

EVALUATION OF SOIL POLLUTION DEGREE IN THE COPȘA MICĂ AREA (ROMANIA) BY MEANS OF RELATIVE INDICES

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Abstract

Regardless the source, soil pollution is a daily reality resulting from anthropising the environment, especially through industrial explosion, increasing consumption and excessive urbanization. Lead, cadmium and other heavy metals provided by human activities accumulate in the soil by bioaccumulation at concentrations that become dangerously high for the biota living in or on the soil. While getting in permanent contact for a long time with the soil, the inhabitants of the polluted areas are exposed daily to the risk entailed by the toxicity of dangerous concentrations of Pb and Cd. The assessment of the pollution degree with heavy metals of the soil can be accomplished by comparing experimental values with the maximum permissible levels regulated by the regional, national or international legislation or by using relative indices such as the Global pollution index Z. Loading with Pb and Cd of some adjacent areas to the point sources of pollution is conditioned by remoteness from the source, the local orography of the land that dictates the concentration, orientation or, as the case may be, the spreading of air mass polluted with the heavy metal component.

Key words: heavy metals, historically polluted area, relative indices, geogenic abundance, Copsa Mica area.

INTRODUCTION

Soil pollution

The discovery and processing of metallic minerals as well as the dispersion in the environment of the waste triggered the pollution and loading over the maximum permissible levels with potentially toxic heavy metals of the soil-component of the terrestrial biotopes that has supported the emergence and development of the human civilization.

What was initially a sporadic phenomenon i.e. the mining-transformed in time, favoured by consequences of the industrial revolution, into a global scale process due to the technological explosion, excessive urbanization and the consumerism the 20th Century and the beginning of the 21st Century (Swartjes, 2011). According to the European Environmental Agency (EEA, 2007) most of the polluted sites are located within or adjacent to the cities.

Soil pollution is a complex phenomenon, the soil being in the position of a polluted system when loaded with polluting substances, or being a polluting agent when it supplies soil particles charged with pollutants or even a depollution system due to its capacity to degrade biodegradable pollutants (Sabău, 2009). The sources of soil pollution can be natural, due to paedogenesis and anthropogenic processes (Galiulin, 2002), generated by human activities. Sabău (2009), Mousavi et al. (2013) presents the causes of the anthropogenic pollution of soils, the main role in this process belonging to the non-ferrous metals processing and mining (including tailings and other residues), the sludge resulting from waste water treatment, irrigation with contaminated water, burning of fossil fuels, treatment of soil with fertilizers, pesticides, exhaust gas from road traffic and waste storage (Popa, 2005). Although it may have further uses, the slag (tailings) resulting

from Pb and Zn melting is considered hazardous waste according to European Commission Decision of 18 December 2014.

The causes of heavy metal soil pollution are natural or anthropogenic.

Geogenic contamination sources load the soil in terms of background concentrations (baseline values or Reference Values), which levels are amended with anthropogenic contribution due to the long-distance transport of anthropogenic emissions. (U.S. EPA, 2002; McLaughlin, 2011).

Heavy metal contamination of soil

Metals are natural constituents of soils (Table 1). Chemical elements such as Fe, Mn, Zn, B, Cu, Mo are called micronutrients and are needed for plant growth. Bolan et al. (2008) show that metals accumulate and persist especially in the superficial layers of the soil.

Soil contamination due to emissions from smelters is generally higher near the point source and decreases exponentially with distance (Haugland et al., 2002).

Acid conditions can remove coal fly ash and bottom ash contained in traces of As, Pb, Cd, Cr, Mn, Cu, Zn, Ni, generating the soil source of pollution (Ahmed et al., 2010). In soils not polluted with metals, the concentration of Zn, Cu, Pb, Ni, Cd, Cr ranges between 0.0001-0.065%.

With the exception of Fe, all heavy metals with concentrations in soil above 0.1% become toxic to plants. Soils rich in Zn (0.1-10% Zn) also have high concentrations of Pb, and soils with a Cu concentration of 0.1 to 3.2%, also contain high amounts of Zn, Pb, Co, Ni, Cd (Bothe, 2011).

Table 1. Natural concentrations of Pb and Cd in soil (Franck and Tölggyessy, 1993)

Chemical element	Concentration in soil	
	Characteristic values in ppm	Concentration ranges
Cd	0.06	0.01-7
Pb	10	2-200

The U.S. Environmental Protection Agency (USEPA) encloses the chemical elements Al, As, Be, Cd, Cr, Cu, Hg, Ni, Pb, Se, Sb, on the major interest index in terms of bioavailability due to the toxic risk they entail to human health (John & Leventhal, 1995).

Soil contamination with Pb

Pb is found in small quantities in the earth's crust and it is a major pollutant of the soil (Gratão et al., 2005). According to Kabata-Pendias (2011) the concentration of Pb is 15 mg/kg, while according to Rudnick and Gao (2003) the ratio is 11 mg/kg. The average Pb concentration in soils according to Fiedler and Rössler (1988) is 15 mg/kg. The global coefficient of geochemical abundance of Pb in soil is 0.94 (Lăcătușu and Ghelase, 1992). Anthropogenic pollution sources consist mainly by ore extraction and processing (Dunnivant and Welborn, 2019). Kabata-Pendias (2010) reported large amounts of Pb in the proximity of these sources of pollution. Kabata-Pendias & Pendias (2001), indicate very high concentrations of Pb in Europe in the proximity of areas adjacent to mines and industrial ore processing platforms e.g. 21.546 mg/kg (England), 13.000 mg/kg (USA), and 18.500 mg/kg (Greece). In Romania, concentrations of 1.083 mg/kg in Baia Mare, 2.248 mg/kg in Zlatna, 3.550 mg/kg in Copșa Mică were recorded by Răuță et al. (1992). According to Kloke et al. (1984) the maximum permissible level for the total content of Pb in soil is 100 mg/kg. Moreover, accumulations of Pb in air due to road traffic are sources of soil contamination (Wang & Zhang, 2018).

Increased soil acidity increases Pb solubility but the mobilization is lower than the rate of accumulation in the rich layer in organic matter (Hough, 2010). According to McBride (1994) Pb is distributed and accumulates generally in the surface soil mainly due to adsorption of organic matter, and high acidity conditions can increase the mobility and bioavailability of this chemical element.

Soil contamination with Cd

Cadmium is one of the environmentally most toxic heavy metal present in ecosystems. Cd abundance in the earth's crust is 0.1 mg/kg according to Kabata-Pendias (2011). According to Lăcătușu & Ghelase (1992), the global coefficient of geochemical abundance of Cd is 2.31, and the average Cd concentration in soil is 0.3 mg/kg (Fiedler & Rössler, 1988). The sources of soil pollution with Cd are the mining and the industry: non-ferrous metals, chemical, and especially phosphatic fertilizers with high Cd content, but also road traffic. Less than 10%

of ambient air Cd concentrations are stored locally, the rest being transported by atmospheric circulation over long distances. Around the decommissioned smelters, concentrations of 100 mg/kg Cd can be measured due to the retention of Cd released over time in the superficial layers of the soil (IPCS INCHEM, 1992).

The objective of this study is to assess the degree of contamination of the soils in the Coșea Mică area affected by historical pollution by a relative index-the Z index.

MATERIALS AND METHODS

The soil sampling was carried out from 15 November to 4 December, 2009, in compliance with soil sampling rules issued by ICPA Bucharest (Methods, Reports, Guidelines, 1981). Between 26 January and 31 March, 2009 the main polluter of the Coșea Mică area i.e. S.C. SOMETRA S.A. ceased its major

industrial activity that has not been resumed so far. The soil sampling depths ranged between 0-5 cm, 10-15 cm, and 30-35 cm. For the undifferentiated assessment in depth of the soil variables, weighted average concentrations were calculated. 79 analytical determinations were made on the Pb and Cd content of the soil sampled from 14 sampling areas containing the following soil types: calcareous regosol, marl faeoziom regosol, typical luvisol, reddish preluvosol and argic horizon soil (xxx, 2009; Estefan et al., 2013). A control sample area "SP1", located at the outskirts of the Blaj town, Alba County at a distance of 26.361 km from the main source of pollution, was considered. The location on the map of the sampling plots from which the samples were collected and subjected to analytical determinations, and having the Coșea Mică industrial platform as focal point of reporting, is presented in Figure 1.

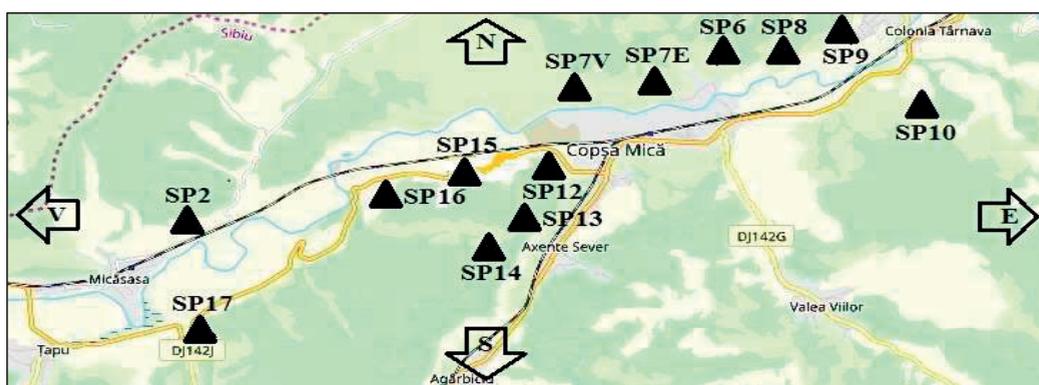


Figure 1. Distribution of the sampling plots in the Coșea Mică area (Romania), from which soil samples were subjected to analytical determinations

Soil sample was homogenized and dried at 105°C. The mineralization of the soil sample and the extraction of the total heavy metal forms studied was carried out by mineralization method using an oxidizing mixture composed of strong acids: HNO₃, HClO₄, and H₂SO₄. In order to quantify the total Pb and Cd content of the soils sampled from sampling areas, we used the flame atomic absorption spectroscopy (FAAS) technique, by the Analyst 200 Atomic Absorption Spectrometer from Perkin Elmer with laminar flame generated by the mixture between pure C₂H₂ and air, using the lamp

specific to the chemical element to be determined, thanks to its high sensitivity, low detection levels, good accuracy, sound and improved methodology, and its relatively low costs (Franck & Tölgyessy, 1993; Csűrös & Csűrös, 2002). For calibration, we made use of a standard solution (Pb, Cd, Zn, Cu,) of 1000 mg/l (Merck) concentration for calibration purpose.

Dry ashing was done with strong acids: HNO₃, HClO₄, H₂SO₄ in a Berzelius glass that contains 1 g of fine soil mixed in a mortar with pestle were added 15 ml of oxidant mixture

HNO₃: HClO₄: H₂SO₄ in a volumetric volume of 2: 1: 0.2 and evaporated to dryness (on a sand bath) (Bader, 2011). After cooling the obtained residue was treated with 5 ml HCl concentrated, the bath sand operation being repeated.

The resulted residue was treated with 0.5 HCl for dissolution, the filterer being passed in a balloon of 50 ml, that was brought to sign with a sol of HCl 0.5 N. The solution that was obtained was aspirated into Perkin Elmer Analyst AA200 spectrometer with the laminar flame generated by the combination of pure C₂H₂ and air, using the lamp specific to the element to be determined. Main parameters of FAAS technique are presented in Table 2. All chemicals used were of analytical reagent grade. Double distilled water obtained with a water purification system (Millipore® Direct-Q, Millipore Corporation, Bedford, MA, USA) was used for all preparations. The glassware and polyethylene containers used for analysis were washed with tap water, then soaked overnight in 10% HNO₃ solution and rinsed several times with double distilled water.

Table 2. FAAS instrumental analytical values of investigated trace elements Pb and Cd

Parameters	Pb	Cd
Wavelength (nm)	217	228.8
Lamp current (HCL) (mA)	9.5	4.0
Flame type	Aer-acetylene	
Air flow rate (l/min)	4	4
Slit width (nm)	0.7	0.7

Concentrations of the Pb and Cd in the soil samples were calculated using the following formula: Concentration (mg/kg) = (Concentration (mg/L)x V)/W (Uwah et al., 2012), where:

V = final volume (50 ml) of solution;
M = initial weight (1g) of sample measured.

Calculation of the Z index for ranking the pollution degree of the soils sampled from the Copșa Mică area

The assessment of the global pollution with Pb and Cd of the surveyed soils was possible by

calculating the Z index based on dedicated literature (Lăcătușu et al., 2012), using Geogenic abundance (AG) through the equation below:

$$Z = \sum_{i=1}^n [AG_i - (n - 1)]$$

where AG_i - geochemical abundance.

Geogenic abundance (AG) is the ratio between the content of the chemical element in soil (C_i) and the clark value (Cl_i), and represents the abundance of the elements in the soil in comparison with that of the lithosphere (Table 3). Clark represents the average content of a chemical element in the lithosphere (Cl), and is calculated as follows:

$$AG_i = \frac{C_i}{Cl_i}$$

Table 3. The Clark values, mean content in soil and global geochemical abundance ratio of elements (mg/kg soil), (Lăcătușu & Ghelase, 1992).

Clark values (after Fiedler & Rösler, 1988)

Chemical element	Clark's value	Mean content in soil	Global geochemical abundance ratio
Pb	16.0	15.0	0.94
Cd	0.13	0.3	2.31

To calculate the Z index value, only the AG_i values ≥ 1.5 are added (n-number of chemical elements surveyed). According to the Z index value, the following levels regarding the degree of soil pollution were established:

- Z < 8 - Minimum pollution;
- 8 < Z < 16 - Low pollution;
- 16 < Z < 32 - Average pollution;
- 32 < Z < 64 - Heavy pollution;
- 64 < Z < 128 - Very strong pollution;
- Z ≥ 128 - Maximum pollution.

RESULTS AND DISCUSSIONS

The Z index values calculated based on the values of the concentrations of Pb and Cd determined experimentally are presented in Table 4.

Table 4. Z and AG index value, calculated on sampling areas depending on the remoteness from the main source of pollution

Sampling surface SP	Remoteness from the main source of pollution (km)	Chemical element determined quantitatively	Geochemical abundance (AG)	Z index calculated value	Z index interpretation
SP1	26.361	Pb	1.44	2.54	Minimum pollution
		Cd	3.54		
SP2	7.818	Pb	4.42	54.88	Very strong pollution
		Cd	52.46		
SP6	2.621	Pb	9.06	78.75	Very strong pollution
		Cd	71.69		
SP7E	1.708	Pb	3.75	177.90	Maximum pollution
		Cd	176.15		
SP7V	1.035	Pb	6.74	24.74	Average pollution
		Cd	20		
SP8	3.045	Pb	5.59	299.20	Maximum pollution
		Cd	295.61		
SP9	5.510	Pb	10.14	137.06	Maximum pollution
		Cd	128.92		
SP10	7.156	Pb	1.87	6.18	Minimum pollution
		Cd	6.31		
SP12	1.145	Pb	12.99	91.14	Very strong pollution
		Cd	81.15		
SP13	2.022	Pb	3.35	33.27	Heavy pollution
		Cd	31.92		
SP14	3.090	Pb	5.40	90.02	Very strong pollution
		Cd	86.62		
SP15	2.994	Pb	13.00	70.08	Very strong pollution
		Cd	59.08		
SP16	4.052	Pb	6.60	73.29	Very strong pollution
		Cd	68.69		
SP17	8.413	Pb	1.85	8.31	Low pollution
		Cd	7.46		

The dispersion of the calculated values of the AG geochemical abundance for Pb and Cd in the soils from the investigated sample surfaces is shown in Figure 2 and shows the tendency of

exponential decrease of the pollution with Pb and Cd reflected in the AG calculated for the soils from the investigated sample surfaces.

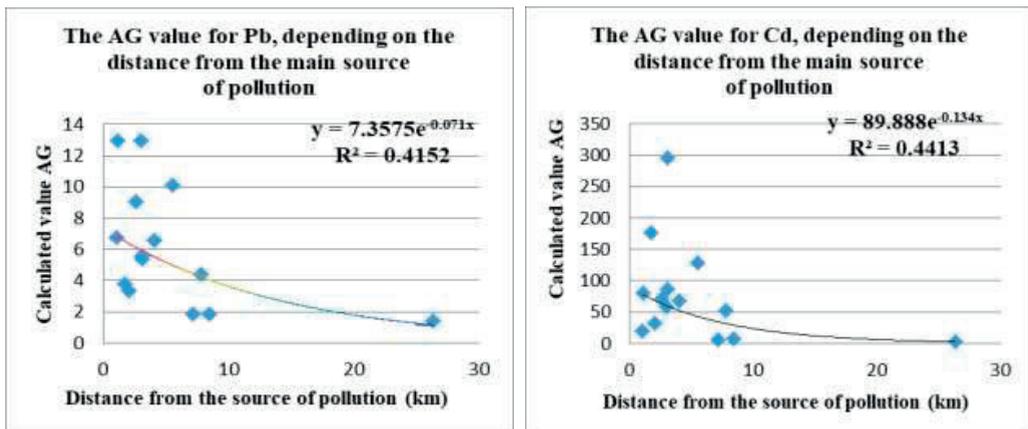


Figure 2. Reporting the distribution of AG values at a distance from the main polluting source

According to the multi-element global pollution index Z, almost half of the soil of the sample surfaces surveyed are **very heavily polluted**, and the sample surfaces with **maximum polluted** soil reaching 20%. The table above indicates that the soil in the control sample area falls into the **minimum pollution** category.

In order to highlight the dynamics of multi-element soil pollution, the distances between the test surfaces and the main source of pollutant emission dispersion located on the Sometra S.A. platform were taken into consideration, as well as the direction of spreading air masses loaded with metal pollutants. The sampling surfaces covered with soils subjected to research are located in the Târnavă Mare valley, upstream (eastward) and downstream (westward) from the town of Coșea Mică, to the left or right bank of the river. Exception is SP1 which is considered as a control sample area located near the town of

Blaj. The Z index value measured at the point of reference considered westward show a decreasing trend between SP12-SP1, the control sample area SP7V being located at a distance of 1.034 km from the source of pollution.

Z-index interpretation of the dynamics of global multi-elemental soil pollution on the emission source-westward direction is as follows: average pollution/very-strong pollution/very-strong pollution/very-strong pollution/very-strong pollution/low pollution/low pollution/minimum pollution (SP7V-SP12-SP15-SP16-SP1-SP2-SP17). Increasing the distance of the test surfaces from where soil was samples eastwards causes a sharp decrease in the Z index values between SP8-SP10 whose interpretations show the following trend: maximum pollution-maximum pollution-minimum pollution (Figures 3 and 4).

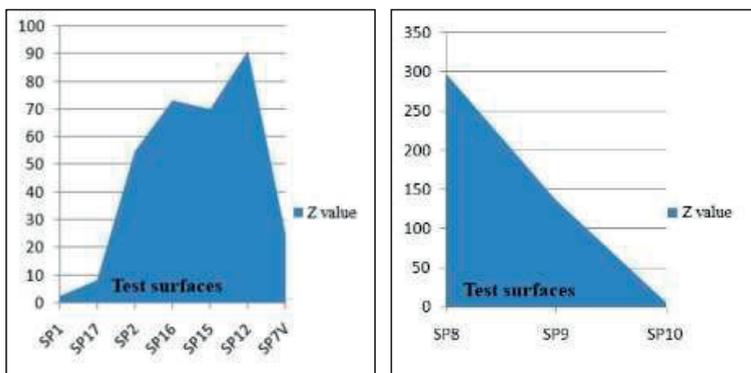


Figure 3. The dynamics of global multi-element pollution of the studied soils according to the index Z and considered cardinal points (W and E)

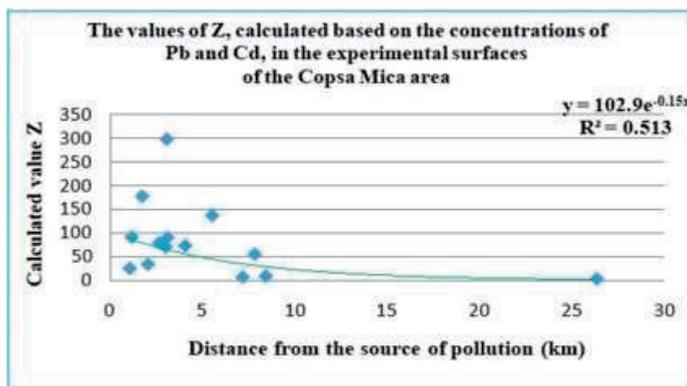


Figure 4. Distribution of the values of Z in the soils of the experimental sample surfaces

When reporting the calculated values of Z index obtained from the sampled soils oriented on the NE and SV direction as against the source of pollutant emissions, a decrease in the pollution degree of the soils was observed from the maximum pollution level (SP7E) to a very strong pollution level in SP6, while increasing distance towards NE. The SV direction determines the following trend of the multi-element global pollution degree of soils: very

strong pollution- strong pollution-very strong pollution (Figure 5). Failure to observe a downward trend in the Z index calculated value in SP13 is due to the location of the test surface outside the Visa valley, compared to the other surfaces exposed more strongly to the pollutants that are located in the afore mentioned valley.

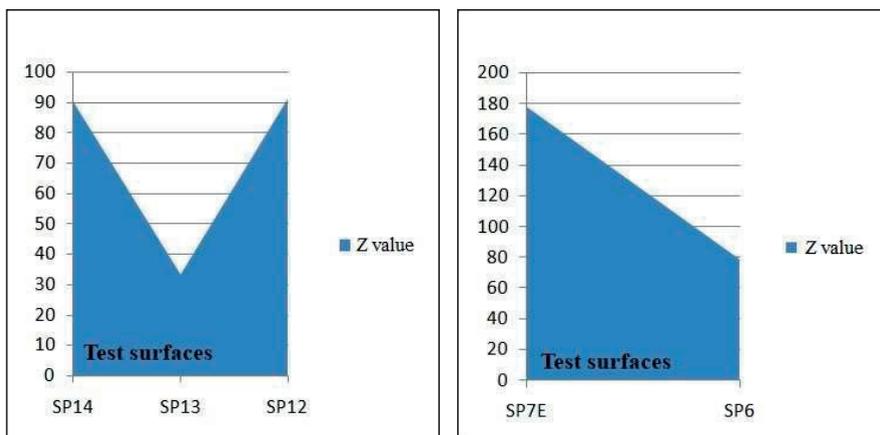


Figure 5. The dynamics of global multi-element pollution of the studied soils according to the index Z and considered cardinal points (NE and SW)

CONCLUSIONS

The calculation of the global multi-elemental pollution index Z allows the classification of soils according to the degree of pollution thereof.

The analysis of the Z index values shows that 20% of the surveyed soils fall into the maximum pollution category, 46% in the very strong pollution category, 7% in strong pollution category, 7% in the average pollution category, 7% in low pollution category, and 13% falls in the minimum pollution category.

It has been determined that increasing the distance from the main source of pollutant emissions westward and eastward is a determining factor in decreasing the degree of global multi-elemental pollution of the surveyed soils.

Besides the dynamic distance, the global pollution of the soils in the Coșșa Mică area, the terrain orography and the local conditions of microclimate are factors that can disturb the trend imposed by the distance or geographical orientation.

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