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

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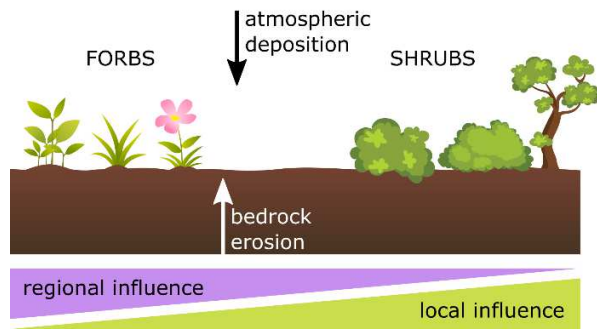
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12

13 **Graphical abstract**

Subarctic ecosystems



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  $330\text{ mm a}^{-1}$  (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  $3.46\text{ m s}^{-1}$  (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 18<sup>th</sup> 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 m<sup>2</sup>-pixel was composed of two crossed transects (green lines, **Figure 2**),  
209 themselves comprising 42 quadrats of 1 m<sup>2</sup>. 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<sup>-1</sup> CaCl<sub>2</sub> extraction solution (solution/soil  
227 ratio of 1:10). The total dissolution of the samples was performed using a mixture of suprapure  
228 acids (HNO<sub>3</sub> and HF) and H<sub>2</sub>O<sub>2</sub>. 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% HNO<sub>3</sub> was added  
231 and kept at room temperature for 6 h before adding 1 mL of 30% H<sub>2</sub>O<sub>2</sub> for 24 h at 40 °C for  
232 evaporation. Finally, we added 1 mL of 65% HNO<sub>3</sub> for 24 h and solutions were diluted with  
233 Milli-Q water to obtain a final acid concentration of 2% HNO<sub>3</sub> 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  $\leq 6.6$ ), 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.

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829		

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

	<b>NJU975</b>	<b>BAD599</b>	<b>NJU383</b>	<b>TOR363</b>
<b>latitude/longitude</b>	68.363°N / 18.716°E	68.317°N / 18.867°E	68.355°N / 18.763°E	68.349°N / 19.057°E
<b>elevation (m a.s.l.)<sup>1</sup></b>	975	599	383	363
<b>slope aspect / steepness<sup>1</sup></b>	SE / ~3.25°	SW / ~1°	ESE / ~2°	N / ~1°
<b>mean annual air temperature (1979–2013; °C)<sup>2</sup></b>	–3.2	–1.2	–0.1	–0.3
<b>precipitation (1979–2013; mm a<sup>-1</sup>)<sup>2</sup></b>	602	449	413	402
<b>average wind speed (1970–2000; m s<sup>-1</sup>)<sup>3</sup></b>	3.2	2.7	2.5	2.3
<b>bedrock</b>	mica-rich metamorphic rocks	calc-silicate rocks	mica-rich metamorphic rocks	quartz-feldspar-rich sedimentary rocks and intrusive rocks
<b>soil type</b>	histosol	histosol	histosol	histosol
<b>soil pH<sup>4</sup> (range)</b>				
<b>organic horizons</b>	4.3–6.4	3.9–4.9	4.2–6.4	3.8–5.2
<b>mineral horizons</b>	5.2–5.9	4.9–5.8	4.7–6.6	5.1
<b>land cover<sup>5</sup></b>	grassland	moor	broad-leaved forest	peat bog
<b>ecological habitat<sup>6</sup></b>	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 <sup>1</sup>based on a 10 m national DEM (*geonorge.no*)832 <sup>2</sup>based on CHELSA project (downscaled model, ERA-Interim climatic analysis; Karger *et al.*, 2017)833 <sup>3</sup>based on interpolated data from the WorldClim 2.0 data base (Fick and Hijmans, 2017)834 <sup>4</sup>pH<sub>CaCl<sub>2</sub>, 0.25 mM</sub>835 <sup>5</sup>CORINE Land Cover 2012 (European Environment Agency, 2014)836 <sup>6</sup>EUNIS 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 <sup>1</sup> ( $\mu\text{g g}^{-1}$ )	
	organic horizon (n = 44)		mineral horizon (n = 21)		median in organic horizon	median in mineral horizon
	median	range	median	range		
<b>Al</b>	16,700	2,170–95,900	104,000	80,100–128,000	2,030	11,787
<b>As</b>	1.67	0.27–11.0	3.13	1.23–5.51	0.81	1.48
<b>Ba</b>	128	19.1–632	480	81.5–605	36	15
<b>Cd</b>	0.24	<0.01–2.92	0.23	0.09–0.67	0.51	0.03
<b>Co</b>	6.12	1.02–202	18.9	6.05–28.6	1.48	6.14
<b>Cr</b>	15.2	1.38–78.2	55.1	36.6–100	3.03	26
<b>Cu</b>	20.9	3.28–204	18.0	2.54–97.9	7.86	13
<b>Fe</b>	20,300	2,570–136,000	42,800	26,000–74,600	3,003	19,419
<b>Mn</b>	224	2.80–1,910	820	83.4–2,270	58	167
<b>Ni</b>	8.03	<0.23–108	21.3	5.73–130	3.23	14
<b>Pb</b>	6.53	1.76–19.5	14.2	6.65–18.3	27	6.61
<b>Sb</b>	0.14	<0.01–1.60	0.40	0.04–3.64	0.33	0.04
<b>Se</b>	2.06	0.39–15.1	2.94	0.39–7.70	0.90	<0.5
<b>Sn</b>	0.58	0.01–2.30	1.90	1.40–2.58	0.75	0.44
<b>Sr</b>	44.4	14.6–214	181	25.8–256	30	6.87
<b>Ti</b>	1,510	163–4,470	4,170	3,660–7,830	161	1,032
<b>V</b>	28.3	2.95–138	91.4	63.5–273	2.91	31
<b>Zn</b>	25.9	0.68–179	69.7	20.2–193	38	27

842 <sup>1</sup>Reimann et al. (2015)

843

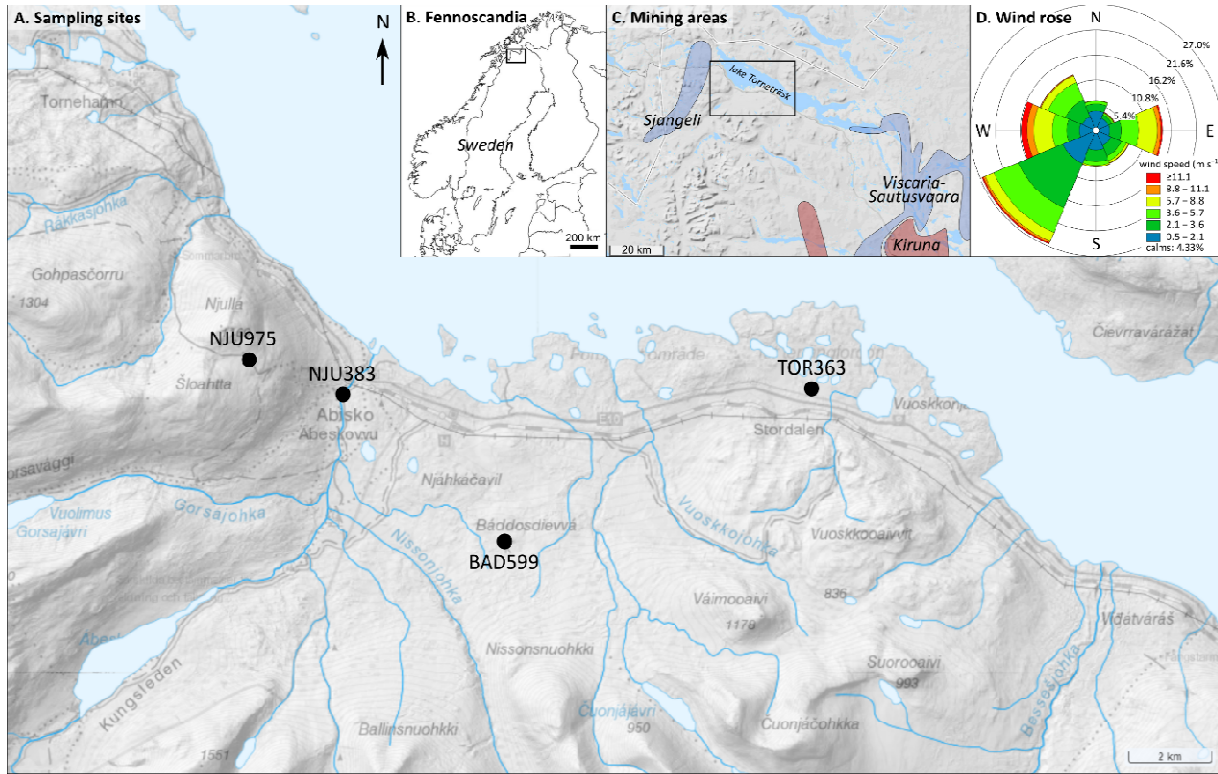
844 **Table 3.** Proportions (in %) of vegetation classes following the adapted Walker (2000)  
 845 classification in the four study sites. Both deciduous and evergreen were identified for trees,  
 846 shrubs, and dwarf shrubs.

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

847

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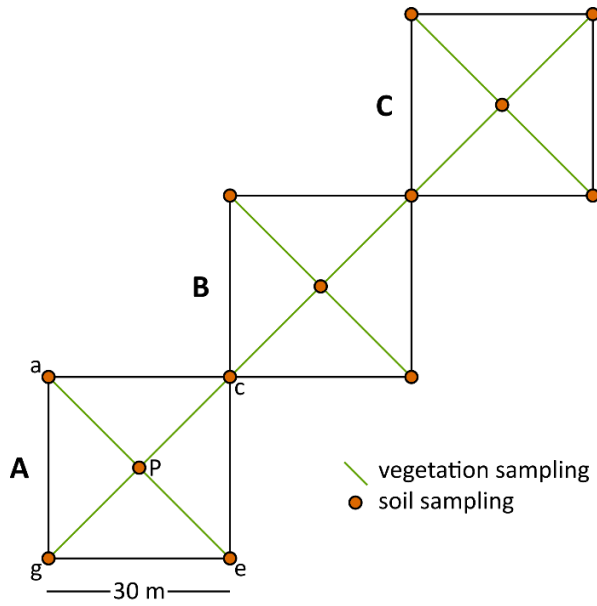
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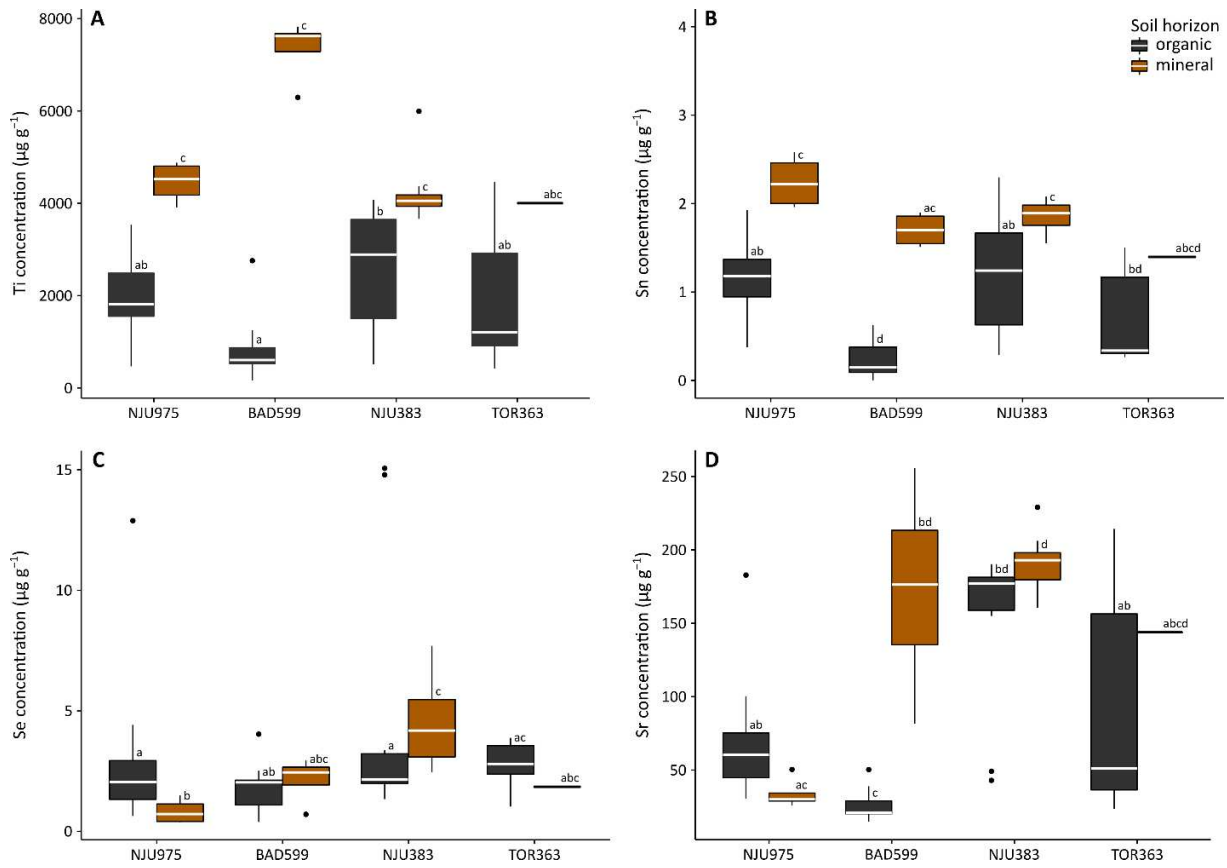
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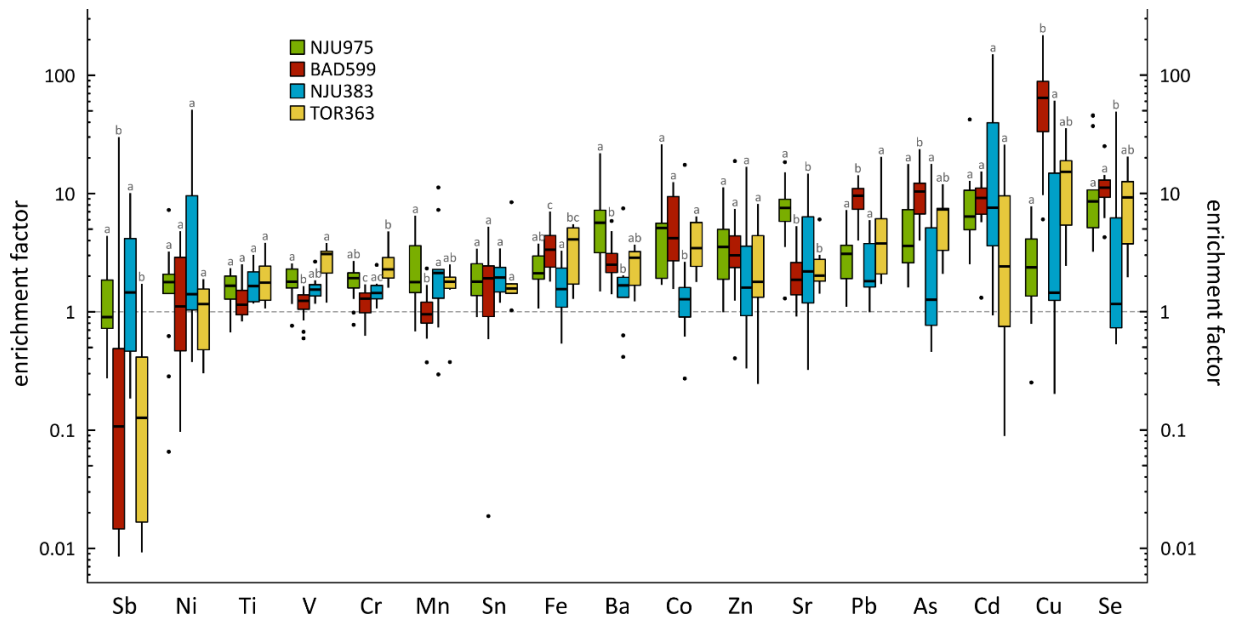
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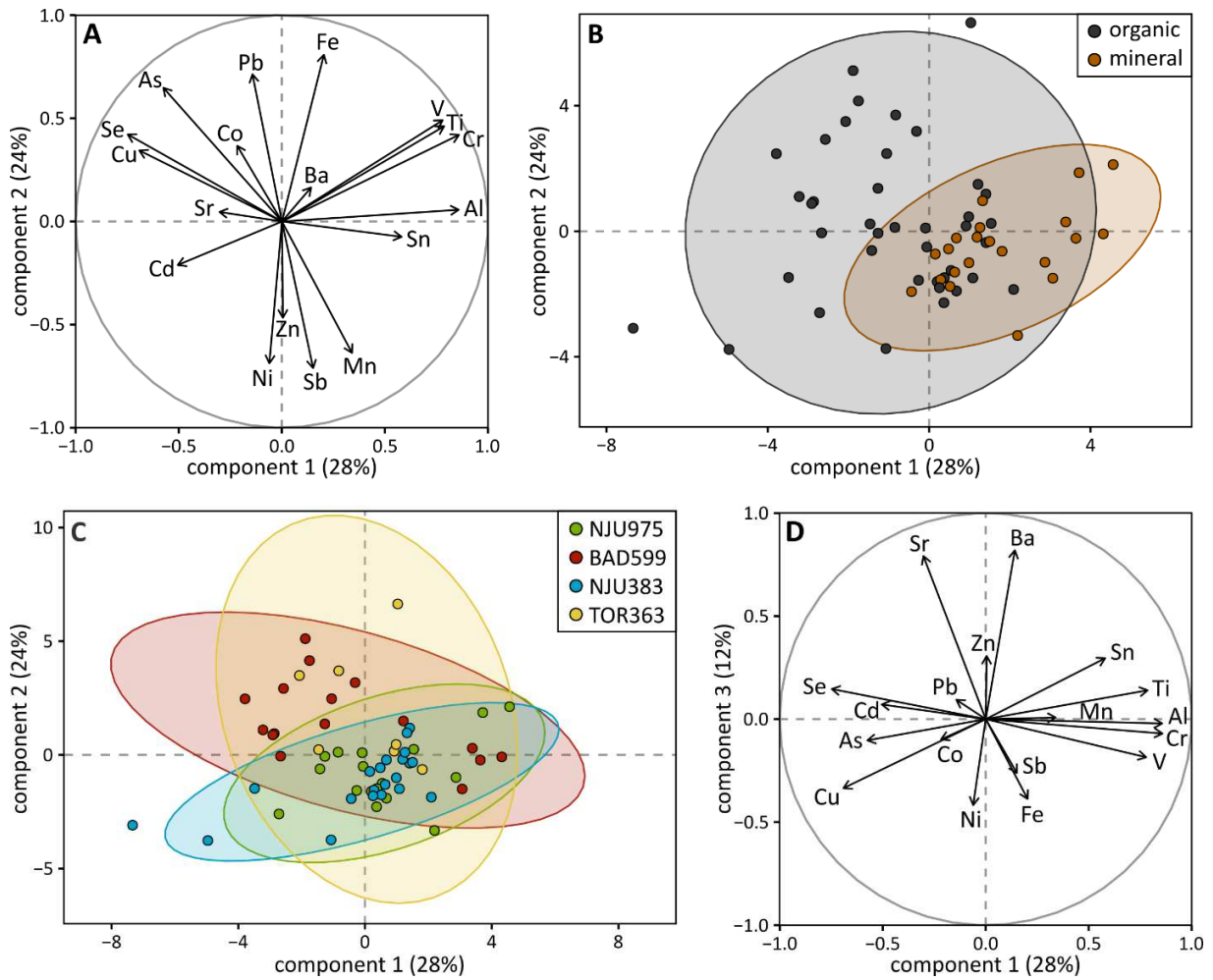
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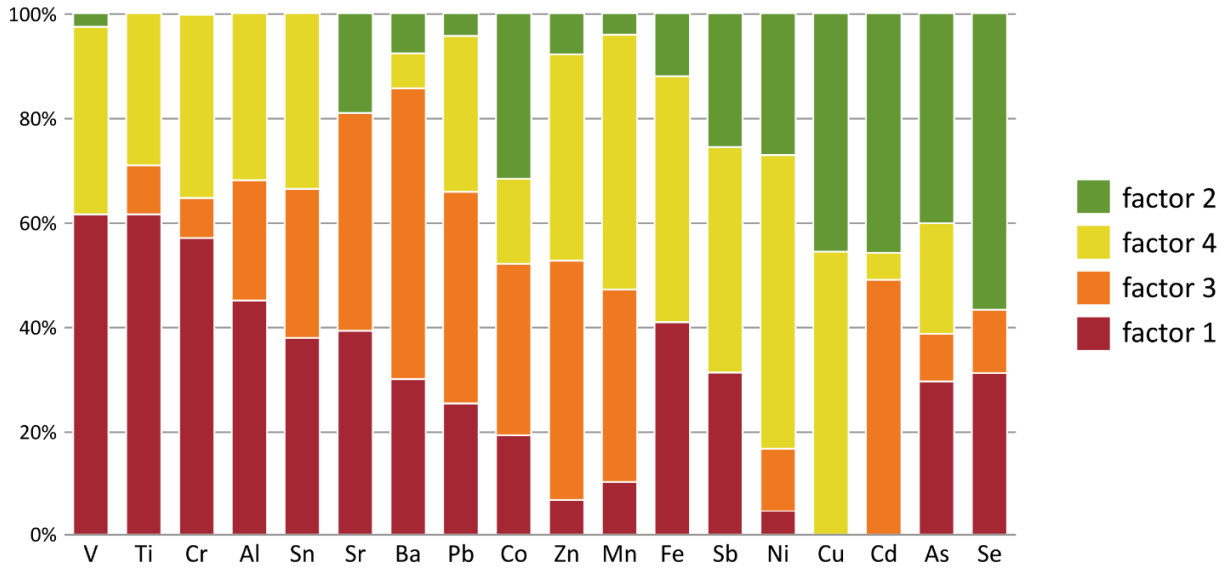
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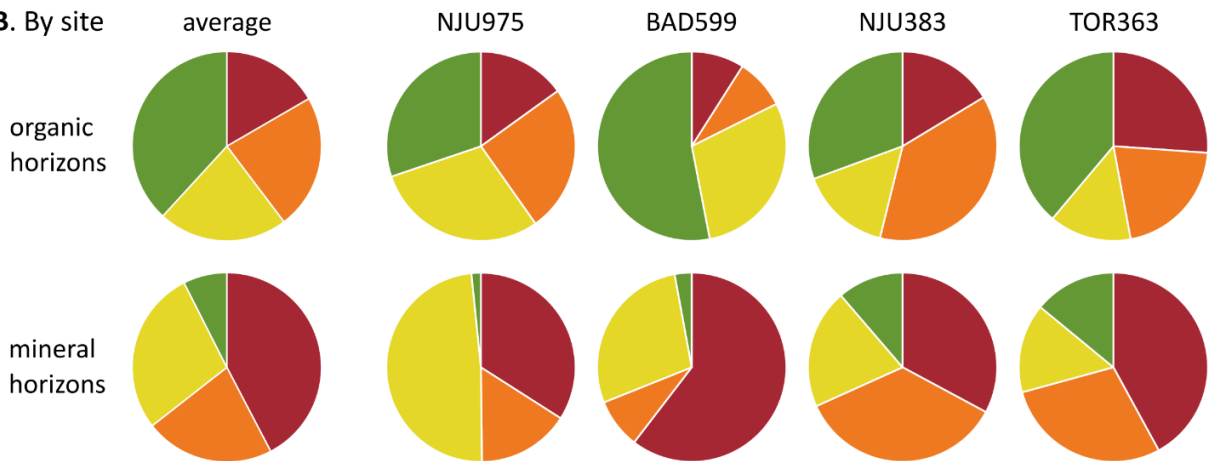
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**A. By element**



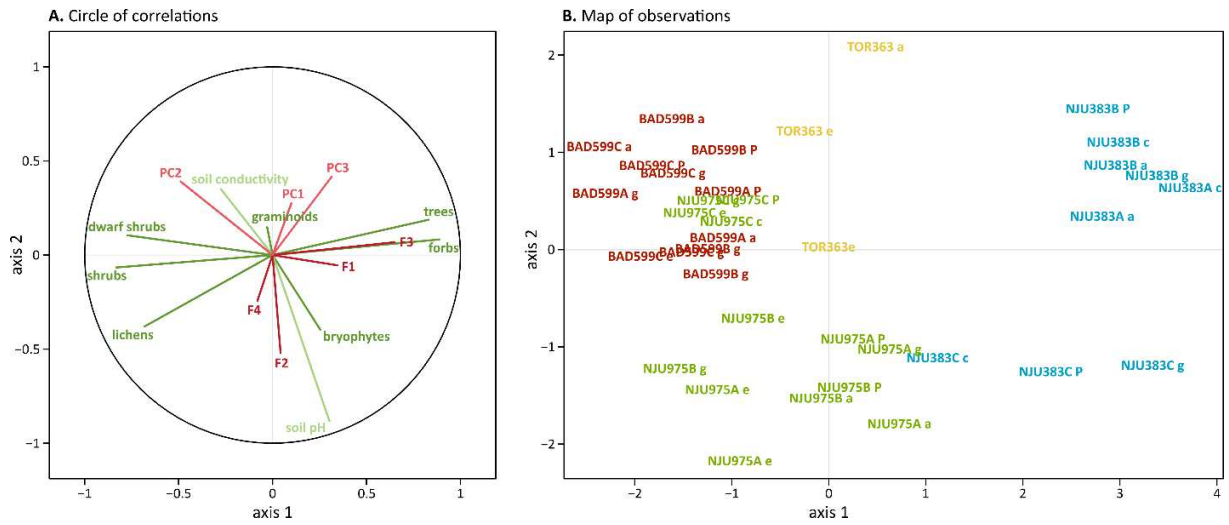
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