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RESEARCH ARTICLE

RESEARCH OF THE RELATIONSHIP BETWEEN THE CONTENT OF HEAVY METALS IN SEDIMENT DUST AND AGRICULTURAL SOIL IN THE AREA SURROUNDING THE STEELWORKS ZENICA.

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Abstract

This paper contains analysis of the results after monitoring the content of heavy metals (Pb, Cd, Fe, Zn, Ni, Cr and Mn) in sediment dust and agricultural soil in nine locations surrounding steelworks Zenica from March to August in 2016. The aim of the paper is to analyze and assess connection of the content of heavy metals in sediment dust and in soil surrounding steelworks Zenica in order to rate soil contamination caused by heavy metals that have been emitted from metallurgic plants, and in order to evaluate the industrial emissions effect on the environment and human health. It has been established that the biggest sample number of sediment dust is being loaded by Nickel and Cadmium, and soil samples by Nickel and Manganese. By applying Pearson coefficient, it has been concluded that there is a strong relation between Lead and Zinc, and a medium strong relation between Iron in sediment dust and soil, while other analyzed metals do not show significant correlation in terms of statistics. This shows that Lead, Zinc and Iron in the soil are probably of techno-genic origin, and the other metals are of primarily geological origin. The correlation analysis of metal substance in soil and sediment dust in measuring areas has shown that there is a statistically significant correlation between Zinc in sediment dust and soil in the following areas: Gradišće, Arnauti and Novo Selo, and correlation of Chromium in sediment dust and soil is present in Šerići.

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Introduction:-

Soil in industrial areas is often exposed to the influence of increased emissions of different pollutants from industrial machines. Research has registered higher concentrations of heavy metals in the soil than those naturally present, and very often this concentration is even higher than limit, which leads to deterioration of physical, chemical and biological characteristics as well as violations of soil usable values.

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Also, the living environment surrounding the steelworks in Zenica, central Bosnia for the past decades has been exposed to the influence of the high emissions of heavy metals and other pollutants emitted from industrial plants. Monitoring results identified a higher content of heavy metals in agricultural soil and in populations of different plants, which is a consequence of more than a couple decades of the environment being loaded by the emissions from industrial plants [1, 2, 3, 4].

Heavy metals from industrial sources are being emitted in the air and they are included into anthropogenic redistribution in the geosphere. The atmospheric deposits of heavy metals reach the soil, where they are accumulated and stored for decades. Heavy metals from the soil are taken out by the harvest and are washed by water and in that way they are included in the geobiocycle and food networks, through which they can cause different consequences for all elements in food networks, including health consequences for the people. Harmful effects of heavy metals on ecosystems, especially on plants, are expected with higher concentration and are being reflected through different changes and disorders, which can reflect on human health [5]. Therefore, knowing all the factors affecting the redistribution of heavy metals in geosphere is of high significance for the protection of the environment, human health, and the stability of the eco-system.[5, 6, 7].

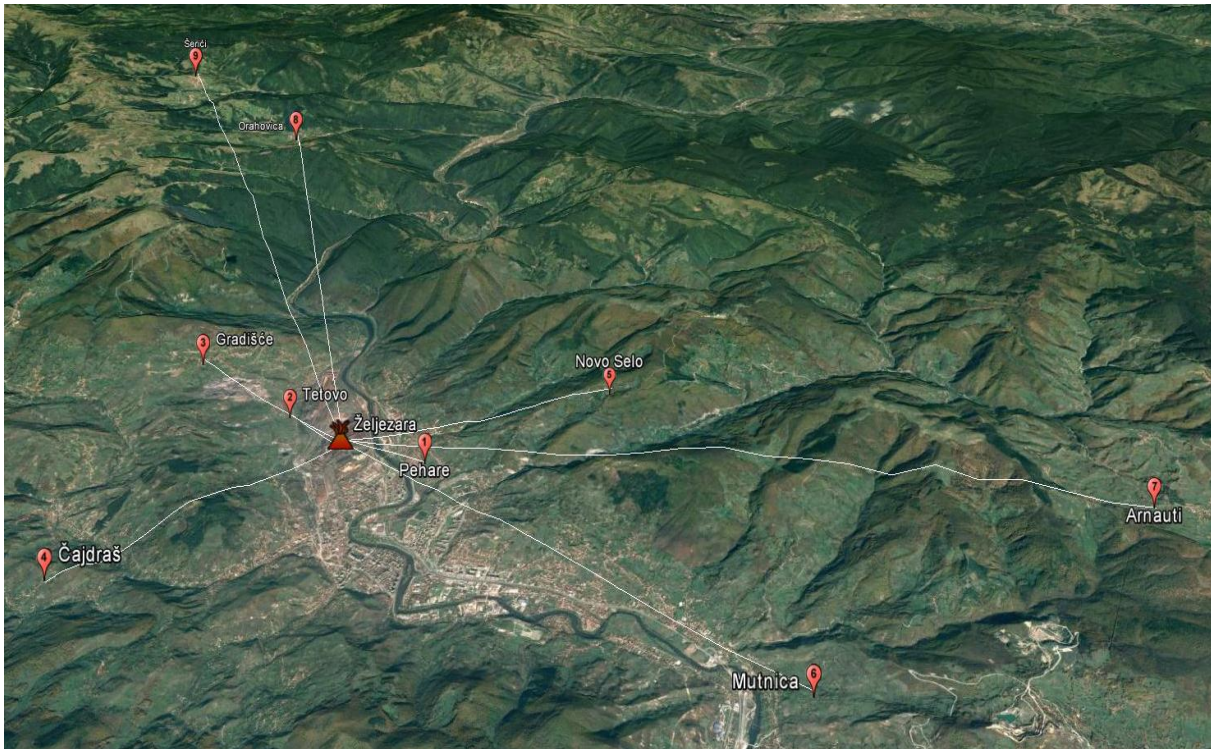
This paper contains analyzed results of heavy metals monitoring of (Lead-Pb, Cadmium-Cd, Iron-Fe, Zinc-Zn, Nickel-Ni, Chromium-Cr and Manganese-Mn) in sediment dust and agricultural soil surrounding steelworks Zenica. The aim of the analysis is to determine the origin of heavy metals in agricultural soil and the origin of soil contamination caused by heavy metals that have been emitted from industrial plants in order to assess the influence of industrial emissions to the environment and people's health.

Material and methods of work:-

Research has taken place in 9 locations (L) surrounding steelworks Zenica, in the Central Bosnia area. The locations are positioned in a cyclical scheme various distances (1,2 – 17,4 linear km) from industrial sources of emissions (Table 1 and Picture 1).

Table 1:-Distance of locations from industrial sources of pollutants emission

Location	Distance (km)	Latitude	Longitude	Altitude
Tetovo – L1	1,2	44 ⁰ 13'35" N	17 ⁰ 53'27" E	338 m
Pehare – L2	1,51	44 ⁰ 12'58" N	17 ⁰ 55'23" E	326 m
Gradišće - L3	3,18	44 ⁰ 14'13" N	17 ⁰ 52'8" E	528 m
Gornji Čajdraš - L4	4,41	44 ⁰ 11'16" N	17 ⁰ 51'49" E	520 m
Novo Selo - L5	4,13	44 ⁰ 13'34" N	17 ⁰ 57'60" E	696 m
Mutnica - L6	8,2	44 ⁰ 10'31" N	17 ⁰ 58'57" E	414 m
Arnauti - L7	11,7	44 ⁰ 12'3" N	18 ⁰ 2'37" E	680 m
Orahovica - L8	12,5	44 ⁰ 19'31" N	17 ⁰ 51'27" E	487 m
Šerići - L9	17,4	44 ⁰ 21'30" N	17 ⁰ 48'83" E	809 m



Picture 1:-Lookup map for locations for sampling of sediment dust and soil

Sampling of sediment dust has been done in nine locations surrounding steelworks Zenica in continuously for a 6 months period, starting from March to August 2016. Sampling has been done with Bergerhoff's device for collecting sediment dust, which consists of a sampling bowl for collecting sediment dust and a stool with wire net. The stool is positioned so that the surface of the bowl is in horizontal position 1, 5-2 m above the surface of the soil. Measuring of the sediment dust has been done according to standard method [8,9,10].

Soil sampling has been done on representative surfaces set in the same locations (Table 1). Soil sampling and preparation of the samples have been done according to standard procedure [11]. In each location, average soil samples have been collected (approximately 2 kg) from more (10-20) single samples taken with chrome-plated probe and with hand plastic tools. The samples are taken from the surface layer of 25 cm in depth of agricultural soil, which has been used for food production for humans and cattle.

According to mechanical composition, the researched soil in 5 locations (L-1, L-3, L-4, L-5 and L-9) has a powder-clay structure, and in 3 locations (L-2, L-6 and L-7) there are argillaceous soil and in location L-8 there is sandy soil.

The chemical analysis of the content of total shape of heavy metals in sediment dust and soil have been performed in the laboratory for the following elements: Pb, Cd, Fe, Zn, Ni, Cr and Mn according to AAS method (Perkin Elmer Analytical Methods for Atomic Absorption Spectrometry). The content of metal in the soil is expressed in mg/kg, and in sediment dust in %.

The correlation between the content of heavy metals in sediment dust and agricultural soil has been done using statistical tools in the Excel programme.

The Pearson coefficient of correlation is used to determine how close observations are to the regression line which best describes linear connection of heavy metals in sediment dust and the soil.

Table 2. Shows the interpretation of linear correlation coefficient, upon which a conclusion on intensity and character of a connection between questioned variables is made.

Table 2:- Criterium of intensity of linear connection between compared values expressed through Pearson correlation coefficient values

Correlation coefficient			Intensity of connection among variables
1			complete
$0,80 \leq$	r	< 1	strong
$0,50 \leq$	r	$< 0,8$	medium strong
$0,20 \leq$	r	$< 0,5$	weak
$0,0 \leq$	r	$< 0,2$	slight
0			completely absent

Connection intensity is the same for negative values of the correlation coefficient as well. For the determination of connectivity of chemical elements in sediment dust and agricultural soil, the correlation coefficients between elements (Table 8), and cluster analysis (Picture 2) have been used. The aim of the cluster analysis is to determine the the group of chemical elements (cluster) that shows that the elements inside the cluster are mutually alike more than the elements of the other cluster. Cluster hierarchy technique has been used in this analysis, which is based on correlation coefficient (r).

In order to assess values of total sediment dust and heavy metal concentration in sediment dust (Table 3), measured values are compared with limit values of polluting substance given in Annex XV: Limit and tolerant values for dedicated measuring according to [12].

Table 3:- Limit values for total sediment dust and heavy metals in sediment dust

Polluting substance	Period of sampling	Average annual value (mg/m ³ d)	High value (mg/m ³ d)
Sediment dust-total	one month	200	350*
Lead (Pb) in sediment dust	one month	0,1	-
Cadmium (Cd) in sediment dust	one month	0,002	-
Zinc (Zn) in sediment dust	one month	0,4	-
Nickel (Ni) in sediment dust	one month	0,015	-

***Note:** It refers to the month in a year with the highest values of precipitation disposition

Limit values of heavy metals in agricultural soil are shown in Table 4, which are determined according to the national standards [13].

Table 4:-Limit values of heavy metals in soil

Heavy metals (Total shape)	Limit values depending on soil texture (mg/kg soil)		
	Sandy soil	Powder-clay soil	Argillaceous soil
Lead (Pb)	50	80	100
Cadmium (Cd)	0,5	1,0	1,5
Nickel (Ni)	30	40	50
Chromium (Cr)	50	80	100
Zinc (Zn)	100	150	200

Limit values for Manganese and Iron, according to H.Resulović (Table 5). These limit values are presented in The Report on monitoring in the area of Municipality of Zenica for the period 2011-2015 [4].

Table 5:-Limit values for Manganese and Iron according to H.Resulović

Manganese (Mn)	1000 mg/kg of soil
Iron (Fe)	5%

Limit values of heavy metals in the soil (Tables 4 and 5) refer to the soil areas with acid reaction. In alkaline and carbonated soil areas, limit values can be increased by 25%.

Discussion and results:-

Content of heavy metal in sediment dust:-

According to the data presented in Table 6, we notice that in locations L-1 and L-3 average values of sediment dust are higher than limit value of 350mg/m²/day. The highest maximum value has been recorded in these locations, which was expected since they are exposed to the highest pressures of industrial emissions due to the proximity to anthropogenic sources of the emissions and to the directions of dominant winds.

Average values of Pb concentrations in sediment dust are lower than the limit value of 0,1mg/m²/day, but maximum concentrations are exceeding limit values in localities L-1, L2, L-7 and L-9, which are located in the direction of dominant winds.

Average concentrations of Cd in sediment dust in most locations (L-1,L-2,L-3,L-5, L-7 and L-9) are higher than the limit value of 0,002 mg/m²/day. Maximum concentrations of Cd in sediment dust are exceeding the proscribed limit value at almost all locations.

Average concentration of Zn in sediment dust in location L-1 (0,721 mg/m²/day) was higher than limit value of 0,4mg/m²/day. Maximum values of Zn in sediment dust were higher in the same location with the value of 1,089 mg/m²/day and in location L-5 with the value of 0,491mg/m²/day.

In all researched locations, values of Ni in sediment dust are higher than the limit value of 0,015 mg/m²/day have been recorded, and these varied from 0,016 – 0,107 mg/m²/day. Maximum values of Ni in sediment dust varied from 0,053 - 0,484 mg/m²/day.

For other heavy metals Fe, Cr and Mn, which are measured in sediment dust as well, the limit values are not proscribed nor does the criteria for their concentration assessment exist.

Table 6:-Average and maximum values of sediment dust and heavy metal concentrations in sediment dust (mg/m²/day)

Location	Sediment dust		Pb		Cd		Fe	
	Average	Max	Average	Max.	Average	Max.	Average	Max.
L1	664	828	0,0645	0,137	0,0041	0,0215	74,16	95,22
L2	279	369	0,042	0,112	0,0024	0,0051	34,03	65,75
L3	367	635	0,026	0,090	0,0038	0,0150	64,56	174,5
L4	198	264	0,035	0,082	0,0013	0,0055	7,705	10,59
L5	250	630	0,026	0,069	0,0035	0,0057	9,05	24,51
L6	203	274	0,024	0,088	0,00045	0,0019	8,56	13,25
L7	157	213	0,034	0,136	0,00285	0,0077	5,67	9,07
L8	188	298	0,0207	0,068	0,0018	0,0062	3,185	4,02
L9	350	595	0,027	0,123	0,006	0,0114	10,717	41,59
Limit value	350		0,1		0,002		-	

Table 6:-(Continued)

Location	Zn		Ni		Cr		Mn	
	Average	Max.	Average	Max.	Average	Max.	Average	Max.
L1	0,721	1,089	0,107	0,484	0,054	0,084	4,163	5,395
L2	0,196	0,267	0,019	0,055	0,019	0,055	1,865	4,915
L3	0,285	0,382	0,0365	0,053	0,035	0,099	2,606	6,754
L4	0,145	0,182	0,0325	0,130	0,009	0,025	0,318	0,378
L5	0,172	0,491	0,0275	0,113	0,020	0,101	0,429	1,600
L6	0,111	0,135	0,0295	0,079	0,015	0,063	1,394	4,723
L7	0,112	0,173	0,023	0,092	0,003	0,002	0,135	0,215
L8	0,072	0,085	0,016	0,078	0,003	0,011	0,107	0,162
L9	0,1525	0,381	0,039	0,173	0,0073	0,027	0,5615	2,037
Limit value	0,4		0,015		-		-	

Note: Bolded values and shadowed fields in the table indicate that the limit values are exceeded.

Content of heavy metals in soil:-

The content of heavy metals in agricultural soils shown in Table 7, where we can see the increased presence of heavy metals in the area surrounding steelworks Zenica, depending of the distance from the industrial sources, type of the soil and wind rose.

Average concentrations of Pb in the soil in zones up to 3,5 km surrounding steelworks Zenica (locations L-1,L-2 and L-3) were higher than the limit values shown in Table 4. Maximum concentrations of Pb in soil were higher than the limit values in five locations (L-1, L-2, L-3, L-8 and L-9).

Average concentrations of Cd in the soil were higher than the limit values in two of the locations (L-4 and L-7), while maximum concentrations of Cd were higher in most of the researched locations.

Average concentrations of Ni in soil were significantly higher than the limit values in almost all researched locations, which proves that the whole area is loaded with this pollutant.

Average concentrations of Cr in the soil were higher than the limit values in two locations (L-5 and L-7), and the maximum values were higher than the limit values in five locations (L-1,L-2, L-5, L-6 and L-7).

The highest concentrations of Zn in soil were recorded in zones up to 3,5 km surrounding steelworks Zenica (locations L-1, L-2 and L-3), and in location L-8 these were higher than limit values shown in Table 4.

In most locations the content of Mn in the soil was higher than the limit values shown is Table 5. Especially high concentrations have been recorded in location L-9, which is the farthest one from the industrial sources of emission to the air.

Analysis of heavy metals concentrations, in soil as shown in Table 7, clearly indicates that distribution of concentrations do not show a decreasing trend with moving from industrial sources of heavy metals emission. This is probably because of the origin of certain types of heavy metals, which is primarily geogenic, and secondarily anthropogenic, because of the configuration of the researched area and wind rose [4, 13]. The results of this research show increased values of certain heavy metals, especially of Mn, even in the farthest location tested.

Table 7:-Average and maximum values of heavy metals concentrations in soil (mg/kg)

Location	Pb		Cd		Fe (%)		Zn	
	Average	Max.	Average	Max.	Average	Max.	Average	Max.
L1	220	280	<0,1	<0,1	4,94	5,82	290	310
L2	92	110	0,9	3	3,98	4,17	247	260
L3	82	150	0,41	2	3,13	3,58	155	180
L4	53,3	80	1,23	6	2,95	3,46	113,3	140
L5	35	60	0,91	5	3,67	3,83	76,6	110
L6	53,3	70	0,41	2	3,64	5,12	108,3	160
L7	40	60	2,88	7	3,95	4,27	83,3	100
L8	48,5	70	0,1	0,1	2,34	2,61	103,3	120
L9	58,3	90	1,06	4	2,26	2,48	116,6	130

Table 8:- (Continued)

Location	Ni		Cr		Mn	
	Average	Max	Average	Max.	Average	Max.
L1	125	200	80	130	1717	2130
L2	122	170	92	140	1647	1890
L3	70	110	43	80	1213	1280
L4	62	130	40	80	743,3	830
L5	136,6	200	91,6	120	1376,6	1640
L6	125	180	90	130	873,3	920
L7	198,3	250	158,3	190	1203,3	1360
L8	50	70	15,16	30	3110	3500

L9	35	80	13,6	40	6561,6	8200
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Investigation of the relationship between heavy metals content in sediment dust and soil:-

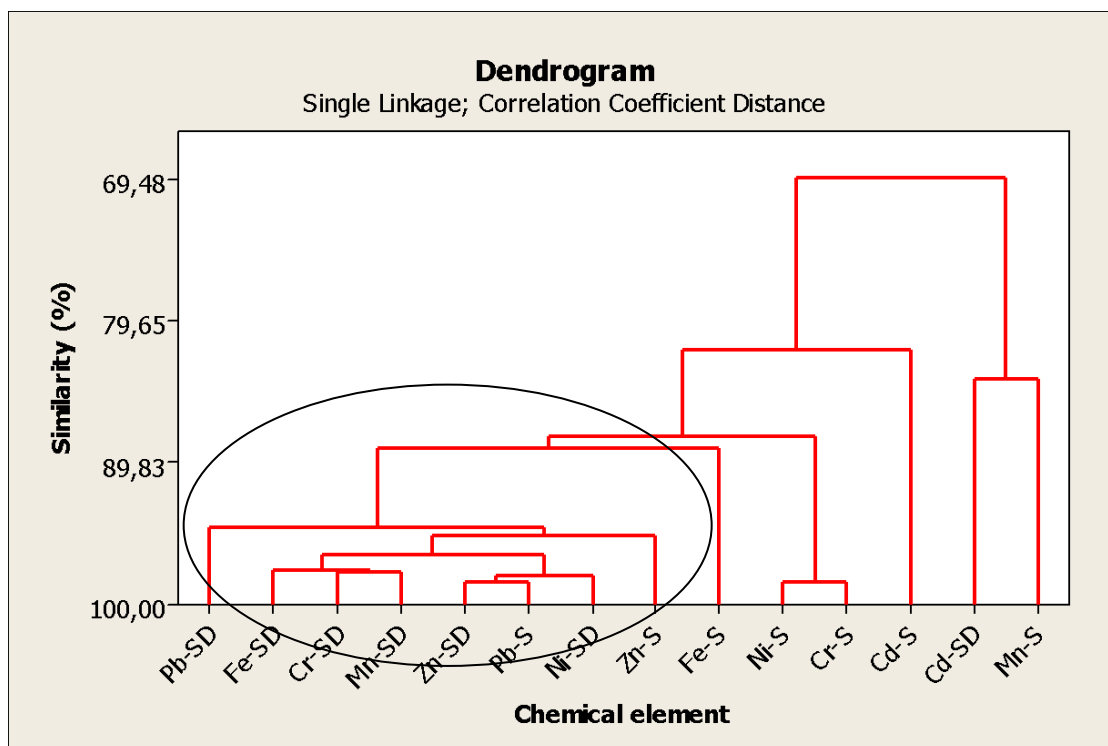
In Table 8 there is a survey of the correlation coefficient between heavy metals concentrations in sediment dust and soil.

Table 9:-Correlation coefficient between heavy metals concentrations in sediment dust and soil

	Pb-SD	Cd-SD	Fe-SD	Zn-SD	Ni-SD	Cr-SD	Mn-SD
Pb-S	0,891	0,284	0,837	0,967	0,920	0,867	0,906
Cd-S	-0,061	0,043	-0,430	-0,379	-0,342	-0,503	-0,508
Fe-S	0,777	-0,074	0,531	0,671	0,577	0,655	0,644
Zn-S	0,836	0,197	0,808	0,804	0,679	0,760	0,864
Ni-S	0,381	-0,251	0,066	0,147	0,093	0,099	0,151
Cr-S	0,282	-0,197	-0,039	0,047	-0,010	0,031	0,029
Mn-S	-0,193	0,676	-0,180	-0,112	0,009	-0,263	-0,204

Note: Dark fields point out strong connections between the same metals in sediment dust (SD) and soil (S)

According to the data presented in Table 8 there is a noticeably strong connection between Pb ($r=0,891$) and Zn ($r=0,804$) in sediment dust and soil, while in the connection between Fe in sediment dust and soil is of medium strength ($r=0,531$). There is a noticeable strong connection between Pb, Fe, Zn, Cr and Mn in sediment dust with Pb and Zn in the soil, as it has been shown in dendrogram in Picture 2. This connection means that these researched chemical elements probably have the same origin and they come from industrial sources of emissions in the air. Ni, Cr, Mn and Cd do not show significant mutual connection in the samples of sediment dust and soil collected in the researched area surrounding steelworks Zenica (Picture 2).



Picture 2:-Dendrogram of cluster analysis– connection of the elements in sediment dust (SD) and soil (S)

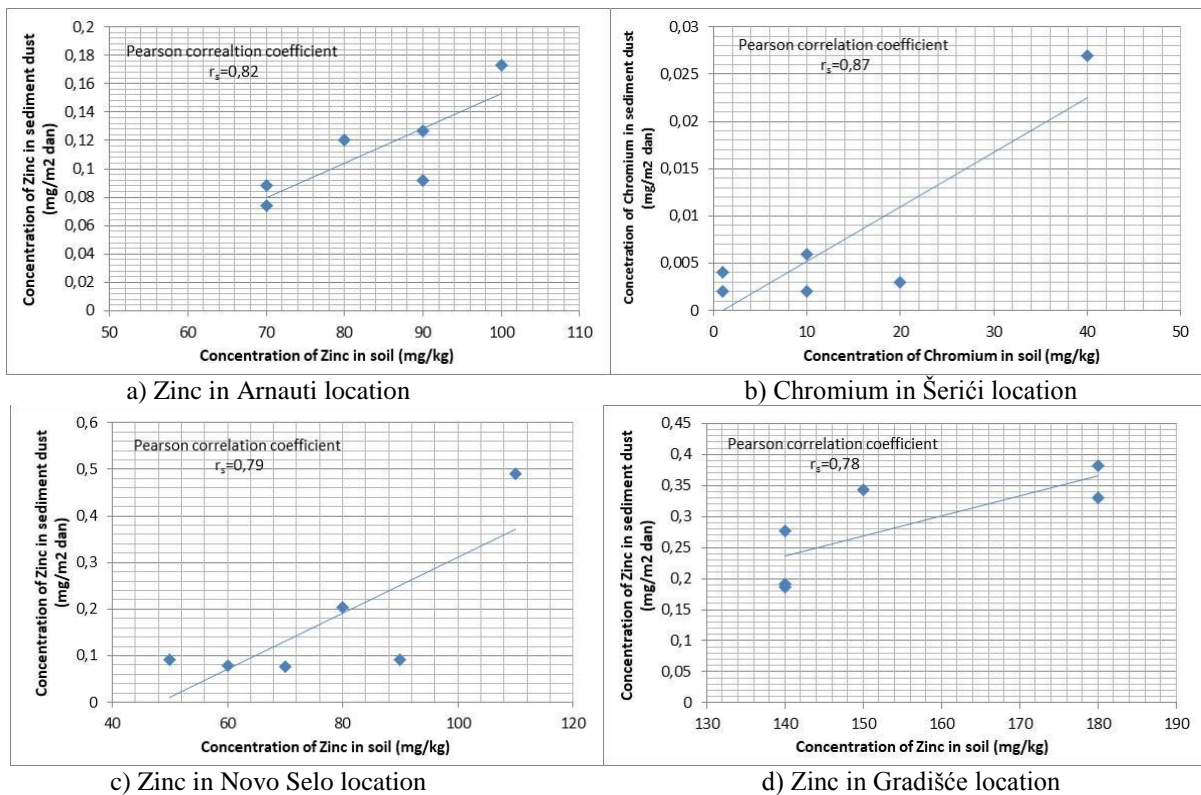
Correlation coefficients between heavy metals in sediment dust and soil in researched locations surrounding steelworks Zenica are presented in Table 9.

Table 10:-Pearson correlation coefficient of concentration of heavy metals in sediment dust and soil in researched locations during the period of March-August 2016.

LOCATION	Pearson correlation coefficient						
	Pb	Cd	Fe	Zn	Ni	Cr	Mn
L1	-0,42	0	0,42	-0,01	0,45	0,40	-0,42
L2	-0,42	0,13	0,45	0,10	-0,15	-0,05	-0,33
L3	-0,45	-0,27	-0,51	0,78	-0,85	0,69	-0,45
L4	0,25	0,09	-0,58	0,60	-0,15	-0,58	0,36
L5	-0,34	0,35	0,51	0,79	-0,18	0,65	-0,16
L6	0,05	-0,17	-0,55	0,04	0,15	-0,07	-0,02
L7	-0,28	-0,08	-0,40	0,82	-0,30	-0,59	0,01
L8	0,55	0,00	0,28	0,70	0,28	-0,62	0,00
L9	0,41	0,11	-0,14	0,53	-0,39	0,87	0,08
Average values for all locations	0,89	0,04	0,53	0,80	0,09	0,03	-0,20

Note:-Dark fields point out strong connections between heavy metal substance in sediment dust and soil

Statistical analysis of data, presented in Table 9, show that the correlation between heavy metal content in sediment dust and soil inside certain locations is weak (negative values are not taken into consideration),except for Zn in locations:L-3 in (Picture 3-d, $r_s=0,78$ – strong connection), L-5 (Picture 3-c, $r_s=0,79$ – middle strong connection) and L-7 (Picture 3-a $r_s=0,82$ – strong connection), and Cr in location L-9 in Picture 3-b, $r_s=0,87$ – strong connection). Therefore, statistically significant connections exist between the heavy metal content in the sediment dust and the soil for Zn in these locations: L-3, L-5 and L7, and for Cr in location L-9.



Picture 3:-Diagram showing dissipation of heavy metals in sediment dust and soil

Conclusion:-

According to the monitoring of in agricultural soil and sediment dust in the area surrounding steelworks Zenica, in central Bosnia, it was found that a large number of the samples of sediment dust were loaded with Ni (25 samples) and Cd (18 samples), with a total number of 54 samples collected. In addition, a large number of the soil samples were loaded with Ni (47 samples) and Mn (42 samples), with a total of 54 samples obtained.

Heavy metal content in sediment dust and agricultural soil in the area surrounding steelworks Zenica shows increase levels compared to the natural state and goes beyond the maximum limit values allowed, which is a consequence of their anthropogenic redistribution caused by present emissions of heavy metals from metallurgic and thermal plants. Application of Pearson correlation coefficient shows strong connection between Pb and Zn as well as a medium level connection between Fe in sediment dust and soil. Other elements that have been analyzed do not show significant statistical interrelation. This shows that increased content of Pb, Zn and Fe in soil is probably of technogenic origin because of industrial emissions of these elements.

Correlation analysis between the content of metals in soil and sediment dust in these researched locations has shown that there is a statistically significant correlation between Zn in sediment dust and soil in following locations: Gradišće, Arnauti and Novo Selo, and of Cr in sediment dust and soil in location Šerići.

The results of this research of the content and dynamics of heavy metals in agricultural soil of Zenica region show that the soil quality is endangered and is risky for food production because of the heavy metal inclusion in food networks and the potential negative impact on human health.

Since this analysis has shown high variation of the results and high number of negative results, it is necessary, in further research, to lower the number of measuring places, increase the number of elements analyzed, and increase number of samples for an extended research period. In this way, we would get more representative data, which would provide better correlation between the content of heavy metals in sediment dust and soil. Based on such detailed examination it is possible to give recommendations for applying suitable technology and measures for remediation of the contaminated soil (e.g. phytoremediation or calcination or adding humus to the soil, etc.).

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