



Investigations of the Atmospheric Deposition of Major and Trace Elements in Western Tajikistan by Using the *Hylocomium splendens* Moss as Bioindicators

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Abstract

The study was performed in a mountainous area of approximately 7000 sq. km of Western Tajikistan, i.e., Turkestan, Zeravshan, Hissar, and Karateghin ridges that are characterized by complex geological settings. Moss biomonitoring was used to assess the concentration level of trace and major elements in atmospheric deposition of the study area. *Hylocomium splendens* (Hedw.) Schimp. moss was used as biomonitor in this study. 43 major and trace-elements were determined by Epithermal Neutron Activation (ENAA) and Atomic Absorption Spectrometry (AAS). GIS maps of the 43 elements showed that the distribution of Mo, Cd, REE, Th, and U could be most probably associated with the Odjuk pegmatite field. Zr, Hf, and W contents are significantly increased in the vicinity of the Sarbo River washout while Cr, Co, Ni, and As showed a maximum content near Kanchoch gold field. The global pollution index based on the local content of presumed pollutants Cr, Ni, Cu, Zn, As, Cd, Sb, and Pb in some places exceeded the threshold limits for a pristine, unpolluted environment. At the same time, the distribution of incompatible Sc, La, Yb, and Th suggested for the airborne material deposited on mosses a continental component, enriched in few places in felsic components.

Bryophyte mosses are widely used in environmental study as trace metals bioindicators of atmospheric deposition; because of the lack of a root system, they take up the pollutants directly from the atmosphere. For this reason, mosses

represent unique natural bioindicators (biomarkers) that can react to environmental pollution, providing relevant data on both spatial distribution and temporal evolution on areas encompassing hundred of thousands of square kilometers within various environments (Gilbert 1968; Kelly and Whitton 1989; McLean and Jones 1975; Berg et al. 1995; Vanderpoorten 1999; Dolegowska et al. 2013; Harmens et al. 2007, 2010, 2015; Culicov et al. 2016; Nickel et al. 2018).

Tajikistan is a landlocked, mountainous country covered by the Pamir Mountains; more than 50% of its area is at altitudes higher than 3000 m above sea level. The only lower land areas are in the southern part along the Kofarnihon and Vakhsh River valleys and in the north partially covering the Fergana valley.

It should be remarked that three northernmost mountains ranges with a length of approximately 200 km: Hissar, Zeravshan, and Turkestan, which average height is more than 3500 m are virtually oriented from the east to the west. This makes them an efficient barrier for winds (Middleton 1986), so that the aerosols transported by them over a large area could be trapped and deposited onto mosses.

To determine the content of elements in mosses, Instrumental Neutron Activation Analysis (INAA) and Atomic

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Absorption Spectrometry (AAS) were used. AAS was selected only for Cu, Cd, and Pb, elements for which INAA cannot assure enough accuracy and precision. Together, these methods allow determining the content of more than 40 elements in different species of mosses and lichens as literature data confirm (Herpin et al. 2001; Spiric et al. 2013; Stihi et al. 2017).

The first mosses biomonitoring in Tajikistan was performed between 2016 and 2017 when the country joined the European Moss Survey in the framework of the International Cooperative Program on Effects of Air Pollution on Natural Vegetation and Crops UNECE ICP Vegetation Program (Harmens 2018; Frontasyeva et al. 2019; ICP 2019). Data thus obtained allowed to consider both spatial and temporal trends in the deposition of heavy metals as well as to evidence the anomalous zones with high depositions of aerosols coming from both distant regions and local sources.

Moreover, the same set of data could be used not only to assess the level of anthropogenic influence on investigated area but also could furnish confident data concerning the source of aeolian sedimentary material retained by mosses (Marinova et al. 2010). For a better quality of this study, we have chosen only the *Hylocomium splendens* (Hedw.) Schimp mosses due to their ability to retain a great variety of elements as well as to their relative abundance in mountainous Tajikistan. It is worth mentioning that among neighboring countries, similar studies using mosses as biomonitors were performed only in Kazakhstan (Omarova et al. 2018). The final results of this study will be further presented and discussed.

Materials and Methods

Investigated Area

The investigated area, i.e., the Western Tajikistan that covers a part of Hercynian geosynclinals-folded Southern Tien Shan Mountains consists mainly of Upper Paleozoic covered in some places with local the Precambrian, Paleozoic, Mesozoic, and Cenozoic sediments. In the Hissar range, the intrusive rocks are most abundant (approximately 50% of its territory), whereas in the neighboring Zeravshan and Hissar ridges they occupy less than 12% of surface, by forming the so-called Hissar pluton. Here, they are mainly developed as basic and ultra-basic intrusions by forming a dyke sector. Adjacent to this lays the Garm median masses consisting of Precambrian gneisses and Middle Devonian gabbro, pyroxenite, peridotite, cordierite, as well as granite with garnets. In some places, the presence of stratified volcanic formations consisting of acid rocks can be associated with Paleozoic (lower, middle, and upper) local formations (CAG 2019).

In Southern Hissar Mountains, the iron pyrite mineralization associated with early geosynclinals magmatites have a significant potential for Ni–Co–Pt associations (Levikov 1980). Due to the granitoids inversion, the mineralization in this region is well developed and manifests itself by an intricate association of cassiterite and wolframite—scheelite, together with pegmatite, greisen, and hydrothermal biotite porphyry-type granitoids from the Late Carboniferous. A skarn iron—rare earth elements with granitoids with enhanced core of the Middle Carboniferous—also was identified. Besides these, on the Early Permian multipartite trachyandesite—dacite—liparite and subvolcanic porphyry is associated with multiple polymetallic formation rich in copper, molybdenum, antimony, and mercury mineralization (CAG 2019).

Therefore, it should be remarked that through the entire Hissar ridge passes antimony–mercury geochemical belt. It is 300-km long and 30-km wide. It starts in the west in Panjikent and stretches to the Alai Valley in Kyrgyzstan and Tajikistan (Bakhtdavlatov 2017). The most common elements in belt are As, Ba, and Sb of which presence should be reflected by the elemental content of various bioindicators found in this region, inclusive the *Hylocomium* mosses.

Sampling and Samples Preparation

For this study, we have collected only fresh specimens of *H. splendens*. To avoid any local contamination, all sampling points were located at least 300–700 m from main highways and populated areas and 200–400 m from country roads (Fig. 1) (Steinnes et al. 1992). In each sampling point, which covered a 50 × 50-m area, 5–10 mosses specimens were collected and blended into one average sample. After removal of alien material, the first three segments of plants were selected, and packed in plastic bags and transported to the Dushanbe Institute of Water Problem where all samples were dried at 35–40 °C at constant weight (Steinnes et al. 1992). Finally, the dried material was sent to the Joint Institute for Nuclear Researches (JINR), Dubna, Russian Federation for INAA and AAS determinations where all samples were previously ground by an agate mortar to a fine powder.

Analytical Measurements

INAA determinations were performed at the JINR Frank Laboratory of Neutron Physics, using the pulsed nuclear reactor IBR-2 for both thermal and epithermal neutrons (Frontasyeva et al. 2005; Barandovski et al. 2008; Marinova et al. 2010; Frontasyeva 2011; Allajbeu et al. 2016). For analysis, powdered samples weighing 0.3 g were packed in polyethylene bags for the determination of short-lived isotopes and in aluminum foil for long-lived ones. This procedure allowed to determine the content of 40 major and trace



Fig. 1 Location of collecting points on investigated area with respect to Tajikistan general map (inset)

elements: Na, Mg, Al, Cl, K, Sc, Ca, Ti, Cr, V, Mn, Ni, Fe, Co, Zn, Se, As, Br, Sr, Rb, Zr, Mo, Sb, I, Ba, Cs, La, Ce, Nd, Eu, Gd, Sm, Tb, Yb, Tm, Hf, Ta, W, Th, and U. More details concerning the sample preparation and INAA measurements can be found in (Culicov et al. 2016; Pavlov et al. 2016).

Complementary to INAA, the content of Cu, Cd, and Pb were measured in the same moss specimens by using a ThermoFisher Scientific™ iCe 3000 series Atomic Absorption Spectrometer provided with an electrothermal atomization. The calibration solutions were prepared from a 1 g/L stock solution: AAS standard solution (Merck, Germany). In this case, approximately 0.3 g of dried moss sample was placed in a Teflon vessel and treated with 5 mL of concentrated nitric acid (HNO₃) and 1 mL of hydrogen peroxide (H₂O₂). Then, samples were introduced in a microwave digestion system (Mars; CEM, USA) for complete moss digestion. Digestion was performed in two steps: i) ramp: temperature 160 °C, time 15 min, power 400 W, and pressure 20 bar; ii) hold: temperature 160 °C, hold time 10 min, power 400 W, and pressure 20 bar. Digests were quantitatively transferred

to 50-mL calibrated flasks and made up to the volume with bi-distilled water. All of the reagents used for this study were of analytical grade: nitric acid trace pure (Merck, Germany); hydrogen peroxide, p.a. (Merck, Germany); and bi-distilled water.

Quality Assurance

In the case of both INAA and AAS, a special attention has been paid to the quality assurance of experimental determinations. The analytical performance of the method was checked by the analyses of a set of appropriate certified standard reference materials (SRM), i.e., National Institute for Standard and Technology (NIST) 1573a (Tomato leaves), 1575a (Pine needles), 1547 (Peach leaves), 1632c (Coal), 1570a (Spinach leaves), 2709 (Saint Joaquin soil), International Atomic Energy Agency (IAEA) 433 (Marine sediment), as well as Food and Agriculture Organization of the United Nations (FAO) 690 cc (Calcareous soil).

The software “Concentration,” developed in FLNP JINR (Pavlov et al. 2016), was used to create the so-called Group Standard Sample (GSS) from all above-mentioned reference materials, which, irradiated together, were used to calculate the content of approximately 40 elements in the analyzed samples with a maximum accuracy. Furthermore, the GSS together with the Concentration software was used to check the quality of analysis by determining the content of the elements of interest. This procedure, applied to certified SRM, allowed to compare the obtained values with the certified ones (Zinicovscaia et al. 2018). In this way, the accuracy expressed as the percentage standard deviation of the mean of three determinations, was below 10% for all elements. In the same conditions, the precision defined as the percentage deviations from the certified values varied between 3 and 10% (Table 2, Supplementary Material).

Results and Discussion

The problem of a correct estimation of the anthropogenic influence onto a given biota is of paramount importance, especially when it needs to discriminate between the natural and anthropogenic processes (Tarzia et al. 2002).

On the other hand, as mentioned, the main source of mosses contamination is represented by the atmospheric transport as dry and wet precipitation deposition. For this reason, the anthropogenic contamination could have either local character or the pollutants could be transported by local atmospheric circulation from distant sources.

We have represented the spatial distribution of the content of each of all 43 elements in mosses samples over the entire investigated area (Fig. 1, Supplementary Material). This allowed us to evidence more locations associated either with anthropogenic activity or agglomeration of rocks rich in REE, Th, or U, such as pegmatite (see further discussion).

It is worth mentioning that among the 43 analyzed elements, only few of them such as V, Cr, Co, Cu, Zn, As, Sb, Cd, and Pb could be considered as having beside natural and an anthropogenic origin while the other one mainly could come from rock weathering, and in this last case, could be used to trace their origin. It is the case of Sc, Zr, REE, Hf, and Th. For this reason, we have concentrated our analysis mainly to these elements.

For a concise description, in Table 1 we have showed for each considered element the average content \pm standard deviation, the total experimental uncertainty estimated by composing the statistic error in determining the peak areas with the certified material uncertainty, as well as the

Table 1 Average values \pm standard deviation (in mg/kg), total uncertainty (TU) (in %), and the corresponding values of reference plant (RP) (in mg/kg) (Markert et al. 2015) and the Contamination Factor (CF) (Fernández et al. 2000)

Element	Average \pm SD	TU	RP	CF	Element	Average \pm SD	TU	RP	CF
Na	3100 \pm 2640	2.6–5.2	200	20 \pm 18	Mo	1.5 \pm 2.2	9.1–5.2	0.5	3.2 \pm 4.7
Mg	11,160 \pm 4510	3.3–6.9	150	54 \pm 22	Cd	0.6 \pm 0.5		0.05	12 \pm 11
Al	17,330 \pm 9215	0.8–5.2	80	213 \pm 119	Sb	2.8 \pm 5	6.5–2.8	0.1	31 \pm 551
Cl	1402 \pm 1400	1.7–3.6	20,000	0.7 \pm 0.7	I	7.2 \pm 8.3	9.9–15.3	3	2.5 \pm 2.9
K	12,045 \pm 6320	1.7–3.4	19,000	6.5 \pm 3.5	Cs	2.9 \pm 4.6	1.9–4.2	0.2	15 \pm 24
Ca	41,410 \pm 44,480	1.3–2.9	10,000	4.2 \pm 4.7	Ba	195 \pm 150	3.2–6.8	40	4.8 \pm 3.7
Sc	3.6 \pm 2.1	1.2–3.1	0.02	167 \pm 94	La	12 \pm 12.9	7.5–11.3	0.2	62 \pm 67
Ti	1120 \pm 640	6.2–9.1	0.05	220 \pm 134	Ce	21 \pm 20	7.2–12.1	0.5	43 \pm 41
V	29 \pm 13	0.4–1.3	0.5	55 \pm 26	Nd	9.1 \pm 9.2	9.9–15.5	0.2	47 \pm 48
Cr	26 \pm 17	1.1–3.1	1.5	16 \pm 9	Sm	1.6 \pm 1.6	11.5–20.5	0.04	41 \pm 42
Mn	352 \pm 351	2.5–4.2	200	1.4 \pm 1.1	Eu	0.4 \pm 0.3	11.5–20.5	0.008	56 \pm 45
Fe	9550 \pm 5750	1.1–2.3	150	59 \pm 36	Gd	2.3 \pm 4.7	12.5–25.9	0.04	59 \pm 122
Co	4.4 \pm 2.9	3.3–6.6	0.2	19 \pm 10	Tb	0.2 \pm 0.2	14.5–29.5	0.008	29 \pm 27
Ni	14 \pm 9	4.4–8.9	1.5	7.9 \pm 3.4	Tm	0.2 \pm 0.1	12.5–22.6	0.004	38 \pm 35
Cu	19 \pm 14		10	1.6 \pm 0.9	Yb	0.7 \pm 0.6	12.1–24.5	0.02	36 \pm 29
Zn	55 \pm 23	2.2–5.3	50	1 \pm 0.4	Hf	1.5 \pm 1.5	1.8–5.1	0.05	30 \pm 31
As	10.3 \pm 11.2	1.3–5.3	0.1	109 \pm 117	Ta	0.3 \pm 0.4	10.2–20.1	0.001	309 \pm 386
Se	2.2 \pm 2.1	5.6–10.1	0.02	101 \pm 105	W	1.3 \pm 1.7	5.5–9.8	0.2	7.1 \pm 8.9
Br	79 \pm 166	2.2–5.3	4	21 \pm 43	Pb	7.4 \pm 4.3		1	7.6 \pm 4.5
Rb	35 \pm 32	1.8–8.7	50	0.7 \pm 0.7	Th	5.4 \pm 9.5	2.2–5.6	0.005	1127 \pm 1987
Sr	162 \pm 218	1.5–4.3	40	3 \pm 4.4	U	10.5 \pm 38.3	1.2–3.4	0.01	1137 \pm 4021
Zr	69 \pm 73	11.2–15.3	0.1	703 \pm 757	–	–		–	–

corresponding content of Reference Plant (RP) (Markert et al. 2015). It should be remarked that in the absence of any national regulations concerning the normal as well as the alarm threshold level of heavy element contaminants in mosses, we have used the RP as the most appropriate reference system free of anthropogenic contamination. We came to this conclusions by comparing the content of certain elements reported in more pristine regions of the world such as Livingstone Island of Antarctica (Culicov et al. 2016) or Canadian High Arctic (Wilkie and La Farge 2011) are of the same order of magnitude as RP.

Therefore, we have considered the RP to assess the environmental situation in the Western Tajikistan as reflected by the *Hylocomium splendens* mosses. Besides it, we have also taken into account for comparison the Upper Continental Crust (Rudnick and Gao 2003), especially in the case of incompatible and insoluble elements, such as Sc, REE, or Th.

According to the data showed in Table 1, the content of enzymatic V, Cr, Fe, Co, Ni, Cu, Zn, and Mo (Markert et al. 2015), as well as of those elements considered pollutants, such as Cr, Co, Zn, Cu, As, Sb, Cd, and Pb, are significantly greater than corresponding RP contents. To these, we could add Th and U of which average content of 6.7 and 16.4 mg/kg is few order of magnitude greater than the corresponding RP, and in the case of Odjuck pegmatite field, even greater than the UCC ones.

To describe in a unitary manner the peculiarities of elements distributions, we have calculated, for each element and for each location the corresponding contamination factor CF (Fernández et al. 2000) defined as the ratio between the content of considered element c_x and the content of the same element c_R in a pristine, uncontaminated medium. In this case, taking into account the total lack of a root system, we have considered, as mentioned before, the RP (Markert et al. 2015), so that the CF an element i has the expression:

$$CF_i = c_{i|mass} / c_{i|RP} \quad (1)$$

where $c_{i|mass}$ represents the content of element x in the Tajikistan mosses and $c_{i|RP}$ represents the content of the same element in RP (Markert et al. 2015) considered an average, universal, and uncontaminated medium.

The average values of the CF for all 43 elements also are illustrated in Table 1. The CF was further used to represent the spatial distribution of all 43 considered elements over the entire investigated area. The resulted 43 maps, presented in Fig. 1, Supplementary Material allowed us to infer the possible sources of natural and anthropogenic contamination. Hence, a careful analysis of the maps depicted in Fig. 1, Supplementary Material showed that, except for major rock-forming elements Na, Mg, and Al common to all rocks, every other elements showed one or

in a few cases two or three places where their content in *Hylocomium* mosses presents a maximum. On the other hand, the higher value of Al, Ti, Mn, and Fe in mosses collected from all sampling points most probably suggest a natural, nonanthropogenic origin, because they cover a large mountainous area, with a restrained industrial activity, as mentioned before.

In the case of the other elements, it should be remarked that their maxima were located around a coal intermediary deposit near Maykhura (S2 on Fig. 1), Yagnob near to the Anzob mining and processing plant (S23 on Fig. 1), which can be relate to the complex mercury-antimony ores of the Dzhizhikrut deposit. Also, golden mines in the vicinity of Kanchoch (S8, S9 on Fig. 1), Kumarg (S23 on Fig. 1, Supplementary Material) and Karatag (S15, on Fig. 1, Supplementary Material) as well as the massive Odjuk pegmatite deposit (S5 and S6 on Fig. 1, Supplementary Material).

At a careful examination of the CF shown in Table 1, it can be remarked that with few exception, such as Cl, Zn, or Rb, the CF all other investigated elements were significantly higher than the unit, reaching in the case of Th and U values greater than 1000. At the same time, for almost all elements, the CF showed a great variability reaching again a maximum value of approximately 350% in the case of U.

The fact that in the case of all investigated elements, regardless of whether they are enzymatic, natural, or have a presumed anthropogenic origin, the CF presented higher value suggests rather a significant influence of the local environment, with as mentioned, a higher degree of mineralization.

Moreover, for a more global characterization of a possible anthropogenic contamination as reflected by the content of Cr, Co, Zn, Cu, As, Sb, Cd, and Pb, we have calculated for each collecting point a more general Pollution Loading Index (PLI) (Tomlinson et al. 1980) following the equation:

$$PLI = \sqrt[n]{\prod_i^n CF_i} \quad (2)$$

where index i designate the above mentioned elements.

It is worth mentioning that the PLI can be used to characterize the spatial distribution of contamination, by taking into account that a $PLI < 1$ suggests no contamination, a $PLI = 1$ represents the lower threshold of contamination while a $PLI > 1$ signifies the presence of contamination so that greater than PLI, higher the contamination.

Accordingly, the PLI values showed in Table 1 suggest a significant degree of contamination over the entire investigated area, of which maxima correspond, as mentioned above, to few industrial objectives presented, i.e.,

Maykhura coal intermediary deposit, Yagnob antimony ore processing plant, Kanchoch and Kumarg golden mines as well as the Karatag, not far away from the aluminum refractory located in the Tursunzoda region.

A more suggestive representation of a possible anthropogenic influence on the investigated area is given by the PLI spatial distribution illustrated by Fig. 2. Indeed, on this figure, the PLI showed few maxima localized around the industrial objectives mentioned above, which also could explain the great dispersion of the PLI as evidenced in Table 1.

Not all elements presented on *Hylocomium* mosses could be connected with an anthropogenic activity. Elements, such as Sc, Zr, REE, Th, or U, are normal components of rocks and not related to any human activity, so that their presence could be used to assess the origin of material accumulated on mosses.

Accordingly, the Sc–La–Th discriminating ternary diagram illustrated in Fig. 3a showed with clarity that all mosses samples, with the exception of those collected in the vicinity of Odjuk pegmatite field, are grouped around the Upper Continental Crust (UCC) (Rudnick and Gao 2003),

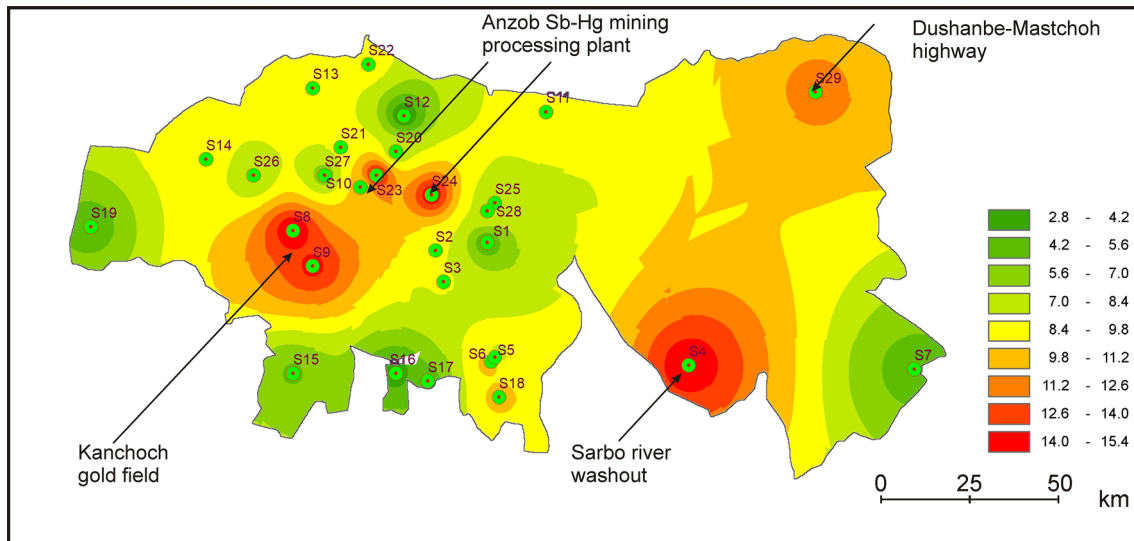


Fig. 2 Spatial distribution map of the Pollution Load Index (PLI) in investigated area

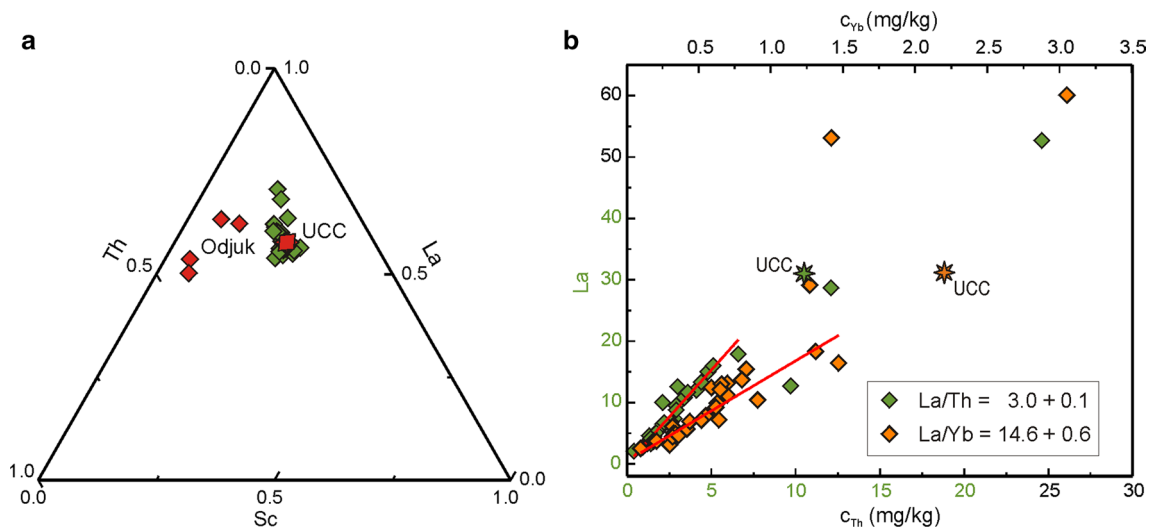


Fig. 3 The Sc–La–Th discriminating ternary diagram (a) and the bi-plots illustrating the correlation Th–La and Yb–La for all locations (b). The sampling points corresponding to Odjuk pegmatite field are illustrated in orange on both plots. For comparison, the data corre-

sponding to UCC (Rudnick and Gao 2003) are shown too. Due to the fact that the La, Yb, and Th experimental data are not normal distributed, we have used the nonparametric Spearman's correlation coefficient. All correlations were significant at $p < 0.01$

suggesting a terigenous component of the material deposited on mosses. This hypothesis is sustained also by the La versus Th and La versus Yb biplots (Fig. 3b), which showed, for all samples with the exception of the Odjuk vicinity ones, a perfect linear correlation characterized by a La/Th ratio of 3.2 ± 0.6 and La/Yb of 14.7 ± 3 very close to the UCC value of 2.95 and 15.5 respectively.

Concerning the mosses samples collected near the Odjuk pegmatite field, their position on the Sc–La–Th diagram (Fig. 3a) suggest a felsic, more acid origin of deposited material, in good concordance with the pegmatite mineralogy, very similar to granite (Linnen et al. 2012).

The Kazakhstan is the nearest to Tajikistan country where the results of a similar moss biomonitoring was reported by Omarova et al. (2018). By comparing our results with the results of Omarova et al. (2018), we have remarked content of almost all elements in Tajikistan exceeds the Kazakhstan ones. On the other hand, the Kazakhstan is a more industrialized country than Tajikistan (World Bank 2018), but, at the same time, our investigation was restrained to a small mountainous area of approximately 7000 km² representing only 4.9% of the Tajikistan entire surface.

In these conditions, one of the most plausible explanation of our finding consists of the existence of local anthropogenic sources of contaminants mainly in the vicinity of industrial objects superposed on a high background consisting of the same elements but due to the complex mineralogical structure of investigated area. Such kind of investigation worth to be extended on the entire Tajikistan territory given its remarkable geodiversity.

Conclusions

Epithermal Neutron Activation Analysis and Inductively Coupled Plasma Optical Absorption Spectrometry were used to determine the content 43 major and trace elements in 29 samples of *H. splendens* mosses collected from the Western Tajikistan covering an area of approximately 7000 km² with a few industrial objectives, which included a Sb ore enrichment plant, two gold mines, and a coal intermediary deposit.

The Contamination Factor (CF) and Pollution Loading Index (PLI) were used to quantify the level of contamination. Despite a reduced industrial activity, both indices showed increased values with reference to the Reference Plant, considered as an uncontaminated environment. Compared with neighboring Kazakhstan moss biomonitoring, and despite a significant lower industrial activity, the Tajikistan one showed a significant increased content of almost all elements.

Although a 2D representation of the PLI showed increased values in the vicinity of some industrial objectives, the higher values of both CF and PLI could not be attributed

only to an anthropogenic activity, but most probably they were influenced by the geochemistry of the investigated area rich in some places in felsic rocks, as in the case of Odjuk pegmatite deposit.

At the same time, the distribution of some incompatible elements, Sc, La, Yb, and Th, pointed towards, with some exceptions, to a dominant continental origin of the airborne material deposited on mosses.

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Data Repository A complete set of experimental data can be found at <http://dx.doi.org/10.17632/myvr4dpvk7.3>.

Compliance with Ethical Standards

Conflict of interest The authors declare that they have no conflict of interest.

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