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**1 Measurement report: Spatial variations in snowpack ionic chemistry and water stable
2 isotopes across Svalbard**

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4 Elena Barbaro^{1,2}, Krystyna Koziol³, Mats P. Björkman⁴, Carmen P. Vega⁵, Christian Zdanowicz⁶, Tonu
5 Martma⁷, Jean-Charles Gallet⁸, Daniel Kępski⁹, Catherine Larose¹⁰, Bartłomiej Luks⁹, Florian Tolle¹¹,
6 Thomas V. Schuler^{12,13}, Aleksander Uszczyk¹⁴ and Andrea Spolaor^{1,2*}

7
8 ¹Institute of Polar Sciences, ISP-CNR, Via Torino 155, 30170 Venice Mestre, Italy

9 ²Department of Environmental Sciences, Informatics and Statistics, Ca' Foscari University of Venice,
10 Via Torino 155, 30172, Venice, Italy.

11 ³Department of Analytical Chemistry, Chemical Faculty, Gdansk University of Technology, G.
12 Narutowicza 11/12, 80-233 Gdańsk, Poland.

13 ⁴Department of Earth Sciences, University of Gothenburg, Box 460, SE-40530 Gothenburg, Sweden.

14 ⁵Dirección Meteorológica de Chile, Dirección General de Aeronáutica Civil, Portales 3450, Santiago,
15 Chile. *Previously at:* Department of Earth Sciences, Uppsala University, Villavägen 16, Uppsala,
16 Sweden.

17 ⁶Department of Earth Sciences, Uppsala University, Villavägen 16, SE-76236, Uppsala, Sweden.

18 ⁷Department of Geology, Tallinn University of Technology, Ehitajate tee 5, 19086 Tallinn,
19 Estonia⁸Norwegian Polar Institute, Tromsø, No-9296, Norway

20 ⁹Institute of Geophysics, Polish Academy of Sciences, Księcia Janusza 64, 01-452 Warsaw, Poland

21 ¹⁰Environmental Microbial Genomics, Laboratoire Ampère, CNRS, University of Lyon, France

22 ¹¹Université de Franche-Comté, Besancon, FEMTO-ST, UMR 6174 CNRS

23 ¹²Departement of Geosciences, University of Oslo, Oslo, Norway

24 ¹³Arctic Geophysics, University Center on Svalbard, UNIS, Longyearbyen, Svalbard, Norway

25 ¹⁴University of Silesia in Katowice, Faculty of Natural Sciences, Będzińska 60, 41-200 Sosnowiec,
26 Poland

27
28 ***Corresponding author**

29 Andrea Spolaor (andrea.spolaor@cnr.it)

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36 Abstract

37 The Svalbard archipelago, between 74° and 81° N, is ~60% covered by glaciers and located at the Arctic
38 sea ice edge. The region experiences rapid variations in atmospheric flow during the snow season (from
39 late September to May) and can be affected by air advected both from lower and higher latitudes, which
40 likely impact the chemical composition of snowfall. While long-term changes in Svalbard snow
41 chemistry have been documented in ice cores drilled from two high-elevation glaciers, the spatial
42 variability of the snowpack composition across Svalbard is comparatively poorly understood. Here, we
43 report the results of the most comprehensive seasonal snow chemistry survey to date, carried out in April
44 2016 across 22 sites on 7 glaciers across the archipelago. At each glacier, three snow pits were sampled
45 along altitudinal profiles and the collected samples were analysed for major ions (Ca^{2+} , K^+ , Na^+ , Mg^{2+} ,
46 NH_4^+ , SO_4^{2-} , Br^- , Cl^- and NO_3^-) and stable water isotopes ($\delta^{18}\text{O}$, $\delta^2\text{H}$). The main aims were to investigate
47 the natural and anthropogenic processes influencing the snowpack and to better understand the influence
48 of atmospheric aerosol transport and deposition patterns on the snow chemical composition. The snow
49 deposited in the southern region of Svalbard was characterized by the highest total ionic loads, mainly
50 attributed to sea salt particles. Both NO_3^- and NH_4^+ in the seasonal snowpack reflected secondary aerosol
51 formation and post-depositional changes, resulting in very different spatial deposition patterns: NO_3^- had
52 its highest loading in northwestern Spitsbergen, and NH_4^+ in the southwest. The Br^- enrichment in snow
53 was highest in northeastern glacier sites closest to areas of extensive sea ice coverage. Spatial correlation
54 patterns between Na^+ and $\delta^{18}\text{O}$ suggest that the influence of long-range transport of aerosols on snow
55 chemistry is proportionally greater above 600-700 m a.s.l.

56

57 1. Introduction

58 Svalbard is a region of the Arctic experiencing rapid climate change, with a mean warming rate of +1.35
59 K per decade, much faster than the global average (Isaksen et al., 2016; Maturilli et al., 2013; Nordli et
60 al., 2014). This archipelago is located at the southern edge of the perennial Arctic sea ice, in the North
61 Atlantic Ocean, and is characterized by a maritime climate with large, rapid temperature variations
62 during winter (Brage et al., 2014). South-westerly inflow of mild oceanic air, associated with a low-
63 pressure system east of Iceland, often brings relatively warm and moist air in winter months, while Arctic
64 air intrusions from the north-east, driven by a high-pressure system over Greenland, result in much
65 colder temperatures (Rinke et al., 2017). In addition to these synoptic fluctuations, intense autumn or
66 winter cyclonic storms associated with anomalous warming events sometimes occur, transporting both
67 heat and moisture from lower latitudes to Svalbard (Rinke et al., 2017).

68

69 The aforementioned meteorological conditions also favor long-range transport of aerosols to the
70 archipelago, including pollutants from continental sources. Depending on the predominant air flow
71 pattern at the time of snowfall, the archipelago may experience regionally different amounts of both
72 snow accumulation (Eneroth et al., 2003; Forland et al., 2011) and chemical loads, the latter reflecting

73 contrasting mixtures of aerosol, varying by source area (Aas et al., 2016; Forsström et al., 2009; Möller
74 and Kohler, 2018). These regional differences are also associated with contrasts in sea ice cover. While
75 all Svalbard coasts are usually ice-free in summer, sea ice can form and cover large parts of the ocean
76 surface in the eastern and northern parts of the archipelago, while the southern and western parts often
77 remain ice-free (Dahlke et al., 2020), and therefore tend to experience greater snowfall owing to the
78 proximity of open water. In addition, the West Spitsbergen Current, a branch of the Atlantic Meridional
79 Overturning Circulation (AMOC) that flows to the west of the archipelago, causes markedly different
80 regional climatic conditions between its eastern and western parts (van Pelt et al., 2019): the west
81 exhibits higher temperatures and precipitation, while the east is less humid and cooler, and has also
82 experienced a stronger warming trend since 1957.

83
84 The seasonal snowpack contains a complex mixture of impurities that are either scavenged from the
85 atmosphere during snowfall or directly received through dry deposition (Kuhn, 2001). On land, the
86 majority of impurities found in seasonal snow are usually eluted during summer melting, influencing
87 terrestrial and aquatic systems (Björkman et al., 2014; Brimblecombe et al., 1987). However, in the
88 accumulation area of Arctic glaciers and ice caps, impurities can be retained within or below the seasonal
89 snow layer (Björkman et al., 2014; Pohjola et al., 2002; Vega et al., 2015b). For this reason, chemical
90 impurities such as major ions (Ca^{2+} , K^+ , Na^+ , Mg^{2+} , NH_4^+ , SO_4^{2-} , Br^- , Cl^- and NO_3^-) in ice cores have
91 been widely used to study past trends of atmospheric and climatic conditions (Barbante et al., 2017;
92 Isaksson et al., 2003; Thompson et al., 2002; Wolff et al., 2010). Previous studies in Svalbard (Goto-
93 Azuma et al., 1994; Nawrot et al., 2016; Semb et al., 1984; Winther et al., 2003) have shown that the
94 chemistry of the seasonal snowpack is dominated by sea salt ions (Hodgkins and Tranter, 2017).
95 However, the region is also a sink for atmospheric contaminants brought in by long-range transport
96 (Vecchiato et al., 2018). Investigations of precipitation and snow cover chemistry have predominantly
97 focused on the central and western parts of the archipelago (Kühnel et al., 2011; Nawrot et al., 2016;
98 Vega et al., 2015a; Virkkunen et al., 2007), due to the accessibility of research facilities in these sectors.

99
100 In the present study, the concentration, mass loading, spatial and altitudinal distribution of major ion
101 species (Ca^{2+} , K^+ , Na^+ , Mg^{2+} , NH_4^+ , SO_4^{2-} , Br^- , Cl^- and NO_3^-) in snow, together with its stable oxygen
102 and hydrogen isotope composition ($\delta^{18}\text{O}$ and $\delta^2\text{H}$), were evaluated in the late winter snowpack at 22
103 glacier sites across Svalbard. This study was part of the larger Community Coordinated Snow Study in
104 Svalbard (C2S3) project and the most comprehensive survey of seasonal snow chemistry in Svalbard to
105 date. The snowpack survey, which was carried out by coordinated teams using a standardized sampling
106 protocol (Gallet et al., 2018) aimed to map and characterize regional differences in the chemical
107 composition and impurity load of the winter snow pack, and interpret the observed differences in relation
108 to meteorological and other environmental factors. In this way, we aim to identify the conditions

controlling the chemistry of Svalbard snow that are susceptible to be influenced by future climate warming across the region.

111

2. Methods

2.1 Sampling location and strategy

During April 2016, the seasonal snowpack was sampled at 22 sites on seven glaciers across Svalbard: Austfonna (AF) on Nordaustlandet, Lomonosovfonna (LF) on central Spitsbergen, Hansbreen (HB) and Werenskiöldbreen (WB) in the Hornsund area of southern Spitsbergen, and Austre Lovénbreen (ALB), Kongsvegen (KVG) and Holtedahlfonna (HDF) in the vicinity of Ny-Ålesund on northwestern Spitsbergen (Table 1 and Figure 1). The glaciers are of different sizes and hypsometries. Each glacier was sampled in the ablation zone, close to the equilibrium line altitude (ELA), and in the accumulation zone (Table 1). The ELA is the elevation at which the surface mass balance is zero, i.e., where the accumulation of snow is exactly balanced by ablation over a period of a year (Cogley et al., 2011). Although the exact elevation range of these zones differ for each glacier, they share enough glaciological similarities to support inter-site comparisons. Snowpit sampling was performed using a standardized common protocol (Gallet et al., 2018) with pre-cleaned equipment (i.e., tubes, plastic scrapers and plastic shovels cleaned with ultrapure water) and protective clothing (powder-free plastic gloves, clean suits and face masks). Samples for ionic chemistry were taken in each discrete snow pit layer, according to the visible stratigraphy, and directly into pre-cleaned, 50 mL polypropylene "Falcon" centrifuge tubes. Samples for the isotopic composition of water were collected at a 5-cm resolution for sites in the Ny-Ålesund area and at a 10-cm or stratigraphic layer resolution for other sites. All sampling was conducted in a safe distance and upwind from potential local pollution sources, such as the snowmobiles used as transport by the sampling team.

132

2.2 Major ion analyses

Samples from glaciers in the Hornsund region (HB, WB) were analysed at the Polish Polar Station Hornsund (Institute of Geophysics, Polish Academy of Sciences), while samples from glaciers near Ny-Ålesund (KVG, ALB, HDF) were shipped frozen to the Institute of Polar Sciences (ISP-CNR) in Venice (Italy). Snow sampled in central Spitsbergen (AF, LF) was shipped frozen to the Department of Earth Sciences at Uppsala University (Sweden). Due to a temporary equipment malfunction in Uppsala, only cations could be analysed there, and the refrozen samples were forwarded to ISP-CNR for anion analysis. All samples and standards were handled and prepared under clean room conditions, wearing powder-free gloves. In all labs except at the Polish Polar Station Hornsund, laminar flow hoods (class 100) were used. Samples were melted immediately before analysis.

143

2.2.1. Hornsund

145 Samples were filtered through 0.45 μm mixed cellulose esters membranes (Merck Millipore S-pak®)
146 prior to analysis. Ion concentrations were determined on a Metrohm 761 Compact IC ion chromatograph
147 equipped with an autosampler (Metrohm, Herisau, Switzerland), with isocratic flow, and chemical
148 suppression for anions (column Metrosep A Supp S + Metrosep A Supp 4/5 Guard 4.0, eluent: NaHCO_3
149 1.0 mmol L^{-1} + Na_2CO_3 3.2 mmol L^{-1}). Cations were determined without suppression (column Metrosep
150 C4 + Metrosep C4 Guard; eluent: HNO_3 1.7 mmol L^{-1} + 2,6-pyridinecarboxylic acid [dipicolinic acid, or
151 DPA] 0.7 mmol L^{-1}). Cation samples were acidified with 2 μL of 2mM HNO_3 per 10 mL sample prior to
152 analysis, as recommended for this device and column. The injection volume was 20 μL in the anion
153 system and 100 μL in the cation system. Nitric acid solutions were prepared from POCH S.A. (Poland)
154 concentrated weighed amounts, while sodium carbonate and hydrogen carbonate, as well as DPA, were
155 dissolved from solid phase (Merck Millipore).

156

157 2.2.2 Uppsala

158 Samples were filtered using 0.22 μm polyethersulfone membranes (Minisart®, Sartorius) and anion
159 determinations were performed using a Metrohm ProflC850 ion chromatograph (Metrohm, Herisau,
160 Switzerland), equipped with an autosampler and a Metrosep C4 column. The mobile phase of 0.02 M
161 DPA and 0.1 M HNO_3 was run in isocratic flow of 0.7 mL min^{-1} . Very low detection limits ($\leq 0.006 \text{ mg L}^{-1}$)
162 were achieved thanks to the sample injection volume of 500 μL .

163

164 2.2.3. Venice

165 Anion determination was performed using a Dionex™ ICS-5000 ion chromatograph
166 (ThermoScientific™, Waltham, US) equipped with an anionic exchange column (Dionex IonPac AS 11,
167 $2 \times 250 \text{ mm}$) and a guard column (Dionex IonPac AG11 $2 \times 50 \text{ mm}$). Sodium hydroxide (NaOH), used
168 as a mobile phase, was produced by an eluent generator (Dionex ICS 5000EG, Thermo Scientific). The
169 injection volume was 100 μL . The IC was coupled to a single quadrupole mass spectrometer (MSQ
170 Plus™, Thermo Scientific™) with an electrospray source (ESI) that operated in negative mode. To
171 determine cations, a capillary ion chromatograph (Thermo Scientific Dionex ICS-5000), equipped with a
172 capillary cation exchange column (Dionex IonPac CS19-4 μm , $0.4 \times 250 \text{ mm}$) and a guard column
173 (Dionex IonPac CG19-4 μm , $0.4 \times 50 \text{ mm}$), was used, coupled to a conductivity detector. The injection
174 volume was 0.4 μL . All details about the anion and cation methods are reported by (Barbaro et al., 2017).

175

176 2.2.4. Instrumental performance of each laboratory

177 For all laboratories, calibration for ions were evaluated using analytical standards (Merck/Sigma
178 Aldrich). The calibrations in each lab gave different linear ranges for each ion due to the different
179 methods used (Table S1). Good linearity was demonstrated in each lab and all calibration curves had
180 $R^2 > 0.99$. Samples that had ion concentrations beyond the calibration range were diluted with ultrapure
181 water before re-analysis. Analytical blanks of ultrapure water ($>18 \text{ M}\Omega\cdot\text{cm}$) were included in the

analysis at all three labs. The method detection limit (MDL) was set to three times the standard deviation of the blank values (Table S1). For Na^+ , Mg^{2+} , Cl^- and SO_4^{2-} , values $<\text{MDL}$ occurred in less than 10% of cases, and for Ca^{2+} and NO_3^- the $<\text{MDL}$ concentrations were noted in 12% and 17% of cases, respectively. However, K^+ and Br^- were detected only in 53% and 46% of all samples, respectively, while NH_4^+ concentration exceeded the MDL only in 36% of all measurements. For the calculation of bulk ionic loading in snow pits, values $<\text{MDL}$ were assumed to be equal to half the MDL.

Accuracy and precision are important parameters to evaluate during method validation. Checks for accuracy were made using certified multi-element standard solutions for anions (F^- , Cl^- , Br^- , NO_3^- , SO_4^{2-} , n° 89886-50ML-F, Sigma Aldrich) and cations (Na^+ , K^+ , Mg^{2+} , Ca^{2+} , n° 89316-50ML-F, Sigma Aldrich), at the concentration of $10 \text{ mg L}^{-1} \pm 0.2\%$. Accuracy is expressed as a relative error calculated as $(Q-T)/T \times 100$, where Q is the determined value and T is the “true” value. The accuracy for each ion in all labs was always $<\pm 10\%$, except for Mg^{2+} measurements at the Hornsund laboratory. The analytical precision was quantified as the relative standard deviation (RSD) for replicates ($n>3$) of standard solutions and was always $<10\%$ for each ion (Table S1).

2.3. Stable water isotopes

The determination of stable isotope ratios of O and H was performed at Tallinn University of Technology using a Picarro L2120-i water isotope analyser with a high-precision AO211 vaporizer. Results are reported in the standard delta notation as $\delta^{18}\text{O}$ and $\delta^2\text{H}$ relative to VSMOW. Reproducibility was $\pm 0.1\%$ for $\delta^{18}\text{O}$ and $\pm 1\%$ for $\delta^2\text{H}$, respectively.

204 3. Results

205 3.1 Spatial distribution of ionic species

206 To investigate differences in snowpack composition across all glaciers, we compared the total mass of
 207 ions that accumulated in snow at the different sampling sites. On average, the snow cover season on
 208 Svalbard lasts from early September to early May, but snow may also fall in summer months at high
 209 elevations. The snow pits in this study were sampled in early to late April 2016 and might therefore not
 210 contain the full annual ionic burden, since deposition might occur also in other months. Therefore, we
 211 report these data as ionic loads (mg m^{-2}) rather than annual fluxes. In each snow pit, the ionic load was
 212 calculated as the cumulative sum of the ionic concentrations multiplied by the snow water equivalent in
 213 each discrete layer. The snowpack chemical characteristics were then compared between glacier zones
 214 (ablation zone, ELA, and accumulation zone; Table 2, Figure 1).

215

216 Snow pits samples collected in the Hornsund area (southern Spitsbergen) showed a markedly higher total
 217 load for all major ions (Figures 1 and 2) than at all other sites. The samples collected in the accumulation
 218 zones of WB and HB had total ionic loads of 8161 and 8023 mg m^{-2} , respectively, four times higher than
 219 those collected in the same zone at KVG (2861 mg m^{-2}), AF (2607 mg m^{-2}) and ALB (1934 mg m^{-2}) and
 220 16 times higher than those sampled at LF (639 mg m^{-2}) and HDF (583 mg m^{-2}). Similar differences were
 221 observed for the snow pits collected at lower altitudes (Figure 2).

222

223 In the accumulation zone of all glaciers (Figure S1), Na^+ and Cl^- were generally the most abundant ionic
 224 species, with percentages ranging from 29% (HDF) to 36% (AF) for Na^+ , and from 34% (LF) to 48%
 225 (HB and WB) for Cl^- , respectively. The snowpack on Hornsund glaciers (HB, WB) had higher Cl^-
 226 percentages (48-49%) compared to that of other glaciers (34-39%), while conversely the SO_4^{2-} percentage
 227 was lower there (9%) than on other glaciers (11-23%). The ionic loads were generally highest in the
 228 accumulation zone of glaciers, and lowest in the ablation zone (Figure 2), mostly due to the lower snow
 229 accumulation and greater wind deflation at lower elevations. This pattern held true for Na^+ , Cl^- , NH_4^+ ,
 230 K^+ , Ca^{2+} , and Mg^{2+} at most glacier sites, except in the Hornsund region. The load of Br^- was similar on
 231 glaciers of the Ny-Ålesund sector (ALB, HDF, KVG) and on LF, but was higher in AF and Hornsund
 232 glaciers (HB, WB; Figure 2). The load of NO_3^- was similar for all glaciers, except for LF, where very low
 233 loads were found. Unlike total SO_4^{2-} , the non-sea-salt fraction of sulphate (nss-SO_4^{2-}), calculated using a
 234 seawater $\text{SO}_4^{2-}:\text{Na}^+$ mass ratio of 0.252, (Millero et al., 2008) showed lower loads on Hornsund glaciers
 235 (15-107 mg m^{-2}) when compared to glaciers in other parts of the archipelago (Figure 1, Table 2). The
 236 nss-SO_4^{2-} loads varied between 22-131 mg m^{-2} at HDF and LF, 75-266 mg m^{-2} at KVG and ALB, and
 237 153-206 mg m^{-2} at AF.

238

239 3.2 Stable water isotopes ($\delta^{18}\text{O}$ and $\delta^2\text{H}$)

Our results provide the first picture of spatial variations in the mean stable water isotope composition of the seasonal snowpack across Svalbard (Table 2, Figure S2). The snow water equivalent (SWE)-weighted mean $\delta^{18}\text{O}$ and $\delta^2\text{H}$ decreased significantly from south to north (Spearman rank correlation p with latitude was -0.69 and -0.65 for $\delta^{18}\text{O}$ and $\delta^2\text{H}$, with $p < 0.001$ and $p < 0.01$, respectively). The isotopically heaviest snow (least negative δ values) occurred on glaciers of the Hornsund region ($\delta^{18}\text{O}$: -11.25 to -9.54 ‰; $\delta^2\text{H}$: -77.62 to -63.64 ‰), and the isotopically lightest (most negative δ values) in AF ($\delta^{18}\text{O}$: -16.00 to -13.89 ‰; $\delta^2\text{H}$: -111.15 to -96.89 ‰). Glacier sites in NW Spitsbergen and on LF had mean $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values that fell within these ranges. On KVG, ALB, HDF and LF, the $\delta^{18}\text{O}$ and $\delta^2\text{H}$ in snow decreased monotonically (becoming gradually more negative) with increasing elevation. However, on other glaciers this pattern did not hold: on AF, we found an increase from AF1 to AF2, but a decrease from AF2 to AF3; on WB the $\delta^{18}\text{O}$ and $\delta^2\text{H}$ were similar at WB1 and WB2, but less negative at WB3. On HB there was no statistical difference between the mean $\delta^{18}\text{O}$ and $\delta^2\text{H}$ value in all snow pits. A general, significant anticorrelation with altitude was found for SWE-weighted mean $\delta^2\text{H}$ ($\rho = -0.63$, $p < 0.01$), and $\delta^{18}\text{O}$ ($\rho = -0.65$, $p < 0.01$).

254

255 4. Discussion

256

There have been few published studies on recent seasonal snow or firn chemistry in Svalbard, hence comparisons of our data with these earlier results are limited to a few sites. (Virkkunen et al., 2007) and (Vega et al., 2015a) and unpublished data) quantified the annual chemical loads of Na^+ , Ca^{2+} , NO_3^- and nss-SO_4^{2-} at Lomonosovfonna summit (LF3) from 2002 to 2011 using snow and firn cores, and our study extends these data to 2016. The range of annual ionic loads at LF3 over the 15-year period is remarkably wide, but no clear temporal trend can be identified (Table 3). At Holtedahlfonna summit (HDF3), firn core measurements by (Spolaor et al., 2013) found a mean Na^+ concentration of $110 \pm 73 \text{ ng g}^{-1}$ over the period 2003-2012, while the mean concentration in the April 2016 snowpack (this study) was 191 ng g^{-1} , hence within the range reported in earlier years.

266

267 4.1 The main ion sources in the Svalbard seasonal snow

The composition of the Svalbard seasonal snowpack sampled during the C2S3 project clearly indicates that the ocean is the main source of ions in snow, as was shown by Hodgkins and Tranter (2017). At all sites, the dominant ions are Na^+ , Cl^- , and SO_4^{2-} , with comparatively minor amounts of K^+ , Ca^{2+} and Mg^{2+} (Figure S1). To help clarify the possible sources and modes of deposition of ions in snow, we computed Spearman rank correlations between total ionic loads (ρ_{load}), as well as between volume-weighted mean ionic concentrations (ρ_{conc}), across all snow pits ($n = 22$; Table 4). The chemical species that are predominantly wet-deposited and sharing common sources and not undergoing significant composition changes in transport should exhibit similar concentration patterns (high ρ_{conc}) (Schüpbach et al., 2018). The concentrations of Mg^{2+} , K^+ and Ca^{2+} were all positively correlated with those of Na^+ and Cl^- ,

277 indicating a common sea spray source. The ρ_{load} correlations are very similar for these ionic species,
 278 which points to both wet and dry deposition being a significant mechanism in their accumulation in
 279 snowpack.

280

281 The concentrations of Mg^{2+} were positively and significantly correlated with both Ca^{2+} and nss-Ca^{2+} (ρ_{conc}
 282 = 0.70 and 0.47, respectively; the latter coefficient was higher for loads at 0.56; Table 4), suggesting they
 283 share some non-marine source(s). Furthermore, all glaciers had greater $\text{Ca}^{2+}:\text{Mg}^{2+}$ ratios than seawater
 284 (0.32; (Millero et al., 2008) (Figure 3). It is likely that the excess Ca^{2+} and Mg^{2+} came from mineral
 285 particles, i.e. CaCO_3 (calcite), and local rock (or soil) dust (Kekonen et al., 2005) derived from calcareous
 286 rocks, especially limestone, dolostone and marble, which are abundant in (Dallmann, 1999) The presence
 287 of carbonate ions in the collected snow samples would explain the missing negative charge in the ionic
 288 balance (anion X^- ; Figure S3).

289

290 Sulphate (SO_4^{2-}) was highly and significantly ($p\text{-value} < 0.05$) correlated with both Na^+ ($\rho_{\text{load}} = 0.92$; ρ_{conc}
 291 = 0.80) and Cl^- ($\rho_{\text{load}} = 0.93$; $\rho_{\text{conc}} = 0.75$), indicating that sea spray is its main source. However $\text{Na}^+/\text{SO}_4^{2-}$
 292 and $\text{Cl}^-:\text{SO}_4^{2-}$ ratios were well below seawater values (Millero et al., 2008) on most glaciers except those
 293 near Hornsund (WB and HB), suggesting inputs of nss-SO_4^{2-} (Figure 3). Biogenic nss-SO_4^{2-} can be
 294 deposited in the snow as an oxidized by-product of dimethyl sulphide (DMS) emitted by marine algal
 295 blooms (Gondwe et al., 2003), typically initiated in April but sometimes later (Ardyna et al., 2013).
 296 Another plausible source of nss-SO_4^{2-} deposition in Svalbard is long-range transport of SO_4^{2-} aerosols
 297 from biomass burning in the spring, or from fossil fuel combustion throughout the winter (Barrie, 1986;
 298 Law and Stohl, 2007; Nawrot et al., 2016). The nss-SO_4^{2-} did not correlate significantly with other ionic
 299 species, suggesting a separate origin. An extra consideration is that in the southern region of the
 300 archipelago, the higher sea spray input could partially mask the nss-SO_4^{2-} signal, which is a derived
 301 variable, because the larger uncertainty for greater Na^+ concentrations would disproportionately affect
 302 estimations of nss-SO_4^{2-} at these sites.

303

304 Bulk ionic loads of SO_4^{2-} in the snow pits were significantly and positively correlated with those of NO_3^-
 305 ($\rho_{\text{load}} = 0.55$) and NH_4^+ ($\rho_{\text{load}} = 0.68$), but the correlations between weighted mean ionic concentrations
 306 were not significant, hinting at co-deposition (wet) rather than shared sources (Table 4). These species
 307 are known to form secondary aerosols (Karl et al., 2019; Schaap et al., 2004) and thus their proportions
 308 in aerosols may differ significantly from those in their source emissions. It is also possible that nitrogen
 309 species underwent further post-depositional photochemical reduction and evasion, thereby reducing their
 310 concentrations in snow (Curtis et al., 2018). Finally, we remark here that the snow pit sampling was done
 311 in April, earlier than the beginning of the oceanic algal bloom in the surrounding Svalbard basin, which
 312 could have led to underrepresentation of biological emissions from late spring in our samples.

313

Spatial variations of ammonium loads (NH_4^+) across Svalbard glaciers mirrored the pattern shown by sea salt ions, with higher loads in the Hornsund region and lower loads in other areas. This was also reflected by significant correlations between bulk loads of NH_4^+ with those of Na^+ and Cl^- ($\rho_{\text{load}} = 0.64$ and 0.73 , respectively), and with Na^+ , K^+ and Mg^{2+} by concentration ($\rho_{\text{conc}} = 0.47$, 0.62 and 0.47 , respectively), the latter relationships suggesting that some ammonium is deposited as coatings on crustal aerosols (Eastwood et al., 2009). Ammonium has been linked to biogenic, forest fire, and anthropogenic agricultural emissions (Trachsel et al., 2019). The higher annual snowpack load of NH_4^+ , determined in the Hornsund region is more likely connected with biological sources than anthropogenic activities, although some contribution from biomass burning events cannot be excluded. The marine primary productivity in spring 2016 (April and May) was higher in the south-eastern ocean sector of the Svalbard archipelago (Figure S4), which could partially explain the higher NH_4^+ load. This would also explain the correlation between ammonium and sea-salt ions (Table 4). Locally, especially for HB, there may be extra NH_4^+ emissions from bird colonies (Keslinka et al., 2019; Wojczulanis K. et al., 2008).

Unlike NH_4^+ , the bulk loading of NO_3^- in snow was highest in northwestern Spitsbergen (Ny-Ålesund area), when compared to other parts of Svalbard. Deposition of NO_3^- in Arctic snow is often associated with the long-range atmospheric transport of NO_x and related N species from anthropogenic source regions at lower latitudes (Björkman et al., 2014; Fibiger et al., 2016; Vega et al., 2015a). Differences in NO_3^- loads in snow in various parts of Svalbard might therefore reflect differences in the transport pathways of precipitating air masses, including formation of secondary aerosols, or post-depositional processes, rather than local emissions. While local shipping routes and the settlement of Ny Ålesund itself may contribute NO_3^- emissions (Winther et al., 2014), the highest share of the total ionic load of NO_3^- was found in the accumulation zone of HDF (9% of the total ionic load; Figure S1). Given that HDF is the most remote site from Ny Ålesund relative to KVG or ALB, it should not capture a high share of local pollution. The highest correlation coefficient for NO_3^- , both in terms of concentrations and loads, was found with nss-Ca^{2+} . This would support both the formation of calcium nitrate in the atmosphere (Gibson et al., 2006) or post-depositional processes removing the NO_3^- from layers poor in Ca^{2+} , since calcium has been hypothesised to stabilise the nitrate in snowpack against post-depositional losses (Kekonen et al., 2017).

4.2. Chlorine depletion

Although Na^+ and Cl^- , the main species of sea salt, were significantly correlated ($\rho_{\text{conc}} = 0.95$), the values of the Cl^-/Na^+ ratio in snow were lower than that in seawater on most studied glaciers, except those near Hornsund (Figure 3), suggesting a Cl^- deficit at the more northerly sites. A possible explanation of this Cl^- deficit might be de-chlorination of the sea spray aerosol during transport or, less likely, at the snow-atmosphere interface. This reaction occurs between sea salt particles, containing NaCl , and NO_3^- , SO_4^{2-} , or organic acids to release gaseous HCl (Zhuang et al., 1999). We calculated the percentage of Cl^-

depletion (Cl^-_{dep}) as $\text{Cl}^-_{\text{dep}} = (\text{Cl}^-_{\text{ss}} - \text{Cl}^-_{\text{meas}}) / \text{Cl}^-_{\text{ss}} \times 100\%$, where $\text{Cl}^-_{\text{ss}} = 1.174 \text{ Na}^+_{\text{meas}}$, and $\text{Cl}^-_{\text{meas}}$ and $\text{Na}^+_{\text{meas}}$ are the measured equivalent concentrations (Yao et al., 2003). Except for site HDF2 ($\text{Cl}^-_{\text{dep}} = 2\%$), the lowest mean Cl^-_{dep} values were obtained for Hornsund glaciers (WB, HB: 10–19%), while values at other glacier sites ranged between 21 and 75% (Table 2). This suggests that sea-salt aerosols travel along a route from southern to northern Svalbard, which gives more time for Cl^- depletion in the ionic mixtures reaching more northerly locations.

4.3. Bromine enrichment

In addition to Cl^- , snowfall can scavenge Br^- (Peterson et al., 2019; Spolaor et al., 2019), and Br^- loads on Svalbard glaciers surveyed in April 2016 were positively and significantly correlated with those of primary sea salt ions Na^+ ($\rho_{\text{load}} = 0.48$), Cl^- ($\rho_{\text{load}} = 0.53$) and Mg^{2+} ($\rho_{\text{load}} = 0.51$) (Table 4). Correlations between weighted mean concentrations were not significant, however, suggesting departures of the Br^- concentrations in snow from typical seawater ionic ratios at some glacier sites. A Br^- enrichment factor (Br_{enr}) can be calculated as $\text{Br}_{\text{enr}} = \text{Br}^- / (0.0065 \text{ Na}^+)$, where 0.0065 is the $\text{Br}^- : \text{Na}^+$ seawater mass ratio (Maffezzoli et al., 2017). The Br_{enr} reflects specific processes (in particular sea ice Br emission) that affect the Br^- concentration and load in the snowpack (Spolaor et al., 2014). Results of our calculations (Table 2, Figure S5) show that on glaciers of the Hornsund area (HB and WB) and NW Spitsbergen (KVG, ALB and HDF), the mean Br_{enr} values are often, but not always, < 1 , indicating some Br^- depletion, in agreement with the findings of (Jacobi et al., 2019) for glaciers in the Ny Ålesund area. The depletion could be a result of snowpack Br re-emission, but this seems unlikely since field measurements near Ny-Ålesund found no evidence of such volatilization of snow-bound Br (Spolaor et al., 2019).

Alternatively, Br^- depletion could occur through BrO loss from marine aerosols and subsequent deposition of these Br -depleted aerosols in snow. In contrast to southern and northwestern Spitsbergen, glaciers in central Spitsbergen (LF) and in Nordaustlandet (AF), showed Br_{enr} values > 1 . These glaciers lie relatively close to areas to the east of the archipelago that are often covered by first-year sea ice. Newly-formed sea ice has been shown to release gas phase Br into the polar atmosphere, thus supplying an extra Br source in addition to sea spray (Spolaor et al., 2016). The spatial distribution of the Br -enriched snow pit sites supports this : sites closest to areas covered by first-year sea ice have the largest Br enrichments, and the latter decrease with greater distance from the eastern shores of Svalbard (Figure S5). A survey of the average sea ice coverage in the period March-May 2016, which is relevant to the Br enrichment phenomenon (data by Norwegian Meteorological Institute) confirms that the north-eastern and eastern shore of Svalbard were indeed covered much more frequently by close and open drift ice than the south or north-west.

4.4 Distribution pattern of $\delta^{18}\text{O}$ and $\delta^2\text{H}$

387 As described earlier, the SWE-weighted mean $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values in glacier snow pits decreased
 388 significantly with increasing latitude across Svalbard, the least negative values occurring on glaciers of
 389 the Hornsund region, and the most negative in Austfonna (Table 2). This pattern follows the climate
 390 gradient across the archipelago, milder in the south, colder in the north. Part of the south-north contrast in
 391 δ values could be explained by the lower mean altitude of glacier sites in the Hornsund region compared
 392 to some of the higher-elevation sites further north on Spitsbergen or on Austfonna, and the relationship
 393 with elevation was similar for both isotopic ratios in the collected dataset.

394 The deuterium excess ($d = \delta^2\text{H} - (8 \cdot \delta^{18}\text{O})$) is mainly influenced by the source region of the precipitating
 395 moisture and in particular by the sea surface temperature. In addition, d is also influenced by the
 396 temperature gradient between the moisture source and precipitation area. The SWE-weighted mean d
 397 values in Svalbard snow pits varied within a relatively narrow range of 6.74‰ (from 10.10 to 16.84 ‰),
 398 and similarly to $\delta^{18}\text{O}$, showed no clear gradient with elevation or longitude. Deuterium excess showed a
 399 significant correlation with latitude, at $p = 0.60$ ($p < 0.01$). A more detailed analysis of d by latitude
 400 showed that only beyond 79.2 °N, i.e. in Austfonna snow pits, was d significantly different than at other
 401 sites (Kruskal-Wallis test, $z = 4.23$, $p < 0.04$). This is consistent with lower temperatures and evaporation
 402 rates in the more northern waters around Svalbard, and suggests that snowfall on AF is at least partly
 403 affected by a different, more northerly moisture source than the rest of the archipelago.

404

405 **4.5 Effect of elevation: a case study of Na**

406 The glacier survey carried out during the C2S3 project afforded the opportunity to investigate the
 407 possible effect of elevation on the ionic composition of the snowpack. To do this, we compared the bulk
 408 load and SWE-weighted mean concentration of Na^+ across all studied snow pits, ordered by elevation
 409 (Figure 4). Overall, both Na^+ loads and concentrations decreased with increasing altitude ($\rho_{\text{load}} = -0.24$,
 410 $p > 0.05$; $\rho_{\text{conc}} = -0.72$, $p < 0.05$). This likely reflects greater local sea spray aerosol deposition at lower,
 411 compared to higher, glacier sites. We then computed linear (Pearson) correlation coefficients (R , with
 412 associated p -values) between log-transformed Na^+ loadings ($\log(\text{Na}_{\text{load}})$) and $\delta^{18}\text{O}$ for all snow pits in the
 413 accumulation zones of glaciers (Figure 5). The calculation was performed with all snow layers. The Na^+
 414 load was used as sea-spray tracer, while the $\delta^{18}\text{O}$ was assumed to vary with moisture source between
 415 discrete snowfall events. We found that the positive correlation between $\log(\text{Na}_{\text{load}})$ and $\delta^{18}\text{O}$ increased
 416 with elevation from $R = 0.1$ (HB3; 396 m a.s.l.) to $R = 0.65$ (LF3; 1193 m a.s.l.), and reached a 95 %
 417 threshold of significance ($R > 0.3$) for glaciers sites above 600 m a.s.l. (KVG, AF, LF and HDF; Figure
 418 5). The average distance was a comparatively negligible factor in explaining the correlation between
 419 $\log(\text{Na}_{\text{load}})$ and $\delta^{18}\text{O}$.

420

421 The increase in strength and significance of the $\log(\text{Na}_{\text{load}})$ - $\delta^{18}\text{O}$ correlation with altitude might be
 422 explained by different contributions of locally-emitted ssNa^+ , relative to those of Na^+ from more distant
 423 sources. Sites located at lower altitudes are proportionally more affected by local sea spray deposition,

with or without snowfall. Conversely, sites at higher elevations likely receive a larger share of their ionic load from more distant sources, and by wet deposition through snowfall. At the four sites (KVG, AF, LF and HDF) where the $\log(\text{Na}_{\text{load}})$ - $\delta^{18}\text{O}$ correlation was significant, increases in $\delta^{18}\text{O}$ in snow layers were often associated with higher Na^+ concentrations. The isotopically heavier (less negative) $\delta^{18}\text{O}$ values suggests that the co-registered Na^+ enhancements were associated with precipitation of relatively warm air, probably advected from lower latitudes. Air masses arriving from the south travel across the ocean for an extended time, which can enrich them in sea spray aerosol and hence in Na^+ . It is also possible that the poorer $\log(\text{Na}_{\text{load}})$ - $\delta^{18}\text{O}$ correlation at lower altitude glacier sites is partly due to stronger post-depositional modification of isotopic and ionic signals in snowpack related to more frequent melt-refreeze episodes.

434

435 5. Summary and Conclusion

We have quantified and described, for the first time, the spatial distribution of major ion loads (Ca^{2+} , K^+ , Na^{2+} , Mg^{2+} , NH_4^+ , SO_4^{2-} , Br^- , Cl^- and NO_3^-) and variations of $\delta^{18}\text{O}$ and $\delta^2\text{H}$ in the snowpack on glaciers across Svalbard for a single accumulation season (2015-2016). The highest total ionic loads were found in the southern region of Spitsbergen (Hornsund area), and exceeded 8 g m^{-2} . Conversely, the lowest total ionic loads ($\leq 0.6 \text{ g m}^{-2}$) were found at sites in central or northwestern Spitsbergen (LF and HDF). Sea salt ions (Cl^- , Na^+ and SO_4^{2-}) dominated the ionic loads at all sites, but their share was highest at sites near Hornsund, for e.g., 48% Cl^- , compared to only 29% on Holtedahlfonna. Relatively elevated $\text{Ca}^{2+}/\text{Mg}^{2+}$ ratios in snow at all sites indicated non-sea-salt Ca^{2+} inputs, most likely in the form of carbonate dust. Unlike other ions, NO_3^- had the highest loads in glaciers of northwestern Spitsbergen, and the lowest at LF. The nitrogen species, NO_3^- and NH_4^+ , showed distinct spatial distribution patterns. The highest NO_3^- loads were found in the northwestern part of Svalbard, while the highest NH_4^+ loads were in the southwest. Bromide (Br^-) was most enriched in snow relative to seawater at AF and LF, the glacier sites located closest to areas with first-year sea ice cover. This supports first-year sea ice being an important source of non-sea salt Br^- in the polar atmosphere.

450

An increasing, positive correlation between $\log(\text{Na}_{\text{load}})$ and $\delta^{18}\text{O}$ as a function of elevation sites suggests that locations above 600-700 m a.s.l. are influenced by a proportionally higher share of ions from distant sources, while the lower sites are exposed to more local sources, especially sea spray. These findings confirm that the optimal sites to study the effects of long-range pollution deposition in Svalbard are those found at higher elevations sites, such as the accumulation zones of HDF or LF, because they are the least impacted by the local aerosol emissions. The current study gives the first picture of the ionic composition in the Svalbard snowpack in different regions across the archipelago, in the context of which processes are relevant in controlling the annual snowpack chemical composition there, especially the influence of local and long-range transport.

460

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652 TABLES

653 **Table 1.** Table 1. laciers and sampling sites included in this study with their main characteristic. AWS:
 654 atmospheric weather station; UiO: University of Oslo; ThèMA: Théoriser & Modéliser pour Aménager,
 655 University of Franche-Comté; NPI: Norwegian Polar Institute; IMAU: Institute for Marine and Atmospheric
 656 Research, Utrecht University; UoS: University of Silesia, IG PAS – Institute of Geophysics, Polish
 657 Academy of Sciences; CNR – Consiglio Nazionale delle Ricerche. . The seven glaciers were considered at
 658 three different altitudes: 1) lower ablation zone; 2) ELA; 3) upper accumulation zone. Exceptionally, two
 659 snow pits (KVG 1 and KVG 1.5) were dug in the ablation zone of Kongsvegen glacier.

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661 **Table 2.** Total load (mg m^{-2}) of major ions, calculated as the sum of loads in all layers of each snow pit. Sea
 662 salt sulphate (ss-SO_4^{2-}) and non-sea-salt sulphate (nss-SO_4^{2-}) are expressed as mg m^{-2} , while chloride
 663 depletion (Cl_{dep}^+) is given as a percentage and bromide enrichment (Br_{enr}) refers to an enrichment compare to
 664 sea water. Average SWE-weighted stable water isotope ratios ($\delta^{18}\text{O}$ and $\delta^2\text{H}$) and average deuterium excess
 665 (d) are also reported.

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667 **Table 3.** Chemical loads (mg m^{-2}) of selected elements from the 2016 sampling and earlier studies
 668 at Lomonosovfonna summit (LF3), corresponding to the concentrations given in Fig. 4.

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670 **Table 4.** Spearman rank order correlations for all 7 glaciers ($n=22$ locations) with a $p < 0.05$: a) of total loads
 671 (mg m^{-2}) of major ions (calculated as the sum of loads in all layers of each snow pit), b) of volume-weighted
 672 average concentrations of each snow pit, calculated by dividing the total load of each snow pit by its total
 673 water equivalent. Non-sea-salt (nss) components were calculated based on the seawater ratios to Na^+ (0.038
 674 and 0.252, respectively, for Ca^{2+} and SO_4^{2-} in mg L^{-1} ; Millero et al. 2008); ns = not significant with p -value
 675 > 0.05 .

FIGURES

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678 **Figure 1.** Total snowpack loads (mg m^{-2}) of major ions in 22 snow pits collected on glaciers during the C2S3
 679 project. Seven glaciers were sampled in three snow pits in the lower ablation zone (1), near the equilibrium
 680 line (2) and in the upper accumulation zone (3), except on Kongsvegen glacier (KVG) where there was an
 681 extra snow pit sampled within the ablation zone. Glacier site abbreviations are given in Table 1.

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Figure 2. Calculated ionic loads in the snowpack (mg m^{-2}) at the 7 glacier sites sampled during the C2S3 project. Snow pits for each glacier are marked with the same colour and ordered from lower (left) to higher altitudes (right). For the KGV another snow pit was dug between glaciers zone 1 and 2.

Figure 3. Panels from top: 1) Cl^-/Na^+ ; 2) $\text{Na}^+/\text{SO}_4^{2-}$; 3) $\text{Cl}^-/\text{SO}_4^{2-}$; 4) the total loads of sea-salt sulphate (ss- SO_4^{2-}) and non-sea-salt sulphate (nss- SO_4^{2-}), and 5) $\text{Ca}^{2+}/\text{Mg}^{2+}$ - for all glaciers investigated during the C2S3 project (in the spring 2016).

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710 **Figure 4.** Sodium load in snow pits ordered by increasing elevation in m a.s.l., indicated by the red lines.

711 The colours identify areas where the snow pits have been excavated: each colour represents a separate glacier

712 (HB – blue; WB – purple; ALB – red; LF – green; KVG – pink; AF – light blue; HDF – orange). IQR =

713 inter-quartile range, i.e. the difference between the value of quartiles 3 and 1.

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726 **Figure 5.** The correlation coefficient between oxygen isotope ratio ($\delta^{18}\text{O}$) and $\log[\text{Na}_{\text{load}}]$ increases with
 727 elevation. The left axis represents the correlation coefficient (R) between $\log[\text{Na}_{\text{load}}]$ and $\delta^{18}\text{O}$, using the
 728 entire dataset for each snow pit (i.e. all layers have been used for the statistical correlation). The x axis
 729 indicates the altitude of the snow pit. The upper panel shows the p-value: correlations have been considered
 730 statistically significant if $p < 0.05$.

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732

Table 1. Glaciers and sampling sites included in this study with their main characteristics. AWS: automatic weather station; UiO: University of Oslo; Théma: Thémiser & Modéliser pour Aménager, Université de Franche-Comté; NPI: Norwegian Polar Institute; IMAU: Institute for Marine and Atmospheric Research, Utrecht University; UoS: University of Silesia, IG PAS – Institute of Geophysics, Polish Academy of Sciences; CNR – Consiglio Nazionale delle Ricerche.

AWS									
Glacier	Site	Zone	Lat. (°N)	Lon. (°E)	Elev. (m)	Date (dd.mm.yyyy)	Air Temp. (°C)	Snow height (cm)	Snow Water Equivalent (SWE) (mm)
Austfonna	AF1	ablation	79.734	22.414	336	21.04.2016	-13.5	106	330.50
	AF2	equilibrium line	79.767	22.825	507	23.04.2016	-7.1	135	439.59
	AF3	accumulation	79.832	24.004	785	24.04.2016	-14.7	181	803.93
Austre Lovénbreen	ALB1	ablation	78.883	12.136	195	25.04.2016	-3.7	81	296.66
	ALB2	equilibrium line	78.889	12.159	340	25.04.2016	-2.8	90	353.17
	ALB3	accumulation	78.861	12.187	513	20.04.2016	-11.3	161	499.67
Kongsvegen	KVG1	ablation	78.830	12.759	226	13.04.2016	-13.9	20	51.29
	KVG1.5	ablation	78.813	12.869	326	13.04.2016	-13.9	75	261.94
	KVG 2	equilibrium line	78.780	13.153	534	11.04.2016	-17.5	162	575.78
	KVG3	accumulation	78.756	13.336	672	12.04.2013	-15.5	234	880.13
Holtedahlfonna	HDF1	ablation	78.931	13.303	570	17.04.2016	-14.5	108	372.98
	HDF2	equilibrium line	79.029	13.531	718	17.04.2016	-14.2	175	625.00
	HDF3	accumulation	79.140	13.394	1119	15.04.2016	-18.1	201	732.08
Lomonosovfonna	LF1	ablation	78.633	17.077	223	10.04.2016	-10.9	27	99.4
	LF2	equilibrium line	78.691	17.150	523	9.04.2016	-5.8	94	277.28
	LF3	accumulation	78.824	17.435	1193	11.04.2016	-24	146	487.01
Hansbreen	HB1	ablation	77.049	15.639	102	25.04.2016	-7.3	102	396.10
	HB2	equilibrium line	77.083	15.639	275	25.04.2016	-6.9	169	640.28
	HB3	accumulation	77.120	15.487	396	29.04.2016	0.7	288	1305.09
Werenskiöldbreen	WB1	ablation	77.075	15.313	166	16.04.2016	-9.2	81	328.34
	WB2	equilibrium line	77.072	15.441	413	16.04.2016	-11.2	110	454.75
	WB3	accumulation	77.092	15.489	528	18.04.2016	-11.1	330	1396.60

Table 2. Total load (mg m^{-2}) of major ions, calculated as the sum of loads in all layers of each snow pit. Sea salt sulphate (ss-SO_4^{2-}) and non-sea-salt sulphate (nss-SO_4^{2-}) are expressed as mg m^{-2} , while chloride depletion (Cl^-_{dep}) is given as a percentage and bromide enrichment (Br_{enr}) refers to an enrichment compare to sea water. Average SWE-weighted stable water isotope ratios ($\delta^{18}\text{O}$ and $\delta^2\text{H}$) and average deuterium excess (d) are also reported.

	Cl^-	Br^-	SO_4^{2-}	NO_3^-	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	ss-SO_4^{2-}	nss-SO_4^{2-}	$\text{Cl}^-_{\text{dep}} \%$	Br_{enr}	$\delta^{18}\text{O}$	$\delta^2\text{H}$	d
KVG1	190	0.4	96	6	421	0.3	7	37	20	106	nd	75	0.2	-9.69	-66.17	11.37
KVG1.5	281	1.9	157	45	327	0.9	6	36	33	82	75	52	1.0	-11.32	-78.25	12.34
KVG2	652	1.2	342	118	605	1.9	9	51	63	152	190	40	0.3	-12.51	-88.62	11.48
KVG3	1039	1.2	509	148	967	2.9	12	86	94	244	266	40	0.2	-12.72	-89.50	12.25
HDF1	373	1.3	144	74	267	2.6	4	23	33	68	79	21	0.9	-13.51	-94.37	13.75
HDF2	423	2.5	192	148	240	4.5	6	18	67	61	131	2	1.7	-13.91	-99.15	12.10
HDF3	227	0.8	65	51	170	4.5	9	12	45	43	22	25	0.8	-15.18	-104.51	16.97
ALB1	446	1.9	226	115	343	1.5	10	31	90	86	139	27	0.9	-11.22	-75.17	14.59
ALB2	294	1.4	158	87	221	1.6	4	17	38	56	107	25	1.2	-12.19	-83.11	14.40
ALB3	729	1.8	342	64	648	4.2	8	56	81	158	165	36	0.4	-12.40	-85.40	13.79
LF1	75	1.1	59	1	95	0.4	3	12	6	31	48	51	2.1	-11.61	-82.79	10.10
LF2	174	2.8	127	18	141	2.6	5	12	7	53	127	38	3.2	-14.54	-105.44	10.90
LF3	216	3.5	144	10	225	2.9	7	22	9	56	93	45	2.7	-15.14	-110.42	10.69
AF1	498	5.7	348	38	578	2.2	27	48	95	127	173	53	1.6	-14.34	-100.76	13.94
AF2	438	6.3	263	15	509	3.4	19	62	26	147	153	51	2.0	-16.00	-111.15	16.84
AF3	928	6.5	439	88	933	8.1	35	89	81	185	206	54	1.5	-13.89	-96.89	14.24
WB1	2041	4.7	332	34	1278	6.7	53	131	91	340	15	10	0.6	-10.17	-70.62	10.75
WB2	1584	2.7	304	38	1051	9.9	44	110	68	220	24	16	0.4	-10.25	-70.14	11.90
WB3	3922	7.2	713	118	2649	26.1	137	277	313	671	37	17	0.5	-9.54	-63.64	12.66
HB1	2680	6.2	482	46	1722	10.5	74	201	110	475	47	13	0.6	-11.14	-75.93	13.19
HB2	2499	2.8	490	185	1667	32.5	105	159	150	350	73	19	0.3	-10.69	-73.34	12.17
HB3	3964	6.5	719	118	2557	28.2	125	281	223	751	107	13	0.4	-11.25	-77.62	12.35

Table 3. Loads (mg m^{-2}) of selected major ions from the 2016 sampling and from earlier studies at Lomonosovfonna summit (LF3), corresponding to the concentrations given in Fig. 4.

Year	Na^+	Ca^{2+}	NO_3^-	nss- SO_4^{2+}	Study
2002	126.7	7.1	27.3	37.1	(Virkkunen et al., 2007)
2009	n.a.	n.a.	33.5	n.a.	<i>Vega C. (unpublished data)</i>
2010	80.1	24.3	52.3	48.1	<i>Vega C. (unpublished data)</i>
2011	262.9	46.2	27.2	34.1	(Vega et al., 2015), <i>Vega C. (unpublished data)</i>
2016	222.2	7.2	11.4	93.0	<i>This study</i>

Table 4. Spearman rank order correlations of a) ionic loads (mg m^{-3}) and b) SWE-weighted mean concentrations of major ions across all 7 glaciers ($n=22$ locations). ns = non-significant correlations (p -value > 0.05). Ionic loads were calculated from all snow pit layers, while SWE-weighted mean concentrations were calculated by dividing the ionic loads in each snow pit by its total SWE. Non-sea-salt (nss) components were estimated based from seawater ratios to Na^+ (0.038 and 0.252, respectively, for Ca^{2+} and SO_4^{2-} in mg L^{-1} ; Millero et al. 2008).

	Cl ⁻	Br ⁻	SO ₄ ²⁻	NO ₃ ⁻	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	nss-SO ₄ ²⁻
a)										
Br ⁻	0.53									
SO ₄ ²⁻	0.93	0.60								
NO ₃ ⁻	0.55	ns	0.55							
Na ⁺	0.94	0.48	0.92	0.44						
NH ₄ ⁺	0.73	0.62	0.68	ns	0.64					
K ⁺	0.82	0.61	0.81	ns	0.85	0.75				
Mg ²⁺	0.90	0.51	0.88	ns	0.98	0.62	0.82			
Ca ²⁺	0.86	ns	0.83	0.69	0.82	0.61	0.76	0.71		
nss-SO ₄ ²⁻	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns
nss-Ca ²⁺	0.76	ns	0.75	0.77	0.68	0.56	0.66	0.56	0.96	ns
b)										
Br ⁻	ns									
SO ₄ ²⁻	0.75	0.58								
NO ₃ ⁻	ns	-0.48	ns							
Na ⁺	0.95	ns	0.80	ns						
NH ₄ ⁺	ns	ns	ns	ns	0.47					
K ⁺	0.83	0.46	0.73	ns	0.88	0.62				
Mg ²⁺	0.92	ns	0.78	ns	0.98	0.47	0.86			
Ca ²⁺	0.85	ns	0.64	0.44	0.76	ns	0.62	0.70		
nss-SO ₄ ²⁻	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns
nss-Ca ²⁺	0.67	ns	0.45	0.56	0.54	ns	ns	0.47	0.91	ns