



HAL
open science

Distribution of trace and major elements in subarctic ecosystem soils: Sources and influence of vegetation

Yannick Agnan, Romain Courault, Marie Alexis, Tony Zanardo, Marianne Cohen, Margaux Sauvage, Maryse Castrec-Rouelle

► To cite this version:

Yannick Agnan, Romain Courault, Marie Alexis, Tony Zanardo, Marianne Cohen, et al.. Distribution of trace and major elements in subarctic ecosystem soils: Sources and influence of vegetation. *Science of the Total Environment*, 2019, 10.1016/j.scitotenv.2019.05.178 . hal-02136021

HAL Id: hal-02136021

<https://hal.science/hal-02136021>

Submitted on 25 Oct 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial 4.0 International License

1 **Distribution of trace and major elements in subarctic ecosystem soils: sources and**
2 **influence of vegetation**

3 Yannick Agnan^{1*}, Romain Courault², Marie A. Alexis¹, Tony Zanardo¹, Marianne Cohen²,
4 Margaux Sauvage³, Maryse Castrec-Rouelle¹

5 ¹Sorbonne Université, CNRS, EPHE, UMR METIS, F-75252, Paris, France

6 ²Sorbonne Université, CNRS, FRE ENeC, F-75006, Paris, France

7 ³Université Paris Diderot, Pôle Image, F-75013, Paris, France

8

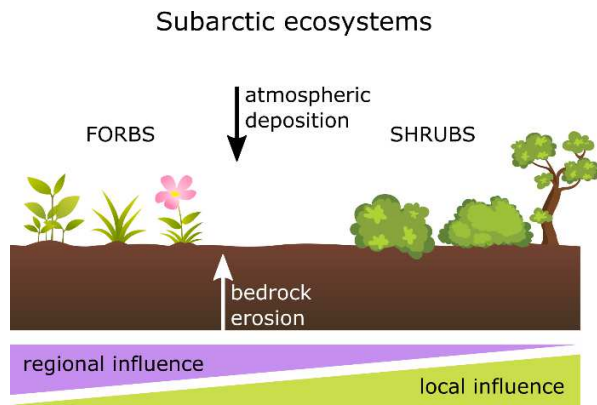
9 *Corresponding author:

10 Yannick Agnan: yannick.agnan@biogeochimie.fr

11

12

13 **Graphical abstract**



14

15

16 Abstract

17 Artic and subarctic environments are particularly sensitive to climate change with a faster
18 warming compared to other latitudes. Vegetation is changing but its role on the biogeochemical
19 cycling is poorly understood. In this study, we evaluated the distribution of trace elements in
20 subarctic soils from different land covers at Abisko, northern Sweden: grassland, moor, broad-
21 leaved forest, and peat bog. Using various multivariate analysis approaches, results indicated a
22 spatial heterogeneity with a strong influence of soil horizon classes considered: lithogenic
23 elements (e.g., Al, Cr, Ti) were accumulated in mineral horizon classes and surface process-
24 influenced elements (e.g., Cd, Cu, Se) in organic horizon classes. Atmospheric influences
25 included contamination by both local mines (e.g., Cu, Fe, Ni) and regional or long-range
26 atmospheric transport (e.g., Cd, Pb, Zn). A non-negative matrix factorization was used to
27 estimate, for each element, the contribution of various sources identified. For the first time, a
28 comparison between geochemical and ecological data was performed to evaluate the influence of
29 vegetation on element distribution. Apart from soil pH that could control dynamics of As, Cu,
30 and Se, two vegetation classes were reported to be correlated to geochemical factors: forbs and
31 shrubs/dwarf shrubs probably due to their annual vs. perennial activities, respectively. Since
32 these are considered as the main vegetation classes that quickly evolve with climate change, we
33 expect to see modifications in trace element biogeochemical cycling in the future.

34

35 **Keywords:** trace elements, soil, ecosystem, vegetation, subarctic, sources

36

37 Highlights:

38 Concentrations of elements were measured in different subarctic soils.

39 Four ecosystems were compared: grassland, moor, broad-leaved forest, and peat bog.

40 Spatial heterogeneity of concentrations resulted from different sources.

41 Only forbs and shrubs showed covariance with trace element distribution.

42 Soil pH controlled the geochemical dynamics of As, Cu, and Se.

43 **1. Introduction**

44 Arctic tundra and subarctic taiga cover 16% of the continental surface area. Biological and
45 climate characteristics of these two biomes imply distinct net primary productions (180 and
46 $380 \text{ g m}^{-2} \text{ a}^{-1}$, respectively), suggesting different carbon dynamics (Chapin *et al.*, 2011). The
47 limit between these two biomes, which could be up to 200 km in width, constitutes an important
48 vegetation transition: the tundra–taiga ecotone (Callaghan *et al.*, 2002a). Furthermore, the high
49 latitudes are sensitive to climate change with faster warming compared to the rest of the globe
50 (Acosta Navarro *et al.*, 2016; IPCC, 2013). Increase in air temperature induces a permafrost
51 warming (up to +2 °C in 30 years; Romanovsky *et al.*, 2010) that modifies Arctic and subarctic
52 landscapes (Callaghan *et al.*, 2002b). In parallel, long-term high latitude vegetation records show
53 increasing biomass (i.e., greening) in Arctic and subarctic ecosystems related to climate change
54 (Beck and Goetz, 2011; Ju and Masek, 2016). Ecosystems are currently displacing to the North
55 in response to the rising temperatures (Berner *et al.*, 2013). Observations performed in Siberia
56 from 1910s to 2000s report a boreal shift of about 300–500 m during the last century (Shiyatov
57 *et al.*, 2007), despite very complex mechanisms implied on the local scale (Callaghan *et al.*,
58 2013).

59 Trace elements are characterized by very low concentrations in the environment (<0.1% in the
60 continental crust). They include various chemical families—metals (e.g., Pb, Cd, and Zn),
61 metalloids (As and Sb), and nonmetals (Se)—which behave differently in the environment in
62 relation to their physicochemical characteristics (Kabata-Pendias, 2010). If some of them are
63 required for many biological activities (such as Cu and Zn), all trace elements present potentially
64 harmful effects on human and ecosystem health in more or less high concentrations (Klassen *et al.*,
65 2010; Ulrich and Pankrath, 1983). In remote areas, such as Arctic and subarctic regions, trace
66 elements come from two major sources: local bedrock erosion (lithogenic fraction) and
67 atmospheric inputs (atmospheric fraction). Indeed, the low air temperature in polar environments
68 increases the atmospheric deposition of contaminants emitted in temperate latitudes (Law and
69 Stohl, 2007). Environmental archives (peat bogs, lake sediments, and ice cores) showed a recent
70 increase of As, Cd, Cu, Pb, and Zn deposition in the Arctic during the last decades attributed to
71 coal and gasoline combustions promoting accumulation of contaminants in the high latitudes

72 (Evenset *et al.*, 2007; McConnell and Edwards, 2008). Some rare metals (e.g., Pt, Pd, Rh) also
73 indicate a dramatic progression since the 1970s linked to their application in various new
74 technologies (Barbante *et al.*, 2001). Distinguishing anthropogenic from natural sources,
75 however, constitutes a challenge in Arctic and subarctic soils (Halbach *et al.*, 2017).
76 Geochemical tracers such as stable isotopes and rare metals (e.g., Ce, La, and Y) can help to
77 assess each source of trace elements (Agnan *et al.*, 2014; Fedele *et al.*, 2008).

78 Despite their pristine conditions, polar regions are accumulating contaminants that exposes
79 wildlife to bioaccumulation and biomagnification within trophic levels (AMAP, 2018; Dietz *et al.*
80 *et al.*, 2000; Tyler *et al.*, 1989). Indeed, data reported in Arctic biota indicate important
81 concentrations in different organisms, which induces direct or indirect health impacts (Gamberg
82 *et al.*, 2005). For example, the increase of some elements in West Greenland caribou, likely
83 linked to atmospheric deposition on lichens, may affect their reproduction functions (Gamberg *et al.*
84 *et al.*, 2016). Both aquatic and terrestrial ecosystems are therefore subjected to a long-term impact
85 of trace elements, and fauna vulnerability was recently shown to be largely dependent on their
86 habitat (Pacyna *et al.*, 2019).

87 Trace elements therefore require to be spatially characterized to better constrain both pools and
88 fluxes of elements in polar regions (Nygård *et al.*, 2012; Steinnes and Lierhagen, 2018). This is
89 particularly critical in ecosystems largely influenced by climate change that modifies element
90 dynamics (AMAP, 2011; IPCC, 2013): e.g., through modifications of hydrological regime
91 related to precipitation and snowpack cover or through the increase of lithogenic pool with
92 permafrost warming. The influence of vegetation on trace element dynamics was already
93 observed, which can manifest in different processes, including atmospheric deposition
94 interception (e.g., Ilyin *et al.*, 2017), element storage and recycling (e.g., Herndon *et al.*, 2015),
95 and modification of soil properties changing trace element speciation (e.g., Ren *et al.*, 2015). It
96 has been recently reported that Arctic tundra vegetation plays an important role on Hg cycling
97 through accumulation of gaseous elemental Hg from the atmosphere (Obrist *et al.*, 2017).
98 Processes involved, however, are currently not well understood and need more investigation.

99 In this context, we expect that vegetation in the tundra–taiga transition ecosystem can modify
100 the current and future dynamics of elements coming from local rock dust, regional mining, and

101 long-range atmospheric deposition. More specifically, we hypothesize that lichens and mosses,
102 as well as herbaceous plant species, accelerate the accumulation of elements from the
103 atmosphere to the soil related to their physiological features (higher bioaccumulation and faster
104 turnover, respectively) compared to ligneous species, such as shrubs. The main objective of this
105 study was to determine the spatial distribution of potentially harmful elements in subarctic soils
106 in northern Sweden (Abisko) comparing different land covers (grassland, moor, broad-leaved
107 forest, and peat bog) in order to specifically: (1) identify the occurrence and sources of trace
108 elements in these soils and (2) evaluate the influence of ecosystems on the element distribution.

109 **2. Materials and methods**

110 **2.1. Study sites**

111 The study area was located around Abisko Scientific Research Station in northern Sweden
112 (68.3°N), about 200 km north to the Arctic circle (**Figure 1**). Four sampling sites, characterized
113 by different land covers, were considered: two located on the east slope of the Njulla Mountain
114 at 975 m a.s.l. (NJU975) and 383 m a.s.l. (NJU383), one on the north-west slope of the
115 Baddosdievvá Mountain at 599 m a.s.l. (BAD599), and one on the south shore of Lake
116 Torneträsk at 363 m a.s.l. (TOR363).

117 **2.1.1. Lithology and soil**

118 All study sites were located in the Scandinavian Mountains characterized by metamorphic
119 lithologies mainly from the Caledonian (490–390 Ma) and Svecofennian (1.85–1.75 Ga)
120 orogenies (**Table 1**). More specifically, NJU975 and NJU383 were characterized by Cambrian
121 and Ordovician metamorphic bedrocks (phyllite, micaschist, quartzite, metaconglomerate),
122 including limestone deposits. BAD599 also presented metamorphic unit (feldspathic
123 metasandstone, calcareous micaschist, quartzite, phyllite, metaconglomerate, amphibolite,
124 eclogite) dated from the Neoproterozoic. Finally, TOR363 was characterized by a distinct
125 lithology due to its location between an Ediacarian and Cambrian platformal sedimentary unit
126 (sandstone, conglomerate, siltstone, shale) and a Palaeoproterozoic lithotectonic unit (granite and
127 pegmatite). Study sites were characterized by acidic organic-rich soils (histosols) that were

128 underlain by a discontinuous permafrost (approximately 4–16 m thick according to the location)
129 with an active layer of 60–80 cm thick during the last decade (Åkerman and Johansson, 2008).

130 **2.1.2. Climate**

131 Abisko is characterized by a cool continental/subarctic climate (Dfc following the Köppen
132 climate classification) with a mean annual air temperature of $-0.1\text{ }^{\circ}\text{C}$ and a mean annual
133 precipitation of 330 mm a^{-1} (averages from 1981–2010, Abisko Meteorologic Station NOAA-
134 NCDC). Recent evolution of Abisko climate showed a significant increase in both temperatures
135 and precipitation: modeled rise is about $+1.75\text{ }^{\circ}\text{C}$ for annual air temperatures and $+69.3\text{ mm a}^{-1}$
136 for annual precipitations between 1967 and 2013 (Courault *et al.*, 2015). The prevailing winds
137 were from the south-west (26% as a percentage frequency, 1987–2017, Abisko Meteorologic
138 Station NOAA-NCDC; **Figure 1**) and remained unchanged over seasons (34% in fall, 36% in
139 winter, and 22% in spring), excepted in summer where winds come from the north-west (22%).
140 The mean annual wind speeds were 3.46 m s^{-1} (1987–2017, Abisko Meteorological Station
141 NOAA-NCDC).

142 Based on downscaled reanalysis (CHELSA project; Karger *et al.*, 2017) and interpolated data
143 from the WorldClim 2.0 data base (Fick and Hijmans, 2017), mean annual air temperatures
144 varied from -3.2 to $-0.1\text{ }^{\circ}\text{C}$ according to the local topoclimatology (*i.e.*, elevation, slope
145 steepness, and exposure; **Table 1**). Average wind speed decelerated from the summits to the
146 Lake Torneträsk (NJU975 to TOR363).

147 **2.1.3. Mining context**

148 Several metallogenic areas, producing base metals (blue shapes in the map) and ferrous metals
149 (red shapes in the map) as far back as the 18th century, are located around the study area (**Figure**
150 **1**; Eilu, 2012). To the west of Abisko, the mining area of Sjangeli (production of Cu, Fe, Pb, Ag,
151 Au, and U) includes two abandoned open pit Cu mines: Kopparåsen (12 km to the North-West)
152 and Sjangeli (28 km to the South-West). In eastern Abisko, two mining areas are close to Kiruna
153 (80 km to the South-East): Viscaria-Sautusvaara mining area that contained large Cu and Fe
154 deposits (production of Cu, Fe, Au, Zn, Ag, and Co) and Kiruna mining area that contained the
155 most important Fe deposit of Europe (production of Fe, Cu, and Au). According to their

156 respective exposure (i.e., east facing for NJU975 and NJU363 vs. north-west facing for
157 BAD599), we expect different mining influences within the study area.

158 **2.1.4. Ecology**

159 Each site is characterized by distinct land cover (grassland, moor, broad-leaved forest, and peat
160 bog; European Environment Agency, 2014) and ecological habitat (Davies *et al.*, 2004; **Table 1**).
161 Ecosystems were strongly structured by the altitudinal gradient (~1000 m) between the Lake
162 Törnetrask and the summits (Slåttatjåkka/Šloahtta and Njulla Mountains). Four bioclimatic belts
163 were identified in the study area: subalpine birch forest, low, middle, and high alpine belt.

164 Lowest altitudes are characterized by the subalpine forest belt, a forest type mainly structured
165 by *Betula pubescens* ssp. *czerepanovii*. Shrub and herbaceous layer shared vegetal species in
166 common with the northern boreal zone, including additional mountain species: *Empetrum*
167 *nigrum* ssp. *nigrum*, *Trollius europaeus*, *Angelica archangelica* ssp. *archangelica*, *Pedicularis*
168 sp. Forest undergrowth are made of heath (*Betula nana*, *Vaccinium myrtillus*) or meadow (*Salix*
169 sp., *Rumex* sp., *Alchemilla* sp., *Saussurea alpina*) communities. Meadows soils were generally
170 less acidic, might locally be calcareous in accordance with Caledonian substratum (Carlsson *et*
171 *al.*, 1999).

172 The low alpine belt is located up to 600 m a.s.l. and vegetation structure is very dependent on
173 micro-topography (snow cover duration, soil acidity, water availability, and sun and wind
174 exposures). As summer air temperatures dropped, the mountain birch forest-line gradually
175 disappeared and gave way to prostrate shrubs: patches of hydrophilic *Salix* sp., *Betula nana* mats
176 on snow-free areas, mosaics of *Vaccinium uliginosum*, *Empetrum nigrum* ssp. *hermaphroditum*,
177 *Arctostaphylos alpinus*. Species line of *Vaccinium myrtillus* was commonly found as the last
178 extent of the low alpine belt, and thus heavily dependent on the snow cover duration. Grass
179 heaths represented the typical vegetation landscape of the middle alpine belt. Poaceae,
180 Cyperaceae, and Juncaceae were pretty common (*Carex bigelowii*, *Festuca ovina*, *Juncus*
181 *trifidus*, *Luzula spicata*, etc.), as well as smaller dwarf shrubs (*Salix polaris*, *Phyllodoce*
182 *caerulea*). In the high alpine belt from 1100 m a.s.l., frost and the almost continuous presence of
183 snow cover prevented vascular plant growth. Plant cover was thus discontinuous or even absent.

184 Patches without snow were a favorable habitat for a very few species, such as *Salix herbacea* or
185 *Ranunculus glacialis*. Mats of mosses and lichens were particularly thick (Carlsson *et al.*, 1999).

186 All along the altitudinal gradient, mires were present where water, sediment, and organic
187 matter were accumulated. These were extended in large flat areas, particularly on Torneträsk
188 banks (**Figure 1**). Water table depth structured mire ecosystem: deep at the center, the water
189 depth was progressively replaced with sediments, organic matter, and vegetation such as
190 *Sphagnum* sp. On the edges, the birch forest rapidly disappeared into hydrophilous woody
191 species, grasses, and sedges, largely dependent on water depth in surface, including *Eriophorum*
192 *vaginatum*, *Rubus chamaemorus*, *Carex* sp. (Rydin *et al.*, 1999).

193 **2.2. Sampling protocol**

194 The sampling protocol applied for both soils and vegetation was aligned on the Landsat image
195 grid (30 m × 30 m). For representativeness purpose, three pixels (A, B, and C) were sampled
196 along the diagonal at BAD599, NJU383, and NJU975 (**Figure 2**). Only one pixel was considered
197 at TOR363.

198 **2.2.1. Soil sampling**

199 Soils of the study sites were between 30-cm and more than 100-cm deep. In this work, the top
200 soil (<1 m deep) was considered and collected using an auger in each corner of the pixels (a, c, e,
201 and g), plus a central sampling point P (orange dots, **Figure 2**). One sample, or two when
202 differentiated horizons occurred, were subsampled. Based on the organic carbon (OC) content,
203 we distinguished two horizon classes: organic (OC content > 5%) and mineral (OC content
204 < 5%).

205 **2.2.2. Vegetation sampling**

206 The sampling strategy was semi-stratified, taking account both the altitudinal gradient of
207 vegetation landscapes and the limited time of acquisition for floristic and edaphic data (Frontier,
208 1983). Each 900 m²-pixel was composed of two crossed transects (green lines, **Figure 2**),
209 themselves comprising 42 quadrats of 1 m². Two protocols have been followed, according to
210 vegetal physiognomy. For field layer vegetation (<50 cm height), floristic inventories were

211 conducted on vascular plants, mosses, and lichens. Their cover has been estimated by a
212 systematic sampling using a long needle imbedded in the herbaceous layer (every meter).
213 Vegetal species in contact with the needle were the main contributors of the vegetation layer.
214 The cover of each species (*sp*) was computed as follows: $\Sigma \text{contact}_{sp} \times 100/42$ needle-points
215 (Daget and Godron, 1995). When tree and shrub layers were encountered, the intercept cover
216 protocol was applied for vegetation up to 50 cm height (Canfield, 1941).

217 To estimate the influence of vegetation on geochemical distribution, percentages of vegetation
218 cover were computed for the five quadrats encompassing each soil sampling achieved (orange
219 dots, **Figure 2**). Then, vegetal taxa have been grouped into eight plant functional groups
220 following the tundra ecosystems key, adapted from Walker (2000): lichens, mosses, forbs,
221 graminoids, dwarf shrubs (deciduous and evergreen), shrubs (deciduous and evergreen), and
222 trees (deciduous and evergreen).

223 **2.3. Soil sample processing and chemical analyses**

224 Soil samples were dried (<40 °C in an oven for several days), sieved (<2 mm), and ground
225 (<200 mm) using a soil grinder (planetary ball mill PM200, Retsch, Haan, Germany). Soil pH
226 and conductivity were measured in the 0.25 mmol L⁻¹ CaCl₂ extraction solution (solution/soil
227 ratio of 1:10). The total dissolution of the samples was performed using a mixture of suprapure
228 acids (HNO₃ and HF) and H₂O₂. All cleaning and analytical procedures used high purity Milli-Q
229 water (18.2 MΩ cm). Approximately 100 mg of ground soil sample were dissolved using 0.5 mL
230 of 50% HF at 90 °C in a Savillex (Teflon bottle) for 24 h. Then, 1 mL of 65% HNO₃ was added
231 and kept at room temperature for 6 h before adding 1 mL of 30% H₂O₂ for 24 h at 40 °C for
232 evaporation. Finally, we added 1 mL of 65% HNO₃ for 24 h and solutions were diluted with
233 Milli-Q water to obtain a final acid concentration of 2% HNO₃ for ICP analysis. A set of 9 trace
234 elements (As, Cd, Co, Cr, Pb, Sb, Se, Sn, and V) was quantified using ICP–MS Agilent 7500 CE
235 (Agilent Technologies, Santa Clara, CA, USA) at the Alysés platform (Sorbonne Université) and
236 a second set of 6 trace elements (Ba, Cu, Mn, Ni, Sr, and Zn) using ICP–OES Agilent 5100
237 SVDV (Agilent Technologies, Santa Clara, CA, USA) at the ALIPP6 platform (Sorbonne
238 Université). In parallel, the concentrations of three major elements (Al, Fe, and Ti), considered

239 as geochemical tracers, were measured in bulk soil samples by XRF Niton XL3t (Thermo
240 Scientific, Billerica, MA USA) performed at IStEP (Sorbonne Université).

241 Limits of detection (LOD) and limits of quantification (LOQ) were calculated for each element
242 using 3- and 10-times, respectively, the standard deviation determined on the blank samples,
243 with the exception of XRF measurements. LOD were $<1000 \mu\text{g g}^{-1}$ for Al, $<40 \mu\text{g g}^{-1}$ for Fe,
244 $<20 \mu\text{g g}^{-1}$ for Ti, $<0.3 \mu\text{g g}^{-1}$ for Cu and Ni, $<0.2 \mu\text{g g}^{-1}$ for Sr and Zn, $<0.1 \mu\text{g g}^{-1}$ for Ba and
245 Se, $<0.02 \mu\text{g g}^{-1}$ for Co and Mn, $<0.01 \mu\text{g g}^{-1}$ for V, $<0.005 \mu\text{g g}^{-1}$ for As, Cd, Cr, and Sn,
246 $<0.002 \mu\text{g g}^{-1}$ for Sb, and $<0.001 \mu\text{g g}^{-1}$ for Pb. We then used $(\text{LOD} + \text{LOQ})/2$ for concentration
247 values below LOQ and $\text{LOD}/2$ for concentration values below LOD.

248 The procedure performance of the dissolution for ICP-MS and ICP-OES was checked for each
249 series of 35 samples by adding two replicates each of soil and sediment certified materials:
250 LKSD-1, CRM-033, BCR-146, BCR-277, and BCR-320. The average recovery
251 $(C_{\text{measured}}/C_{\text{certified}} \times 100)$ was calculated for each analyte: $100 \pm 5\%$ for As, Ba, Cd, Cu, Mn, Pb, V,
252 and Zn, $100 \pm 10\%$ for Se and Sr, $100 \pm 20\%$ for Sn, and $100 \pm 30\%$ for Co, Cr, Ni, and Sb. The
253 absence of contamination during the digestion procedure was checked through 2-4 blank
254 samples analyzed per series. A multi-element quality control samples ($1 \mu\text{g L}^{-1}$ standard) was
255 used every eight samples to correct the analytical deviation of the ICP-MS. Additionally, a
256 control liquid sample (CMR TMDA 64.2) was analyzed at the beginning of each series to check
257 the measurement quality (recovery between 90 and 110%).

258 In parallel, OC content was measured using a CHNS elemental analyzer (Elementar® vario
259 PYRO cube, Langenselbold, Germany) at the Institut d'écologie et des sciences de
260 l'environnement (CISE platform, iEES, Paris, France). Tyrosine was used as internal analytical
261 standard every four samples and analytical precision was estimated to 0.2%.

262 **2.4. Data processing and statistical analyses**

263 **2.4.1. Enrichment factor**

264 The enrichment factor (EF; Chester and Stoner, 1973; Vieira et al., 2004) was calculated for
265 each trace and major element (X) in organic soil horizon class samples using Al as normalized
266 element and mineral horizon class samples as reference material:

$$EF = \frac{(X/Al)_{\text{organic}}}{(X/Al)_{\text{mineral}}}$$

267

268 A comparison between Al and Ti as normalized element was made: it should be noted that values
269 with Al represented the maximum EFs.

270 2.4.2. Statistical analyses

271 All statistics were performed using R 3.5.1 (R Foundation for Statistical Computing, Vienna,
272 Austria) and RStudio 1.1.453 (RStudio Inc., Boston, Massachusetts, USA). Statistically
273 significant differences were tested using the non-parametric Kruskal-Wallis test ($\alpha = 0.05$) and a
274 post-hoc Dunn test ($\alpha = 0.05$) using the *dunn.test* package.

275 To identify the relationships between trace and major elements, a principal component analysis
276 (PCA) was performed with *FactoMineR* and *factoextra* packages on element concentrations after
277 centered log-ratio transformation using *rgr* package (*clr* function). Contributions of element
278 sources were estimated using a non-negative matrix factorization (NMF)—an innovative
279 statistical method recently used in environmental geochemistry (Christensen *et al.*, 2018)—with
280 *NMF* package (Gaujoux and Seoighe, 2010). The number of factors to best fit the model was
281 determined to be four. Finally, a partial least squares (PLS) regression was performed using
282 *plsdepot* package (*plsreg2* function) to determine the influence of environmental variables
283 (vegetation and soil parameters) on trace and major element distribution in organic horizon
284 classes.

285 3. Results and discussions

286 3.1. Occurrence of trace and major elements in subarctic soils

287 3.1.1. Concentration levels of Abisko area soils

288 Concentrations of trace and major elements were measured in both organic (OC content > 5%)
289 and mineral (OC content < 5%) soil horizon classes from Abisko (**Table 2**). Values showed a
290 wide range of median concentrations from Sb (0.14 $\mu\text{g g}^{-1}$) to Fe (20300 $\mu\text{g g}^{-1}$) in organic

291 horizon classes and from Cd ($0.23 \mu\text{g g}^{-1}$) to Al ($104000 \mu\text{g g}^{-1}$) in mineral horizon classes. This
292 concentration variability between elements is frequent in natural Arctic and subarctic
293 environments, such as in soil (Reimann *et al.*, 2015), snow (Shevchenko *et al.*, 2017), water
294 (Manasypov *et al.*, 2015), or vegetation (Wojtuń *et al.*, 2013). For each considered element,
295 concentrations were widely distributed: from 10- (Pb) to more than 1000-fold (Ni) between the
296 highest and the lowest values. Mineral horizon classes showed higher median values compared
297 to organic ones, particularly for Al, Sr, Ba, Mn, Cr, Sn, V, and Co (between 3- and more than 4-
298 fold). The opposite trend was only observed for Cu and Cd.

299 Compared to data from Nord-Trøndelag (Central Norway), a region also belonging to the
300 Fennoscandian range (Reimann *et al.*, 2015), the present data set showed generally higher
301 median element concentrations in both organic and mineral horizon classes (**Table 2**): on
302 average, 4- and 7-fold higher at Abisko, respectively. The maxima were observed for Ba and Sr
303 that could be explained by distinct lithologies between the two regions (West and East slope of
304 the Scandinavian Mountain). This was particularly obvious for V, Ti, Al, Fe, and Cr in organic
305 horizon classes (from 5- to 10-fold higher) and for Ba, Sr, Al, Cd, and Se for mineral horizon
306 classes (up to 32-fold). It should be mentioned that this difference can be largely attributed to the
307 preparation methods for chemical analyses: total HF dissolution in the present study (except for
308 Al, Fe, and Ti analyzed on the solid soil fraction by XRF) vs. aqua regia dissolution in Reimann
309 *et al.* (2015). Some elements showed opposite trend in organic soil horizon classes with lower
310 median concentrations observed at Abisko: Pb, Sb, Cd, Zn, and Sn. We suggest that either
311 Abisko was less atmospherically-influenced by these trace elements due to lower atmospheric
312 depositions, or that Abisko's distinct surface environment (such as vegetation) influenced metal
313 and metalloid recycling (Kabata-Pendias, 2004).

314 **3.1.2. Comparison of concentrations across subarctic ecosystems**

315 We compared concentrations measured in both organic and mineral soil horizon classes
316 between the different study sites. Four groups of elements were identified according to their
317 spatial and vertical distribution patterns (**Figure 3**). The first group included Al, Co, Cr, Fe, Mn,
318 Ni, Ti, and V (illustrated by Ti; **Figure 3A**) and showed statistically significantly higher
319 concentrations in mineral horizon classes. This element association was generally related to a

320 lithogenic origin (Halbach et al., 2017; Kabata-Pendias, 1993). The concentrations reported in
321 BAD599 were statistically significantly higher compared to other study sites. Because of the
322 complex silicate-rich local bedrock, it remains difficult to clearly identify the rock type and/or
323 minerals responsible for such high concentrations specifically observed in BAD599.

324 The second group of elements included Pb, Sb, Sn, and Zn (illustrated by Sn; **Figure 3B**).
325 These elements presented a similar pattern as previously observed (i.e., statistically significantly
326 higher concentrations in mineral horizon classes), with the exception of BAD599 that did not
327 show the same extremely high values in mineral horizon classes (compared to Ti for example,
328 **Figure 3A**). This indicates the absence of important mineral contribution related to the BAD599
329 lithology. Moreover, some of these elements were well known to be part of the long-range
330 atmospheric deposition even in the Arctic, such as Pb and Zn (McConnell and Edwards, 2008;
331 Shevchenko et al., 2003).

332 The third group of elements included As, Cd, Cu, and Se (illustrated by Se; **Figure 3C**) and
333 showed clearly distinct pattern compared to the two previous groups. Concentrations in mineral
334 horizon classes were either lower (NJU975), or higher (NJU383) than those measured in organic
335 horizon classes, while concentrations were constant across the study sites in organic horizon
336 classes. This implies the absence of major bedrock influence for these elements. Dynamics of
337 As, Cu, and Se are known to be largely controlled by soil oxidoreduction and pH (Masscheleyn
338 et al., 1991; Shaheen et al., 2013), parameters largely variables in organic-rich and temporarily
339 water-saturated soils. These elements are also influenced by biological activities, such as
340 methylation processes (Carbonell et al., 1998; Winkel et al., 2015). Cadmium was already
341 observed abundantly in surface soil horizons because of its strong dependence on atmospheric
342 deposition (Halbach et al., 2017).

343 Finally, the fourth group of elements included two alkaline earth metals: Ba and Sr (illustrated
344 by Sr; **Figure 3D**). This association presented a very specific behavior with high concentrations
345 in mineral horizon classes in BAD599, NJU383, and possibly TOR363 (only one data point in
346 TOR363). These two elements are well known to be chemically similar to Ca, facilitating
347 substitutions (Lucas et al., 1990). Indeed, BAD599 was characterized by a calc-silicate bedrock
348 and NJU383 was overhung by a limestone deposit 300–400 m higher. A dilution process may

349 explain the fact that Sr and Ba were weakly concentrated in organic soil horizon classes from
350 BAD599, while NJU383 should be affected by carbonate-rich dust coming from the top of the
351 mountain. The influence of carbonates, however, was limited given the acidic soil pH (**Table 1**).

352 **3.2. Sources of trace and major elements in subarctic soils**

353 **3.2.1. Enrichment of elements**

354 The EF is frequently used to identify natural vs. anthropogenic origins (Agnan *et al.*, 2015;
355 Reimann and de Caritat, 2005). In this study, we only considered organic soil horizon classes and
356 compared to mineral ones for identifying surface inputs (i.e., atmospheric deposition, vegetation,
357 etc.) after lithologic normalization. This method makes study sites comparable by eliminating the
358 local lithology influence. Results, presented for each site on a logarithmic scale, were compared
359 to the reference value of 1 (**Figure 4**): enrichment if $EF > 1$ and depletion if $EF < 1$. Because EFs
360 calculated using Al as normalized element were higher compared to those using Ti, we used the
361 threshold of $EF = 2$ for considering an enrichment.

362 Antimony was the only element showing a depletion in both BAD599 and TOR363 (on
363 average close to 0.1 and up to <0.01 for some samples). This may result from either an Al
364 enrichment in organic horizon classes (that could induce depletion of other elements), or a Sb-
365 rich lithology attenuating the low surface inputs in these two aforementioned sites. Six elements
366 (Cr, Mn, Ni, Sn, Ti, and V), most of them previously included in the first group (**Figure 3A**), did
367 not show enrichment ($EF < 2$) in the four study sites (with the exception of Mn in NJU383 and
368 Cr and V in TOR363). This corroborates the main lithogenic origin for these elements.
369 Considering the lithology normalization, Ba and Sr only showed enrichment in NJU975 that
370 probably reflects a local atmospheric deposition of carbonate-rich dust from the nearby limestone
371 deposit despite the absence of such minerals in the soil profile. This also illustrates the lack of Ba
372 and Sr anomalies in BAD599 and NJU383, as observed in **Figure 3D**.

373 The lowest EF values were observed in BAD599, while Fe was slightly enriched in BAD599
374 and TOR363 (EF between 3 and 4, respectively). In the same way, average EFs of Co, Pb, and
375 Zn were >3 , with important site heterogeneity. The highest enrichments were observed in
376 NJU975 and BAD599, and to a lesser extent in TOR363 (up to $EF > 10$). This illustrates the

377 large influence of surface processes, even for Co previously characterized as lithogenic element
378 (**Figure 3A**). Overall, the specific enrichment of Cu, Fe, and Pb observed in BAD599 can be
379 related to direct atmospheric influences from Sjangeli mining area (from southwestern prevailing
380 winds). Conversely, NJU975 presented the highest enrichments of Co, Mn, and Zn that we may
381 attribute to a regional influence of Viscaria mining area (from eastern winds).

382 Finally, As, Cd, Cu, and Se showed the highest enrichments (up to >100 for Cd and Cu).
383 Statistically significant differences were evidenced between sites, with the same trend for As,
384 Cu, and Se: BAD599 > TOR363 > NJU975 > NJU383. This indicates distinct intensities for
385 surface processes responsible for the presence of these chemicals (factor of 8–10 for As and Se
386 and up to 44 for Cu between BAD599 and NJU383). Cadmium, however, only presented lower
387 EFs in TOR363 (median EF of 4), while median EF reached 6–10 for the three other sites. Since
388 the EF cannot fully distinguish the origins of elements (e.g., mining, long-range atmospheric
389 deposition, vegetation), we suggest the use of multivariate analyses to identify the distinct
390 sources and quantify their contribution.

391 **3.2.2. Principal component analysis: covariance between elements**

392 A PCA was thus performed on trace and major element concentrations measured in Abisko soil
393 samples to identify the covariance between elements. The first three components explained 64%
394 of the data variance. We represented both observations (soil samples) and variables (elements) in
395 biplot diagrams (**Figure 5**). Principal component 1 (28% of the data variance) allowed the
396 grouping of Al, Cr, Sn, Ti, and V with positive scores and As, Cd, Cu, and Se with negative ones
397 (**Figure 5A**). This principal component was mainly driven by soil horizon classes considered
398 (**Figure 5B**): mineral samples with positive scores vs. organic samples with negatives scores.
399 This demonstrates that the nature of horizon class represented the first driver for the distribution
400 of trace and major elements. We previously discussed the lithogenic origin of the first group
401 (**Figure 3A**), including Al, Cr, Ti, and V. Surprisingly, Fe was not included in principal
402 component 1, despite its high concentrations in mineral samples (**Table 2**), as frequently
403 observed in the literature (Reimann *et al.*, 2007). At the opposite end, As, Cd, Cu, and Se were
404 already highlighted as part of the third group related to surface processes, including both
405 biological influence and atmospheric deposition (**Figure 3C**).

406 Principal component 2 (24% of the data variance) associated As, Fe, and Pb with positive
407 scores and Mn, Ni, and Sb with negative scores (**Figure 5A**). This principal component seemed
408 to be partly controlled by the location: in organic horizon class samples (i.e., negative scores of
409 principal component 1), BAD599 and TOR363 were mostly located in the positive scores while
410 NJU383 and NJU975 were mostly in the negatives scores (**Figure 5C**). This grouping was not
411 observed in mineral horizon classes and probably resulted from surface processes, such as
412 different atmospheric inputs. Indeed, as previously discussed, geographical features may explain
413 this site differentiation: NJU383 and NJU975 were both located on the same mountain slope with
414 similar eastern exposure, promoting the influence of East mining areas (e.g., Viscaria-
415 Sautusvaara). Due to the low influence of Fe, we assume that Kiruna mining area was either too
416 far, or not adequately located to atmospherically transport contaminants to the study area.
417 Conversely, BAD599, and to a lesser extent TOR363, were under the influence of South-West
418 winds (e.g., Sjangeli mining area), which are sources of Cu, Fe, and Pb, among other elements,
419 resulting in the presence of these chemicals in principal component 2. It should be noted that this
420 mixed source of Fe may explain the absence of Fe in the lithogenic element group.

421 Finally, principal component 3 (12% of the data variance) only highlighted two elements
422 previously grouped together: Ba and Sr (**Figure 5D**). We assume that this resulted from
423 carbonate-rich dust in BAD599 and NJU383, as previously discussed (**Figure 3D**). Considering
424 the acidic soil pH (mineral soil pH ≤ 6.6), we assume that this source was limited in the overall
425 geochemical composition. In the next section, we present the NMF analysis to evaluate the
426 contribution of the various sources to each single element measured.

427 **3.2.3. Non-negative matrix factorization: contribution of different sources**

428 The NMF, recently applied to environmental geochemistry (Christensen et al., 2018), was
429 performed to identify the contribution of different sources to trace and major element
430 concentrations. The relative contribution of the four factors selected are represented for each
431 element (**Figure 6A**) and sampling site by soil horizon class (**Figure 6B**). For clarity in the
432 graphical representations, we reordered the factors following the discussion.

433 First, factor 1 contributed significantly (>35%) to Al, Cr, Fe, Sn, Sr, Ti, and V (**Figure 6A**).
434 With high prevalence in mineral horizon classes (on average, 42 vs. only 17% in organic ones;

435 **Figure 6B**), this factor likely represented a strong lithogenic source relatively homogeneously
436 distributed in the study area (Halbach et al., 2017), confirming the results obtained by EFs
437 (**Figure 4**) and PCA (**Figure 5**). The highest contribution in mineral horizon classes was
438 observed in BAD599 at 60%, corroborating the low EFs observed in this site for Cr, Mn, V, and
439 Ti (**Figure 4**).

440 Factor 3 showed a high contribution (>35%) to Ba, Cd, Pb, Mn, Sr, and Zn, including an
441 important inter-site heterogeneity with the highest contribution in NJU383 (36%) and the lowest
442 one in BAD599 (9%). Surprisingly, it was almost equally distributed between organic and
443 mineral horizon classes in all study sites. This illustrates a site specificity that might result from
444 different lithologies between Njulla (NJU383 and NJU975) and Baddosdievvá (BAD599)
445 Mountains. The complexity of geological terrain of the study area cannot confirm this
446 assumption. However, the important contrast between sites could result from distinct
447 atmospheric inputs: Njulla Mountain sites were mainly influenced by eastern winds (e.g., from
448 Viscaria mining area). Besides, considering the chemical association (e.g., Cd, Pb, Zn), we may
449 suggest that factor 3 also constituted a source for global long-range atmospheric deposition
450 (Nygård et al., 2012). We thus attribute this factor to a mixed origin mainly driven by both
451 regional mining influence and global atmospheric deposition.

452 Factor 4 contributed importantly (>35%) to Cr, Cu, Fe, Mn, Ni, Sb, V, and Zn. The
453 contribution decreased in the following order: NJU975 > BAD599 > NJU383 > TOR363. On
454 average, mineral horizon classes had higher contribution than organic ones (28 vs. 22%,
455 respectively). In NJU975, factor 4 accounted for almost half of the total contribution in mineral
456 horizon classes. Considering the geochemical composition (Cu, Fe, Pb), we attribute factor 4 to a
457 local mining influence (e.g., from southwestern Sjangeli mining area), also illustrated in the
458 positive scores of the principal component 2 (**Figure 5A**). Since the contribution was important
459 in mineral horizon classes, we assume that factor 4 was also associated with the local lithology
460 particularly enriched in several base metals (Reimann et al., 2015).

461 Finally, factor 2 showed dominant contribution (>35%) to As, Cd, Cu, and Se. This factor
462 contributed 38% in organic horizon classes (mainly in BAD599 and TOR363 with 53 and 39%,
463 respectively) and only 7% in mineral ones. The differing contribution between the horizon

464 classes could suggest surface process influence being captured by factor 2 (**Figure 3C** and
465 **Figure 5A**). All of these elements were also reported to be accumulated in organic horizons in
466 European boreal forests due to their atmospheric deposition and their strong link to soil organic
467 matter (Gustafsson and Johnsson, 1992; Räisänen *et al.*, 1997). To better constrain the influence
468 of vegetation on elements distribution, we now propose a covariance analysis between
469 geochemical and vegetation data using a PLS regression.

470 **3.3. Influence of vegetation on trace and major element distribution**

471 Vegetation was sampled in each study site, corresponding to distinct ecosystems and land
472 covers according to decreasing altitude: grassland (NJU975), moor (BAD599), broad-leaved
473 forest (NJU383), and peat bog (TOR363). Following the Walker (2000) classification for Arctic
474 and subarctic vegetation, we adapted and simplified the classes to: trees, shrubs, dwarf shrubs,
475 graminoids, forbs, mosses, and lichens. We then distinguished deciduous and evergreen
476 vegetation for trees, shrubs, and dwarf shrubs. Vegetation classes were heterogeneously
477 distributed across the sites with dominance of dwarf shrubs (42%) in BAD599, forbs (42%) in
478 NJU383, and graminoids (87%) in TOR363 (**Table 3**). In NJU975, dwarf shrubs and graminoids
479 were almost equal proportions (22–26%). Trees were only found in NJU383 at a paltry 5% for
480 both deciduous (*Betula pubescens* ssp. *czerepanovii* and *Sorbus aucuparia*) and evergreen
481 (*Juniperus communis*) species. Shrubs and dwarf shrubs were mostly found in NJU975 and
482 BAD599. Only deciduous shrubs (*Betula nana* and *Salix phylicifolia*) were collected, while both
483 deciduous (e.g., *Vaccinium uliginosum*, *Rubus saxatilis*, *Salix herbacea*) and evergreen (e.g.,
484 *Empetrum nigrum* ssp. *hermaphroditum*, *Vaccinium vitis-idaea*, *Dryas octopetala*, *Loiseleuria*
485 *alpinum*) species were reported for dwarf shrubs. The graminoid species collected at Abisko
486 belonged to Cyperaceae (e.g., *Eriophorum vaginatum*, *Eleocharis acicularis*, *Carex vesicaria*),
487 Juncaceae (e.g., *Luzula* sp.), and Poaceae; the species *Carex vesicaria* was specifically found in
488 TOR363 with an important frequency. Forbs were highly found in NJU383, including *Epilobium*
489 *angustifolium*, *Trollius europaeus*, *Saussurea alpina*, *Myosotis* sp., *Viola biflora*, and *Paris*
490 *quadrifolia*. Finally, mosses were mostly found in NJU383 (21%) and lichens (e.g., *Cetraria*,
491 *Cladonia*) in NJU975 and BAD599 (16% in both cases).

492 We performed a PLS regression combining geochemical data (first three PCA components and
493 four NMF factors) as dependent variables with environmental data (frequency of vegetation
494 classes and soil pH and conductivity) as independent variables. The data set consists of
495 37 observations collected in each sampling point (9–13 per study site, except for TOR363). To
496 better understand the influence of vegetation, we only considered organic soil horizon classes
497 thereby limiting any lithogenic inputs. The first two axes represented 53% of the data variance
498 for independent variables and 23% for dependent variables.

499 The first axis of the correlation circle showed factor 3 (i.e., mixed influence, including
500 lithogenic, regional mining, and long-range atmospheric deposition) with positive scores and
501 principal component 2 (i.e., local mining influence) with negative ones (**Figure 7A**).
502 Surprisingly, factor 4 (i.e., local mining influence) was not correlated to principal component 2.
503 This can result from the opposite influence for principal components (positive scores vs. negative
504 ones), whereas factors corresponded to single sources. Vegetation classes that were influenced
505 by the first axis were: trees and forbs (positive scores) and shrubs, dwarf shrubs, and lichens
506 (negative scores). Because of the limited representativeness of trees in the study area, it is
507 difficult to correctly interpret their role in the geochemical distribution. Moreover, in NJU383,
508 pixels A and B differed from pixel C by forb and bryophyte proportions, and not by tree
509 proportions. This indicates that positive scores of the first axis were mainly driven by forbs. The
510 first axis clearly differentiated NJU383 with positive scores from BAD599 and NJU975 with
511 negative scores (**Figure 7B**), corresponding to their distinct vegetation classes. The fact that
512 NJU975 and NJU383 are presented diametrically despite their geochemically similar inputs
513 (**Figure 5** and **Figure 6**) supports the limited bedrock effect along the first axis. The annual
514 activity of forbs may contribute to the recycling of lithogenic elements (factors 1 and 3).
515 Conversely, perennial shrubs and dwarf shrubs present a higher bioaccumulation (Wojtuń *et al.*,
516 2013), and therefore a lower recycling because of the element retention. When differentiating
517 deciduous and evergreen species, results showed similar covariances as previously observed
518 (data not shown). We attribute this finding to a physiological effect (i.e., perennial vs. annual)
519 rather than a leaf biomass production effect (i.e., deciduous vs. evergreen). Finally, despite
520 similar phytosociological behaviors in the ecosystems, lichens and bryophytes, two classes of
521 vegetation well known to efficiently accumulate trace elements that permit to accurately evaluate

522 atmospheric deposition (Agnan *et al.*, 2015; Steinnes *et al.*, 2003), were opposed along the first
523 axis, probably resulting from distinct bioaccumulated elements between these two vegetation
524 classes (Gandois *et al.*, 2014).

525 The second axis of the correlation circle showed factor 2 (i.e., surface processes), and to a
526 lesser extent factor 4 (i.e., local mining influence), with negative scores and principal component
527 3 (i.e., calcareous lithology) with low positive scores (**Figure 7A**). Despite both lichens and
528 bryophytes were slightly influenced along the negative scores, soil pH appeared as the main
529 driver of the second axis, controlled by the sampling sites: the more acidic soil pH were observed
530 in BAD599, TOR363, pixels A and B of NJU383, and pixel C of NJU975 (**Figure 7B**). This
531 illustrates the higher element solubility in more acidic soils, promoting their mobilization
532 (Shaheen *et al.*, 2013). Conversely, the less acidic soils retained trace elements such as As, Cd,
533 and Cu. Selenium was also associated to factor 4 while its adsorption decreased with increasing
534 pH (Söderlund *et al.*, 2016). Thus, soil pH does not seem to be the only parameter that controls
535 the dynamics of these elements. This observation implies more complex surface processes that
536 we cannot identify in this study.

537 Following the results observed here, two hypotheses could be formulated: (1) vegetation
538 influenced the distribution through distinct bioaccumulation and recycling processes (Shahid *et al.*
539 *et al.*, 2017) or (2) soil geochemistry influenced the distribution of vegetation as an edaphic
540 variable (Thuiller, 2013). Both hypotheses seem possible following the two PLS axes (axes 1 and
541 2, respectively). Unfortunately, the influence of vegetation on trace element distribution is poorly
542 documented in the literature, particularly in Arctic and subarctic ecosystems. We also assume
543 that vegetation may be impacted by the altitudinal gradient (from 363 to 975 m a.s.l.) that likely
544 masked the edaphic influence. However, considering the findings from this study and the quick
545 changes in Arctic and subarctic vegetation composition recently observed at Abisko (Callaghan
546 *et al.*, 2013), we expect modifications in trace element cycling. This is of particular concern since
547 shrubs, one of the main vegetation groups that was correlated with trace elements, are known to
548 be currently expanding with climate change (Myers-Smith *et al.*, 2015). Because of herbivore
549 mammals in the high latitudes, such as reindeer, are largely dependent on vegetation as intake,
550 such modifications may impact the whole trophic network (Pacyna *et al.*, 2019). These

551 preliminary results, however, need to be confirmed, and trace element dynamics in the Arctic
552 and subarctic environments, better characterized in further research.

553 **4. Conclusions**

554 In this study, we measured trace and major element concentrations in both organic and mineral
555 soil horizon classes from four different subarctic land covers: grassland, moor, broad-leaved
556 forest, and peat bog. The main objectives were to: (1) identify the occurrence and sources of
557 elements and (2) evaluate the influence of vegetation on the element spatial distribution. Results
558 showed non-negligible concentrations that partially resulted from natural regional enrichments.
559 Several trends were observed according to the study site and the considered soil horizon class.
560 The association between elements showed a strong influence of the soil horizon class: lithogenic
561 (Al, Cr, Ti) vs. surface influenced (As, Cu, Se) elements. Atmospheric influences, including
562 contamination from local and regional mines (Cu, Fe, Mn, Ni, Pb), as well as long-range
563 atmospheric transport (Cd, Zn), were observed in surface organic horizon classes. Every source
564 identified was not exclusive for each trace and major element: a non-negative matrix
565 factorization allowed quantification of contribution of each single source. To our knowledge, this
566 study is the first to combine geochemical with ecological data in high latitude environments to
567 evaluate the covariance between vegetation and element concentrations using a partial least
568 square regression. Forbs and shrubs/dwarf shrubs were highlighted as the main vegetation
569 classes controlling the geochemical dynamics likely due to their annual vs. perennial activities,
570 respectively. In addition, the control of geochemical dynamics by soil pH could be evidenced for
571 As, Cu, and Se. We thus expect that the evolution of vegetation in Arctic and subarctic regions
572 related to climate change may modify the dynamics of trace elements, either directly by
573 accumulating elements from the atmosphere to the soil, or indirectly by modifying the soil pH,
574 and up to the whole trophic network through herbivore mammals. Further research is therefore
575 required to confirm the findings of the present study.

576 **Acknowledgements**

577 Thanks to ENVEXX team for sampling. We also thank Emmanuel Aubry for analytical
578 support, Benoit Caron for ICP–OES (ALIPP6), Florence le Cornec and Irina Djoureaev for ICP–
579 MS (ALIZE), and Laurence Le Callonnec and Nathalie Labourdette for XRF. The funding was
580 provided by Sorbonne Universités (Collège des licences, ENVEXX project) and element analysis
581 by TETRISE project (supported by the Needs Environment 2012 program, CNRS-ANDRA-
582 IRSN-EDF). We also would like to acknowledge the Abisko Research Station for their
583 hospitality and the scientific support. We thank Jimmy Ong for his proofreading of the
584 manuscript. We also thank two anonymous reviewers for their valuable comments to improve the
585 manuscript.

586 **References**

- 587 Acosta Navarro, J.C., Varma, V., Riipinen, I., Seland, Ø., Kirkevåg, A., Struthers, H., Iversen,
588 T., Hansson, H.-C., Ekman, A.M.L., 2016. Amplification of Arctic warming by past air
589 pollution reductions in Europe. *Nat. Geosci.* 9, 277–281.
590 <https://doi.org/10.1038/ngeo2673>
- 591 Agnan, Y., Séjalon-Delmas, N., Claustres, A., Probst, A., 2015. Investigation of spatial and
592 temporal metal atmospheric deposition in France through lichen and moss
593 bioaccumulation over one century. *Sci. Total Environ.* 529, 285–296.
594 <https://doi.org/10.1016/j.scitotenv.2015.05.083>
- 595 Agnan, Y., Séjalon-Delmas, N., Probst, A., 2014. Origin and distribution of rare earth elements
596 in various lichen and moss species over the last century in France. *Sci. Total Environ.*
597 487, 1–12. <https://doi.org/10.1016/j.scitotenv.2014.03.132>
- 598 Åkerman, H.J., Johansson, M., 2008. Thawing permafrost and thicker active layers in sub-arctic
599 Sweden. *Permafr. Periglac. Process.* 19, 279–292. <https://doi.org/10.1002/ppp.626>
- 600 AMAP, 2018. AMAP assessment 2018: biological effects of contaminants on Arctic wildlife and
601 fish. Arctic Monitoring and Assessment Programme (AMAP), Oslo.
- 602 AMAP, 2011. Snow, water, ice and permafrost in the Arctic (SWIPA): climate change and the
603 cryosphere. Arctic Monitoring and Assessment Programme (AMAP), Oslo.
- 604 Barbante, C., Veysseyre, A., Ferrari, C., Van De Velde, K., Morel, C., Capodaglio, G., Cescon,
605 P., Scarponi, G., Boutron, C., 2001. Greenland snow evidence of large scale atmospheric
606 contamination for platinum, palladium, and rhodium. *Environ. Sci. Technol.* 35, 835–
607 839. <https://doi.org/10.1021/es000146y>
- 608 Beck, P.S.A., Goetz, S.J., 2011. Satellite observations of high northern latitude vegetation
609 productivity changes between 1982 and 2008: ecological variability and regional
610 differences. *Environ. Res. Lett.* 6, 045501. <https://doi.org/10.1088/1748-9326/6/4/045501>

- 611 Berner, L.T., Beck, P.S.A., Bunn, A.G., Goetz, S.J., 2013. Plant response to climate change
612 along the forest-tundra ecotone in northeastern Siberia. *Glob. Change Biol.* 19, 3449–
613 3462. <https://doi.org/10.1111/gcb.12304>
- 614 Callaghan, T.V., Crawford, R.M.M., Eronen, M., Hofgaard, A., Payette, S., Rees, W.G., Skre,
615 O., Sveinbjörnsson, B., Vlassova, T.K., Werkman, B.R., 2002b. The dynamics of the
616 tundra-taiga boundary: an overview and suggested coordinated and integrated approach
617 to research. *Ambio Special Report* 12, 3–5.
- 618 Callaghan, T.V., Jonasson, C., Thierfelder, T., Yang, Z., Hedenas, H., Johansson, M., Molau, U.,
619 Van Bogaert, R., Michelsen, A., Olofsson, J., Gwynn-Jones, D., Bokhorst, S., Phoenix,
620 G., Bjerke, J.W., Tommervik, H., Christensen, T.R., Hanna, E., Koller, E.K., Sloan, V.L.,
621 2013. Ecosystem change and stability over multiple decades in the Swedish subarctic:
622 complex processes and multiple drivers. *Philos. Trans. R. Soc. B Biol. Sci.* 368,
623 20120488. <https://doi.org/10.1098/rstb.2012.0488>
- 624 Callaghan, T.V., Werkman, B.R., Crawford, Robert.M.M., 2002a. The tundra-taiga interface and
625 its dynamics: concepts and applications. *Ambio Special Report* 12, 6–14.
- 626 Canfield, R.H., 1941. Application of the line interception method in sampling range vegetation.
627 *J. For.* 39, 388–394. <https://doi.org/10.1093/jof/39.4.388>
- 628 Carbonell, A.A., Aarabi, M.A., DeLaune, R.D., Gambrell, R.P., Patrick Jr, W.H., 1998. Arsenic
629 in wetland vegetation: availability, phytotoxicity, uptake and effects on plant growth and
630 nutrition. *Sci. Total Environ.* 217, 189–199. [https://doi.org/10.1016/S0048-](https://doi.org/10.1016/S0048-9697(98)00195-8)
631 [9697\(98\)00195-8](https://doi.org/10.1016/S0048-9697(98)00195-8)
- 632 Carlsson, B.Å., Karlsson, P.S., Svensson, B.M., 1999. 6. Alpine and subalpine vegetation, in:
633 Rydin, H., Snoeijs, P., Diekmann, M. (Eds.), *Swedish Plant Geography, Acta*
634 *Phytogeographica Suecica*. Svenska Växtgeografiska Sällskapet, Uppsala, pp. 75–89.
- 635 Chapin, F.S.I., Matson, P.A., Vitousek, P., 2011. *Principles of terrestrial ecosystem ecology*.
636 Springer, New York.
- 637 Chester, R., Stoner, J.H., 1973. Pb in particulates from the lower atmosphere of the Eastern
638 Atlantic. *Nature* 245, 27–28. <https://doi.org/10.1038/245027b0>
- 639 Christensen, E.R., Steinnes, E., Eggen, O.A., 2018. Anthropogenic and geogenic mass input of
640 trace elements to moss and natural surface soil in Norway. *Sci. Total Environ.* 613–614,
641 371–378. <https://doi.org/10.1016/j.scitotenv.2017.09.094>
- 642 Courault, R., Cohen, M., Ronchail, J., 2015. Régimes de circulation atmosphérique, impact du
643 changement climatique et variation démographique des rennes dans le nord de la
644 Scandinavie, in: *Modélisations et Variabilités : Actes Du XXVIII^e Colloque de*
645 *l'Association Internationale de Climatologie*. Presented at the XXVIII^e Colloque de
646 *l'Association Internationale de Climatologie*, Liège, Belgium, pp. 123–128.
- 647 Daget, P., Godron, M., 1995. *Pastoralisme. Troupeaux, espaces et sociétés*. Hatier, Aupelf, Uref,
648 Universités francophones, Paris.
- 649 Davies, C.E., Moss, D., Hill, M.O., 2004. EUNIS habitat classification revised 2004. European
650 Topic Centre on Nature Protection and Biodiversity, Paris.

- 651 Dietz, R., Riget, F., Cleemann, M., Aarkrog, A., Johansen, P., Hansen, J., 2000. Comparison of
652 contaminants from different trophic levels and ecosystems. *Sci. Total Environ.* 245, 221–
653 231. [https://doi.org/10.1016/S0048-9697\(99\)00447-7](https://doi.org/10.1016/S0048-9697(99)00447-7)
- 654 Eilu, P. (Ed.), 2012. Mineral deposits and metallogeny of Fennoscandia, Special paper /
655 Geological Survey of Finland. Geological Survey of Finland, Espoo.
- 656 European Environment Agency, 2014. CLC2012: Addendum to CLC2006 technical guidelines.
657 Office for Official Publications of the European communities, Luxembourg.
- 658 Evenset, A., Christensen, G.N., Carroll, J., Zaborska, A., Berger, U., Herzke, D., Gregor, D.,
659 2007. Historical trends in persistent organic pollutants and metals recorded in sediment
660 from Lake Ellasjøen, Bjørnøya, Norwegian Arctic. *Environ. Pollut.* 146, 196–205.
661 <https://doi.org/10.1016/j.envpol.2006.04.038>
- 662 Fedele, L., Plant, J.A., Vivo, B.D., Lima, A., 2008. The rare earth element distribution over
663 Europe: geogenic and anthropogenic sources. *Geochem. Explor. Environ. Anal.* 8, 3–18.
664 <https://doi.org/10.1144/1467-7873/07-150>
- 665 Fick, S.E., Hijmans, R.J., 2017. WorldClim 2: new 1-km spatial resolution climate surfaces for
666 global land areas. *Int. J. Climatol.* 37, 4302–4315. <https://doi.org/10.1002/joc.5086>
- 667 Frontier, S., 1983. *Stratégies d'échantillonnage en écologie*. Masson, Paris.
- 668 Gamberg, M., Braune, B., Davey, E., Elkin, B., Hoekstra, P.F., Kennedy, D., Macdonald, C.,
669 Muir, D., Nirwal, A., Wayland, M., Zeeb, B., 2005. Spatial and temporal trends of
670 contaminants in terrestrial biota from the Canadian Arctic. *Sci. Total Environ.* 351–352,
671 148–164. <https://doi.org/10.1016/j.scitotenv.2004.10.032>
- 672 Gamberg, M., Cuyler, C., Wang, X., 2016. Contaminants in two West Greenland caribou
673 populations. *Sci. Total Environ.* 554–555, 329–336.
674 <https://doi.org/10.1016/j.scitotenv.2016.02.154>
- 675 Gandois, L., Agnan, Y., Leblond, S., Séjalon-Delmas, N., Le Roux, G., Probst, A., 2014. Use of
676 geochemical signatures, including rare earth elements, in mosses and lichens to assess
677 spatial integration and the influence of forest environment. *Atmos. Environ.* 95, 96–104.
678 <https://doi.org/10.1016/j.atmosenv.2014.06.029>
- 679 Gaujoux, R., Seoighe, C., 2010. A flexible R package for nonnegative matrix factorization. *BMC*
680 *Bioinformatics* 11, 367. <https://doi.org/10.1186/1471-2105-11-367>
- 681 Gustafsson, J.P., Johnsson, L., 1992. Selenium retention in the organic matter of Swedish forest
682 soils. *J. Soil Sci.* 43, 461–472. <https://doi.org/10.1111/j.1365-2389.1992.tb00152.x>
- 683 Halbach, K., Mikkelsen, Ø., Berg, T., Steinnes, E., 2017. The presence of mercury and other
684 trace metals in surface soils in the Norwegian Arctic. *Chemosphere* 188, 567–574.
685 <https://doi.org/10.1016/j.chemosphere.2017.09.012>
- 686 Herndon, E.M., Jin, L., Andrews, D.M., Eissenstat, D.M., Brantley, S.L., 2015. Importance of
687 vegetation for manganese cycling in temperate forested watersheds. *Glob. Biogeochem.*
688 *Cycles* 29, 2014GB004858. <https://doi.org/10.1002/2014GB004858>

- 689 Ilyin, I., Rozovskaya, O., Travnikov, O., Aas, W., 2017. Assessment of heavy metal
690 transboundary pollution on regional and national scales, transition to the new EMEP grid
691 (EMEP Status Report 2/2017). Meteorological Synthesizing Centre-East & Norwegian
692 Institute for Air Research, Moscow - Kjeller.
- 693 IPCC, 2013. Climate Change 2013: the physical science basis. Contribution of the Working
694 Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate
695 Change. Cambridge University Press, Cambridge - New York.
- 696 Ju, J., Masek, J.G., 2016. The vegetation greenness trend in Canada and US Alaska from 1984–
697 2012 Landsat data. *Remote Sens. Environ.* 176, 1–16.
698 <https://doi.org/10.1016/j.rse.2016.01.001>
- 699 Kabata-Pendias, A., 2010. Trace elements in soils and plants, 4th ed. CRC Press.
- 700 Kabata-Pendias, A., 2004. Soil–plant transfer of trace elements—an environmental issue.
701 *Geoderma*, Biogeochemical processes and the role of heavy metals in the soil
702 environment 122, 143–149. <https://doi.org/10.1016/j.geoderma.2004.01.004>
- 703 Kabata-Pendias, A., 1993. Behavioural properties of trace metals in soils. *Appl. Geochem.*,
704 *Environmental Geochemistry* 8, 3–9. [https://doi.org/10.1016/S0883-2927\(09\)80002-4](https://doi.org/10.1016/S0883-2927(09)80002-4)
- 705 Karger, D.N., Conrad, O., Böhrer, J., Kawohl, T., Kreft, H., Soria-Auza, R.W., Zimmermann,
706 N.E., Linder, H.P., Kessler, M., 2017. Climatologies at high resolution for the earth’s
707 land surface areas. *Sci. Data* 4, 170122. <https://doi.org/10.1038/sdata.2017.122>
- 708 Klassen, R.A., Douma, S., Rencz, A.N., 2010. Environmental and human health risk assessment
709 for essential trace elements: considering the role for geoscience. *J. Toxicol. Environ.*
710 *Health A* 73, 242–252. <https://doi.org/10.1080/15287390903340906>
- 711 Law, K.S., Stohl, A., 2007. Arctic air pollution: origins and impacts. *Science* 315, 1537–1540.
712 <https://doi.org/10.1126/science.1137695>
- 713 Lucas, J., El Faleh, E.M., Prevot, L., 1990. Experimental study of the substitution of Ca by Sr
714 and Ba in synthetic apatites. *Geol. Soc. Lond. Spec. Publ.* 52, 33–47.
715 <https://doi.org/10.1144/GSL.SP.1990.052.01.04>
- 716 Manasypov, R.M., Vorobyev, S.N., Loiko, S.V., Kritzkov, I.V., Shirokova, L.S., Shevchenko,
717 V.P., Kirpotin, S.N., Kulizhsky, S.P., Kolesnichenko, L.G., Zemtsov, V.A., Sinkinov,
718 V.V., Pokrovsky, O.S., 2015. Seasonal dynamics of organic carbon and metals in
719 thermokarst lakes from the discontinuous permafrost zone of western Siberia.
720 *Biogeosciences* 12, 3009–3028. <https://doi.org/10.5194/bg-12-3009-2015>
- 721 Masscheleyn, P.H., Delaune, R.D., Patrick, W.H., 1991. Arsenic and selenium chemistry as
722 affected by sediment redox potential and pH. *J. Environ. Qual.* 20, 522–527.
723 <https://doi.org/10.2134/jeq1991.00472425002000030004x>
- 724 McConnell, J.R., Edwards, R., 2008. Coal burning leaves toxic heavy metal legacy in the Arctic.
725 *Proc. Natl. Acad. Sci.* 105, 12140–12144. <https://doi.org/10.1073/pnas.0803564105>
- 726 Myers-Smith, I.H., Elmendorf, S.C., Beck, P.S.A., Wilmking, M., Hallinger, M., Blok, D., Tape,
727 K.D., Rayback, S.A., Macias-Fauria, M., Forbes, B.C., Speed, J.D.M., Boulanger-

- 728 Lapointe, N., Rixen, C., Lévesque, E., Schmidt, N.M., Baittinger, C., Trant, A.J.,
729 Hermanutz, L., Collier, L.S., Dawes, M.A., Lantz, T.C., Weijers, S., Jørgensen, R.H.,
730 Buchwal, A., Buras, A., Naito, A.T., Ravolainen, V., Schaepman-Strub, G., Wheeler,
731 J.A., Wipf, S., Guay, K.C., Hik, D.S., Vellend, M., 2015. Climate sensitivity of shrub
732 growth across the tundra biome. *Nat. Clim. Change* 5, 887–891.
733 <https://doi.org/10.1038/nclimate2697>
- 734 Nygård, T., Steinnes, E., Royset, O., 2012. Distribution of 32 elements in organic surface soils:
735 contributions from atmospheric transport of pollutants and natural sources. *Water. Air.*
736 *Soil Pollut.* 223, 699–713. <https://doi.org/10.1007/s11270-011-0895-5>
- 737 Obrist, D., Agnan, Y., Jiskra, M., Olson, C.L., Colegrove, D.P., Hueber, J., Moore, C.W., Sonke,
738 J.E., Helmig, D., 2017. Tundra uptake of atmospheric elemental mercury drives Arctic
739 mercury pollution. *Nature* 547, 201–204. <https://doi.org/10.1038/nature22997>
- 740 Pacyna, A.D., Frankowski, M., Koziół, K., Węgrzyn, M.H., Wietrzyk-Pełka, P., Lehmann-
741 Konera, S., Polkowska, Ż., 2019. Evaluation of the use of reindeer droppings for
742 monitoring essential and non-essential elements in the polar terrestrial environment. *Sci.*
743 *Total Environ.* 658, 1209–1218. <https://doi.org/10.1016/j.scitotenv.2018.12.232>
- 744 Räisänen, M.L., Kashulina, G., Bogatyrev, I., 1997. Mobility and retention of heavy metals,
745 arsenic and sulphur in podzols at eight locations in northern Finland and Norway and the
746 western half of the Russian Kola Peninsula. *J. Geochem. Explor.* 59, 175–195.
747 [https://doi.org/10.1016/S0375-6742\(97\)00014-9](https://doi.org/10.1016/S0375-6742(97)00014-9)
- 748 Reimann, C., Arnoldussen, A., Englmaier, P., Filzmoser, P., Finne, T.E., Garrett, R.G., Koller,
749 F., Nordgulen, O., 2007. Element concentrations and variations along a 120-km transect
750 in southern Norway - Anthropogenic vs. geogenic vs. biogenic element sources and
751 cycles. *Appl. Geochem.* 22, 851–871. <https://doi.org/Article>
- 752 Reimann, C., de Caritat, P., 2005. Distinguishing between natural and anthropogenic sources for
753 elements in the environment: regional geochemical surveys versus enrichment factors.
754 *Sci. Total Environ.* 337, 91–107.
- 755 Reimann, C., Schilling, J., Roberts, D., Fabian, K., 2015. A regional-scale geochemical survey of
756 soil O and C horizon samples in Nord-Trøndelag, Central Norway: Geology and mineral
757 potential. *Appl. Geochem.* 61, 192–205.
758 <https://doi.org/10.1016/j.apgeochem.2015.05.019>
- 759 Ren, Z.-L., Tella, M., Bravin, M.N., Comans, R.N.J., Dai, J., Garnier, J.-M., Sivry, Y., Doelsch,
760 E., Straathof, A., Benedetti, M.F., 2015. Effect of dissolved organic matter composition
761 on metal speciation in soil solutions. *Chem. Geol.* 398, 61–69.
762 <https://doi.org/10.1016/j.chemgeo.2015.01.020>
- 763 Romanovsky, V.E., Smith, S.L., Christiansen, H.H., 2010. Permafrost thermal state in the polar
764 Northern Hemisphere during the international polar year 2007-2009: a synthesis.
765 *Permafr. Periglac. Process.* 21, 106–116. <https://doi.org/10.1002/ppp.689>

- 766 Rydin, H., Sjörs, H., Löfroth, M., 1999. 7. Mires, in: Rydin, H., Snoeijs, P., Diekmann, M.
767 (Eds.), *Swedish Plant Geography, Acta Phytogeographica Suecica*. Svenska
768 Växtgeografiska Sällskapet, Uppsala, pp. 91–112.
- 769 Shaheen, S.M., Tsadilas, C.D., Rinklebe, J., 2013. A review of the distribution coefficients of
770 trace elements in soils: Influence of sorption system, element characteristics, and soil
771 colloidal properties. *Adv. Colloid Interface Sci.* 201–202, 43–56.
772 <https://doi.org/10.1016/j.cis.2013.10.005>
- 773 Shahid, M., Dumat, C., Khalid, S., Schreck, E., Xiong, T., Niazi, N.K., 2017. Foliar heavy metal
774 uptake, toxicity and detoxification in plants: A comparison of foliar and root metal
775 uptake. *J. Hazard. Mater.* 325, 36–58. <https://doi.org/10.1016/j.jhazmat.2016.11.063>
- 776 Shevchenko, V., Lisitzin, A., Vinogradova, A., Stein, R., 2003. Heavy metals in aerosols over
777 the seas of the Russian Arctic. *Sci. Total Environ.* 306, 11–25.
778 [https://doi.org/10.1016/S0048-9697\(02\)00481-3](https://doi.org/10.1016/S0048-9697(02)00481-3)
- 779 Shevchenko, V.P., Pokrovsky, O.S., Vorobyev, S.N., Krickov, I.V., Manasyrov, R.M., Politova,
780 N.V., Kopysov, S.G., Dara, O.M., Auda, Y., Shirokova, L.S., Kolesnichenko, L.G.,
781 Zemtsov, V.A., Kirpotin, S.N., 2017. Impact of snow deposition on major and trace
782 element concentrations and elementary fluxes in surface waters of the Western Siberian
783 Lowland across a 1700 km latitudinal gradient. *Hydrol. Earth Syst. Sci.* 21, 5725–5746.
784 <https://doi.org/10.5194/hess-21-5725-2017>
- 785 Shiyatov, S.G., Terent'ev, M.M., Fomin, V.V., Zimmermann, N.E., 2007. Altitudinal and
786 horizontal shifts of the upper boundaries of open and closed forests in the Polar Urals in
787 the 20th century. *Russ. J. Ecol.* 38, 223–227.
788 <https://doi.org/10.1134/S1067413607040017>
- 789 Söderlund, M., Virkanen, J., Holgersson, S., Lehto, J., 2016. Sorption and speciation of selenium
790 in boreal forest soil. *J. Environ. Radioact.* 164, 220–231.
791 <https://doi.org/10.1016/j.jenvrad.2016.08.006>
- 792 Steinnes, E., Berg, T., Sjobakk, T.E., 2003. Temporal trends in long-range atmospheric transport
793 of heavy metals to Norway. *J. Phys. IV Proc.* 107, 1271–1273.
794 <https://doi.org/10.1051/jp4:20030532>
- 795 Steinnes, E., Lierhagen, S., 2018. Geographical distribution of trace elements in natural surface
796 soils: Atmospheric influence from natural and anthropogenic sources. *Appl. Geochem.*
797 88, 2–9. <https://doi.org/10.1016/j.apgeochem.2017.03.013>
- 798 Thuiller, W., 2013. On the importance of edaphic variables to predict plant species distributions -
799 limits and prospects. *J. Veg. Sci.* 24, 591–592. <https://doi.org/10.1111/jvs.12076>
- 800 Tyler, G., Pålsson, A.-M.B., Bengtsson, G., Bååth, E., Tranvik, L., 1989. Heavy-metal ecology
801 of terrestrial plants, microorganisms and invertebrates: A review. *Water. Air. Soil Pollut.*
802 47, 189–215. <https://doi.org/10.1007/BF00279327>
- 803 Ulrich, B., Pankrath, J., 1983. *Effects of accumulation of air pollutants in forest ecosystems.*
804 Springer, Dordrecht.

- 805 Vieira, B.J., Freitas, M.C., Rodrigues, A.F., Pacheco, A.M.G., Soares, P.M., Correia, N., 2004.
806 Element-enrichment factors in lichens from Terceira, Santa Maria and Madeira Islands
807 (Azores and Madeira archipelagoes). *J. Atmospheric Chem.* 49, 231–249.
808 <https://doi.org/Proceedings Paper>
- 809 Walker, D.A., 2000. Hierarchical subdivision of Arctic tundra based on vegetation response to
810 climate, parent material and topography. *Glob. Change Biol.* 6, 19–34.
811 <https://doi.org/10.1046/j.1365-2486.2000.06010.x>
- 812 Winkel, L., Vriens, B., Jones, G., Schneider, L., Pilon-Smits, E., Bañuelos, G., 2015. Selenium
813 cycling across soil-plant-atmosphere interfaces: a critical review. *Nutrients* 7, 4199–4239.
814 <https://doi.org/10.3390/nu7064199>
- 815 Wojtuń, B., Samecka-Cymerman, A., Kolon, K., Kempers, A.J., Skrzypek, G., 2013. Metals in
816 some dominant vascular plants, mosses, lichens, algae, and the biological soil crust in
817 various types of terrestrial tundra, SW Spitsbergen, Norway. *Polar Biol.* 36, 1799–1809.
818 <https://doi.org/10.1007/s00300-013-1399-0>
- 819
- 820

821 **List of tables**

822	Table 1. Environmental characteristics of the four study sites.	34
823	Table 2. Summary of trace and major element concentrations in organic (OC content > 5%) and	
824	mineral (OC content < 5%) soil horizon classes and comparison with soil concentrations from	
825	Nord-Trøndelag (Central Norway).	35
826	Table 3. Proportions (in %) of vegetation classes following the adapted Walker (2000)	
827	classification in the four study sites. Both deciduous and evergreen were identified for trees,	
828	shrubs, and dwarf shrubs.....	36
829		

830 **Table 1.** Environmental characteristics of the four study sites.

	NJU975	BAD599	NJU383	TOR363
latitude/longitude	68.363°N / 18.716°E	68.317°N / 18.867°E	68.355°N / 18.763°E	68.349°N / 19.057°E
elevation (m a.s.l.)¹	975	599	383	363
slope aspect / steepness¹	SE / ~3.25°	SW / ~1°	ESE / ~2°	N / ~1°
mean annual air temperature (1979–2013; °C)²	–3.2	–1.2	–0.1	–0.3
precipitation (1979–2013; mm a⁻¹)²	602	449	413	402
average wind speed (1970–2000; m s⁻¹)³	3.2	2.7	2.5	2.3
bedrock	mica-rich metamorphic rocks	calc-silicate rocks	mica-rich metamorphic rocks	quartz-feldspar-rich sedimentary rocks and intrusive rocks
soil type	histosol	histosol	histosol	histosol
soil pH⁴ (range)				
organic horizons	4.3–6.4	3.9–4.9	4.2–6.4	3.8–5.2
mineral horizons	5.2–5.9	4.9–5.8	4.7–6.6	5.1
land cover⁵	grassland	moor	broad-leaved forest	peat bog
ecological habitat⁶	moss and lichen tundra (F1.2) / acid alpine and subalpine grassland (E4.3)	shrub tundra (F1.1)	broadleaved swamp woodland on acid peat (G1.5)	raised bog (D1.1)

831 ¹based on a 10 m national DEM (*geonorge.no*)832 ²based on CHELSA project (downscaled model, ERA-Interim climatic analysis; Karger *et al.*, 2017)833 ³based on interpolated data from the WorldClim 2.0 data base (Fick and Hijmans, 2017)834 ⁴pH_{CaCl₂, 0.25 mM}835 ⁵CORINE Land Cover 2012 (European Environment Agency, 2014)836 ⁶EUNIS habitat (Davies *et al.*, 2004)

837

838 **Table 2.** Summary of trace and major element concentrations in organic (OC content > 5%) and
 839 mineral (OC content < 5%) soil horizon classes and comparison with soil concentrations from
 840 Nord-Trøndelag (Central Norway).

841

	soil concentration in Abisko ($\mu\text{g g}^{-1}$)				soil concentration in Central Norway ¹ ($\mu\text{g g}^{-1}$)	
	organic horizon (n = 44)		mineral horizon (n = 21)		median in organic horizon	median in mineral horizon
	median	range	median	range		
Al	16,700	2,170–95,900	104,000	80,100–128,000	2,030	11,787
As	1.67	0.27–11.0	3.13	1.23–5.51	0.81	1.48
Ba	128	19.1–632	480	81.5–605	36	15
Cd	0.24	<0.01–2.92	0.23	0.09–0.67	0.51	0.03
Co	6.12	1.02–202	18.9	6.05–28.6	1.48	6.14
Cr	15.2	1.38–78.2	55.1	36.6–100	3.03	26
Cu	20.9	3.28–204	18.0	2.54–97.9	7.86	13
Fe	20,300	2,570–136,000	42,800	26,000–74,600	3,003	19,419
Mn	224	2.80–1,910	820	83.4–2,270	58	167
Ni	8.03	<0.23–108	21.3	5.73–130	3.23	14
Pb	6.53	1.76–19.5	14.2	6.65–18.3	27	6.61
Sb	0.14	<0.01–1.60	0.40	0.04–3.64	0.33	0.04
Se	2.06	0.39–15.1	2.94	0.39–7.70	0.90	<0.5
Sn	0.58	0.01–2.30	1.90	1.40–2.58	0.75	0.44
Sr	44.4	14.6–214	181	25.8–256	30	6.87
Ti	1,510	163–4,470	4,170	3,660–7,830	161	1,032
V	28.3	2.95–138	91.4	63.5–273	2.91	31
Zn	25.9	0.68–179	69.7	20.2–193	38	27

842 ¹Reimann et al. (2015)

843

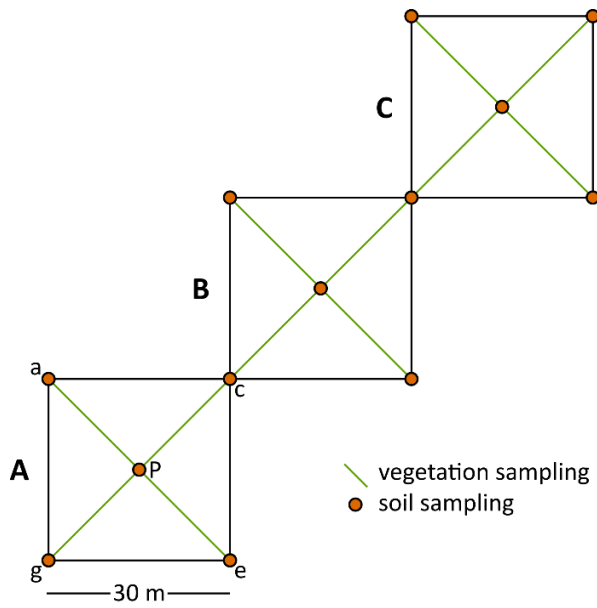
844 **Table 3.** Proportions (in %) of vegetation classes following the adapted Walker (2000)
 845 classification in the four study sites. Both deciduous and evergreen were identified for trees,
 846 shrubs, and dwarf shrubs.

	NJU975	BAD599	NJU383	TOR363
trees	–	–	5	–
<i>deciduous</i>	–	–	2	–
<i>evergreen</i>	–	–	3	–
shrubs	10	10	1	2
<i>deciduous</i>	10	10	1	2
<i>evergreen</i>	–	–	–	–
dwarf shrubs	26	42	13	5
<i>deciduous</i>	6	16	6	2
<i>evergreen</i>	20	26	7	5
graminoids	22	19	14	87
forbs	11	1	42	1
mosses	15	12	21	3
lichens	16	16	4	2

847

848 **List of figures**

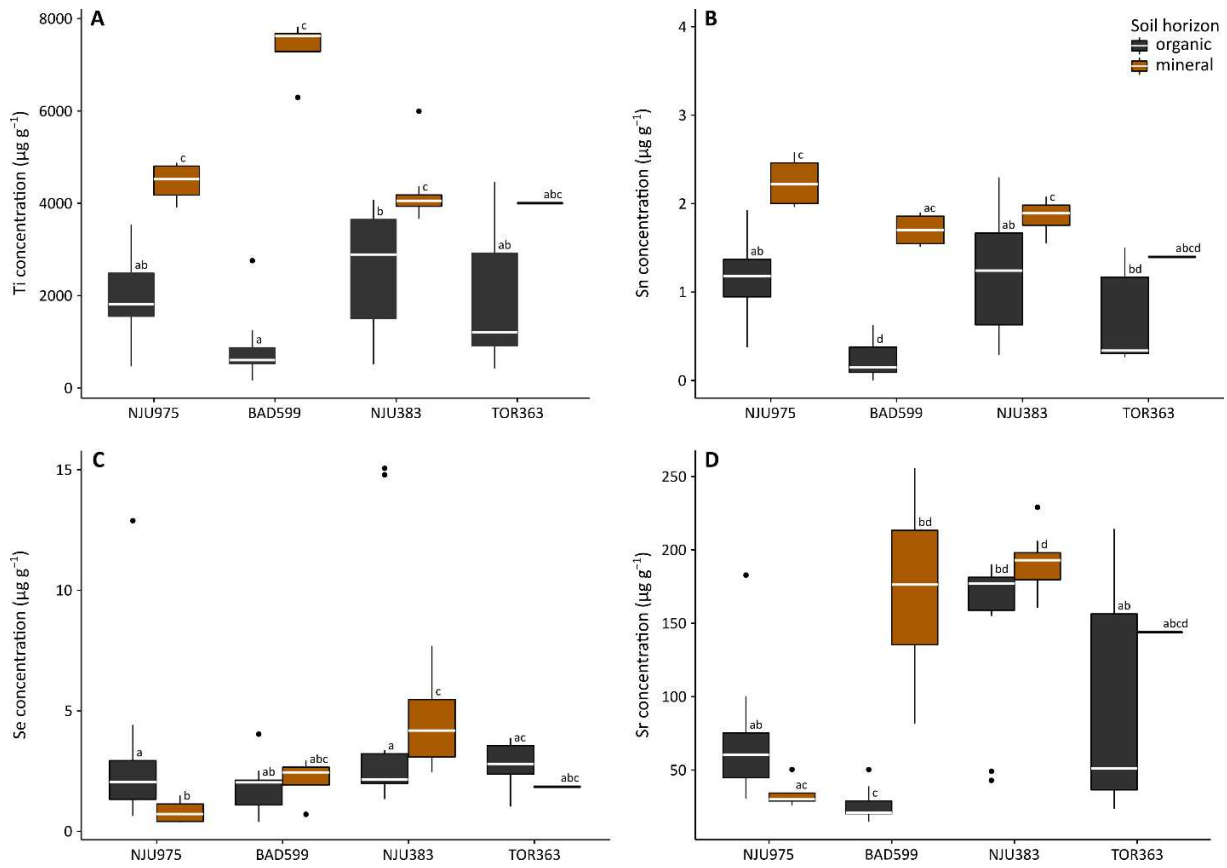
- 849 **Figure 1.** Location of the four study sites around Abisko Scientific Research Station
 850 (www.sgu.se; **A**) in Northern Sweden (**B**). The mining areas were indicated by colored shapes of
 851 the regional map (**C**): Cu mines in blue and Fe mines in red (Eilu, 2012). Wind rose is presented
 852 for Abisko (1987–2017, Abisko Meteorological Station NOAA-NCDC; **D**). 38
- 853 **Figure 2.** Sampling protocol for soils and vegetation applied in each study sites considering
 854 three pixels (A, B, and C) following the Landsat imagery grid, with the exception of TOR363
 855 where only one pixel was sampled. Soil sampling considered four corner points (a, c, e, and g)
 856 and a central point (P) in each pixel..... 39
- 857 **Figure 3.** Concentrations of Ti (**A**), Sn (**B**), Se (**C**), and Sr (**D**) in both organic (gray, n = 44) and
 858 mineral (brown, n = 21) soil horizon classes for the four study sites in Abisko. Letters represent
 859 statistically significant differences using Kruskal-Wallis and Dunn post-hoc tests ($p < 0.05$). ... 40
- 860 **Figure 4.** Enrichment factor of trace and major elements in organic soil horizons normalized to
 861 Al and mineral soil horizons of the same study sites. The dash gray line indicates the enrichment
 862 factor of 1. Letters represent statistically significant differences using Kruskal-Wallis and Dunn
 863 post-hoc tests ($p < 0.05$)..... 41
- 864 **Figure 5.** Principal component analysis of log-ratio transformed trace and major element
 865 concentrations in soil samples collected at Abisko (n = 61): representation of elements for
 866 components 1 vs. 2 (**A**) and components 1 vs. 3 (**D**) and representation of observations by soil
 867 horizon class (**B**) and by study site (**C**) for components 1 vs. 2. 42
- 868 **Figure 6.** Non-negative matrix factorization of trace and major element concentrations in soil
 869 samples collected at Abisko (n = 61): contribution of the four factors for each element (**A**) and
 870 sampling site by soil horizon class (**B**). Each factor represents distinct source: lithogenic (factor
 871 1), surface (factor 2), mixed (factor 3), and mining (factor 4)..... 43
- 872 **Figure 7.** Partial least squares regression using geochemical data (the first three principal
 873 components from the PCA [PC1, PC2, and PC3] and the four factors from the NMF [F1, F2, F3,
 874 and F4]) as dependent variables (red) and environmental data (vegetation classes according to
 875 the Walker (2000) classification and soil pH and conductivity) as independent variables (green):
 876 circle of correlations (**A**) and map of observations (**B**). Only organic soil horizon classes were
 877 considered (n = 37). 44
- 878



885

886 **Figure 2.** Sampling protocol for soils and vegetation applied in each study sites considering three pixels
 887 (A, B, and C) following the Landsat imagery grid, with the exception of TOR363 where only one pixel
 888 was sampled. Soil sampling considered four corner points (a, c, e, and g) and a central point (P) in each
 889 pixel.

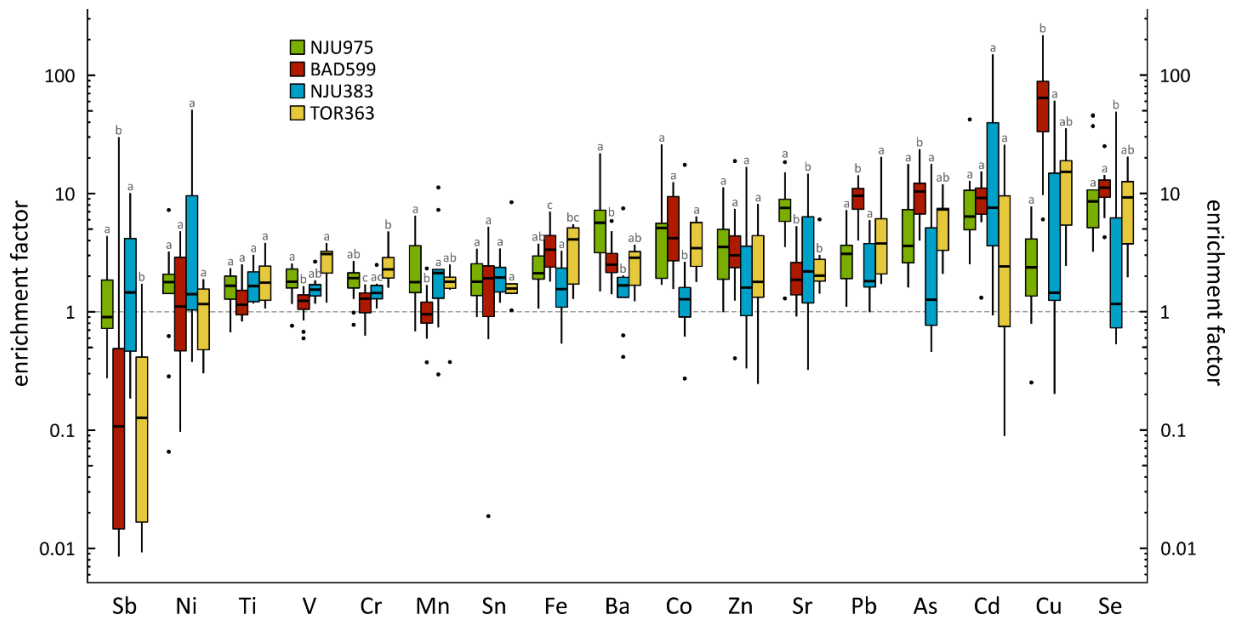
890



891

892 **Figure 3.** Concentrations of Ti (A), Sn (B), Se (C), and Sr (D) in both organic (gray, n = 44) and mineral
 893 (brown, n = 21) soil horizon classes for the four study sites in Abisko. Letters represent statistically
 894 significant differences using Kruskal-Wallis and Dunn post-hoc tests ($p < 0.05$).

895



896

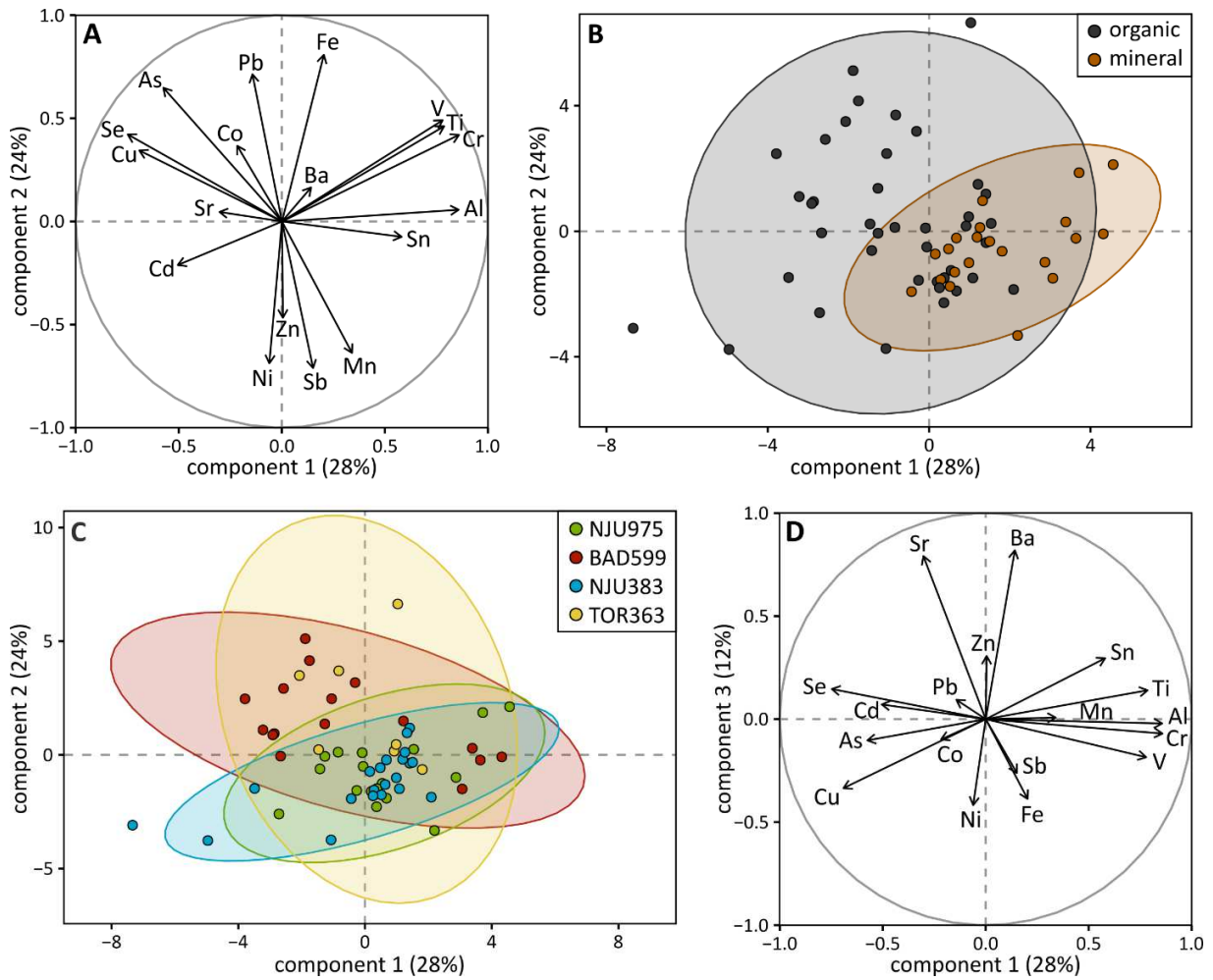
897 **Figure 4.** Enrichment factor of trace and major elements in organic soil horizons normalized to Al and

898 mineral soil horizons of the same study sites. The dash gray line indicates the enrichment factor of 1.

899 Letters represent statistically significant differences using Kruskal-Wallis and Dunn post-hoc tests

900 ($p < 0.05$).

901



902

903

904

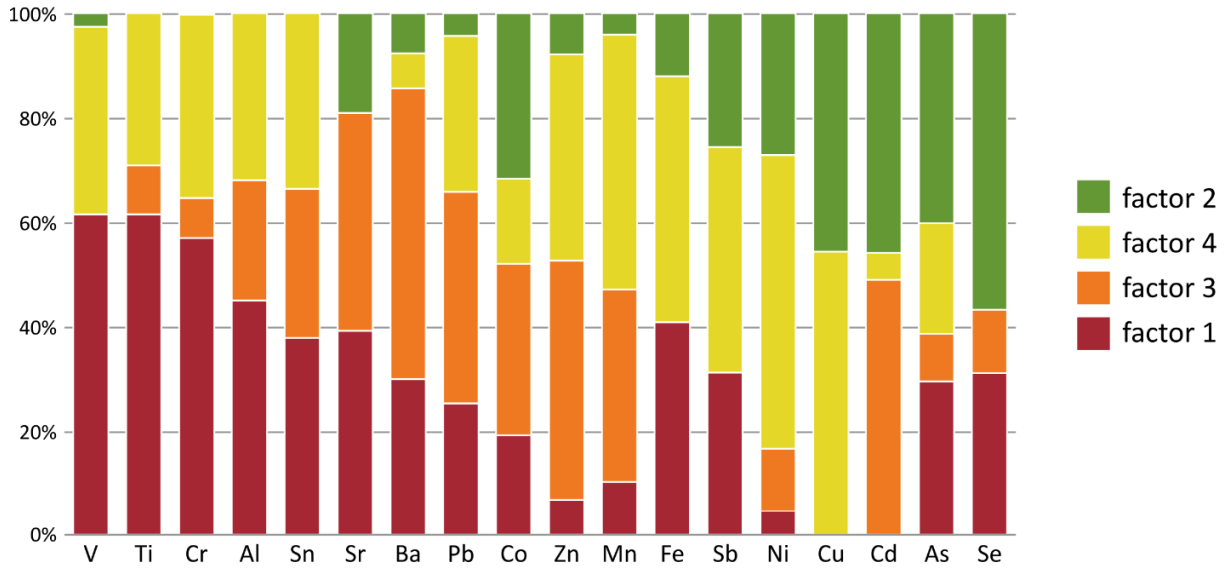
905

906

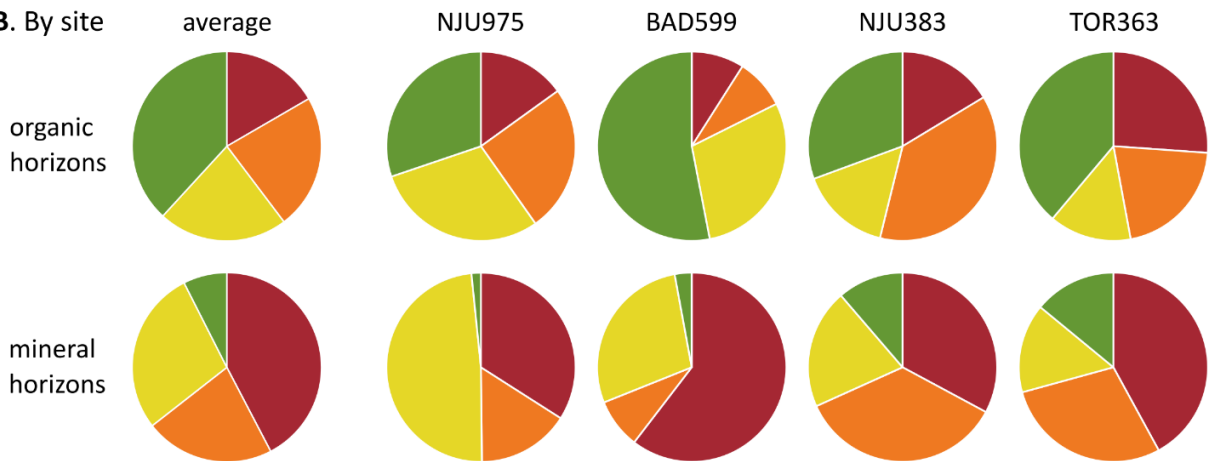
907

Figure 5. Principal component analysis of log-ratio transformed trace and major element concentrations in soil samples collected at Abisko ($n = 61$): representation of elements for components 1 vs. 2 (A) and components 1 vs. 3 (D) and representation of observations by soil horizon class (B) and by study site (C) for components 1 vs. 2.

A. By element



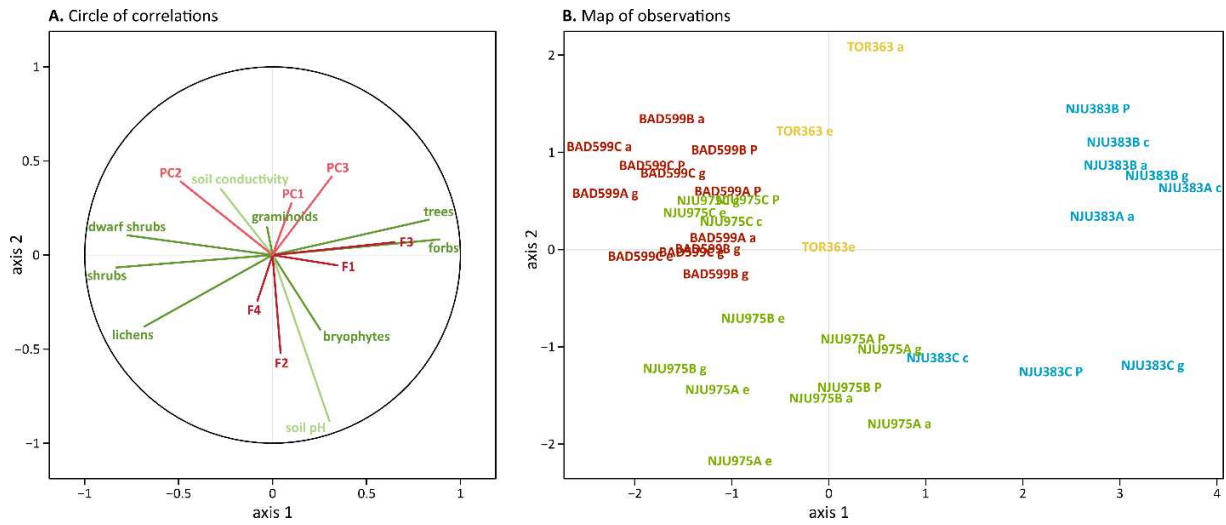
B. By site



908

909 **Figure 6.** Non-negative matrix factorization of trace and major element concentrations in soil samples
 910 collected at Abisko (n = 61): contribution of the four factors for each element (A) and sampling site by
 911 soil horizon class (B). Each factor represents distinct source: lithogenic (factor 1), surface (factor 2),
 912 mixed (factor 3), and mining (factor 4).

913



914

915 **Figure 7.** Partial least squares regression using geochemical data (the first three principal components
 916 from the PCA [PC1, PC2, and PC3] and the four factors from the NMF [F1, F2, F3, and F4]) as
 917 dependent variables (red) and environmental data (vegetation classes according to the Walker (2000)
 918 classification and soil pH and conductivity) as independent variables (green): circle of correlations (A)
 919 and map of observations (B). Only organic soil horizon classes were considered (n = 37).

920