STUDY OF SOIL POLLUTION WITH HEAVY METALS IN THE TOWN OF COPȘA MICĂ

Camelia MICLĂUȘU[1*], Cristian POP[2], Bogdan VÂLCAN[2], Anamaria RADU[2], Cristina ROȘU[3]

[1]ECO TERRA S.R.L., C-tin Lepadatu 37C, Cisnădie, Sibiu, Romania
[2] Environmental Health Center S.R.L., Busuiocului 58, Cluj Napoca, Romania
[3] Babeș-Bolyai University, Faculty of Environmental Science and Engineering, Fântânele 30, Cluj-Napoca, Romania.
*Corresponding author: eco_camelia@yahoo.com

ABSTRACT. Copşa Mică is known to be one of the most polluted towns in Romania because of the historical pollution generated by non-ferrous metallurgy. The purpose of this study is to investigate soil quality in urban areas, where land is sensitive and where the population is most exposed. Heavy metals (Pb, As, Cd, Cu, Zn) from the horizon of the surface soil were analyzed using descriptive statistical analysis, geostatistical analysis and pollution indices. The results of the analysis showed that the most affected areas of Copșa Mică are in S of the industrial platform in N-E of the town – in the Târnăvioara neighborhood and also in E-SE of neighborhood of the industrial platform. In areas with different land uses, the average concentrations of heavy metals have been decreasing in the following way: in public areas > in access areas (streets) > in yards > in gardens. The level of heavy metals in soil is influenced by anthropogenic activities in each functional area. The pollution load index (PLI) shows a level of soil pollution between moderate and very high. The biggest contribution through the single pollution index (PI) is represented in descending order by the following: Pb, Zn and Cd. There is no correlation between the calculated values of the PLI and the distance from the emission source (the metallurgical unit basket). Because a large area of land is affected by pollution, it is not feasible to apply greening measures, so interventions should be focused on decreasing the exposure of the population to heavy metals.

Key words: *soil, heavy metals, Copșa Mică.*

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INTRODUCTION

Copșa Mică is a heavily polluted industrial area where the retention of heavy metals in the soil is long-lasting and the effects on the health of the population are still present. This affirmation is supported by the results of population health studies (Gurzău et al., 2008, 2010b), but also the fact that the industrial platform of Copșa Mică is included in the preliminary inventory of potentially contaminated sites (A.P.M. Sibiu, 2017).

Over time, in Copșa Mică, soil quality was investigated through a series of studies. The first investigations carried out (Răuţă et al., 1987, Vădineanu et al., 1991, Toti et al., 1993, Comănescu et al., 2010, Lăcătuşu and Lăcătuşu, 2010) have shown a polluted area of approximately 180,750 ha.

According to Damian et al. (2008) the highest concentrations of heavy metals are found in the horizons of soil surface where normal Pb values have been exceeded (5.61 to 27.68 times), for Zn (2.16 to 25 times) and for Cd (5.99 to 133.3 times).

According to Gurzău and Neamţiu (2010a), a research project carried out in Copșa Mică (and the area of Sălişte) has shown that here the level of pollution with Pb and Cd remained high.

Other investigations by Szanto et al. (2012), found that for Pb and Cd the intervention threshold for sensitive uses is exceeded. The highest concentration for Pb is in the surface horizon but is decreasing with depth and when the distance from the source is increasing. For Cu and Zn, the highest concentrations are recorded in the surface horizon where the alert thresholds for sensitive uses are exceeded.

According to Lăcătuşu (2014), the soil pollution level at Copşa Mică is assessed based on the values of the pollution index. The result showed that there is a moderate to high soil pollution of Cd and Pb, a moderate pollution of Zn and a low pollution of Cu.

Regarding soil pollution, effects are felt on land use. Thus, their economic value is diminished, there occur some conditions for use in case of future development of localities, and the perspective of territorial planning and urban development is not favorable. This means that the socio-economic environment has no optimistic prospects. Taking into consideration the land surface that has been affected by heavy metal pollution, methods for greening are unfeasible in such areas.

It is well known that historically polluted localities have low socio-economic development potential, which are *"hot spots"* at local and national level, not only from this point of view, but also as a theme in social, health and environmental policies.

MATERIALS AND METHODS

Sampling methods

The study was carried out in two research campaigns in 2014 and 2018, when samples were taken and analyzed from the urban area of Copșa Mică, from the superficial horizon of the soil (0-5 cm). Determination of soil sampling depth was made taking into account that most research in the area showed that the highest concentrations of heavy metals are recorded in the horizon of soil surface (Damian et al., 2008). The research area was established in the urban areas of Copșa Mică, on lands with sensitive uses, because they are currently used by the population and the population is exposed for the longest time.

In 2014, a number of 20 individual households from Copșa Mică were investigated. Samples were collected from the access area in households (at the front of the street), yards and gardens, resulting 60 soil samples analyzed.

In 2018, other 27 soil samples were collected from public areas, the most transited by the population, especially areas frequently used by children like schools, kindergartens, playgrounds, sports grounds, public administration, cult institutions, market and supermarket. Focus is on these areas because the group of population with the highest susceptibility to exposure to heavy metals in the soil is represented by children (Gurzău et al., 2008, 2010b), and the study about soil pollution was continued with a population exposure study to heavy metals in the soil.

The investigated points have been established to be uniformly distributed across all directions in the urban area of Copșa Mică. They were marked with G1-G20 (households), the investigated points in 2014, and P1-P27 (public areas), the investigated points in 2018. These points are located according to figure 1.
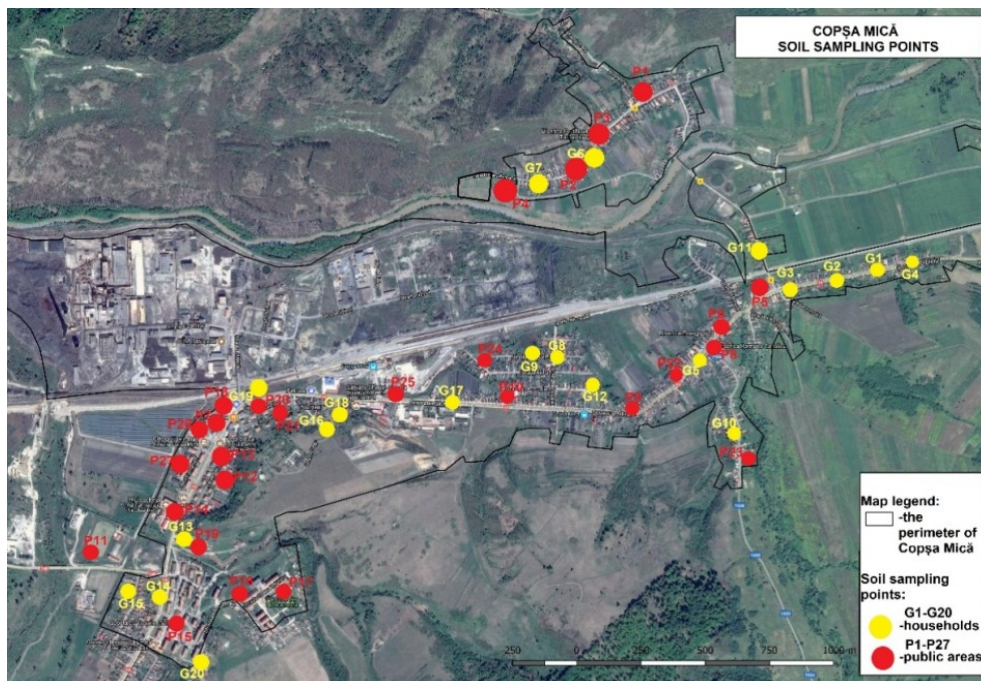
Fig. 1. Copşa Mică, soil sampling points

When soil samples were taken, the land was ment to be without filler materials, not affected by various material or waste and not being cemented or asphalted. Soil samples were collected after the vegetation was completely removed, the surface of the soil was cleared by removing dust, roots, leaves or other residues. The samples were handle carefully, not to be contaminated during sampling, storage, transport and during laboratory analysis.

To determine concentrations of heavy metals in soil (Pb, Cd, Cu, Zn and As), the analyzes were performed in the Environmental Health Center laboratory, Cluj.

Analytical method for heavy metals in soil is by X-Ray fluorescence (XRF) spectrometry.

Analytical equipment used: Niton™ XL3t XRF Analyzer (Thermo Scientific™).

Method for laboratory analysis used: US EPA Method 6200 reference method.

Descriptive statistical analysis

The results were centralized into a .xlsx document and were interpreted by descriptive statistics such as the measurement of the central trend (mean, median, frequency of occurrence of certain values / histogram) and by measuring the variability (data dispersion interval – value minimum and maximum, standard deviation). The statistical data analysis tool was used from Surfer 13 program.

Geostatistical Analysis

Spatial distribution of heavy metal concentrations in soil was performed using the Kriging Interpolation Method in the ArcGIS software, a method which helps determine and assign values to missing points based on measured values in their vicinity. Ordinary Kriging is a commonly used interpolation method to predict the overall trend of soil pollution.

Noticing similarities in the spatial distribution of heavy metals in urban areas, statistical testing was used through the correlation method. For the calculation of the correlation coefficient between heavy metals in the soil, the DataAnalysis tool in the Excel program was used and the matrix of the correlation coefficients was accomplished.

The correlation coefficient r (Pearson) takes values between -1 and +1, passing through 0, which indicates a null correlation. For the positive interpretation of the correlation coefficient (r) we use: $0 < r \leq 0.1$ – very week correlation; $0.1 < r \leq 0.39$ - week correlation; $0.4 < r \leq 0.69$ – moderate correlation; $0.7 < r \leq 0.89$ – strong correlation; $0.9 < r \leq 1$ – very strong correlation (Schober et al., 2018).

Soil contamination assessment methods

The assessment of the soil pollution degree was performed by calculating the pollution load index (PLI). This index shows the level of heavy soil contamination and was calculated based on the single pollution index (PI). These indices are calculated separately for each analyzed metal (Kovalska et al., 2018).

PLI is calculated as a geometric mean of PI according to the formula:

$$PLI = n\sqrt{(PI_1 \times PI_2 \times PI_3 \times ... PI_n)},$$

where: n – number of pollutant assesses; PI – single pollutant index of each metal assesses (Varol, 2011).

PLI values vary from 0 (unpolluted) to 10 or more (highly polluted) as follows: PLI = 0 – background concentration; $0 < PLI \leq 1$ – unpolluted; $1 < PLI \leq 2$ – moderately to unpolluted; $2 < PLI \leq 3$ – moderately polluted; $3 < PLI \leq 4$ – moderately to highly polluted; $4 < PLI \leq 5$ – highly polluted; PLI > 5 – very highly polluted (Zhang et al., 2011 and Kowalska et al., 2018).

The single pollution index (PI) is used to determine the metal that represents the greatest threat to soil (Zhang et al., 2011 and Kowalska et al., 2018). Calculate with the formula:

$$PI = Cn / GB,$$

where: Cn – concentration of metal in the soil sample; GB – geochemical background (mg/kg).

PI levels and significance: PI < 1 – non polluted; $1 \leq PI < 2$ – slightly polluted; $2 \leq PI < 3$ – moderately polluted; $PI \geq 3$ – highly polluted (Jorfi et al., 2017).

When the individual pollution index (IP) was calculated, the level of geochemical background in Romania was taken into account (Utterman et al., 2006).

RESULTS AND DISCUSSIONS

The results of the analyzes performed were centralized into a .xlsx document and were interpreted through summary statistical methods.

Table 1. Values of minimum, mean and maximum concentrations for lead (Pb) and arsenic (As) in soil in different function areas (mg/kgSU)

| Sampling area	Pb			As		
	min	med	max	min	med	max
Access area (street)	157.73	1550.22	8141.12	16.85	63.06	302.51
Yard	214.98	1465.79	4496.61	12.97	62.84	152.29
Garden	270.54	**907.55**	2595.45	20.43	**36.93**	69.39
Public area	36.27	2920.11	18514.66	18.62	199.02	803.57

Table 2. Values of the minimum, mean and maximum concentrations for cadmium (Cd), copper (Cu) and zinc (Zn) in soil, in different function areas (mg/kgSU)

Sampling area	Cd			Cu			Zn		
	min	med	max	min	med	max	min	med	max
Access are (street)	9.54	46.26	236.90	57.68	232.91	1123.46	543.75	2843.85	17661.06
Yard	11.33	32.81	84.84	56.19	224.59	1373.17	531.21	2566.05	8221.26
Garden	8.84	**21.82**	53.28	61.76	**131.37**	233.87	610.00	**1640.81**	3599.07
Public area	11.49	48.76	151.85	13.14	250.43	1272.93	34.76	2810.99	11346.43

The results of the analyzes showed that in 2014 average values of concentrations were recorded in all sampling areas in descending order Zn > Pb > Cu > As > Cd, with the following values (mg/kgSU): Zn – 2350.23; Pb – 1307.85; Cu – 196.29; As – 54.94; Cd – 32.88.

For the year 2018 heavy metals have recorded average concentrations in descending order Pb > Zn > Cu > As > Cd, with the following values (mg/kgSU): Pb – 2920.11; Zn – 2810.99; Cu – 250.43; As – 199.02; Cd – 48.76. Unlike in 2014, higher average concentrations were observed for all metals analyzed. One reason is that in 2014, more than 20% of soil samples were taken from heavily disturbed areas of gardens, where there are permanent interventions on the soil through usual farm work, manure fertilization and gardening etc.

It is known that soil intervention by mechanical work can lead to dilution of pollutant concentrations in the horizon of soil surface. Irrigation can also favor vertical transport. This fact is also highlighted in the tables, where it can be noticed that average concentration of heavy metals in the samples taken from the gardens are significantly lower than the other sampling areas.
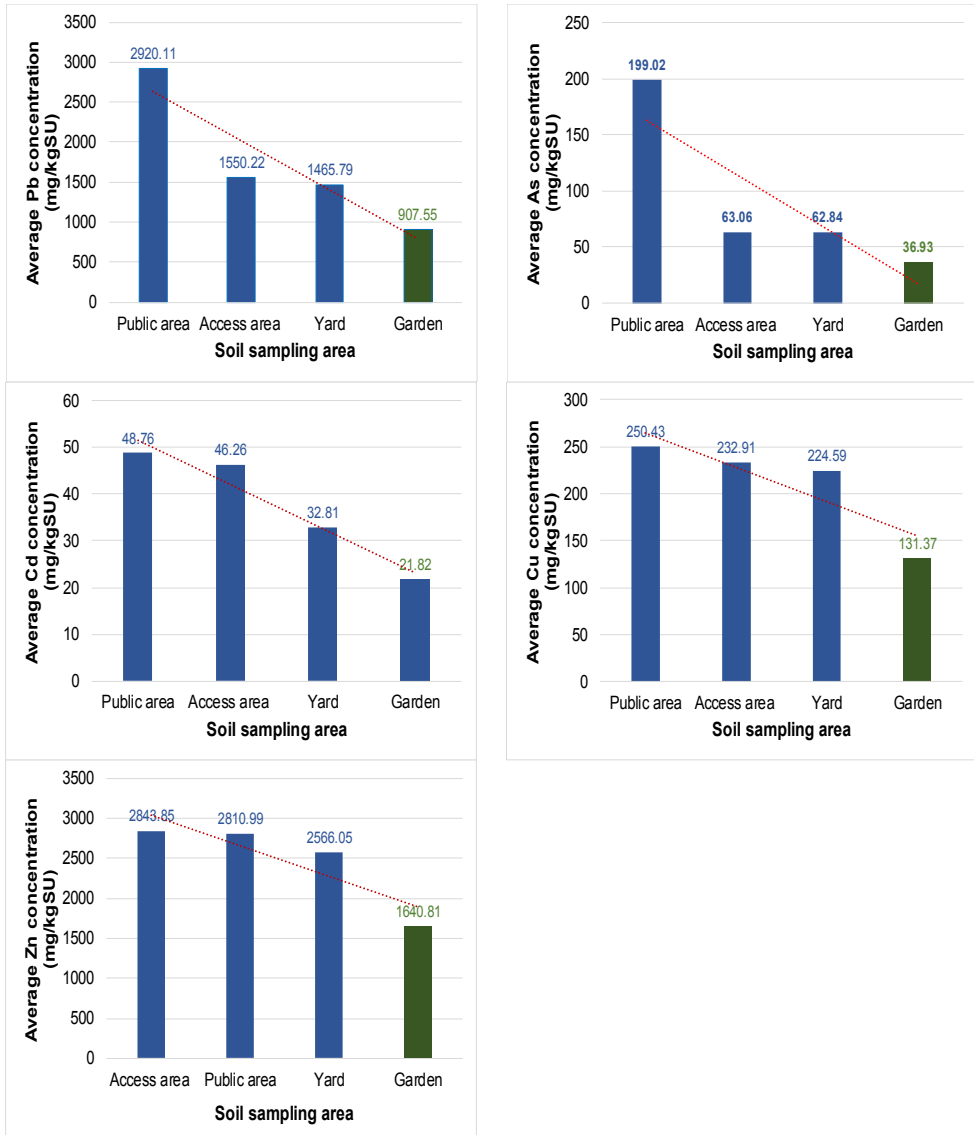
Fig. 2. Average concentration of heavy metals content of soil in different areas

According to graphics, the highest average concentrations were recorded in descending order in the public area > in the access area (street) > in the yard > in the garden. The exception is Zn, where the average concentration recorded on the access zones is slightly higher than the recorded average for the samples taken from public areas, but the difference is not significant (1.16%).
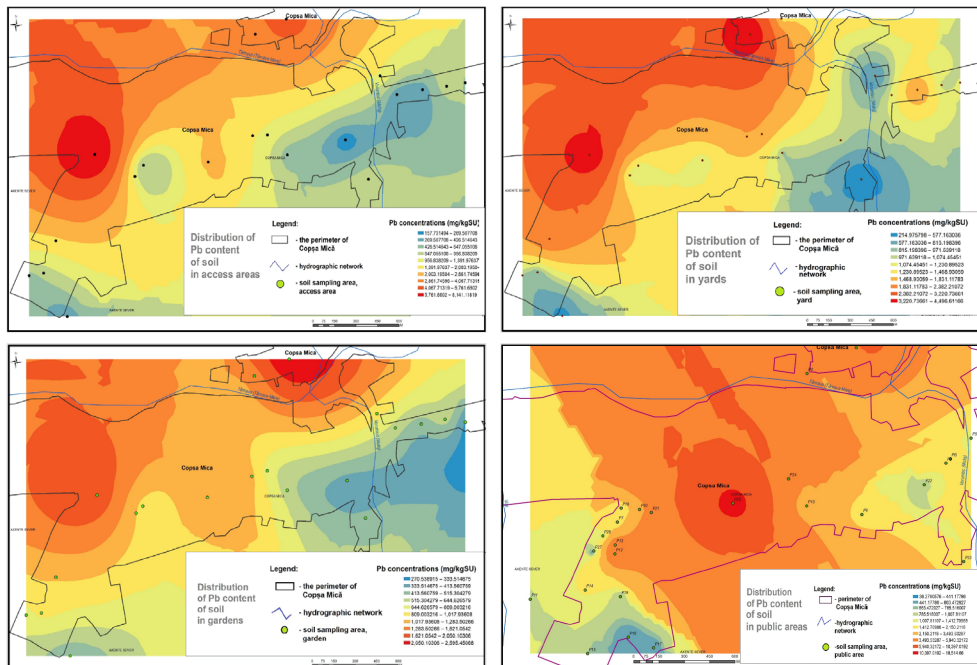
The highest average of heavy metals is recorded outside households. Theoretically, inside them there are frequently interventions, resulting in disturbing the superficial horizon of soil and dilution of pollutant concentrations. In gardens, interventions are most frequent, due to annual usual farm work. In public areas, interventions are lower compared to access areas in the household, where there are frequent interventions for the arrangement and maintenance to access households and only exceptional, with excavations for infrastructure works (water network construction between 2013-2014).

Spatial distribution of heavy metals content of soil in Copșa Mică

The spatial distribution of heavy metal content of soil registered in 2014 and 2018 in Copșa Mică, highlights the areas most affected by pollution compared to the emission source, which is the industrial platform SOMETRA, the flue-stack of the metallurgical unit.

Fig. 3. The distribution map of Pb content of soil in different areas, Copșa Mică

We can see a rule on average concentrations of heavy metals depending on the field of land use. The highest concentrations being recorded in 2018 in public areas, with the lowest concentrations in 2014 in gardens (except Zn, insignificant difference). It was considered relevant to generate separate distribution maps for the four soil sampling areas: home access (street), yard, garden and public areas.

The distribution maps of Pb content of soil in different areas show similarity between them in terms of concentration peaks, the highest values being recorded in the vicinity of the industrial platform in S of it, in the Târnăvioara neighborhood – in the NE and in the E-SE of the industrial platform, indicating that these are the areas most affected by pollution.

Even if there are frequent soil interventions in the gardens, it is noted that the highest concentrations are maintained in the same areas of the town. It is explained by the fact that the interventions on the soil, the existing concentrations of pollutants, by dilution due to agricultural works, don't manage to go down to normal values.

As shown above, the average concentration recorded in the gardens is the lowest compared to the averages recorded in the other sampling areas, but the high concentrations of pollutants persist here as well.

The problem is that in the neighborhood located at the E-SE by the industrial platform and in Târnavioara, rural traditions are maintained meaning the population is cultivating vegetables for their own consumption, gardens drenched with water from fountains. The areas least affected by Pb pollution are those located in the E and SE parts of Copșa Mică, towards Mediaș and Valea Viilor.

To observe the distribution of other heavy metals in the soil (As, Cd, Cu, Zn), the distribution maps in the access area (street) are also presented, as an example.

The spatial distribution of the highest concentrations for As, Cd, Cu and Zn respects the model of Pb distribution.

For 2014, all heavy metals have recorded the highest concentration values in the vicinity of the industrial platform, in S of it, in the Târnăvioara neighborhood and in the E-SE neighborhood of the industrial platform. The exit from Copșa Mică to Mediaș and to Valea Viilor, the areas on the E and SE side of Copșa Mică are the least affected by pollution.
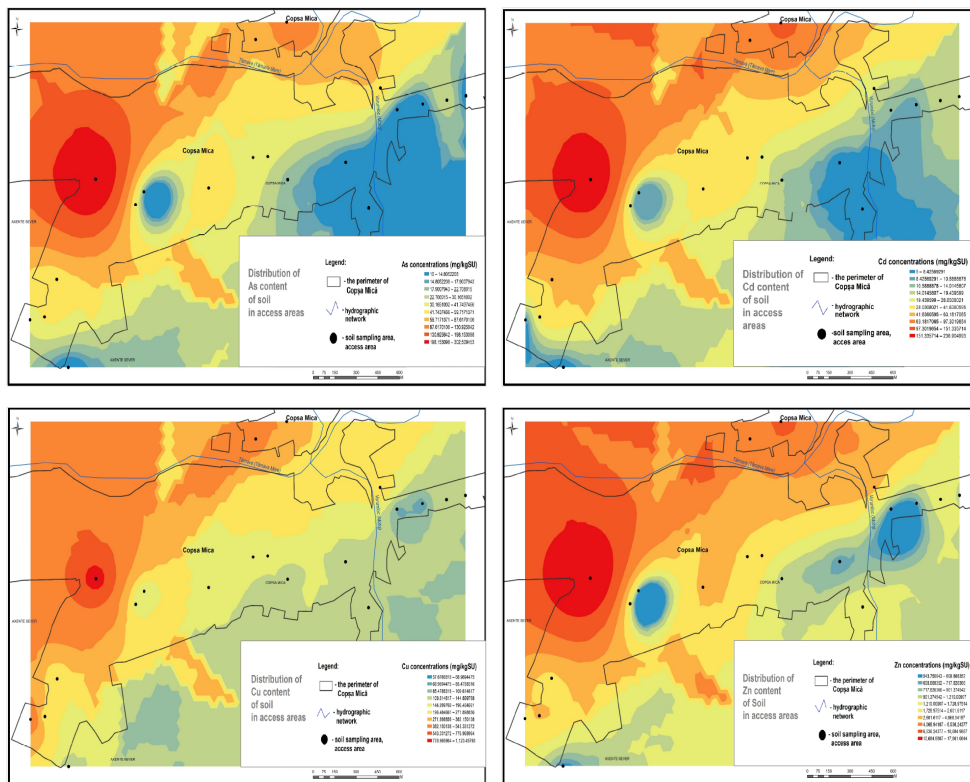
Fig. 4. The distribution map of As, Cd, Cu and Zn content of soil, in the access areas in households, Copșa Mică.

The spatial distribution maps of the heavy metal concentrations in the soil indicate that a correlation is possible between them in Copșa Mică.

Correlation test for heavy metal concentrations in soil

Using the DataAnalysis tool in Excel, the matrix of correlation coefficients was accomplished.

Table 3. Correlation coefficient matrix for heavy metals in soil,
Copşa Mică, 2014 and 2018

|	Pb	As	Cd	Cu	Zn
Pb	1				
As	0.873956	1			
Cd	0.708955	0.741514	1		
Cu	0.687042	0.791314	0.754622	1	
Zn	0.688901	0.718573	0.951336	0.877277	1

There is a very strong correlation between the concentrations of heavy metals for Zn-Cd, a strong correlation for Pb-As and Zn-Cu, a moderate correlation between Pb-Cd, Pb-Cu, Pb-Zn, As-Cd, As-Cu, As-Zn, Cu-Cd and Cu-Zn.

The lowest values of the correlation coefficients are for Pb and Cu in relations with the other metals.

The correlation between these five metals is not by accident, and they are influenced by the same soil pollution phenomenon that has been present for decades in Copșa Mică.

Calculation of pollution indices (PLI, PI)

The following graphs present the results of the calculations for PLI, for all samples taken in 2014 and 2018.
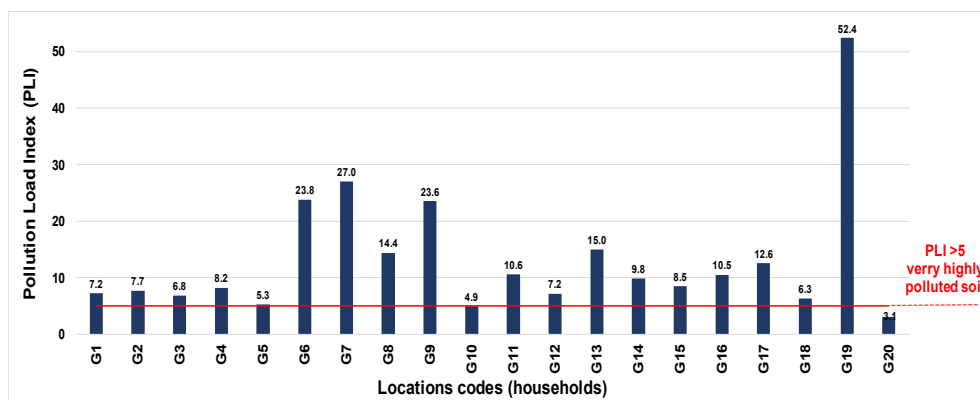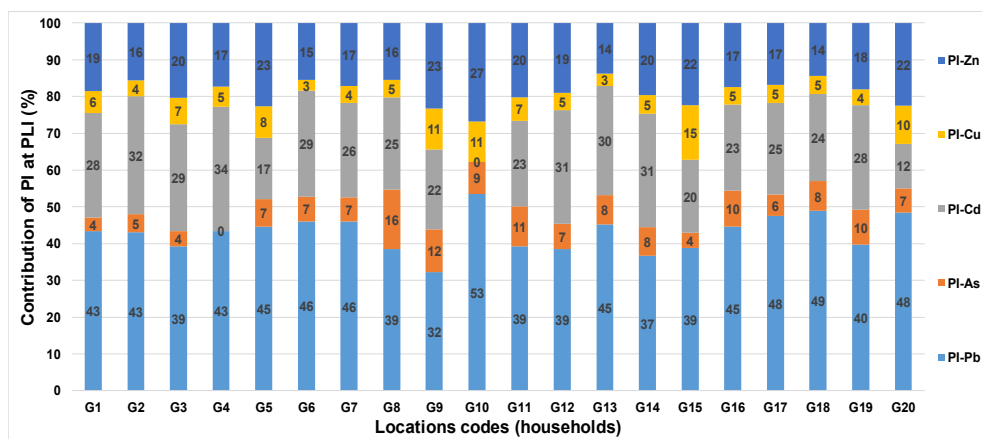

Fig. 5. Values of PLI for soil, Copșa Mică, 2014

PLI values indicate a moderately to highly polluted soil in one of the investigated households G20 (PLI = 3.1), a highly level of pollution in one household G10 (PLI = 4.9) and very highly polluted soil in the remaining of 18 households investigated.

The lowest levels of PLI are recorded in two households, one in SW of Copșa Mică to Axente Sever (G20), and one in SE of Copșa Mică to Valea Viilor.

The highest values of PLI were registered in the investigated households: G19 (PLI = 52.4), G7 (PLI = 27.0), G6 (PLI = 23.8) and G9 (PLI = 23.6). These households are located in the S of industrial platform, in the Târnăvioara neighborhood and in the E-SE neighborhood of the industrial platform.

To highlight the contribution of each metal to the total level of PLI in the soil (2014), graphical and percentage indices of individual pollution (PI) were represented in figure 6.


Fig. 6. Percentage contribution of PI at the level of PLI for soil, Copșa Mică, 2014

The highest contribution to the general level of PLI by individual pollution indices (PI) is in descending order Pb > Cd > Zn > As > Cu, with the following values: Pb – 42.9%; Cd – 24.5%; Zn – 18.8%; As – 7.4%; Cu – 6.4%.

For the year 2018, the calculated values of PLI (figure 7) indicate an unpolluted soil in two investigated points, P27 – playground Castanilor street (PLI=0.4) and P16 – playground no.1, 1st December neighborhood (PLI=0.9). The explanation could be that the soil was disturbed by recent arrangement of these areas, especially in P16.

For an investigated point P19, there was a low level of pollution (PLI=1.62), being the Copşa Mică Orthodox cemetery area. For two investigated points resulted a moderately polluted soil P1 (PLI=2.48) and P17 (PLI=2.34) – these were located in Tarnăvioara (playground) and playground no.2 in Copşa Mică, the explanation being that they were newly arranged and the soil being disturbed.
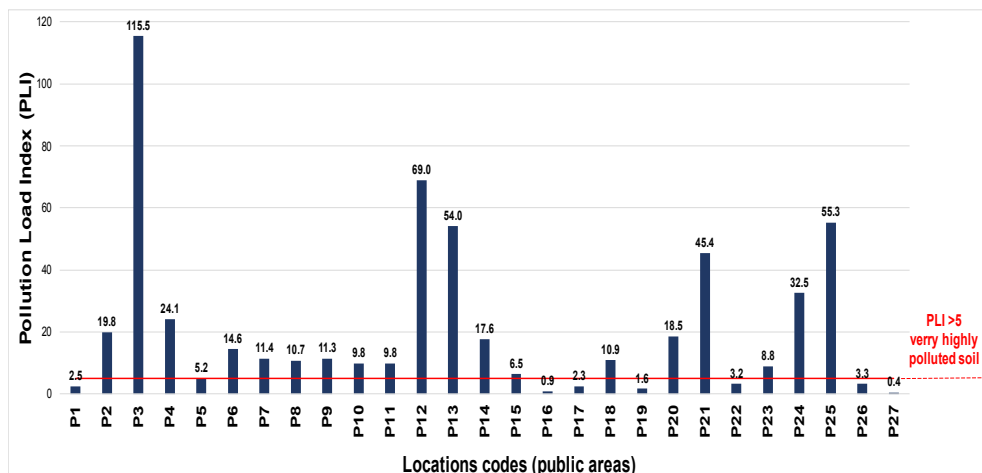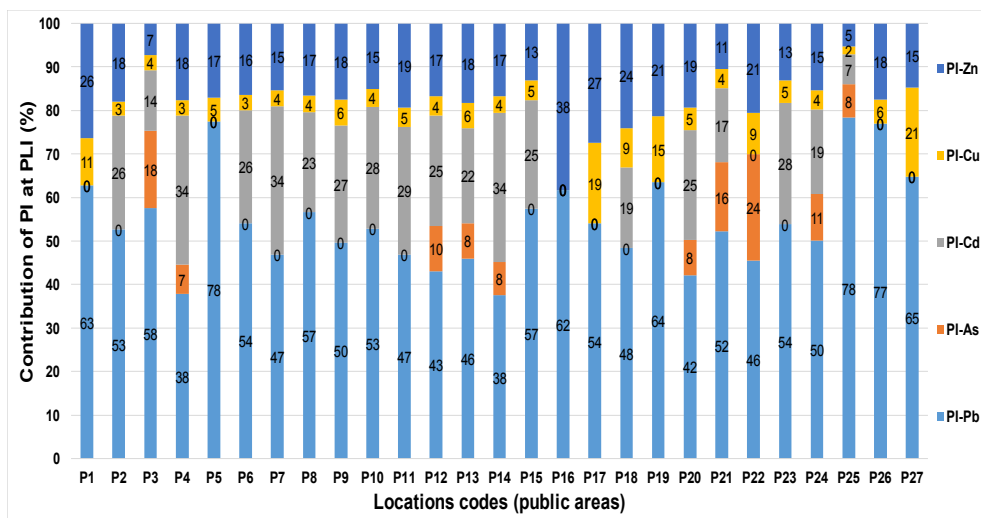

Fig. 7. Values of PLI for soil in Copșa Mică, 2018

There was a moderatey to highly polluted soil in points P22 (PLI=3.23), P26 (PLI=3.26), P22 (PLI=3.27) and P26 (PLI=3.3). A verry highly polluted soil (PLI > 5) resulted in the remaining 20 investigated points.

The highest levels of the PLI were recorded in the following points: P3-Orthodox Church Târnăvioara (PLI=115.47); P12-Kindergarten no.1 of Copşa Mică (PLI=69.0); P25-Copşa Mică Sport Hall (PLI=55.32); P13-playground Kindergarten no.1 (PLI=54.04) and P21-Copşa Mică playground (PLI=45.43). Most of these points are relatively close to the industrial platform and are frecvently used by children.

To highlight the contribution of each metal to the total level of PLI in the soil (2018), graphical and percentage indices of individual pollution (PI) were represented in figure 8.

Fig. 8. Percentage contribution of PI at the level of PLI for soil,
Copşa Mică, 2018

The highest contribution to the general level of PLI by individual pollution indices (PI) is in decreasing order Pb > Zn > Cd > As > Cu, with the following values: Pb – 54.5%; Zn – 17.8%; Cd – 17.1%; Cu – 6.3%; As – 4.3%.

Both in 2014 and 2018, the highest contribution to PLI is represented by PI calculated for Pb, Zn and Cd.

To verify if there is a correlation between the value of the general pollution index (PLI) and the distance from the industrial platform SOMETRA, especially the flue-stack of the metallurgical unit, a correlation test was performed using the DataAnalysis tool in Excel.

Table 4. Correlation test between PLI level and

distance from emission source

	Distance	PLI
Distance	1	
PLI	-0.16907	1

There is a very weak, inversely negative correlation between the PLI level and the distance to the flue-stack of the metallurgical unit.

CONCLUSIONS

The highest concentration averages for the analyzed heavy metals were recorded for Pb, Zn, Cu, and then for As and Cd, the latter being recognized as having toxic potential even at low concentrations in the environment.

In 2018, higher concentration averages for heavy metals were found in the soil due to the fact that in the 2014 campaign at least 20% of soil samples were taken from areas disturbed by agricultural and infrastructure works (construction of water supply network, 2013-2014).

For the heavy metals analyzed, depending on the use of lands, highest concentration averages occur in a descending order: in the public area > in the access area (street) > in the yard > in the garden, this correlating with the anthropic intervention on these areas. The exception is Zn, where the concentration average recorded on access areas in households is slightly higher than the average recorded for public areas, but is insignificant (1.16%).

From the distribution maps of heavy metals content of soil it can be observed that the most affected areas are: in the vicinity of the industrial platform, in S of it; in Târnăvioara neighborhood, in the NE towards the industrial platform; and in E-SE of the industrial platform neighborhood.

The calculated pollution indices show mostly a level between moderate to very high soil pollution. The highest contribution to PLIs, by individual pollution indices (PIs), is in decreasing order: Pb, Zn and Cd.

Following the statistical correlation test between the PLI levels and the distance to the emission source (the flue-stack of the metallurgical unit), there was no statistically significant correlation between the PLI level and the distance from the emission source.

Like previous studies, in 2014 and 2018 results of the analyses obtained further indicate soil pollution with heavy metals at a level between moderate and very high, according to the pollution indices. Because the area affected by pollution is large, measures for greening the area are not feasible but there can be implemented measures to decrease the exposure of population in the most affected areas and within certain functional areas.

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