Assessment of heavy metal contamination in paddy soils from Kočani Field (Republic of Macedonia): part I

Ocena onesnaženja s težkimi kovinami v tleh riževih polj iz Kočanskega polja (Republika Makedonija): 1. del

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Abstract: The research focuses on the assessment of heavy metals contamination in paddy soils from Kočani Field (Republic of Macedonia). Very high concentrations of Ag, As, Cd, Cu, Mo, Ni, Pb, Sb and Zn were found in the paddy soils samples from the vicinity of the Zletovska River (western part of Kočani Field). This river drains the untreated effluents from the Pb-Zn mine in Zletovo-Kratovo region and it is used for irrigation of the surrounding paddy fields. Heavy metal contamination of Kočani paddy soils was evaluated by using contamination factor and contamination degree, which confirmed a very high heavy metal contamination status for Kočani paddy soils, especially around Zletovska River area. Taking into account the results of this research the area around the Zletovska River is considered as the most anthropogenically impacted part of Kočani Field.

Izvleček: V tej študiji smo ocenili in opredelili onesnaženje s težkimi kovinami v tleh riževih polj Kočanskega polja (Republika Makedonija). Zelo visoke koncentracije Ag, As, Cd, Cu, Mo, Ni, Pb, Sb in Zn smo ugotovili v vzorcih tal iz bližine reke Zletovske (zahodni del Kočanskega polja). Reka Zletovska odvaja neprečiščene odplake in odpadke z območja rudnika Pb-Zn Zletovo, kmetje pa jo uporabljajo za namakanje riževih polj. Visoke vrednosti težkih kovin v tleh riževih polj območja Kočanskega polja smo potrdili tudi z izračunom faktorja onesnaževanja in stopnje onesnaževanja. Na podlagi zelo

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visokih koncentracij težkih kovin v vzorcih tal riževih polj smo ugotovili, da je območje okoli reke Zletovske resna ekološka grožnja bližnjim ekosistemom.

Key words: assessment of contamination, heavy metals, paddy soil, Kočani Field, Republic of Macedonia

Ključne besede: ocena onesnaženja, težke kovine, tla riževih polj, Kočansko polje, Republika Makedonija

Introduction

Soil is a specific component of the biosphere because it is not only treated as geochemical sink for contaminants, but also as a natural buffer system controlling the transport of chemical elements and substances into the atmosphere, hydrosphere and biota. It has always been important to humans and their health, especially as a resource that can be used for shelter and food production (Kabata-Pendias & Pendias, 2001). Unfortunately, heavy metal contamination of soils has become a widespread global problem over the past two decades and represents a long-term risk to ecosystem health and quality (Kabata-Pendias & Pendias, 2001).

Base-metal mining activities together with milling and grinding operations, concentrating ore and the disposal of tailings, acid mine and mill wastewater provide important sources of heavy metals entering into the environment (Adriano, 1986). Consequently, very high concentrations of heavy metals can be found in and around abandoned and active mines, which can affect nearby agricultural soils, food crops, riverine water and stream sediments (Adriano, 1986; Jung, 2001; Jung & Thornton, 1997; Korre et al., 2002; Lee et al., 2001; Liu et al., 2005; Lu & Zhang, 2005; Simmons et al., 2005; Witte et al., 2004, Wong et al., 2002; Yang et al., 2004).

Numerous studies worldwide have investigated the heavy metal concentrations in soils (Kabata-Pendias & Pendias, 2001). Although mining is one of the most important industries in the Republic of Macedonia, studies about heavy metal concentrations in soils originating from historical or current base-metal mining activities are scarce, especially in the area of Kočani region (Dolenec et al., 2007; Rogan et al., 2009; Rogan et al., 2010).

In this context, the major aims of the presented study are:

- to detect the total heavy metal concentrations and distribution in paddy soil samples from Kočani Field and compare these results with permissible levels of heavy metals in arable soils;

- to assess the soil contamination by environmental indexes, contamination factor (Hakanson, 1980; Loska et al., 2004) and degree of contamination (Hakanson, 1980) and;
- to evaluate the overall environmental risk of the soil system in the Kočani Field area.

Materials and methods

Study area

Kočani Field is located in eastern Macedonia, about 32 km from the city of Štip and 115 km from the capital city Skopje. With an average length of 35 km and width of 5 km, Kočani Field lies in the valley of the Bregalnica River between the Osogovo Mountains in the north and Plačkovica Mountains in the south (Figure 1).

Figure 1. Study area, Kočani Field

The broader region is well known as an agricultural and mining province, with significant thermal activity. The municipality of the Kočani area comprises 28 settlements with 38,092 inhabitants.

The Bregalnica River, together with its tributaries, represents the principal drainage system in the investigated area and is, therefore, an important water supply for the irrigation of the surrounding paddy fields. The main tributaries of the Bregalnica River are the Kamenica River in the northeastern part of the study area and the Zletovska River in the western side of Kočani Field (Figure 1). The Zletovska River originally drained the central part of the Zletovo-Kratovo volcanic complex as well as the untreated mine effluents from the Pb-Zn Zletovo mine and its ore processing facilities. Both rivers are used by local farmers for the irrigation of the nearby paddy fields. The Kočanska and Orizarska Rivers are two small tributaries of the Bregalnica River (Figure 1) that drain the southern part of the Osogovo Mountains, as well as the untreated municipal wastes and domestic sewage of the cities of Kočani and Orizari. The riverine water of the Kočanska and Orizarska Rivers is also used for the irrigation of the paddy fields located in the northeastern part of Kočani Field.

The paddy soil of Kočani Field was estimated to originate from the composite material of the sediment derived from igneous, metamorphic and sedimentary rocks located in the wider area of Kočani. The sediment material was transported by the Bregalnica River and its tributaries and deposited in the Kočani depression (Dolenec et al., 2007). The soil mineralogy and elemental composition are closely related to the acidic and intermediate rocks of the Kočani region (Dolenec et al., 2007).

Zletovo-Kratovo ore district

The Zletovo-Kratovo Pb-Zn ore district is situated 5 km northwest of the Zletovo village and about 7 km from the city of Probistip (Figure 1). It is located in the central part of the Zletovo-Kratovo volcanic complex. The mineral association comprises galena (principal ore mineral) and sphalerite, with subordinate pyrite, lesser amounts of siderite and chalcopyrite and occasionally pyrrhotine, marcasite and magnetite. Minor occurrences of U-mineralisation have also been discovered (pitchblende). The Zletovo mine has an annual capacity of 350,000 tons of Pb-Zn concentrate (8 % Pb and Zn) and significant concentrations of Ag, Bi, Cd and Cu. Ore is concentrated at the flotation processes at Probistip, and tailings material is stored in two impoundments in the adjacent valleys (Alderton et al., 2005).

Paddy soil sampling and analysis

The sampling of the paddy soils was carried out in autumn 2005. The sampling locations are shown in Figure 2. Paddy soil samples were assembled at 38 locations from seven profiles across Kočani Field (Sections I–VII).

Near surface paddy soils (0–20 cm in depth) were collected because it is impossible to distinguish the A, B and C horizons in the agricultural soil. The paddy soils were sampled using a plastic spade to avoid any heavy metal contamination. Each paddy soil sample comprised a composite of five subsamples taken within a (1 × 1) m^2.

The paddy soil samples were air dried at 25 °C for one week and sieved through a 2 mm polyethylene sieve to remove plant debris, pebbles and stones. Afterwards they were ground in a mechanical agate grinder into a fine powder for subsequent physico-chemical parameters and geochemical analysis.

All paddy soil samples were analysed for heavy metal concentrations in a certified commercial Canadian laboratory (Acme Analytical Laboratories, Vancouver, B. C., Canada) by extraction for 1 h with 2-2-2-HCl-HNO_3-H_2O at 95 °C with ICP-MS.

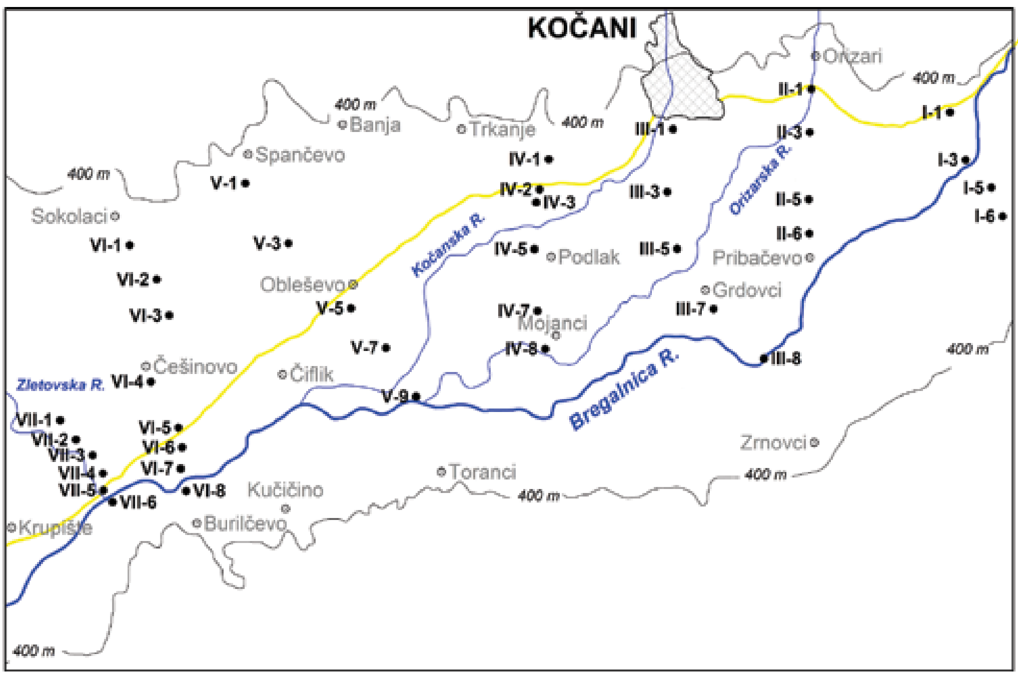

Figure 2. Paddy soil sampling locations map

The accuracy and precision of the paddy soil analysis were assessed using an international reference material such as Canadian Certified Reference Material Project (CCRMP) SO-1 (soil) and United States Geological Survey (USGS) G-1 (granite). The analytical precision and accuracy were better than ±5 % for the investigated elements. This was indicated by the results of the duplicate measurements in 10 soil samples as well as duplicate measurements of the G-1 and SO-1 standards.

RESULTS AND DISCUSSION

Heavy metal concentration in the paddy soil

Table 1 shows the concentrations of Ag, As, Cd, Cu, Mo, Ni, Pb, Sb and Zn determined in the paddy soil samples from Kočani Field together with the assumed permissible level (1st limiting level) of heavy metals adopted by the National Environmental Protection Agency of Slovenia (URADNI LIST RS, 1996), typical heavy metal contents of comparable soils around the world (BOWEN, 1979) and the maximum allowable concentrations (MAC) of trace elements in agricultural soil proposed by the GERMAN FEDERAL MINISTRY OF ENVIRONMENT (1992). The permissible level of heavy metals and the MAC of trace metals signify the values above which toxicity is considered possible.
Table 2 displays the descriptive statistical parameters (mean, median, range and standard deviation (SD)).

The Ag, Mo, Ni, Sb concentrations defined in the paddy soil samples from Kočani Field were corresponding to the average worldwide values for Ag (WEDEPOHL, 1974) and Sb (BOWEN, 1979) and did not exceed the limit values for Mo and Ni reported by the Environmental Protection Agency of Slovenia (Table 1).

Very high concentrations of As, Cd, Cu, Pb and Zn (Table 1) were found in the paddy soils samples from Section VII. The determined values significantly exceeded the typical As content within comparable soils around the world (BOWEN, 1979), as well as the limit values for As, Cd, Cu, Pb and Zn suggested by the environmental protection agencies of Slovenia and Germany (Table 1).

To compare the distribution of each heavy metal in the paddy soil samples between Sections I–VII, whisker plots were employed (Figure 3). All soil samples in Sections I–VI had similar levels of median values, suggesting that their intersection differences were not substantial, whereas significant variations existed only between Section VII and the other sections. The only exception and consequently difference was is the distribution of the Ni values in the paddy soil samples.

Table 1. Total heavy metal concentrations in the paddy soil of Kočani Field: (1) typical contents of Ag, As, Cd, Cu, Mo, Ni, Sb, Pb and Zn in comparable soils around the world; (2) limits for elemental concentrations in soil (Environmental Protection Agency of Slovenia (Uradni list RS, 1996); (3) maximum allowable concentrations (MAC) of trace elements in agricultural soils proposed by the German Federal Ministry of the Environment (1992).

Element (μg/g) Location	Ag	As	Cd	Cu	Mo	Ni	Pb	Sb	Zn
I-1	0.7	3.4	0.1	14.9	0.5	15.6	19.1	0.2	53
I-3	0.2	18.7	0.5	40.3	1	31.1	81.3	0.6	162
I-5	0.1	5.6	0.2	32	0.5	29.7	18.8	0.2	85
I-6	0.7	5.1	0.1	25.9	0.5	25.7	13.1	0.2	70
II-1	0.1	6.1	0.2	26.7	0.5	29.4	26.6	0.2	93
II-3	0.1	5	0.1	29.1	0.4	28.2	21	0.2	81
II-5	0.1	5.3	0.2	24.9	0.5	22	20.8	0.3	80
II-6	0.1	11.8	0.3	25.7	0.6	20.8	32.1	0.8	100
III-1	0.1	6.1	0.2	27.2	0.6	27.5	18.1	0.2	89
III-3	0.1	3.1	0.4	32.6	0.3	25.4	20.4	0.2	95
III-5	0.1	8.3	0.2	32.7	0.5	36.8	23.9	0.2	102
III-7	0.7	8.2	0.2	16.8	0.3	13.9	22.1	0.4	64
III-8	0.7	4.2	0.1	21.8	0.5	20.8	10.5	0.1	64
IV-1	0.1	3.8	0.1	28.5	0.7	21.7	13.9	0.2	74
IV-2	0.7	3.5	0.2	17.1	0.6	17.3	17.9	0.2	68
IV-3	0.7	4.3	0.3	23.4	0.5	24.1	19.1	0.2	102
IV-5	0.1	6.3	0.2	25.7	0.6	25.5	15.4	0.2	94
IV-7	0.1	8.1	0.2	26.2	0.7	26.1	17	0.2	83
IV-8	0.1	5.9	0.2	28.9	0.6	28.6	18.1	0.2	96
V-1	0.1	10	0.2	19	0.7	9.1	30.8	0.3	85
V-3	0.1	7.8	0.2	20	0.6	9.5	26.9	0.2	73
V-5	0.1	6.5	0.2	25	0.6	22.8	22.1	0.2	98
V-7	0.1	6.4	0.2	22.8	0.4	17.2	17.8	0.3	67
V-9	0.1	4.5	0.2	20.8	0.3	16.7	19.2	0.3	69
VI-1	0.1	5	0.2	21.4	0.3	17.1	24.1	0.3	71
VI-2	0.1	6.8	0.3	24.5	0.6	9.9	39.4	0.2	76
VI-3	0.1	10.1	0.3	34.1	0.6	22.3	35.4	0.3	86
VI-4	0.1	9.9	0.3	28.6	0.5	25.2	41.4	0.4	105
VI-5	0.1	10.5	0.3	25	0.5	21.7	39.6	0.4	94
VI-6	0.1	11.5	0.3	28.3	0.6	22	45.1	0.5	107
VI-7	0.7	7.7	0.1	19.3	0.5	16.4	20	0.4	66
VI-8	0.7	6.2	0.2	29.5	0.5	30.5	16.3	0.2	72
VII-1	0.9	22.2	2.7	48.8	1.3	10.9	411.9	1.8	531
VII-2	2	42	5.6	99.4	1.8	15.4	892.4	2.5	1134
VII-3	1.4	35.1	4.5	89.4	1.4	15.5	726.7	2	893
VII-4	2.1	47.6	6.4	80.1	1.8	11.9	983.1	3	1245
VII-5	1.6	39.6	5	68.3	1.6	14.1	745.1	2.2	928
VII-6	0.6	20.7	2	47.1	0.9	19.8	295.7	1	384
1	0.1-8	6	0.35	30	/	/	35	1	90
2	/	20	1	60	10	50	85	/	200
3	/	/	1.5	60	/	/	100	/	200

Table 2. Descriptive basic statistic parameters for heavy metals in the paddy soils of Kočani Field

Elements (μg/g)	Ag	As	Cd	Cu	Mo	Ni	Sb	Pb	Zn
Mean	0.306	11.4	0.9	33	0.682	21.0	0.57	128	206
Median	0.100	6.7	0.2	26	0.600	21.7	0.25	22	88
Minimum	0.066	3.1	0.1	15	0.300	9.1	0.10	11	53
Maximum	2.100	47.6	6.4	99	1.800	36.8	3.00	983	1245
S.D.	0.541	11.3	1.7	20	0.389	6.8	0.72	260	310

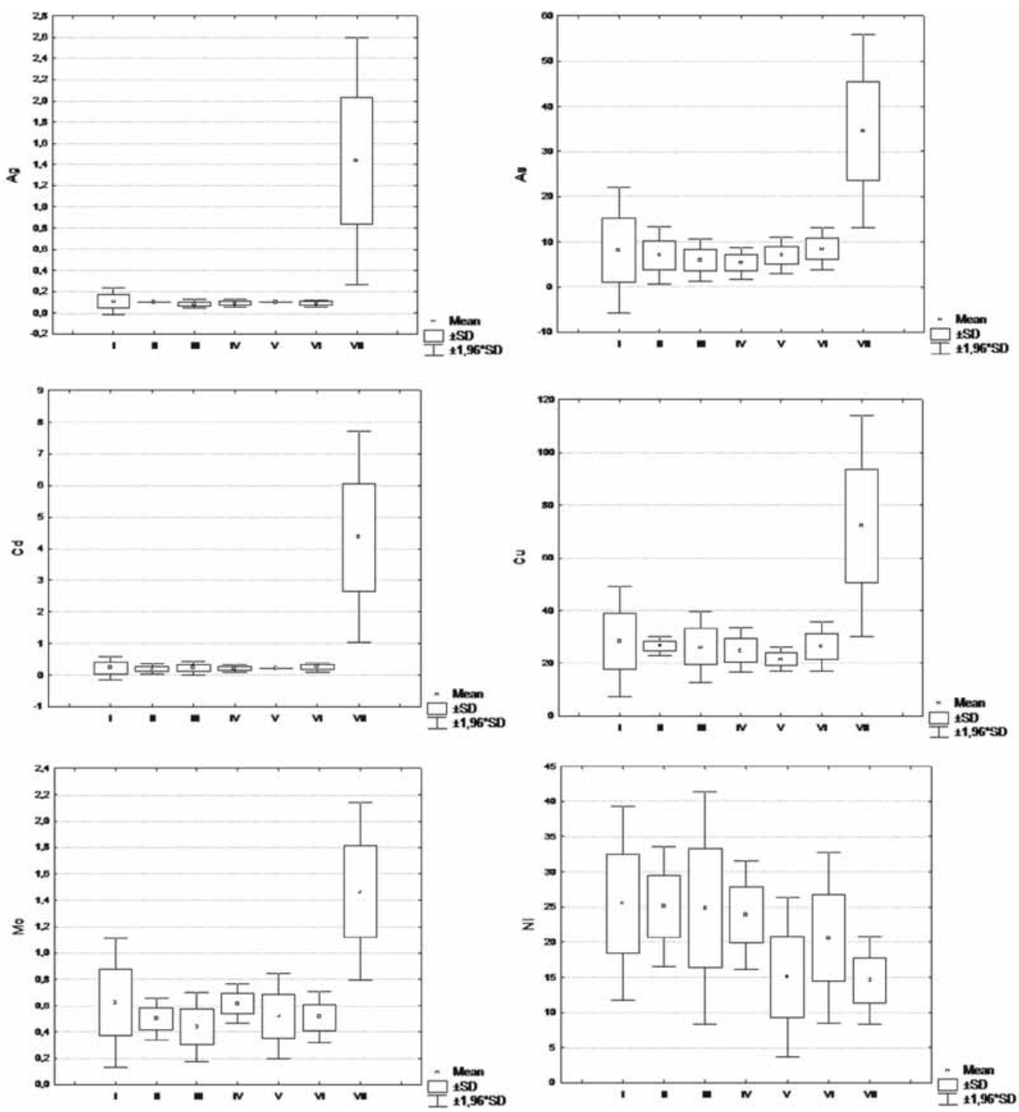

Figure 3. Whisker plots of Ag, As, Cd, Cu, Mo, Ni, Pb, Sb and Zn for ddy soil samples.

Contamination factor (c_f^i) and contamination degree (C_{deg})

According to contamination factor (c_f^i) values (mean), the paddy soils from the Kočani Field were classified as slightly (low) contaminated with Mo, moderately contaminated with Cu and Ni, considerably contaminated with Ag, Pb, Sb and Zn and very highly contaminated with Cd and As (Figure 4). The highest Ag, As, Cd, Pb, Sb and Zn c_f^i values (very high contamination factor) and the highest Cu and Mo c_f^i values (moderate contamination factor) were found in the paddy soil samples from Section VII (close to Zletovska River area). The highest Ni c_f^i values (moderate contamination factor) were determined in Section I. The calculated c_f^i values entirely confirmed the I_{geo} results.

The complete assessment of the paddy soil contamination was evaluated by degree of contamination (C_{deg}). The C_{deg} (mean) of the studied soils samples was 36.2 (Table 3), which signified very high contamination. Cd and As represented the largest contamination degree factors with 23 % and 20 %, respectively. Ag and Pb both contributed 16 %, Sb 9 % and Zn 8 %. Cu, Ni and Mo negligibly influenced the soil contamination with 4 %, 3 % and 1 %, respectively.

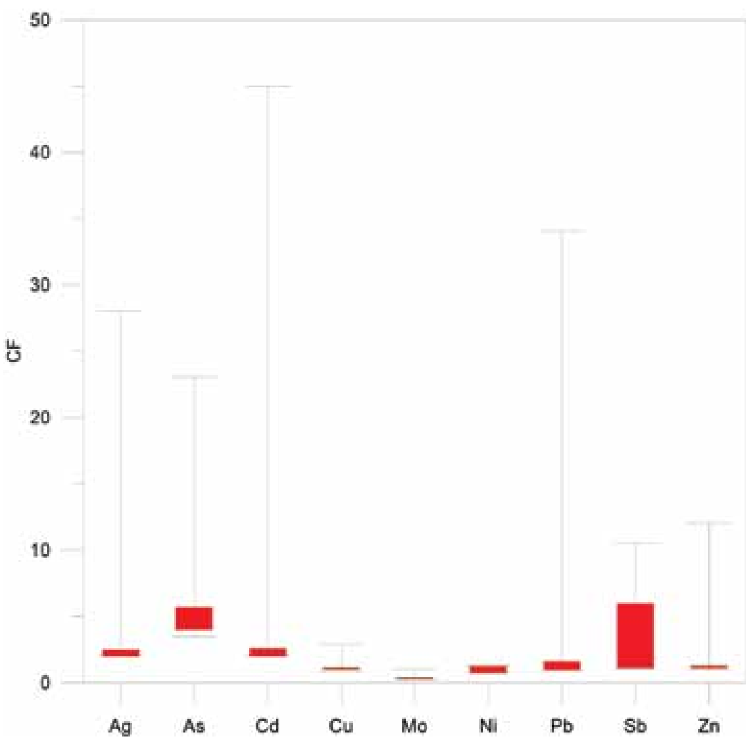

Figure 4. Contamination factor (c_f^i) values for the paddy soils from the Kočani Field

Table 3. Contamination factors and degrees for heavy metals in the paddy soils.

Metal	Range	Average
Ag	2.0–28.0	5.8
As	3.5–23.0	7.3
Cd	2.0–45.0	8.3
Cu	0.9–2.9	1.3
Mo	0.3–1.0	0.4
Ni	0.7–1.3	1.1
Pb	0.9–34.0	5.9
Sb	1.0–10.5	3.4
Zn	0.9–12.0	2.7
Degree of contamination	12.2–157.7	36.2

The paddy soil samples from Section VII, located in the vicinity of the Zletovska River and Zletovo-Kratovo ore district, received a comparatively higher input of anthropogenically derived heavy metals than other parts of the Kočani area. The As, Cd, Cu, Pb and Zn concentrations determined in the paddy soils from Section VII significantly exceeded maximum permissible levels. The distinction between heavy metal contents in Section VII and the other sections was also confirmed by statistical analysis. The pollution in Section VII is undoubtedly related to the irrigation of the paddy fields with the riverine water from the Zletovska River, which drains acidic mine waters and untreated mining waste effluents from the Zletovo-Kratovo mine (ALDERTON et al., 2005).

Elevated concentrations of As, Cd, Cu, Pb and Zn were observed in other paddy soil sections (especially Sections V and VI). This elevation originates from the discharges of the untreated municipal and domestic waste from the city of Kočani and the village of Orizari into the riverine systems of the Kočanska and Orizarska Rivers, which are both used for irrigation purposes. The increased concentrations of the investigated heavy metals could also be explained by the agricultural application of various fertilisers and pesticides, urban and traffic sources and atmospheric deposition.

Although the concentrations of Ag, Mo, Ni and Sb were below the mentioned threshold values, their enrichment in the paddy soil samples near the Zletovska River was also noticeable. This situation confirms the higher input of heavy metals in the area around the Zletovska River.

The high anthropogenic impact on the paddy soils in Section VII was as well demonstrated by the calculation of the following environmental indexes: contamination factor and degree of contamination. The elevation of the heavy metals present in the paddy soil samples from the other parts of Kočani Field was similarly verified with the results of the environmental indexes. From an environmental point of view, it is evident that the paddy soil samples from Section VII represent a serious risk to the surrounding ecosystems (Dolenec et al., 2007; Rogan et al., 2009, Rogan et al., 2010).

Conclusions

As, Cd, Cu, Pb and Zn concentrations determined in the paddy soils from Section VII significantly exceeded the permissible maximum levels. Although the concentrations of Ag, Mo, Ni and Sb were below the mentioned threshold values, their enrichment in the paddy soil samples near the Zletovska River was noticeable, consequently confirming the higher input of heavy metals in the area around the Zletovska River.

The high anthropogenic impact on the paddy soils in Section VII was demonstrated by the calculation of the contamination factor and degree of contamination. The elevation of the heavy metals present in the paddy soil samples from other parts of Kočani Field was similarly verified with the results of the environmental indexes. According to the results of the heavy metal concentrations present in the paddy soil the area around the Zletovska River is considered as the most anthropogenically impacted part of Kočani Field. The highly elevated concentrations of the analysed heavy metals were undoubtedly related to past and present mining activities, especially in the Zletovo-Kratovo ore district.

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