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SPATIAL DISTRIBUTION OF HEAVY METALS IN AGRICULTURAL SOILS OF ROMANIA: REVIEW OF THE CURRENT SITUATION

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Abstract

Contamination of soils and crops by heavy metals is considered one of the most serious environmental problem due to their non-biodegradable nature, the long biological half-life and also their potential accumulation in different body parts of plants. In Romania there are areas of thousands of hectares of agricultural land polluted with heavy metals. The distribution of heavy metals in agricultural soil can serve as a basis for a better management of soil quality, as well as to protect human health and the soil environment. This paper aims to present an overview based on the data found in other studies and to obtain a mapping of each the most common heavy metals met in soils of Romania. Due to the fact that there are not enough studies for the whole country, some areas will be estimated and also, is possible that others will not be covered at all.

Key words*: heavy metals, mapping, pollution, Romania, soil contamination.*

INTRODUCTION

It is a well-known fact that heavy metals are part of the soil structure, and they occur there naturally. Depending on their type and concentration, these metals can be harmful to environment, and further, to human health.

Besides the natural causes (erosions, volcanic eruptions) (Cyraniak et al., 2014), the heavy metal pollution can also originate from human activities like mining industry, transportation, urbanization, agriculture and others (Dumitrel et al., 2013). With a few exceptions (some sites with high levels of natural resources), the pollution in Romania was caused by population, through unmonitored industry centers (Boros et al., 2015; Muntean et al., 2010; Sur et al., 2012).

Although in general Romania has safe areas from agricultural point of view, there are still some sites where the heavy metal pollution exceeds the limits provided by laws in force. Even more, soil is considered as a key source of socio-economic development (Dimitriu, 2014).

The only study that offers complete information about the levels of heavy metals in all areas of Romania was conducted by ICPA (National Research and Development Institute for Soil Science, Agrochemistry and Environment) in 2000. The network of soil monitoring sites uses 942 investigation points spread in all the country, as 16 x 16 km squares. For this reason, the points rarely hit areas with severe problems regarding pollution. Yet, this kind of approach is most representative for soil characterization.

A similar report was published in 2011 by the same institution (Soil Quality Monitoring in Romania, Dumitru et al., 2011) but it does not provide the same detailed information.

Using this study as a base, the current review aims to update the existing data with others found in most recent studies, and also to add the plots with high values of pollutants found in other studies as well, to form some maps as accurately as possible.

MATERIALS AND METHODS

The most common heavy metals that are polluting the soil are cadmium, lead, nickel, chromium, zinc, cobalt, copper, arsenic (Masindi et al., 2017).

In order to create the map distributions for some of these metals, some parameters that describe the levels of interest from current law were used (Table 1).

Using the data from ICPA report, six distribution maps were created, one for each element shown above in the Table 1. Also, a table with some statistical data was created for each element. Only two studies that have expanded their research on the entire country were found.

Table 1. Reference values for trace chemical elements in soil (Order 756, 1997)

Element	Reference value (ppm)	Alert threshold sensitive soils (ppm)	Intervention threshold sensitive soils
Zn	100	300	600
Cd	1	3	5
Cu	20	100	200
Pb	20	50	100
Ni	20	75	150
Co	15	30	50

Apart from ICPA report, another study was performed with the same scope, in 2012 (Moldoveanu, 2014). It is interesting that the second study analyses a large range of heavy metals from both urban and rural areas. Unfortunately, there are some areas that are not covered at all. Since it not provides any numeric values, a direct comparison can hardly be made.

Other assays that have limited their research on a small area provided some valuable information to update the map distribution. All the studies that have been analysed had the same mode for preparing the soil samples: depth 0-20 cm, dried, grinded, sieved and analysed through different detection methods.

RESULTS AND DISCUSSIONS

Each metal has been analysed separately in order to structure the information efficiently.

Zinc

Zinc is an essential micronutrient required by plants and it has multiple role in their development (Hassan et al., 2017). In many cases, the zinc deficiency is a very important problem that requires different amendments to raise its concentration (Berbecea et al., 2011). But there are some cases that due to different anthropogenic activities such as mining or industrial emissions the situation can be reversed (Dumitrel et al., 2013).

In Romania exists a few areas where the concentration of zinc exceeds both, alert or intervention threshold.

A series of studies highlights the zones who have encountered this problem and also offers some quantitative values (Buruiana et al., 2016; Dumitrel et al., 2013; Ene et al., 2010; Marin et al., 2010; Munteanu et al., 2012; Sur et al., 2012; Elekes, 2014; Nimirciag, 2012; Albulescu et al., 2012; Popa et al., 2016; Big et al., 2012; Gămăneci et al., 2011; Morar et al., 2010; Stefu et al., 2013; Ungureanu et al., 2017; Fâciu et al., 2012; Cojoc, 2011; Lăcătușu et al., 2009). These levels are shown in the table below, together with the average values from ICPA report (Table 2). The table contains only the counties were zinc levels were studied in recent works.

Table 2. Zinc concentration in different areas (ppm)

County	ICPA study					Other studies					
	Mean	Number of sites	Median	Min	Max	Mean	Number of sites	Median	Min	Max	Year
Alba	153.59	27	100	41	540	834	15	725	100	3997	2013
Bacau	51.27	26	50.5	20	111	75.48	95	-	24.97	126.38	2013
B Nasaud	84.19	21	65	35	275	578.14	35	197.5	58.7	5144.2	2012
Braila	84.38	21	60	32	235	88.80	3	92.75	70.80	102.86	2009
Caras-Severin	138.42	33	83	45	500	98.61	9	53.9	29.1	294	2012
Dambovita	99	18	7538		215	125.5	5	36.5	22	600.4	2014
Galati	155.06	16	170	25	250	57.66	8	58.16	33.62	72.69	2009
Gorj	68.91	22	65	37	110	61.56	17	52	31.8	133	2016
Iasi	83.05	22	49	23	405	45.36	1030	30.31	11.6	702.61	2008
Maramures	210.20	25	230	5	360	311.44	10	144.38	89.04	1110.2	2012
Mures	91.96	27	70	40	270	69.87	5	56.6	53.4	99.43	2010
Sibiu	95.75	20	68.5	35	255	463.4	94	-	64.04	1938	2011
Suceava	89.60	35	68	23	285	95.8	63	-	33.6	332.8	2013
Timis	193.06	34	200	36	500	60.43	18	57.62	39.82	98.72	2013
Vaslui	62.74	23	56	34	118	68.22	193	63	31	192	2017

Comparing the average values from ICPA study with the average values from other studies, it shows a significant difference between these set of values (T test result: 0.15). The difference is somehow explainable regarding the fact that most of studies directed their research on areas with problems. These areas are pointed out in the figure below (Figure 1). From a total of 1043 investigated sites, 767 (73.53%) have a smaller concentration of zinc than the reference value, 246 (23.58%) between reference value and alert threshold, 79 sites (7.57%) exceeds alert threshold and only 18 (1.72%) exceeds intervention threshold. These top values represent some well-known high polluted areas: Zlatna (Alba): 3997 ppm, Certej (Hunedoara): 2200 ppm.

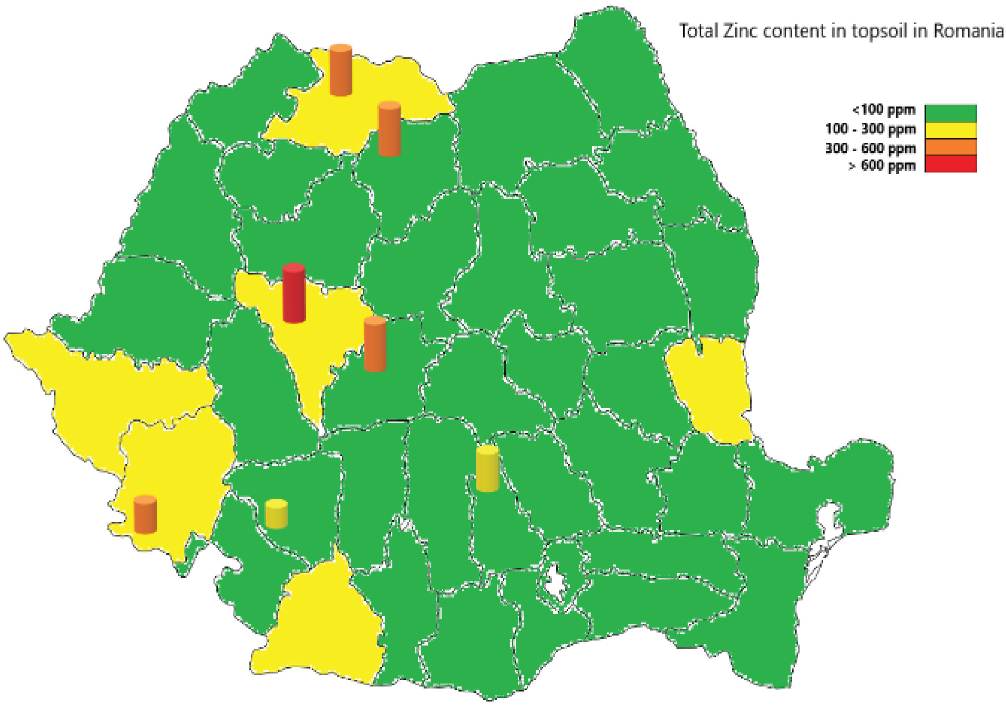

Figure 1. Zinc content in topsoil of Romania counties

Copper

Alongside zinc, copper is an important element in development of plants as well. Copper has an important role in metabolism and also participates to the respiration process (Laţo et al., 2012). Generally, a level of 4-20 ppm Cu is sufficient for any type of culture (Sutradhar et al., 2017).

A deficiency of copper can be possible as well, and this is one of the major constraints for crop productivity in many countries of the world (Corches et al., 2017).

The availability of copper depends on soil characteristics, such as organic carbon content, texture and pH and threshold values have been established as functions of these soil properties (Ballabio et al., 2018).

In Romania, almost all areas have sufficient quantities of copper, in some of them being in excess.

In the table below are presented the counties that have been mentioned in recent studies (Dinu et al., 2018; Suciu et al., 2008; Zgripcea, 2013; Popescu et al., 2013; Morar et al., 2010; Buruiana et al., 2016; Dumitrel et al., 2013; Ene et al., 2010; Marin et al., 2010; Munteanu et al., 2012; Sur et al., 2012; Elekes, 2014; Nimirciag, 2012; Albulescu et al., 2012; Popa et al., 2016; Big et al., 2012; Gămăneci et al., 2011; Morar et al., 2010; Stefu et al., 2013; Ungureanu et al., 2017; Hura et al., 2013; Iancu et al., 2008) to have this problem (Table 3).

Table 3. Copper concentration in different areas (ppm)

County	ICPA study					Other studies					
	Mean	Number of sites	Median	Min	Max	Mean	Number of sites	Median	Min	Max	Year
Alba	24.80	25	25	6	46	223.95	16	82.18	18.7	914.1	2013
Bacau	16.90	29	15	9	33	35.84	89	-	12.1	82.1	2013
B Nasaud	25.33	21	20	10	68	43.37	33	36.9	15.2	102.8	2012
Braila	31.14	21	25	12	125	33.87	6	31.28	25.06	52.78	2009
Caras-Severin	25.15	33	23	10	60	58.92	9	56.66	25.06	52.78	2012
Cluj	21.62	26	20	10	32	46.26	18	47.44	28.2	52.84	2008
Dambovita	20.47	17	20	6	50	125.5	5	36.5	22.0	600.4	2014
Galati	25.06	16	22.5	15	75	24.54	14	23.96	18.38	31.59	2009
Gorj	27.75	20	21	15	114	11.31	12	7.67	1	34.34	2015
Hunedoara	21.81	32	15	10	59	100.21	6	44.85	15.3	378	2018
Iasi	23.30	23	21	8	46	45.36	1030	30.31	11.6	702.61	2008
Maramures	19.52	25	20	5	40	112.98	10	80.11	33.29	310.82	2012
Mures	26.56	27	25	10	43	22.41	18	17.5	1.28	72.8	2008
Sibiu	26.38	21	25	10	49	31.73	56	-	9.03	114.6	2011
Suceava	23.42	36	20	8	75	36	63	-	17.85	112.75	2013
Timis	19.14	35	17	8	45	26.22	18	30.89	0	49.44	2013
Valcea	33.14	22	25	10	100	32.86	6	33.85	10.48	53	2013
Vaslui	23.26	23	21	8	45	29.83	193	27	14	300	2017

The average values from ICPA study and the average values from other studies are significantly different (T test result: 0.034).

Some major differences are shown on the map below (Figure 2).

A total of 1103 points were analysed. Less than half, 418 (37.89%) had a copper level smaller than the reference value, 663 (60.10%) points were situated between reference value and alert threshold, 14 (1.26%) points exceeds alert threshold but not the intervention threshold, and 7 (0.63%) exceeds intervention threshold. The highest recorded value is 914.1 ppm, near Zlatna region.

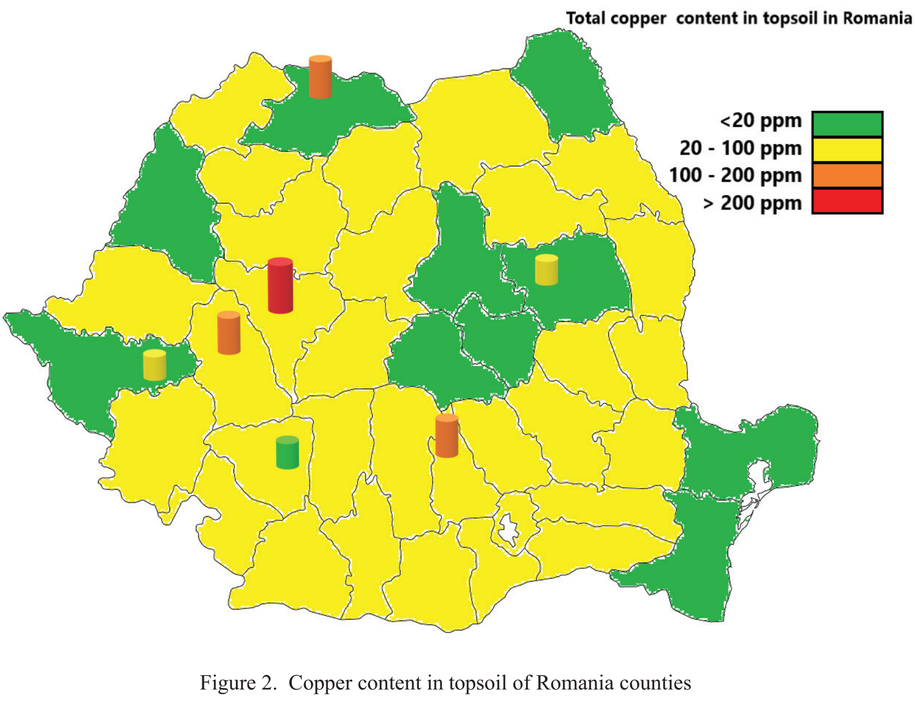

Figure 2. Copper content in topsoil of Romania counties

Lead

Lead is well known to be toxic and his harmful effects were intense studied (Roba et al., 2015). Unlike other heavy metals like zinc or cooper, lead has no role for plant or animal development. His presence in soils has only negative effects. Still, in a lot of areas the high concentration of lead causes serious problems, especially in zones with big industry centers. Some of these areas are mentioned in the table below (Table 4), according to the data presented in a set of studies (Mihali et al., 2013; Damian et al., 2013; Curcă, 2011; Dumitrel et al., 2013; Elekes et al., 2014; Bird et al., 2005; Buruiana et al., 2016; Sirbu et al., 2012; Dinu et al., 2018; Suciu et al., 2008; Zgripcea, 2013; Popescu et al., 2013; Morar et al., 2010; Buruiana et al., 2016; Dumitrel et al., 2013; Ene et al., 2010; Marin et al., 2010; Munteanu et al., 2012; Sur et al., 2012; Elekes, 2014; Nimirciag, 2012; Albulescu et al., 2012; Popa et al., 2016; Big et al., 2012; Gămăneci et al., 2011; Morar et al., 2010; Stefu et al., 2013; Ungureanu et al., 2017; Chirilă, 2013).

Table 4. Lead concentration in different areas (ppm)

County	ICPA study					Other studies					
	Mean	Number of sites	Median	Min	Max	Mean	Number of sites	Median	Min	Max	Year
Alba	32.11	19	30	13	84	207.24	18	0	0	1537	2013
Bacau	38.58	26	39	20	58	21.370	89	-	2.94	56.73	2013
B Nasaud	50.80	20	47	19	145	242.79	35	78.7	3.5	3687.2	2012
Braila	32.13	23	30	19	56	19.27	3	18.15	16.47	23.18	2009
Caras-Severin	43.19	32	40	7	98	31.00	9	25.59	21.63	47.08	2012
Cluj	30.41	27	30	15	51	75.36	18	0	0	735	2008
Dambovita	25.79	19	25	6	40	76.4	5	43.3	0.6	294.3	2014
Galati	19.81	16	20	10	30	20.17	8	18.54	6.50	35.72	2009
Gorj	39.50	22	35.5	28	67	10.89	12	1	1	64	2015
Harghita	43.31	26	35	20	84	102.08	12	67.5	30	260	2013
Hunedoara	41.86	29	30	15	99	373.5	6	270	120	888	2018
Iasi	32.65	23	28	14	69	27.73	1030	20.04	4.5	1995.43	2008
Maramures	32.77	26	30	10	81	365.27	10	261.88	163.28	804.09	2012
Mures	29.41	29	25	10	64	13.87	9	14.4	7.05	19.1	2010
Prahova	31.55	20	31.5	2	102	11.79	9	3.5	0	68.9	2006
Sibiu	43.05	19	35	20	170	680.9	56	-	19.61	2863	2011
Suceava	34.14	35	30	10	95	24.9	63	-	14.75	102.4	2013
Timis	21.51	35	20	11	40	6.58	18	0	0	26.09	2013
Valcea	33.45	22	33	15	45	16.90	4	14.84	12.43	25.5	2013
Vaslui	32.83	23	28	14	69	25.27	193	24	16	84	2017

The two sets of average values also differ significantly (T test value 0.049). The map shown in Figure 3 express some of the differences.

A total of 1115 points was used to obtain the map. Only 179 (16.05%) points had a smaller level than the reference value. The majority of sites, 759 (68.07%) had a value situated between reference and alert threshold. Surprisingly, a high number of sites, 131 (11.74%) have a high level of lead contamination, between alert and intervention threshold. Still, an impressive amount of 46 (4.12%) sites comes with a very high level of contamination, over intervention threshold.

Considering that some studies were focused on these areas, the results are expected. The highest value was found around Rodna mining perimeter (3687.2 ppm).

Cadmium

Cadmium is found among the most toxic elements. Like lead, cadmium is not an essential microelement and his presence in the environment in concentrations that exceed the normal values may seriously affect living organisms (Oprea et al., 2011). The normal content of Cd in soil is 1 ppm as defined by the Romanian regulations.

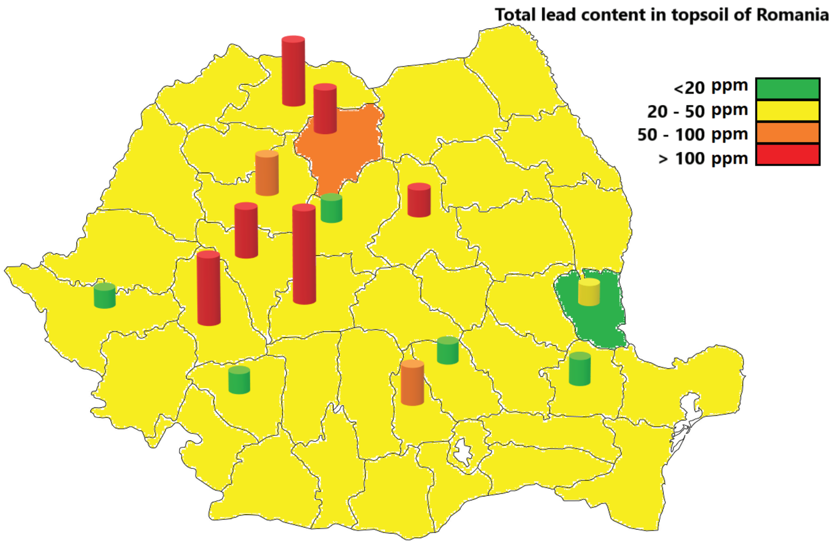

Figure 3. Lead content in topsoil of Romania counties

The cadmium pollution is described in the Table 5 (Buruiana et al., 2016; Ene et al., 2010; Popa et al., 2016; Albulescu et al., 2012; Damian et al., 2008; Stefu et al., 2013; Dumitrel et al., 2013; Oprea et al., 2011; Muntean et al., 2008; Fâciu et al., 2012; Iordache et al., 2015; Roşu et al., 2011; Chira et al., 2014; Sur et al., 2012; Nimirciag, 2012; Ungureanu et al., 2017; Dinu et al., 2018; Buzatu et al., 2018; Mandoc et al., 2013; Trîmbiţasu et al., 2006).

Table 5. Cadmium concentration in different areas (ppm)

County	ICPA study					Other studies					
	Mean	Number of sites	Median	Min	Max	Mean	Number of sites	Median	Min	Max	Year
Alba	1	26.00	1.00	0.1	2	0.53	4	0.42	0.35	0.95	2008
Bacau	0.9	27.00	0.83	0.5	1.1	0.595	89		0	1.45	2013
Braila	0.5	21.00	0.82	0.5	1.8	0.82	17	0.9	0.3	1.2	2009
Caras-Severin	1.5	34.00	1.54	0.9	3	2.06	9	1.85	1.28	3.22	2012
Dolj	1	29.00	0.82	0.4	1.6	0.6	2	0.6	0.05	1.15	2018
Galati	1	16.00	1.00	1	1	0.31	23	0.4	0	0.81	2009
Gorj	1	21.00	1.06	0.5	2.7	0.47	16	0.4	0	1.4	2015
Hunedoara	1	29.00	1.09	0.2	1.9	3.93	6	2.7	1.01	11.4	2018
Iasi	0.7	22.00	0.71	0.4	1.4	0.49	1030	0.36	0	15.44	2008
Maramures	1	25.00	1.19	0.5	2	3.8	10	2.4	1.52	12.6	2012
Prahova	2	21.00	1.88	1.1	2.5	0.59	9	0.5	0.2	1.05	2006
Sibiu	1.25	20.00	1.39	0.5	5	9.54	95	-	0.774	52.0	2011
Suceava	1	35.00	1.07	0.5	2	0.76	63	-	0.2	1.34	2013
Timis	1	35.00	1.11	0.4	1.5	0	18	0	0	0	2013
Valcea	1	23.00	1.12	0.5	1.8	1.96	5	2.1	1	2.1	2013
Vaslui	0.8	23.00	0.73	0.4	1	0.32	193	0.31	0.02	0.8	2017

In the cadmium analysis, the average values from ICPA study and the other studies are significantly different (T test = 0.307), as before. Form a total of 1035 point analysed, 708 (68.40%) are smaller than the reference value, 317 (30.62%) are situated between reference and alert threshold, just 6 points (0.57%) exceeds alert threshold to intervention threshold, and only 4 (0.38%) exceeds intervention threshold. The maximum value for cadmium contamination was 52.01 ppm, found in Sibiu County, near Copşa Mica, one of the world most polluted city.

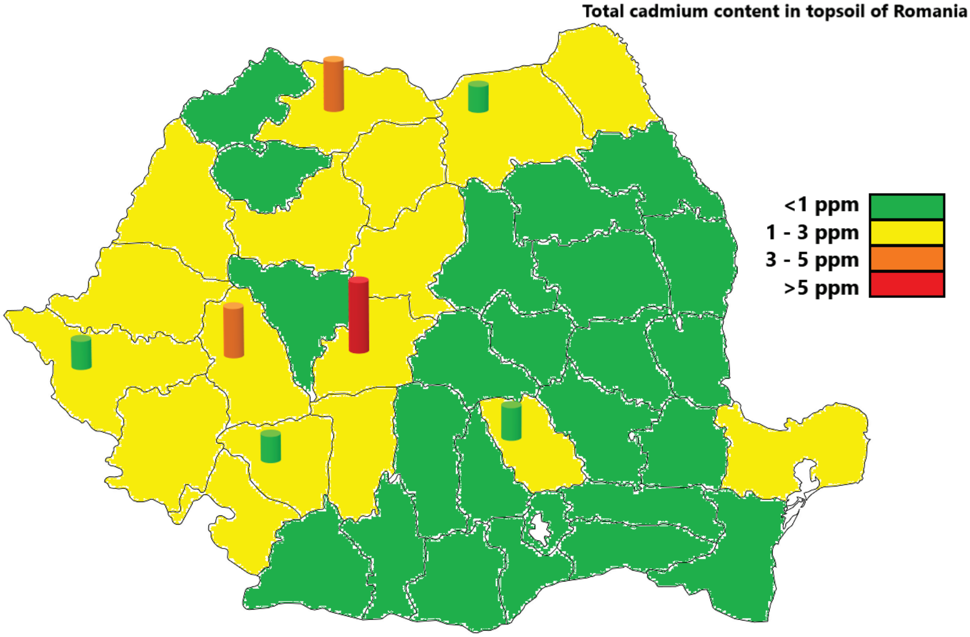

Figure 4. Cadmium content in topsoil of Romania counties

Giving an overview to the analysed heavy metals, the situation can be expressed in the chart below (Figure 5).

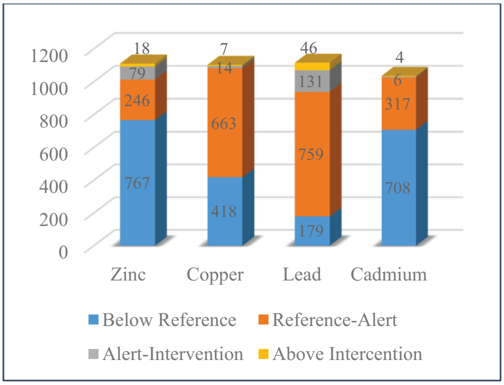

Figure 5. Distribution of examined sites by the pollution level

CONCLUSIONS

There are a lot of areas in Romania that are poorly investigated, investigated a long time ago or which have not been studied at all.

Even that the most part of Romania had safe lands for agriculture, there are some areas with very high pollution which exceeds Romanian regulations, according to the reviewed papers.

Copper and cadmium exceed intervention threshold in less than 1% of the examined points.

Lead is “leader”, regarding the points who exceeds reference value, with 83.95% of sites in this category. At the opposite pole, zinc has 26.47% of sites in this category.

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