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(Article begins on next page)

1 **Soil properties and trace elements distribution along an**
2 **altitudinal gradient on the southern slope of Mt. Everest, Nepal**

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10 **Abstract**

11 The absence of significant local inputs of pollution makes remote mountain ecosystems suitable to
12 assess the atmospheric deposition of contaminants, such as trace elements, which can derive from
13 both natural and anthropogenic sources. The Himalayan range is a potential target for the atmospheric
14 deposition of pollutants because of the regional monsoon climate and the presence of contaminant
15 source regions in its vicinity (e.g. Kathmandu Valley). Studies of elevation gradients of trace elements
16 in topsoils and soil profiles in the Himalaya are very limited. The main goal of this study was to
17 determine the distribution of trace elements (Co, Cd, Cu, Zn, Cr, Pb, Ni, and Mn) in soils on the
18 southern slope of Mt. Everest as a function of elevation, soil depth, and pedogenic properties. Trace
19 elements were measured in 30 topsoils along an altitudinal gradient (3570-5320 m a.s.l.) and in 11
20 different soil profiles opened under different land uses and topographical settings. The contents of
21 Co, Zn, Cr, and Ni in the topsoil were found positively correlated with the altitude, and on average
22 reached a peak at 4900-5000 m a.s.l.. The results showed a limited contamination, similar to the one
23 measured in soils from other high mountain regions. Differently from the expectations, both soil depth
24 and organic carbon, which significantly decreased with the altitude, were not found significant factors
25 controlling the altitudinal distribution of trace elements. Pedogenic processes were associated with
26 different depth trends of trace metals along the elevation gradient, with depletion in surface and
27 accumulation in illuvial horizons where podzolization was active; at higher altitude, a weaker
28 leaching resulted in higher surface concentrations.

29

30 **Key Words:** Himalaya, Depth distribution, Inorganic contaminants, Pedogenesis, Toposequence.

31

32 **1. Introduction**

33 Mountain soils are a fragile ecosystem, and their high spatial variability derives from their strong
34 dependency on factors such as parent materials, climate, relief, biosphere, and human impact, which
35 are particularly striking in high-elevation areas (Florinsky, 2012; Zanini et al., 2015).

36 Soil parent material is a primary source of trace elements particularly in weakly developed soils
37 (D'Amico et al., 2015). However, additions may occur from atmospheric deposition from both natural
38 and anthropogenic sources. The long-range transport of trace elements from centers of human activity
39 to the biosphere has been recognized in the past years (e.g. Elgmork et al. 1973, Zoller et al. 1974).

40 Trace elements occur in the atmosphere as, or adsorbed to, airborne particles, and these atmospheric
41 aerosols can travel long distances before being redeposited, reaching remote areas such as the Arctic
42 (Camarero et al., 2009), and high elevations such as the Himalayan Mountains (Yeo and Langley-
43 Turnbaugh, 2010; Cong et al., 2015). The deposition degree of air-borne trace elements is related to
44 several factors. These include distance from potential sources, intensity and frequency of precipitation
45 and wind, and the aerosol capture by intercepting surfaces (Reiners et al., 1975; Bacardit and
46 Camarero, 2010). Their deposition is especially influenced by precipitation and wind, particularly in
47 regional convergence zones, such as mountain ranges, that trap atmospheric contaminants because of
48 cold condensation and enhance atmospheric deposition (the so-called "orographic effect") (Lovett
49 and Kinsman 1990; Loewen et al. 2005; Wegmann et al. 2006; Cong et al., 2015). Since precipitation
50 tends to increase with altitude until a specific elevation limit and then tends to decrease (Salerno et
51 al., 2015), total deposition might be expected to increase accordingly (Reiners et al., 1975). In
52 addition, the soil trace elements content is susceptible to chemical (e.g. weathering, pedogenesis,
53 leaching) and biological (e.g. microbial decomposition or organo-metal chelates) post-depositional
54 processes. It is well known that soil characteristics are closely related to trace elements retention,
55 uptake by plants and mobility into the ecosystem with potential negative effects on living organisms
56 and water sources (Navas and Lindhorfer, 2005). According to USEPA (2008), one-tenth of the

57 world's population is using high mountain water every day, emphasizing the importance of a more
58 detailed knowledge of elemental contents in high altitude areas.

59 Few studies, with contrasting results, have examined the soil trace elements distribution in high-
60 elevation soil profiles, whose characteristics and pedogenic properties were less studied compared to
61 lower altitude soils (e.g. Magnani et al., 2017a). Mountain soils are strongly influenced by elevation
62 and local topographical settings (Zanini et al., 2015) and at higher elevation, terrain features (e.g.
63 concavities and convexities) may influence the amount of water, the accumulation of sediments and
64 the snow cover duration (Magnani et al., 2017b), with significant effects on soil development. Soil
65 physicochemical properties, such as organic matter content, oxides, pH, and redox reactions
66 determine trace elements mobility along the profile and the more so in extreme settings such as a high
67 mountain range (Navas and Lindhorfer, 2005). A valuable opportunity to study both the soil
68 development and the associated trace elements distribution is offered in the Himalayan range, which
69 presents both a wide altitudinal range and a traditionally considered pristine environment, and thus is
70 suitable for studying the effects of remote pollution sources. It is known that the orographic effect is
71 quite pronounced in the Himalayan range because of dramatic elevation temperature and precipitation
72 gradients relative to contaminant source regions in its vicinity, and because of the regional monsoon
73 climate that has been shown to deliver air pollutants to higher altitudes (Loewen et al., 2005). The
74 Kathmandu Valley, which is widely recognized as an area characterized by a severe air pollution due
75 to high concentrations of airborne particulate matter (Gurung and Bell, 2012), can be considered a
76 potential regional source of contamination for the southern slope of Mt. Everest, especially during
77 the monsoon period when the prevailing direction of the wind is S-N and SW-NE (Ichiyanagi et al.,
78 2007).

79 Following these considerations, this study was designed along an elevation transect in remote areas
80 in the Sagarmatha National Park (central southern Himalaya) to test these hypotheses: 1) The soil
81 pedogenic properties would change along the altitudinal gradient and on a natural high-elevation

82 toposequence; 2) Trace element contents would increase with elevation, but would then decline above
83 the treeline due to a decline in rainfall and interceptive vegetative surfaces; 3) Trace element contents
84 along the profiles would be influenced by organic matter dynamics and differential leaching
85 associated with pedogenic processes.

86 Soil characteristics and trace element contents were analyzed on topsoil samples and in soil profiles
87 across a wide altitudinal gradient (from 3700 to 5320 m a.s.l.), and on one high-elevation
88 toposequence (5055 – 5070 m a.s.l.) in order to point out background values, external additions and
89 possible local contamination due to the increasing touristic pressure. A special attention was given to
90 factors and pedogenic trends, which may influence trace elements distribution, such as elevation,
91 topography, land cover, and local or remote sources of contamination.

92

93 **2. Material and Methods**

94 *2.1. Study area*

95 The study area is located in the Mt. Everest region in the Sagarmatha National Park and Buffer Zone
96 (SNPBZ) (27.75° to 28.11° N; 85.98° to 86.51° E), which lie in eastern Nepal in the southern part of
97 the central Himalaya (Fig. 1). The park area (1148 km²) extends in elevation from 2845 to 8844 m
98 a.s.l. covering the upper Dudh Koshi Valley, and represents the highest protected area in the world
99 (Amatya et al., 2010).

100 Carosi et al. (1999) described the geology of the Higher Himalayan Crystallines, while Searle et al.
101 (2003) summarized the main lithological units in the SNP. The Khumbu Valley substratum is
102 dominantly made up of crystalline rocks (e.g. sillimanite gneisses calc-silicates, amphibolites and K-
103 feldspar augen gneisses) and leucogranites that intrude the metasediments as dykes, sills and bodies.
104 The overlying Everest series (weakly metamorphosed shales and pelites with limestone bands), the
105 Yellow band unit (limestones, marbles and calc-silicates), and the Ordovician limestones are only
106 present in the eastern part of the study area. Extensive glacial till of different ages, alluvial and glacio-

107 fluvial deposits, host sub-surface aquifers in close connection with the hydrographic network (Tartari
108 et al., 1998; Salerno et al., 2016).

109 According to Nepal and Nepal (2004), the soils in the high valleys are primarily Regosols. Below
110 4000 m a.s.l., Podzols have developed in forested areas, which are mainly located in the north-facing
111 slopes. The extensive grassland and shrubland areas in the southern slopes below 3750 m a.s.l. were
112 characterized by Cambisols and Regosols.

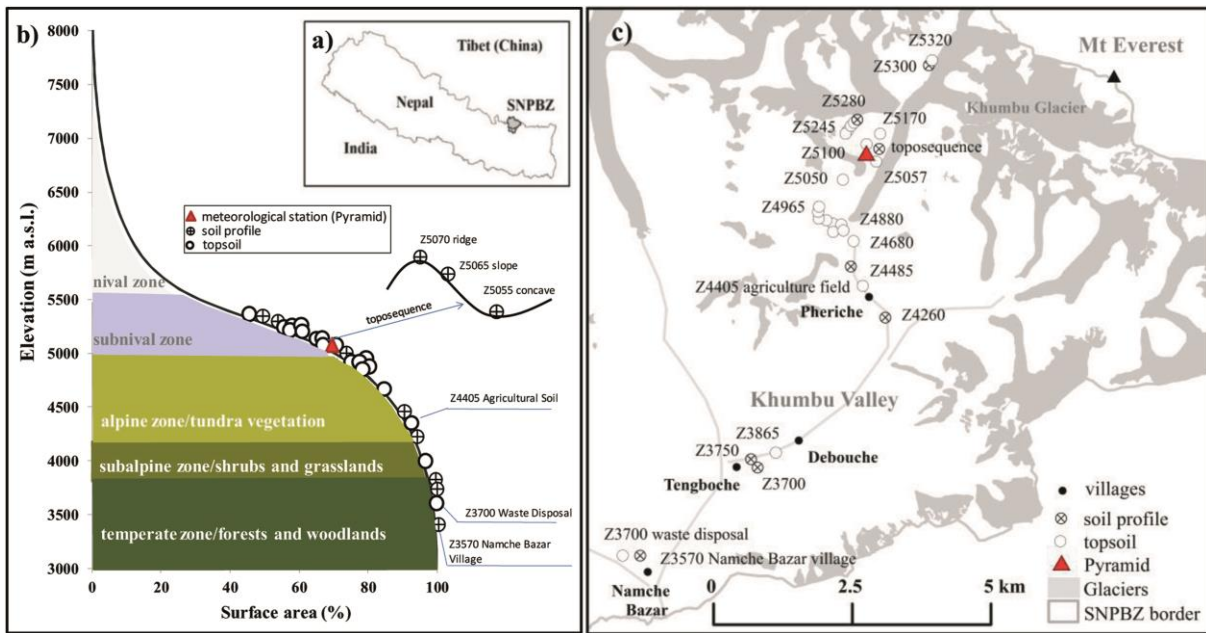
113 The SNPBZ can be divided into three vegetation zones based on altitude. The lower altitudinal belt
114 (below 3800 m a.s.l.), consisting of temperate forests and woodlands, the middle zone (3800 to 4200
115 m a.s.l.) of subalpine forests and shrublands, and the upper zone (above 4200 m a.s.l.) of alpine tundra
116 vegetation (Nepal and Nepal, 2004). Shrubs, grasslands and diverse varieties of herbs characterize
117 the vegetation from 3600 to 4000 m a.s.l. on the southern slope. The alpine zone from 4200 to 5500
118 m a.s.l. is divided into a lower area characterized by moist alpine shrubs of dwarf rhododendrons
119 (*Rhododendron setosum* D. Don, *Rhododendron nivale* Hook. f., *Rhododendron lepidotum* Wall. ex
120 G. Don, *Rhododendron anthopogon* D. Don) and prostrate junipers (*Juniperus recurva* Buch.-Ham.
121 Ex D.Don, *Juniperus indica* Bertol.), and an upper area dominated by *Kobresia pygmaea*
122 (C.B.Clarke) C.B.Clarke mats and cushion plants such as *Anaphalis cavei* Chatterjee and
123 *Leontopodium monocephalum* Edgew. (Byers, 2005).

124 Glaciers are located above 4300 m a.s.l., with more than 75% of the glacier surfaces lying between
125 5000 and 6500 m a.s.l. (Thakuri et al., 2014; 2016). The climate is characterized by monsoons, with
126 a prevailing S-N direction. During the 1994-2013 period, the mean annual precipitation at the
127 Pyramid meteorological station (5050 m a.s.l.) was 446 mm, with a mean annual temperature of -2.45
128 °C. In total, 90% of the precipitation falls between June and September. The probability of snowfall
129 during these months is very low (4%) but reaches 20% at the annual level. Precipitation linearly
130 increases to an elevation of 2500 m a.s.l. and exponentially decreases at higher elevations (Salerno et
131 al., 2015; Derin et al., 2016).

132 The SNPBZ, which represents one of the most attractive mountain sites in the world, reports an
133 exponential increase of tourists trekking in the last thirty years, reaching even 30,000 visitors in 2008
134 (Salerno et al., 2013). Nepal and Nepal (2004) reported soil erosion on trails, while Byers (2005)
135 showed how erosion processes were exacerbated by harvesting of soil-binding shrub juniper for fuel
136 wood, primarily used in tourist lodges. Salerno et al. (2010) reported a 20% decrease of forest cover
137 since 1992 in order to sustain the tourism demand for fuel. Furthermore, the increasing number of
138 visitors caused the localized buildup of litter and pollution from human waste and the necessity to
139 manage solid waste that is usually collected and burnt in specific areas (Stevens, 2003; Manfredi et
140 al., 2010). Local sources of contamination could therefore contribute to an increase of trace elements
141 in soil, whose distribution has been studied extensively in sites that are close to human activity centers
142 (Li et al., 2009; Biasioli et al., 2012), in agricultural areas (Wilcke, 2000; Liu et al., 2014) and in
143 forest soils (Hernandez et al., 2003).

144 In our study area, at low elevations (agropastoral zone below 4000 m a.s.l.), the farming activities of
145 Sherpa people has contributed to the development of soils rich in organic matter by leveling, stone
146 removal and manure distribution. Forests are an important part of village life and provide fuel wood,
147 structural timber, litter, and grazing areas (Stevens, 1993). The main impacts that tourism has had on
148 local vegetation are the forest thinning in the Khumbu temperate and subalpine forests, which was
149 more intensive and extensive in Pharak, and the loss of alpine shrub juniper in some areas of eastern
150 and central Khumbu (Stevens, 2003). The forest is preserved in many sacred sites near the
151 monasteries, such as in the Kathmandu Valley, and appears as vegetative islands in an intensively
152 used landscape, like spiritual oases (Hamilton, 2002).

153



154

155 **Fig. 1.** a) Location of the study area (SNPBZ) in Nepal (southern Himalaya); b) Hypsometric curve,
 156 bioclimatic zones and location of the sampling sites in the study area; c) Spatial distribution of soil
 157 sampling sites along the Khumbu Valley.

158

159

160 *2.2. Soil sampling*

161 Soil samples were collected in July 2012 from 30 sites in the Dudh Koshi Valley along an altitudinal
 162 gradient that started from the village of Namche Bazar at the elevation of 3570 m a.s.l., and ended in
 163 the proximity of the Khumbu Glacier at 5320 m a.s.l. (Fig. 1). At 11 of these 30 sites, complete soil
 164 profiles were described according to IUSS Working Group WRB (2015) and each genetic horizon
 165 was sampled, chosen because of the representativeness of land cover and topography, in order to
 166 observe the soil forming processes that might influence the accumulation or leaching of trace
 167 elements in soil profiles. In order to better investigate the topography as soil forming factor, 3 soil
 168 profiles were opened close to the Pyramid Laboratory, at an elevation of about 5100 m a.s.l., along a
 169 toposequence characterized by concavities, gentle slope and convexities (ridge), in an “old”
 170 hummocky moraine (Hambrey et al., 2008). A different vegetation cover corresponded to different

171 catena positions, represented by sparse *Rhododendron* and herbaceous species in the concave site, by
172 a dense *Rhododendron* cover on the gentle slope and by a biological crust (bryophytes and algae) on
173 the ridge. In the other cases, due to logistic constraints, only topsoil samples were collected (0-10 cm
174 depth). Among these topsoils, one was sampled in the Namche Bazar Village (3570 m a.s.l.), one was
175 collected close to a waste disposal site at 3700 m a.s.l., near the same village, and one was collected
176 in an agricultural area, around the village of Pheriche (4405 m a.s.l.) (later named as village, waste
177 disposal, and agricultural soil, respectively).

178 Approximately 0.5-1 kg of soil material was collected from each sampled horizon or topsoil.
179 Undisturbed samples were collected with core cylinders for the measurement of bulk densities.
180 At each soil sampling site, a temperature sensor with a data logger (I-button) was placed at 10 cm
181 depth in July 2012 to measure the hourly soil temperature till October 2013.

182

183 2.3. Soil chemical and physical analysis

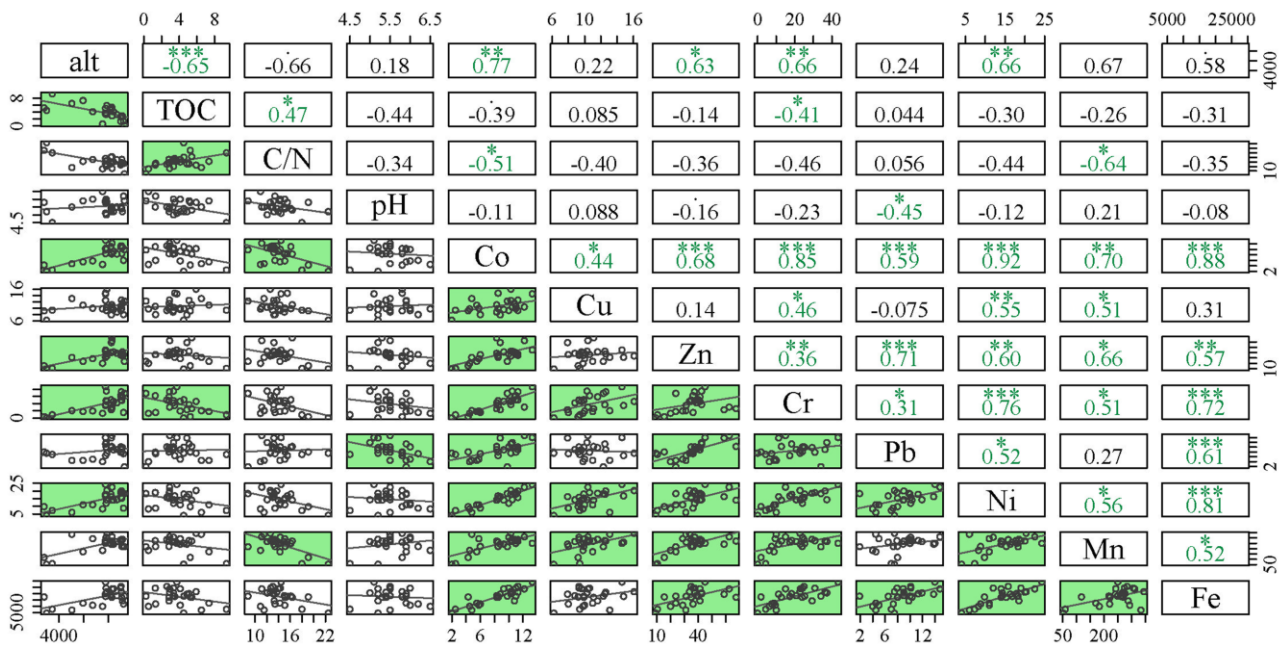
184 The soil chemical and physical analyses were performed according to standard methods (Colombo
185 and Miano, 2015). All samples were air-dried and sieved to separate the fine earth (<2 mm) from the
186 coarse fraction. The pH was measured in water (soil:water=1:2.5). Total carbon (corresponding to the
187 total organic carbon, TOC, given the absence of carbonates) and nitrogen (TN) were analyzed by dry
188 combustion with a CN elemental analyzer (CE Instruments NA2100, Rodano, Italy). The cation
189 exchange capacity (CEC) was measured with ammonium acetate (pH 7) and the exchangeable bases
190 were measured by FAAS (Flame Atomic Absorption Spectrometer, Analyst 400, Perkin Elmer,
191 Waltham, MS, USA) in the ammonium acetate extracts. The particle size distribution was determined
192 by the pipette method. The pseudo-total contents of Co, Cd, Cu, Zn, Cr, Pb, Ni, Mn and Fe were
193 measured with FAAS after a microwave-assisted *aqua regia* (3:1 HCl:HNO₃) digestion
194 (MILESTONE Start D Digester). Cd contents were always below detection limits so they were not
195 used in the elaborations.

196 Pedogenic Fe-oxides (Fed) were extracted by dithionite-citrate-bicarbonate (DCB) solution. Non-
197 crystallized or poorly crystallized Fe-oxides, hydroxides, and associated gels (Feo) were extracted by
198 acid ammonium oxalate solution (Van Reeuwijk, 2002).

199

200 *2.4. Statistical analysis*

201 Initially, the degree of correlation among the data was verified through the Pearson correlation
202 coefficient (r) and the normality of single variables and residuals were tested using the Shapiro–Wilk
203 test (Shapiro and Wilk, 1965; Hervé, 2015). We found that the altitude of the sampled soils did not
204 follow a normal distribution and even after the log-transformation, the requirements for normality
205 were not reached (Venables and Ripley, 2002), so the non-parametric Spearman’s correlation test was
206 performed in order to test the significance of the relationships. Although the majority of variables and
207 relationships were normally distributed, we preferred to apply the non-parametric test to all data set
208 and comparisons. Results are summarized in the Spearman’s correlation matrix of Fig. 2. This
209 analysis was performed using the “cor.test” function selecting “spearman” as method (R
210 Development Core Team, 2010). Generally, the non-parametric tests are more robust than the
211 parametric ones (Venables and Ripley, 2002). The Spearman’s test showed similar correlations
212 among the variables, but with a lower significance level. Therefore, the correlations presented and
213 discussed in this work are more precautionary than the ones that include parametric assumptions.



214

215 **Fig. 2.** Spearman's correlation between altitude (alt), TOC, C/N, pH, trace elements, and Fe in
 216 topsoils (n=27) along the altitudinal gradient. Village, Waste Disposal and Agricultural Soil sites
 217 were excluded from the analysis. * Indicates significant correlation at $p < 0.05$; ** at $p < 0.01$; *** at
 218 $p < 0.001$. A single dot indicates slightly significant correlation at $p < 0.1$.

219

220

221 Moreover, we conducted a principal component analysis (PCA) in order to obtain information on the
 222 relationships among altitude, soil characteristics, and trace elements. This analysis was performed
 223 using the "prcomp" and "biplot" functions (e.g. Salerno et al., 2014; Salerno et al. 2016). All tests
 224 were implemented in the software R (R Development Core Team, 2010) with a significance level at
 225 $p < 0.05$. All data were normalized and autoscaled before implementing the PCA. As widely discussed
 226 by Jolliffe (2002) and Wilks (2006), the PCA is a purely geometrical technique and therefore there is
 227 no need for a statistical hypothesis: multivariate normality is not a critical assumption. This
 228 assumption is required only when inferential statistics are to be derived from the principal
 229 components.

230

231 3. Results

232 3.1. Soil properties and trace elements distribution along the altitudinal gradient

233 Most soils were developed in sandy or sandy loamy materials, almost free of coarse fragments, which
234 however became abundant in the deepest horizons (Table 1). The most common soil types changed
235 with altitude as normally observed in mountain areas. Podzols were common below subalpine forest
236 and *Rhododendron* sp., below 4500 m a.s.l., and were characterized by a strong Fe-Al oxi-hydroxide
237 redistribution from the bleached eluvial to the illuvial Bs/Bhs horizons. The Fe_o/Fe_d ratio was
238 relatively high along the soil profiles, in particular in the A horizon. TOC redistribution with depth
239 was not always visible, but a particularly high C/N characterized these soils. Other soil types were
240 the less developed Arenosols, Umbrisol, and Regosols (Table 1). Arenosols were mostly observed
241 below sparse high altitude vegetation or under cryoturbated hummocks.

242 In the agricultural land the topsoil was characterized by the highest TOC (16.6%). The pH and the
243 percentage of rocks were similar to the other topsoils. The topsoil sampled close to the landfill, where
244 the solid wastes are burnt in an open disposal, was characterized by high pH and TOC values. The
245 characteristics of the topsoil sampled close to Namche Bazar village (3570 m a.s.l.) were in general
246 similar to the other topsoils (Table 1).

247 There was no clear altitudinal trend of topsoil pH values, while topsoil TOC was inversely correlated
248 with elevation (i.e. from subalpine forest to alpine tundra, $\rho=-0.65$; $p<0.001$) (Fig. 2). Topsoil trace
249 elements showed a significant increasing trend with the elevation, except for Cu, Pb, Mn, and Fe (Fig.
250 2), and a peak in trace elements was measured at 4900-5000 m of altitude. TOC was not correlated
251 to most trace element contents in the topsoils, and a significant negative correlation between TOC
252 and Cr was observed ($\rho=-0.41$; $p<0.05$) (Fig. 2). No significant positive correlations were observed
253 between the considered trace elements and C/N, and only Co and Mn presented significant, but
254 negative correlations ($\rho=-0.51$; $p<0.05$ and $\rho=-0.64$; $p<0.05$, respectively) (Fig. 2). The soil pH

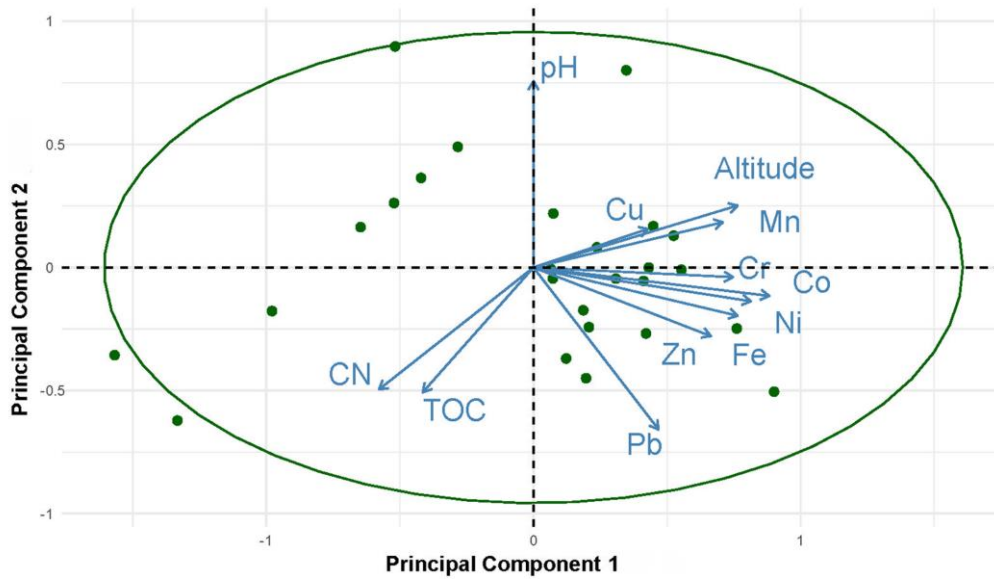
255 did not show any significant correlation with trace element contents as well, except with Pb ($\rho=$
256 0.45; $p<0.05$) (Fig. 2).

257 Concerning the soil profiles, in subalpine podzolic soils many trace elements showed very low
258 contents in the surface E horizon, and higher ones in the underlying illuvial Bs and Bhs horizons
259 (particularly Co, Cu, Ni, Table 2). The increasing trend of trace elements with soil depth was generally
260 not observed above about 4400 m of altitude, where non-podzolic soils showed higher contents in
261 surface A and lower in subsurface layers. The largest difference between trace element-“rich” surface
262 horizons and subsurface ones was measured in profile Z4955, where contents of most trace elements
263 were almost doubled in the surface. Above this altitude, the difference between surface and
264 subsurface horizons tended to disappear (Table 2).

265 Trace elements in the waste disposal showed a content of the selected elements below the critical
266 limits for contaminated soils reported by USEPA (2006) and only Cu, Zn, and Pb showed values
267 closer to the lower limit reported by this guideline. In both village and agricultural soils, trace element
268 contents were similar or even lower than in the other topsoils (Table 2).

269 In the PCA biplot shown in Fig. 3, PC1 (explaining 56% of the variance) was highly positively
270 associated with altitude and trace element contents, negatively with TOC and C/N (Table 3), and it
271 can thus be interpreted as an altitudinal gradient. Siderophile elements, such as Fe, Co, Ni, Cr, Mn,
272 with the addition of Zn, were very closely associated among them along the gradient described by
273 PC1, while Cu and Pb were less strongly related. PC2 (explaining 20% of the variance) was strongly
274 associated with soil pH and negatively with TOC, C/N and Pb; it showed the strong Pb bind with
275 TOC in surface horizons. The peculiarity of Cu content variations along the altitudinal gradient was
276 evidenced by axis 3, which was associated with this element and, weakly, with Pb (Table 3).

277



278

279 **Fig. 3.** PCA analysis biplot performed between altitude (alt), TOC, C/N, pH, trace elements, and Fe
 280 recorded in the topsoil along the altitudinal gradient (n=27). Village, Waste Disposal and Agricultural
 281 Soil sites were excluded from the analysis. Topsoil samples scores are identified by their position,
 282 and the length of the arrows refers to the factor loadings. The ellipse represents the 95% confidence
 283 interval for the data.

284

285

286 *3.2. Soil properties and trace elements distribution in the high-elevation toposequence*

287 The soil depth in the high-elevation toposequence ranged between 30 and 85 cm, revealing a high
 288 variability, and all soils showed well-developed sandy Bw horizons (Table 4). In all topographic
 289 positions, the proportion of stone fragments in the upper soil horizons was negligible. The sand and
 290 coarse silt fractions dominated with more than 80% in all horizons. The soil texture was sandy loam
 291 in the highest horizons of ridge and concave sites, while it was loamy sand in the slope site. Stone
 292 rich layers appeared at depths of about 40, 80, and 21 cm in the concave, slope, and ridge sites,
 293 respectively (Table 4).

294 At all sites the content of TOC decreased regularly along the soil profile. The mean TOC was higher
 295 in the concave and slope sites than in the ridge, as does the C/N (Table 4).

296 There were no significant differences in mean trace element contents among the three sites of the
297 toposequence. However, their depth distribution showed different patterns for the different elements
298 (Table 5). In all soils, the maximum values of Pb were recorded in subsurface horizons (AB horizons).
299 Cu, Zn, Cr and Ni content in the ridge zone were the highest in the upper soil horizons, and in the
300 slope and concave sites the Zn recorded the maximum values in the subsurface soil horizons, with a
301 decreasing trend at greater depths. In the latter sites, the maximum values of all the other trace
302 elements were recorded at greater depths, showing an accumulation in the subsoil. A relative
303 enrichment of trace elements in the surface horizons was not measured (Table 5).

304

305 **4. Discussion**

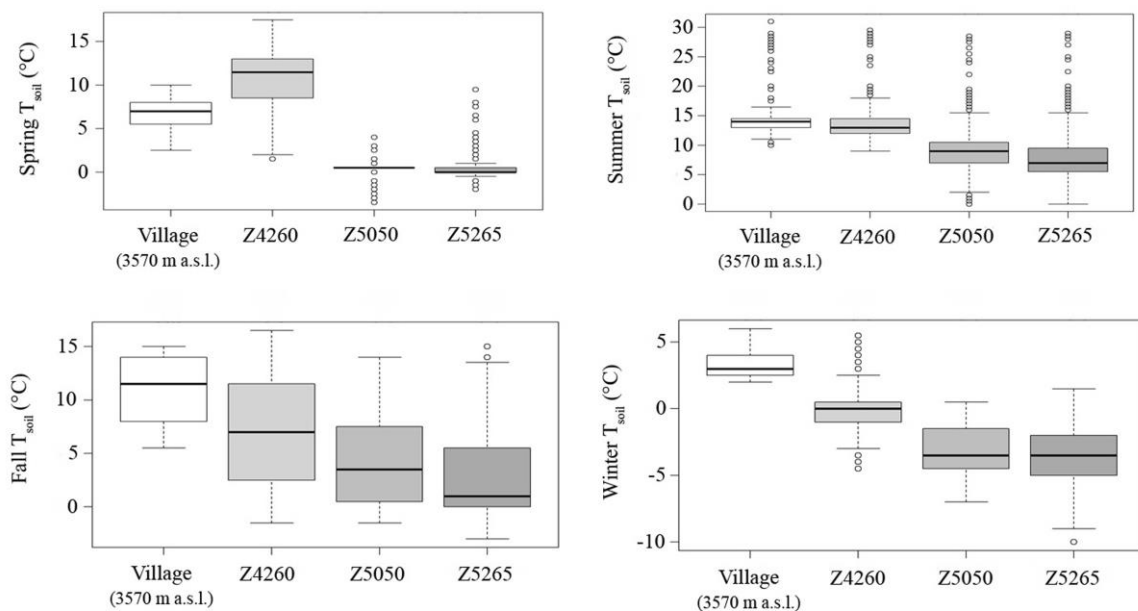
306 *4.1. Topsoils and soil profiles along the altitudinal gradient*

307 A pedogenic trend with altitude was observed from podzolic soils below subalpine forest and
308 *Rhododendron* sp. to Umbrisols, Regosols and Arenosols above the treeline (Righi and Lorphelin,
309 1987; Bäumler and Zech, 1994). The relatively high Fe_o/Fe_d ratio along most soil profiles revealed
310 the dominance of poorly crystalline Fe oxides and metal-organic complexes. A decreasing weathering
311 degree with altitude was shown, as expected (Table 1), by the decreasing Fe_d/Fe_t ratio; it showed the
312 highest values in subalpine Podzols and the lowest values in the other soil types (Arenosols,
313 Umbrisols, and Regosols), and it was associated with a weaker Fe and Al redistribution with depth.
314 The almost total absence of coarse fragments in the soil profiles can be considered as evidence of a
315 widespread presence of wind-blown loess-like materials, as already reported for high-elevation soils
316 in Eastern Nepal by Bäumler (2001) (Table 1).

317 The ecosystem gradient from subalpine forest to alpine grassland and tundra was associated with a
318 decreasing trend in topsoil TOC and an increase in the C/N ratio of the organic matter (Fig. 2), in
319 agreement with various studies showing an increasing trend with altitude from the montane to the
320 subalpine belt, followed by a strong decrease in the alpine one (e.g. Egli et al., 2008). The differences

321 along the altitudinal gradient reflected a changing balance of soil C inputs and soil C losses that are
 322 potentially related to changes in both abiotic (e.g. temperature) and biotic (e.g. litter quality) factors
 323 (Garten and Hanson, 2006; Egli et al., 2008). TOC was low at high altitudes (Table 1), as a result of
 324 harsh climate and short growing seasons limiting net primary productivity and C inputs to the soil
 325 above the treeline (Garcia-Pausas et al., 2007). In fact, we observed that the mean topsoil winter
 326 temperature drops below 0°C at elevations higher than 4000 m, with values close to -5°C at elevations
 327 above 5000 m (Fig. 4). Soil freezing and the consequent cryoturbation are strong limitations to plant
 328 growth (Körner, 2003).

329



330

331 **Fig. 4.** Topsoil mean temperature (T_{soil} , °C) during the different meteorological seasons (Spring,
 332 Summer, Fall, Winter) along the altitudinal gradient (from 3570 to 5265 m a.s.l.). Data recorded every
 333 four hours from July 2012 to October 2013.

334

335

336 Most of the limited available information on the distribution of trace elements in the Himalayas has
 337 been determined only in topsoil samples (e.g. Loewen et al., 2005; Li et al., 2008; Yeo and Langley-

338 Turnbaugh, 2010). In particular, trace element contents in topsoils measured in the southern side of
339 Mt. Everest (Table 2) were similar to those recorded in soils from other remote areas, such as the
340 Tibetan Plateau (Li et al., 2008) and the northern side of Mt. Everest (Yeo and Langley-Turnbaugh,
341 2010), and were below the critical limits for contaminated soils reported by USEPA (2006) at all
342 altitudes. Only the soil sampled close to the waste disposal showed element contents close to these
343 critical limits; trace elements released from the uncontrolled open burning of electronic waste could
344 penetrate the soils where vegetables and crops are grown by contaminating irrigation water and
345 through direct deposition by air (Luo et al., 2011).

346 Considering the larger number of topsoil trace elements data, we can see a peak in trace element
347 contents measured at 4900-5000 m of altitude. This peak did not correspond to the peak of
348 precipitation observed at 2500 m in this area (Salerno et al., 2015). We did not observe the
349 hypothesized vegetation interceptive effect (Reiners et al., 1975), since the forest sites showed the
350 lowest values of trace elements (Table 2). Our data agreed with those measured by Bergamaschi et
351 al. (2002), who found a slight increase of the enrichment factor for Br, Sb, Zn and Pb in foliose lichens
352 between 3800 and 5090 m in the same area. They supposed that some of these elements were
353 associated to the finest airborne particles and could be subjected to long transport phenomena in the
354 higher tropospheric layer.

355 Although organic matter is an efficient scavenger of trace elements (Reiners et al., 1975), TOC was
356 not correlated to most topsoil trace elements; only Pb was well associated with organic matter content
357 in PCA axis 2, while Cu was weakly negatively correlated with TOC, despite its high measured
358 contents in A horizons (Fig. 3). Soil organic matter with low C/N is usually considered to decompose
359 more rapidly than organic matter with high C/N (Finzi et al., 1998), and the fast turnover of organic
360 matter may cause a rapid loss of trace elements (e.g. Pb) from soils (Watmough et al. 2005). The
361 absence of significant positive correlations between trace elements and C/N, and between trace
362 elements and pH (Fig. 2), revealed that the sorption potential determined by the organic matter content

363 and quality, and soil pH as factors influencing the mobility of trace elements, were not generalized
364 causes of the differences in elemental contents in the topsoils, and their influence was different on
365 each trace element, as also reported by Bacardit and Camarero (2010). Pedogenic processes, on the
366 contrary, have a strong impact on trace element content in topsoils, and the leaching caused by
367 cheluviation in subalpine podzolic soils can explain the low values below treeline.

368 The depth trend of trace elements throughout the soil profile is less known. Depth trends can be used
369 to detect possible surface contamination or to assess their mobility in the soil profile, associated with
370 soil properties such as structure or with pedogenic processes (Li et al., 2014). In general, the relative
371 enrichment of trace elements in the upper soil horizons implies the contributions of anthropogenic
372 inputs (Li et al., 2014). Our results showed that, in subalpine podzolic soils, most siderophile elements
373 (i.e. Ni, Cr, Co, Mn), but also Cu and Zn, were subjected to leaching in podzolic soils (e.g. D'Amico
374 et al., 2008). The lowest values observed in topsoils of podzols can be ascribed to cheluviation
375 processes. Cu often showed the highest values in A horizons, in line with the capacity of organic
376 matter to bind this element (Alloway, 2013), or possibly with anthropogenic enrichment. In other soil
377 types, no significant increase in deep layers was observed (Table 2).

378 The trace element depth trend in alpine soils, showing the highest values in surface horizons,
379 particularly around 5000 m of elevation (Table 2), could be caused by air-borne contamination. In
380 fact, air-borne contamination cannot be excluded, and is maybe driven by particular micro-climatic
381 or topographic conditions (e.g. Bergamaschi et al. 2002; Cong et al., 2015). An increase in Pb content
382 with altitude has been recorded in other high-elevation areas such as in soils across the Pyrenees
383 (McGee and Vallejo, 1996) and on northeastern United States mountains in New Hampshire (Reiners
384 et al., 1975). Several studies in the European Alps suggested that there is generally an increase in
385 deposition as altitude increased (Camarero et al., 2009), but other studies (e.g. Kang et al., 2007)
386 found no obvious trends in trace element accumulation with altitude. Thus, these pristine ecosystems
387 can undergo long-range atmospheric contamination (Bacardit and Camarero, 2010; Cong et al.,

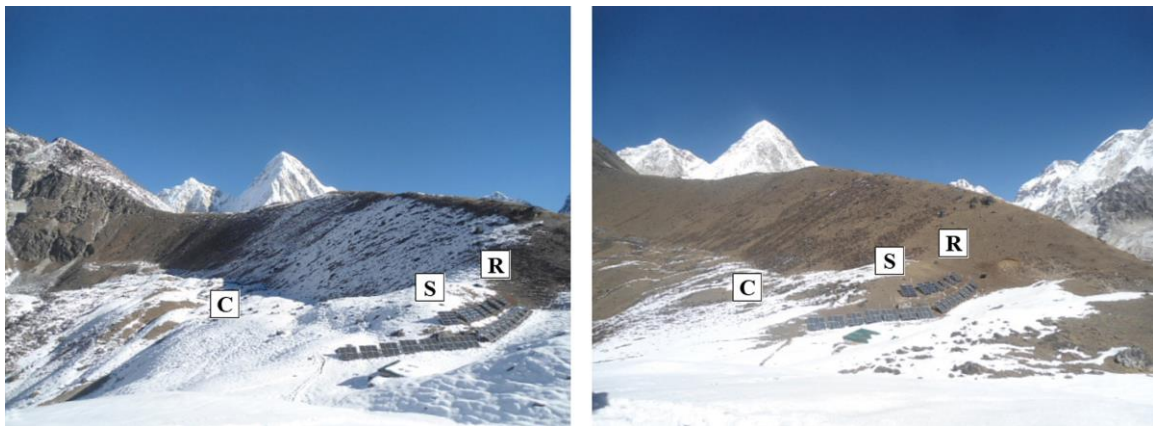
388 2010). Soil trace element deposition levels are especially influenced by precipitation amounts and
389 high mountains are regional convergence zones that trap atmospheric contaminants as a result of cold
390 condensation and enhanced atmospheric deposition (Zechmeister, 1995; Wegman et al., 2006). A
391 stronger air born contamination (possibly associated with higher rainfall) could however be masked
392 in lower altitude soils, where pedogenic processes lead to leaching from surface layers.

393

394 4.2 High-elevation toposequence

395 Soil development in high mountain regions is strongly controlled by processes of weathering and
396 cryoturbation, which in turn are significantly influenced by the topographic conditions associated
397 with different humidity, wind, snow cover depth, and duration (Fig. 5). In fact, rugged surfaces shape
398 snow deposition and translocation patterns on slopes, affecting the freeze-thaw cycles and melt-water
399 runoff (Stres et al., 2013), thus influencing soil development degree and chemical properties.

400



401

402 **Fig. 5.** Snow distribution patterns in the high-elevation toposequence (R: ridge; S: slope; C: concave).
403 On the left: October 24th, 2013. On the right: November 11th, 2013.

404

405 All soils were characterized by well-developed sandy Bw horizons (Table 4), in contrast with that
406 reported by Bäumler and Zech (1994), who indicated the prevalence of shallow Regosols in the alpine
407 mountain zone above the forest line and in the zone of alpine turf vegetation. The presence of stone-

408 rich bottom layers at all sites evidenced a depositional discontinuity explained by a widespread
409 presence of wind-blown materials (Bäumler 2001). The clear increasing trend in pH values with soil
410 depth (Table 4) could imply a multi-layer stratigraphy of the solum (Bäumler, 2001), but could be
411 easily explained also by leaching during the monsoon season and to the acidifying effect of
412 *Rhododendron* shrubs (Guggenberger et al., 1998). The Fed/Fet ratio evidenced a higher weathering
413 degree at the slope and concave sites than at the ridge one, probably because of the greater erosion
414 intensity in the latter site (Table 4). In the ridge site, the Fed/Fet decreased with increasing soil depth,
415 while in the other two sites local maxima were located in the subsoil. The greater Feo/Fed ratio in the
416 AB and B horizons at all sites revealed a weathering trend leading to the formation of amorphous and
417 poorly crystalline iron oxides. According to Bäumler (2001) the Fed, which ranged between 1 and 7
418 g kg⁻¹, reveals how the deposits are of Late Pleistocene origin or younger, since older soils of
419 interglacial origin have much higher amounts of well-crystallized iron oxides up to 40 g kg⁻¹ in eastern
420 Nepal.

421 The TOC and C/N were higher in the concave and slope sites than in the ridge, revealing a higher
422 degree of organic matter mineralization in the ridge site or a lower TOC input associated with a
423 sparser vegetation cover devoid of *Rhododendron* shrubs (Table 4).

424 Water fluxes are also important in element leaching or accumulation through the soil. In particular,
425 snowmelt or monsoon precipitation can cause high water contents in soils and rapid water flow in
426 preferential pathways, with a potential reduction of retention by sorption (Bacardit and Camarero,
427 2010). Thus, the observation of trace element contents in soils developed across a toposequence
428 characterized by different snow accumulation and wind processes can give insights on their behavior
429 in these high-elevation Himalayan soils. In fact, the coarse-texture of these soils could facilitate the
430 downward migration of trace elements resulting in an accumulation at greater soil depth (Li et al.,
431 2014). Water is the vehicle for transport of solutes, including trace elements, through soil. The always

432 lower content in the stone rich deep horizons is likely associated with a mineralogical discontinuity
433 caused by the thick aeolian layer (Table 5) (Guggenberger et al., 1998).

434

435 **5. Conclusions**

436 Soils types and pedogenic processes change along the altitudinal gradient in the Dudh Koshi Valley.

437 At elevations higher than 4000 m a.s.l. soil temperatures below 0°C were reported, which caused soil
438 freezing and consequent cryoturbation. Thus, the plant growth was limited at high elevations and it
439 was reflected in the negative correlations found between altitude and TOC.

440 Trace element contents along the altitudinal gradient were similar to those recorded in soils from
441 other remote areas, with the exception of the area close to the open waste disposal where higher values
442 were recorded, but still below the USEPA (2006) thresholds, revealing a potential localized source
443 of contamination. Contents of Co, Zn, Cr, and Ni, in the topsoil were positively correlated with the
444 altitude, as reported in other mountain areas, a phenomenon potentially ascribable to the orographic
445 effect on trace element deposition and/or to different biogeochemical processes leading to leaching
446 at lower altitudes. Among the soil characteristics influencing the mobility of trace elements along the
447 altitudinal gradient, both organic matter and pH seemed not to be determinant of the differences in
448 elemental contents in the topsoils. The degree of weathering and pedogenesis influenced the
449 distribution of trace elements through the soil profiles. In the lower elevation podzolic soils, low
450 surface concentrations of trace elements were measured, with higher contents in the illuvial Bs and
451 Bhs horizons. The difference between surface and subsurface horizons tended to disappear among
452 soil profiles with the increase in elevation. No significant differences in mean trace element contents
453 were found in the high-elevation toposequence despite the observed different patterns of depth
454 distribution.

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| Sample | Altitude | Classification | Horizon | Depth | Color | Stones | TOC | C/N | pH | Fed | Feo | Feo/Fed | Fed/Fet | Alo | IS |
|--------|----------|----------------------------------|---------|--------|------------|--------|-----|------|-----|--------------------|--------------------|---------|---------|--------------------|-----|
| | m a.s.l. | IUSS 2015 | | cm | | % vol | % | | | g kg ⁻¹ | g kg ⁻¹ | | | g kg ⁻¹ | |
| Z3700 | 3700 | Albic Podzol | A1 | 0-7 | 7.5 YR 2/3 | 5.0 | 7.6 | 19.0 | 5.4 | 12.6 | 12.0 | 1.0 | 0.7 | 9.5 | 1.6 |
| | | | A2 | 7-11 | 7.5 YR 4/3 | 5.0 | 2.9 | 29.0 | 6.2 | 8.4 | 6.4 | 0.8 | 0.7 | 3.7 | 0.7 |
| | | | E1 | 11-18 | 7.5 YR 5/2 | 5.0 | 2.6 | 26.0 | 6.1 | 4.0 | 2.7 | 0.7 | 0.6 | 2.0 | 0.3 |
| | | | Bs | 18-25 | 10 YR 5/6 | 5.0 | 2.4 | 24.0 | 5.9 | 17.1 | 15.1 | 0.9 | 0.9 | 4.9 | 1.3 |
| Z3750 | 3750 | Ortsteinic Albic Podzol (Arenic) | A | 3-9 | 10 YR 5/3 | 0.0 | 4.5 | 22.5 | 5.2 | 3.5 | 3.3 | 0.9 | 0.8 | 0.6 | 0.2 |
| | | | E | 9-17 | 10 YR 5/4 | 0.0 | 1.5 | 15.0 | 5.0 | 3.4 | 1.8 | 0.5 | 0.6 | 0.8 | 0.2 |
| | | | Bhs | 17-43 | 10 YR 4/6 | 0.0 | 4.5 | 45.0 | 5.3 | 21.9 | 14.2 | 0.6 | 0.8 | 18.4 | 2.6 |
| | | | BC | 43-60+ | 10 YR 5/4 | 0.0 | 1.4 | 14.0 | 5.6 | 7.0 | 4.6 | 0.7 | 0.4 | 7.8 | 1.0 |
| Z3865 | 3865 | | A | 0-10 | 10 YR 3/3 | 1.5 | 9.4 | 18.0 | 4.5 | | | | | | |
| Z4260 | 4260 | Brunic Dystric Arenosol | A | 0-10 | 7.5 YR 2/2 | 0.0 | 6.5 | 10.8 | 5.5 | 2.5 | 2.3 | 0.9 | 0.4 | 1.3 | 0.3 |
| | | | AB | 10-20 | 10 YR 6/4 | 0.0 | 0.7 | 7.0 | 5.8 | 1.1 | 0.6 | 0.6 | 0.1 | 0.6 | 0.1 |
| | | | Bw | 20-30 | 7.5 YR 6/6 | 0.0 | 0.7 | 7.0 | 5.9 | 1.1 | 0.9 | 0.8 | 0.1 | 0.9 | 0.1 |
| Z4485 | 4485 | Brunic Dystric Arenosol | A | 0-7 | 10 YR 3/4 | 5.0 | 7.4 | 14.8 | 5.9 | 3.9 | 3.2 | 0.8 | 0.3 | 1.5 | 0.3 |
| | | | Bw | 7-10+ | 10 YR 5/3 | 0.0 | 1.9 | 19.0 | 5.1 | 2.5 | 1.9 | 0.8 | 0.2 | 1.3 | 0.3 |
| Z4680 | 4680 | | AC | 0-10 | 10 YR 4/3 | 37.9 | 4.1 | 15.1 | 6.0 | | | | | | |
| Z4880 | 4880 | | AC | 0-10 | 10 YR 7/2 | 38.5 | 0.5 | 10.9 | 6.5 | | | | | | |
| Z4940a | 4940 | | A | 0-10 | 10 YR 4/3 | 0.0 | 3.7 | 14.4 | 5.1 | | | | | | |
| Z4940b | 4940 | | A | 0-10 | 10 YR 4/3 | 0.9 | 5.1 | 13.2 | 5.9 | | | | | | |
| Z4940c | 4940 | | A | 0-10 | 10 YR 5/4 | 6.4 | 3.5 | 15.5 | 5.8 | | | | | | |
| Z4950 | 4950 | | A | 0-10 | 10 YR 4/3 | 0.4 | 3.3 | 14.3 | 6.0 | | | | | | |
| Z4955 | 4955 | Dystric Arenosol | A | 0-7 | 10 YR 4/2 | 10.0 | 3.4 | 11.3 | 5.0 | 2.5 | 1.1 | 0.5 | 0.1 | 0.8 | 0.1 |
| | | | C | 7-20 | 10 YR 5/2 | 10.0 | 0.1 | 4.7 | 5.6 | 0.6 | 0.2 | 0.4 | 0.1 | 0.2 | 0.0 |
| Z4960 | 4960 | | A | 0-10 | 10 YR 5/3 | 0.0 | 0.1 | 4.9 | 6.0 | | | | | | |

| | | | | | | | | | | | | | | | | |
|-------------------|------|----------------------------------|----|-------|------------|------|------|------|-----|------|------|-----|-----|------|-----|--|
| Z4965 | 4965 | | A | 0-10 | 10 YR 3/3 | 3.3 | 5.9 | 12.6 | 5.8 | | | | | | | |
| Z5050 | 5050 | | A | 0-10 | 10 YR 2/3 | 0.0 | 4.5 | 16.1 | 5.4 | | | | | | | |
| Z5100 | 5100 | | A | 0-10 | 10 YR 3/3 | 0.0 | 4.1 | 14.0 | 5.2 | | | | | | | |
| Z5170 | 5170 | | A | 0-10 | 10 YR 4/3 | 0.0 | 3.3 | 15.0 | 5.5 | | | | | | | |
| Z5245 | 5245 | | A | 0-10 | 10 YR 5/3 | 0.0 | 1.3 | 12.7 | 5.5 | | | | | | | |
| Z5250 | 5250 | | A | 0-10 | 10 YR 4/3 | 1.5 | 2.6 | 13.0 | 5.5 | | | | | | | |
| Z5260 | 5260 | | A | 0-7 | 10 YR 2/2 | 5.8 | 2.9 | 13.7 | 5.4 | | | | | | | |
| Z5265 | 5265 | | A | 0-10 | 10 YR 3/4 | 20.0 | 2.8 | 15.1 | 5.4 | | | | | | | |
| Z5280 | 5280 | Haplic Umbrisol (Arenic, Turbic) | A1 | 0-7 | 10 YR 2/3 | 20.0 | 2.9 | 14.5 | 5.2 | 5.2 | 2.6 | 0.5 | 0.4 | 1.7 | 0.3 | |
| | | | A2 | 7-20 | 10 YR 3/3 | 0.0 | 2.3 | 11.5 | 5.5 | 6.3 | 2.6 | 0.4 | 0.2 | 0.2 | 0.2 | |
| | | | AB | 20-30 | 10 YR 3/3 | 0.0 | 2.0 | 20.0 | 5.9 | 6.9 | 3.8 | 0.6 | 0.4 | 0.3 | 0.2 | |
| Z5300 | 5300 | Brunic Dystric Arenosol | A | 0-10 | 10 YR 5/3 | 0.0 | 1.3 | 13.0 | 6.2 | 1.3 | 0.5 | 0.4 | 0.1 | 0.7 | 0.1 | |
| | | | AB | 10-20 | 10 YR 6/3 | 0.0 | 0.7 | 7.0 | 5.9 | 1.2 | 0.6 | 0.5 | 0.0 | 0.8 | 0.1 | |
| | | | Bw | 20-25 | 10 YR 6/3 | 10.0 | 0.6 | 6.0 | 6.4 | 1.5 | 0.6 | 0.4 | 0.1 | 1.0 | 0.1 | |
| Z5320 | 5320 | | A | 0-7 | 10 YR 6/1 | 28.1 | 0.2 | 8.8 | 5.9 | | | | | | | |
| Village | 3570 | Dystric Regosol | A | 0-10 | 7.5 YR 3/3 | 2.0 | 6.7 | 13.4 | 5.8 | 15.5 | 8.2 | 0.5 | 0.5 | 9.9 | 1.4 | |
| | | | AC | 10-30 | 10 YR 3/4 | 2.0 | 4.6 | 15.3 | 5.7 | 19.0 | 12.0 | 0.6 | 0.6 | 15.3 | 2.1 | |
| Waste disposal | 3700 | | A | 0-10 | 10 YR 4/2 | 0.0 | 5.3 | 22.1 | 8.3 | | | | | | | |
| Agricultural soil | 4405 | | A | 0-10 | 10 YR 2/3 | 1.7 | 16.6 | 11.9 | 5.7 | | | | | | | |

Table 1. Topsoil and soil profile characteristics along the altitudinal gradient. IS: index of spodicity ($1/2 * Fe_o + Al_o$). Stone content (% vol) was estimated qualitatively in the field.

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| Sample | Altitude m a.s.l. | Horizon | Co | Cu | Zn | Cr | Pb | Ni | Mn | Fe |
|--------|----------------------|---------|------|------|------|---------------------|------|------|-------|--------------------|
| | | | | | | mg kg ⁻¹ | | | | g kg ⁻¹ |
| Z3700 | 3700 | A1 | 4.3 | 9.2 | 15.5 | 1.8 | 8.0 | 9.8 | 118.0 | 17.4 |
| | | A2 | 2.5 | 5.2 | 8.0 | 0.0 | 10.5 | 4.3 | 81.0 | 12.3 |
| | | E1 | 2.6 | 6.3 | 11.0 | 3.8 | 11.4 | 4.8 | 92.3 | 6.8 |
| | | Bs | 2.9 | 8.7 | 10.0 | 5.9 | 18.5 | 5.4 | 83.0 | 18.3 |
| Z3750 | 3750 | A | 2.0 | 6.2 | 9.3 | 0.0 | 7.5 | 4.0 | 53.0 | 4.4 |
| | | E | 1.7 | 1.8 | 9.7 | 5.4 | 29.7 | 0.5 | 54.0 | 6.1 |
| | | Bhs | 7.0 | 7.9 | 35.0 | 23.9 | 29.2 | 17.5 | 163.0 | 29.0 |
| | | BC | 7.6 | 7.7 | 37.9 | 21.4 | 25.9 | 20.0 | 218.1 | 19.5 |
| Z3865 | 3865 | A | 2.5 | 9.4 | 10.5 | 4.3 | 7.0 | 7.3 | 59.4 | 7.5 |
| Z4260 | 4260 | A | 3.5 | 6.4 | 16.3 | 5.8 | 3.5 | 9.9 | 90.0 | 7.0 |
| | | AB | 4.9 | 5.1 | 20.1 | 6.6 | 7.8 | 8.2 | 98.0 | 8.5 |
| | | Bw | 5.5 | 5.4 | 20.4 | 7.1 | 7.9 | 8.6 | 120.0 | 8.6 |
| Z4485 | 4485 | A | 5.5 | 11.0 | 23.8 | 8.8 | 4.5 | 11.0 | 171.0 | 12.4 |
| | | Bw | 4.8 | 8.4 | 30.9 | 7.2 | 9.2 | 9.2 | 119.0 | 10.6 |
| Z4680 | 4680 | AC | 5.8 | 10.0 | 28.8 | 9.5 | 5.0 | 11.0 | 293.0 | 10.8 |
| Z4880 | 4880 | AC | 4.5 | 9.8 | 20.0 | 6.3 | 4.0 | 8.3 | 183.0 | 9.9 |
| Z4940a | 4940 | A | 8.5 | 14.8 | 31.3 | 17.3 | 6.5 | 16.8 | 260.5 | 17.9 |
| Z4940b | 4940 | A | 10.3 | 12.5 | 42.5 | 18.3 | 11.3 | 17.8 | 275.5 | 20.0 |
| Z4940c | 4940 | A | 13.3 | 14.6 | 45.0 | 43.3 | 14.0 | 24.3 | 250.0 | 29.4 |
| Z4950 | 4950 | A | 10.0 | 9.3 | 37.5 | 24.1 | 10.1 | 12.0 | 306.3 | 24.5 |
| Z4955 | 4955 | A | 10.5 | 10.3 | 42.5 | 37.3 | 11.9 | 14.3 | 266.0 | 20.6 |
| | | C | 4.8 | 7.3 | 27.5 | 6.8 | 6.5 | 8.3 | 79.0 | 8.9 |
| Z4960 | 4960 | A | 6.5 | 6.6 | 26.3 | 22.3 | 2.1 | 13.0 | 173.8 | 15.1 |
| Z4965 | 4965 | A | 10.3 | 16.1 | 33.8 | 22.8 | 7.5 | 20.3 | 344.3 | 21.6 |

| | | | | | | | | | | |
|-------------------|------|----|------|-------|--------|------|-------|------|-------|------|
| Z5050 | 5050 | A | 11.0 | 9.9 | 40.0 | 25.3 | 10.0 | 18.0 | 248.8 | 18.1 |
| Z5100 | 5100 | A | 9.3 | 9.6 | 41.3 | 23.0 | 8.9 | 14.0 | 242.5 | 24.0 |
| Z5170 | 5170 | A | 8.3 | 8.8 | 38.8 | 20.8 | 9.0 | 14.3 | 266.8 | 18.4 |
| Z5245 | 5245 | A | 11.0 | 10.6 | 36.3 | 30.8 | 7.9 | 19.0 | 273.8 | 24.5 |
| Z5250 | 5250 | A | 11.3 | 9.3 | 35.0 | 25.0 | 10.1 | 15.8 | 283.8 | 25.9 |
| Z5260 | 5260 | A | 11.3 | 12.6 | 37.3 | 36.3 | 9.0 | 20.0 | 271.3 | 21.3 |
| Z5265 | 5265 | A | 8.5 | 9.9 | 35.0 | 24.0 | 8.0 | 15.3 | 221.3 | 17.0 |
| Z5280 | 5280 | A1 | 10.8 | 11.6 | 41.3 | 29.3 | 9.8 | 18.5 | 288.0 | 14.3 |
| | | A2 | 10.5 | 11.9 | 37.5 | 26.5 | 10.3 | 19.0 | 270.0 | 27.5 |
| | | AB | 11.3 | 10.7 | 59.3 | 19.0 | 18.6 | 20.6 | 365.0 | 16.6 |
| Z5300 | 5300 | A | 6.0 | 8.0 | 36.3 | 7.5 | 7.3 | 8.8 | 221.0 | 11.8 |
| | | AB | 6.7 | 9.4 | 48.6 | 11.2 | 11.6 | 12.2 | 199.1 | 24.1 |
| | | Bw | 7.9 | 8.0 | 49.4 | 12.2 | 13.6 | 12.7 | 227.0 | 11.7 |
| Z5320 | 5320 | A | 9.8 | 12.3 | 23.8 | 34.1 | 1.9 | 18.3 | 217.5 | 18.1 |
| Village | 3570 | A | 9.5 | 14.9 | 37.5 | 20.3 | 20.3 | 20.8 | 273.0 | 28.5 |
| | | AC | 10.1 | 13.9 | 51.7 | 19.6 | 20.8 | 22.8 | 270.0 | 32.0 |
| Waste disposal | 3700 | A | 18.5 | 428.5 | 1360.0 | 90.0 | 241.5 | 45.0 | 868.0 | 34.5 |
| Agricultural soil | 4405 | A | 2.5 | 19.5 | 12.5 | 4.0 | 2.5 | 15.5 | 56.2 | 6.2 |

716 **Table 2.** Topsoil and soil profile trace element contents along the altitudinal gradient.

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| | Biplot scores | | |
|-------------------------------|---------------|--------|--------|
| | PC1 | PC2 | PC3 |
| Eigenvalue | 6.697 | 2.409 | 0.893 |
| Cumulative variance explained | 0.558 | 0.759 | 0.833 |
| Altitude | 0.998 | 0.453 | 0.160 |
| TOC | -0.565 | -0.829 | -0.376 |
| C/N | -0.441 | -0.906 | 0.164 |
| pH | 0.180 | 0.843 | -0.163 |
| Co | 1.152 | -0.157 | -0.058 |
| Cu | 0.737 | -0.331 | -0.790 |
| Zn | 1.068 | -0.117 | 0.288 |
| Cr | 1.060 | -0.055 | 0.064 |
| Pb | 0.609 | -0.819 | 0.439 |
| Ni | 1.058 | -0.169 | -0.326 |
| Mn | 1.038 | 0.056 | -0.061 |
| Fe | 1.014 | -0.242 | 0.043 |

719 **Table 3.** PCA eigenvalues, variance explained, and biplot scores of the main soil properties and trace elements in topsoil samples (KMO measure of
720 sampling adequacy = 0.7). Village, waste disposal and agricultural soil sites were excluded from the analysis.

| Sample | Altitude m a.s.l. | Classification IUSS 2015 | Horizon | Depth cm | Stones % vol | Color | TOC % | C/N | pH | Fed g kg ⁻¹ | Feo g kg ⁻¹ | Feo/Fed % | Fed/Fet % | Alo g kg ⁻¹ | Clay % | Fine silt % | Coarse silt % | Fine sand % | Coarse sand % |
|-------------|----------------------|--|-----------|-------------|-----------------|-----------|----------|------|------|---------------------------|---------------------------|--------------|--------------|---------------------------|-----------|----------------|------------------|----------------|------------------|
| Concave (C) | 5055 | Dystric Cambisol (Loamic, Raptic) | A | 0-9 | 0 | 10 YR 4/3 | 4.9 | 13.2 | 5.2 | 4.3 | 2.5 | 57.9 | 22.1 | 1.4 | 3.1 | 8.9 | 18.1 | 58.7 | 11.2 |
| | | | AB | 9-18 | 0 | 10 YR 4/4 | 2.1 | 12.1 | 5.4 | 7.4 | 3.9 | 53.0 | 32.1 | 1.8 | 2.9 | 11.3 | 20.3 | 57.6 | 8.0 |
| | | | Bw1 | 18-35 | 0 | 10 YR 4/3 | 1.9 | 12.3 | 5.5 | 6.4 | 4.8 | 75.5 | 29.9 | 2.8 | 2.9 | 8.2 | 15.0 | 62.8 | 11.1 |
| | | | Bw2 | 35-40 | 0 | 10 YR 4/3 | 1.5 | 14.2 | 5.6 | 4.2 | 2.5 | 59.3 | 22.6 | 3.2 | 2.1 | 5.6 | 7.6 | 58.2 | 26.5 |
| | | | 2BC | 40-45+ | 20 | 10 YR 4/3 | 1.3 | 13.9 | 5.7 | 3.2 | 1.9 | 60.4 | 17.1 | 2.8 | 2.3 | 5.4 | 5.9 | 52.9 | 33.4 |
| Slope (S) | 5065 | Brunic Dystric Arenosol (Aeolic, Raptic) | A1 | 0-3 | 0 | 10 YR 3/3 | 6.1 | 16.8 | 5.3 | 4.3 | 2.0 | 47.2 | 22.4 | 1.2 | 1.5 | 5.0 | 14.1 | 49.5 | 29.9 |
| | | | A2 | 3-9 | 0 | 10 YR 4/3 | 4.7 | 15.8 | 5.3 | 5.4 | 2.2 | 41.4 | 25.4 | 1.3 | 1.6 | 7.7 | 14.1 | 56.0 | 20.6 |
| | | | A3 | 9-19 | 0 | 10 YR 3/4 | 3.4 | 15.0 | 5.6 | 5.6 | 2.8 | 50.1 | 26.8 | 1.8 | 1.8 | 9.1 | 16.1 | 61.3 | 11.7 |
| | | | A4 | 19-30 | 0 | 10 YR 3/3 | 3.4 | 15.9 | 5.8 | 7.0 | 2.6 | 37.2 | 34.3 | 1.8 | 2.8 | 10.8 | 20.6 | 58.1 | 7.8 |
| | | | AB1 | 30-50 | 0 | 10 YR 3/4 | 2.4 | 13.8 | 5.9 | 6.5 | 3.3 | 51.7 | 30.4 | 2.3 | 2.3 | 10.9 | 14.3 | 62.9 | 9.5 |
| | | | AB2 | 50-60 | 0 | 10 YR 4/3 | 2.3 | 17.5 | 5.9 | 6.3 | 4.0 | 64.4 | 33.2 | 3.1 | 1.7 | 8.6 | 11.0 | 65.7 | 13.0 |
| | | | BC | 60-80 | 0 | 10 YR 4/3 | 1.2 | 12.4 | 5.8 | 3.3 | 1.8 | 52.8 | 14.5 | 2.3 | 1.5 | 6.7 | 6.4 | 56.9 | 28.5 |
| 2C | 80-85+ | 50 | 10 YR 5/3 | 0.7 | 11.0 | 6.0 | 1.1 | 0.8 | 65.9 | 10.9 | 1.5 | 2.4 | 12.6 | 7.0 | 34.0 | 44.0 | | | |
| Ridge (R) | 5070 | Dystric Cambisol (Loamic, Raptic) | A1 | 0-5 | 0 | 10 YR 3/3 | 3.2 | 10.7 | 5.4 | 5.2 | 1.3 | 24.3 | 21.8 | 1.6 | 3.3 | 12.4 | 20.6 | 51.3 | 12.4 |
| | | | A2 | 5-12 | 0 | 10 YR 4/3 | 2.6 | 10.6 | 5.7 | 4.0 | 1.8 | 44.5 | 17.1 | 2.2 | 4.1 | 12.4 | 16.8 | 57.5 | 9.3 |
| | | | AB | 12-21 | 0 | 10 YR 4/4 | 2.3 | 11.2 | 5.8 | 3.9 | 2.3 | 57.9 | 18.0 | 2.4 | 3.9 | 11.0 | 15.0 | 56.4 | 13.8 |
| | | | 2Bw | 21-30+ | 40 | 10 YR 5/4 | 0.8 | 10.1 | 6.0 | 1.0 | 0.6 | 56.7 | 9.3 | 2.1 | 2.0 | 13.2 | 4.6 | 31.2 | 48.9 |

722 **Table 4.** Soil characteristics in the high-elevation toposequence. Stone content (% stones) was estimated qualitatively in the field.

| Sample | Altitude m a.s.l. | Horizon | Co | Cu | Zn | Cr mg kg ⁻¹ | Pb | Ni | Mn | Fe g kg ⁻¹ |
|----------------|----------------------|---------|------|------|------|---------------------------|------|------|-------|--------------------------|
| Concave (C) | 5055 | A | 8.8 | 7.9 | 58.8 | 11.3 | 13.8 | 16.3 | 241.0 | 19.6 |
| | | AB | 12.5 | 7.3 | 63.5 | 12.0 | 16.8 | 13.0 | 524.0 | 23.0 |
| | | Bw1 | 12.3 | 9.8 | 54.8 | 10.0 | 16.3 | 12.3 | 464.1 | 21.3 |
| | | Bw2 | 11.0 | 9.8 | 56.0 | 15.3 | 12.3 | 16.8 | 268.0 | 18.8 |
| | | BC | 9.3 | 9.0 | 44.8 | 13.5 | 10.0 | 15.3 | 243.0 | 18.9 |
| Slope (S) | 5065 | A1 | 9.3 | 9.4 | 60.0 | 14.0 | 11.3 | 16.0 | 280.0 | 19.3 |
| | | A2 | 9.0 | 8.9 | 68.8 | 15.5 | 11.0 | 16.0 | 256.4 | 21.3 |
| | | A3 | 8.8 | 8.1 | 61.3 | 12.5 | 11.0 | 14.0 | 229.0 | 21.0 |
| | | A4 | 8.5 | 8.1 | 43.8 | 13.3 | 13.5 | 14.3 | 243.0 | 20.4 |
| | | AB1 | 8.0 | 9.0 | 51.0 | 11.5 | 14.3 | 12.0 | 249.2 | 21.3 |
| | | AB2 | 7.3 | 7.5 | 38.5 | 9.0 | 12.0 | 13.3 | 190.1 | 18.9 |
| | | BC | 10 | 9.8 | 36.0 | 20.5 | 11.3 | 19.3 | 240.0 | 23.0 |
| | | C | 3.8 | 4.8 | 21.0 | 8.5 | 6.8 | 11.3 | 94.0 | 10.5 |
| Ridge (R) | 5070 | A1 | 12.2 | 10.4 | 62.5 | 15.0 | 14.8 | 22.5 | 311.0 | 23.9 |
| | | A2 | 12.3 | 9.6 | 57.5 | 12.8 | 13.5 | 20.5 | 303.0 | 23.1 |
| | | AB | 11.0 | 10.3 | 51.0 | 11.3 | 16.5 | 14.5 | 259.3 | 21.6 |
| | | Bw | 4.5 | 5.3 | 16.0 | 4.3 | 8.5 | 10.3 | 96.1 | 10.4 |

724 **Table 5.** Soil trace elements in the high-elevation toposequence.