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2 **Spatial Distribution and Temporal Trend of Airborne Trace Metal Deposition in**
3 **Albania Studied by Moss Biomonitoring**
4

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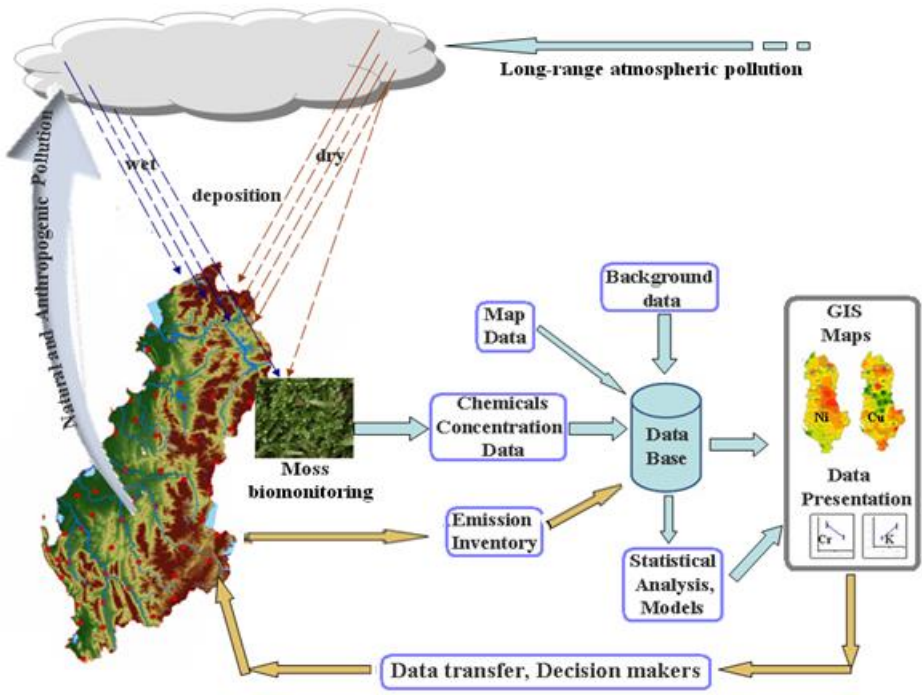
24 **ABSTRACT**

25 This study deals with the assessment of air quality in Albania by using the method of moss
26 biomonitoring. Bryophyte moss (a traditional bio-indicator), *Hypnum cupressiforme* (Hedw)
27 *sps.* was used in this study. Mosses were collected during August and September 2015 at
28 55 sampling sites homogeneously distributed over the entire territory of the country. The
29 spatial distribution and temporal trend of trace metal atmospheric deposition was studied
30 through the determination of twenty inorganic elements, Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe,
31 Hg, K, Li, Mg, Mn, Na, Ni, Pb, Sr, V, and Zn, in moss samples. ICP-AES, AAS (As and Cd)
32 and CV AAS (Hg) analysis were used to determine the concentration of the elements. The
33 obtained data show significant differences in spatial distribution of the elements that derived
34 from different emission sources. High emission loads of anthropogenic elements (Cr, Ni, Cu,
35 As and Zn) were detected in the East, and of sea salt elements, particularly Na and K, in

36 the Western coastal line. The distribution trend of the trace metals was studied by
37 comparing the current data with the data of a similar study performed in 2010. Due to
38 different factors, particularly the meteorological conditions and seasonal changes that
39 differed from year to year, the 2015 concentration data of Cr, Ni, Cu, As, Zn and Na were
40 increased, while Ba, K, Mg and Hg moss concentration data were declined. Other elements
41 remained mostly at the same level. The contamination levels that were evaluated by the
42 values of the contamination factor (CF) of each element provided similar results for current
43 study and 2010 moss survey study. Factor analysis (FA) was applied to identify the
44 possible sources of elements in current moss samples. Five dominant factors were
45 extracted from FA that represent long-range and local atmospheric transport of wind
46 blowing soil dust particles (F1); local emission from chromium industry and wind blowing
47 mineral dust particles (F2); anthropogenic sources of traffic emission and wind blowing fine
48 mineral dust particles derived from geogenic factors (F3); natural transport from the marine
49 environment (F4); and long-range atmospheric deposition (F5). These approaches reflect
50 the important role of the bioindicators and moss biomonitoring that in combination with data
51 analysis and the inventory of emission sources are important tools to interpret the moss
52 concentration data and to assess air quality of the studied area.

53

54 **Key words:** atmospheric deposition; moss biomonitoring; trace elements; ICP-AES
55 analysis; factor analysis; GIS; Albania.



56

57 **Graphical Abstract**

58

59 **Highlights:**

- 60 - The origin of trace metal in moss samples is assessed through different methods.
61 - Mosses reflect spatial variability and temporal trend of trace elements air pollution.
62 - Mosses show similar contamination scale of elements on 2015 and 2010 surveys.
63 - Weather conditions affect the processes of accumulation of Trace metal in moss.
64 - FA is a useful tool identifying the origin sources of elements causing air pollution.

65

66 **Abbreviations:**

67 AAS – Atomic absorption spectroscopy

68 AMS – Albanian moss survey

69 BC - background concentration

70 CF – contamination factor

71 C_i – Concentration of element i

72 CVAAS - cold vapor atomic absorption spectrometry

73 ETAAS - electrothermal atomic absorption spectrometry

74 EWMA - Exponentially weighted moving average

75 FA – Factor analysis

76 FPM – Fine Particulate Matter

77 GIS - Geographic Information System

78 ICP-AES - inductively coupled plasma atomic emission spectrometry

79 L_i - individual loading of element i

80 LRTAP – Long-Range Transboundary Air Pollution

81 MB – Moss biomonitoring

82 NBC – national background concentration

83 TM – trace metal

84

85 **1. Introduction**

86 Air quality is a global problem affected by different contaminants present in the atmosphere,
87 such as particulate matter, toxic inorganic pollutants and heavy metals, reactive nitrogen,
88 and persistent organic pollutants. The atmosphere is a carrier of different organic and
89 inorganic chemicals transported to by natural and anthropogenic emission sources. The
90 largest anthropogenic sources of TM in the atmosphere are fine soil dust particles, dust

91 dispersion from metal processing, fossil fuel combustion, vehicle exhaust, emission from
92 cement factories and other human activities (Lazo et al., 2018; Harmens et al., 2010; Duffus,
93 2007). The main global source of atmospheric mineral dust is the Sahara desert, which
94 produces about half of the annual mineral dust (Karanasiou et al., 2012) and is directly
95 affecting the Mediterranean region (Flentje et al., 2015; Baqueros et al., 2013; Karanasiou
96 et al., 2012; Nastos, 2012; Theodosi et al., 2010; Tafuro et al., 2006).

97 Wet and dry deposition are the most important processes that remove these chemicals by
98 depositing those on terrestrial ecosystems (Amodio et al., 2014; Fang et al., 2014; Huang et
99 al., 2012). The deposition rate depends on several factors such as meteorology (wind
100 velocity, relative humidity, and temperature), particle size and shape, and the chemical
101 forms of the elements. Differences in the deposition fluxes of the pollutants in different
102 times or between different monitoring sites may be due to the differences in their
103 atmospheric concentrations and their scavenging ratios from the atmosphere to the
104 terrestrial ecosystems. On the other hand, the variability of ambient conditions, such as
105 precipitation, acidity, temperature, humidity and altitude, may affect to the state of elemental
106 occurrences as soluble or non soluble forms and to their bioavailability, that are important
107 during the accumulation process of inorganic elements in moss (Boutique et al., 2014;
108 Amodio et al., 2014; Steinnes, 1994). The cumulative long-term deposition of metals from
109 the atmosphere to the ecosystem is important as they may have adverse impacts on the
110 environment and human health, directly through the inhalation and through the food chain
111 (Cao et al., 2016; Zeng et al., 2016; Dockery, 2009). High content of heavy metals in the air
112 that we breath may cause different acute and chronic symptoms on human health (Chen
113 and Lippman, 2009; Kampa and Castanas, 2008; Schaumann et al., 2004).

114 Different monitoring models were applied to understand and to monitory the air pollution
115 (EUR/03/5042688, 2003; Amodio et al., 2014), as well as to investigate the effects of trace
116 elements airborne pollution (Gao et al., 2005). Besides the technical deposition samplings,
117 some alternative methods are developed to evaluate the degree and extent of airborne
118 contamination by investigating the level of contaminants in plants that are sensitive and
119 good accumulators to various pollutants. Naturally growing bryophyte mosses are good
120 bioindicators for assessing air quality impacted by atmospheric deposition of TM, organic
121 pollutants and nitrogen (Harmens et al., 2017, 2010). Carpet-forming bryophyte mosses are
122 rootless plants that obtain most of the nutrients directly from wet and dry deposition, and
123 the uptake from the substrate soil is not significant (Harmens et al., 2015, 2013, 2010; Berg
124 et al., 1996; Rühling and Tyler, 1968). They have a high capacity to retain TM and are an

125 ideal sampling medium for TM deposition from the atmosphere to vegetation. Moss
126 biomonitoring (MB) provides a time-integrated measurement of TM deposition (Harmens et
127 al., 2013; Schröder et al., 2013; Rühling and Steinnes, 1998) that may cover in a broad
128 range of spatial scales of the same method at a high spatial density after selecting a proper
129 moss species as biomonitor (Schröder et al., 2016; Harmens et al., 2015, 2010; Qarri et al.,
130 2013; Vuković et al., 2013; Steinnes et al., 2011, 1997a).

131 Since 2010, Albania had contributed to the data of the European moss survey (EMS), which
132 is repeated at five-yearly intervals. Albania is exposed to high levels of TM particularly for
133 elements linked with geogenic factors, mining operations, and mineral mine wastes. The
134 country has been appointed as a “hotspot” of heavy metal contamination in Europe (Lazo et
135 al., 2018; Harmens et al., 2015; UNDP-Albania, 2010). The data collected from 2015 AMS
136 are presented in this study. The aim of this study is the assessment of the air quality in
137 Albania, and the objectives that may lead to are: 1) the evaluation of TM atmospheric
138 deposition in Albania via MB; 2) the investigation of the spatial distribution and temporal
139 trend of TM concentrations in moss since 2010, 3) the identification of the probable factors
140 that may affect to TM concentration levels in current moss samples.

141

142 **2. Material and method**

143 *2.1. Description of the study area*

144 Albania is a Mediterranean country, positioned in south-west of the Balkan Peninsula. It is
145 characterized by warm and dry summer, and mild and wet winter. Due to the high diversity
146 of the topology of the country the climate varies from one region to another. The western
147 part of the country is affected by a Mediterranean climate, and the eastern part is mainly
148 affected by continental climate that is characterized by mild summer and cold winter.
149 Average annual precipitation varies from more than 2,500 mm in the North to less than 760
150 mm along the most part of the eastern border. The southwestern part of the country suffers
151 from summer droughts. Albania is highly exposed to several natural hazards like drought,
152 heavy rainfall, wind storms, heat waves, landslides, forest fires, airborne sand intrusion
153 from North African deserts that are directly or indirectly related to hydrology, meteorology
154 and weather conditions of the country (Hoxha et al., 2014). The 2000s have been
155 characterized by rising temperature and prolonged dry periods in Southern Europe and the
156 Mediterranean area (Hoerling et al., 2012). The low precipitation in the Mediterranean basin
157 favored the long time of PM in the atmosphere that showed an impact on air quality

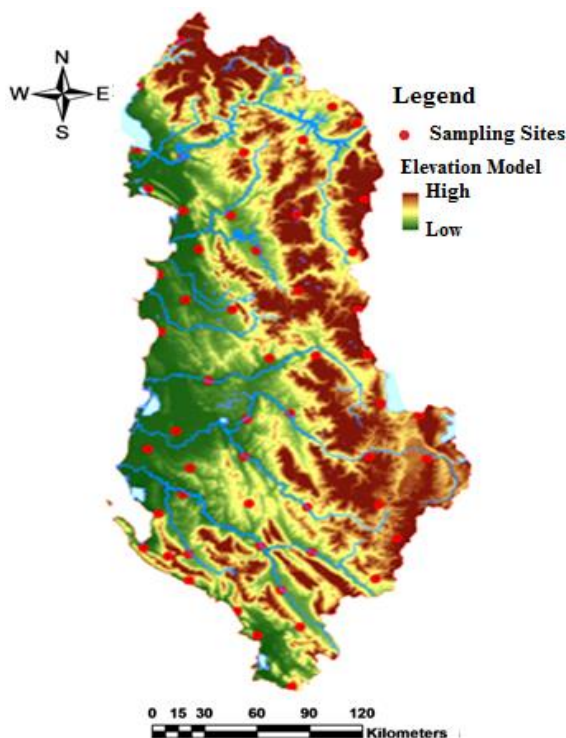
158 (Karanasiou et al., 2012). The longest drought events in Southern Europe (in particular in
159 the Mediterranean area) have been reported in the period between 1991 and 2010 (Spinoni
160 et al., 2017, 2015) that is also reflected in the climate of Albania.

161

162 2.2. Sampling procedure

163 Sampling and sample preparation was done by following the recommendations of the
164 European moss network (Frontasyeva and Harmens, 2015). Carpet forming moss species,
165 *Hypnum cupressiforme* (Hedw.) is selected for trace metal biomonitoring because it grows
166 almost in the natural environment. In addition, it has a wide range distribution in Albania
167 (Marka and Sabovljevic, 2011) and in other European areas (Sardans et al., 2005). It is
168 used in our previous biomonitoring study (Lazo et al., 2018; Allajbeu et al., 2017; Bekteshi
169 et al., 2015; Qarri et al., 2013) and in European trace element biomonitoring (Harmens et
170 al., 2015, 2010, 2008). Thus, it is useful for comparing the data with the other areas of
171 Europe (Sardans et al., 2005). The same scientists from the University of Tirana are
172 involved in field samplings of 2010 and 2015 AMS. They are carefully trained and instructed
173 by Dr. Jani Marka, a scientist in the field of bryophytes at the University of Tirana, Albania.
174 On the other hand, *Hypnum cupressiforme* (Hedw.) is easily identified in the field by non
175 bryophyte specialists. Mosses were collected during August and September 2015 at 55
176 sampling sites homogeneously distributed over the entire territory (Fig. 1). Geographic
177 coordinates of sampling sites are shown in Table 1S (S, as supplement material).

178 Composite samples of ten sub-samples collected within an area of 50 m x 50 m were used
179 as a representative site sample. A one liter glass container was used for approximately
180 measuring the volume of the moss samples after manually pressed in the container.
181 Samples were stored and transported to the laboratory in tightly closed paper bags. Coarse
182 contaminants were carefully removed and the moss samples were dried at room
183 temperature until a constant weight. Unwashed green and green-brown parts of the moss
184 stems that represent 3 years of the moss growth, just between two monitoring periods,
185 were selected for analysis (Frontasyeva and Harmens, 2015) because it may reduce the
186 effect of tissue ageing on cation uptake/retention capacity. Thus, it may prevent the
187 chronological relationship between the concentrations of TM in moss tissues and
188 atmospheric deposition (Boquete et al., 2014). The selected dry moss samples were
189 manually gridded to small parts before analysis. Disposable polyethylene gloves were used
190 to avoid the potential contamination of samples during sampling and sample preparation.



191

192 **Fig. 1.** GIS map for the localization of sampling sites (latitude, 41°00'N of the equator;
 193 longitude, 20°00'E of Greenwich)

194

195 **2.3. Chemical analysis**

196 Microwave digestion (MARS, CEM, USA) was used for total digestion of moss samples.
 197 The method was presented in previous publications by Stafilov et al., (2017), Qarri et al.,
 198 (2013) and Balabanova et al. (2008). The content of Al, Ba, Ca, Co, Cr, Cu, Fe, K, Li, Mg,
 199 Mn, Na, Ni, Pb, Sr and V in moss samples was determined by inductively coupled plasma–
 200 atomic emission spectrometry (ICP-AES) (Varian, 715ES), As and Cd by electrothermal
 201 atomic absorption spectrometry (ETAAS) (Varian, SpectrAA 640Z), performed at the
 202 Institute of Chemistry, Faculty of Science, Sts. Cyril and Methodius University, Skopje,
 203 Macedonia. The optimal conditions for each analytical technique are given by Stafilov et al.,
 204 (2017). Hg was determined by cold vapor atomic absorption spectrometry (CVAAS) (Varian
 205 10+) and homemade cold vapor system (Lazo and Cullaj, 2002) at the Department of
 206 Chemistry, Faculty of Natural Sciences, University of Tirana. Three replicates per moss
 207 sample were digested and three measurements per digest were done during the analysis.
 208 The detection limits of the elements were calculated as 3SD of the lowest instrumental
 209 measurements of the blanks, and were reported by Stafilov et al., (2017) on previous
 210 publications.

211

212 2.4. Quality control

213 The quality control of the results obtained by ICP-AES and AAS analysis was checked by
214 multiple analyses of the examined samples and simultaneous analysis of M2 and M3 moss
215 reference materials (Stafilov et al., 2017; Steinnes et al., 1997b). M2 and M3 were prepared
216 for the EMS (Steinnes et al., 1997b; Harmens et al., 2010) in the frame of the International
217 Cooperative Programme on Effects of Air Pollution on the Natural Vegetation and Crops
218 (<https://icpvegetation.ceh.ac.uk>). The recommended concentration values of elements were
219 issued based on an inter-comparison exercise with the participation of about 30 laboratories
220 from a number of European countries employing different analytical techniques (Steinnes et
221 al., 1997b). M2 and M3 reference moss samples were used for analytical quality control
222 among all participants in the joint European MS (Harmens et al., 2015, 2010, 2008). The
223 obtained values of the analyzed elements were within recommended values (Steinnes et al.,
224 1997b; Harmens et al., 2010). The Table with reference data of M2 and M3 moss reference
225 materials is shown also in Qarri et al., (2013).

226 Quality control of CVAAS results was checked by multiple analyses of the examined
227 samples and by simultaneous analysis of the inter-country plant reference material IAEA-
228 140/TM (*Fucus homogenate*). The differences between the mean contents of Hg and the
229 certified values of reference material were within 15%. The method of standard addition
230 was also applied, and quantitative recoveries were achieved for most of the elements. The
231 recovery of the investigated elements ranged between 98.5 and 101.2 % for ICP-AES, 96.9
232 to 103.2 % for AAS, and 92.4 to 103.6 % for CVAAS analysis.

233

234 2.5. Data processing and statistical analyses

235 The contents of TM in moss samples were calculated on a dry matter basis. Descriptive
236 statistics was applied to the elemental concentration data set as the first step of statistical
237 analysis. The type of distribution of the concentration data and the possible outliers of each
238 element were examined through the frequency distribution confirmed by the statistical
239 significance levels at $P > 0.05$.

240 Time series analysis was applied to atmospheric pollution data (Qarri et al., 2014b; Salcedo
241 et al., 1999; Morawska et al., 1998) and exponentially weighted moving average (EWMA)
242 univariate charts were used to detect the trend of the variables (Haridy and Wu, 2009; Lam
243 et al., 1999), potential shifts in location and scale (Liu et al., 2013), and to investigate the
244 country background level (NBC) of each element (Qarri et al., 2014b). Most of the
245 guidelines for environmental legislation are based on background values and the toxicity

246 levels of the contaminants (Carlon, 2007). Statistical method was used to evaluate the NBC
247 level of TM in moss samples of this study. The median values and the 2 maximum absolute
248 standard deviation (MAD) (Matschullat et al., 2000; Reimann et al., 2005) are usually used
249 to identify the outlier values of each element. The values of [median+2 STDEV] are referred
250 as the upper concentration limit (UCL) of the geochemical background variation, and was
251 suggested as a “threshold level” for the cleanup goal of environmental legislation (Reimann
252 et al., 2005). The values of [median–2 STDEV] are referred as the lower concentration limit
253 (LCL) of geochemical background, or as the background level. UCL and LCL levels of the
254 elements that show high variation (CV % > 75 %) were expressed as [median ± STDEV].
255 NBC was calculated on the basis of the LCL level of each element re-plotted after excluding
256 the outlier points higher than the respective UCL levels of the sorted original data (Qarri et
257 al., 2014b). The average value of the outlier points lower than LCL level was used as NBC.
258 The contamination level was evaluated by calculating the contamination factors (CF) given
259 by Fernandez et al., (2000). The CF values for each element were calculated as the ratio of
260 the median concentration of the element (C_i) to its respective value of NBC_i. The most
261 probable emitting sources were discussed based on the CF level of each element.

262

$$263 \quad CF = C_i / NBC_i$$

264

265 The data obtained in 2010 and 2015 AMS are compared for statistically significant
266 differences by using Mann-Whitney U test that investigates the significance of the
267 differences between two different data sets. The significance of null hypothesis, for no
268 difference between two data sets, is tested by p-value higher than 0.05.

269 Spearman correlation analysis was carried out to measure the strength of the association
270 between two ranked monotonic variables that changes together in the same sense. Beside
271 it the Pearson correlation analysis was tested to distinguish the association between
272 elements with strong linear combinations. The significance of Pearson correlation was
273 examined by linear regression analysis, based on the slope of the regression line (s≠0) and
274 P<0.05 for R² (slope) ≠ 0. The formal hypothesis test for the relationship between variables
275 is confirmed by the overall F-test that determines whether this relationship is statistically
276 significant. If the P value of the obtained overall F-test is less than the significance level
277 (0.05), it can conclude that the relationship is true and R-squared value is significantly
278 different from zero.

279 Factor analysis (FA) was applied as an extension of the Pearson correlation analysis that
280 may explore hidden multivariate structures in the data (Astel et al., 2008; Reimann et al.,
281 2002) and may clarify the links between elements that tend to have similar origins or appear
282 similar associations on the data matrix. The most important factors extracted from FA are
283 discussed. The statistical analyses were conducted using the MINITAB 17 software
284 package.

285 Geographic Information System (GIS) maps are plotted to explain the spatial distribution of
286 selected elements and the factor loadings of each factor extracted from FA. Arc-GIS 10.2
287 was used in combination with local deterministic methods and the inverse distance
288 weighting for mapping the spatial distribution of the factor loadings (FL_{site}) and for spatial
289 interpolation. FL_{site} is calculated as a percentile of the sum of the product of individual loads
290 of each element and the respective site concentration (Lazo et al., 2018; Allajbeu et al.,
291 2017; Barandovski et al., 2008):

$$293 \quad FL_{site} = average(100 \frac{\sum(C_i)_{site}}{\sum(C_i)_{max}} L_i)$$

294
295 where, C_i is the site concentration of the i^{th} element and L_i is the factor loading of the same
296 element.

298 3. Results and discussion

299 3.1. Frequency distribution

300 The frequency distribution of the data was examined through the frequency plots of each
301 element. Current data represent different statistical distributions by indicating that the data
302 are affected by different factors. Most of the elements follow a lognormal distribution
303 ($P>0.05$) that is characteristic for the lithogenic origin of crustal elements (Vinogradov,
304 1962), so their presence in moss may be derived from atmospheric deposition of windblown
305 fine dust particles. The concentration data onto As, Cd, Pb, Zn and Na do not follow the
306 lognormal distribution ($P<0.05$). After excluding the outlier sites with high concentration
307 level of TM in sites (3 points for As, Pb, Na, and 5 for Zn), the remained data onto these
308 elements follow the lognormal distribution ($P>0.05$). The outlier sites of Cd are greater than
309 5. No rule was observed for Cd outlier points.

311 3.2. Elemental concentration and contamination scale

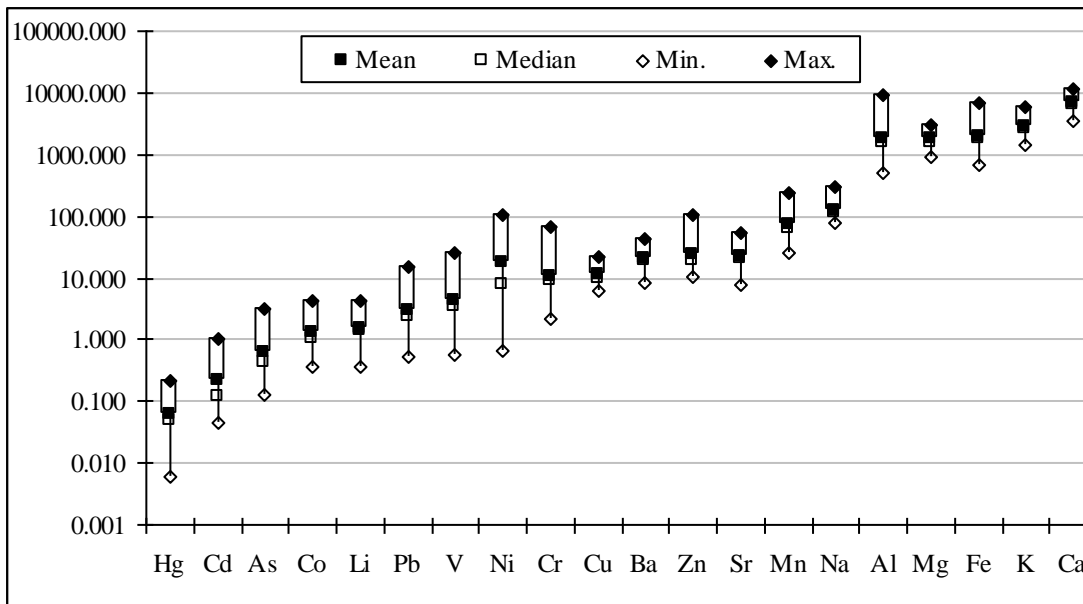
312 The observed concentrations could be used to determine the origin of the deposits (natural
 313 or anthropogenic), and the distribution of the TM (Meyer et al., 2015) in the study area, and
 314 to evaluate the trend of the distribution by comparing the data with previous moss
 315 biomonitoring. Descriptive statistic (Table 1) analysis and contamination scale were used to
 316 interpret the results.

317

318 **Table 1** Descriptive statistics of the concentration data (mg/kg, DW) in moss samples (n=55)

Elements	Mean	Median	St. Dev.	CV %	Kurtosis	Skewness	Min.	Max.
Al	1,745	1,521	1,267	73	19.9	3.8	486	9,022
As	0.621	0.418	0.565	91	8.6	2.7	0.13	3.18
Ba	19.7	18.2	7.58	38	0.8	1	8.39	43.3
Ca	6,667	6,509	1,910	29	-0.7	0.2	3,543	11,188
Cd	0.222	0.116	0.233	105	3.3	2	0.045	1.02
Cr	10.7	9.27	9.69	91	19.5	3.7	2.21	66.2
Co	1.32	1.05	0.85	65	3.3	1.8	0.35	4.24
Cu	10.8	10	3.69	34	1.2	1.2	6.1	22.4
Fe	1,854	1,735	1,007	54	11.6	2.7	685	6,956
Hg	0.059	0.049	0.04	67	4.1	1.8	0.013	0.213
K	2,824	2,577	1,001	35	1.5	1.3	1,435	5,824
Li	1.54	1.35	0.79	51	1.6	1	0.371	4.34
Mg	1,740	1,550	591	34	-0.9	0.5	910	2,970
Mn	72.9	63.2	42.4	58	4.4	1.8	24.5	244
Na	126	114	44.9	36	6.5	2.3	76.1	297
Ni	17.1	7.57	23.5	138	6.5	2.6	0.68	108
Pb	3.01	2.38	2.8	93	5.5	2.1	0.51	14.5
Sr	21.2	19.7	9.06	43	1.8	1.1	7.98	53.5
V	4.14	3.32	3.74	90	22.4	4.07	0.56	26.2
Zn	23.3	18.4	16.2	70	14.1	3.48	10.3	108

319



320
 321 **Fig. 2.** Box-plot diagram of concentration data (mg/kg, DW)

322
 323 The order of the median concentration of the elements in moss samples was Ca > K > Fe >
 324 Mg > Al > Na > Mn > Sr > Zn > Ba > Cu > Cr > Ni > V > Pb > Li > Co > As > Cd > Hg. The
 325 concentration data onto As, Cd, Cr, Ni, Pb and V show high disparity (CV% > 75 %)
 326 followed by high values of kurtosis, and positive skewness that may indicate the data was
 327 affected by different factors (Wang et al., 2010). The concentration data of other elements
 328 were more stable and show moderate CVs (%) (25 % < CV % < 75 %). For a better
 329 interpretation of the results, the contamination factors (CF) were calculated (Table 2).

330
 331 **Table 2** The contamination factors (CFs) and contamination classification (Fernandez et al.,
 332 2000) for metal concentrations in current mosses

Elements	As	Cd	Cr	Cu	Fe	Hg	Ni	Pb	V	Zn
CF (2015, n=55)	3.2 ³	2.6 ³	4.2 ⁴	1.6 ²	2.5 ³	3.8 ³	11.1 ⁵	4.7 ⁴	5.9 ⁴	1.8 ²
CF (2010, n=62)	3.3 ³	1.9 ²	8.8 ⁵	1.6 ²	7.8 ⁵	-	5.1 ⁴	2.1 ³	3.8 ³	0.5 ¹
Elements	Al	Sr	Mn	Co	Ca	K	Mg	Na	Ba	Li
CF (2015, n=55)	3.1 ³	2.5 ³	2.6 ³	3.0 ³	1.8 ²	1.8 ²	1.7 ²	1.5 ²	2.2 ²	3.7 ³
CF (2010, n=62)	8.3 ⁵	1.4 ²	2.2 ²	-	2.4 ³	1.9 ²	1.4 ²	1.3 ²	1.3 ²	2.1 ³

333 ¹ No contamination; ² Suspected contamination; ³ Slight contamination; ⁴ Moderate
 334 contamination; ⁵ Serious contamination

335
 336 The CF of Ni is associated with the 5th category of the contamination scale, C5 (serious
 337 contamination). The CFs of Cr, Pb and V are associated with the 4th category of the

338 contamination scale, C4 (moderate contamination), all are probably linked with long-range
 339 atmospheric transport of pollutants from other parts of Europe (Harmens et al., 2015). In
 340 addition, it is most likely derived from local factors such as fuel combustion, wind blowing
 341 fine mineral dust particles originating from industrial waste deposits, mine and metal
 342 processing industry, and geogenic factors (Harmens et al., 2010; Lazo et al., 2018). The
 343 CFs of As, Cd, Co, Fe, Hg, Al, Li, Sr and Mn are associated with the 3rd category of the
 344 contamination scale, C3 (slight contamination). The CFs of Cu, Zn, Ca, Mg, Ba, Na and K
 345 are associated with the second category of the contamination scale, C2 (suspended
 346 contamination), that is easily obtained from natural variation (Fenandez et al., 2000).
 347 Similar results were obtained for CF values of the elements As, Cu, Mg, Ba, Na, K and Li
 348 for current data and AMS 2010 data (Qarri et al., 2013). The differences observed in CFs of
 349 Cd, Cr, Ni, Fe, Mn, Pb, V, Al, Sr, Mn, Ca and Zn are mostly affected by different
 350 background levels (BC_i) used for 2010 AMS (data from a pristine area in Norway, Steinnes
 351 et al., 2007) and 2015 (country background level, NBC), and by the differences between the
 352 respective median concentration values (C_i) of the elements on 2010 and 2015 AMS (Table
 353 2S).
 354 The median concentrations of 2015 AMS are compared with similar national studies done in
 355 Macedonia (a neighboring country) (Stafilov et al., 2017) and Norway, selected as a clean
 356 area in Europe (Steinnes et al., 2016) (Table 3).

357

358 **Table 3** The comparison of 2015 AMS median concentration with respective data of
 359 Macedonia and Norway

Elements	Albania ¹		Macedonia ²		Norway ³
	Median	Range	Median	Range	Median
Al	1,521	486 - 9,022	2,100	750 - 7,400	461
As	0.42	0.13 - 3.2	0.54	0.13 - 1.4	0.13
Ba	18.2	8.4 - 43	42.0	9.7 - 180	25
Ca	6,509	3,543 - 11,188	6,900	3,500 - 13,000	3,030
Cd	0.12	0.045 - 1.0	0.23	0.018 - 0.88	0.08
Co	1.1	0.35 - 4.2	0.60	0.16 - 2.0	0.20
Cr	9.3	2.21- 66	5.7	1.8 - 31	0.66
Cu	10.0	6.1 - 22	4.6	3.0 - 8.3	4.2
Fe	1,735	685 - 6,956	1,700	510 - 4,600	310
Hg	0.049	0.013 - 0.21	0.084	0.02 - 0.25	0.06

K	2,577	1,435 - 5,824	6,000	3,100 - 14,000	3,560
Li	1.35	0.37 - 4.3	0.79	0.32 - 3.51	0.16
Mg	1,550	910 - 2,970	1,900	1,200 - 3,800	1,350
Mn	63.2	25 - 244	160	33 - 510	400
Na	114	76 - 297	190	140 - 380	208
Ni	7.6	0.68 - 108	3.5	0.68 - 63	1.2
Pb	2.38	0.51 - 14.5	4.9	2.2 - 14	1.6
Sr	19.7	8.0 - 54	25	6.5 - 220	13
V	3.3	0.56 - 26	3.3	0.47-11	1.2
Zn	18	10 - 108	30	12.1 - 66	31

360 ¹ current work, ² Stafilov et al.015, ³ Steinnes et al. 2016

361

362 Some differences were observed onto median values of the elements among different
363 countries. Moss tissue appears to be affected by the equilibrium established between the
364 moss and the environment that may increase/or decrease the concentrations of elements in
365 moss samples caused by chemical, biological and environmental factors (Boquete et al.,
366 2014). The observed differences onto median values of the elements among different
367 countries are probably affected by different local natural and anthropogenic emission
368 sources (Lazo et al., 2018; Stafilov et al., 2017; Steinnes et al., 2016; Harmens et al., 2013)
369 and local geochemistry of each country that strongly affect the chemical composition of soil
370 dust. Our attention was focused on the differences observed for Zn and Hg between Balkan
371 countries (Albania and Macedonia) and Norway. The same differences were found also in
372 2010 EMS for Zn. The lowest Zn median values of 2010 EMS were reported for Albania,
373 Faroe Islands, Macedonia, Iceland and Bulgaria (Harmens et al., 2015), while the highest
374 levels of Hg on 2010 EMS were found in Albania and Macedonia, followed by Italy (Bolzano
375 region), Poland and France. Relatively high levels of Hg were also reported for Norway and
376 the Faroe Islands, and it has been increased since 2005 in some parts of Norway (Harmens
377 et al., 2015). To investigate the spatial distribution of elements present in moss samples,
378 the GIS maps are plotted for some elements of different origin (Fig. 3).

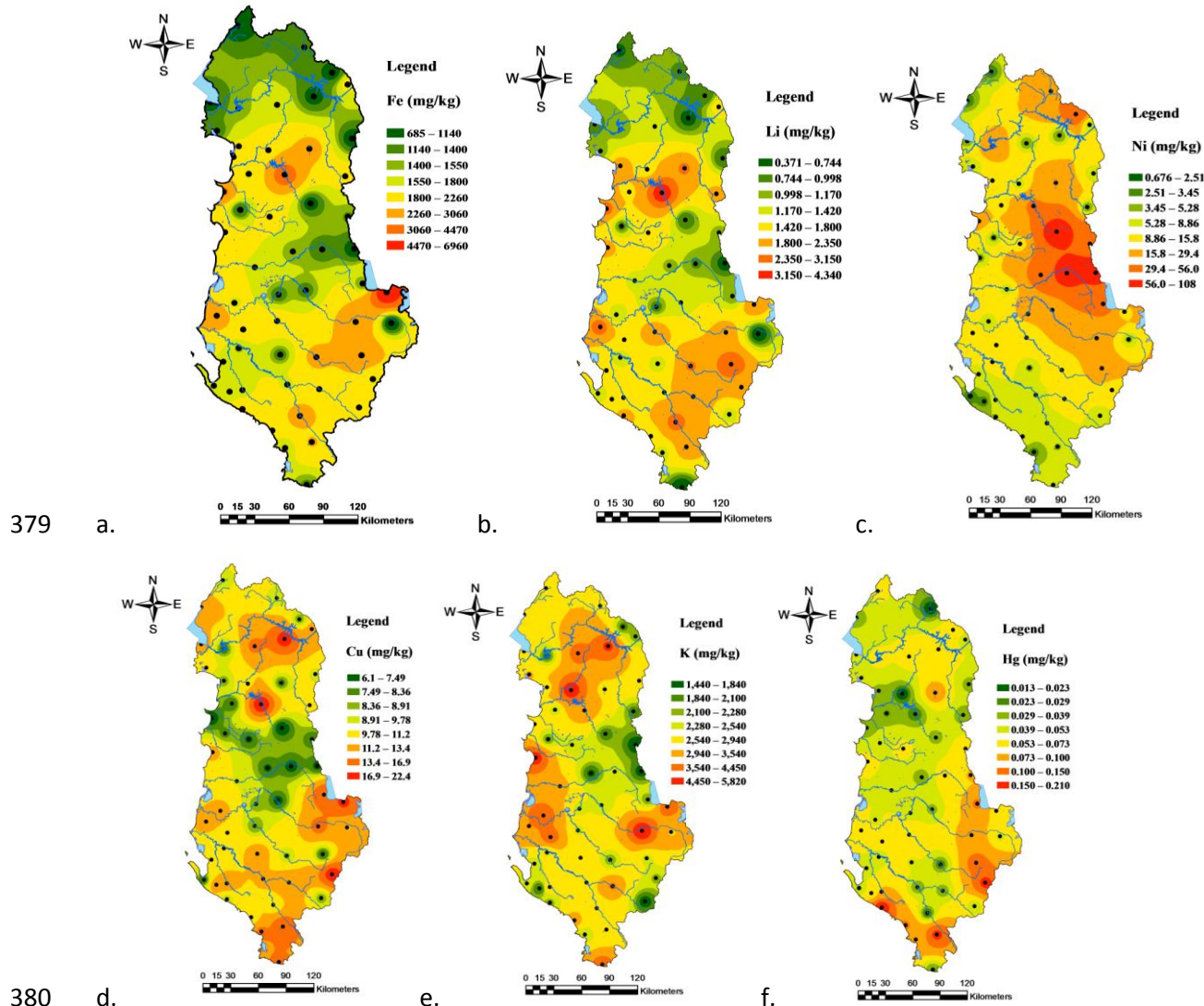


Fig. 3. GIS maps of TM atmospheric deposition in Albania. a. Fe, b. Li, c. Ni, d. Cu, e. K, f. Hg

The distribution pattern of the elements showed the presence of local emitting sources that are distinguished from the relatively high content of moss elements. The elements that are linked with geogenic factors (Ni and Cu) and metal processing industry (Ni) show an enrichment in the eastern part of the country that is characterized by high primarily metallogenic mineral deposits, particularly of Ni-Fe, Cr and Cu minerals (Lazo et al., 2018). High content of potassium was found in the Western coastal area and in the N-E part of the country. K^+ and Mg^{2+} were found in the sea-spray, Saharian dust, in regional mineral dust, smoke (K^+) and in plant debris (Flentje et al., 2015), and thus the comment on spatial distribution of K is relatively difficult. The western coastal area is a low land. It is the main agriculture and industrial area of Albania, and high potassium content in mosses collected

394 in this area was probably affected by different factors such as biomass burning, sea salt
395 and industrial emission (Qiu et al., 2014; Zhang et al., 2014; Jorge et al., 2014; Quennehen
396 et al., 2012; Sardans et al., 2005) and the use of K fertilizers in agricultural activities
397 (Sardans et al., 2005). It is estimated that the erosion washes away 16,000 tons per year of
398 potassium salts in Albania (Laze and Kovaçi, 1996). The anomalies of K content on the N-E
399 are probably derived from biomass burning, K fertilizer used in vinery yards, soil dust
400 emission and geogenic factors, such as (Na, K)-feldspars of 8-9% alkali content and 4-7%
401 K₂O of Korabi, Gashi, and Mirdita zones (http://albanian_mining_today). However, the
402 relation of the K fluxes in the atmosphere and the role of the soils and anthropogenic
403 activities is not clear till now (Quennehen et al., 2012).

404 The distribution pattern of Hg is different from the other elements. It is more or less
405 homogeneously distributed over the country, with the highest Hg concentration in the S-E
406 direction, and in the cross-border between Albania, Greece and Macedonia. As was shown
407 from the EMEP data (EMEP Report, 2015) that were collected from the measurements of
408 EMEP monitoring network and were evaluated on the basis of the model calculations
409 complemented by joint analysis of measurement data (Colette, et al., 2016), high Hg
410 concentration in S-E direction is probably affected by trans-boundary pollution from
411 neighboring countries. The highest Hg concentration level on the 2015 AMS shows a
412 similar geographic position as was observed by EMEP (EMEP Report, 2015). It confirms
413 that the ecosystems of Albania are significantly affected by Hg trans-boundary pollution and
414 only a small part come from long-range atmospheric transport.

415

416 3.3. Temporal trends of TM in mosses

417 The data onto 2015 and 2010 AMS (Qarri et al., 2013) are compared for providing further
418 information on temporal trends of TM concentrations on the mosses of Albania. By
419 comparing the medians of the 2015 and 2010 AMS data, the metal concentrations on
420 mosses can be separated into two different categories.

421 *The first category* is composed by the elements that show high decline, such as Hg (165 %),
422 Mg (51.4 %) and K (32.5 %), Ba (16.5 %) and Sr (10 %). The elements Li, Ca, Al, V and Pb
423 show small declines, respectively 5.3 % (Li), 3.5 % (Ca), 7.7 % (Al), 5.7 % (V) and 1.4 %
424 (Pb). The declines lower than 10 % are probably derived from significant differences of
425 analytical and method variation (RSD % < 10 %) during the determination of the elements,
426 sample preparation and sampling procedure.

427 *The second category* is composed by the elements that show a high increase onto the
428 concentrations data of 2015, such as Cr (95.2 %), Cu (79.2 %), As (37.1 %), Zn (33.7 %),
429 Ni (29.3 %) and Na (30.6 %). The elements Mn (12.2 %), Cd (8.1 %) and Fe (7.2 %) show a
430 small increments in 2015 compared to the 2010 AMS.

431 The concentration data of both MS showed the same statistical distribution shape (mostly
432 log-normal distribution) that made it possible to apply the Man-Whitney U Test for
433 statistically testing the significant differences ($p > 0.05$) between the same elements. The
434 concentration data of Cd, Fe, Pb, V, Al, Ba, Li, Mn, Ca and Sr did not show statistically
435 significant differences ($p > 0.05$) between the 2015 and 2010 AMS. Statistically significant
436 differences ($p < 0.05$) were found between 2015 and 2010 AMS data of As, Cr, Cu, Hg, Mg,
437 Ni, Zn, K and Na. The results of Man-Whitney U Test ($p = 0.05$) are shown in Table 3S.
438 There are different factors affecting the uptake of inorganic elements in moss tissue that
439 may cause significant differences in TM contents in moss samples. TM may be absorbed
440 on the moss from the atmosphere as soluble chemical species during wet deposition or
441 from fine particles during dry deposition (Amodio et al., 2014). Several factors, such as
442 physic-chemical characteristics of the elements, the uptake mechanisms and bioavailability
443 of the elements during deposition (Varela et al., 2015), the competition effect of cations, the
444 wind blowing mineral dust from local soil, the transport of soluble compounds from the soil
445 to the moss tissue, particularly during periods with excessive soil/water contact, the
446 differences in growth rate of the moss within the region, variations in precipitation chemistry
447 and different natural and anthropogenic emission levels, may affect the uptake of the TM in
448 moss (Steinnes, 1995). These factors are mostly linked with the meteorology, which affect
449 to the dispersion and the chemistry of the atmospheric pollutants (Wu et al., 2018; Thunis et
450 al., 2016; Miranda et al., 2015; Amodio et al., 2014). Moreover, the rain scavenging and
451 gravity sedimentation during wet and dry deposition, are the main transport mechanisms of
452 TM from the air to the other terrestrial ecosystems (Miranda et al., 2015).

453 The dry period of 1991 and 2010 was characterized as the most severe drought events in
454 the Mediterranean area (Spinoni et al., 2017, 2015). Some separated intense rainfalls
455 events happened in December 2009 to January 2010 at the North of Albania (UNDP CO-
456 ALBANIA, 2010), while 2015 showed high rainfall in January to March and September to
457 November 2015, moderate rainfall on April to July, and very small raining on August
458 (<https://...../albania/precipitation>). Faced with different raining conditions, but no changes in
459 emission inventory and emission sources during both sampling and moss growing periods,
460 we suggest that meteorological conditions and seasonal variability were the most important

461 factor which probably affected to the significant differences on the concentration level of
462 some TMs in moss samples of Albania. The decrease of Mg, K, Ba and Sr in 2015 mosses
463 may be linked with the leaching process of these elements from moss samples that may
464 cause a decline in their concentrations during wet weather. Similar findings are reported
465 from other authors for different study areas (Amodio et al., 2014; Baqueros et al., 2013;
466 Connan et al., 2013; Steines et al., 2003). Beside it, drastically decline was observed on
467 2015 on Hg concentration compared to 2010. Based on the EMEP gridded emission data
468 (EMEP Report, 2015) that provide information on the levels of emissions of heavy metals
469 (Cd, Pb and Hg) it was found that the trend in Hg deposition for 2015 showed a decline
470 compared to 2010 (EMEP Report, 2015). These results are in the same line with Hg 2015
471 and 2010 AMS data. The decline observed in the Hg content of moss samples was
472 probably linked with the retention process of Hg^{2+} during wet deposition of 2015 and a
473 considerable supply of Hg^0 to the moss of 2010 that may occur during the high temperature
474 and dry deposition (Wu et al., 2018; Steinnes et al., 2003; Baquero et al., 2013). A mercury
475 “hotspot” with a huge content of metallic Hg in soils that was caused by the destroyed
476 Chlor-alkali Plant in 1997, posed a serious Hg^0 supply in the atmospheric air of the area
477 (Lazo and Reif, 2013). The 2010 EMS, ranked Albania among the countries with the
478 highest Hg level in Europe followed by Macedonia, Italy (Bolzano region), Poland and
479 France (Harmens et al., 2015). The Hg content in the soil of Vlora area was drastically
480 declined after the remediation of the polluted area on the end of 2009 that corresponds
481 before the period of 2015 moss growth. The Hg median concentration of 2015 AMS was
482 similar with Croatia (0.043 mg/kg, DW) (Špirić et al., 2013), and lower than Macedonia
483 (0.084 mg/kg, DW) (Stafilov et al., 2017).

484 High and significant differences have been found onto the concentrations data of 2015 and
485 2010 AMS for Cr, Cu, As, Zn, Ni and Na may be derived by the same climatic factor, that
486 may affect to the concentrations of these elements in atmospheric deposition (Wu et al.,
487 2018; Amodio et al., 2014) and is reflected also in TM concentration in moss samples
488 (Amodio et al., 2014; Baqueros et al., 2013; Steines et al., 2003). Wet deposition is
489 associated with the scavenging ability of the rain in removing pollutants from the
490 atmosphere (Amodio et al., 2014; Fang et al., 2014) particularly in high contaminated areas,
491 such as the N-E part of Albania that is characterized by high primarily metallogenic mineral
492 deposits, mining and metallurgical industry, and mineral mine wastes of ex-mining industry
493 (Lazo et al., 2018). Na shows the same behavior as anthropogenic elements (Ni, Cr, Cu)

494 during wet deposition. It is probably derived from relatively high wet deposition flux of Na
495 and by high solubility of Na in coastal areas (Miranda et al., 2015; Baqueros et al., 2013).
496 Wet deposition is often considered as an important natural factor in cleaning the
497 atmosphere and removing of pollutants from the atmosphere (Yang et al., 2012). The wet
498 deposition rate for single events is several orders of magnitude greater than dry deposition
499 rates measured for periods between precipitation events (Lindberg and Harriss, 1981), but
500 dry deposition is more important than wet deposition throughout the study period
501 (Muezzinoglu and Cizmecioglu, 2006). From this point of view, to avoid the effect of the
502 meteorological variations on deposition rates of pollutants during wet and dry deposition,
503 perhaps more precise criteria should be recommended on the atmospheric conditions
504 under which the sampling process should be carried out.

505

506 *3.4 Multivariate analysis*

507 Spearman and Pearson correlation (Table 4S) analysis was carried out to measure the
508 strength of the association between two ranked monotonic variables, and to distinguish the
509 association between elements with strong linear combinations. Similar results were
510 obtained by Spearman and Pearson correlations mostly for the elements with strong and
511 significant correlation coefficients. Some differences were found only for low correlation
512 coefficients ($r^2 < 0.4$, $P < 0.001$). On the other hand, the Pearson correlation is examined by
513 linear regression analysis, mostly for the paired elements with high and significant
514 correlations ($r^2 > 0.4$, $P < 0.001$). Based on the values of the slope of the regression lines and
515 the analysis of the variances (F-test, $P < 0.05$) it was found that the correlations with $r^2 > 0.4$,
516 $P < 0.001$ are true and can be used for further statistical analysis. The obtained results of
517 the overall F-test and the respective P values are shown in Table 5S and Fig. 1S. Factor
518 analysis (FA) of the correlated data matrix was carried out, and the main factors were
519 extracted and interpreted as source categories contributing to element concentrations on
520 moss samples. The main criteria in selecting the number of optimal factors is that of
521 Eigenvalues larger than 1. The identification of source categories was done by the
522 examination on the profiles of the factors and factor loadings of the elements, after varimax
523 rotation. The factors were interpreted based on the values of factor loadings of the
524 estimated parameters and the associations of the elements with high loadings in the same
525 factor (Reimann et al., 2002). Factors are composed by the loadings that are affected by
526 different reasons, such as chemical properties of the elements, their geochemical
527 associations in soil and dust, local and long-range transport of the elements, the inventory

528 of the local emission sources of the elements in the study area and the previous knowledge
 529 of the atmospheric concentrations (Miranda et al., 2015). Emission inventory of air
 530 pollutants can supply the essential information to understand regional and local emission
 531 sources (Qiu et al., 2014). These are important reasons for linking the TM with their sources
 532 of origin or factors affecting their presence in the study area. However, most of the results
 533 are governed by different sources and could be predicted by using the pre-existing
 534 information (Reiman et al., 2002). Factor loadings (FL) larger than 0.4 were used for
 535 interpreting each factor (Table 4). The FL lower than 0.4 represents low correlation between
 536 elements and were not under consideration (Pauperio et al., 2014; Costello and Osborne,
 537 2005).

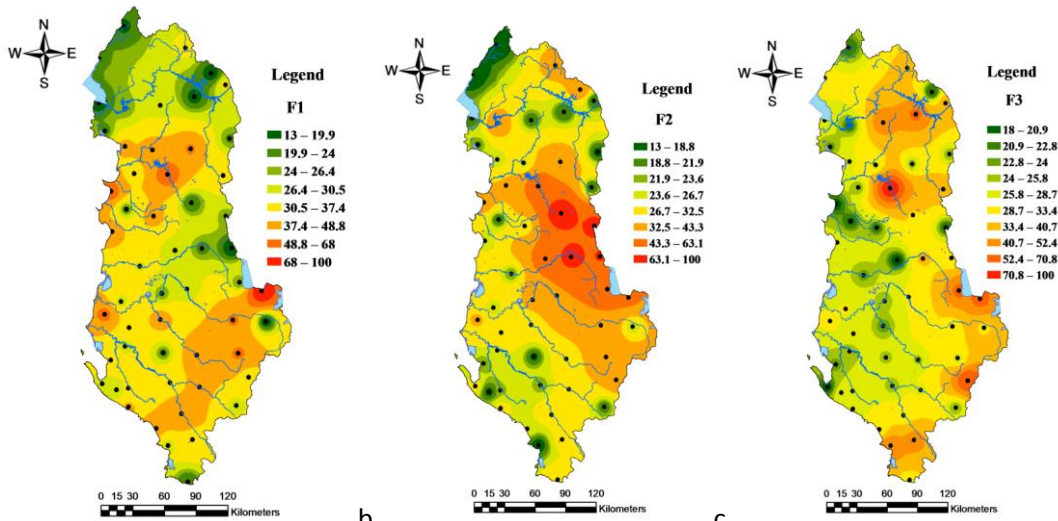
538

539 **Table 4** The results of the FA analysis of the correlation matrix (Sorted Varimax Rotated
 540 Factor Loadings for values > 0.4)

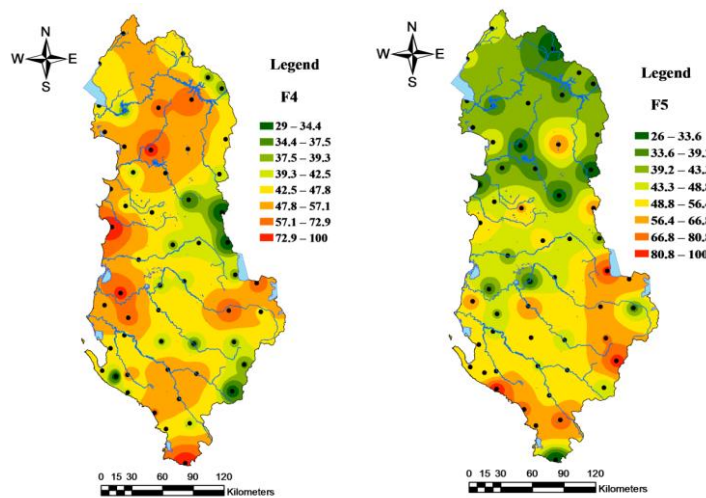
Variable	Factor1	Factor2	Factor3	Factor4	Factor5	Communality
Fe	0.938					0.944
Al	0.913					0.899
V	0.910					0.889
Ba	0.853					0.773
Li	0.818					0.712
Sr	0.758					0.713
Mn	0.582					0.558
Ni		0.905				0.896
Co		0.870				0.800
Cr		0.788				0.697
Mg		0.670				0.617
Pb		0.491	0.466			0.561
Cu			0.846			0.881
Zn			0.737			0.780
Cd			0.703			0.616
As			0.633			0.585
K				0.843		0.833
Na				0.818		0.702
Hg					0.714	0.585

Ca					0.665	0.731
Variance	5.516	3.248	2.768	1.824	1.415	14.77
% Var	0.276	0.162	0.138	0.091	0.071	0.739

541



542



543

544 **Fig. 4.** GIS maps of factor loadings for each factor extracted from FA. a. F1, b. F2, c. F3, d.
545 F4, e. F5

546

547 Five main factors that represent 73.9 % of the total variance were identified. The
548 associations of the elements on the same factor are analyzed as follows:

549 *Factor 1* (F1) is the strongest factor that presents 27.6 % of the total variance. It is
550 characterized by high loadings of Fe, Al, V, Li, Ba and Sr (FL > 0.76) and lower loading of
551 Mn (FL = 0.58). These elements are naturally distributed as typical soil elements (Rudnick
552 and Gao, 2004) and crustal materials by indicating their origin from soil dust particles. The

553 presence of Al in this factor confirms this assumption, since Al compounds are insoluble
554 and most of the Al found in biological systems comes from soil and dust contamination
555 (Qarri et al., 2013). Similar associations of the elements are reported on 2010 AMS and in
556 other regions of Europe (Harmens et al., 2015; Steinnes and Lierhagen, 2017) and Balkan
557 countries (Stafilov et al., 2017; Barandovski et al., 2015; Špirić et al., 2013).

558 *Factor 2* (F2) is the second strongest factor, with 16.2 % of the total variance. It is
559 associated by high loadings of Ni, Co, Cr and Mg (FL > 0.67) and lower loading of Pb (FL =
560 0.49). Ni, Co, Cr and Mg are typical elements present in chromites and nickel ores, and are
561 probably derived from mining industry, ferrochromium and ferronickel metallurgy of Elbasan
562 (Lazo et al., 2013), from the geogenic origin and the wind blowing mineral dust particles of
563 mine wastes (Lazo et al., 2018). The presence of Pb in this factor is probably derived from
564 long-range transport (Harmens et al., 2015), traffic emission and the emission from
565 metallurgy, as another probable factor (Harmens et al., 2016, 2015, 2014, 2012, 2010).

566 *Factor 3* (F3) represent 13.8 % of the total variance. It is characterized by high loads of Cu,
567 Cd, As and Zn (FL > 0.63) and lower loads of Pb (FL = 0.47). These elements are
568 distinguished as typical elements derived from anthropogenic sources that are mainly linked
569 with traffic emissions, geogenic origin, and wind blowing mineral dust particles of sulfide
570 mineral mine wastes (Lazo et al., 2018).

571 *Factor 4* (F4) is a weak factor that represents only 9.1 % of the total variance. It is
572 associated with high loadings of K and Na (FL > 0.82). Those are typical sea salt elements
573 that are probably originated from the Adriatic and Ionian coastal area positioned along the
574 western part of Albania. On the other hand, a wide potassium anomaly (see Fig. 3e) is
575 present in N-E part of Albania. Higher K content in this area is probably linked with
576 geochemical settings, mostly from serpentine soils in the East, where soils were derived
577 from K feldspars, gabbros and ultrabasic rocks, the last rich also in K (Shallari et al., 1998).

578 *Factor 5* (F5) is associated with high loads of Hg and Ca (FL > 0.66). It is probably
579 associated with long-range atmospheric transport of Hg, a global pollutant (Harmens et al.,
580 2015), entrapped to wind blowing soil dust fine particles that should be identified from the
581 presence of Ca (Wu et al., 2018) which is proposed as Saharan dust tracer (Flentje et al.,
582 2015). The Mediterranean basin is affected by Saharan dust intrusions during dry weeks
583 when the deposition rate of fine particulate matter is relatively high which may increase the
584 content of FPM in the atmosphere (Flentje et al., 2015; Baqueros et al., 2013; Karanasiou
585 et al., 2012; Nastos, 2012; Theodosi et al., 2010; Tafuro et al., 2006). Similar results were
586 extracted from FA of 2015 and 2010 AMS (Qarri et al., 2013).

587

588

4. Conclusions

589

The following conclusions can be drawn from this study:

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- MB in combination with statistical analysis of the moss data and GIS technology provides a suitable complementary method for spatial investigation of atmospheric deposition flux that may identify the areas at risk caused by high atmospheric deposition of toxic metals.

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The differences in moss metal concentrations of different parts of the study area reflect local variation in TM deposition.

594

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- The observed concentrations of the contaminants, their relationships and associations, and the knowledge about the inventory of the probable emitting sources of the contaminants, in combination with GIS technology and factor analysis could be useful to establish the most probable emission sources of the elements in site specific areas.

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- Factor analysis proved to be a useful tool for the classification and the identification of the probable factors affecting air pollution.

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- Different climatic conditions during wet and dry deposition, is another key factor that impact on the concentrations of TM in atmospheric deposition. The significant differences on the concentration level of TM in different MS may partly affect by the variations on meteorology, such as dispersion and atmospheric conditions that differ from year to year.

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From this point of view, to avoid the effect of the meteorological variations on deposition

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rates of pollutants during wet and dry deposition, perhaps more precise criteria should be

607

recommended on the atmospheric conditions under which the sampling process should be

608

carried out

609

- The increment observed on 2015 AMS concentration level of anthropogenic trace elements (Cr, Cu, As, Ni and Zn) is an important finding for continuing future monitoring trends at a high spatial resolution and for strengthening the environmental masses for a cleaner environment with less contamination level of different contaminants.

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- Aiming to avoid the effect of meteorological and climatic variations on deposition rates of pollutants during wet and dry deposition, perhaps more precise criteria should be recommended on the atmospheric conditions under which the sampling process should be carried out.

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- The use of MB is a suitable tool that provides a cheap and effective alternative method of atmospheric deposition analysis. It indicates the potential role of vegetation in the removal of particular pollution.

618

619

620 • The obtained results may certify the approaches on how research indicators can be
621 transformed into direct application for management purposes and may help the policy
622 makers and regulators taking proper decisions that protect the environment.

623

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635

636 **References:**

- 637 Allajbeu, S., Qarri, F., Marku, E., Bekteshi, L., Ibro, V., Frontasyeva, V.M., Stafilov, T.,
638 Lazo, P., 2017. Contamination scale on atmospheric deposition for assessing air quality in
639 Albania evaluated from most toxic heavy metal and MB. *Air. Qual. Atmos. Health.* 10, 587–
640 599.
- 641 Amodio, M., Catino, S, Dambruoso, P.R., de Gennaro, G., Di Gilio, A., Giungato, P.,
642 Laiola, E., Marzocca, A., Mazzone, A, Sardaro, A., Tutino, M., 2014. Atmospheric Deposition:
643 Sampling Procedures, Analytical Methods, and Main Recent Findings from the Scientific
644 Literature. *Adv. Meteorology.* 2014, 1-27. <http://dx.doi.org/10.1155/2014/161730>.
- 645 Astel, A., Astel, K., Biziuk, A., 2008. PCA and multidimensional visualization
646 techniques united to aid in the bioindication of elements from transplanted Sphagnum
647 palustre moss exposed in Gdansk city area. *Environ. Sci. Pollut. Res.* 15(1), 41–50.
- 648 Barandovski, L., Frontasyeva, V.M., Stafilov, T., Šajin, R., Ostrovnaya, M.T., 2015.
649 Multielement atmospheric deposition in Macedonia studied by the moss biomonitoring
650 technique. *Environ. Sci. Pollut. Res.* 22,16077–16097. doi:10.1007/s11356-015-4787-x.
- 651 Barandovski, L., Cekova, M., Frontasyeva, M.V., Pavlov, S.S., Stafilov, T., Steinnes,
652 E., Urumov, V., 2008. Atmospheric deposition of trace element pollutants in Macedonia
653 studied by the moss biomonitoring technique. *Environ. Monit. Assess.* 138,107–118.
- 654 Bekteshi, L., Lazo, P., Qarri, F., Stafilov, T., 2015. Application of the normalization

655 process in the survey of atmospheric deposition of heavy metals in Albania through Moss
656 Biomonitoring. *Ecol. Indic.* 56, 50–59. doi:10.1016/j.ecolind.2015.03.001.

657 Berg, T., Pedersen, U., Steinnes, E., 1996. Environmental indicators for long-range
658 atmospheric transported heavy metals based on country MS. *Environ. Monit. Assess.* 43,
659 11–17.

660 Colette, A., Aas, W., Banin, L., Braban, C.F., Ferm, M., Ortiz, G.A, Ilyin, I., Mar, K.,
661 Pandolfi, M., Putaud, J.P., Shatalov, V., Solberg, S., Spindler, G., Tarasova,
662 O., Vana, M., Adani, M., Almodovar, P., Berton, E., Bessagnet, B., Bohlin-
663 Nizzetto, P., Boruvkova, J., Breivik, K., Briganti, G., Cappelletti, A., Cuvelier,
664 K., Derwent, R., D'Isidoro, M., Fagerli, Funk, C., Vivanco, M.G., Haeuber, R.,
665 Hueglin, C., Jenkins, S., Kerr, K., de Leeuw, F., Lynch, J., Manders, A., Mircea,
666 M., Pay, M.T., Pritula, D., Querol, X., Raffort, V., Reiss, I., Roustan, Y.,
667 Sauvage, S., Scavo, K., Simpson, D., Smith, I.R., Tang, Y.S., Theobald, M.,
668 Tørseth, K., Tsyro, S., van Pul, A., Vidic, S., Wallasch, M., Wind, P. Air pollution trends in the
669 EMEP region between 1990 and 2012. Kjeller, Norway, Norwegian Institute for Air Research,
670 105pp. (EMEP: CCC-Report 1/2016).

671 Costello, A.B., Jason, O., 2005. Best practices in exploratory factor analysis: four
672 recommendations for getting the most from your analysis. *Practical Assessment Research
673 and Evaluation.* 10(7), 1-9. Available online: <http://pareonline.net/getvn.asp?v=10&n=7>

674 Cao, J., Xu, X., Hylkema, M.N., Zeng, E.Y., Sly, P.D., Suk, W.A., Bergman, A.L., Huo,
675 X., 2016. Early-life exposure to widespread environmental toxicants and health risk: a focus
676 on the immune and respiratory systems. *Ann. Glob. Health.* 82(1), 119-31.
677 doi:10.1016/j.aogh.2016.01.023.

678 Carlon, C., 2007. Derivation methods of soil screening values in Europe. A review and
679 evaluation of national procedures towards harmonisation. European Commission, Joint
680 Research Centre, Ispra. EUR 22805-EN.

681 Chen L.C., Lippmann, M., 2009. Effects of metals within ambient air particulate
682 matter (PM) on human health. *Inhal. Toxicol.* 21, 1–31.

683 Connan, O., Maro, D., Hébert, D., Roupsard, P., Goujon, R., Letellier, B., Le Cavelier, S.,
684 2013. Wet and dry deposition of particles associated metals (Cd, Pb, Zn, Ni, Hg) in a rural
685 wetland site, Marais Vernier, France. *Atmos. Environ.* 67, 394-403.
686 doi:10.1016/j.atmosenv.2012.11.029

687 EMEP Report, Country-specific report for Albania within the CLRTAP and its related
688 Protocols. 2015. <http://www.msceast.org/index.php/albania>

689 Dockery, D.W., 2009. Health effects of particulate air pollution. *Ann. Epidemiol.*
690 19, 257–263.

691 Duffus, J.H., 2007. Carcinogenicity classification of vanadium pentoxide and

692 inorganic vanadium compounds, the NTP study of carcinogenicity of inhaled vanadium pent-
693 oxide and vanadium chemistry. Regul. Toxicol. Pharmacol. 47(1), 110–114.

694 EUR/03/5042688 (2003) Health Aspects of Air Pollution with Particulate Matter, Ozone and Nitrogen
695 Dioxide. Report on a WHO Working Group, Bonn, Germany. 1-94.

696 Fang, G.C., Chang, S.C., Chen, Y.C., Zhuang, Y.J., 2014. Measuring metallic elements of
697 total suspended particulates (TSPs), dry deposition flux, and dry deposition velocity for
698 easonal variation in central Taiwan. Atmos. Res. 143, 107–117.
699 doi:10.1016/j.atmosres.2014.02.002

700 Fernandez, J.A., Carballeira, A., 2000. Evaluation of contamination by different
701 elements in terrestrial mosses. Arch. Environ. Contam. Toxicol. 40, 461–468.
702 doi:10.1007/s002440010198.

703 Flentje, H., Briel, B., Beck, C., Collaud Coen, M.C., Fricke, M., Cyrus, J., Gu, J., Pitz, M.,
704 Thomas, W., 2015. Identification and monitoring of Saharan dust: An inventory
705 representative for south Germany since 1997. Atmos. Environ. 109, 87-96.
706 doi.10.1016/j.atmosenv.2015.02.023

707 Frontasyeva, M., Harmens, H., in collaboration with the participants., 2015. Monitoring of
708 Atmospheric Deposition of Heavy Metals, Nitrogen and Pops in Europe Using Bryophytes.
709 Monitoring Manual, 2015 Survey.
710 [https://icpvegetation.ceh.ac.uk/publications/documents/MossmonitoringMANUAL-2015-](https://icpvegetation.ceh.ac.uk/publications/documents/MossmonitoringMANUAL-2015-17.07.14.pdf)
711 [17.07.14.pdf](https://icpvegetation.ceh.ac.uk/publications/documents/MossmonitoringMANUAL-2015-17.07.14.pdf)

712 Gao, X., Yu, Q., Cheng, L.M., 2005. Health Effects of Airborne Particulate Matter
713 Trace Elements. Biomed. Environ. Sci. 18, 349-355.

714 Haridy, S., Wu, Z., 2009. Univariate and multivariate control charts for monitoring dynamic-
715 behavior processes: a case study. J. Ind. Eng. Manag. 2(3), 464–498.
716 doi:10.3926/jiem.2009.v2n3.

717 Harmens, H., Mill, G., Hayes, F., Sharps., K., Frontasyeva, M. and the participants of
718 the ICP Vegetation.,2016. Air Pollution and Vegetation ICP Vegetation Annual Report
719 2015/2016 <http://icpvegetation.ceh.ac.uk>

720 Harmens, H., Norris, D.A., Sharps, K., Mills, G., Alber, R., Aleksiyenak, Y., Blum, O.,
721 Cucu-Man, S.M., Dam, M., De Temmerman, L., Ene, A., Fernández, J.A., Martínez-Abaigar,
722 J., Frontasyeva, M., Godzik, B., Jeran, Z., Lazo, P., Leblond, S., Liiv, S., Magnússon, S.H.,
723 Maňková, B., Phil-Karlsson, G., Piispanen, J., Poikolainen, J., Santamaria, J.M.,Skudnik,
724 M., Spiric, Z., Stafilov, T., Steinnes, E., Stihi, C., Suchara, I., Thöni, L., Todoran, R.,
725 Yurukova, L., Zechmeister, H.G., 2015. Heavy metal and nitrogen concentrations in mosses
726 are declining across Europe whilst some “hotspots” remain in 2010. Environ. Pollut. 200, 93–
727 104.

728 Harmens, H., Norris, D.A., Steinnes, E., Kubin, E., Piispanen, J., Alber, R.,

729 Aleksiyenak, Y., Blum, O., Coskun, M., Dam, M., et al., 2010. Mosses as biomonitors of
730 atmospheric heavy metal deposition: spatial patterns and temporal trends in Europe. *Environ.*
731 *Pollut.* 158, 3144–3156.

732 Harmens, H., Norris, D., Mills, G., and the participants of the moss survey., 2013.
733 Heavy metals and nitrogen in mosses: spatial patterns in 2010/2011 and long-term temporal
734 trends in Europe. <http://icpvegetation.ceh.ac.uk>

735 Harmens, H., Norris, D.A., Koerber, G.R., Buse, A., Steinnes, E., Rühling, Å., 2008.
736 Temporal trends (1990–2000) in the concentration of cadmium, lead and mercury in mosses
737 across Europe. *Environ. Pollut.* 151, 368–76.

738 Harmens, H., Norris, D.A., Koerber, G.R., Buse, A., Steinnes, E., Rühling, Å., 2007.
739 Temporal trends in the concentration of arsenic, chromium, copper, iron, nickel, vanadium
740 and zinc in mosses across Europe between 1990 and 2000. *Atmos. Environ.* 41, 6673–87.

741 Hoerling, M., Eischeid, J., Perlwitz, J., Quan, X., Zhang, T., Pegion, P., 2012. On the
742 increased frequency of Mediterranean drought. *J. Clim.* 25 (6), 2146–2161.

743 Hoxha, P., Lipo, S., Dollma, M., 2014. GIS Technology on Natural Disasters Management
744 in Albania. *Int. J. Sci. Res.* 3(11), 1759-1764. ISSN (Online): 2319-7064. www.ijsr.net
745 <https://icpvegetation.ceh.ac.uk>
746 http://www.globalbispartners.com/uploads/albanian_mining_today.pdf
747 <https://tradingeconomics.com/albania/precipitation>

748 Huang, J., Kang, S., Zhang, Q., Yan, H., Guo, J.M., Jenkis M.G., Zang, G.S., Wang, K.,
749 2012. Wet deposition of mercury at a remote site in the Tibetan Plateau: concentrations,
750 speciation, and fluxes. *Atmos. Environ.* 62, 40–550.

751 Kampa, M., Castanas, E., 2008. Human health effects of air pollution. *Environ. Poll.*
752 151, 362-367. <https://doi.org/10.1016/j.envpol.2007.06.012>

753 Karanasiou, A., Moreno, N., Moreno, T., Viana, M., de Leeuw, F., Querol, X., 2012. Health
754 effects from Sahara dust episodes in Europe: Literature review and research gaps. *Environ.*
755 *Int.* 47, 107–114. doi:10.1016/j.envint.2012.06.012.

756 Lam, G.C.K., Leung, D.Y.C., Niewiadomski, M., Pang, S.W., Lee, A.W.F., Louie,
757 P.K.K., 1999. Street-level concentrations of nitrogen dioxide and suspended particulate
758 matter in Hong Kong. *Atmos. Environ.* 33,1–11.

759 Laze, P., Kovaçi, V., 1996. Soil erosion and physico-chemical nature of eroded materials. 9th
760 Conference of the International Soil Conservation Organization (ISCO), Bonn, Germany.
761 Extended Abstracts.

762 Lazo, P., Steinnes, E., Qarri, F., Allajbeu, S., Stafilov, T., Frontasyeva, M., Harmens,
763 H., 2018. Origin and spatial distribution of metals in moss samples in Albania: A hotspot of
764 heavy metal contamination in Europe. *Chemosphere.*190, 337-349.
765 <https://doi.org/10.1016/j.chemosphere.2017.09.132>

766 Lazo, P., Reif, J., 2013. Vlora, an Abandoned PVC Factory at the Mediterranean Coast.
767 Mercury Pollution, Threat to Humans, and Treatment Options, in: Wagner-Döbler, I. (Eds.),
768 Bioremediation of Mercury: Current Research and Industrial Applications. Caister Academic
769 Press, Germany, pp. 67-79.

770 Mettam, G.R., Adams, L.B., 2009. How to prepare an electronic version of your article, in: Jones,
771 B.S., Smith, R.Z. (Eds.), *Introduction to the Electronic Age*. E-Publishing Inc., New York, pp.
772 281–304.

773 Lazo, P., Bekteshi, L., Shehu, A., 2013. Active moss biomonitoring technique for
774 atmospheric deposition of heavy metals study in Elbasan city, Albania. *Fresen. Environ. Bull.*
775 23(1a), 213–219.

776 Lazo, P., Cullaj, A., 2002. Determination of the different states of mercury in seawater near the
777 Vlora and Durres Bays. [Anal. Chem.](#) 374, 1034-1038.

778 Liu, L., Zi, X., Zhang, J., Wang, Z., 2013. A sequential rank-based nonparametric
779 adaptive EWMA control chart. *Commun. Stat. Simul. Comput.* 42(4), 841–
780 859.

781 Lindberg, S. E., Harriss, R. C., 1981. The role of atmospheric deposition in an eastern
782 U.S. deciduous forest. *Water Air Soil Pollut.* 16, 13–31. doi:10.1007/BF01047039.

783 Marka, J., Sabovljevic, M., 2011. New bryophyte records from Albania. *J. Biol.* 33(1),
784 74–76.

785 Matschullat, J., Ottenstein, R., Reimann, C., 2000. Geochemical background—can we
786 calculate it? *Environ. Geol.* 39(9), 990–1000.

787 Meyer, C., Diaz-de-Quijano, M., Monna, F., Franchi, M., Toussaint, M-L., Gilbert, D.,
788 Bernard, N., 2015. Characterisation and distribution of deposited trace elements transported
789 over long and intermediate distances in north-eastern France using Sphagnum peatlands as
790 a sentinel ecosystem. *Atmospheric Environment*. 101, 286-293.
791 doi.org/10.1016/j.atmosenv.2014.11.041

792 Miranda, A., Silveira, C., Ferreira, J., Monteiro, A., Lopes, D., Relvas, H., Borrego, C.,
793 Roebeling, P., 2015. Current air quality plans in Europe designed to support air quality
794 management policies. *Atmos. Pollut. Res.* 6, 434-443.
795 <https://doi.org/10.5094/APR.2015.048>.

796 Morawska, L., Thomas, S., Bofinger, N., Wainwright, D., Neale, D., 1998.
797 Comprehensive characterization of aerosols in a subtropical urban atmosphere: particle size
798 distribution and correlation with gaseous pollutants. *Atmos. Environ.* 32, 2467–2478.

799 Morales-Baquero, R., Pulido-Villena, E., Reche, I., 2013. Chemical signature of
800 saharan dust on dry and wet atmospheric deposition in the south-western Mediterranean
801 region. *Tellus B:Chemical and Physical Meteorology*. 65 (1).
802 <http://dx.doi.org/10.3402/tellusb.v65i0.18720>

803 Muezzinoglu, A. and Cizmecioglu, S. C., 2006. Deposition of heavy metals in a
804 Mediterranean climate area, *Atmos. Res.* 81, 1–16. doi:10.1016/j.atmosres.2005.10.004.

805 Nastos, P.T., 2012. Meteorological Patterns Associated with Intense Saharan Dust Outbreaks
806 over Greece in Winter. *Adv. Meteorology.* 1-17. doi:10.1155/2012/828301.

807 Pachon, J.E., Weber, J.R., Zhang, X., Mulholl, J.A., Russell, A.G., 2013. Revising the
808 use of potassium (K) in the source apportionment of PM_{2.5}. *Atmos. Pollut. Res.* 4, 14-21.

809 Paupério, A., Severo, M., Lopes, C., Moreira, P., Cooke, L., Oliveira, A., 2014. Could
810 the Food Neophobia Scale be adapted to pregnant women? A confirmatory factor analysis in
811 a Portuguese sample. *Appetite.* 75, 110–116.

812 Paupério, A., Severo, M., Lopes, C., Moreira, P., Cooke, L., Oliveira, A., 2014. Could the Food
813 Neophobia Scale be adapted to pregnant women? A confirmatory factor analysis in a
814 Portuguese sample. *Appetite.* 75, 110–116.

815 Qarri, F., Lazo, P., Stafilov, T., Frontasyeva, M., Harmens, H., Bekteshi, L., Baceva,
816 K., Goryainova, Z., 2013. Multi-elements atmospheric deposition study in Albania. *Environ.*
817 *Sci. Pollut. Res.* 21, 2506-2518. doi:10.1007/s11356-013-2091-1

818 Qarri, F., Lazo, P., Bekteshi, L., Stafilov, T., Frontasyeva, M., Harmens, H., 2014a.
819 The effect of sampling scheme in the survey of atmospheric deposition of heavy metals in
820 Albania by using moss biomonitoring. *Environ. Sci. Pollut. Res.* 22(3), 2258-2271.
821 doi:10.1007/s11356-014-3417-3.

822 Qarri, F., Lazo, P., Stafilov, T., Bekteshi, L., Baceva, K., Marka, J., 2014b. Survey of
823 atmospheric deposition of Al, Cr, Fe, Ni, V and Zn in Albania by using Moss biomonitoring
824 and ICP-AES. *Air. Qual. Atmos. Health.* 7, 297-307. doi:10.1007/s11869-014-0237-z.

825 Reimann, C., Filzmoser, P., Garrett, R.G., 2005. Background and threshold: critical
826 comparison of methods of determination. *Sci. Total. Environ.* 346(1–3), 1–16.
827 doi:10.1016/j.scitotenv.2004.11.023

828 Reimann, C., Filzmoser, P., Garrett, R., 2002. Factor analysis applied to regional
829 geochemical data: Problems and possibilities. *Appl. Geochemistry.* 17(3),185-206. doi:
830 [10.1016/S0883-2927\(01\)00066-X](https://doi.org/10.1016/S0883-2927(01)00066-X)

831 Rudnick, R.L., Gao, S., 2004. Composition of the continental crust. *Treatise on Geochem.* 3, 1–64.

832 Rühling, A., 1994. Atmospheric heavy metal deposition in Europe—estimation based
833 on moss analysis. *NORD 1994*, 9. Nordic Council of Ministers, Copenhagen, Denmark.

834 Rühling, Å., Steinnes, E., 1998. Atmospheric heavy metal deposition in Europe 1995–1996, *NORD*
835 *environment 1998*, 15. Nordic Council of Ministry, Copenhagen, Denmark.

836 Rühling, Å., Tyler, G., 1968. An ecological approach to the lead problem. *Bot. Notiser.*
837 122, 248–342.

838 Quennehen, B., Schwarzenboeck, A., Matsuki, A., Burkhart, J.F., Stohl, A., Ancellet,

839 G., Law, K.S., 2012. Anthropogenic and forest fire pollution aerosol transported to the Arctic:
840 observations from the POLARCAT-France spring campaign. *Atmos. Chem. Phys.* 12, 6437–
841 6454. doi:10.5194/acp-12-6437-2012

842 Qiu, P., Tian, H., Zhu, C., Liu, K., Gao, J., Zhou, J., 2014. An elaborate high
843 resolution emission inventory of primary air pollutants for the Central Plain Urban
844 Agglomeration of China. *Atmos. Environ.* 86, 9-101. doi.org/10.1016/j.atmosenv.2013.11.062

845 Salcedo, R.L.R., Alvim Ferraz, M.C.M., Alves, C.A., Martins, F.G., 1999. Time series
846 analysis of air pollution data. *Atmos. Environ.* 33, 2361–2372.

847 Sardans, J., Penuelas, J., 2005. Trace element accumulation in the moss *Hypnum*
848 *cupressiforme* Hedw. and the trees *Quercus ilex* L. and *Pinus halepensis* Mill. in Catalonia.
849 *Chemosphere.* 60, 1293–1307

850 Schaumann, F., Borm, P.J., Herbrich, A., Knoch, J., Pitz, M., Schins, R.P., Luettig, B.,
851 Hohlfeld, J.M., Heinrich, J., Krug, N., 2004. Metal-rich ambient particles (particulate matter
852 2.5) cause air way inflammation in healthy subjects. *Am. J. Respir. Crit. Care. Med.* 170,
853 898–903.

854 Schröder, W., Nickel, S., Schönrock, S., Meyer, M., Wosniok, W., Harmens, H.,
855 Frontasyeva, M.V., Alber, R., Aleksiyayena, J., Barandovski, L., Carballeira, A., Danielsson,
856 H., de Temmermann, L., Godzik, B., Jeran, Z., Karlsson, G.P., Lazo, P., Leblond, S.,
857 Lindroos, A.J., Liiv, S., Magnússon, S.H., Mankovska, B., Martínez-Abaigar, J., Piispanen,
858 J., Poikolainen, J., Popescu, I.V., Qarri, F., Santamaria, J.M., Skudnik, M., Špirić, Z., Stafilov,
859 T., Steinnes, E., Stihl, C., Thöni, L., Uggerud, H.T., Zechmeister, H.G., 2016. Spatially valid
860 data of atmospheric deposition of heavy metals and nitrogen derived by MS for pollution risk
861 assessments of ecosystems. *Environ. Sci. Pollut. Res.* 23 (11), 10457-10476.
862 doi:10.1007/s11356-016-6577-5.

863 Schröder, W., Pesch, R., Hertel, A., Schonrock, S., Harmens, H., Mills, G., Ilyin, I.,
864 2013. Correlation between atmospheric deposition of Cd, Hg and Pb and their
865 concentrations in mosses specified for ecological land classes covering Europe. *Atmos.*
866 *Pollut. Res.* 4, 267-74.

867 Shallari, S., Schwartz, C., Hasko, A., Morel, J.L., 1998. Heavy metals in soils and plants of
868 serpentine and industrial sites of Albania. *Sci. Tot. Environ.* 209, 133-142

869 Spinoni, J., Naumann, G., Vogt, J.V., 2017. Pan-European seasonal trends and recent
870 changes of drought frequency and severity. *Glob. Planet. Change.* 148, 113–130.
871 doi.org/10.1016/j.gloplacha.2016.11.013 0921-8181

872 Spinoni, J., Naumann, G., Vogt, J.V., Barbosaa, P., 2017. The biggest drought events in
873 Europe from 1950 to 2012. *Journal of Hydrology: Reg. Stud.* 3, 509–524.
874 doi.org/10.1016/j.ejrh.2015.01.001 2214-5818.

875 Špirić, Z., Vučković, I., Stafilov, T., Kušan, V., Frontasyeva, V.M., 2013. Air pollution

876 study in Croatia using moss biomonitoring and ICP–AES and AAS analytical techniques.
877 Arch. Environ. Contam. Toxicol. 65, 33–46.

878 Stafilov, T., Šajin, R., Barandovski, L., Bačeva, A.K., Malinowska, S., 2018. Moss
879 biomonitoring of atmospheric deposition study of minor and trace elements in Macedonia. Air.
880 Qual. Atmos. Health. 11 (2), 137–152. <https://doi.org/10.1007/s11869-017-0529-1>.

881 Steinnes, E., Uggerud, H.T., Pfaffhuber, K.A., Berg, T., 2016. Atmospheric deposition
882 of heavy metals in Norway–country Moss Survey 2015. M–595. Norwegian Institute for Air
883 Research, Trondheim.

884 Steinnes, E., Berg, T., Uggerud, H.T., 2011. Three decades of atmospheric metal deposition in
885 Norway as evident from analysis of moss samples. Sci. Total. Environ. 412/413, 351-358.
886 doi: 10.1016/j.scitotenv.2011.09.086.

887 Steinnes, E., Berg, T., Sjøbakk, T.E., 2003. Temporal and spatial trends in Hg
888 deposition monitored by moss analysis. Sci. Total. Environ. 304, 215–219.

889 Steinnes, E., Allen, R.O., Petersen, H.M., Rambaek, J.P., Varskog, P., 1997a. Evidence of large
890 scale heavy- metal contamination of natural surface soils in Norway from long-range
891 atmospheric transport. Sci. Total. Environ. 205, 255-266.

892 Steinnes, E., Rühling, Å., Lippo, H., Mäkinen, A., 1997b. Reference materials for large-scale metal
893 deposition surveys. Accred. Qual. Assur. 2, 243-249.

894 Steinnes, E., 1995. A critical evaluation of the use of naturally growing moss to monitor the
895 deposition of atmospheric metals. Sci. Total. Environ. 160/161, 243-249.

896 Wang, Z., Darilek, J.L., Zhao, Y., Huang, B., Sun, W., 2010. Defining soil
897 geochemical baselines at small scales using geochemical common factors and soil organic
898 matter as normalizers. J. Soils. Sediments. 11, 3-14. doi:10.1007/s11368-010-0269-4.

899 Tafuro, A.M., Barnaba, F., De Tomasi, F., Perrone, M.R., Gobbi, G.P., 2006. Saharan dust
900 particle properties over the central Mediterranean. Atmos. Res. 81, 67– 93.
901 doi:10.1016/j.atmosres.2005.11.008

902 Theodosi, C., Markaki, Z., Tselepidis, A., Mihalopoulos, N., 2010. The significance of
903 atmospheric inputs of soluble and particulate major and trace metals to the eastern
904 Mediterranean seawater. Mar. Chem. 120, 154–163. doi:10.1016/j.marchem.2010.02.003

905 Thunis, P., Miranda, A., Baldasano, J.M., Blond, N., Douros, J., Graff, A., Janssen, S., Rezler,
906 J.K., Karvosenoja, N., Maffeis, G., Martilli, A., Rasoloharimahefa, M., Real, E., Viaene, P.,
907 Volta, M., White, L., 2016. Overview of current regional and local scale air quality modelling
908 practices: Assessment and planning tools in the EU. Environ. Sci. Policy. 65, 13–21.

909 UNDP-Albania., 2010. Ferrochrome smelter in Elbasan; Fwienv contract n_ 1-BH-0381. In:
910 Consultancy Services to Conduct Environmental Impact Assessment of Ten High Priority
911 Environmental Hotspots to Form the Basis for a Major Remediation Programme, for the

912 Project: "Identification and Prioritization of Environmental Hot Spots in Albania" Eia Report
913 Final.

914 UNDP CO- ALBANIA., 2018. Contribution to Sub-Regional HDR. [https://info.undp.org](https://info.undp.org/docs/pdc/Documents/ALB/Albania%20Regional%20HDR%20contribution%20July%2018-20Strengthening%20resilience%20LGU%20Atlas%2004120.pdf)
915 /docs/pdc/Documents/ALB/Albania%20Regional%20HDR%20contribution%20July%2018-
916 20Strengthening%20resilience%20LGU%20Atlas%2004120.pdf

917 Varela, Z., Fernandez, J.A., Real, C., Carballeira, A., Aboal, J.R., 2015. Influence of the
918 physicochemical characteristics of pollutants on their uptake in moss. *Atmos. Environ.* 102,
919 130-135. <http://dx.doi.org/10.1016/j.atmosenv.2014.11.061>

920 Vuković, G., Aničić, U.M., Razumenić, I., Goryainova, Z., Frontasyeva, V.M.,
921 Tomašević, M., Popović, A., 2013. Active moss biomonitoring of small-scale spatial
922 distribution of airborne major and trace elements in the Belgrade urban area. *Environ. Sci.*
923 *Pollut.* 20, 5461–5470.

924 Vinogradov, A.P., 1962. Average contents of chemical elements in the principal type of igneous
925 rocks of the Earth's crust. *Geokhimiya.* 7, 641–664.

926 Wu, Y., Zhang, J., Ni, Z., Liu, S., Jiang, Z., Huang X., 2018. Atmospheric deposition of trace
927 elements to Daya Bay, South China Sea. *Mar. Pollut. Bull.* 127, 672–683.
928 [doi:10.1016/j.marpolbul.2017.12.046](https://doi.org/10.1016/j.marpolbul.2017.12.046)

929 Yang, F., Tan, J., Shi, Z. B., Cai, Y., He, K., Ma, Y., Duan, F., Okuda, T., Tanaka, S., Chen, G.,
930 2012. Five-year record of atmospheric precipitation chemistry in urban Beijing, China. *Atmos.*
931 *Chem. Phys.* 12, 2025–2035. [doi:10.5194/acp-12-2025-2012](https://doi.org/10.5194/acp-12-2025-2012).

932 Zeng, X., Xu, X., Zheng, X., Reponen, T., Chen, A., Huo, X., 2016. Heavy metals in
933 PM_{2.5} and in blood, and children's respiratory symptoms and asthma from an e-waste
934 recycling area. *Environ. Poll.* 210, 346–353.

935 Zhang, F., Cheng, H., Wang, Z., Lv, Zhu, Z., Zhang, G., Wang, X., 2014. Chemical compositions,
936 seasonal variations and regional pollution events. *Atmos. Environ.* 86, 193-20.