

ORIGIN OF NATURAL ARSENIC AND ANTIMONY CONTENTS IN THE PERMIAN TO LOWER TRIASSIC SILICICLASTIC ROCKS OF THE WESTERN MECSEK MOUNTAINS, SW HUNGARY

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Abstract: Arsenic and antimony has been determined by neutron activation analysis in a large number (109) of siliciclastic sedimentary rock samples from the western part of the Mecsek Mountains (Tisia Block), Hungary. Additionally, a total of 84 samples were selected for XRD mineralogy. The goal of this work is to determine the distribution and origin of the local natural background contents of these toxic elements. Concentrations of As and Sb in different Lower Permian to Lower Triassic lithostratigraphic units are higher than the normal global ranges represented by the Upper Continental Crust composition (1.5 and 0.2 mg/kg, respectively). In the studied mudrock samples, the median values for As and Sb range from 3.5 to 30.5 mg/kg, and from 1.1 to 2.0 mg/kg, respectively. In the sandstone samples, the median As contents range from 2.3 to 20.5 mg/kg, while the values for Sb range from 0.4 to 0.9 mg/kg. The highest metalloid contents were found in the Upper Permian Cserdi and Boda formations, which could be only of local importance as a source of As in the contamination process of soils and natural waters. The relatively enhanced metalloid content of these rocks could be related to hydrothermal activity in the region. Another possibility is that the source for background levels of As and Sb in these siliciclastic rocks could be the metalloid-bearing Fe oxides and oxyhydroxides coating mineral grains.

Keywords: siliciclastic rocks, environmental analysis, background level, metalloids, origin, sulphide minerals

1. INTRODUCTION

Arsenic (As) and antimony (Sb) are ubiquitously present in the Earth's crust, with average concentrations of 1.5 and 0.2 mg/kg for Upper Continental Crust (UCC), respectively (McLennan, 2001). Because of the high As toxicity, numerous publications have been dedicated to its chemical behavior in natural systems (Matschullat, 2000; Navas & Machín, 2002; Smedley & Kinniburgh, 2002; Garcia-Sanchez & Alvarez-Ayuso, 2003; Smedley et al., 2005) and in acid mine drainage (Smedley, 2008; Garcia-Sanchez et al., 2010; Nagy-Korodi et al., 2011). Antimony is a Group V element in the Periodic table and it is similar to As in many aspects of chemical behaviour and toxicity to animals (Filella et al., 2002; Ashley et al., 2003; Filella et al., 2007). Their contamination of soils and surface waters or groundwater represents a great threat to

human health due to their high potential to enter into the food chain (Garcia-Sanchez & Alvarez-Ayuso, 2003; Filella et al., 2007; Garcia-Sanchez et al., 2010). Consequently, the presence of elevated levels of these metalloids in soils and rocks may be an issue of environmental concern.

The main natural occurrences of As and Sb are ore deposits with sulfide minerals, and some rock types such as shales, sandstones and phosphate rocks, which can contain up to some hundreds of mg/kg (Filella et al., 2002; Ashley et al., 2003; Garcia-Sanchez & Alvarez-Ayuso, 2003; Smedley, 2008; Garcia-Sanchez et al., 2010). The nature of the soil parent material appears to be the main factor determining the As and Sb contents in soils, although due to their low supergene mobility, the soils are slightly enriched in As and Sb compared with their parent rocks (Filella et al., 2002; Navas & Machín, 2002; Garcia-Sanchez & Alvarez-Ayuso,

2003). Therefore, it could be interesting to know the natural contents and distributions of these toxic metalloids in potential parent rocks (Filella et al., 2002; Smedley, 2008; Varga et al., 2012).

On the other hand, legal regulation of heavy metal and metalloid contents is an important issue in many countries (e.g., Navas & Machín, 2002; Smedley, 2008). In the last decade, consequently, a countrywide geochemical map of surface and near surface rocks was prepared by the Geological and Geophysical Institute of Hungary (former Geological Institute of Hungary; Fügedi, 2004; Gondi et al., 2004; Fügedi et al., 2007). Additionally, the geochemical background levels of different elements, including metalloids, and the extent of accumulation of toxic elements were also established (Fügedi, 2004; Fügedi et al., 2007). Nevertheless in the Western Mecsek Mountains and its surroundings (SW Hungary), very little is known about the distribution and origin of such elements in the natural, non-polluted soils, sediments, and rocks (Varga et al., 2012). Therefore, as a first step to determine the reference levels of toxic elements, it is necessary to know their contents in soil parent rocks. For this reason, this study was aimed at providing a contribution to a database on the metalloid status (natural background) of the Lower Permian to Lower Triassic sedimentary rock units in this region. Another objective of the present study is to discuss the origin of natural As and Sb contents in a local to regional framework. It is beyond the scope of this manuscript to characterize the major and trace element geochemistry of these formations. This is addressed in detail in previous separate papers (e.g., Varga et al., 2005, 2006, 2007; Varga & Raucsik 2009).

2. MATERIALS AND METHODS

The western part of the Mecsek Mountains (Tisia Block) is located in SW Hungary in the drainage basin of the Danube River, and covers an area about of 300 km². Outcropping rocks are mostly Paleozoic and Mesozoic siliciclastic sedimentary rocks consisting mainly of claystone, siltstone, sandstone and conglomerate. In basinal areas, these rocks are overlain by a thick sequence of sediments of Miocene to Quaternary age, which constitutes the most important aquifer systems in the studied area.

To eliminate an influence of outcrop weathering, a total of 109 mudrock and sandstone core samples from different exploration boreholes penetrating the Lower Permian–Lower Triassic siliciclastic succession were collected for the present study. Lithostratigraphic units studied in this paper are the Lower Permian Korpád Sandstone (KoF), Upper Permian Cserdi Conglomerate (CF), Upper

Permian Boda Claystone (BF), Upper Permian to Lowermost Triassic Kővágószőlős Sandstone (KvF), and Lower Triassic Jakabhegy Sandstone (JF) formations. The KoF, CF, BF and KvF were deposited in continental redbed environments (fluvial systems including arid-zone alluvial-fan sequences and playa deposits), whereas the rocks of the JF represent a fluvial to deltaic sequence (Haas et al., 1999; Fülöp, 1994; Barabás & Barabásné Stuhl, 1998; Bércziné Makk et al., 2004).

In a large number (109) of siliciclastic sedimentary rock samples chemical analysis of arsenic and antimony were performed by neutron activation analysis (NAA) at the ACME Analytical Laboratories Ltd. (Vancouver, British Columbia, Canada). Detection limits for As and Sb are 0.5 and 0.1 mg/kg, respectively. As quality control, duplicate analyses were performed on selected samples.

A total of eighty-four samples (N=54 for sandstones and N=30 for mudrocks) were selected for mineralogy. The mineralogical analyses of the whole rocks and their clay-sized fraction were performed at the Department of Earth and Environmental Sciences of University of Veszprém (Hungary) by X-ray diffraction (XRD) using a Philips PW 1710 diffractometer, Cu-K α radiation and diffracted-beam graphite single crystal monochromator. Rock samples were disaggregated under standard conditions using jaw crusher. Random powder of the bulk sample was used for characterization of the whole rock mineralogy. Prior the clay fraction (<2 μ m) separation, the samples were treated for removing the easily soluble carbonates by using 10% acetic acid and properly dispersed by using ultrasonic deflocculation. The clay fraction was separated from the stabilised aqueous suspension and oriented specimens were prepared by smearing a paste of <2 μ m fraction onto a glass slide to minimize size fractionation of the clay particles.

3. RESULTS AND DISCUSSION

3.1. Total arsenic and antimony concentrations in the studied samples

Summary statistics for the analysed elements in the formations studied are presented in table 1 for mudrock samples and in table 2 for sandstone samples. The median is used as the preferred statistical value than the arithmetic mean because it is not dependent on outliers.

Arsenic and antimony show a high variability in most of the sedimentary rock units studied. The median As contents calculated for each mudrock

units, ranges between 3.5 mg/kg (KoF) and 30.5 mg/kg (BF). High As values occur in the CF, BF and JF samples. However, the median value for JF represents the analysis of a minor number of samples. Median As contents for sandstone units vary from 2.3 to 20.5 mg/kg in the KoF and CF samples, respectively. Other relative high values (around 8 mg/kg) are found in the BF sandstone samples. Median Sb contents for mudrocks vary between 1.1 mg/kg (JF) and 2.0 mg/kg found in the BF samples. The same values for the sandstone units vary from 0.4 to 0.9 mg/kg in the JF and BF samples, respectively.

3.2. Mineralogy

Mineralogical composition of the examined Lower Permian to Lower Triassic sandstone and mudrock samples is listed in table 3.

KoF samples consist dominantly of quartz, albite, illite±muscovite, chlorite, hematite, and carbonate minerals (calcite and/or dolomite). Minor proportions of kaolinite, siderite, K-feldspar and smectite also occur. Based on XRD, pyrite is also present in some sandstone samples.

CF samples consist mostly of quartz, albite, K-feldspar, illite±muscovite and hematite. Additionally, dolomite, smectite, kaolinite and chlorite also occur. Anhydrite is present only in a single sandstone sample.

BF samples are predominantly composed of quartz, albite, illite±muscovite, hematite and

carbonates. Additionally, smectite, chlorite, rare K-feldspar and kaolinite also occur. The claystone samples generally have higher illite±muscovite, hematite and chlorite contents relative to the sandstone samples. In contrast, the BF rock types with coarser grain-size have higher albite, quartz and carbonate contents relative to the claystone samples.

KvF samples are composed of variable amounts of quartz, K-feldspar, albite, illite±muscovite and chlorite. Moreover, there are some accessory minerals such as hematite, calcite, dolomite, pyrite and smectite.

In JF samples, quartz, illite±muscovite, smectite and K-feldspar are the main components, and albite, hematite, dolomite and magnesite appear in low proportions.

3.3. Metalloid content versus reference values

The relative abundance of the elements studied in different terrestrial systems, including local geochemical backgrounds of the silicate and carbonate rock covered catchment areas in the Mecsek and Villány Mts. (SW Hungary), is given in table 4. The concentration of As in sedimentary rocks is typically in the range of 5–10 mg/kg (Smedley & Kinniburgh, 2002), i.e. slightly above average terrestrial abundance represented by the upper continental crust composition (UCC, Wedepohl, 1995; McLennan, 2001; Rudnick & Gao, 2003).

Table 1. Descriptive basic statistics of the As and Sb contents (mg/kg) in the mudrock samples

Mudrocks (N=35)		KoF N=11	CF N=4	BF N=10	KvF N=8	JF N=2
As	min	1.4	19.0	18.0	5.5	19.0
	max	8.7	37.0	43.0	69.6	38.0
	median	3.5	26.0	30.5	11.0	28.5
Sb	min	0.5	1.4	1.6	0.9	1.1
	max	4.7	2.2	19.0	1.9	1.1
	median	1.5	1.5	2.0	1.2	1.1

Abbreviations: N: number of analyses; KoF: Korpád Sandstone Formation; CF: Cserdi Conglomerate Formation; BF: Boda Claystone Formation; KvF: Kővágószőlős Sandstone Formation; JF: Jakabhegy Sandstone Formation

Table 2. Descriptive basic statistics of the As and Sb contents (mg/kg) in the sandstone samples

Sandstones (N=74)		KoF N=15	CF N=18	BF N=3	KvF N=33	JF N=5
As	min	0.6	5.2	6.3	2.1	2.2
	max	12.0	74.2	16.0	80.6	7.5
	median	2.3	20.5	8.4	5.3	2.7
Sb	min	0.4	0.4	0.7	0.4	0.2
	max	1.8	2.7	1.3	1.8	0.7
	median	0.8	0.8	0.9	0.7	0.4

Abbreviations: N: number of analyses; KoF: Korpád Sandstone Formation; CF: Cserdi Conglomerate Formation; BF: Boda Claystone Formation; KvF: Kővágószőlős Sandstone Formation; JF: Jakabhegy Sandstone Formation

Table 3. Mineralogical composition of the examined Lower Permian to Lower Triassic sandstone and mudrock samples (Southern Transdanubia, SW Hungary)

Lithostratigraphy	No of analyses	Major components	Accessories
JF sandstones	N=5	q, 10A ± sme, kfp	kfp, sme, hem, do ± ab, mgs
JF mudrocks	N=2	10A, q	kfp, ab, sme, hem, do, mgs
KvF sandstones	N=18	q, kfp, ab ± 10A, chl, sme	10A ± chl, kfp, hem, cc, do, sme, pyr
KvF mudrocks	N=2	10A, chl, q, ab ± do	kfp, cc, do, hem
BF sandstones	N=3	q, ab ± cc, 10A, do, hem	kfp, chl ± do, hem, 10A, sme
BF mudrocks	N=10	10A, ab, hem ± q, do, cc	chl ± cc, do, kfp, sme, kao
CF sandstones	N=11	q, ab, 10A ± hem, kfp, anh	kfp, do, sme, hem ± hem, kao, chl
CF mudrocks	N=5	q, 10A, hem, ab, kfp	chl, kao ± do
KoF sandstones	N=17	q, ab ± chl, 10A, kao, hem, do, sme	kao, 10A, cc ± do, sid, chl, sme, hem, pyr
KoF mudrocks	N=11	q, ab, 10A ± chl, hem, cc, do	chl, do, hem, kao ± ab, kfp, sid

Abbreviations: JF: Jakabhegy Sandstone Formation; KvF: Kővágószőlős Sandstone Formation; BF: Boda Claystone Formation; CF: Cserdi Conglomerate Formation; KoF: Korpád Sandstone Formation; 10A: 10 Ångström phases (micas±illite); ab: albite; anh: anhydrite; cc: calcite; chl: chlorite; do: dolomite; hem: hematite; kao: kaolinite; kfp: K-feldspar; mgs: magnesite; pyr: pyrite; q: quartz; sid: siderite; sme: smectite

Sands and sandstones tend to have the lowest concentrations, reflecting the low metalloid contents of their dominant framework minerals: quartz and feldspars (both K-feldspar and plagioclase). Average sandstone As concentrations are around 4 mg/kg (Smedley & Kinniburgh, 2002), although other work (Matschullat, 2000) gave a lower average value of 0.5–1.0 mg/kg (Table 4). The concentration of Sb in sedimentary rocks is generally somewhat lower, varying in the range of 0.2–2 mg/kg (Filella et al., 2002). Regarding local natural background, As and Sb values of the silicate and carbonate rock covered catchment areas in the South Transdanubian region (Fügedi et al., 2007) are comparable to world-average siliciclastic sedimentary rock metalloid concentrations (Table 4).

Table 4. Typical elemental concentrations (in mg/kg) for reference material

Material*	As	Sb
UCC (1)	2.0	0.31
UCC (2)	1.5	0.2
UCC (3)	4.8	0.4
Shales	3–13	1–2
Sandstones	0.5–4.1	
Pennsylvanian metasandstones	2–40	0.4–2.5
Background (1)	5.3	0.38
Background (2)	6.6	0.5

*UCC values were used from (1) Wedepohl (1995), (2) McLennan (2001) and (3) Rudnick & Gao (2003); shale and sandstone data come from Condie (1993), Wedepohl (1995), Matschullat (2000), McLennan (2001), Filella et al. (2002) and Smedley & Kinniburgh (2002); Pennsylvanian Tésény Metasandstone data come from Varga et al. (2012); additionally, background data for (1) silicate and (2) carbonate rock covered catchment areas come from Fügedi et al. (2007)

In the Permian to Lower Triassic siliciclastic rocks of the Western Mecsek Mountains, compared

to the sandstone samples, the mudrock units have generally higher As and Sb contents (Tables 1 and 2), which could be a consequence of the natural enrichment of these elements in the pelitic material (Garcia-Sanchez & Alvarez-Ayuso, 2003).

The contents of As and Sb compared with those reported for the UCC (McLennan, 2001) as a measure of the analysed elements are shown for each of the formations studied in the plots of figure 1. Concentrations of As in the mudrock and sandstone samples are uniformly higher than the normal global value (UCC); moreover, they are locally higher than the background levels (0.2–16 mg/kg) reported for different sedimentary rock types such as clays and mudstones, sandstones, arkoses and conglomerates (Garcia-Sanchez & Alvarez-Ayuso, 2003; Fügedi et al., 2007). In general, the highest medians of As were found in the Upper Permian CF and BF samples. Their enrichment factors for As relative to the UCC is of the order of 20 times, which could be of local importance as a potential source of this metalloid in the contamination process of soils during pedogenesis. On the other hand, the siliciclastic sedimentary rock samples are only slightly enriched in Sb relative to the UCC; and the median Sb contents are similar or slightly lower in the samples studied than in average shales (1–2 mg/kg; Filella et al., 2002). Therefore, the Lower Permian to Lower Triassic siliciclastic sedimentary rocks are not considered to be direct pollution sources of Sb during soil forming processes.

3.4. Origin of local background

Areas of sulfide mineralization have long been associated with environmental As problems.

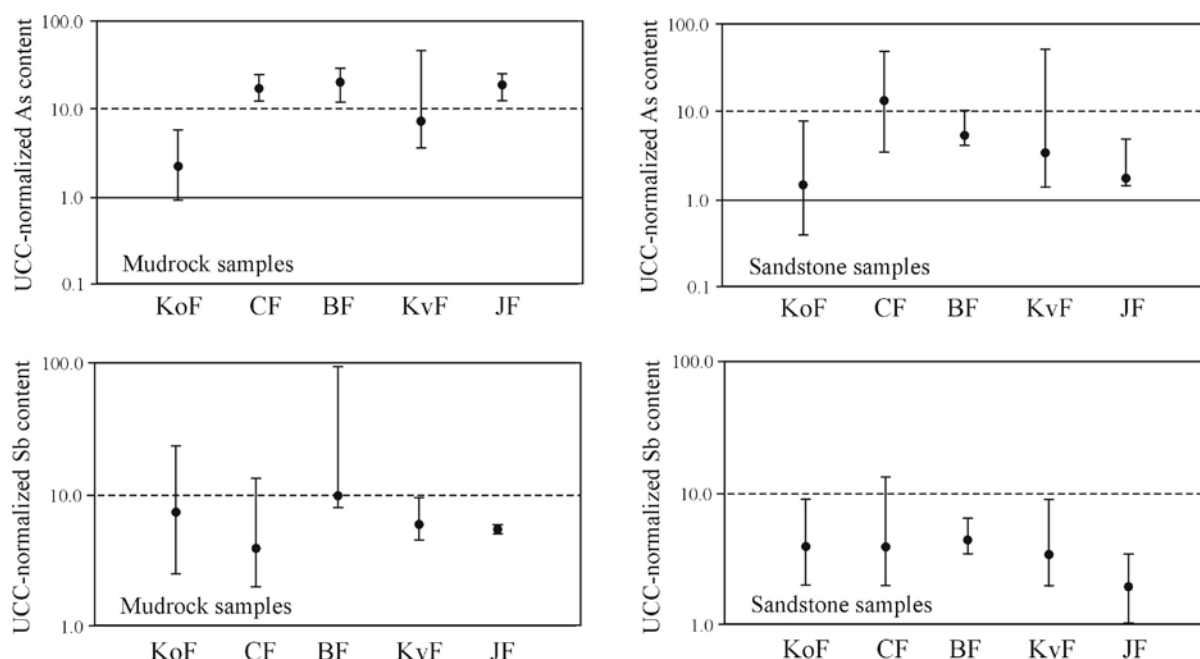


Figure 1. Upper Continental Crust (UCC)-normalized plots of As and Sb contents (mg/kg) for each studied lithostratigraphic unit. The bar represents minimum–maximum range, the dot represents the median

Sulfide minerals (e.g., arsenopyrite, arsenian pyrite, realgar, orpiment) can contain very high concentrations of As and oxidation of the minerals during weathering can lead to As release (Smedley, 2008; Appelo & Postma, 2009). Arsenic is also present at high concentrations in the more common sulfide minerals such as pyrite, chalcopyrite, sphalerite, galena and marcasite (several weight %), the most abundant of which is pyrite (FeS_2) with arsenic concentration range 100–120,000 mg/kg (Smedley, 2008). Therefore, hydrothermally altered and mineralized rocks, and associated veins constitute the principal source of As and Sb in the natural environments (Garcia-Sanchez & Alvarez-Ayuso, 2003; Filella et al., 2002; Ashley et al., 2003).

On the other hand, high concentrations of As can also be found in many oxide and oxyhydroxide minerals such as hematite (up to 29,000 mg/kg) and iron(III) oxyhydroxide (up to 29,000 mg/kg; Smedley, 2008). In siliciclastic sedimentary systems, hematite can also occur as a grain-coating cement mineral in oxidizing environment (red beds; Tucker, 1991; Chan et al., 2000). Sorption plays an important role in As speciation. Amorphous iron oxides in particular are well-known to have strong sorption capacities for As and therefore exert a strong influence on As mobility (Smedley, 2008; Appelo & Postma, 2009; see Fig. 2).

Within a regional context, Transdanubian part of the Tisia Block (Mecsek and Villány Mts. in South Hungary) and Southern Tisia (Slavonian Mts., Moslavačka Gora and Požeška Gora in Croatia) is not rich in ore deposits (Haas et al., 1999; Jurković,

2003; Palinkaš et al., 2008). The rare commodities of economic value in South Tisia are part of metamorphic sequences (e.g., graphite), products of igneous activity (e.g., pegmatites), and sedimentary-epigenetic uranium occurrences. Hydrothermal quartz veinlets with sulfides have only mineralogical significance in the Moslavačka Gora (Jurković, 2003; Palinkaš et al., 2008).

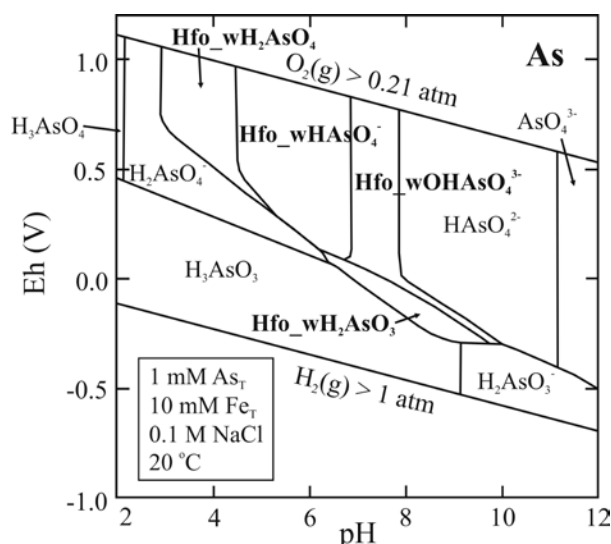


Figure 2. Predominant species in the system As-Fe-H₂O where sorption of As species onto hydrous ferric oxide (Hfo) is taken into consideration (modified after Smedley, 2008). Amorphous iron oxides exert a strong influence on As mobility (Smedley, 2008; Appelo & Postma, 2009)

Regarding the crystalline basement of the SW part of the Pannonian Basin hydrothermal ore

indications occur in the rock body (Tarnai, 1998; Fintor et al., 2011; Vincze et al., 2011). These veins contain only minor amounts of pyrite; additionally, chalcopyrite and sphalerite grew sporadically in non-sulfide host minerals, like epidote, diopside, chlorite, quartz and calcite. The ore phase mostly form thin veinlets filled by pyrite–pyrrhotite ore assemblages, but at some places, several centimetres thick polymetallic sulfide veins also can be found (Tarnai, 1998). According to Vincze et al., (2011), economically insignificant uranium ore veinlets were developed along the fractures of the mylonitized granitoid rocks in the western part of the Mecsek Mts. Uranium mineralization forms uraninite, coffinite, pyrite and calcite in veins, representing ascendant hydrothermal (epi-telethermal) mineralizing fluids genetically related to an Alpine rejuvenation of an earlier pre-Alpine event (Vincze et al., 2011).

Additionally, within the Permian sequences (Korpád Sandstone Formation and the historical uranium deposit of the Kővágószőlős Sandstone Formation), sedimentary-epigenetic uranium-copper mineralization events were recorded (Vincze, 1985; Fülöp, 1994; Vincze et al., 2011; Bodor et al., 2012). It comprises uraninite, coffinite, pyrite, galena, chalcopyrite, marcasite, sphalerite, sulfosalt minerals, chalcocite, covellite, and arsenopyrite (Vincze, 1985; Fülöp, 1994; Bodor et al., 2012).

In the Mecsek Mts. considerable sulfide mineralization does not occur within the Lower Permian to Lower Triassic sequences, however, within these sedimentary rocks fracture-controlled vein systems dominated by quartz and carbonates are present. Based on our petrographic observations, these hydrothermally altered zones contain minor disperse grains of sulfide minerals such as pyrite. Furthermore, XRD mineralogical results also indicate pyrite occurrence in some KoF and KvF sandstone samples (Table 3). Therefore, the possible source of metalloids could be the sulfide minerals contained in these rocks with heterogeneous distribution of metalloid-bearing phases. This interpretation is in agreement with the trace elemental geochemistry of the Pennsylvanian Tésény Metasandstone Formation (TMF, SW Hungary). In TMF subsurface rocks, some samples have naturally elevated As and Sb contents (up to 40 and 2.5 mg/kg, respectively) reflecting hydrothermally altered, pyritized rocks (Varga et al., 2012). Furthermore, another source of As and Sb could be the metalloid-bearing Fe oxides and oxyhydroxides coating mineral grains, such as quartz, feldspar, etc (Garcia-Sanchez & Alvarez-Ayuso 2003; Smedley, 2008). However, none of the

studied samples shows anomalous high local background concentrations of naturally-occurring As and Sb (Table 1).

4. CONCLUSIONS

In the Lower Permian to Lower Triassic mudrock samples from the western part of the Mecsek Mountains, the median As and Sb contents range from 3.5 to 30.5 mg/kg, and from 1.1 to 2.0 mg/kg, respectively. In the associated sandstone samples, the median As contents range from 2.3 to 20.5 mg/kg, while the median Sb contents range from 0.4 to 0.9 mg/kg. The highest As and Sb concentrations were found in the Upper Permian Cserdi and Boda formations, which could be only of local importance as a source of metalloids in the contamination process of soils and waters.

The source for background levels of As and Sb in the Lower Permian to Lower Triassic siliciclastic sedimentary rocks could be related to hydrothermal activity in the region (hydrothermal quartz-carbonate veinlets with sulfides). Another source of metalloids could be the Fe oxides and oxyhydroxides coating mineral grains, which are host of both elements.

The results obtained contribute to our knowledge on the As and Sb compositions of the sedimentary units studied and are the first attempt to establish the origin of these toxic elements in SW Hungary. The data will be of use for further detailed studies in the region as well as to complement other geochemical databases.

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