RESEARCH ARTICLE OPEN

Agricultural Land Of Rural Areas And Way Of Removing Heavy Metals

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ABSTRACT

The aim of this paper is to determine Pb, Cd and Zn in: I) agricultural land of rural areas; II) selected plants; and III) establishing a new method for removing heavy metals from the soil. Potentiometric striping analysis (PSA) was used to determine the metal concentration. The obtained results indicate: I) the presence of heavy metals in the analyzed soil; II) the presence of heavy metals in selected plants, and III) reduction of metal concentration in soil after planting.

Key words:heavy metals, rural land, plants.

Date Of Submission:18-10-2018 Date Of Acceptance: 04-11-2018

I. INTRODUCTION

Due to the production method [1], toxicity, plant adoption and inclusion in the food chain, heavy metals increasingly appear as dominant carriers of environmental pollution [2,3]. In addition to the polluters, in the northern part of Kosovo and Metohija, they represent a good indicator of the effects of the production processes and the existing passive and active ore dumps of the Trepca mine located in this area.

Due to the process in these complex formations, erosion, the effects of watercourses and atmospheric precipitation, heavy metals come into the air, water, and soil. Processes in the soil and the presence of substances that are very important for plants dictate product properties and fertility of the soil [4]. Pollution of the soil by heavy metals, i.e. knowledge of the mechanism of adoption, distribution, metabolism, and accumulation in plants is of great importance [5,6]. Plants react differently to elevated metal concentrations. The presence of heavy metals affects the life processes of plants through nutrition, water regime, photosynthesis, respiration, or through all physical-biochemical processes [7].The effects of heavy metals on plants are the reduction in the production of organic matter and changes in the chemical composition of plants [8,9]. Plants adopt heavy metals in the form of ions or organic complexes [10]. The process of adoption depends on the characteristics of the soil itself, the content of organic matter, accessible nutrients, the processes in the rhizosphere, and the application of phosphate, lime, and pH, the intensity of light and temperature, as well as cultivated genotype [11–14]. There is a frequent occurrence of damage to the mechanisms of regulation and adoption of ions by plants. Some authors have found that among metals there are antagonisms and a different reaction of plants to the presence of heavy metals [15,16]. By basic soil analysis (in order to evaluate interdependence and computation of correlation matrices), the dominant role of certain ions was also determined [17].

The scope of research in this paper is to determine the concentration of Pb, Cd, and Zn in soil and plants. The land for analysis was taken in the gotof village Mali Zvecan, Zitkovac, Guvniste, Belo Brdo in the northern part of Kosovo and Metohija.

From plants analyzed are: *LactucaSative*, *Allium cepe* as a plant that is intensively cultivated in the area; *Lotus corniculatus* L. and *Trifolium pretense* L., plants used in animal nutrition; as well as grasses (weed plants): *Cynodondactylon* and *Festuca arundinaceous Schreb*. These plants have been selected for application in the process cleaning of contaminated soil [18–21]. The concentration of the tested metals was determined at the beginning and at the end of the experiment using potentiometric stripping analysis [22–27].

II. EXPERIMENTAL PART

1.1. Apparatus

Determination of Pb, Cd, and Zn was performed using the Striping analyzer M1 (Faculty of Technology, Novi Sad, and *Symmetry*, Leskovac, Serbia) [28–30]. The investigated metals were determined in the same analytical step at a constant electricity of -48.90 μA over a time of 300 seconds and the potential of separation -1.40 V [22].

1.2. Chemicals

To perform the experimental part of this work, solutions from high purity chemicals (suprapur) manufactured by *Merck* (Darmstadt, Germany) were prepared. The basic solutions were prepared from the standards of lead, cadmium, zinc and mercury (1.000 g dm^{-3}) while the working solutions were prepared from the bases in the concentration range of 50 – 90 mg dm^{-3}. In addition to the standard solutions, acid solutions were used: chloride (HCl, 30%), nitrate (HNO_3, 65%); Salt solutions: potassium chloride (KCl), copper sulphate ($CuSO_4$), gallium chloride ($GaCl_3$), and acetone ($CH3COCH_3$, 99.5%). The solutions are stored in polyethylene bottles.

1.3. Sampling and preparation of samples

Soil samples were taken using a system of concentric circles from a depth of up to 30 cm using a hand probe. After drying (105 ºC), shredding and screening, the gram of powder obtained that way was converted into a solution by digestion with concentrated nitric and chloric acid, after which evaporation was carried out. The residual mass was dissolved in a 2% solution of chloric acid and stored in measuring vessels of 100 cm^3 until analysis [22].

To determine the concentration of Pb, Cd, and Zn, used plants are after washing, drying, and annealing (500 ºC), the gram of ash was soluble in nitric acid (5 cm^3, conc.) and evaporated. Evaporation was repeated after adding a few drops of concentrated chloric acid. The remaining white matter was dissolved with the chloric acid (5 cm^3, 2% solution), and prepared for analysis in 100 cm^3 measuring bottles [22,25,26]. Tests in this work were carried out through vegetation experiments, in six series with four tests per 2 kg of soil, figure 1.

Figure 1. The vegetation experiment

In one series it was for *Lactuca Sative*, in the second series *Allium cepe*, in the third *Lotus corniculatus* L. in the fourth *Trifolium pretens*e L. in the fifth *Cynodon dactylonea* and in the sixth *Festucea arundinaceaeSchreb*.Experiments were exhibited in the same conditions (sunlight, watering deionized water, without the addition of nutrients and atmospheric effects) from March to June.

III. RESULTS AND DISCUSSION

The results of determining the concentrations of Pb, Cd, and Zn in soil samples in the northern part of Kosovo and Metohija are shown in Table 1.

Table 1. The concentrations of Pb, Cd and Zn in the analysed soil samples

$\overline{X}$ µg g^{-1}	Measuring location				*MDK* µg g^{-1}
	Mali Zvecan	Zitkovac	Guvniste	Belo Brdo	
Pb	68.49	**112.08**	12.41	**370.46**	100.0
Cd	2.75	**3.58**	2.04	**12.13**	3.0
Zn	299.40	**379.34**	281.86	**434.89**	300.0

number of measurements n=5

Recent studies have shown that the average concentration in the uncontaminated soil is 2 – 200 µg g^{-1} Pb; 0.01 – 0.7 µg g^{-1} Cd and 10 – 300 µg g^{-1} Zn [9,31]. The obtained results (Table 1) show that at measuring points Zitkovac and Belo Brdo the concentration of metals in soil exceeded the limits of maximum allowable values [32]. Such results indicate that analyzed soil samples belong to the category of contaminated soils. The cause of the increased concentration of tested heavy metals in the soil in the northern part of Kosovo and Metohija is the influence of the production processes and the Trepca mines. The highest concentration of investigated metals was determined in the sample at the measuring points: of Belo Brdo located in the immediate vicinity of the mine with the same name, and Zitkovac located in the vicinity of the active and passive mines of the Trepca. Guvniste is one of the farthest places in terms of landfills, so the lowest concentration of tested metals has been determined in this soil sample.

The results of the determination of the concentration of Pb, Cd, and Zn in plants: *LactucaSative* and *Allium cepa*, planted on the analyzed soil, are shown in Table 2.

Table 2. The concentrations of Pb, Cd and Zn in *Lactuca Sative* and *Allium cepa*

	$\overline{X}$ / µg g^{-1} SM										
		Mali Zvecan		Zitkovac		Guvniste		Belo Brdo			
		root	leaf	root	leaf	root	leaf	root	leaf	*C*c	*C*t
Lactuca Sative	Pb	7.99	4.24	8.13	4.09	6.49	3.73	11.29	9.71	10.0	20.0
	Cd	2.32	3.48	2.42	3.55	2.05	2.60	4.05	5.38	5.0	10.0
	Zn	38.33	59.28	38.75	58.26	33.89	52.68	93.47	130.45	150.0	200.0
Allium cepa	Pb	4.38	2.08	4.60	3.26	4.37	2.15	10.09	10.43	10.0	20.0
	Cd	1.64	1.87	1.72	1.96	1.53	1.99	3.19	4.12	5.0	10.0
	Zn	29.76	51.12	33.21	56.94	24.14	53.67	62.83	128.49	150.0	200.0

$\overline{X}$ –average of the measurements; number of measurements, *n=5; SM*-dry matter*; C*c-critical concentration*; C*t-toxic concentration.

Adoption and accumulation of tested metals in plants depend to a great extent on the nature and the kinetics of enzymatic reactions, as well as on the concentration of metals in the soil [9–11, 13]. Based on the results shown (Table 2), the plants mostly accumulated zinc (as the essential element), where the concentration of Zn in the leaves is higher in relation to the root (observed for all measuring points), which is in correlation with the content of Zn in the soil and in consistency with the range of concentrations shown in the literature [9,33].

The plants easily adopt Zn and move it further into the above-ground organs. The process of adopting, moving and accumulating Zn in plants depends on a number of factors: soil pH, temperature [2,9], the presence of bicarbonate [34], phosphate [35], ion exchange capacity, organic matter content, redox conditions, chloride ion content [36]. In addition, Zn it helps the adoption of Cd, so that both plants: *Lactuca Sativa* and *Allium cepa* largely absorb Cd over the root and transport in overground organis [8]. The amount of Cd in the leaf of *Lactuca Sativa*, at the measuring site of Belo Brdo, is higher (5.38 µg g^{-1} SM) than the amount of Cd at the root (4.05 µg g^{-1} SM), which is in agreement with the results of other researchers that plants that have a developed leaf surface accumulate more Cd [8,9,13]. The concentration of Cd in the *Lactuca Sative* is higher in relation to the *Allium cepa* observed for all the analyzed soil samples. The adoption of Cd has a great influence on the concentration of Zn in the soil. At lower concentrations of Cd and Zn in the soil, plants (*Lactuca Sative* and *Allium cepa*) accumulate Cd more, and where the concentrations of Cd and Zn are larger, plants accumulate Zn more.Based on the results shown in Table 2, it is noted that the concentration of Pb higher at the root than in the leaf regarding both analysed plants. Plants adopt Pb most often in an inorganic form, where the degree of adoption is low as well as the mobility in the above-ground organisms [8–10,13]. Similar to Cd, it could be expected that the concentration of Pb is higher in the leaf than at the root of the plants with a developed leaf surface, which was not the case for the *Lactuca Sative*. The obtained results are in agreement with the fact that mobility lead increases with the increase of acidity of the soil (soil samples had pH values between 6.90 and 7.10) [37]. In addition, it has been observed that the concentration of metals in cultivated plants is positively correlated with the concentration of metals in the soil. The highest concentration of metals in *Lactuca Sative* and *Allium cepe* plants, both at the root and in the leaf, is found in samples grown on the soil from Belog Brda, and the smallest on samples of the plants grown on the soil from Guvnista. The results of determining the concentration of metals in leguminous plants (fodder plants) and grasses (weed plants) grown on the analysed soil are shown in Table 3.

Table 3. The results of determination of the concentration of metals in plants grown on the investigated soils

$\overline{X}$ μg g^{-1}*SM*					
		Mali Zvecan	Zitkovac	Guvniste	Belo Brdo
Lotus corniculatus L.	Pb	3.38	3.82	2.46	4.46
	Cd	/	/	/	/
	Zn	54.39	56.07	51.56	68.13
Trifolium pretense L.	Pb	1.71	3.51	1.50	3.63
	Cd	/	/	/	/
	Zn	68.15	69.54	66.7	70.41
*Cynodon dactylon*ea	Pb	54.94	68.56	41.03	**119.30**
	Cd	/	/	/	1.48
	Zn	60.71	96.88	58.94	114.78
Festucea arundinacea	Pb	21.61	27.05	16.25	47.24
	Cd	/	/	/	2.01
	Zn	86.41	137.88	84.07	163.36

$\overline{X}$ –average of the measurements; number of measurements, n=5

Determination of the concentration of Pb, Cd, and Zn in the plants (Table 3) was performed in the total biomass (leaf and root) in order to determine the total accumulation potential. Plants of *Lotus corniculatus* L. and *Trifolium pretense* L. represent legumes used as fodder plants. The amount of Pb and Zn in plants does not exceed the values of critical concentrations in plant tissue in which there may be up to 10% loss of biomass (10−20 μg g^{-1} Pb, 100−500 μg g^{-1} Zn), while for most of the analyzed samples, the amount of Cd ispod the maximum tolerable level for feeding animals (1 μg g^{-1}) [38], ie the analysis did not show the presence of Cd after planting of these plants.The amount of Zn in total biomass of *Trifolium pretense* L. was around 68.15−70.41 μg g^{-1} SM for all measuring points, which means that the total amount of Zn in the analyzed soil and the characteristics of the soil itself do not affect to the greatest extent on the sorption of this element. Pb was most accumulated in the *Cynodon dactylonea*L. in Belom Brdu (119.30 μg g^{-1} SM). In this plant has Zn 96.88 μg g^{-1} SM in Zitkovac and 114.78 μg g^{-1} SM in Belo Brdo.

The application of the *Festucea arundinacea* plant also significantly reduced the content of the analyzed metals in the soil. The remaining quantities of metal do not exceed the maximum permitted level except for Pb in Belo Brdo when the *Cynodon dactylone* plant was used.Obtained results for of these two plants, they demonstrate good Pb and Zn accumulation ability: *Cynodon dactylone* has a higher accumulation capacity according to Pb, while *Festucea arundinacea* for Zn. The amount of the metal in the soil was determined also after the removal of the plant material. The content of metal ions in the analyzed soil after planting, expressed in%, is shown in Table 4.

Table 4. The content of metal ions in the analyzed soil after planting, expressed in %

		%			
		Mali Zvecan	Zitkovac	Guvniste	Belo Brdo
Pb^{2+}	*Lactuca sativa*	89.23	89.76	89.48	90.12
	Allium cepa	91.24	90.86	91.58	91.30
	Lotus corniculatus	89.33	89.28	89.39	89.34
	Trifoliumpratense	89.52	89.49	89.38	89.31
	Cynodon dactylon	31.45	31.40	31.31	31.49
	Festucea arundinacea	70.21	70.17	70.19	70.32
Zn^{2+}	*Lactuca sativa*	88.18	88.18	88.79	80.75
	*Allium*cepa	87.19	87.12	86.89	87.31
	Lotus corniculatus	85.43	85.46	85.54	85.65
	Trifoliumpratense	85.23	85.59	85.87	85.92
	Cynodon dactylon	57.65	57.76	57.65	57.32
	Festucea arundinacea	39.41	39.28	40.37	38.98

Within this paper, the idea was to verify how the selected plant species influence the concentration of ions in the investigated metals in the soil. Six different plant species are selected, each of which has a different vegetation period (life span). Since at least the maximum vegetation time of one plant (*Latuca sativa*) is 120 days, the selected monitoring time for all plants is 4 months. During this period, the maximum amount of ion that a plant can adopt is not reached. Analyzed plants: *L. sativa*, *A. cepa* and leguminosa (*L. corniculatus* and *T. pratense*) adopted from 9 − 11% Pb, and 11 − 19% Zn. The species *C. dactylon* adopted about 69% Pb, and 43% Zn, while the species *F. arundinacea* adopted 39 − 40% Zn, and Pb and up to 70%. On a number of measuring points, the applied technique after the application of plants Cd was not detected.

Based on the content of the adopted metals, it can be concluded that the plants *C. dactylon* and *F. arundinacea* have the greatest ability to adopt the selected metals. As *C. dactylon* intensively adopts Zn, and *F. arundinacea*Pb, sowing their mixture on abandoned and non-agricultural soils where is high concentration of these metals, can lead to a decrease in their concentration. As far as weeds are concerned, it is recommended to remove these plants from the soil before classing. The smallest adoption capacity for all three metals was shown by *L. sativa* and *A. cepa.*

IV. CONCLUSION

The results shown in this study indicate that the concentration of ion Pb^{2+}, Cd^{2+} and Zn^{2+} in the analyzed soil samples increased at the measuring points Zitkovac and Belo Brdo. By applying plants, a reduction in the concentration of metal in the soil is achieved. Expressed in percentages, analyzed vegetable plants (*Latuca sativa* L. and *Allium cepa* L.) and legumes (*Lotus corniculatus* L. and *Trifolium pratense* L.) from the soil accumulated 9 − 11% Pb and 11 − 19% Zn. Species *Cyndon dactylon* (L.) *Pers.* it has adopted about 69% Pb and 43% Zn, while the species is *Festuca arundinaceae Schreb*. adopted 39 − 40% Zn and Pb up to 70%. On a number of measuring points, the applied technique after the application of plants Cd was not detected.

The obtained results indicate a high degree of tolerance of *Cyndon dactylon* and *Festuca arundinacea* plants and the possible application of these plants to reduce the concentration of metals in contaminated soil. Using of these plants for the observed period did not achieve the maximum amount of ion that one plant can adopt, because observation was not performed to saturation due to the vegetation period.

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Ljiljana M. Babincev "Original Scientific PaperAgricultural Land Of Rural Areas And Way Of Removing Heavy Metals"International Journal of Engineering Research and Applications (IJERA), vol. 8, no.10, 2018, pp81-87