2021

654 Barbaro, E., Padoan, S., Kirchgeorg, T., Zangrando, R., Toscano, G., Barbante, C., Gambaro,
 655 A.: Particle size distribution of inorganic and organic ions in coastal and inland Antarctic aerosol,
 656 *Environ. Sci. Pollut. Res.*, 24, 2724-2733, <https://doi.org/10.1007/s11356-016-8042-x>, 2017

657 Barbaro, E., Zangrando, R., Vecchiato, M., Piazza, R., Cairns, W.R.L., Capodaglio, G.,
 658 Barbante, C., Gambaro, A.: Free amino acids in Antarctic aerosol: Potential markers for the evolution
 659 and fate of marine aerosol, *Atmos. Chem. Phys.*, 15, 5457–5469, [https://doi.org/10.5194/acp-15-](https://doi.org/10.5194/acp-15-5457-2015)
 660 5457-2015, 2015

661 Bazzano, A., Ardini, F., Becagli, S., Traversi, R., Udisti, R., Cappelletti, D., Grotti, M.: Source
 662 assessment of atmospheric lead measured at Ny-Ålesund, Svalbard, *Atmos. Environ.*, 113, 20-26,
 663 <https://doi.org/10.1016/j.atmosenv.2015.04.053>, 2015

664 Bazzano, A., Bertinetti, S., Ardini, F., Cappelletti, D., Grotti, M.: Potential Source Areas for
 665 atmospheric Lead reaching Ny-Ålesund from 2010 to 2018, *Atmos.*, 12, 388, 1-17,
 666 <https://doi.org/10.3390/atmos12030388>, 2021

667 Beaudon, E., Moore, J.: Frost flower chemical signature in winter snow on Vestfonna ice cap,
 668 *Nordautlandet, Svalbard, Cryosphere*, 3, 147–154, <https://doi.org/10.5194/tc-3-147-2009>, 2009

669 Beine, H.J., Dominé, F., Ianniello, A., Nardino, M., Allegrini, I., Teinilä, K., Hillamo, R.:
 670 Fluxes of nitrates between snow surfaces and the atmosphere in the European high Arctic, *Atmos.*
 671 *Chem. Phys.*, 3, 335-346, <https://doi.org/10.5194/acp-3-335-2003>, 2003

672 Bertò, M., Cappelletti, D., Barbaro, E., Varin, C., Gallet, J.-C., Markowicz, K.,
 673 Rozwadowska, A., Mazzola, M., Crocchianti, S., Poto, L., Laj, P., Barbante, C., Spolaor, A.:
 674 Variability in black carbon mass concentration in surface snow at Svalbard, *Atmos. Chem. Phys.*, 21,
 675 12479-12493, <https://doi.org/10.5194/acp-21-12479-2021>, 2021

676 Björkman, M., Kühnel, R., Partridge, D.G., Roberts, T.J., Aas, W., Mazzola, M., Viola, A.,
 677 Hodson, A., Ström, J., Isaksson, E.: Nitrate dry deposition in Svalbard, *Tellus B*, 65(1),
 678 <https://doi.org/10.3402/tellusb.v65i0.19071>, 2013

679 Chylek, P., Folland, C., Klett, J.D., Wang, M., Hengartner, N., Lesins, G., Dubey, M.K.:
 680 Annual Mean Arctic Amplification 1970-2020. Observed and Simulated by CMIP6 Climate Models,
 681 *Geophys. Res. Lett.*, 49 (13), e2022GL099371, <https://doi.org/10.1029/2022GL099371>, 2022

682 D'Amico, M., Kallenborn, R., Scotto, F., Gambaro, A., Gallet, J.-C., Spolaor, A., Vecchiato,
683 M.: Chemicals of Emerging Arctic Concern in north-western Spitsbergen snow: Distribution and
684 sources, *Sci. Total Environ.*, 908, 168401, <https://doi.org/10.1016/j.scitotenv.2023.168401>, 2024

685 Dethloff, K., Maslowski, W., Hendricks, S., Lee, Y.J., Goessling, H.F., Krumpen, T., Haas,
686 C., Handorf, D., Ricker, R., Bessnov, V., Cassano, J.J., Kinney, J.C., Osinski, R., Rex, M., Rinke, A.,
687 Sokolova, J., Sommerfeld, A.: Arctic sea ice anomalies during the MOSAiC winter 2019/20,
688 *Cryosphere*, 16, 981-1005, <https://doi.org/10.5194/tc-16-981-2022>, 2022

689 Draxler, R.R.: An overview of the HYSPLIT_4 modelling system for trajectories, dispersion
690 and deposition, *Aust. Meteorol. Mag.*, 47, 295–308, 1998

691 Dunn, O.J.: Multiple comparisons among means, *J. Am. Stat. Assoc.*, 56, 293,
692 <https://doi.org/10.1080/01621459.1961.10482090>, 1961

693 Eleftheriadis, K., Vratolis, S., Nyeki, S.: Aerosol black carbon in the European Arctic:
694 Measurements at Zeppelin station, Ny-Ålesund, Svalbard from 1998-2007, *Geophys. Res. Lett.*, 36,
695 1–5, <https://doi.org/10.1029/2008GL035741>, 2009

696 Feltracco, M., Barbaro, E., Hoppe, C.J.M., Wolf, K.K.E., Spolaor, A., Layton, R., Keuschnig,
697 C., Barbante, C., Gambaro, A., Larose, C.: Airborne bacteria and particulate chemistry capture
698 Phytoplankton bloom dynamics in an Arctic fjord, *Atmos. Environ.*, 256, 118458,
699 <https://doi.org/10.1016/j.atmosenv.2021.118458>, 2021

700 Feltracco, M., Barbaro, E., Tedeschi, S., Spolaor, A., Turetta, C., Vecchiato, M., Morabito,
701 E., Zangrando, R., Barbante, C., Gambaro, A.: Interannual variability of sugars in Arctic aerosol:
702 Biomass burning and biogenic inputs, *Sci. Total Environ.*, 706, 136089,
703 <https://doi.org/10.1016/j.scitotenv.2019.136089>, 2020

704 Geng, H., Ryu, J., Jung, H.J., Chung, H., Ahn, K.H.O., Ro, C.U.N.: Single-particle
705 characterization of summertime arctic aerosols collected at Ny-Ålesund, Svalbard, *Environ. Sci.*
706 *Technol.*, 44, 2348–2353, <https://doi.org/10.1021/es903268j>, 2010

707 George, B.J., Gains-Germain, L., Broms, K., Black, K., Furman, M., Hays, M.D., Thomas,
708 K.W., Simmons, J.E.: Censoring trace-level environmental data: statistical analysis considerations to
709 limit bias, *Environ. Sci. Technol.*, 55(6), 3786-3795, <https://doi.org/10.1021/acs.est.0c02256>, 2021

710 Gerland, S., Pavlova, O., Marnela, M., Divine, D.V., Kohler, J., Renner, A., Skoglund, A.:
711 Sea ice extent variability in Kongsfjorden, Svalbard during 2003-2021, based on visual observations
712 from the mountain Zeppelinfjellet, Norwegian Polar Institute,
713 <https://doi.org/10.21334/npolar.2022.d6d31f5b>, 2022

714 Gjermundsen, A., Seland Graff, L., Bentsen, M., Anders Breivik, L., Boldingh Debernard, J.,
715 Makkonen, R., Olivié, D.J.L., Seland, Ø., Zieger, P., Schulz, M.: How representative is Svalbard for
716 future Arctic climate evolution? An Earth system modelling perspective (SvalCLIM), SESS Report
717 2020 – The State of Environmental Science in Svalbard, <https://doi.org/10.5281/zenodo.4034104>,
718 2020

719 Hansen, B.B., Isaksen, K., Benestad, R.E., Kohler, J., Pedersen, Å., Loe, L.E., Coulson, S.J.,
720 Larsen, J.O., Varpe, Ø.: Warmer and wetter winters: Characteristics and implications of an extreme
721 weather event in the High Arctic, *Environ. Res. Lett.*, 9, [https://doi.org/10.1088/1748-](https://doi.org/10.1088/1748-9326/9/11/114021)
722 [9326/9/11/114021](https://doi.org/10.1088/1748-9326/9/11/114021), 2014

723 Hodgkins, R., Tranter, M.: Solute in High Arctic glacier snow cover and its impact on runoff
724 chemistry, *Ann. Glaciol.*, 26, 156-160, <https://doi.org/10.3189/1998AoG26-1-156-160>, 1998

725 Jacobi, H.-W., Obleitner, F., Da Costa, S., Ginot, P., Eleftheriadis, K., Aas, W., Zanatta, M.:
726 Deposition of ionic species and black carbon to the Arctic snowpack: combining snow pit
727 observations with modeling, *Atmos. Chem. Phys.*, 19, 10361-10377,
728 <https://doi.org/10.3402/tellusb.v65i0.19071>, 2019

729 Jung, J., Furutani, H., Uematsu, M., Park, J.: Distributions of atmospheric non-sea-salt
730 sulphate and methanesulfonic acid over the Pacific Ocean between 48°N and 55°S during summer,
731 *Atmos. Environ.*, 99, 374-384, <https://doi.org/10.1016/j.atmosenv.2014.10.009>, 2014

732 Koziol, K., Uszczyk, A., Pawlak, F., Frankowski, M., Polkowska, Ż.: Seasonal and Spatial
733 Differences in Metal and Metalloid Concentrations in the Snow Cover of Hansbreen, Svalbard, *Front.*
734 *Earth Sci.*, 8, 1–8, <https://doi.org/10.3389/feart.2020.538762>, 2021

735 Lai, A.M., Shafer, M.M., Dibb, J.E., Polashenski, C.M., Schauer, J.J.: Elements and inorganic
736 ions as source tracers in recent Greenland snow, *Atmos. Environ.*, 164, 205–215,
737 <https://doi.org/10.1016/j.atmosenv.2017.05.048>, 2017

738 Lawrence, Z.D., Perlwitz, J., Butler, A.H., Manney, G.L., Newman, P.A., Lee, S.H., Nash,
739 E.R.: The Remarkably Strong Arctic Stratospheric Polar Vortex of Winter 2020: Links to Record-
740 Breaking Arctic Oscillation and Ozone Loss, *J. Geophys. Res. Atmos.*, 125, 1–21,
741 <https://doi.org/10.1029/2020JD033271>, 2020

742 Maffezzoli, N., Spolaor, A., Barbante, C., Bertò, M., Frezzotti, M., Vallelonga, P.: Bromine,
743 iodine and sodium in surface snow along the 2013 Talos Dome-GV7 traverse (northern Victoria Land,
744 East Antarctica), *Cryosphere*, 11, 693-705, <https://doi.org/10.5194/tc-11-693-2017>, 2017

745 Millero, F.J., Feistel, R., Wright, D.G., McDougall, T.J.: The composition of Standard
746 Seawater and the definition of the Reference-Composition Salinity Scale, *Deep-Sea Res.*, 55, 50–72,
747 2008

748 Morales, J.A., Pirela, D., de Nava, M.G., de Borrego, B.S., Velásquez, H., Durán, J.: Inorganic
749 water soluble ions in atmospheric particles over Maracaibo Lake Basin in the western region of
750 Venezuela, *Atmos. Res.*, 46, [https://doi.org/10.1016/S0169-8095\(97\)00071-9](https://doi.org/10.1016/S0169-8095(97)00071-9), 1998

751 Nawrot, A.P., Migąła, K., Luks, B., Pakszys, P., Głowacki, P.: Chemistry of snow cover and
752 acidic snowfall during a season with a high level of air pollution on the Hans Glacier, Spitsbergen,
753 *Polar Sci.*, 10, 249–261, <https://doi.org/10.1016/j.polar.2016.06.003>, 2016

754 Platt, S.M., Hov, Ø., Berg, T., Breivik, K., Eckhardt, S., Eleftheriadis, K., Evangeliou, N.,
755 Fiebig, M., Fisher, R., Hansen, G., Hansson, H.-C., Heintzenberg, J., Hermansen, O., Heslin-Rees,
756 D., Holmén, K., Hudson, S., Kallenborn, R., Krejci, R., Krognes, T., Larssen, S., Lowry, D., Lund
757 Myhre, C., Lunder, C., Nisbet, E., Nizzetto, P.B., Park, K.-T., Pedersen, C.A., Pfaffhuber, K.A.,
758 Röckmann, T., Schmidbauer, N., Solberg, S., Stohl, A., Ström, J., Svendby, T., Tunved, P., Tørnkvist,
759 K., van der Veen, C., Vratolis, S., Yoon, Y.L., Yttri, K.E., Zieger, P., Aas, W., Tørseth, K.:
760 Atmospheric composition in the European Arctic and 30 years of the Zeppelin Observatory, Ny-
761 Ålesund, *Atmos. Chem. Phys.*, 22, 3321-3369, <https://doi.org/10.5194/acp-22-3321-2022>, 2022

762 Pomeroy, J.W.: A process-based model of snow drifting, *Ann. Glaciol.*, 13, 237-240,
763 <https://doi.org/10.3189/S0260305500007965>, 1989

Rankin, A.M., Wolff, E.W.: Frost flowers: Implications for tropospheric chemistry and ice core interpretation, *J. Geophys. Res.*, 107, D23, 4683, <https://doi.org/10.1029/2002JD002492>, 2002

Rantanen, M., Karpechko, A.Y., Lipponen, A., Nordling, K., Hyvärinen, O., Ruosteenoja, K., Vihma, T., Laaksonen, A.: The Arctic has warmed nearly four times faster than the globe since 1979, *Commun. Earth Environ*, 3, 168, <https://doi.org/10.1038/s43247-022-00498-3>, 2022

Reimann, C., De Caritat, P.: Intrinsic flaws of element Enrichment Factors (EFs) in environmental geochemistry, *Environ. Sci. Technol.*, 34, <https://doi.org/10.1021/es001339o>, 2000

Rinke, A., Maturilli, M., Graham, R.M., Matthes, H., Handorf, D., Cohen, L., Hudson, S.R., Moore, J.C.: Extreme cyclone events in the Arctic: Wintertime variability and trends, *Environ. Res. Lett.*, 12, <https://doi.org/10.1088/1748-9326/aa7def>, 2017

Roscoe, H.K., Brooks, B., Jackson, A.V., Smith, M.H., Walker, S.J., Obbard, R.W., Wolff, E.W.: Frost flowers in the laboratory: Growth, characteristics, aerosol, and the underlying sea ice, *J. Geophys. Res.*, 116, D12301, <https://doi.org/10.1029/2010JD015144>, 2011

Ruppel, M.M., Khedr, M., Liu, X., Beaudon, E., Szidat, S., Tunved, P., Ström, J., Koponen, H., Sippula, O., Isaksson, E., Gallet, J.-C., Hermanson, M., Manninen, S., Schnelle-Kreis, J.: Organic compounds, radiocarbon, trace elements and atmospheric transport illuminating sources of elemental carbon in a 300-year Svalbard ice core, *J. Geophys. Res.: Atmos.*, 128, <https://doi.org/10.1029/2022JD038378>, 2023

Salzano, R., Cerrato, R., Scoto, F., Spolaor, A., Valentini, E., Salvatore, M., Esposito, G., Sapio, S., Taramelli, A., Salvatori, R.: Detection of winter heat wave impact on surface runoff in a periglacial environment (Ny-Ålesund, Svalbard), *Remote Sens.*, 15(18), 4435, <https://doi.org/10.3390/rs15184435>, 2023

Schoeberl, M.R., Newman, P.A.: Middle Atmosphere: Polar Vortex, *Encyclopedia of Atmospheric Sciences*, Second Edition, Elsevier, 12-17, <https://doi.org/10.1016/B978-0-12-382225-3.00228-0>, 2015

Schwikowski, M., Döschner, A., Gäggeler, H.W., Schotterer, U.: Anthropogenic versus natural sources of atmospheric sulphate from an Alpine ice core. *Tellus B: Chemical and Physical Meteorology*, 51:5, 938-951, <https://doi.org/10.3402/tellusb.v51i5.16506>, 1999

Scoto, F., Pappaccogli, G., Mazzola, M., Donato, A., Salzano, R., Monzali, M., de Blasi, F., Larose, C., Gallet, J.-C., Decesari, S., Spolaor, A.: Automated observation of physical snowpack properties in Ny-Ålesund, *Front. Earth Sci.*, 11:1123981, 1-9, <https://doi.org/10.3389/feart.2023.1123981>, 2023

Serreze, M.C., Barry, R.G.: Processes and impacts of Arctic amplification: A research synthesis, *Glob. Planet. Change*, 77, 85-96, <http://dx.doi.org/10.1016/j.gloplacha.2011.03.004>, 2011

Sherrell, R.M., Boyle, E.A., Harris, N.R., Falkner, K.K.: Temporal variability of Cd, Pb, and Pb isotope deposition in central Greenland snow, *Geochem Geophys*, 1 (1), 1-22, <https://doi.org/10.1029/1999GC000007>, 2000

Sobota, I., Weckwerth, P., Grajewski, T.: Rain-On-Snow (ROS) events and their relations to snowpack and ice layer changes on small glaciers in Svalbard, the high Arctic, *J. Hydrol*, 590, 125279, <https://doi.org/10.1016/j.jhydrol.2020.125279>, 2020

804 Song, C., Becagli, S., Beddows, D.C.S., Brean, J., Browse, J., Dai, Q., Dall'Osto, M., Ferracci,
805 V., Harrison, R.M., Harris, N., Li, W., Jones, A.E., Kirchg  bner, A., Kramawijaya, A.G., Kurganskiy,
806 A., Lupi, A., Mazzola, M., Severi, M., Traversi, R., Shi, Z.: Understanding sources and drivers of
807 size-resolved aerosol in the High Arctic islands of Svalbard using a receptor model coupled with
808 machine learning, *Environ. Sci. Technol.*, 56, 11189-11198, <https://doi.org/10.1021/acs.est.1c07796>,
809 2022

810 Song, J., Zhao, Y., Zhang, Y., Fu, P., Zheng, L., Yuan, Q., Wang, S., Huang, X., Xu, W., Cao,
811 Z., Gromov, S., Lai, S.: Investigation of biomass burning on atmospheric aerosols over the western
812 South China Sea: Insights from ions, carbonaceous fractions and stable carbon isotope ratios,
813 *Environ. Pollut.*, 242, 1800-1809, <https://doi.org/10.1016/j.envpol.2018.07.088>, 2018

814 Spolaor, A., Scoto, F., Larose, C., Barbaro, E., Burgay, F., Bjorkman, M., Cappelletti, D.,
815 Dallo, F., de Blasi, F., Divine, D., Dreossi, G., Gabrieli, J., Isaksson, E., Kohler, J., Martma, T.,
816 Schmidt, L.S., Schuler, T.V., Stenni, B., Turetta, C., Luks, B., Casado, M., Gallet, J.-C.: Climate
817 change is rapidly deteriorating the climatic signal in Svalbard glaciers, *Cryosphere*, 18, 307-320,
818 <https://doi.org/10.5194/tc-18-307-2024>, 2024

819 Spolaor, A., Varin, C., Pedeli, X., Christille, J.M., Kirchgeorg, T., Giardi, F., Cappelletti, D.,
820 Turetta, C., Cairns, W.R.L., Gambaro, A., Bernagozzi, A., Gallet, J.C., Bj  rkman, M.P., Barbaro, E.:
821 Source, timing and dynamics of ionic species mobility in the Svalbard annual snowpack, *Sci. Total*
822 *Environ.*, 751, 141640, <https://doi.org/10.1016/j.scitotenv.2020.141640>, 2021b

823 Spolaor, A., Moroni, B., Luks, B., Nawrot, A., Roman, M., Larose, C., Stachnik,   ., Bruschi,
824 F., Kozio  , K., Pawlak, F., Turetta, C., Barbaro, E., Gallet, J.-C., Cappelletti, D.: Investigation on the
825 Sources and Impact of Trace Elements in the Annual Snowpack and the Firn in the Hansbreen
826 (Southwest Spitsbergen), *Front. Earth Sci.*, 8, 1–10, <https://doi.org/10.3389/feart.2020.536036>, 2021a

827 Spolaor, A., Barbaro, E., Cappelletti, D., Turetta, C., Mazzola, M., Giardi, F., Bj  rkman, M.P.,
828 Lucchetta, F., Dallo, F., Aspmo Pfaffhuber, K., Angot, H., Dommergue, A., Maturilli, M., Saiz-
829 Lopez, A., Barbante, C., Cairns, W.R.L.: Diurnal cycle of iodine, bromine, and mercury
830 concentrations in Svalbard surface snow, *Atmos. Chem. Phys.*, 19, 13325–13339,
831 <https://doi.org/10.5194/acp-19-13325-2019>, 2019

832 Spolaor, A., Angot, H., Roman, M., Dommergue, A., Scarchilli, C., Vard  , M., Del Guasta,
833 M., Pedeli, X., Varin, C., Sprovieri, F., Magand, O., Legrand, M., Barbante, C., Cairns, W.R.L.:
834 Feedback mechanisms between snow and atmospheric mercury: Results and observations from field
835 campaigns on the Antarctic plateau, *Chemosphere*, 197, 306–317,
836 <https://doi.org/10.1016/j.chemosphere.2017.12.180>, 2018

837 Spolaor, A., Opel, T., McConnell, J.R., Maselli, O.J., Spreen, G., Varin, C., Kirchgeorg, T.,
838 Fritzsche, D., Saiz-Lopez, A., Vallenga, P.: Halogen-based reconstruction of Russian Arctic sea ice
839 area from the Akedemii Nauk ice core (Severnaya Zemlya), *Cryosphere*, 10, 245-256,
840 <https://doi.org/10.5194/tc-10-245-2016>, 2016

841 Spolaor, A., Vallenga, P., Gabrieli, J., Martma, T., Bj  rkman, M.P., Isaksson, E., Cozzi, G.,
842 Turetta, C., Kj  er, H.A., Curran, M.A.J., Moy, A.D., Sch  nhardt, A., Blechschmidt, A.-M., Burrows,
843 J.P., Plane, J.M.C., Barbante, C.: Seasonality of halogen deposition in polar snow and ice, *Atmos.*
844 *Chem. Phys.*, 14, 18, 9613-9622, <https://doi.org/10.5194/acp-14-9613-2014>, 2014

845 Stein, A.F., Draxler, R.R., Rolph, G.D., Stunder, B.J.B., Cohen, M.D., Ngan, F.: Noaa's
846 hysplit atmospheric transport and dispersion modeling system, *Bull. Am. Meteorol. Soc.*, 96, 2059–
847 2077, <https://doi.org/10.1175/BAMS-D-14-00110.1>, 2015

848 Stohl, A., Berg, T., Burkhardt, J.F., Fjæraa, a. M., Forster, C., Herber, A., Hov, Ø., Lunder, C.,
849 McMillan, W.W., Oltmans, S., Shiobara, M., Simpson, D., Solberg, S., Stebel, K., Ström, J., Tørseth,
850 K., Treffeisen, R., Virkkunen, K., Yttri, K.E.: Arctic smoke – record high air pollution levels in the
851 European Arctic due to agricultural fires in Eastern Europe, *Atmos. Chem. Phys. Discuss.*, 6, 9655–
852 9722, <https://doi.org/10.5194/acpd-6-9655-2006>, 2006b

853 Stohl, A., Berg, T., Burkhardt, J.F., Fjæraa, a. M., Forster, C., Herber, A., Hov, Ø., Lunder, C.,
854 McMillan, W.W., Oltmans, S., Shiobara, M., Simpson, D., Solberg, S., Stebel, K., Ström, J., Tørseth,
855 K., Treffeisen, R., Virkkunen, K., Yttri, K.E.: Arctic smoke – record high air pollution levels in the
856 European Arctic due to agricultural fires in Eastern Europe, *Atmos. Chem. Phys. Discuss.*, 6, 9655–
857 9722, <https://doi.org/10.5194/acpd-6-9655-2006>, 2006a

858 Turetta, C., Feltracco, M., Barbaro, E., Spolaor, A., Barbante, C., Gambaro, A.: A year-round
859 measurement of water-soluble trace and rare earth elements in arctic aerosol: Possible inorganic
860 tracers of specific events, *Atmosphere*, 12, <https://doi.org/10.3390/atmos12060694>, 2021

861 Udisti, R., Traversi, R., Becagli, S., Tomasi, C., Mazzola, M., Lupi, A., Quinn, P.K.: Arctic
862 Aerosols, in: *Physics and Chemistry of the Arctic Atmosphere*, Springer Polar Sciences, edited by:
863 Kokhanovsky, A., Tomasi, C., 209-329, https://doi.org/10.1007/978-3-030-33566-3_4, 2020

864 Udisti, R., Bazzano, A., Becagli, S., Bolzacchini, E., Caiazzo, L., Cappelletti, D., Ferrero, L.,
865 Frosini, D., Giardi, F., Grotti, M., Lupi, A., Malandrino, M., Mazzola, M., Moroni, B., Severi, M.,
866 Traversi, R., Viola, A., Vitale, V.: Sulphate source apportionment in the Ny-Ålesund (Svalbard
867 Islands) Arctic aerosol, *Rend. Fis. Acc. Lincei*, 27, Suppl 1: S85-S94, [https://doi.org/10.1007/s12210-](https://doi.org/10.1007/s12210-016-0517-7)
868 016-0517-7, 2016

869 Van Hecke, T.: Power study of anova versus Kruskal-Wallis test, *J. Stat. Manag. Syst.*, 15, 2-
870 3, <https://doi.org/10.1080/09720510.2012.10701623>, 2013

871 Vecchiato, M., Barbante, C., Barbaro, E., Burgay, F., Cairns, W.R.L., Callegaro, A.,
872 Cappelletti, D., Dallo, F., D'Amico, M., Feltracco, M., Gallet J.-C., Gambaro, A., Larose, C.,
873 Maffezzoli, N., Mazzola, M., Sartorato, I., Scoto, F., Turetta, C., Vardè, M., Xie, Z., Spolaor, A.: The
874 seasonal change of PAHs in Svalbard surface snow, *Environ. Pollut.*, 340, 122864,
875 <https://doi.org/10.1016/j.envpol.2023.122864>, 2024

876 Vecchiato, M., Barbaro, E., Spolaor, A., Burgay, F., Barbante, C., Piazza, R., Gambaro, A.:
877 Fragrances and PAHs in snow and seawater of Ny-Ålesund (Svalbard): Local and long-range
878 contamination, *Environ. Pollut.*, 242, 1740-1747, <https://doi.org/10.1016/j.envpol.2018.07.095>, 2018

879 Vega, C.P., Björkman, M.P., Pohjola, V.A., Isaksson, E., Pettersson, R., Martma, T., Marca,
880 A., Kaiser, J., Vega, C.P., Björkman, M.P., Pohjola, V.A., Isaksson, E., Pettersson, R., Martma, T.,
881 Marca, A., Kaiser, J., Pohjola, V.A., Isaksson, E., Pettersson, R., Vega, C.P., Bjo, M.P.: Nitrate stable
882 isotopes and major ions in snow and ice samples from four Svalbard sites Nitrate stable isotopes and
883 major ions in snow and ice samples from four Svalbard sites, *Polar Res.*, 2015, 34, 23246,
884 <https://doi.org/10.3402/polar.v34.23246>, 2015

885 Wagenbach, D., Preunkert, S., Schäfer, J., Jung, W., Tomadin, L.: Northward transport of
 886 Saharan dust recorded in a deep alpine ice core, in: The impact of desert dust across the
 887 mediterranean, Springer, The Netherlands, edited by: Guerzoni, S., Chester, R., 291-300, 1996

 888 Wedepohl, K.H.: The composition of the chemical crust. *Geoch. Cosm. Act.*, 59(7), 1217-
 889 1232, [https://doi.org/10.1016/0016-7037\(95\)00038-2](https://doi.org/10.1016/0016-7037(95)00038-2), 1995

 890 Yttri, K.E., Lund Myhre, C., Eckhardt, S., Fiebig, M., Dye, C., Hirdman, D., Ström, J.,
 891 Klimont, Z., Stohl, A.: Quantifying black carbon from biomass burning by means of levoglucosan -
 892 A one-year time series at the Arctic observatory Zeppelin, *Atmos. Chem. Phys.*, 14, 6427–6442,
 893 <https://doi.org/10.5194/acp-14-6427-2014>, 2014a

 894 Zhan, J., Gao, Y., Li, W., Chen, L., Lin, H., Lin, Q.: Effects of ship emissions on summertime
 895 aerosols at Ny-Alesund in the Arctic, *Atmos. Pollut. Res.*, 5, 500–510,
 896 <https://doi.org/10.5094/APR.2014.059>, 2014

 897 Zhuang, H., Chan, C.K., Fang, M., Wexler, A.S.: Size distributions of particulate sulphate,
 898 nitrate, and ammonium at a coastal site in Hong Kong, *Atmos. Environ.*, 33, 843-853, 1999

 899
 900