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

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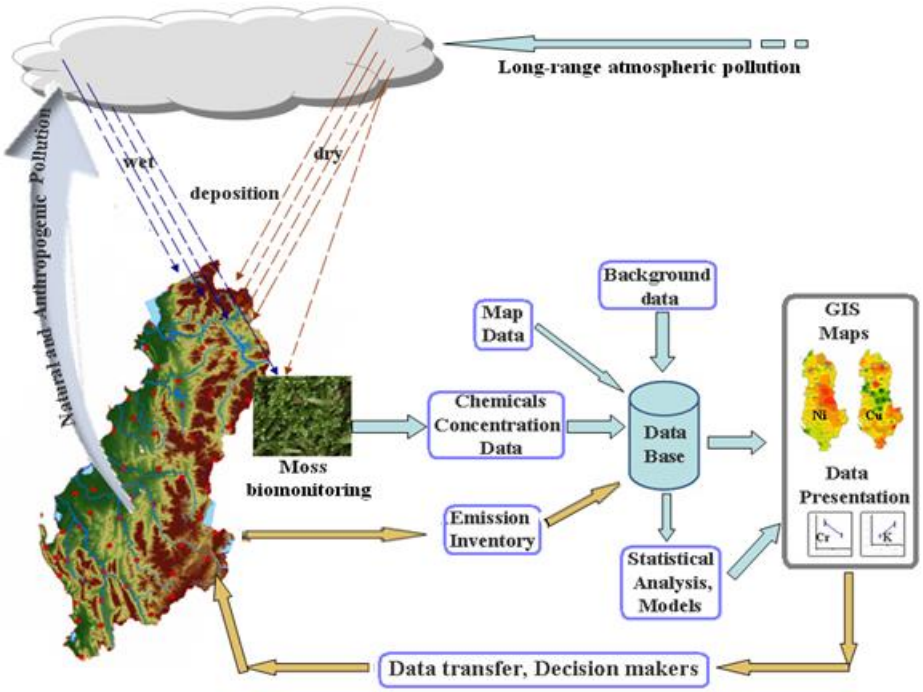
ABSTRACT

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

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

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



Graphical Abstract

Highlights:

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

Abbreviations:

AAS – Atomic absorption specroscopy
AMS – Albanian moss survey
BC - background concentration
CF – contamination factor
C_i – Concentration of element i
CVAAS - cold vapor atomic absorption spectrometry
ETAAS - electrothermal atomic absorption spectrometry
EWMA - Exponentially weighted moving average
FA – Factor analysis
FPM – Fine Particulate Matter
GIS - Geographic Information System
ICP-AES - inductively coupled plasma atomic emission spectrometry
L_i - individual loading of element i
LRTAP – Long-Range Transboundary Air Pollution
MB – Moss biomonitoring
NBC – national background concentration
TM – trace metal

1. Introduction

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

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

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

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

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

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

2. Material and method

2.1. Description of the study area

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

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

2.2. Sampling procedure

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

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

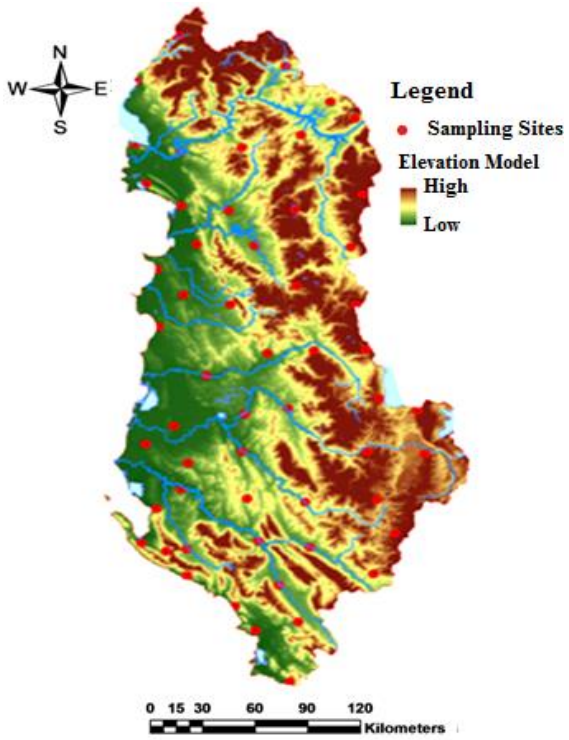


Fig. 1. GIS map for the localization of sampling sites (latitude, 41°00′N of the equator; longitude, 20°00′E of Greenwich)

2.3. Chemical analysis

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

2.4. *Quality control*

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

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

2.5. *Data processing and statistical analyses*

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

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

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

$$CF = C_i / NBC_i$$

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

Spearman correlation analysis was carried out to measure the strength of the association between two ranked monotonic variables that changes together in the same sense. Beside it the Pearson correlation analysis was tested to distinguish the association between elements with strong linear combinations. The significance of Pearson correlation was examined by linear regression analysis, based on the slope of the regression line ($s \neq 0$) and $P<0.05$ for R^2 (slope) $\neq 0$. The formal hypothesis test for the relationship between variables is confirmed by the overall F-test that determines whether this relationship is statistically significant. If the P value of the obtained overall F-test is less than the significance level (0.05), it can conclude that the relationship is true and R-squared value is significantly different from zero.

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

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

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

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

3. **Results and discussion**

3.1. Frequency distribution

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

3.2. Elemental concentration and contamination scale

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

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

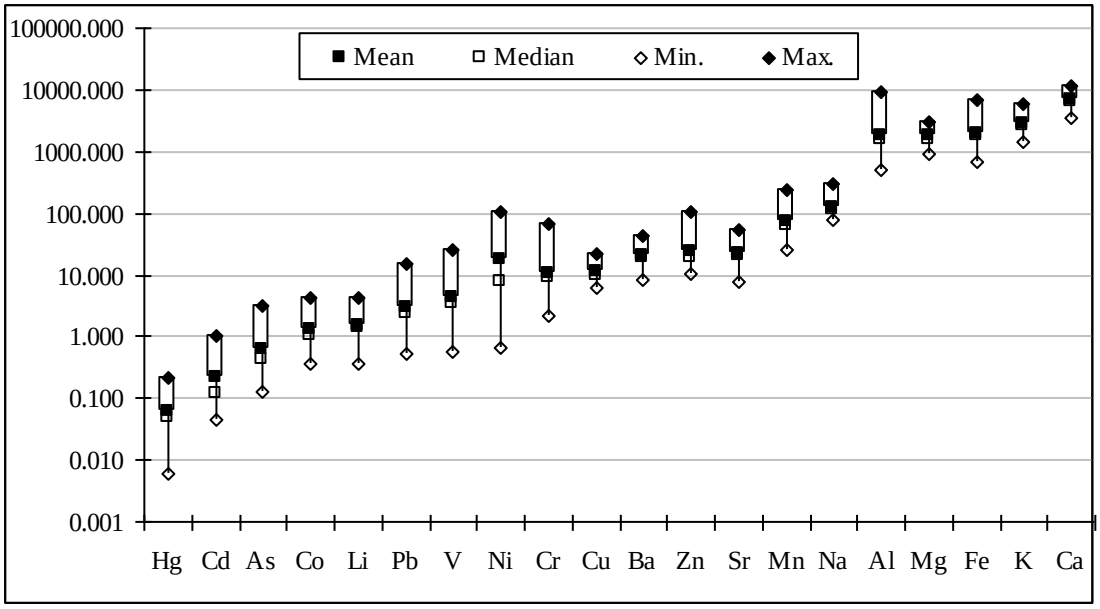


Fig. 2. Box-plot diagram of concentration data (mg/kg, DW)

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

Table 2 The contamination factors (CFs) and contamination classification (Fernandez et al., 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]

[1] No contamination; [2] Suspected contamination; [3] Slight contamination; [4] Moderate contamination; [5] Serious contamination

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

contamination scale, C4 (moderate contamination), all are probably linked with long-range atmospheric transport of pollutants from other parts of Europe (Harmens et al., 2015). In addition, it is most likely derived from local factors such as fuel combustion, wind blowing fine mineral dust particles originating from industrial waste deposits, mine and metal processing industry, and geogenic factors (Harmens et al., 2010; Lazo et al., 2018). The CFs of As, Cd, Co, Fe, Hg, Al, Li, Sr and Mn are associated with the 3rd category of the contamination scale, C3 (slight contamination). The CFs of Cu, Zn, Ca, Mg, Ba, Na and K are associated with the second category of the contamination scale, C2 (suspended contamination), that is easily obtained from natural variation (Fenandez et al., 2000).

Similar results were obtained for CF values of the elements As, Cu, Mg, Ba, Na, K and Li for current data and AMS 2010 data (Qarri et al., 2013). The differences observed in CFs of Cd, Cr, Ni, Fe, Mn, Pb, V, Al, Sr, Mn, Ca and Zn are mostly affected by different background levels (BC_i) used for 2010 AMS (data from a pristine area in Norway, Steinnes et al., 2007) and 2015 (country background level, NBC), and by the differences between the respective median concentration values (C_i) of the elements on 2010 and 2015 AMS (Table 2S).

The median concentrations of 2015 AMS are compared with similar national studies done in Macedonia (a neighboring country) (Stafilov et al., 2017) and Norway, selected as a clean area in Europe (Steinnes et al., 2016) (Table 3).

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

	Albania[1] n=55		Macedonia[2] n=72		Norway[3] n=464
Elements	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

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

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

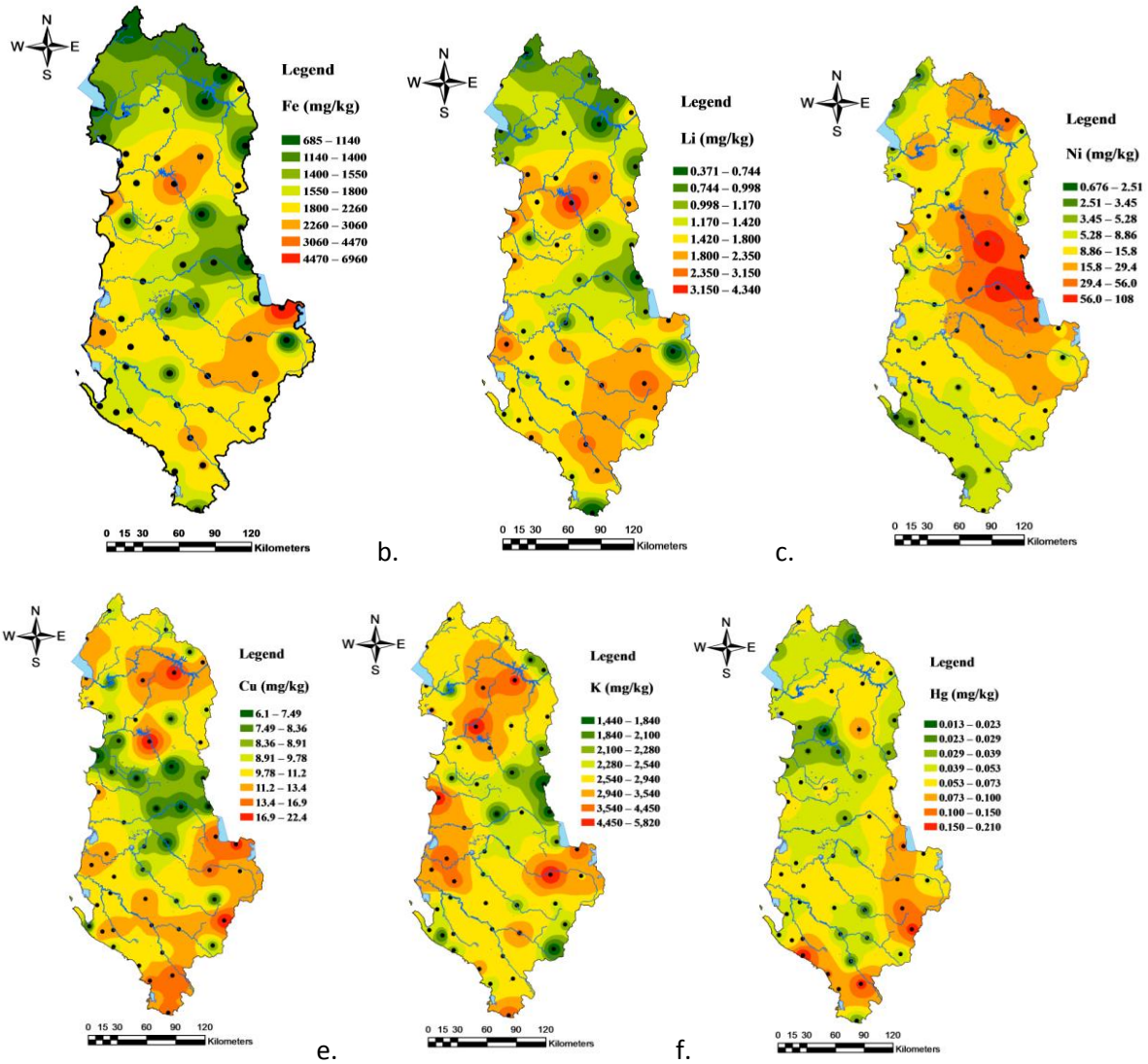


Fig. 3. GIS maps of TM atmospheric deposition in Albania. a. Fe, b. Li, c. Ni, d. Cu, e. K, f. Hg

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

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

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

3.3. Temporal trends of TM in mosses

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

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

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

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

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

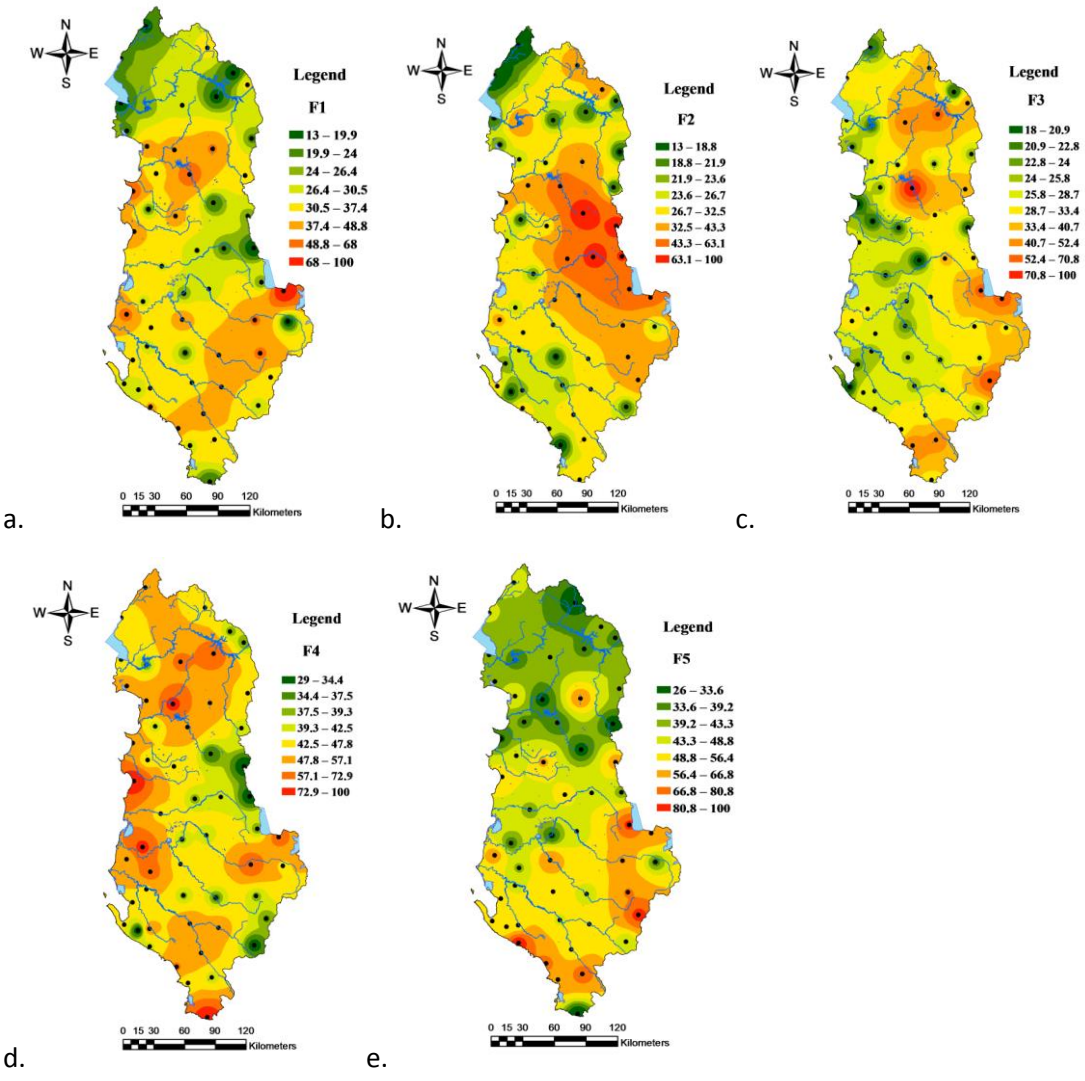
494 during wet deposition. It is probably derived from relatively high wet deposition flux of Na
495 and by high solubility of Na in coastal areas (Miranda et al., 2015; Baqueros et al., 2013).
496 Wet deposition is often considered as an important natural factor in cleaning the
497 atmosphere and removing 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

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

Table 4 The results of the FA analysis of the correlation matrix (Sorted Varimax Rotated 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



a. b. c.

d. e.

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

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

Factor 1 (F1) is the strongest factor that presents 27.6 % of the total variance. It is characterized by high loadings of Fe, Al, V, Li, Ba and Sr (FL > 0.76) and lower loading of Mn (FL = 0.58). These elements are naturally distributed as typical soil elements (Rudnick 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).

4. Conclusions

The following conclusions can be drawn from this study:

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

● 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.

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

● 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. From this point of view, to avoid the effect of the meteorological 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

● 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.

● 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.

● 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.

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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