Romanian Reports in Physics **XX**, XYZ (2018)

SPATIAL DISTRIBUTION OF HEAVY METALS IN URBAN SOILS

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Received January 17, 2018

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Abstract. Heavy metals in the surface soils from 40 points of Targoviste City, were analyzed to assess the industrial and anthropogenic impact on soil pollution. Approximately 700 surface soil samples were collected and analyzed for major heavy metals including Mn, Fe, Ni, Cu, Zn, As, Cd and Pb by using microwave-assisted digestion and inductively coupled plasma–mass spectrometry (ICP-MS). The results indicate that Fe, Cu, Zn, Pb and Cd in the surface soils were primarily derived from industrial and anthropogenic sources, while As content in the surface soils were controlled by both natural and anthropogenic sources. Approximately 10% of the soil samples were polluted from moderately to heavily by these elements.

Key words: heavy metal, ICP-MS, spatial distribution, urban soil.

1. INTRODUCTION

Soil pollution with heavy metals, represents a significant worldwide problem due to the complexity raised by this phenomena. The most known heavy metals are not found to be soluble in water or, if they really exist, the chemical species are complexed with organic or inorganic ligands, which increase their toxicity [1-5]. Heavy metals are naturally occurring elements which are found throughout the earth's crust, in different concentrations, from ppm to ppb (which means that range from ppb to less than 10 ppm) [5, 6]. The most usually toxic elements found in contaminated sites are lead, chromium, cadmium, mercury, nickel, arsenic, zinc, manganese and copper. It is well known that their bioavailability is influenced by physical factors (e.g. temperature, adsorption, phase association and isolation), chemical factors, which influence the speciation and complexation processes, as well as biological factors (e.g. characteristics of species, biochemical process, and trophic interactions) [7-11]. Several studies have shown the fact that heavy metals in soil may pose risks and hazards to humans [12-16] or to ecosystem [17-19], as well. Systematic toxic pollutants, including heavy metals, exert their effects on human body, the results being specific to the substance in question [19]. Their spreading into the environment is increasing and, the fact that are going to be accumulated in the environment or in the human body, as well adding the high risks of serious diseases occurrence possibility is quite disturbing [14, 15]. Contaminants found in different vegetables [20-25] or plants [26-30] grown in urban area are usually derived from previously contaminated soil or atmospheric pollution.

The purpose of this study was to determine the concentrations and spatial distribution of heavy metals (e.g. Mn, Fe, Ni, Cu, Zn, As, Cd, and Pb), to assess the heavy metal contamination in soils of Targoviste City in order to identify the potential risk of heavy metal intake by the urban population.

2. MATERIALS AND METHODS

2.1. SITE DESCRIPTION AND SAMPLING

Targoviste City is situated in the middle of Dambovita County, between two relief stages, one representing the plain area and the other, the hill region at approximately 260 meters above sea level. It is crossed by the parallel of 44°56‘ and the meridian of 25°26’. On a distance of about 18 km, in the north-west direction to the southeast, the city is crossed by the Ialomita River [31]. The climate is temperate - continental and offers gentle winters due to the hills that surrounds the city and deviates the path of cold air, and also, cold summers with moderate precipitation [32, 33]. The soils from Targoviste area are brownish-red argiloiluvial soils with a humus horizon of 20-40 centimeters, which offer good fertility for the crops plants. This area is characterized by a moderate pollution due to urbanization and industrial activities, especially in the southern area of the city. As mineral resources, can be mentioned [34, 35]: the gravel and sands in a highly alluvial area, the oil and gas, wells exploited by the Targoviste Petroleum Schele and coal (lignite) extracted near to the city.

This study was carried out in two years, 2015 and 2016, from early spring to late autumn on 40 sampling points. These points have been distributed in such a way that will ensure a uniform distribution on the drawn maps (Figure 1).

Approximately 700 samples were collected and analyzed during 2015 and 2016. Soil samples were collected in repeated points from the urban surface using a disposable plastic scalpel. Sampling procedure was in according with Romanian legislation and the points were located by GPS (Global Position System).

Fig. 1 – Sampling map of urban studied area.

2.2. CHEMICALS, SAMPLE PREPARATION AND ANALYTICAL TECHNIQUE

All reagents, including nitric acid (Merck), hydrochloric acid (Aldrich), and sulphuric acid (Aldrich) were analytical reagent grade. Ultrapure water (Thermo Fisher Scientific, Germany) was used for standard solutions preparation and blank, as well.

The soil samples were dried at 105°C for 48 h. To avoid possible losses of arsenic as volatile element, the drying process was performed on separate portions. The dried material was grinded in an agate ball-grinder in order to ensure uniformity of chemical composition throughout the mass of the sample. To improve the sample homogeneity and to achieve a final particle size distribution, each sample was additional grinded and carefully sieved through a 100 μm sieve.

Approximately 0.5 g from each sample was introduced in the digestion Teflon vessels of TopWave (Analytik Jena) microwave digestion system and then, in according to EPA method 3052, each soil sample was mineralized. After digestion, the vessels were cooled for 30 minutes at room temperature. The obtained solutions were filtered and brought with deionized double-distilled water to 50 mL graduated flasks. The concentrations of heavy metals of final solutions were determined by an iCAP Qc Inductively Coupled Plasma-Mass Spectrometer (Thermo Scientific, USA). The measurements were performed in triplicate in the standard mode (STD) and the quality control of ICP-MS measurements was provided by using standard reference material SRM 2711a Montana Soil (National Institute of Standards and Technology-NIST). The standard reference sample was prepared using the same sample procedure and the ICP-MS measurements were performed respecting the same parameters. The recovery rates for the analyzed heavy metals were reasonably good (77–109 %). The relative standard deviation (RSD) values were less than 10% (Tables 2 and 3). The data were expressed as mg/kg dried weight (d.w.) material.

3. RESULTS AND DISCUSSION

The soil chemistry highlights heavy metals as a special group of elements due to their toxic effect on peoples and plants upon their concentrations exceed the admitted limit. In this respect, the mean concentrations of investigated elements including manganese, iron, nickel, copper, zinc, arsenic, cadmium and lead in surface soil samples from Targoviste City, collected in 2015 and 2016, are presented in Tables 1 and 2.

The mean value for Mn in surface soils sampled in the urban sites was 547.89 mg/kg with a range from 177.22 to 1,036.52 mg/kg, in 2015 and 314.99 mg/kg with a range from 164.40 to 872.90 mg/kg, in 2016. The maximum values for Mn was obtained in samples collected near industrial area of Targoviste City (Figure 2 and Tables 1 and 2). The mean concentration of Cu in urban surface soils ranges from 10.11 to 404.92 mg/kg with a mean value of 125.02 mg/kg (Table 1), in 2015, and from 8.70 to 364.46 mg/kg with a mean value of 118.36 mg/kg (Table 2), in 2016. The mean value for Zn in selected points, in 2015, was 1,072.92 mg/kg with a range from 631.24 -1,721 mg/kg was slightly higher than mean value 958.81 mg/kg of the year 2016, with a range from 679.07 to 1,379.18 mg/kg. The mean value for Pb in the collected points, in 2015, was 12.59 mg/kg with a range of 1.35 (in residential area) to 56.17 mg/kg, compared to the mean value of the year 2016, it can see approximately same values 11.59 mg/kg ranged from 1.16 to 50.24 mg/kg. Average As concentration in soil samples was similar, with lower differences, such as 27.23 mg/kg varied from 14.42 to 39.36 mg/kg, for year 2015, and 24.58 mg/kg with a range of 15.91 to 31.54 mg/kg, for the year 2016. The average Cd concentrations in soil samples in 2015 was 2.51 mg/kg ranged from 0.47 to 6.69 mg/kg and in 2016 and in 2016 was 2.12 mg/kg with a range from a minimum 0.51 to 5.94 mg/kg as maximum value obtained in industrial area. The mean value for Ni in surface soils sampled in the urban sites for the years 2015 and 2016 was 19.16 mg/kg and 17.95 mg/kg, respectively.

Surfer 9.0 software were used for drawn the heavy metal distribution maps of surface soil in Targoviste City (Figures 2-9).

Table 1.

Mean concentration [mg/kg d.w.] of metals in soil samples collected in 2015.

Sample	Mn	Fe	Ni	Cu	Zn	As	Cd	Pb
ST_1'	470.65	7,098.79	19.23	138.84	1,553.34	28.20	4.77	15.90
ST_2'	218.39	6,875.57	4.47	57.30	1,132.97	23.03	1.55	3.63
ST_3'	263.39	3,502.38	50.49	212.77	1,011.89	20.81	1.96	36.00
ST_4'	742.00	7,080.60	11.22	72.31	1,041.69	26.65	1.71	15.60
ST_5'	546.18	9,102.86	1.22	15.70	813.92	31.76	0.69	2.83
ST_6'	359.76	7,560.23	0.98	15.74	781.67	26.51	0.59	1.38
ST_7'	367.28	6,880.92	7.78	66.40	956.23	27.76	1.64	7.50
ST_8'	862.66	8,669.04	2.20	27.19	1,064.84	32.51	1.78	3.35
ST_9'	1,036.52	9,618.45	4.66	35.19	1,001.57	34.95	1.28	12.79
ST_10'	934.69	10,826.48	34.37	137.19	1,721.22	39.36	4.43	46.01
ST_11'	898.96	5,979.93	43.39	*id**	1,498.53	33.71	3.48	31.93
ST_12'	857.81	6,374.07	22.07	158.74	1,351.66	27.32	3.50	15.15
ST_13'	696.76	1,253.81	106.15	387.64	1,526.19	28.55	6.69	47.57
ST_14'	698.14	2,687.79	94.74	320.84	1,534.02	27.78	6.16	56.17
ST_15'	719.81	491.62	56.69	404.92	1,206.11	25.16	5.10	10.50
ST_16'	525.77	5,944.42	34.42	216.08	876.30	29.07	1.43	21.26
ST_17'	475.59	9,145.88	7.34	87.88	1,066.52	36.12	0.89	5.30
ST_18'	456.04	5,975.43	2.73	42.67	1,248.83	22.16	3.94	2.22
ST_19'	773.79	7,410.61	6.62	77.60	931.12	24.65	1.48	4.50
ST_20'	514.35	9,339.46	0.45	*id**	799.27	31.53	0.60	1.95
ST_21'	367.19	8,370.88	0.56	10.11	892.20	31.89	1.13	1.35
ST_22'	981.14	3,774.20	15.05	160.07	978.18	17.89	4.51	5.13
ST_23'	831.60	4,770.39	15.32	188.80	1,262.97	22.53	2.51	3.71
ST_24'	177.72	7,157.40	7.22	70.20	984.01	24.63	2.01	4.99
ST_25'	444.22	6,788.23	6.15	*id**	860.84	23.61	1.83	4.13
ST_26'	332.30	4,946.24	33.10	231.01	1,088.46	29.82	0.96	14.29
ST_27'	409.39	6,447.12	16.24	86.25	891.93	28.93	1.10	23.58
ST_28'	407.53	6,151.41	9.29	98.22	929.28	24.43	1.56	4.90
ST_29'	328.72	6,845.36	4.02	53.59	1,002.88	27.79	1.53	2.61
ST_30'	325.26	3,672.20	30.58	244.25	1,128.15	26.82	5.50	12.44
ST_31'	222.14	6,735.10	6.09	53.51	1,105.90	23.27	1.42	7.42
ST_32'	756.00	6,013.83	39.76	188.87	866.68	30.21	1.65	33.32
ST_33'	531.76	8,055.19	1.16	16.12	775.19	30.95	0.85	1.82
ST_34'	448.27	5,169.22	16.34	*id**	963.53	31.25	1.32	6.94
ST_35'	284.75	3,393.61	4.96	75.39	631.24	14.42	0.47	1.66
ST_36'	710.19	4,167.85	9.79	105.01	832.61	17.58	3.44	4.37
ST_37'	352.12	5,042.46	12.03	120.98	1,010.65	21.83	3.45	6.30
ST_38'	444.37	6,973.67	9.07	77.92	1,229.76	30.17	4.34	9.02
ST_39'	643.91	5,537.54	10.37	87.21	1,141.93	27.19	2.89	8.24
ST_40'	498.55	6,853.58	7.90	86.72	1,222.39	26.31	4.11	5.81
Average value	547.89	6,217.10	19.16	125.02	1,072.92	27.23	2.51	12.59
Minimum value	177.72	491.62	0.45	10.11	631.24	14.42	0.47	1.35
Maximum value	1,036.52	10,826.48	106.15	404.92	1,721.22	39.36	6.69	56.17

RSD [%]	0.01-3.11	0.01-3.40	0.01-4.46	0.01-5.35	0.01-4.89	0.01-4.51	0.01-4.59	0.01-3.76

id - indeterminate*

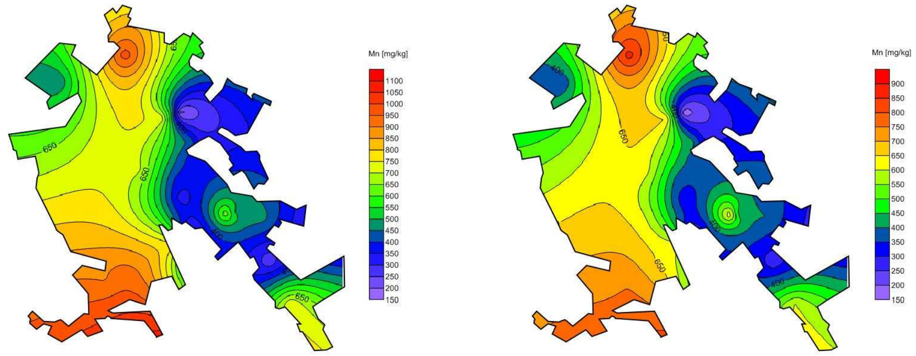
Table 2.

Mean concentration [mg/kg d.w.] of metals in soil samples collected in 2016.

Sample	Mn	Fe	Ni	Cu	Zn	As	Cd	Pb
ST_1"	400.55	6,041.52	16.37	118.16	1,321.99	24.00	4.06	13.53
ST_2"	219.46	6,876.64	5.54	58.37	1,134.04	24.10	2.62	4.70
ST_3"	268.22	3,566.58	51.42	216.67	1,030.44	21.20	1.99	36.66
ST_4"	602.27	5,747.24	9.11	58.69	845.53	21.63	1.39	12.66
ST_5"	526.69	8,778.07	1.18	15.14	784.88	30.62	0.66	2.73
ST_6"	356.55	7,492.79	0.97	15.60	774.70	26.27	0.59	1.37
ST_7"	362.21	6,785.92	7.67	65.48	943.03	27.38	1.62	7.40
ST_8"	712.35	7,158.58	1.82	22.45	879.31	26.84	1.47	2.77
ST_9"	788.23	7,314.41	3.54	26.76	761.65	26.58	0.97	9.73
ST_10"	748.95	8,675.06	27.54	109.93	1,379.18	31.54	3.55	36.87
ST_11"	722.64	4,807.02	34.88	*id**	1,204.61	27.10	2.80	25.67
ST_12"	655.32	4,869.42	16.86	121.27	1,032.59	20.87	2.68	11.57
ST_13"	618.24	1,112.52	94.19	343.96	1,354.21	25.33	5.94	42.21
ST_14"	624.45	2,404.11	84.74	286.98	1,372.11	24.85	5.51	50.24
ST_15"	647.89	442.51	51.03	364.46	1,085.61	22.64	4.59	9.45
ST_16"	427.80	4,836.79	28.01	175.82	713.02	23.65	1.17	17.30
ST_17"	363.60	6,992.26	5.61	67.19	815.38	27.62	0.68	4.05
ST_18"	356.00	4,664.66	2.13	33.31	974.89	17.30	3.08	1.73
ST_19"	700.90	6,712.51	6.00	70.29	843.41	22.33	1.34	4.08
ST_20"	437.00	7,934.97	0.38	*id**	679.07	26.79	0.51	1.66
ST_21"	316.00	7,203.85	0.48	8.70	767.81	27.45	0.98	1.16
ST_22"	872.90	3,357.83	13.39	142.41	870.27	15.91	4.01	4.56
ST_23"	825.00	4,732.53	15.20	187.30	1,252.95	22.35	2.49	3.68
ST_24"	164.40	6,621.09	6.68	64.94	910.28	22.78	1.86	4.62
ST_25"	414.00	6,326.41	5.73	*id**	802.27	22.00	1.71	3.85
ST_26"	304.30	4,529.52	30.31	211.55	996.76	27.31	0.88	13.09
ST_27"	399.40	6,289.87	15.84	84.15	870.18	28.23	1.08	23.00
ST_28"	872.90	3,357.83	13.39	142.41	870.27	15.91	4.01	4.56
ST_29"	825.00	4,732.53	15.20	187.30	1,252.95	22.35	2.49	3.68
ST_30"	164.40	6,621.09	6.68	64.94	910.28	22.78	1.86	4.62
ST_31"	414.00	6,326.41	5.73	*id**	802.27	22.00	1.71	3.85
ST_32"	304.30	4,529.52	30.31	211.55	996.76	27.31	0.88	13.09
ST_33"	399.40	6,289.87	15.84	84.15	870.18	28.23	1.08	23.00
ST_34"	384.10	5,797.74	8.76	92.57	875.85	23.03	1.47	4.62
ST_35"	318.22	6,626.68	3.89	51.88	970.84	26.90	1.48	2.53
ST_36"	293.82	3,317.25	27.62	220.64	1,019.11	24.23	4.97	11.24
ST_37"	202.50	6,139.56	5.55	48.78	1,008.11	21.21	1.30	6.76
ST_38"	611.16	4,861.63	32.14	152.68	700.63	24.42	1.33	26.94
ST_39"	531.76	8,055.19	1.16	16.12	775.19	30.95	0.85	1.82
ST_40"	418.55	4,826.54	15.26	*id**	899.65	29.18	1.23	6.48
Average value	489.39	5,593.91	17.95	118.36	958.81	24.58	2.12	11.59
Minimum value	164.40	442.51	0.38	8.70	679.07	15.91	0.51	1.16
Maximum value	872.90	8,778.07	94.19	364.46	1,379.18	31.54	5.94	50.24

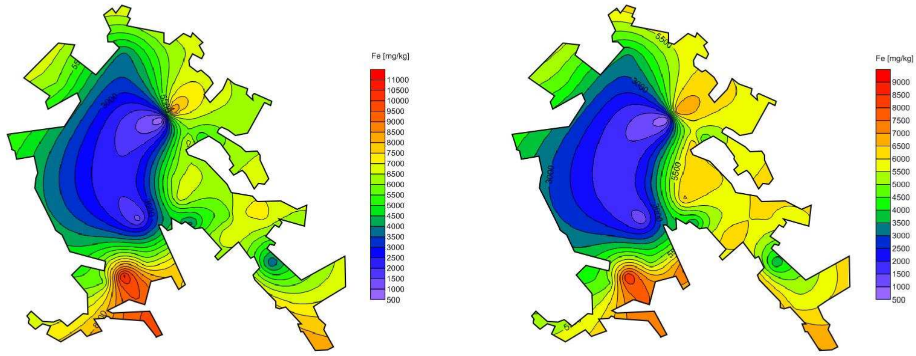
RSD [%]	0.01-3.11	0.01-3.40	0.01-4.46	0.01-5.35	0.01-4.89	0.01-4.51	0.01-4.59	0.01-3.76

id - indeterminate*



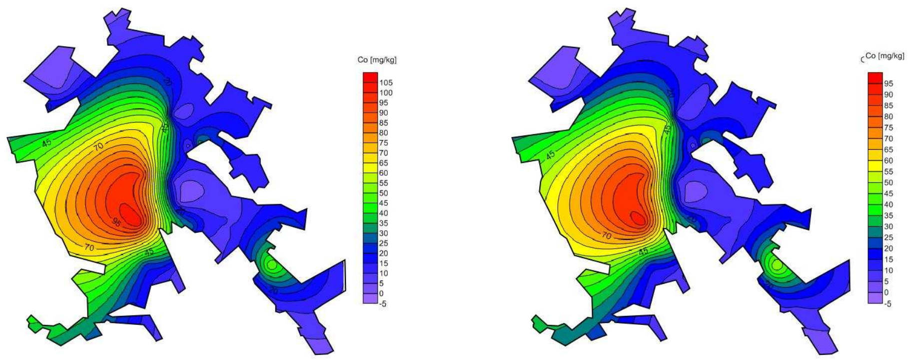
2015 2016

Fig. 2 – Spatial distribution of manganese level at surface soil of Targoviste City.



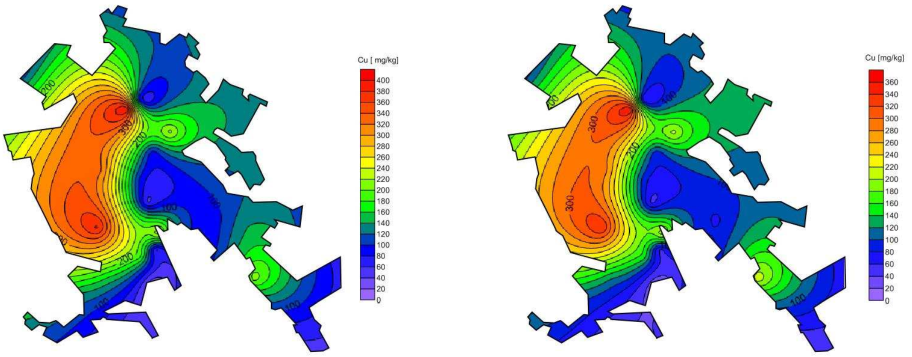
2015 2016

Fig. 3 – Spatial distribution of iron level at surface soil of Targoviste City.



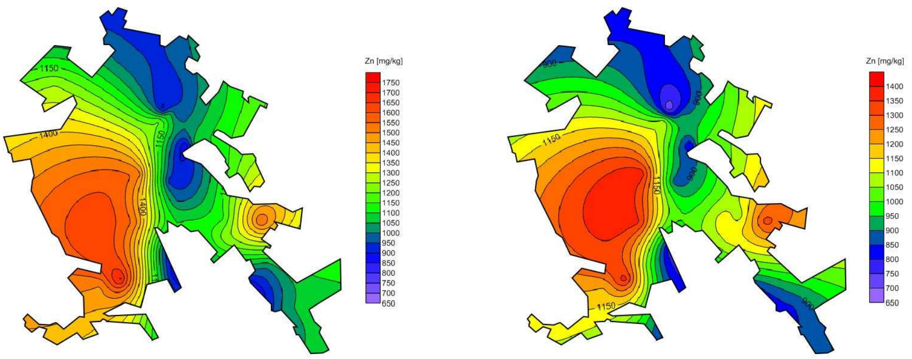
2015 2016

Fig. 4 – Spatial distribution of nickel level for Targoviste City surface soil.



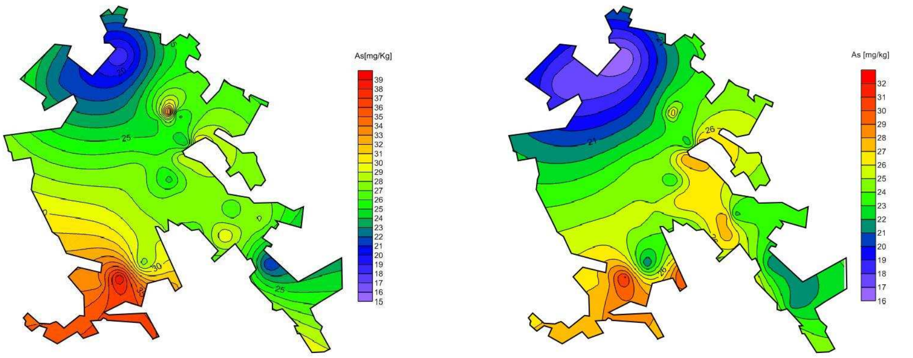
2015 2016

Fig. 5 – Spatial distribution of cooper level for Targoviste City surface soil.



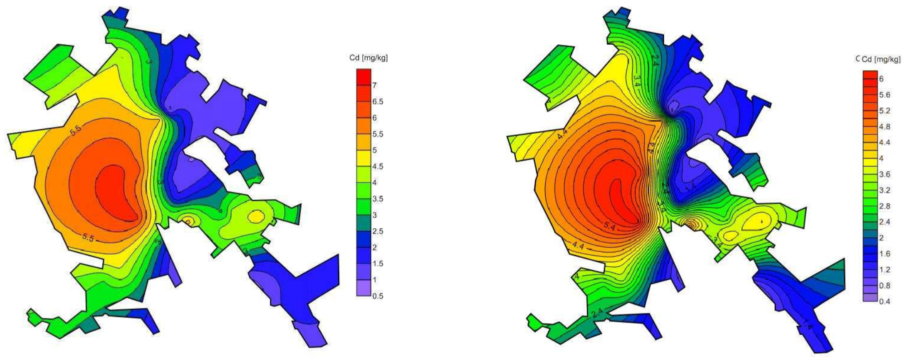
2015 2016

Fig. 6 – Spatial distribution of zinc level at surface soil of Targoviste City.



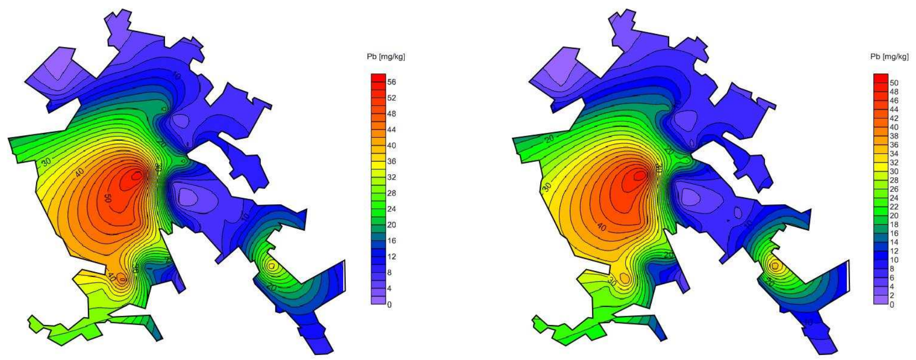
2015 2016

Fig. 7 – Spatial distribution of arsenic level for Targoviste City surface soil.



2015 2016

Fig. 8 – Spatial distribution of cadmium level for Targoviste City surface soil.



2015 2016

Fig. 9 – Spatial distribution of lead level for Targoviste City surface soil.

Figure 2 shows that manganese distribution is the same in 2016 as it was in 2015. The spreading is mostly noticed in north and also in industrial area of Targoviste City. Figure 3 shows small changes in the iron distribution from one year to another, so in 2016, the iron spreading was higher than the same period of 2016. The areas were higher concentrations were observed are the industrial area of the city and the east side. In 2016 is observed a little more pronounced iron spread on the eastern side of the city. Figure 4 shows that the distribution of cobalt does not change from one year to another so that in 2015, compared to 2016, the area of spread of cobalt was concentrated in the central-western area of the city. Figure 5 shows that copper distribution is the same in 2015 as it is in 2016, the spreading area was concentrated in the central-western area of the city. Figure 6 shows that zinc distribution does not change from one study year to another. Zinc spreading was concentrated in the central-western area of the city. Figure 7 shows that the distribution of arsenic changes very little from one study year to another, therefore in 2015 compared to 2016, the arsenic was concentrated on the industrial area and also in the north of the city. In 2016, there is a slight increase of arsenic in the northern part of the city, but also in the east side and a reduction of spreading on the industrial area. From Figure 8, it can be observed that the cadmium distribution does not change and the spreading area is concentrated in the central-western side of the city. Figure 9 shows that the distribution of lead does not change in 2016 and that lead is concentrated on the central-western side of the city.

When compared with the maximum levels of these heavy metals in Romanian legislations (Table 3), Fe, and Zn showed maximum concentrations more than two times higher than the maximum threshold, for two years of soil monitoring, suggested that the surface soils of Targoviste City had been polluted by both industrial and anthropogenic sources.

Table 3.

Concentration [mg/kg d.w.] of metals in soil according with Romanian and European legislations

Elements		**Mn**	**Fe**	**Ni**	**Cu**	**Zn**	**As**	**Cd**	**Pb**
Ord. 756/1997	Normal value	900.00	3,000.00	20.00	20.00	100.00	5.00	1.00	20.00
	Maximum threshold	2,000.00	4,500.00	200.00	250.00	700.00	25.00	5.00	250.00
	Intervention threshold	4,000.00	7,000.00	500.00	500.00	1,500.00	50.00	10.00	1,000.00
European average value*		650.00	35,100.00	37.30	13.00	52.00	7.03	0.145	22.60

**Geochemical Atlas of Europe [36]*

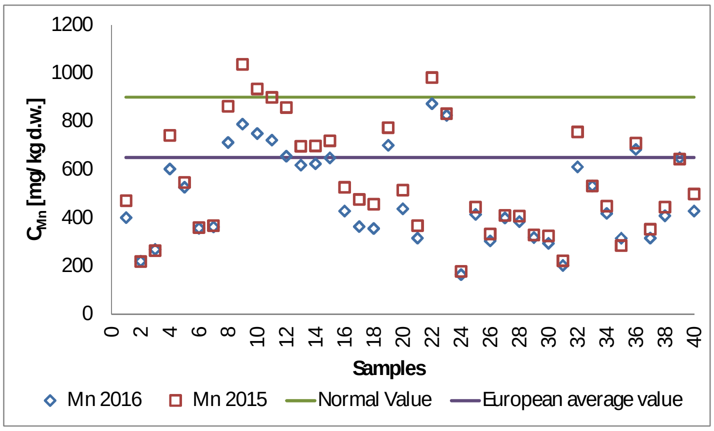


Fig. 10 – Comparison between the values of manganese concentration from surface soil samples collected in Targoviste City and the values provided by the legislation.

Meanwhile, the mean concentrations of Cu, Zn, Cd and As in urban soils were higher than European average value (Table 3) but comparable with other studies [37-39]. Based on these criteria it can concluded that approximately 10% of the soil samples were moderately or heavily polluted by Fe, Cu, Zn, Pb, Cd and As, respectively.

Figure 10 shows the quantitative distribution of manganese in the surface soil samples, compared with values admitted by Romanian and European legislations. The concentration of manganese in the surface soil samples of the studied areas recorded average values close to the European average and the normal value admitted by the Romanian legislation, but well below the maximum threshold (Table 3).

Figure 11 shows the quantitative distribution of iron in the surface soil samples, compared with the values provided in Romanian and European legislations (Table 3).

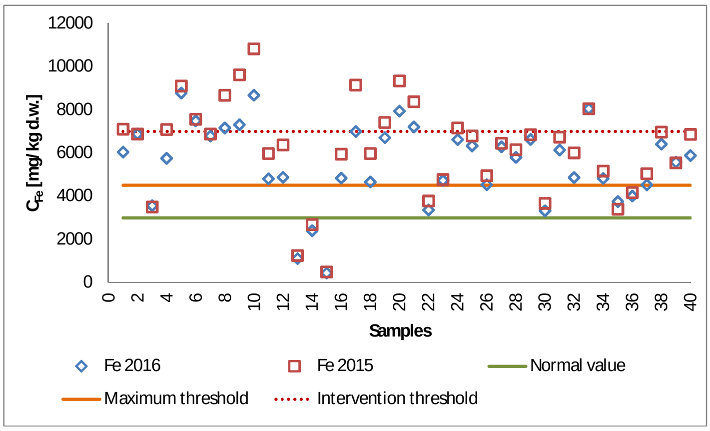


Fig. 11 – Comparison between the values of iron concentration from surface soil samples collected in Targoviste City and the values provided by the legislation.

The iron concentration in the surface soil samples of the studied areas recorded values well below the European average but also with a maximum value that exceeds the intervention threshold regulated by Romanian legislation (Table 3), with a minimum of 491.62 mg/kg; an average of 6,217.10 mg/kg and a maximum of 10,826.48 mg/kg in 2015 and a minimum of 442.51 mg/kg; an average of 5,551.40 mg/kg and a maximum of 8,778.07 mg/kg in 2016.

Figure 12 shows the quantitative distribution of nickel in the surface soil samples, compared with the values from Romanian and European regulation. The concentration of nickel in the surface soil samples of the studied areas recorded average and maximum values well above the European average but within the limits regulated by the legislation from Romania, with a minimum of 0.45 mg/kg in residential area and a maximum of 106.15 mg/kg in points of industrial area, in 2015, and a minimum of 0.38 mg/kg and a maximum of 94.19 mg/kg in 2016 in the same points.

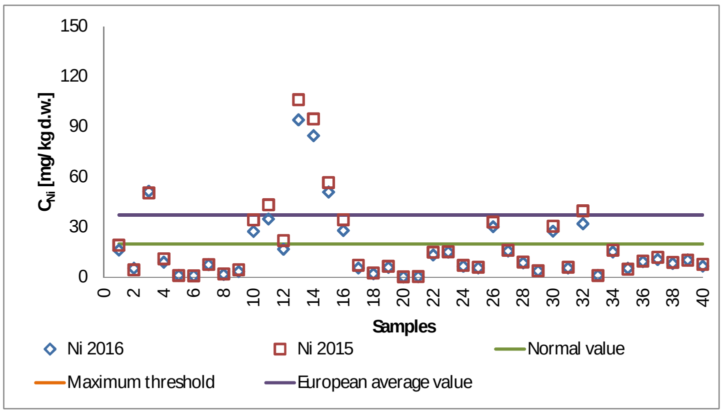


Fig. 12 – Comparison between the values of nickel concentration from surface soil samples collected in Targoviste City and the values provided by the legislation.

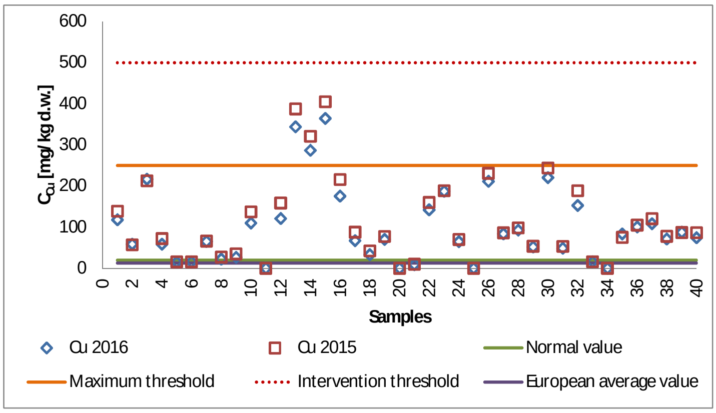


Fig. 13 – Comparison between the values of copper concentration from surface soil samples collected in Targoviste City and the values provided by the legislation.

Figure 13 shows the quantitative distribution of copper in the surface soil samples, compared with the values provided by Romanian and European legislations (Table 3). The concentration of copper in the surface soil samples of the studied areas recorded values well above the European average, but within the limits of Romanian legislation, with a mean, minimum, maximum values, for both 2015 and 2016 years, presented in Tables 1 and 2. Figure 14 shows the quantitative distribution of copper in the surface soil samples, compared with the values from Romanian European legislations (Table 3). The zinc concentration in the surface soil samples of the studied areas, registered values well above the European average, but within the limits provided by the Romanian legislation.

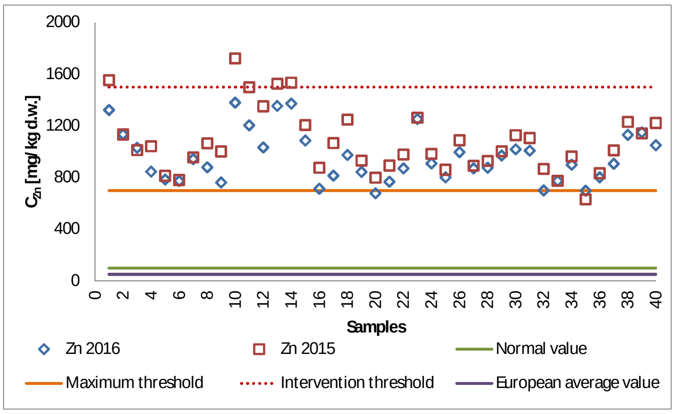


Fig. 14 – Comparison between the values of zinc concentration from surface soil samples collected in Targoviste City and the values provided by the legislation.

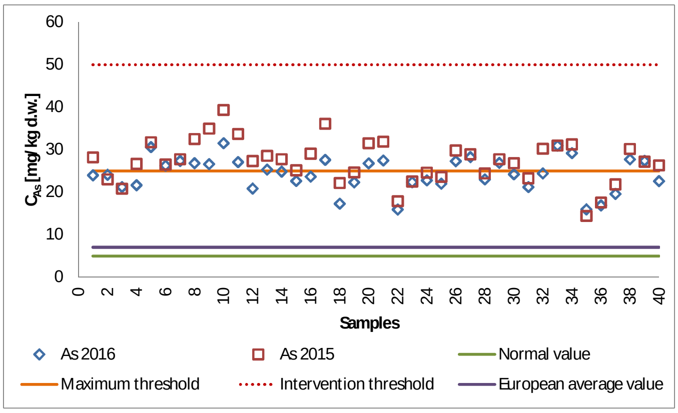


Fig. 15 – Comparison between the values of arsenic concentration from surface soil samples collected in Targoviste City and the values provided by the legislation.

Figure 15 and 16 shows the quantitative distribution of arsenic respectively cadmium, of the surface soil samples, compared with the values provided by Romanian and European regulations (Table 3). The concentration of arsenic and cadmium, respectively in the surface soil samples on the studied areas registered values well above the European average, but within the limits of Romanian legislation.

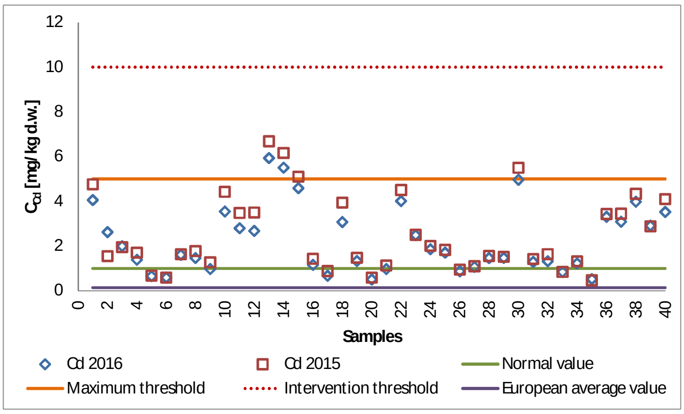


Fig. 16 – Comparison between the values of cadmium concentration from surface soil samples collected in Targoviste City and the values provided by the legislation.

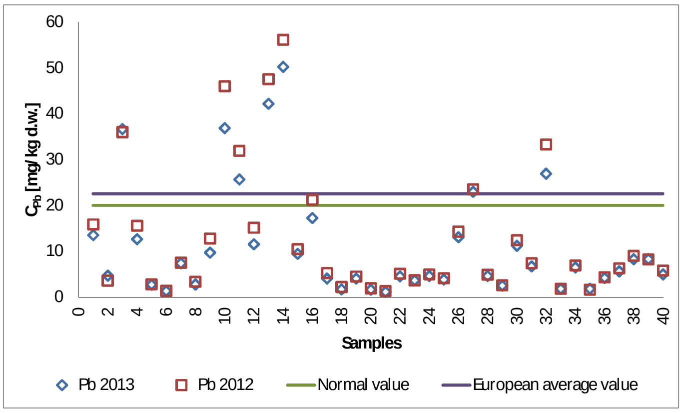


Fig. 17 – Comparison between the values of lead concentration from surface soil samples collected in Targoviste City and the values provided by the legislation.

Figure 17 shows the quantitative distribution of lead in the surface soil samples, compared with the values admitted by Romanian and European legislations (Table 3). The concentration of lead in the surface soil samples of the studied areas registered close values to the European average and within the limits of Romanian regulation.

CONCLUSIONS

The purpose of this study was to investigate the heavy metal levels in surface soil of Targoviste City in order to assess the industrial and anthropogenic impact on urban soil pollution. Clear accumulations of Fe, Zn, Cu, Zn, Cd, Pb and As were observed through the investigation of over 700 soil samples from 40 representative sites of Targoviste City, and it can concluded that approximately 10% of the soil samples were moderately to heavily polluted by these elements. This investigation suggested that the surface soils of Targoviste City had been polluted by both industrial and anthropogenic sources.

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