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LEAD, ZINC AND COPPER IN THE BIOACCUMULATIVE HORIZON OF SOILS FROM IAŞI AND THE SURROUNDING AREAS

Cristian Vasilică SECU, Ovidiu Gabriel IANCU & Nicolae BUZGAR
University Al. I. Cuza, Iaşi, Bld. Carol I, no 11, 700506, Iaşi, Romania, email
cristian.secu@gmail.com
University Al. I. Cuza, Iaşi, Bld. Carol I, no 11, 700506, Iaşi, Romania, email
ogiancu@yahoo.com
University Al. I. Cuza, Iaşi, Bld. Carol I, no 11, 700506, Iaşi, Romania, email
nicolae.nizgar@uaic.ro

Abstract. Study of distribution of Pb, Zn and Cu in the bioaccumulative horizon of soils in the municipality of Iaşi and its surrounding areas indicates differences of concentration between urban and rural spaces. In the urbanised area, the bulk of the concentration is Pb, with an average of 60.5 mg/kg, and Zn, with an average of 207.2 mg/kg, both elements being associated with urban technosols. Pb has a larger area distribution, being associated with industrial units, the areas flanking roads with heavy traffic, and around fuel dumps. Zn can be observed in obvious concentrations in the industrial zone, with values reaching the lesser sensitive alert threshold (0.29%) or the lesser sensitive intervention threshold (1%). As a result of the position of the industrial zone and the prevailing wind direction, a non-uniform concentration of zinc can be observed in the soils (regosols, erodosols, black earth) at the level of the north-facing slope that flanks the urban zone but also in aluviosols at the level of the River Bahlui flood meadows to the east of Iaşi. Cu, in contrast to the other two elements, has a greater concentration in the viticultural and pomicultural area to the S and N of Iaşi, and is primarily associated with horticultural anthrosols. Tests whose values reached or exceeded the less sensitive intervention threshold are situated in the vineyard to the south of Iaşi (Bucium), which is situated in the vicinity of farm outbuildings (tanks, stores etc.).

Key words. heavy metals, pollution, soil cover, land use, SIG analysis

1. INTRODUCTION

Studies on heavy-metal soil pollution in Romania have focussed on mining and steel industry areas (Răuță et al., 1988, Damian et al., 2008) and to a lesser extent on urban (Bulgariu & Bulgariu, 2007, Lăcătuşu et al., 2008) and peri-urban spaces.

This paper will present the natural geochemical background for Pb, Zn and Cu in the bioaccumulative horizon of soils in Iaşi and its surrounding areas, as well as the effects of human activity in polluting the soil with these elements.

After 1989, the metallurgic and motorcar manufacturing industries in Iaşi have seen a decline in production against a backdrop of restructuring of sales markets. Some units have disappeared, with retail complexes springing up in their place. Traditional winegrowing plantations have diminished in size as a result of the Municipality of Iaşi extending its territory or through partial replacement with other crops.

The map of land use is necessary in order to take measures that stipulate restriction of human activity in certain areas, to reconstruct natural ecosystems etc. (Van der Windt et al., 2007), and a knowledge of soils polluted with heavy metals is an important stage in planning land use.

2. MATERIALS AND METHODS

2.1. Site location

The area studied is in NE Romania (Fig. 1), representing a polygon of 258.8 km^2, which includes the Municipality of Iaşi and 28 rural localities.

Between the hilly relief to the north (180-220 m in altitude) and to the south (reaching an altitude of 340 m), lies the Bahlui River flood plain, which to the east continues into the Jijia flood plain.

Figure 1. Study area position, Google-Map Data, 2008

2.2. Materials and methods

Soil samples (ca. 1,500 g) were collected from the bioaccumulative horizon or from other surface horizons, in the case of eroded soils. Sampling was carried out according to a grid of 500 x 500 m, over a surface area of approximately 16 x 16 km^2, with 1,023 samples being collected. The squaring was adjusted depending on local obstructions (buildings, lakes, water courses, etc.). Chemical analysis of heavy metals (Pb, Cu and Zn) was carried out using the Atomic Absorption Spectroscopy method (AAS – Solaar type) and X Ray Fluorescence Spectrometry (EDXRF Epsylon 5). pH analysis was performed using a Corning M-555 pH/Ion Meter. Cartography of land use was carried out using the Corine Land Cover (CLC) methodology (Büttner & Kosztra, 2007), adapted to the specific local terrain. For the maping of soils and land use we used topographical maps and plans, satellite imaging (ASTER 2000 and SPOT), and analysis of soil profiles. The results of analyses for heavy metals were processed using TNT mips 6 software and superimposed upon SRTM (2004).

2.3. Soils and land use description

The soils specific to the area under study, in accordance with SRTS (2003) nomenclature, are typical chernozems (Chernozems, WRB-SR, 2006), and are frequent at the level of the long south-facing slopes (Rusenii Vechi and Cristeşti Hills) and terraces of the Bahlui River. Cambic chernozems, situated at an altitude higher than typical chernozems, are to be found on Rediul Aldei Hill, Patrici Hill etc., while argic varieties can be found insularly on less abrupt inter-fluvial summits and only beneath forest vegetation.

Molic preluvosols evolved over a long period beneath forest vegetation, but are now in agricultural use, and are more frequent in forest clearings.

The class of anthrisols is represented by both types: erodosols and anthrosols (Anthrosols, WRB-SR, 2006). Erodosols are more frequent on the right slope of the Bahlui River, a cuesta ridge, in areas where the forest has been replaced by meadows and in those where diluvia have been shaped by landslides. The hortic anthrosols (Hortic Anthrosols, WRB-SR, 2006) are associated with the agriculturally terraced slopes, planted with vines and fruit trees, of Şorogari, Opincii and Cetăţuia Hill.

In the class of protisols, the following have been identified: aluviosols, regosols, and technosols. The aluviosols (Fluvisols, WRB-SR, 2006) of the Bahlui River flood plain connected to the Municipality of Iaşi have morphological properties strongly modified by the position of various structures; those in the vicinity of the drainage canals, at the eastern extremity (at the confluence with the Jijia) and western extremity have partially lost their stagnic and gleic characters; those in the Jijia flood plain have gleic characters in the abandoned meanders and in places are salinised. The urbic technosols (WRB-SR, 2006) are representative for the territory of the Municipality of Iaşi, where in the abandoned sectors of the Tomeşti household waste dump garbic technosols (WRB-SR, 2006) have formed.

The chemical characteristics of the soil as well as the organic carbon content, pH, forms of oxides, carbonates and some physical properties, such as clay content, can influence the content of chemical elements (He et al., 2005, Horckmans et. al., 2005). Chernozems in the area of Cetăţuia Monastery have a weak alkaline reaction (7.64) and clayey texture, and the proportion of humus is on average 3% (Căpşună et al., 2005). In the higher areas of the southern part (Opincii Woods and Săndulea), the soils are well supplied with humus (>5%), have a weak acid pH (6.5), calcium carbonate no longer appears in the bioaccumulative horizon, and the texture varies in profile from dusty-clayey to clayey (Filipov et al., 2005).

On the Bahlui flood plain, the soils have physical and chemical characteristics that are slightly differently determined by the phreatic, the presence of salts, the stratification of sedimentary deposits, and man-made influences in pedogenesis. The garbic technosols in the perimeter of the Tomeşti household waste dump have an alkaline reaction (>8), the humus content is large in Am (3.06%), the level of saturation in bases is very great, and the texture is clayey (Murariu et al., 2007).

The total distribution of $CaCO_3$ in the soil differs depending on the pedogenetic evolution of the soils, erosion processes or man-made impact. Thus, the black soils can reveal reduced concentrations of carbonates (often below 1%) even in the bioaccumulative horizon (Am), in zones affected by erosion where the carbonato-accumulative horizon appears at the surface, its values increase significantly (>8%). In the urban space, carbonates often originate from sources allochthonous to the soil, such as the rubble resulting from building work, the contribution made by wind-borne particles, building abrasion etc. (Norra et al., 2006).

pH plays an important role in the accessibility or immobilisation of heavy metals. Thus, the capacity of the soil to retain heavy metals increases simultaneous to pH, reaching a maximum at around neutral levels, and accessibility for plants decreases for Cu and Zn when pH increases within the interval 5-8 (Adriano, 2001).

Analysis of *land use* has allowed the separation of four classes, each having 1-4 categories (Tab. 1). In the sample under study, the reduced pollution of the soils is explicable also by the percentages that are ascribable to land use, with separation of agricultural land, permanent crops (vines) and rural spaces whose polluting potential is still reduced (Tab. 1).

Table 1. Characteristics of classes and categories of land in Iaşi and the surrounding areas

Class of land	Categories		Surface area (km^2)	Surface area (%)	Sources of pollution
Artificial areas class	*The urban space*		34.6	13	Road, rail, and air traffic etc.
	Rural localities		27.7	11	Small industrial units
	Industrial, retail, and transport spaces		7.6	3	The steel, plastics, building materials, food, transport etc. industries
	Artificial recreation spaces		0.9	0.1	Unidentified
	Household waste dump (Tomeşti)		0.3	0.1	Unsorted waste
Class of agricultural areas	*Arable land*		77.9	29	Fertilisers and fungicides
	Heterogeneous agricultural areas		39.2	15	
	Permanent crops	*Vines*	20.5	8	Fertilisers and fungicides
		Orchards	7.1	3	
Class of forests and semi-natural areas	*Plantations with native and foreign species*		6.9	3	Unidentified
	Natural meadows		31	13	Unidentified
Class of natural or artificial water areas	*Rivers and lakes*		1.2	0.1	Unidentified

Possible sources of heavy-metal soil pollution, also presented in the literature, are those that belong to the steel industry, due to the dust resulting from smelting, which pollutes the soil with zinc and lead (Schulin, 2007). To these can be added those that utilise pigments in the ceramics and plastics industries, and which involve a risk of Pb pollution. Some industrial objectives have reduced their activity, while others have disappeared. In the last category, we can mention the Nicolina Mechanical Plant, which initially repaired boxcars and locomotives, and later was equipped with stations for preparing asphalt mixtures and melting bitumen etc.; the Agricultural and Food Industry Mechanical Works had a smelting section (Barbu & Ungureanu, coord., 1987); on the site of these plants, retail complexes have been built, and some factories and areas of land are presently in a state of conservation.

In order to limit air pollution, the majority of industrial units were sited in the SE of the city, taking into account the prevailing wind direction (NW-SE) (Ungureanu et al., 2002) and wind speed (5.1 m/s NW and 4.2 m/s N), but at certain points some objectives remained within the interior of the city (the electro-thermal plant) or at the western extremity (SC Antibiotics SA).

Compared to the urban space, the sources of heavy-metal pollution of arable land are reduced, but not excluded. Thus, use of pesticides and fertilisers, which contain phosphates, can constitute a source of Pb and Cu pollution (Chen et al., 2008).

Modification of land use in the last two to three years has led to the expansion of the Bucium district into the viticultural zone, and the passing of viticultural parcels into private hands has led to a decline in this plantation, with some parcels having a complex use, which inscribes them in the category of heterogeneous agricultural areas. While in the first case the soil cover is profoundly disturbed, potential pollution is lost when the soil is mobilised by building construction, in the second case the values continue at high levels even if the land has a different use.

3. RESULTS AND DISCUSSION

3.1. Statistical analysis of concentration of the heavy metals

Of the total number of samples analysed for ***Pb***, half (50.2%) fall within the limit of normal values (NV) (≤20 mg/kg). The most accentuated diminution in the proportion of the samples comes at the transition from the sensitive alert threshold (SAT) (41.85%) to the sensitive intervention threshold (SIT) (1.38%). The downward curve reaches sub-unitary values together with the transition from the lesser sensitive alert threshold (LSAT) (5.8%) to the lesser sensitive intervention threshold (LSIT) (0.29%).

In the area studied, the types from the class of chernisols (typical and cambic chernozems), used in agriculture, are characterised by the lowest values for Pb content (<20 ppm), which is confirmed at a global level also by molisols from China (with an average of 18 ppm, Chen et al., quoted by Adriano, 2001), these being close to those of parental materials such as clay (16-50 mg/kg) and marl (He et al., 2005).

Statistical analysis of the samples for ***Zn*** reveals that 78% of them fall within NV (100 mg/kg); the interval between NV and SAT (300 mg/kg) is 18.5%; 2% of the total samples fall between SAT and SIT (600 mg/kg); values decreases sub-unitarily for LSAT (0.29%) and remain at a very low level (1%) for the LSAT – LSIT interval.

Of the total number of samples analysed for ***Cu*** only 3% fall within NV (31 samples) and 89.2% correspond to the NV-SAT interval. The situation is not worrying because the values regarded as normal differ according to national or regional legislation. Thus, in Danube flood plain of the Donau-Auen national park (Austria), the maximum values have reached 91 mg/kg^{-1}, slightly below the recommended maximum for EU agricultural soils (Graf et al., 2007).

Beginning with the SAT (100 mg/kg) – SIT (200 mg/kg) threshold, the proportion of the number of samples decreases considerably (5.8%) towards LSAT

(1%), becoming sub-unitary in the case of LSIT (0.79%).

Comparative analysis of the average concentrations of the three elements in the bioaccumulative horizon of the soil indicates greater averages in the urban as compared to the rural space, highlighting the influence of industrial activity on the soil (Tab. 2). With the exception of Cu the maximum concentrations are also associated with the urban space and the minimum concentrations in the urban zone are greater than those of extra-urban zones.

Table 2 Statistical parameters of heavy metals content in upper soil horizon (mg/kg)

Indicator/element	**Pb**	**Cu**	**Zn**
	The entire territory studied		
Average	27.4	44.7	111.0
Maximum	1995.4	702.6	5624
Minimum	4.5	11.6	10
Median	20	30.1	69.7
STDV	66.4	45.5	271.5
	Urban space (Iaşi)		
Average	60.5	47.8	207.2
Maximum	1995.4	188.3	5624
Minimum	7.4	20.6	14.9
Median	37.9	36.7	109.1
STDV	150.4	33.3	463.7
	Rural space		
Average	20.1	43.7	90.5
Maximum	209.8	702.6	4460.8
Minimum	4.5	11.6	10
Median	18.6	28.3	67.2
STDV	12.5	47.4	201.7

Geochemical comparisons of soil samples from the urban and rural space, identified on the basis of multi-varied statistical analysis, indicate a more reduced Euclidean distance for Pb and Zn in the urban compared to the rural environment (Fig. 2). Analysis has been preferred only for these two elements inasmuch as average values are close and comparison can be made at the same NV (20 mg/kg) in Romanian legislation.

3.2. Sources of pollution and spatial distribution of heavy metals

The distribution of **lead** in the bioaccumulative horizon differs according to the typology of the soils, the parental materials upon which these have formed, the manner in which the land is used, and, above all, the current or past impact of industrial activities on soils.

The largest concentration of samples whose Pb content exceeds NV are associated with urban technosols, respectively with the urban space of the Municipality of Iaşi. On the basis of the above we can conclude that the presence of Pb within the intervention and alert limits is the exclusive result of man-made intervention. The possibility of capturing exactly lead pollution of soils in the Municipality of Iaşi is conditional upon two factors. The first relates to deviation of the soil sample collection point from the established network, caused by the presence of buildings or other types of structure, which profoundly modify the soil (railway sidings). The second restrictive factor resides in the mixing of the soil horizons along certain alignments (along the courses of heating ducts, drains etc.), coverage of the soil with earthy material, generally originating from the bioaccumulative horizon, in the area of green spaces, and even uncovering and replacement of the bioaccumulative horizon. All human activities induce the risk of collecting samples that have a very large or very small concentration of a certain element.

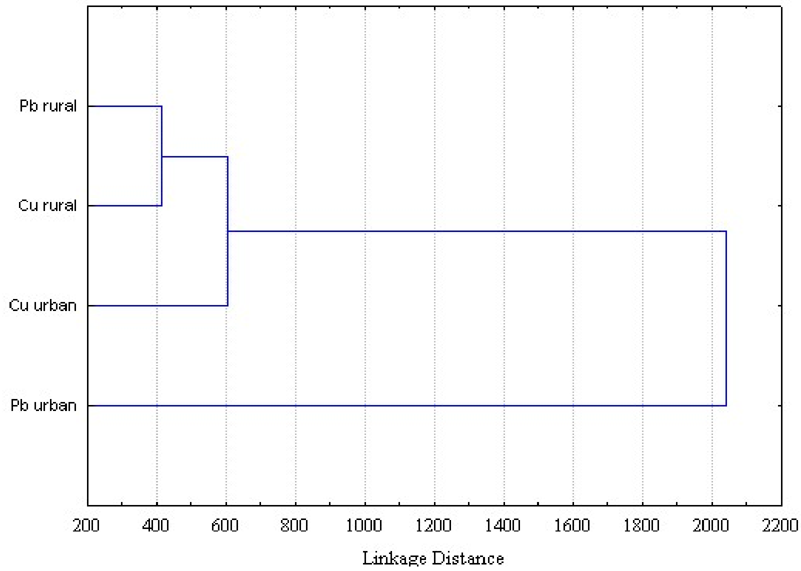

Fig. 2. Tree diagram for the Pb and Cu relation in the rural/urban space

In the Municipality of Iaşi, both distribution and concentration of lead in the soil are varied. The concentration has NV (<20 ppm) only in a few parts in the N, corresponding to the Copou district, and E-NE, corresponding to Eroilor Cemetery, Armenesc and the Ţicău district. Normal levels of lead in the soil are the result of good protection of the soil cover over a prolonged period of time, within the framework of man-made ecosystems. Soil protection in the Copou district is assured by the way in which structures are combined (buildings with a small number of storeys and in a low density) plus forest and shrub vegetation, which limits the penetration and spread of traffic pollution from adjacent areas.

The greatest proportion of samples falls in the SAT (50 mg/kg) – SIT (100 mg/kg) interval. From analysis of the spatial distribution of samples in this interval, there are no obvious poles of concentration at one or other of the two thresholds (Fig. 3).

Nevertheless, what can be remarked is an E-W alignment in the flood meadow common to the River Bahlui and Vămeşoaia Stream, continuing W into the flood plain of the Nicolina River, along a line to the Frumoasa district. The accumulation of Pb in the soil above NV is put down to activity in the industrial zone and the concentration of railway lines for freight traffic. The second, smaller surface area is situated between the railway line, to the W of Iaşi Station and the former auto repair plant.

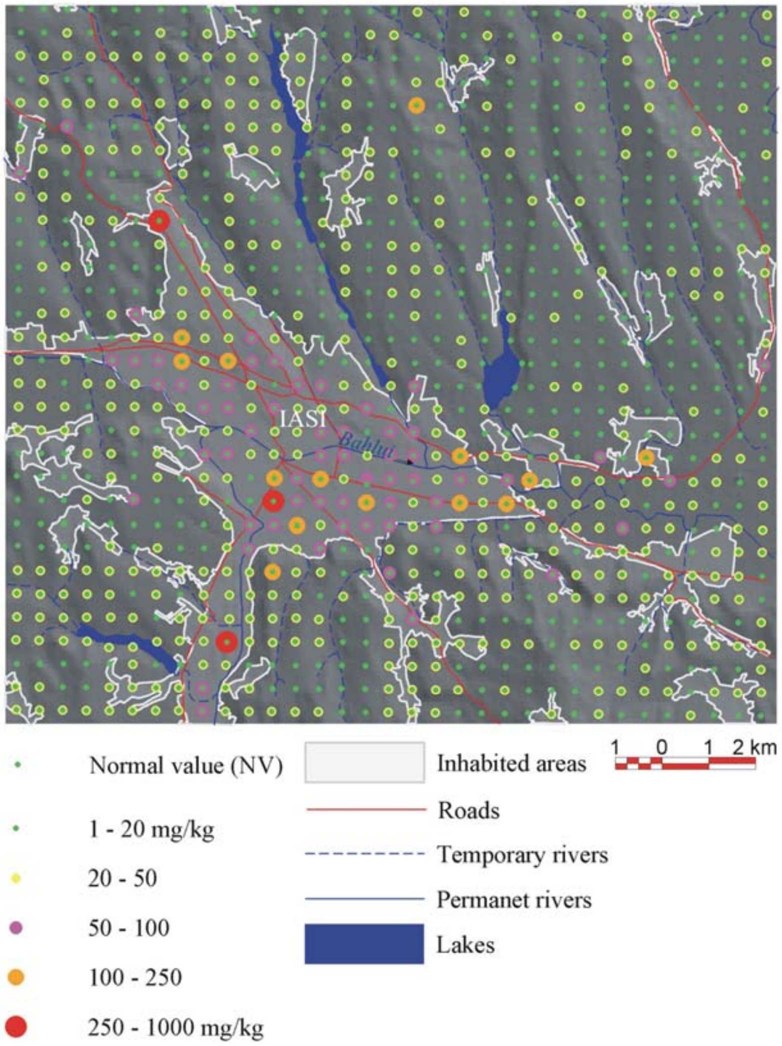

Figure 3. Distribution of Pb in Iaşi and the surrounding areas

The concentration related to LSAT (<250 mg/kg) is connected to industrial units (near the Electro-thermal Plant on Tudor Vladimirescu Boulevard at the intersection with Ţuţora, between the former Cigarette Factory and the PECO Fuel Dump) and the nearby transport routes (Ţuţora Boulevard) or the heavy traffic routes (Păcurari Road). It is estimated that the man-made source of Pb in the soil from the modern period is connected to the lead tetraethyl content in petrol (Sze et al., 2004).

The samples whose values fall within LSAT (350 mg/kg) and LSIT (1,000 mg/kg) or even exceeding the latter threshold are situated in the proximity of industrial units that include smelting sections (the former Nicolina Mechanical plant and Fortus SA), and for the third point accidental pollution is possible, in the northern part of the Municipality of Iaşi.

In the zone adjacent to the Municipality of Iaşi, the Pb content in the bioaccumulative horizon (Am) of the soil falls within NV (<20 mg/kg), excepting a few points (1.2% of the total number of samples), included in SAT and SIT. In this case too, where NV is exceeded, there can be found transport activity or small workshops for the repair of agricultural equipment. Thus SIT is exceeded in the area of Holboca, near the former Agricultural Mechanisation Station, and SAT was recorded to the east of Cristeşti, near the railway station and in the perimeter of Breazu locality.

In the literature, there is mention of a decrease in the concentration of ***zinc*** with depth, while presence of the element in excess is the result of activities specific to the steel industry (Horckmans et al., 2005). The mobility of zinc and other elements (Cu and Pb) is strongly dependent upon pH: the least mobility is specific to the neutral/weak alkaline interval, while modification of the reaction towards the high alkaline, through use of amendments, leads to an increase in mobility (Kumpiene et al., 2008).

Spatial distribution of Zn concentration reveals differences between the urban space, where the majority of samples exceed NV allowed by national legislation (Order no. 756/1997) and the extra-urban space, where, with a few exceptions, the concentration of the element is regarded as being normal (Fig. 4).

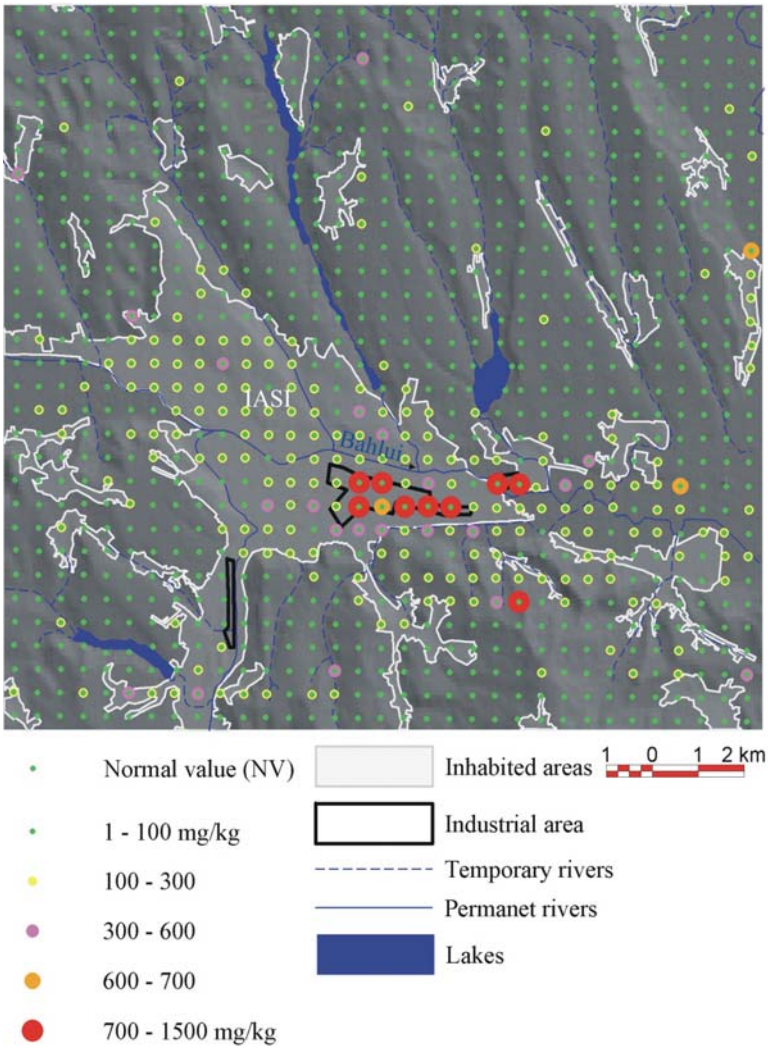

Figure. 4. Distribution of Zn in Iaşi and the surrounding areas

In the Municipality of Iaşi, samples whose concentration does not exceed NV are associated with the "green belts" of the Copou and Galata districts, where there is still forest vegetation, and also in the districts where there are houses with gardens (Tătăraşi). The greatest number of samples from within the perimeter of the municipality fall within the NV-SAT interval. In the industrial zone, values are different, but all are in excess of SAT. Soil pollution with zinc in the sector mentioned previously is arranged linearly on either side of Ţuţora Boulevard, to the E of CET. The source of zinc in the soil is industrial activity (a factory making zinc coated pipes), ash from combustion of coal, and possibly sludge from the purification station, to which can be added motor traffic.

Outside the city of Iaşi, the concentration of zinc in the soil has NV, with the exception of the eastern part of the urban space, where there are two isolated points above LSIT, respectively at Holboca and on Vlădiceni Hill. The area differences of Zn content can be observed at the level of the soils of the Bahlui River flood plain, between the western part, with NV, and the eastern part of the city, where zinc concentration is below or over 300 mg/kg. The cause is pollutants from the industrial zone carried by the prevailing wind direction (NW-SE), without excluding a slight influence from the orientation of the flood plain (W-E) and the high ground to the S of Iaşi. This is also confirmed by the higher than normal values from the first alignment level of the hills that flank the industrial zone (Bălan, Vlădiceni, Mândrului and Cierului Hills). Supplementary Zn deposits have occurred up to 200 m in altitude, respectively as far as the zone in which masses of air have been capable of transporting solid particles. To the W of the previous sector, also at hill level (Cetățuia şi Găureni) normal levels of Zn in the soil confirm the hypothesis according to which the cumulative effect of pollution from the industrial zone and channelled movement of masses of air have only partially affected the hilly area to the S of Iaşi. Zinc can be propagated as far as 5-10 km in the proximity of industrial areas, the main sources being the electro-thermal energy industry, to which can be added pollution resulting from the internal combustion engine (Davydova, 2005).

Channelling of masses of air along the flood plains may create local differences. Thus, values of 100 and 300 mg/kg extend as far as an alignment that would correspond to the easternmost extremity of the Holboca locality, to the left of the flood plain, after which it proceeds into the right sector, as far as the confluence with the Jijia. This can be explained by masses of air being thrust along the Jijia-Prut Valley towards the right sector of the Bahlui flood plain. Samples from the sector of the Jijia flood plain between Golăeşti and Coada Stâncii, which fall within normal values, as well as soils that do not significantly change their characteristics are points of support for the particulate character of pollutant dispersion.

In the peripheral zone of the Municipality of Iaşi, zinc has NV in the soil, excepting the sectors presented above and an alignment that follows the main road and railway line in the area of Cristeşti.

The average values of ***copper*** in the soil are questionable, and the differences, in the opinion of some authors (Aubert and Pinta, 1977, quoted by Adriano, 2001) are correlated with climatic zones, reaching a few hundred ppm in the temperate zone. There is unanimous acceptance of Cu in the soil above NV in zones in which soils have been modified by human actions (Maisto et al. 2004), with the highest values being associated with surface area horizons, as a result of atmospheric interference. In the urban environment, pollution results from industrial activities (electricity, steel industry, waste incineration, motor traffic etc.) (Huang at al., 2007), and in the case of agricultural crops, copper is present in a large spectrum of fungicides, pesticides (EPA, 2006) and fertilisers (Shomar et al., 2005).

In the perimeter of the Municipality of Iaşi a very reduced number of samples

(6) have values within SAT. Due to territorial dispersion, it has not been possible to identify the source of pollution, but it may originate from pieces of copper wire or sheeting buried in the soil.

In the territory under study, the distribution of copper may correlate with the dispersion of vine plantations and orchards (Fig. 5).

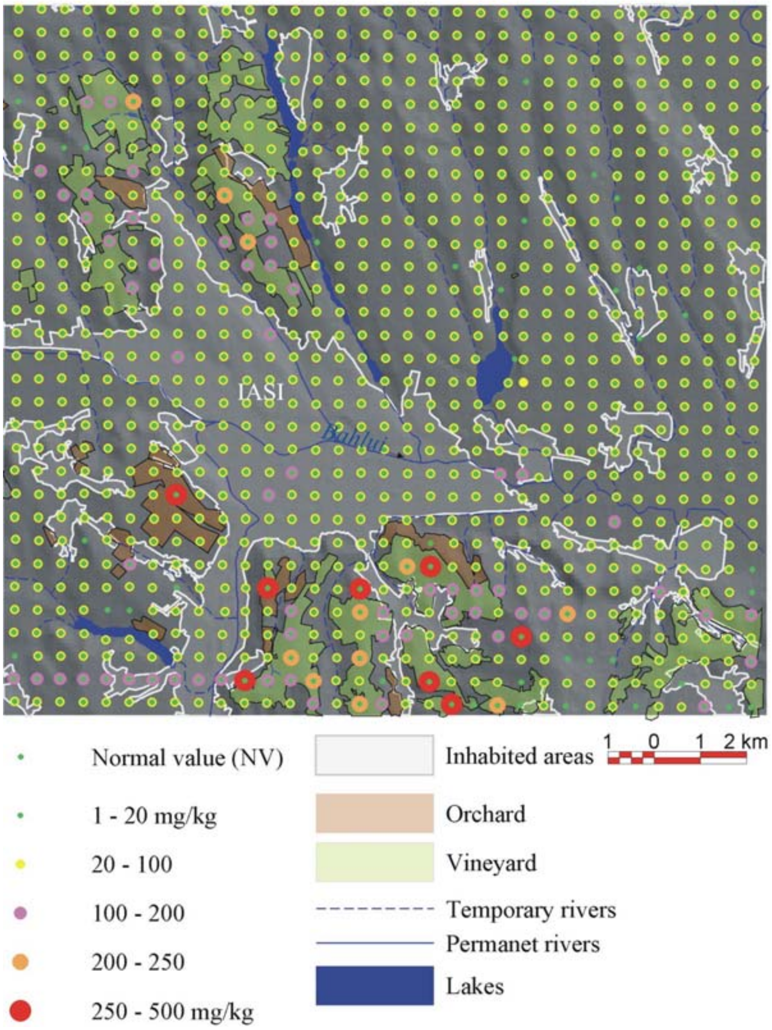

Figure 5. Distribution of Cu in Iaşi and the surrounding areas

To the north of Iaşi, analyses highlighted an area between the Ciric Valley to the east, Rediul Valley to the west, and the alignment of the localities Aroneanu-Şorogari-Cârlig and Breazu to the north, within the framework of which values fall between SAT and SIT (200 mg/kg). The presence of copper in greater concentrations in the soil is strictly associated with orchard and vine plantations, and returns to NV on the right slope of the Ciric Valley, which is covered with forest vegetation.

On the other hand, it is possible to remark a differentiation between the plots of smallholders, often cultivated with hybrid species, which necessitate a minimum of spraying, and to which correspond NV of Cu concentrations, and noble varieties, cultivated over large surface areas, owned by specialist producers, which are characterised by larger concentrations. Within the framework of the latter, application of fungicide has been carried out regularly, over a long period of time, if we take into account the fact that in 1957 the Iaşi Horticultural and Viticultural Research Station was set up (Bazgan & Bazgan, 2005), which administered the vineyards, but residues of the element in the soil may even be older, given that in the past the same land was also cultivated with vines.

4. CONCLUSIONS

Compared to the northern part, the orchard/viticultural area to the south of Iaşi is characterised by a higher number of samples within LSIT and LSAT. The samples whose values are situated below or above LSIT are, to a large extent, to be found in the viticultural zones (Bucium) but also around buildings or specific structures (stores, cisterns) on viticultural farms.

Alteration of land use in the last two-three years has led to the extension of the Bucium district into the viticultural zone and some plots of vines have entered into decline after passing into private hands, some of them having multiple uses and falling into the category of heterogeneous agricultural land. While in the first case, the soil cover is profoundly disturbed, in the second case values continue to be high even if the land is used for other purposes. The mobility of Cu in the soil is dependent on reaction, decreasing together with pH (Kumpiene et al., 2008), on organic content, clay (Adriano, 2001) and carbonates (Graf et al., 2000). Consequently, changes in land use and bioaccumulation, against a backdrop of alteration in the hydric regime in the sense of removing carbonates towards intermediary horizons, might lead in future to the mobilisation of copper.

The presence of copper (and Pb) in the upper horizon of the soil in the area under study is also explainable due to the electric characteristics of the latter, which determine a heightened affinity for organic matter in the soil (humus), and the absorption of elements by organic matter increases together with pH (Basta et al., 2005). Conservation of organic matter in the soil and maintenance within the normal values of soil reaction in the studied samples constitutes a premise for the immobilisation of heavy metals.

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