

1 **Seasonal and interannual variability on the chemical composition of the**
2 **Svalbard surface snowpack**
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32 **Abstract**

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

50 **1. Introduction**

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

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

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

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

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

107 **2. Methodology**
108 *2.1 Sampling and processing*

109 Three sampling campaigns were conducted in Svalbard between 2018 and 2021, covering the period
110 from October to May according to the onset of the snowpack formation and melting. Sample
111 collection followed the protocol presented in Spolaor et al. (2019), further adopted by Bertò et al.
112 (2021), where consecutive and adjacent sampling was carried in a 3x3 meters snow sampling area,
113 within a clean sampling snowfield of 100 m wide. Each sample was collected 10 cm apart from the
114 previous one, along a precise path. This method was designed to minimise the temporal variability
115 between consecutive samples and reduce the impact of potential spatial variability (within the 5-15%
116 range, according to Spolaor et al., 2019).
117 During the first sampling campaign, carried out from October 4th, 2018 to May 10th, 2019, 114 surface
118 snow samples were collected in a delimited snow field located ~ 100 m south of the “Dirigibile Italia
119 Station” in Ny-Ålesund (78.92° N 11.93° E, Ny-Ålesund, Svalbard). The surface snow was sampled
120 within the upper 3 cm, as this is the snow layer most impacted by aerosol deposition and exchange
121 processes at the snow-atmosphere interface (Spolaor et al., 2018, 2021b). This choice also minimised
122 the effect of different physical snow conditions (density, crystal shape, and size).
123 Concurrently, additional 133 snow samples were collected at the Gruebadet Snow Research Site
124 (GSRS) to evaluate the spatial variability with respect to the snow samples collected in Ny-Ålesund.
125 The GSRS is a clean-area located about 1 km south of Ny-Ålesund, nearby the Gruebadet
126 Atmospheric Laboratory (GAL), dedicated to the chemical and physical monitoring of the seasonal
127 snowpack (Scoto et al., 2023; Fig. 1). Throughout the season, the sampling resolution varied based
128 on light conditions. During the polar night (from October to early March), snow sampling was carried
129 out daily at Ny-Ålesund, and every 3-5 days at the GSRS. With the beginning of the polar day, daily
130 sampling was conducted both in Ny-Ålesund and at the GSRS in March, and then continued only at
131 the GSRS until the end of the snow season in June due to the lower contamination of the site, more
132 distant from the fervent local activities. This sampling resolution overlap during March ensured a
133 good comparison of results in both snow fields (Fig. S1).

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140 Starting from the second campaign, snow sampling activities were conducted only at the GSRS site,
141 since clean conditions of the field in Ny-Ålesund could not be guaranteed due to construction works.
142 The snow sampling was carried out from October 26th, 2019 to May 25th, 2020, with a total of 107
143 samples collected. The surface snow layer was sampled every 3-5 days during the polar night (until
144 February 24th, 2020), and daily from the beginning of the polar day until the end of the snow season.
145 Consecutive samples represent the same snow layer in the absence of snowfall or wind drift/erosion.
146 However, factors such as snow aging, potential element re-emission, transformation, and dry
147 deposition can introduce variability, making continuous monitoring essential.
148 Finally, during the third snow sampling campaign, lasting from October 27th, 2020 to June 15th, 2021,
149 a weekly sampling was conducted at GSRS, with a total of 32 samples collected.

150 During snow sampling, the temperature and density of surface snow were measured, and the density
151 of snow was calculated based on weighting a 100 cc cylinder. After collection, snow samples were
152 melted, and two different aliquots were obtained and stored in separate vials. In a 1.5 mL
153 polypropylene (PP) vial, 1 mL of sample was stored for ionic species, while another aliquot was
154 stored in a 5 mL LDPE vials for trace elements analysis. PP vials designated to ionic species analysis
155 were previously sonicated for 30 min in UltraPure Water (UPW) (18 MΩ cm⁻¹ at 25 °C) for
156 decontamination. LDPE vial used for trace elements analysis were instead conditioned with HNO₃
157 2% and sonicated for 30 min. All sample aliquots were stored at -20°C in dark conditions and
158 transported to the Venice ISP-CNR laboratories.

159 To assess potential contamination during sampling, handling, and transport, field blanks were
160 collected during each campaign. Metal-free vials (Avantor, VWR Centrifuge Tubes, CHN) were
161 opened to ambient air at the sampling sites for a few minutes without collecting snow, then sealed
162 and transported to the Ny-Ålesund laboratory. There, they were filled with 2% HNO₃ and stored under
163 the same conditions as the snow samples. In parallel, analytical blanks were prepared by opening
164 vials to air, sealing them, and transporting them directly to Venice, where they were filled with 2%
165 HNO₃ and ultrapure water from the laboratory. Both field and analytical blanks were analyzed
166 alongside the snow samples, confirming that background contamination levels were consistently
167 below LODs or one order of magnitude lower than the lowest concentration detected for all analytes.

169 Furthermore, seawater temperatures and salinity at 10 m depth were also monitored in Kongsfjorden
170 (Kb3; 78°57.228'N, 11°57.192'E) during 2019-2021 spring seasons, with data collected every 3-6
171 days (Assmy et al. 2023). Data was derived from Conductivity Temperature Depth (CTD) casts with
172 either a MiniSTD model SD-204 (SAIV A/S, Bergen, Norway) or a XR-620 CTD (RBR Ltd, Ottawa,

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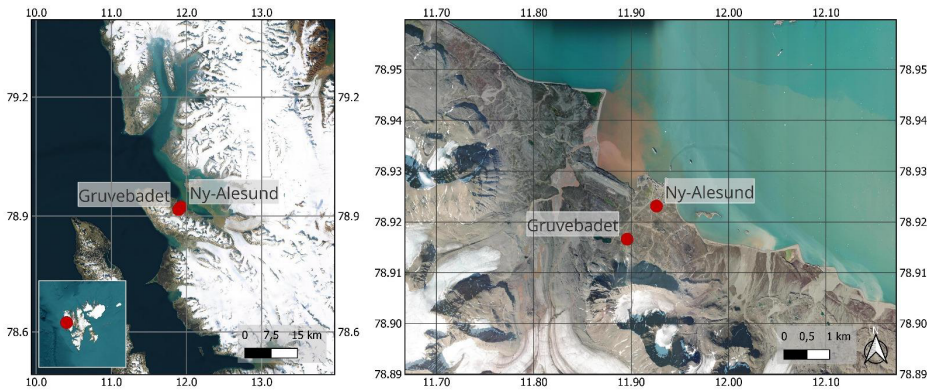
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173 Canada). Combined casts of both instruments conducted in May 2020 and 2021 did not reveal
174 differences in temperature or salinity in the reported accuracy (two post comma digits).

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176 **Fig. 1.** Sampling sites of Ny-Alesund and Gruvebadet, located in the Brøgger peninsula (Svalbard Islands).

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177 **2.2 Analysis of ionic species**

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178 The analysis of anionic species (Cl^- , Br^- , NO_3^- , SO_4^{2-} , MSA) was carried out using an ion
179 chromatograph (IC, Thermo Scientific Dionex™ ICS-5000, Waltham, MA, USA) coupled with a
180 single quadrupole mass spectrometer (MS, MSQ Plus™, Thermo Scientific, Bremen, Germany). The
181 separation was performed using an anionic exchange column (Dionex Ion Pac AS 19 2 mm ID $\times$ 250
182 mm length) equipped with a guard column (Dionex Ion Pac AG19 2 mm ID $\times$ 50 mm length). Sodium
183 hydroxide (NaOH), used as mobile phase, was produced by an eluent generator (Dionex ICS 5000EG,
184 Thermo Scientific). The NaOH gradient with a 0.25 mL min⁻¹ flow rate was: 0–6 min at 15 mM; 6–
185 15 min gradient from 15 to 45 mM; 15–23 min column cleaning with 45 mM; 23–33 min equilibration
186 at 15 mM. The injection volume was 100 μL . A suppressor (ASRS 500, 2 mm, Thermo Scientific)
187 removed NaOH before entering the MS source. The IC-MS operated with a negative electrospray
188 source (ESI) with a temperature of 500°C and a needle voltage of 3 kV. The other MS parameters are
189 reported by Barbaro et al. (2017). The same IC system was simultaneously used to determine cationic
190 species (Na^+ , K^+ , Ca^{2+} and NH_4^+). However, Ca^{2+} was not measured within the samples collected
191 during the second campaign due to instrumental limitations.

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192 The separation occurred with a capillary cation-exchange column (Dionex Ion Pac CS19–4 mm 0.4
193 mm ID $\times$ 250 mm length), equipped with a guard column (Dionex Ion Pac CG19–4, 0.4 mm ID $\times$ 50
194 mm length), and the species were determined using a conductivity detector. Analytical blanks of
195 ultrapure water ($> 18 \text{ M}\Omega \text{ cm}$) were included in the analysis, and the Method Detection Limit (MDL)

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196 was set to 3 times the standard deviation of the blank values. Checks for accuracy were made using
197 certified multi-element standard solutions for anions (Cl^- , Br^- , NO_3^- , SO_4^{2-} ; no. 89886-50ML-F, Sigma
198 Aldrich) and cations (Na^+ , K^+ , Ca^{2+} ; no. 89316-50ML-F, Sigma Aldrich) at a concentration of 10 mg
199 $\text{L}^{-1} \pm 0.2\%$. The analytical precision was quantified as the relative standard deviation (RSD) for
200 replicates ($n > 3$) of standard solutions and was always $< 10\%$ for each ion.

201 *2.3 Trace Elements analysis*

202 Twenty-six elements (Li, Be, Mg, Al, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Ag, Cd, Sb,
203 Cs, Ba, Tl, Pb, Bi and U) were analyzed on samples previously melted and acidified to 2% v/v with
204 HNO_3 (UpA grade, Romil, UK) for 24 hours before analysis (Spolaor et al., 2018; Spolaor et al.,
205 2021a).
206 The analysis was performed using Inductively Coupled Plasma Mass Spectrometry (ICP-MS, iCAP
207 RQ, Thermo Scientific, US). The ICP-MS was equipped with an ASX-560 autosampler (Teledyne
208 Cetac Technologies), PolyPro PFE nebulizer, PFE cyclonic spray chamber thermostated at 2.7°C ,
209 sapphire injector, quartz torch and Ni cones. The acquisition was performed at 1550 W of plasma RF
210 power in Kinetic Energy Discrimination (KED) – high matrix mode, using He as collision gas (4.3
211 mL min^{-1}). Instrument parameters were optimized for best sensitivity in the whole mass range,
212 minimum oxides ($< 1\%$) and double charges ($< 3\%$). Quantification was obtained by external
213 calibration with multi-elemental standards prepared in ultrapure water ($18 \text{ M}\Omega \text{ cm}^{-1}$ at 25°C) with
214 2% v/v ultrapure grade HNO_3 (UpA grade, Romil, UK), with a combination of certified level multi-
215 elemental solutions IMS-102 and IMS-104 from UltraScientific. Analytical quality control was
216 performed by memory test blank (repeated analysis of ultrapure grade HNO_3 2% v/v blank solution)
217 after each sample and calibration verification (repeated analysis of reference standards) every 11
218 samples. More details are found in Spolaor et al., 2021a.

220 *2.4 Transport modelling, sea ice, Kongsfjorden condition, and polar vortex*

222 The Lagrangian particle dispersion model HYSPLIT (Draxler, 1998; Stein et al., 2015) was used to
223 determine the source region of air masses over Ny-Ålesund. This model has previously been shown
224 to be an effective tool for the prediction of transport pathways into and within the Arctic and Antarctic
225 regions (Barbaro et al., 2015; Feltracco et al., 2021). The simulations were driven using
226 meteorological data from the Global Data Assimilation System (GDAS) one-degree archive, set the
227 top of the model at 10000 m and the height source equal to the GSRS altitude. Back-trajectories were
228 calculated every 6 h, with a propagation time of 120 h for each sampling period. The choice of a 6-

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

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

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

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

2.5 Statistical procedures and Enrichment Factors (EFs) Analysis

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

The Wilcoxon rank-sum test was applied on data from the 2018-19 sampling campaign to assess whether there was a statistically significant difference in the surface snow contamination levels (ionic loads) between the two sites: Ny-Ålesund and Gruvebadet. This model assumes that the data is sampled from two matched or dependent populations, and data is assumed to be continuous. Because it is a nonparametric test, it does not require a particular probability distribution of the dependent variable in the analysis. Furthermore, Enrichment Factors (EFs) using Ba as a crustal element of reference (Widepohl, 1995; Spolaor et al., 2021a; Ruppel et al., 2023), were calculated to explore the mixed sources of the investigated elements. To complement the EFs analysis, a Hierarchical Cluster

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275 Analysis (HCA) was performed using Ward's algorithm and Euclidean distances as clustering
276 criteria, to determine the presence of some clusters and simplify the interpretation of the dataset.
277 Additionally, to examine the inter-annual and seasonal variations in chemical's concentrations and to
278 identify the species most affected by these changes, a Kruskal-Wallis' test was employed, followed
279 by Dunn's post-hoc test for pairwise comparisons. The Kruskal-Wallis' test, a nonparametric
280 alternative to Anova, which is typically applied when samples do not follow a normal distribution
281 (Van Hecke, 2013), was chosen to assess differences in chemical concentrations across the years of
282 sampling campaigns and multiple seasons. This allowed for an objective evaluation of temporal
283 differences in concentration levels, while Dunn's post-hoc analysis, which uses the same shared
284 rankings and pooled variance assumed under the Kruskal-Wallis null hypothesis, helped identify
285 which specific seasons showed statistically significant differences from each other. The Bonferroni
286 correction was used for the Dunn's test to control for false positives when performing multiple
287 pairwise comparisons (Dunn, 1961).
288 The ultimate goal of this analysis was to uncover potential correlations between meteorological-
289 climatic conditions and chemical concentrations in surface snow, helping to better understand how
290 environmental factors contribute to contamination trends.
291 These combined statistical approaches provided a comprehensive framework for analysing the
292 complex dataset.

293 **3. Results**

294 *3.1 Comparison between concentration trends at Gruvebadet and Ny-Ålesund*

295 The concentration variations between an undisturbed area in Ny-Ålesund village and GSRS sites were
296 compared during the 2018-19 sampling campaign to better understand the effect of spatial variability
297 between the two sampling sites. The concentration trends of Na⁺, as sea salt tracer, Pb as
298 anthropogenic species, and Ca²⁺ as crustal tracer, are reported in Fig. S1, for both sampling sites.
299 Although the difference in time resolution between sites is apparent in Fig. S1, the difference in
300 concentration trends appears not statistically significant (p values < 0.05, Wilcoxon test) for all the
301 analysed species, except for sporadic peaks in sea salt and crustal tracers present in the Ny-Ålesund
302 record from November to February. These peaks do not consistently correlate with positive
303 temperature anomalies and precipitation events, as some occurred under low temperatures and
304 without significant precipitation (e.g., January and early February), suggesting that other processes,
305 such as long-range transport, wind-driven deposition, or dry deposition may also play a role (Fig. S1).
306 Concordant Pb trends emerge at Ny-Ålesund and Gruvebadet, with the highest concentrations
307 observed from February to May.

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313 To evaluate the differences in concentration range and spatial distribution of surface snow impurity
314 content, we applied the Wilcoxon test for the 2018-19 sampling period by comparing the distributions
315 for positive and negative differences of the ranks of their absolute values. **This comparison is crucial**
316 **for assessing spatial variability in the measured parameters, providing context and serving as a**
317 **reference for local-scale differences. However, at** a significance level of 0.01, the two distributions
318 from GSRS and Ny-Ålesund sites **appeared** not statistically different for all the trace elements and
319 most of the inspected ions, **except for Na⁺, K⁺, NO₃⁻, Br⁻, and MSA – which are typical sea salt and**
320 **biological species (MSA).**
321 For this reason, only the GSRS temporal trend has been considered throughout the manuscript,
322 referring to ionic loads (mg m⁻²) instead of concentrations (ng g⁻¹), to highlight the seasonal trends of
323 specific tracers. The ionic load is calculated as ionic concentrations multiplied by the density and the
324 thickness of sampled strata.

325 *3.2 Interannual trends of chemical species on the surface snow*

326 Three consecutive snow seasons were evaluated to define the chemical composition of the surface
327 snow in the Arctic site of GSRS. The sea salt ions Cl⁻ (50 %), **and** Na⁺ (23%) represent the most
328 abundant species, followed by SO₄²⁻ (11 %), Mg (7 %), Ca (2%), Fe (1%) and Al (1%). Similar
329 relative **concentration** abundances were also found in previous studies on the snow of the Svalbard
330 Archipelago (Beaudon and Moore, 2009; Vega et al., 2015; Barbaro et al., 2021; Spolaor et al.,
331 2021b).

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

338 **Table 1.** Average ionic loads of the most abundant (>1%) ionic and elemental species in the surface snow during each
339 season of the three consecutive sampling campaigns. The standard deviation is shown in brackets, while in the case of
340 nss-SO₄²⁻ the brackets represent the percentage of nss-SO₄²⁻ compared to the total SO₄²⁻. “n” indicates the number of
341 samples considered for the calculation of the average.

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)	116 (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)

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spring 2019 (n=51)	76 (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)
autumn 2019 (n=15)	214 (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)	339 (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)	273 (132)	110 (91)	52 (77)	28 (25)	15 (53%)	21 (21)	6 (9)	13 (16)	4 (2)	2 (4)	5 (8)
autumn 2020 (n=6)	803 (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)	181 (92)	107 (86)	36 (26)	16 (12)	7 (43%)	9 (10)	4 (7)	1 (2)	3 (2)	2 (2)	1 (1)

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The average loads of the first sampling year were lower compared to the other campaigns (Fig. S2; Table 1).

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

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The non-sea-salt sulphate (nss-SO_4^{2-}), calculated using a seawater $\text{SO}_4^{2-}/\text{Na}^+$ mass ratio of 0.252 (Millero et al., 2008), was the most abundant fraction of the total sulphate in autumn 2019 and winter 2020-21, while in autumn 2018 and 2020 sea salt sulphate (ss-SO_4^{2-}) was the dominant fraction. No clear predominance between the two fractions was achieved during the other investigated seasons, which may reflect a more mixed influence of marine and continental sources (Table 1).

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The abundance of all chemical species investigated is quite similar for all years (Fig. S2), although the sampling campaign 2019-20 showed higher percentage of calcium ranging between 3% and 5%, in contrast to the typical concentrations < 1% found in the other two campaigns.

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To further examine differences in species concentrations across seasons and years, a Kruskal-Wallis test was performed. The test indicates significant differences ($\chi^2 > 15.51$, $\text{df} = 8$, $p\text{-value} = 0.05$) for several species across the nine seasons of the three sampling campaigns. These species include Br ($\chi^2 = 16.9$), 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 ($\chi^2 = 31.9$), 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 ($\chi^2 = 40.9$), Zn ($\chi^2 = 29.9$). To identify which seasons accounted for these differences, a Dunn's test with Bonferroni correction was applied as a post-hoc test, revealing significant seasonal differences for multiple elements (Fig. S3), with pronounced differences particularly between autumn and winter, and between spring and winter ($P_{\text{adj}} < 0.05$; Table S5).

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380 Finally, correlations between meteo-climatic variables (air temperature, wind speed, wind direction,
381 precipitation, AO index) and the concentrations of the selected species were computed to assess
382 potential meteorological influences. The results indicated generally weak positive correlations ($p <$
383 0.5) for the variables considered. Notably, MSA showed a correlation with air temperature ($p = 0.2$),
384 as did Bi ($p = 0.2$), Cu ($p = 0.2$), Fe ($p = 0.2$), Pb ($p = 0.1$), Ni, ($p = 0.2$), Ag ($p = 0.2$), U ($p = 0.3$), V (p
385 $= 0.3$), and Zn ($p = 0.3$).

386 3.3 Polar vortex and Arctic Sea ice extent in 2019-20

387 According to the 2023 survey conducted by the National Snow and Ice Data Center (NSIDC), the
388 maximum extent of Arctic Sea ice since 2014 has been recorded in March 2020, with 14.73 million
389 square kilometres of the Arctic Ocean surface, in a decadal trend characterized by a -2.53% of decline,
390 due to the Arctic Amplification. Considering the Kongsfjorden area, the total sea ice extent varied
391 from 63.94 km² in March 2019 to 129.81 km² in March 2020, and was with 46.26 km² lowest in
392 March 2021 (Gerland et al., 2022). Specifications on Drift Ice (DI), Fast Ice (FI), and Open Water
393 (OW) extent are reported in Table S2. The 2020 maximum sea ice extent followed the exceptionally
394 strong and cold stratospheric polar vortex that took place in the Northern Hemisphere (NH) during
395 the 2019-20 polar winter, together with low wave activity from the troposphere, which allowed the
396 polar vortex to remain relatively undisturbed (Lawrence et al., 2020). Notably, the 2020 Arctic Sea
397 ice extent is 16% and 9% higher than previous (2018-19) and following (2020-21) records (dataset
398 NSIDC, NOAA). Lower surface mean air temperatures, (-14.7°C, compared to the -11.4°C recorded
399 in 2018-19, and the -8.0°C recorded in the 2020-21), average reduced precipitations, (2.1 mm
400 compared to 2.6 mm in 2018-19, and 3.2 mm in 2020-21), higher maximum mean wind speed (17.7
401 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),
402 and higher mean snow height (62.63 cm compared to 58.06 cm in 2018-19, and 57.77 cm in 2020-
403 21) were observed during the 2019-20 winter season, attributed to the influence of a strong polar
404 vortex triggered by a net positive Arctic Oscillation (AO) phase. The 2020 anomalous AO index is
405 displayed in Fig. S4. Seasonal values of mean air temperatures (°C), mean precipitation (mm),
406 maximum mean wind speed (m sec⁻¹) and mean snow depth (cm) during the three consecutive
407 sampling campaigns are reported in Table S3. Temperature data were provided by the Norwegian
408 Centre for Climate Services (NCCS), while sea ice extent data were supplied by National Snow and
409 Ice Data Center (NSIDC). Seawater temperature data collected at 10 m depth at a mid-fjord station
410 near Ny-Ålesund (Kb3) was found to be colder during 2020 compared to 2019 and 2021 spring
411 seasons (Table S4). Although these temperatures were still above the freezing point for the observed

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415 salinity levels, they contributed to colder overall conditions in Kongsfjorden, supporting the
416 formation and prolonged presence of sea ice when combined with cold atmospheric conditions.

417 **4. Discussion**

418 *4.1 Ny-Ålesund seasonal and interannual trends variability in surface snow*

419 The three consecutive sampling campaigns conducted from 2018 to 2021 confirmed the dominance
420 of sea salt input in the surface snow of Svalbard, due to the proximity of the sampling sites to the
421 coastline (Barbaro et al., 2021). The dominant ions are Na⁺, Cl⁻, and SO₄²⁻, associated with the
422 scavenging precipitation of marine aerosol (Hodgkins and Tranter, 1998). The observed mean
423 seasonal trends (Fig. S2) display the highest concentrations of marine species in autumn 2020,
424 followed by 2020-21 and 2019-20 winter seasons. However, wintry concentrations are presumably
425 linked to weakened (2019-20) or destroyed (2020-21) polar vortex (Fig. 2) and intense cyclonic
426 storms, associated with anomalous warming events capable of transporting both heat and moisture
427 from lower latitudes to Svalbard (Rinke et al., 2017). In addition to these factors, it is important to
428 account for snow ablation or erosion by wind. The change in snow height (Δh) between consecutive
429 sampling events provides valuable insights into such processes. A decrease in snow height signals
430 ablation can indeed result from snow melting (under positive temperatures), snow aging, sublimation,
431 or snow drift. In particular, when wind speeds exceed 5 m s⁻¹, the threshold commonly accepted for
432 initiating snow drift and ablation episodes (Pomeroy, 1989), we can infer the snow erosion due to
433 wind drift likely occurred (Fig. 2). This effect may explain some of the variability in snow ion
434 concentrations, especially during intense storms or strong winds.

435 Autumn 2020 represents most likely an outlier, due to scarce precipitations (Fig. 3) that led to more
436 concentrated impurities in the surface snow. In contrast, late spring 2020 saw a notable increase in
437 typical marine ions (Na⁺, Cl⁻, Br⁻, MSA, SO₄²⁻) and geogenic elements (Al, Ca, Mn, Fe, Sr), compared
438 to spring 2019 (Fig. 3). This increase may be attributed to the very close drift Arctic Sea ice presence
439 in Kongsfjorden (Table S2), which reached its maximum extent in March 2020 due to low-
440 temperature anomalies and intensified atmospherically driven sea ice transport and deformation,
441 caused by higher than normal winter wind speeds (Fig. S5). These conditions likely enhanced the
442 production of sea spray aerosols, which, when carried by winds, may have increased the deposition
443 of marine species onto the snowpack. The increased deposition of geogenic elements might also have
444 been influenced by low temperature anomalies (as seen with Fe), and/or by stronger wind speeds,
445 although significant correlations have not emerged in this preliminary statistical analysis.
446 Additionally, an outstanding positive phase of the Arctic Oscillation (AO) in the troposphere (Fig. 2)

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was recorded in January-March 2020 (Lawrence et al., 2020; Dethloff et al., 2022). Although the entire period was remarkable, January and February featured as clear outliers in the historical timeseries (1950–2023) reported by the NOAA service, while March 2020 exhibited significantly elevated values but did not exceed the statistical threshold for outliers (Fig. S4).

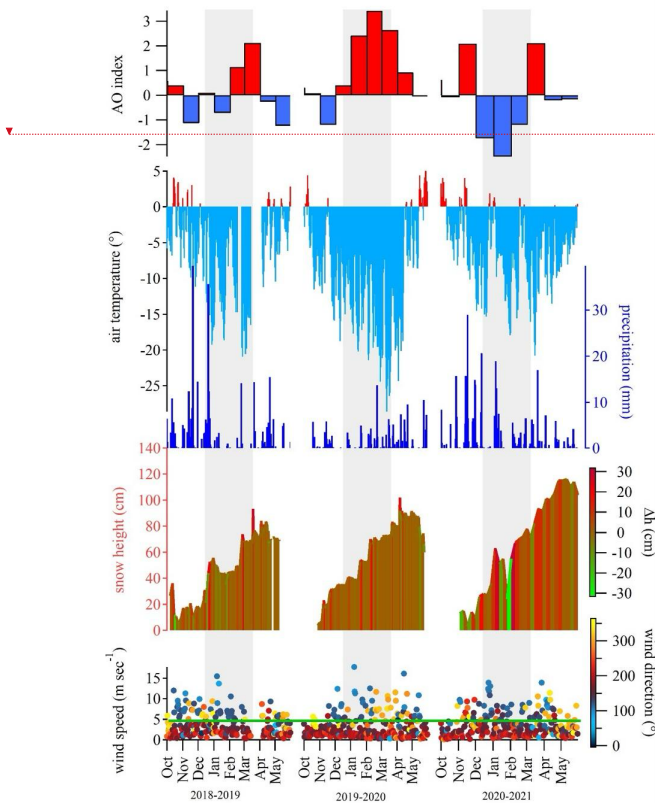


Fig. 2. AO Index, air temperature (°C), precipitation (mm), snow height (cm), Δh snow height (cm), wind speed (m sec⁻¹), and wind direction (°) from the NCEP/NCAR Reanalysis data. The green horizontal line above the wind speed graph indicates the 5 m sec⁻¹ threshold, above which wind drift may occur on surface snow layers. The colour of the line refers to the Δh color scale, which indicates negative values of Δh , NOAA Physical Sciences Lab's daily composites tool was used to calculate the near-surface air temperatures across the Northern Hemisphere from October to May. Grey bands indicate the winter periods.

A 2021 spring peak of marine species was also recorded, although more attenuated than in spring 2020 (Fig. 3, Fig. S2). This variation is likely attributable to different extents of sea ice in the fjord. Nonetheless, seawater temperatures in 2021, similar to those in 2020 and 2.3°C colder than in 2019

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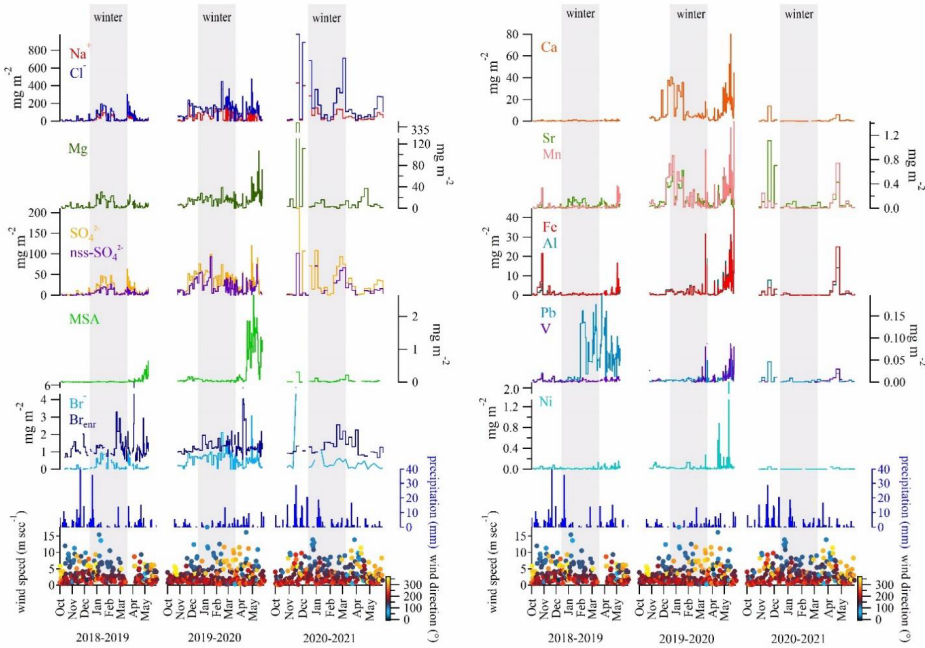
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(Table S4), along with comparable wind speed conditions (Fig. S5), may also have contributed to the observed trends in marine species concentrations. Similarly, the spring peak of Mg, Sr, Mn, Fe, Al, and V in 2021 seems to reflect the high wind speed and positive AO index recorded from March to April 2021. Positive anomalies for air temperatures (A) and wind speed (W) conditions, together with negative anomalies in seawater (O) conditions were observed during the 2020-21 campaign, while negative A and O conditions were accompanied to positive W during 2019-20. On the contrary, 2018-19 diverges from the other campaigns for positive O condition associated to negative W condition anomalies. These observations provide valuable insights into how shifts in atmospheric and oceanic conditions impact the concentrations of ionic and elemental species in surface snow, enhancing our understanding of the underlying mechanisms that govern these changes in the context of AA conditions.

Fig. 3. Wind direction (°), wind speed (m sec⁻¹), and ionic loads (mg m⁻²) of Na⁺, Cl⁻, Mg, SO₄²⁻, nss-SO₄²⁻, MSA, Br⁻, Ca, Sr, Mn, Fe, Al, Pb, V, Ni in the surface snow for the three sampling campaigns: 2018-19, 2019-20, 2020-21. Seasonal trends are here presented for specific elements to provide a detailed view of how concentrations vary across distinct sampling periods.



A singular case is represented by Pb, which showed a 12.5-fold increase during winter-spring 2019 period compared to autumn 2018. The EF for Pb during these seasons exceeded 100, indicating a

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strong anthropogenic contribution. In contrast, the peak observed in 2020 no longer shows such elevated EF values (i.e., EF = 26), suggesting a mixed source (Fig. 4). This implies that the April-May 2020 peak is largely of crustal origin, as the overlap with V (EF < 10) also suggests, possibly due to local dust events driven by strong winds exceeding the 5 m sec⁻¹ threshold (Fig. 3; Table S3).

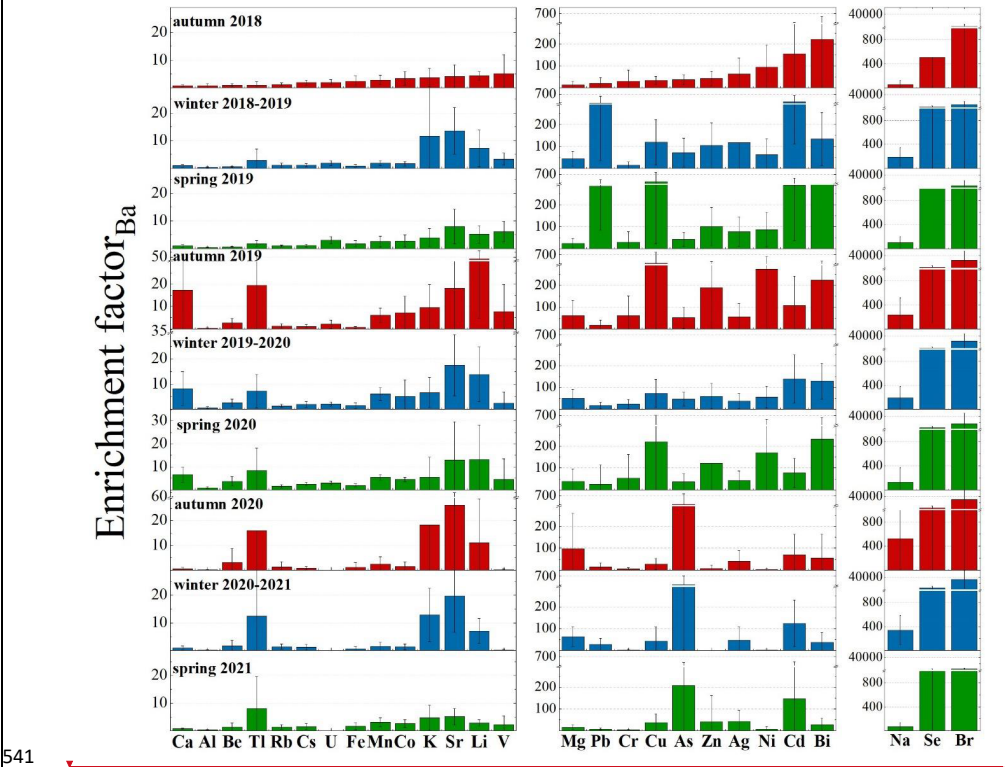


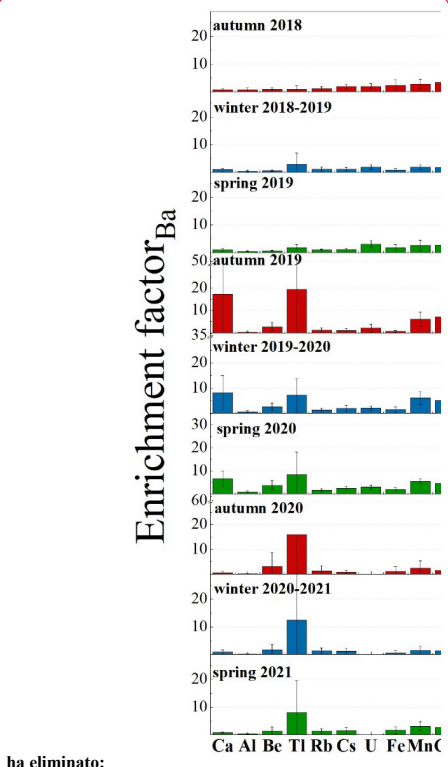
Fig. 4. Enrichment Factors (EFs) calculated on all the presented trace elements for the three sampling campaigns: 2018-19, 2019-20, 2020-21. Calculating EFs for the full dataset offers a more robust assessment of potential sources and enrichment patterns, minimizing the variability inherent in individual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

The calculated EFs (Fig. 4) for Be, Al, V, Mn, Fe, Co, Rb, Cs, and U were consistently below 10, indicating a predominantly crustal (geogenic) origin for these elements (Wedepohl, 1995; Gabrieli et al., 2011). In contrast, Ni, and Sr displayed EFs greater than 10, suggesting contributions from mixed sources. Notably, Ni exhibited exceptionally high EF values – above 100 – during autumn 2019 and spring 2020 (Ni). Mixed sources were also recognised for Li, K, Cr, Cu, Zn, As, Ag, Cd, and Ti with

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occasional EF values over 100. This suggests significant anthropogenic contributions, especially for Cu (spring and autumn 2019; spring 2020), As (from autumn 2020 to spring 2021), Zn (autumn 2019) and Cd (from autumn 2018 to spring 2019; winter 2019-20).

The 2019 springtime Pb concentration maxima are typically consistent with a mixture of eastern and western European sources (Sherrell et al., 2000; Bazzano et al., 2015, 2021). In this study, cluster mean trajectories obtained for winter 2018-2019 highlighted a 25% of air mass provenance from Russian Arctic and a 13% from eastern Siberia (Fig. S6), possibly explaining the higher concentrations of Pb revealed in spring 2019, following a reduced precipitation regime that occurred in January 2019. A local anthropogenic origin can be excluded though, since no activities (ordinary-extraordinary maintenance or particular events) were recorded in the vicinity of the sampling site in the 2019 sampling season. In addition, both GSRS and Ny-Ålesund (Fig. 1), located at 1 km of distance from each other, recorded comparable high concentrations of Pb, thus ruling out a possible contamination. However, at present, the long-range transport of Pb remains a hypothesis, likely supported by the breakdown of the wintry polar vortex (Fig. 2).

Backward trajectories (Fig. S6) for Ny-Ålesund area (78.92° N, 11.89° E) appear mostly in line with literature findings (Platt et al., 2022; Vecchiato et al., 2024), showing three main seasonal characters: a prevalent mass movement from ice-covered Central Arctic Ocean, Kara Sea, and Greenland Sea during autumn, a main provenance from Central Arctic Ocean and Kara Sea during winter, and a predominant trajectory from Northern Canada in addition to air masses arriving from Arctic Ocean and Kara seas during spring.

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

Examining the dominant ions associated with marine aerosol, we found Cl⁻/Na⁺ median ratios ranging from 1.3 to 1.5 w⁻¹, slightly lower than the expected value of 1.8 w⁻¹ in the pure seawater (Zhuang et al., 1999), pointing the occurrence of a minimum Cl⁻ depletion in aerosol, quantified as 14% for the 2018-19 and 2019-20 campaigns, and as just 2% for the 2020-21 campaign. A possible explanation for this phenomenon could be the de-chlorination of sea-spray aerosol during transport. This reaction occurs when sea-salt particles containing NaCl interact with acids such as HNO₃, H₂SO₄, or organic acids, leading to the release of gaseous HCl (Zhuang et al., 1999, and reference therein). Although less likely, this process could also occur at the snow-atmosphere interface. On the contrary, a possible influence of biomass burning on Cl⁻ depletion process has been excluded by the

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591 very low correlation (0.18, p value < 0.05) found between Cl⁻ depletion values and nss-K⁺/K⁺ ratios,
592 which is a tracer of relative contribution of biomass burning (Song et al., 2018).

593 Positive correlations between Mg, Ca, and K⁺ with Na⁺ and Cl⁻ (Fig. 5, p value < 0.05) suggest a
594 common sea-spray source. However, the concentrations of Mg are also positively correlated with nss-
595 Ca ($\rho_{load} = 0.55$), calculated according to Morales et al. (1998), indicating a shared non-marine source.
596 This suggests that in addition to their marine origin, these ions (Mg, Ca) also have contributions from
597 a non-marine, crustal source. Further evidence comes from the Ca/Mg ratios in surface snow samples
598 collected during the three campaigns, which were higher than those found in seawater (0.32, Millero
599 et al., 2008). This excess indicates the likely presence of mineral particles (i.e., calcite and dolomite),
600 potentially originating from local rock or soil dust (e.g., limestone, dolostone, and marble, which are
601 abundant in Svalbard), as previously observed by Barbaro et al. (2021). To further explore the mixed
602 sources of these elements, we calculated the Enrichment Factors (EFs) of Mg and Ca. For Mg, the
603 EF values were consistently above 10 in all the seasons analysed, indicating a significant non-crustal
604 source (e.g., marine). In contrast, Ca displayed EF values greater than 10 only during autumn 2019,
605 suggesting that its non-crustal contribution (likely from sea spray) was more pronounced during that
606 specific season. Based on these findings, we conclude that Mg and Ca effectively share a common
607 sea spray origin, but the sea-salt contribution of Ca was mainly significant in autumn 2019, while the
608 excess of Ca in other seasons likely reflects inputs from crustal source, such as local mineral dust.

609 Similarly, sulphate (SO₄²⁻) is highly and significantly correlated (p < 0.05) with both Na⁺ ($\rho_{load} =$
610 0.80) and Cl⁻ ($\rho_{load} = 0.91), indicating that sea-spray is its main source. Nonetheless, Na⁺/SO₄²⁻ and
611 Cl⁻/SO₄²⁻ ratios are significantly lower than typical seawater values (3.97 and 7.13, respectively,
612 according to Millero et al., 2008) for the former two campaigns (2018-19, 2019-20). The elevated
613 sulphate concentrations compared to sodium, also in winter snow, suggest the absence of a strong
614 frost flower signature. Additionally, the lack of significant depletion in nss-SO₄²⁻ further supports the
615 minimal role of frost flowers in contributing to the snow composition. Therefore, while frost flowers
616 are known to impact snow chemistry in Svalbard (Rankin et al., 2002; Beaudon and Moore, 2009;
617 Roscoe et al., 2011), our analysis indicates that their contribution to the observed sea salt peaks in
618 Kongsfjorden during the 2018-19, 2019-20 campaigns was likely limited. This analysis highlights
619 that, while sea ice extent supports higher sea-salt concentrations in snow, the specific sulphate and
620 ion ratios observed point to sea spray as the main source, with only a minor role for frost flower-
621 related inputs.$

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

629 To estimate the crustal fraction of sulphate (cr-SO_4^{2-}), the nss-Ca (as crustal marker) content was
630 multiplied by 0.59 ($\text{SO}_4^{2-}/\text{Ca}$ w/w ratio in the uppermost Earth crust - Wagenbach et al. 1996),
631 obtaining variable contributions for the three sampling campaigns, ranging from 2.45% up to 12.94%.
632 Conversely, the anthropogenic contribution to nss-SO_4^{2-} concentrations was investigated by the
633 application of the $[\text{ex-SO}_4^{2-}]$ concentration formula (Schwikowski et al., 1999), considering the
634 average concentration of $[\text{Ca}]$ instead of the average ionic concentration $[\text{Ca}^{2+}]$ because Ca^{2+}
635 concentrations were not measured in the samples collected during the second campaign due to
636 instrumental limitations:

637
$$[\text{ex-SO}_4^{2-}] = [\text{SO}_4^{2-}] - (0.12 [\text{Na}^+]) - (0.175 [\text{Ca}^{2+}])$$

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

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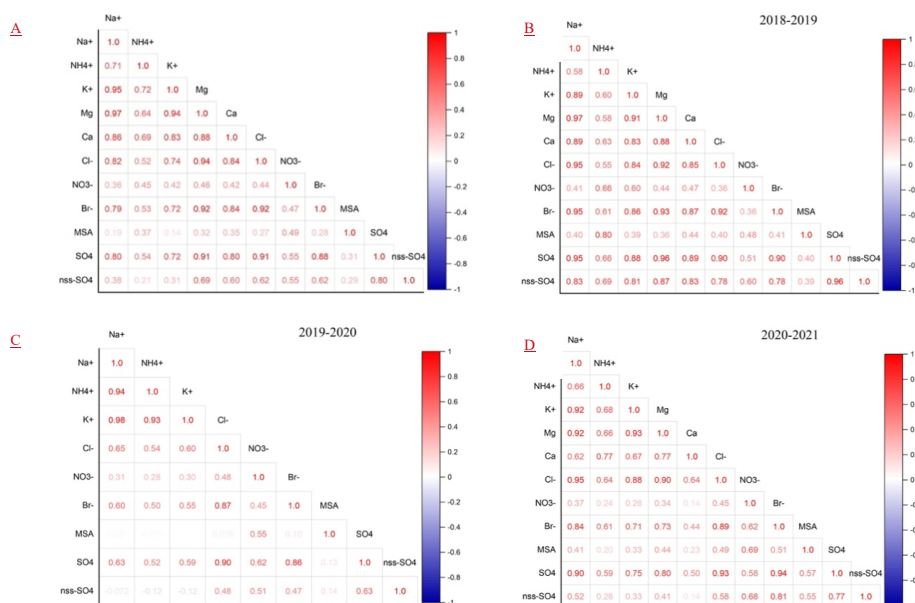


Fig. 5. Spearman correlation plots. A) Correlation plot for the three sampling seasons; B) Correlation plot for the 2018-2019 season; C) Correlation plot for the 2019-2020 season; D) Correlation plot for the 2020-2021 season.

To quantify the biogenic nss-SO₄²⁻ contribution, the methanesulfonic acid (MSA) loads - the final product of DMS oxidation - were multiplied by 3.0 (Udisti et al., 2016), revealing biogenic SO₄²⁻ contributions ranging from 0.15% (2018-19, 2020-21) up to 0.38% (2019-20). Furthermore, the MSA/nss-SO₄²⁻ ratio was inspected, revealing a mean value of 0.02 ± 0.03 during the first (2018-19) and the third (2020-21) sampling campaigns, and a maximum ratio equal to 0.06 ± 0.18 reached during the second campaign (2019-20), similar to the subarctic western North Pacific ratio found by Jung et al. (2014). However, several factors can influence MSA formation, a univocal marker of biogenic emissions, including higher biological productivity related to higher nutrient input; the concentrations of NO₃ radicals as key oxidants for DMS decomposition (higher NO₃ gives higher MSA); and lower air temperatures, which tend to yield higher MSA levels (Andreae et al., 1985; Udisti et al., 2020).

For the 2019-20 campaign, it seems likely that a combination of these three factors, together with the positive expansion of sea ice and the very close drift ice presence in March 2020, as revealed from satellite reconstructions (Fig. S7), contributed to the increased release of MSA in aerosol, and its consistent deposition in surface snow (Fig. 3). Indeed, DMS was likely accumulated under the sea ice cover in the fjord and surrounding areas, and then being released and oxidised in the atmosphere when the ice broke off and melted (April-May). Furthermore, lower temperatures, (-17.3°C in March

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2020, compared to -16.1°C in March 2019, and -9.8°C in March 2021), positive correlation between MSA and NO_3^- ($\rho_{\text{load}} = 0.49$), and high-speed short-range transport (wind directions between 0°-60° and speeds > 5 m sec⁻¹) from the source to the near-coast sink site (GSRS) would have aided elevated concentrations of MSA in atmospheric depositions. However, the dominant south and southwest winds (180°-240°) during the major MSA peak in April 2020 (Fig. 3) likely transported marine aerosols and DMS from open ocean regions, further facilitating the increased MSA concentrations observed.

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

Finally, the ammonium (NH_4^+) load showed positive correlations (Fig. 5) with Na^+ ($\rho_{\text{load}} = 0.71$), Cl^- ($\rho_{\text{load}} = 0.52$) and K^+ ($\rho_{\text{load}} = 0.72$), as well as with SO_4^{2-} ($\rho_{\text{load}} = 0.54$), NO_3^- ($\rho_{\text{load}} = 0.45$), MSA ($\rho_{\text{load}} = 0.37$) and Br^- ($\rho_{\text{load}} = 0.53$), suggesting a close link with sea-salt ions and biogenic emissions. However, some contribution from biomass burning events and potential influence from anthropogenic activities cannot be excluded.

4.3 Bromine enrichment

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

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727 phase Br into the polar atmosphere, thus supplying an extra Br⁻ source in addition to sea spray
728 (Spolaor et al., 2016).

729 4.4 *Anthropogenic and natural sources of ions and particulate trace elements*

730 To complement the EFs analysis and further distinguish possible anthropogenic contributions from
731 natural ones (marine and geogenic) for ions and particulate trace elements, a Hierarchical Cluster
732 Analysis (HCA) method was carried out. Results of clustering (Fig. 6) clearly disentangle marine
733 (Na⁺, Cl⁻, K⁺, NH₄⁺, SO₄²⁻, NO₃⁻, Br⁻, Mg, Sr, bio-SO₄²⁻, MSA), anthropogenic (As, Co, Ag, Ba, Cd,
734 Zn, Pb, Bi, Cr, Cu, Ni), and geogenic (nss-Ca, Tl, Li, Al, Cs, Rb, Fe, Mn, U, Be, Se, V) sources of
735 ionic and elemental species, considering the whole sampling campaign period (2018-2021).
736 Interestingly, nss-SO₄²⁻ is brought together with the marine cluster, suggesting that its presence is
737 largely influenced by marine biogenic sources, alongside contributions from secondary sulphate
738 formation in the atmosphere. This indicates that nss-SO₄²⁻, despite having a variety of sources such
739 as human contribution or dust, is closely linked to the marine environment. One important reason for
740 this is the emission of DMS by phytoplankton. Additionally, secondary sulphate formation may have
741 further contributed to the nss-SO₄²⁻. Co was grouped within the anthropogenic cluster in HCA, in
742 contrast with EFs that suggested a crustal origin. This apparent discrepancy may reflect the relatively
743 larger uncertainties typically associated with EF calculations, which can inherit errors from the choice
744 of reference element, assumptions about crustal composition, and variability in background
745 concentrations, compared to the more integrative approach of HCA (e.g., Reimann and de Caritat,
746 2000). Moreover, the trend for cobalt closely aligns with anthropogenic ones (e.g., As), while distinct
747 trends are evident for crustal elements. Therefore, while the HCA results offer a reliable perspective
748 on source differentiation and clustering patterns, they are best interpreted as complementary to the
749 EF calculations rather than directly integrated with them. Incorporating EFs directly into the HCA
750 could introduce significant inaccuracies, as EFs rely on reference element ratios that may vary and
751 thus add complexity to the clustering process, potentially skewing the results. Consequently, HCA
752 independently provides robust insights in this context, enhancing our understanding without the
753 additional uncertainties that EF-based clustering might introduce.

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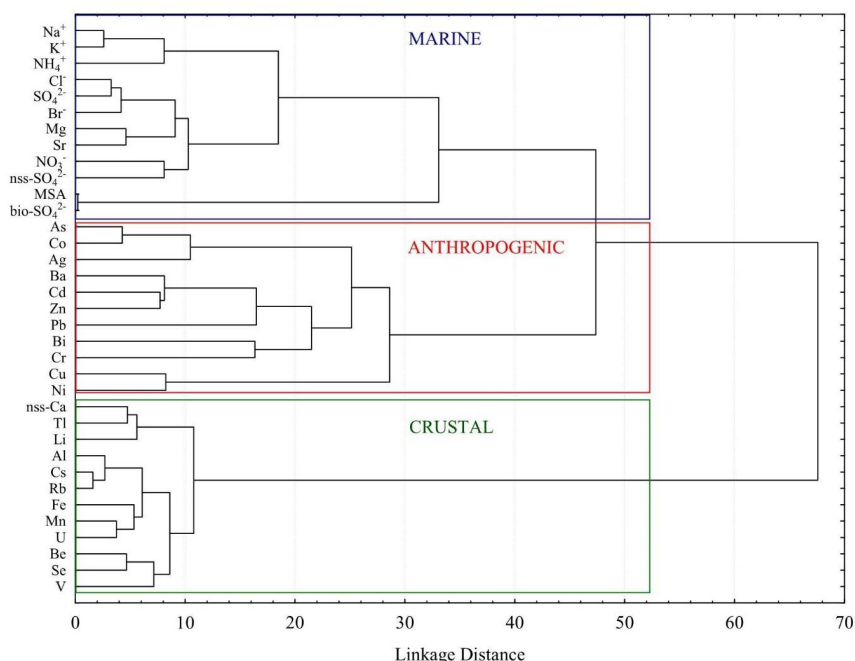


Fig. 6. Hierarchical cluster analysis applied to further disentangle the particulate trace element non-crustal sources.

5. Summary and Conclusion

In this study, trace elements and major ions were investigated in surface snow samples collected in Ny-Ålesund between October 2018 to June 2021. Seasonal and interannual variations of impurities have been observed, with general higher concentrations of marine species revealed in late spring 2020, associated to more extensive sea ice in Kongsfjorden in March 2020, promoted by negative temperature anomalies in both atmosphere and ocean and likely related to higher air mass recycle within the Arctic. These results provide direct evidence of how sea ice extent modulates the storage, release, and transport of marine-derived impurities, thereby influencing snow-atmosphere chemical exchange processes under varying climatic conditions. Higher concentrations in spring 2020 for geogenic and anthropogenic species are consistent with the influence of higher wind speeds, low atmospheric temperature anomalies, and generally drier conditions resulting from the exceptional occurrence of a strong and cold stratospheric polar vortex, accompanied by an unprecedentedly positive phase of the Arctic Oscillation in the troposphere during January-March 2020. While correlations

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815 between meteorological variables and concentrations were generally weak, the overall meteorological
816 context suggests that these unusual conditions likely contributed to the observed patterns. Such
817 findings illustrate how large-scale atmospheric circulation anomalies associated with Arctic
818 Amplification may significantly alter the deposition patterns of both natural and anthropogenic
819 species in the snowpack. This is particularly evident especially in cold years, when the production of
820 sea spray related aerosol likely derives by a larger extension of sea ice and stronger local Arctic
821 circulation. The identification of geogenic, marine, and anthropogenic sources in the snowpack was
822 allowed by a chemometric approach (HCA), which clarified the EFs results. The use of chemometric
823 techniques (HCA) and back-trajectory analysis enabled a clearer attribution of sources and transport
824 pathways, improving the interpretation of snow composition in relation to meteorological drivers.
825 Specifically, the distinct seasonal air mass patterns revealed, characterised by dominant Arctic-origin
826 air masses in fall and winter, and a lack of expected mid-latitude inputs in spring, underscore the
827 changing dynamics of snow-atmosphere interactions in a warming Arctic. Overall, these insights
828 advance our understanding of how recent climatic anomalies, such as altered sea ice extent, shifts in
829 Arctic Oscillation phases, and stronger polar vortices, modulate the chemical composition of the
830 snowpack in Svalbard. Our findings highlight the sensitivity of snow-atmosphere exchanges to both
831 local and large-scale climatic processes, offering important context for interpreting snow chemistry
832 trends in a rapidly changing Arctic environment. Further statistical investigations, including
833 multivariate and longer-term analyses, would be valuable for better quantifying these correlations.

834 **Data availability**

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

838 **Author contribution**

839 AS: Conceptualization, Data curation, Investigation, Writing-original draft, Writing-review and
840 editing. EB: Conceptualization, Field work, Data curation, Formal Analysis, Writing-original draft,
841 Writing-review and editing, Funding acquisition. MF: Formal Analysis, Field work, Data curation,
842 Writing-review and editing. FS: Field work, Formal analysis, Investigation, Writing-review and
843 editing. MV: Writing-review and editing. MV: Field work, Writing-review and editing. MM:
844 Investigation. FB: Investigation, Writing-review and editing. FB: Investigation. Field work. CJMH:
845 Investigation, Data curation, Writing-review and editing. AB: Field work, Data curation, Writing-

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review and editing. AG: Resources, Supervision, Validation, Writing-review and editing, Funding acquisition. CB: Resources, Supervision, Validation, Writing-review and editing, Funding acquisition. AS: Funding acquisition, Supervision, Validation, Writing-review and editing.

Competing interests

The authors declare that they have no conflict of interest.

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Formattato: Regola lo spazio tra testo asiatico e in alfabeto latino, Regola lo spazio tra caratteri asiatici e numeri

ha eliminato: <https://doi.org/10.1088/1748-9326/aa7def>

ha eliminato: <https://doi.org/10.1029/2010JD015144>

ha eliminato: <https://doi.org/10.1029/2022JD038378>

ha eliminato: <https://doi.org/10.3390/rs15184435>

ha formattato: Inglese (Regno Unito)

ha eliminato: [https://doi.org/10.1016/B978-0-12-382225-3.00228-0...](https://doi.org/10.1016/B978-0-12-382225-3.00228-0)

ha eliminato: <https://doi.org/10.3402/tellusb.v51i5.16506>

ha formattato: Legature: Nessuna

ha formattato: Inglese (Regno Unito)

ha eliminato: [http://dx.doi.org/10.1016/j.gloplacha.2011.03.004...](http://dx.doi.org/10.1016/j.gloplacha.2011.03.004)

ha formattato: Inglese (Regno Unito)

ha eliminato: <https://doi.org/10.1029/1999GC000007>

ha formattato: Inglese (Regno Unito)

ha eliminato: <https://doi.org/10.1016/j.jhydrol.2020.125279>

ha formattato: Inglese (Regno Unito)

ha eliminato: <https://doi.org/10.1021/acs.est.1c07796>, 2022

ha formattato: Inglese (Regno Unito)

ha eliminato: 29¶

ha formattato: Colore carattere: Nero

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ha eliminato: <https://doi.org/10.5194/tc-18-307-2024>

ha formattato: Inglese (Regno Unito)

ha eliminato: <https://doi.org/10.1016/j.scitotenv.2020.141640>

ha formattato: Inglese (Regno Unito)

ha formattato: Inglese (Regno Unito)

ha eliminato: <https://doi.org/10.3389/feart.2020.536036>

ha formattato: Inglese (Regno Unito)

ha eliminato: <https://doi.org/10.5194/acp-19-13325-2019>

ha formattato: Inglese (Regno Unito)

ha eliminato: <https://doi.org/10.1016/j.chemosphere.2017.12.180>

ha formattato: Inglese (Regno Unito)

ha eliminato: <https://doi.org/10.5194/tc-10-245-2016>

ha formattato: Inglese (Regno Unito)

ha eliminato: <https://doi.org/10.5194/acp-14-9613-2014>

ha formattato: Inglese (Regno Unito)

ha eliminato: <https://doi.org/10.1175/BAMS-D-14-00110.1>

ha formattato: Inglese (Regno Unito)

ha spostato (inserimento) [2]

ha eliminato: 30¶

ha formattato: Colore carattere: Nero

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ha formattato: Inglese (Regno Unito)

Formattato: Regola lo spazio tra testo asiatico e in alfabeto latino, Regola lo spazio tra caratteri asiatici e numeri

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ha formattato: Inglese (Regno Unito)

ha formattato: Inglese (Regno Unito)

ha formattato: Inglese (Regno Unito)

ha eliminato: <https://doi.org/10.3390/atmos12060694>

ha eliminato: https://doi.org/10.1007/978-3-030-33566-3_4

ha eliminato: <https://doi.org/10.1007/s12210-016-0517-7>

Formattato: Regola lo spazio tra testo asiatico e in alfabeto latino, Regola lo spazio tra caratteri asiatici e numeri

ha eliminato: <https://doi.org/10.1016/j.envpol.2023.122864>

ha eliminato: <https://doi.org/10.1016/j.envpol.2018.07.095>

ha formattato: Inglese (Regno Unito)

ha formattato: Motivo: Trasparente, Evidenziato

ha formattato: Inglese (Regno Unito)

ha eliminato: <https://doi.org/10.3402/polar.v34.23246>

ha eliminato: [https://doi.org/10.1016/0016-7037\(95\)00038-2](https://doi.org/10.1016/0016-7037(95)00038-2)

ha formattato: Inglese (Regno Unito)

ha eliminato: <https://doi.org/10.5194/acp-14-6427-2014>

ha eliminato: 31

ha formattato: Colore carattere: Nero

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ha formattato: Inglese (Regno Unito)

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ha formattato: Inglese (Regno Unito)

ha eliminato: 32 ¶

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