# **Atmospheric deposition of trace elements in Greece using moss**  *Hypnum cupressiforme* **Hedw. as biomonitors**

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Received: 21 December 2018 / Published online: 8 May 2019 © Akadémiai Kiadó, Budapest, Hungary 2019

#### **Abstract**

Mosses have been used for monitoring trace elements airborne pollution due to their ability to accumulate elements directly from wet and dry deposition. During the 2015/2016 European moss survey, ninety-fve samples of *Hypnum cupressiforme* Hedw. were collected in Northern Greece. Mosses were analyzed to the content of Neutron Activation Analysis and the concentrations of thirty-three elements were determined. The contamination factors for the following elements Al, As, V, Ni, Fe, Cr, Zn were calculated. Information about air quality was provided through the study of atmospheric deposition of trace elements in mosses in the vicinity of Northern Greece.

**Keywords** Atmospheric deposition · Trace elements · Moss · Neutron activation analysis · Groundwater

# **Introduction**

Human activities have a great impact on atmospheric chemistry. Since industrial revolution and due to the increasing anthropogenic activities and urbanization, a lot of changes in atmosphere have occurred  $[1, 2]$  $[1, 2]$  $[1, 2]$  $[1, 2]$ . Heavy metals can enter in all ecosystems while they are emitted to the atmosphere in different forms gas or particulate matter  $[3, 4]$  $[3, 4]$ . There are diferent sources of the atmospheric emitted heavy metals

**Electronic supplementary material** The online version of this article [\(https://doi.org/10.1007/s10967-019-06535-4\)](https://doi.org/10.1007/s10967-019-06535-4) contains supplementary material, which is available to authorized users.

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such as housing and commercial properties, industrial processes, road transportations and agriculture. Heavy metals can be transported over long distances and be deposited many kilometers away from their emitting sources, impacting on human health and the environment [\[5](#page-8-4)]. Depending on their concentrations, they can be hazardous for plants, animals and humans afecting the whole food chain. The increase of atmospheric pollutants in urban and industrial areas and their infuence in human health and environment represents one of the main European scientifc concerns since the 1970's  $[2, 6]$  $[2, 6]$  $[2, 6]$  $[2, 6]$ . As a result, controlling the air quality is considered necessary.

During the last 20 years, diferent instructions (1999/30/ EC, 2002/3/EC, 2004/107/EC and 2008/50/EC) have been officially followed by European community for air quality assessments related to metals, nitrogen oxides and dioxides, Sulphur dioxide, and ozone concentrations in the atmosphere [[6\]](#page-8-5). The international cooperation for the reduction of air pollution during this period, has signifcantly contributed to the decrease of trace elements emissions in Europe [[5,](#page-8-4) [7](#page-8-6)]. For monitoring the quality of air, diferent complementary modelling techniques are used, and they are mostly based on physico-chemical measurements of diferent air pollutants [[8,](#page-8-7) [9](#page-8-8)]. Monitoring the quality of ambient air is a complex procedure, as it requires expensive analytic instrumentations, large number of potentially dangerous substances and temporal and spatial variations in the input rates of diferent pollutants  $[2, 9-11]$  $[2, 9-11]$  $[2, 9-11]$  $[2, 9-11]$ . Beyond all the previous, the conventional methods that are used for the atmospheric deposition of heavy metals, require a big number of samples in frequent intervals, so the under investigation area can be pretty limited  $[4, 12-14]$  $[4, 12-14]$  $[4, 12-14]$  $[4, 12-14]$ . Due to these difficulties, different alternatives were under investigation and fnally the idea of biomonitoring became a reality in the late 1960's.

Biomonitoring is a technique that gives the possibility to detect spatial and temporal variations in the atmospheric deposition of diferent pollutants and to defne their possible sources [\[2](#page-8-1), [9](#page-8-8), [15](#page-8-12)[–25](#page-9-0)]. This technique uses living organisms as biomonitors either passively or actively, based on their sensitivity to air pollution or their capacity of accumulating pollutants in their tissues [[2,](#page-8-1) [26](#page-9-1), [27](#page-9-2)]. During the recent decades and among the diferent living organisms (vascular plants, mosses and lichens) that can be used as bioaccumulators of airborne pollutants [\[2](#page-8-1), [28](#page-9-3)[–31\]](#page-9-4), mosses and lichens have been systematically chosen for environmental monitoring. Mosses are special living organisms- one of the earliest land plants in evolutionary terms [\[32](#page-9-5)], and they have reasonably attracted the interest of the researchers as they present very unique characteristics that are not noted in the other plants [\[33](#page-9-6)].

Using mosses as bioindicators of atmospheric fallout, is based on the fact that elemental concentrations in mosses are closely related to atmospheric deposition [[34–](#page-9-7)[38](#page-9-8)]. Mosses are able to accumulate airborne pollutants such as heavy metals  $[19, 35, 37, 39-42]$  $[19, 35, 37, 39-42]$  $[19, 35, 37, 39-42]$  $[19, 35, 37, 39-42]$  $[19, 35, 37, 39-42]$  $[19, 35, 37, 39-42]$  $[19, 35, 37, 39-42]$  $[19, 35, 37, 39-42]$  and organic compounds  $[25]$ due to their morphological and physiological characteristics [[2\]](#page-8-1). Mosses are non-vascular plants and they develop rhizoids while being in close contact to their substrate [[43,](#page-9-13) [44](#page-9-14)]. They have no rooting system and all the nutrients and the water are obtained mainly from precipitation and dry deposition. The uptake of trace elements from the ground is not significant  $[22]$  $[22]$ . Due to their slow growth rate they are able to accumulate trace elements over a large time period. Each annual growth segment of mosses reveals the elemental concentrations of each current year [[45\]](#page-9-16). Some moss species are appropriate for assessing the quality of air on large geographic scale due to the fact that they compose extensive populations [[2,](#page-8-1) [20](#page-8-14)[–24\]](#page-9-17). The concentrations of elements in mosses in a large geographic scale may vary because the accumulation of elements in moss tissues can be seriously infuenced by local pollution sources and different environmental factors  $[2, 11, 19]$  $[2, 11, 19]$  $[2, 11, 19]$  $[2, 11, 19]$  $[2, 11, 19]$  $[2, 11, 19]$ . The moss analysis technique is an easy and less expensive method than the conventional deposition analysis as no special deposition collectors are required and a high sampling density can be carried out [\[22,](#page-9-15) [46,](#page-9-18) [47](#page-9-19)]. Mosses do not provide direct quantitative measurements of elements deposition in mosses. However, relating the information from diferent moss surveys to deposition monitoring data for each country, information about elemental concentrations can be extracted [\[22](#page-9-15), [48,](#page-9-20) [49](#page-9-21)].

More specifcally, the measured concentrations of elements in mosses and in wet deposition can be compared by using linear regression analysis. Then the elemental moss concentrations can be transformed to absolute deposition rates by using the calculated regression equations [\[48](#page-9-20)].

Since 1990 the European moss survey takes place every 5 years providing data for trace elements depositions from the atmosphere to naturally growing mosses [[5,](#page-8-4) [7](#page-8-6), [20](#page-8-14), [22,](#page-9-15) [25](#page-9-0), [50–](#page-9-22)[55](#page-10-0)]. This European survey is being coordinated by the ICP Vegetation (International Cooperative Program on Efects of Air Pollution on Natural Vegetation and Crops) Coordination Centre at the Centre for Ecology and Hydrology, Bangor, UK [[22\]](#page-9-15). Moss surveys provide spatial and temporal trends in trace elements concentration and the identifcation of local or transboundary sources [[56,](#page-10-1) [57\]](#page-10-2). Greece provides for the frst time data on concentrations of several moss monitored metals for the North part of the country. *Hypnum cupressiforme* is the most frequently moss species that can be found in the territory of Greece, due to the dry climate and the mountainous morphology in the biggest part of the country.

The objective of this study is to present the spatial atmospheric deposition of trace elements in the province of Northern Greece for identifcation of possible sources, as well as the comparison of the obtained results with the results of the neighboring and other European countries and set up the frst database of the country for future surveys.

# **Experimental**

#### **Study area and moss sampling**

The study area was situated in the region of Northern Greece (Fig. [1](#page-2-0)). The sampling net included 95 sites in the Hellenic prefectures of West, Central, East Macedonia and Thrace, where the recommended moss species (*Hypnum Cupressiforme* Hedw., *Pseudoscleropodium purum*) could have been found in required quantities. More specifcally sampling sites were located from 39.97° North to 41.65° North and from 20.97° East to 26.26° East covering a grid of 30 m $\times$ 30 m. Moss samples were collected from diferent elevations from 30 m to 1450 m above the mean sea level.

All samples of fresh plant material were collected during the end of summer 2016. There was no rain during the sampling. Samples were taken from open regions in most of the sampling sites avoiding possible contact of mosses with surface water. The sampling sites were located at least 300 m from main roads, 100 m from local roads and 200 m from populated areas. All samples were collected according to the requirements of the Protocol of the European Survey ICP Vegetation [[50\]](#page-9-22).



<span id="page-2-0"></span>**Fig. 1** Moss sampling net in 95 sites along the study area of Northern Greece

## **Sample preparation and analysis**

All the samples were analyzed by means of Neutron Activation Analysis (NAA) performed in the radio-analytical Frank Laboratory of Neutron Physics, Joint Institute for Nuclear Research (JINR) in Dubna, Russia.

After sampling and identifcation of the moss species, mosses were cleaned from impurities very carefully and were stored in deep frozen until further treatment. No chemical treatment was used and mosses were not washed. Before analysis mosses were air-dried at 40 °C for 3 h. After that, the dried material was well homogenized on an agate mortar. From the homogenized material samples with weight of 0.3 g were measured on a scale and formed into the shape of pills by means of a pneumatic press and ceramic matrices. The pills were then packed in two separate ways—in plastic bags for short irradiation and in aluminum foil cups for long term irradiation.

Using a pneumatic rabbit system, the prepared and packed samples were sent to two diferent irradiation channels of the pulsed fast IBR-2 reactor of the JINR for long and short term irradiation respectively. The plastic packed samples were irradiated for 180 s to determine the short lived isotopes (Na, Mg, Al, Cl, Ti, V, Mn, Ni, I, In, Ag), while the aluminum packed samples were irradiated for 4–5 days to determine long lived isotopes (K, Ca, Sc, Cr, Fe, Co, Zn, As, Se, Br, Rb, Sr, Zr, Mo, Sb, Ba, La, Sm, Gd, Au, Th, U). The irradiated samples were then measured on HPGe detectors with relative efficiency 40% and 1.74 keV FWHM at the 1332 keV line of  ${}^{60}Co$ . To determine the short lived isotopes, samples that were irradiated for a short period, were measured immediately for 15 min. For the determination of long lived isotopes, samples that were irradiated for a long term, were measured once 3 days after the irradiation for 5 h, and then a second time, 20 days after the irradiation for 20 h. For the analysis of the gamma spectra the Genie-2000 software by Canberra was used. The elemental concentrations were calculated by means of a special software that was developed in FLNP, JINR [[58\]](#page-10-3). Certifed reference materials were used for the calculation of the elemental concentrations.

High quality certifed reference materials (SRM) were used for the quality control of the NAA results, such as peach leaves-1547, coal fy ash-1633b and 1633c, coal-1632c, Montana soil-2710, San Joaquin Soil-2709 and Calcareous soil-ERM-CC690. All the reference materials were provided by the National Institute of Standards & Technology (NIST) and the Institute for Reference Materials and Measurements (ERM). The SRMs were packed and irradiated together with the moss samples under the same conditions in each transport container. The results that were obtained were compared with the certifed values. The reference material that presented the least deviation between the measured and the certifed values of each elemental concentration was chosen.

# **Results and discussion**

The concentrations of thirty-three elements were determined using the NAA. The results of the descriptive statistical analysis of the elemental concentrations in moss samples (mean, median, min, max, and standard deviation) are presented in Table [1](#page-3-0). Spatial distributions of elemental concentrations (Fe, Ni, As, Zn, Cr) are shown in Figs. [2,](#page-4-0) [3,](#page-4-1) [4,](#page-5-0) [5](#page-5-1) and [6](#page-6-0).

This is the frst attempt of studying the atmospheric deposition of trace elements in mosses, in such a big territory in Greece. Some years earlier, in Athens, close to a highly polluted area, moss bags were used for investigating the elemental atmospheric deposition [\[59\]](#page-10-4). According to this previous study, the concentrations of the elements Al, Zn, Fe, Cr, Ni, V were higher in the sites that were closer to the industrial zone, but there was not clear association between the anthropogenic activities and the observed elements.

Furtheremore, there was one study of biomonitoring, concerning the transboundary transport of trace elements between Bulgaria and the Northeastern part of Greece [\[57](#page-10-2)]. According to this study [\[57](#page-10-2)], in the Greek territory, the relative high concentrations of the elements As, Cr, Fe, Ni and V were due to old mines, soil contamination by windblown dust and road transportation. The same study showed that, the Pb–Zn complex close to Kardzahali town (Bulgaria) was responsible for the rise of the concentrations of Cr, Fe, Ni and As in the territory of Bulgaria.

The current study verifes the results of the previous investigation regarding the pollutants' chemical composition in the region of Northeastern part of Greece, next to the Bulgarian borders. Regions that are characterized by high concentrations of Al, As, Fe and V are observed in both of the studies, with some diferences concerning the concentration levels of each element. More specifcally, the concentration of As close to 'Kerkini Lake', presents a reduction of almost 30% from the previous study, while Al remains in similar levels. In specifc regions close to Bulgarian boarders, the concentrations of Zn, Cr, Fe, Ni and V are two to three times higher than the previous observed concentrations. The reasons of this rise, ten years after the previous study, can be partially clarifed. It might be due to the windblown dust coming from manufacturing and metals industries (Fe and Zn), or from coal mining activities and even from the ophiolitic rocks (Cr and Ni) that are present in the Greek territory. However, it might be due to transboundary transport from sources in Bulgarian boarders and further investigation needs to be done.

Moreover, in the areas that are close to the borders with F.Y.R. Macedonia, the elements Fe, V, Zn, As, Co, Ni and Cr show lower concentrations than in the Kavadarci region [\[56,](#page-10-1) [60](#page-10-5)] of F.Y.R. Macedonia, which is known for the ferronickel mining and other metallurgical activities. These concentrations verify the correlation of the above elements with the ferronickel mining, and maybe indicate a possible transboundary transfer of elements from F.Y.R. Macedonia to the Greek territory.

<span id="page-3-0"></span>Diferences can be observed between the elemental concentrations of the current data and the data of the other countries that participated in the 2005/2006 and 2010/2011





<span id="page-4-0"></span>**Fig. 2** Concentrations of As in ( $\mu$ g g<sup>-1</sup>) in moss samples in Northern Greece



<span id="page-4-1"></span>**Fig. 3** Concentrations of Cr in ( $\mu$ g g<sup>-1</sup>) in moss samples in the area of Northern Greece

moss surveys (Table [2\)](#page-6-1) [[25,](#page-9-0) [50](#page-9-22)]. More specifcally, data from countries that are close to the vicinity of Greece, as well as far from Greece were chosen to be compared with the data of the current study. Greece presents lower maximum concentrations of Zn, Mn, Co than Norway, in contrast to the max elemental concentrations of Bulgaria and Switzerland. Ni max concentrations are almost in the same levels like in Bulgaria, but lower than in Norway and Albania. The Greek median value of As is similar to the Serbian one, but higher than F.Y.R. Macedonia and Albania. V and Zn are in the same levels in the Greek, Serbian and Croatian territory according to the 2005/2006 moss survey results.

In this study, there are evidence that elevated concentrations of trace elements are both observed in groundwater and mosses. More specifcally, elevated concentration of Cr was observed in mosses as well as in groundwater in the central Macedonia, nearby the Anthemountas basin [[71](#page-10-6)]. In Sarigiol basin, it has been reported the contribution of



<span id="page-5-0"></span>**Fig. 4** Concentrations of Fe in ( $\mu$ g g<sup>-1</sup>) in moss samples in the North part of Greece



<span id="page-5-1"></span>**Fig. 5** Concentrations of Ni in ( $\mu$ g g<sup>-1</sup>) in moss samples in Northern Greece

dispersed fy ash from the coal consumption to the pollution of groundwater [[72\]](#page-10-7) and soils and sediments [[73\]](#page-10-8), while Cr and Ni in mosses could also origin form the ophiolitic rocks in the study area. In literature, few studies have investigated linking between groundwater and mosses. Štechová et al. [[74\]](#page-10-9) studied the relationship of *Hamatocaulis vernicosus* with groundwater quality in Czech. Molchanov [\[75](#page-10-10)] studied the Gas exchange in sphagnum mosses nearby shallow groundwater.

There is no direct transfer of elements from the groundwater to mosses, as the last ones do not have roots, and all the nutrients and water they need come from wet and dry deposition. Trace elements in groundwater are linked mainly with the geological background. Trace elements in mosses are linked mainly with their concentrations in air. Elevated concentrations of trace elements in both mosses and in groundwater might be an indicator of pollution transfer from the air to mosses as well as from the air



<span id="page-6-0"></span>**Fig. 6** Concentrations of Zn in ( $\mu$ g g<sup>-1</sup>) in moss samples in the region of Northern Greece

<span id="page-6-1"></span>**Table 2** The concentrations of seven trace elements (As, Cr, Fe, Ni, V, Zn, Al) in moss samples (mg  $kg<sup>1</sup>$ ) from the current survey and from other European countries derived from the 2005/2006 and 2010/2011 moss survey

As Median $(min-max)$	Cr Median $(min-max)$	Fe Median $(min-max)$	Ni Median $(min-max)$	$\mathbf{V}$ Median $(min-max)$	Zn Median $(min-max)$	$\mathbf{Al}$ Median $(min-max)$	
1.44	11.5	3770	7.26	8.17	37.6	5840	Current study
$(0.52 - 17.9)$	$(2.04 - 222)$	$(1010 - 28,700)$	$(1.72 - 90.2)$	$(2.61 - 33.4)$	$(14.6 - 282)$	$(1350 - 46, 100)$	
	2.43	1399	2.99	3.88	27.9	1495	Bulgaria, 2005
-	$(0.79 - 57.8)$	$(186 - 9493)$	$(0.92 - 90)$	$(0.77 - 24.3)$	$(9.38 - 366)$	$(426 - 10, 394)$	[61]
0.63	2.06	1101	2.61	3.07	22.2	1245	Bulgaria, 2010
$(0.15 - 10.8)$	$(0.72 - 38.1)$	$(307 - 8546)$	$(0.84 - 82.1)$	$(0.96 - 22.4)$	$(8.22 - 286)$	$(402 - 8886)$	[25]
0.68	6.79	2239	5.82	6.38	35.6	3600	F.Y.R. Macedonia, 2005
$(0.18 - 4.32)$	$(2.09 - 82)$	$(999 - 8130)$	$(1.8-43.1)$	$(2.5-31.9)$	$(16.4 - 91.3)$	$(1466 - 25,860)$	$\lceil 62 \rceil$
0.48	6.46	1900	4.3	3.8	29	2400	F.Y.R. Macedonia, 2010
$(0.23-1.9)$	$(2.46-35)$	$(890 - 5400)$	$(1-55)$	$(1.5-14)$	$(13-94)$	$(1100 - 6800)$	$[63]$
0.12	0.58	273	1.24	1.40	31.4	255	Norway, 2005
$(0.004 - 4.61)$	$(0.099 - 65.5)$	$(50.4 - 9972)$	$(0.055 - 1016)$	$(0.25 - 22.1)$	$(8.04 - 694)$	$(58.3 - 12, 121)$	$[64]$
0.13	0.59	278	1.16	1.41	30.7	283	Norway, 2010
$(0.02 - 4.84)$	$(0.16 - 47.9)$	$(27-24,684)$	$(0.15 - 857)$	$(0.29 - 25.9)$	$(7.4 - 368)$	$(46 - 4581)$	$\lceil 65 \rceil$
1.41	6.44	2267	4.43	5.76	29.0	3946	Serbia, 2005
$(0.22 - 21.6)$	$(2 - 78.8)$	$(670 - 16, 100)$	$(1.7-23.8)$	$(1.94 - 32.7)$	$(13.2 - 259)$	$(117 - 31, 180)$	[66]
0.305	4.75	1618	5.85	3.51	13.8	1638	Albania, 2010
$(0.05 - 2.86)$	$(1.62 - 31.8)$	$(469 - 5488)$	$(1.56 - 131)$	$(1.15 - 16.9)$	$(1-68.1)$	$(535 - 6974)$	[67, 68]
0.37	2.8	1000	2.7	3.1	29	1350	Croatia, 2005
$(0.1-6)$	$(0.76 - 33)$	$(320-12,140)$	$(0.66 - 18)$	$(0.91 - 32)$	$(12 - 283)$	$(398 - 21,460)$	[69]
0.36	1.94	789	3.16	2.55	24.8	878	Croatia, 2010
$(0.05-1)$	$(0.41 - 8.55)$	$(85 - 4028)$	$(1.04 - 14.66)$	$(0.23 - 37.3)$	$(11.6 - 77.1)$	$(112 - 4493)$	[70]
0.15	1.2	261	1.59	0.67	31.4	$\overline{\phantom{a}}$	Switzerland, 2005
$(0.053 - 1.07)$	$(0.33 - 7.96)$	$(95.4 - 2380)$	$(0.5 - 7.77)$	$(0.21 - 3.55)$	$(10.1 - 179)$	$\overline{\phantom{m}}$	$[61]$

through soil to groundwater. Obviously, the trace elements concentrations in mosses should be compared with nearby soil concentrations, linked with the geological formations. The aforementioned constitutes the next step of this study.

Another method for identifying the most afected areas by the atmospheric deposition of elements is the calculation of the contamination factor. The contamination factor shows the pollution status of the under study area [[76–](#page-10-21)[81](#page-10-22)]. The contamination factors (CF) for each sampling site for the elements Al, As, Fe, Ni, V, Cr, Zn were calculated according to the following formula (Eq. [1](#page-7-0)).

$$
CF = \frac{Concentration\ of\ element\ (\mu g\ g^{-1})}{Background\ level\ concentration\ (\mu g\ g^{-1})}
$$
 (1)

where the background level is considered to be the concentration of each element in a "clean- virgin" area in the territory of Greece, which is as less as possible afected by human activities and other local sources, such as metal industry, mining and agricultural activities, oil combustion and road transportation.

The dispersion of contaminants in the atmosphere can occur independently of the media or the organisms on which they are deposited  $[68-70]$  $[68-70]$  $[68-70]$  $[68-70]$ . Considering this, there are diferent contamination factor scales [[77](#page-10-23), [82](#page-10-24)] and each area can be characterized by diferent contamination levels, according to the calculated CF values. According to Mouvet [[83\]](#page-11-0), who had studied the elemental concentrations in aquatic mosses, areas with  $CF < 2$  can be considered non polluted due to the natural variability that can be found into an ecosystem.

There is another scale of classifcation of the CF, which was established by Fernández et al. [\[78\]](#page-10-25) based on specifc approaches in terrestrial mosses:

- $CF < 1$ , it's a non-polluted area (natural origin of the element).
- $1 < CF < 2$ , it's a suspected area.
- $2 < CF < 3.5$  is a slightly polluted area.
- $3.5 < C$ F $< 8$  it's a moderately polluted area.
- 8 < CF < 27 it's a serious polluted area.
- CF > 27 it's an extreme polluted area.

In the present study, more than 50% of the sampling sites have  $CF < 1$ , concerning the elements Al, As, V, Fe, Zn., while this percentage drops down to 30% concerning Cr and Ni elements. This indicates that most of the above elements in mosses in these regions are not afected by any anthropogenic factor but they have natural origin. Around 30% of the areas are considered suspected areas  $(1 < CF < 2)$  for almost all the elements, while less than 15% are characterized slightly polluted areas  $(2 <$  CF  $<$  3.5). The rise of these elemental concentrations

is probably due to human activities and other local sources. Around 5% of the areas, (except of Cr and Ni that are around 20%), are described moderately polluted areas  $(3.5 < C$ F $< 8$ ) indicating that human activities have even stronger impact on these sites. These regions are mostly in the prefecture of Central Macedonia. Finally, around 5% of the regions (mostly those in the prefecture of West Macedonia)present high enough concentrations of Cr and Ni, in order to be marked as serious polluted areas  $(8 < C$ F $<$  27). The regions where Cr and Ni present high concentrations are in accordance with those regions whose CF>1, indicating that they might be connected not only with ophiolitic rocks (geogenic source of origin) but also with human activities such as coal mining and industry.

<span id="page-7-0"></span>The contamination factor was calculated in other studies too for the characterization of the areas. For example, Maxhumi et al. [\[84](#page-11-1)] mentions that in Kosovo the elements Fe, Ni and Zn are related with the frst two categories of the contamination factor scale, indicating non polluted areas, while Cr is associated with the third and fourth level of contamination factor, the so called slightly and moderately polluted areas. According to Jiang et al. [[81](#page-10-22)], in China, Cr is connected with regions that have experienced moderate contamination, while As, Ni, V and Zn characterize areas with low contamination level (first category of the CF scale). In Galicia (northwest Spain), according to Fernández and Carballeira [\[77](#page-10-23)], most of the areas concerning Al, Cr, Fe, Ni and Zn belong to the second category of CF, while there is a 2% of areas with serious contamination level associated with Al and Fe elements. In Albania, Qarri et al. [[67\]](#page-10-17) reported that Zn is correlated with the frst two levels of CF scale, while As is characterizing areas that are slightly polluted. The CF of the elements Fe, Ni, V and Al describe areas that are moderately and serious polluted in Albanian territory.

# **Conclusions**

Moss biomonitoring is a really efficient and economical technique for the determination of the atmospheric deposition of trace elements. Ninety-fve samples of mosses were collected from the vicinity of Northern Greece. They were analyzed using NAA and the concentrations of 33 elements were determined. A detailed spatial distribution of the elements was achieved.

The values of the present study were compared with the results of the previous EU moss surveys, and more precisely with those of the 2005/2006 and 2010/2011 moss survey. The regions with the higher concentrations of Al, As, Fe and V of this study are in accordance with the data of the previous study that was conducted in the same region by Yurukova et al. [[57](#page-10-2)]. In specific sites close to Bulgarian boarders, the elements Zn, Cr, Fe, Ni and V are found in higher concentrations than in the previous study. The reasons of this rise, has not be defned yet. It might be due to transboundary transport or due to local sources in the Greek territory. Further research should be done for the defnition of the sources that lead to this rise.

The relative higher concentrations of Cr and Ni in the territory of Greece are probably due to geogenic sources (ophiolitic rocks), and maybe due to coal mining activities. Al, Fe and Zn might be connected with metal and manufacturing activities. Future investigation about the determination of potential sources of the studied elements in mosses is required. Also the case of transboundary transfer of metals should be examined more carefully.

Elevated concentrations of trace elements are co observed in groundwater and mosses, indicating the pollution transfer from the air to mosses and from the air through soil to groundwater.The elemental concentrations in mosses should be compared with nearby soil concentrations, linked with the geological formations.Α further and more detailed analysis is necessary to understand potential process between mosses and groundwater.

The contamination profle of each area is also formed using the contamination factor. The CF results indicate that the elements Al, As, Fe, V, Zn are associated with the frst category of the contamination scale—the unpolluted areas, revealing a pretty low pollution level of atmospheric deposition of the above elements. The elements Cr and Ni are mostly related with the suspected areas—second category of the CF, emphasizing on the impact of human activities on their atmospheric deposition.

Finally, this study provides a baseline dataset for future studies, in order to evaluate the air quality of Greece and compare with data from other neighboring countries.

**Acknowledgements** This research has been fnancially supported by General Secretariat for Research and Technology (GSRT) and the Hellenic Foundation for Research and Innovation (HFRI) (Scholarship Code: Contr. No 131183/I2, Code 569, 1st action 2016). Also, part of the sampling was fnancially supported by the West Macedonia Development Company (ANKO) SA.

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