

SOIL POLLUTION IN THE HUNGARIAN-ROMANIAN BORDER REGION (VALLEY OF KÖRÖS-CRIȘ RIVERS)

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Abstract: The investigation assessed and evaluated the soil polluting activities in 30 Romanian and Hungarian settlements in the region of the Double, Black and White Körös-Criș Rivers. Pb concentrations are mainly higher than the limit values with maximum values registered in the area of landfill and near the roads. The concentration of Cu and Zn in soils of the region is below the alarming values. In some places Cd levels in soils reached the alert threshold. Plant samples mostly do not contain higher than permitted levels of heavy metals. One of the main goals of the study is to contribute to the future sustainable use of the environment and conservation actions in the region in order to implement regional development policy. We must therefore address the social activities that involve a risk of soil pollution in the region, especially in the Körös-Criș Valley. Science-based assessment of the level of pollution can lead to the development of implementing procedures to reduce the impact of these activities. The most important result of the quality and quantity analysis of polluting activities in the region is territorial definition of soil heavy metal load.

Keywords: Environmental protection, soil pollution, land use, spatial development, Romania, Hungary

1. INTRODUCTION

Soil quality conservation is one of the key elements of biotic preserve and improvement of the ecological status of the region's ecosystem. The soil pollution sources are diverse: agricultural chemical residues with toxic effects, industrial waste, sewage, pollution from transport and mechanization, indirect effects (oil spill), etc. The contaminated soil as an ecosystem habitat is not only harmful to the ecosystem, but also directly to humans, as people lose the supply of ecosystem services, which are bound to the ground.

Soil pollution in Romania and Hungary was investigated by a number of authors (Farsang, 2000; Nemeth-Conda et al., 2002; Covaci et al., 2003; Dragan et al., 2006; Lăcătușu et al., 2008; Farsang et al., 2009; Ferencz & Balog, 2010; Prundeanu &

Buzgar, 2011; Szolnoki et al., 2013).

It is now accepted that the historical routine use of **agrochemicals** (such as pesticides and fertilizers) may have resulted in undesirable concentrations of trace elements, such as As, Cd, Cu, Hg, Pb, and Zn accumulating in some soils (Van Gaans et al., 1995; Webber & Wang, 1995; Harris et al., 2000). Mermut et al., (1996) reported that the use of fertilizers and pesticides in agricultural lands has led to an increase of Zn and Cd concentrations in agricultural soils in Canada. The contamination of soil with elevated concentrations of trace elements can have adverse effects on soil biology (microbiological communities and invertebrates) and hence soil ecosystem function (Giller et al., 1998; Merrington et al., 2002). Elevated concentrations of trace elements in soils can also have phototoxic effects and result in trace element contamination of edible crops (Merry et al., 1986,

Cobb et al., 2000).

A limited number of soils shows slightly enhanced levels for As, Be, Cu, Co, Cr, Mn, Ni, Pb and Zn in Northern Belgium (De Temmerman et al., 2003). In most cases, this could be linked to the regional industrial activities, but in some regions, the Cu and Zn contents were slightly raised, possibly due to excessive fertilization with animal manure in the past. As, Cd, Cr, Cu, Mn, Pb and Zn show high concentration values in almost all topsoil samples in Northern Greece (Papastergios et al., 2004). Because information exists that connects these elements with the production and usage of fertilizers and pesticides, as well as with combustion of petrol, these human activities in the area could be, at least partially, responsible for their elevated concentrations. Significant accumulation of As, Cd and Cr was found in soils of arable land in China (Luo et al., 2007). Based on correlation and cluster analysis, it can be concluded that Cd and Zn originate mainly from phosphate fertilizer, Pb from the use of insecticides, fertilizers, and sewage sludge as well as air deposition, and Cu from copper-based fungicides, while As, Ni and Cr might come from parent soil material. Several authors have reached conclusions that elevated Cu concentrations in soil are related to the high use of fertilizers and pesticides (Facchinelli et al., 2001; Sanghi & Sasi, 2001; Yu et al., 2004; Acosta et al., 2011; Huzum et al., 2012).

The pollution of soils by heavy metals from **transport**, especially automobile sources is a serious worldwide environmental issue. Pb, Cd, Cu and Zn are the major metal pollutants of the roadside environments and are released from burning of fuel, wearing out of tires, leakage of oils, and corrosion of batteries and metallic parts such as radiators etc. (Akbar et al., 2006; Dolan et al., 2006; Yoshinori et al., 2010). The level of Pb had a correlation with the traffic density attributing its origin to vehicular exhaust in Dubai (Aslam et al., 2013). The high concentration of Pb from automobile has been reported previously by numbers of authors (Foner, 1987; Gratani et al., 1992; Bahemuka & Mubofu, 1999; Renberg et al., 2000; Yaman et al., 2000; Andrews & Sutherland, 2004; Finster et al., 2004; Yakupoglu et al., 2008; Yoshinori et al., 2010). It has also been confirmed that load on heavy metal contents in topsoils and their variability depend upon the traffic density and the distance (Motto et al., 1970; Bai et al., 2009; Nakayama et al., 2010).

One of the most environmentally harmful activities in the conventional waste management is dumping unseparated waste in **landfill** disposal (Esakku et al., 2003). Leachate from landfills contains a wide range of contaminants: dissolved organic

matter, inorganic cations and anions, heavy metals, and xenobiotic organic compounds (Christensen et al., 2001). The potential pollution caused by leachates is the result of several factors, including the release of ammonia, chlorinated and non-chlorinated organic compounds and heavy metal ions into the environment, all of which are toxic to living organisms (Baccini et al., 1987; Christensen et al., 2001). The problematic metal contaminations originated mainly from the disposed materials/wastes have lead to leachate seepage through the underlying soils (Elwali, 2005; Adnan et al., 2013).

Some key points relevant to **impacts of soil contamination on health** mentioned in Van Camp et al., (2004) report are: i) in most cases, soil pollution from point sources is unintended and happens due to handling spills or accidents or insignificant but continual losses/emissions; ii) consumer behavior and the industrial sector are contributing to the increase in the number of potential sources of contamination, such as municipal waste disposal, energy production and transport, mainly in urban areas; iii) in Central and Eastern Europe, many problems stem from past activities and poor management practices (inefficient technologies and uncontrolled emissions). One of the major impacts is groundwater contamination and related health problems; iv) monitoring is specific to each individual site and is not very representative of other locations, unless there are a larger number of similar sites; v) data on concentrations of contaminants at individual sites are not necessarily relevant for EU policy discussions; vi) the soil's capacity and resilience in terms of holding onto and transforming contaminants mean that damage is not perceived until it is far advanced.

Valley of Körös-Criş Rivers has a rich and delicate ecological balance of natural and cultural heritage. The area is suitable for development principles and objectives, and for recreational and tourism activities based on the regional regulation (National Development Plan of Hungary, Regional Operative Programme of South Plain, County Development Strategy of Arad 2007-2013). Concern for the natural, built and cultural heritage and good ecological status of socio-cultural and livable environment of the area are objectives for the local people and visiting guests.

The main objectives of this study are the definition of pollution sources and contamination level of soils and vegetation in the Valley of Körös-Criş Rivers in the Hungarian-Romanian border region. The test parameters were heavy metals Cu, Zn, Cd and Pb and phosphate (PO_4^{3-}) and nitrate (NO_3^-).

2. MATERIAL AND METHODS

2.1. Sampling Area

The analysis took place within the administrative boundaries of villages (176,000 ha) through which flow White (in Hungarian: *Fehér-Körös*; in Romanian: *Crișul Alb*) and Black (in Hungarian: *Fekete-Körös*; in Romanian: *Crișul Negru*) Körös-Criș Rivers (Fig. 1). They are located in one of the Great Plain depressions that have SW direction and are gradually raising towards the Bihar foothill areas in Romania.

The Hungarian part of the study area is a 1-10 m low-lying perfect plain. This landscape continues for a few km into Romania and gradually rises towards the Transylvanian hilly area. The region's rivers Black, White and Double (in Hungarian: *Kettős-Körös*) Körös-Criș Rivers played a key role in surface shaping. Springs of these rivers are located in the mountains belonging to Romania (Réz Mountains, Bihar Mountains and Metaliferi Mountains).

The water catchment area starts from 50 to 150 km east from the border and the majority of sediment is transported from here. The majority of the plain is thus interpreted as an alluvial cone of the rivers. The surface is covered from 20 to 50 m thick fluvial sediments (sand, loess infusion). Mainly there are developed alluvial meadow soils (Fluvisols) and saline areas (Solonchacs and Solonetz). Groundwater level is

at major depth in the internal parts of the landscape contrary to its peripheral parts. Climate is moderately dry with precipitation amount from 540 mm to 600 mm.

The Körös-Criș Rivers are the most important determinants of the natural environment in this area. The Double Körös-Criș River flows only in Hungary with a small part of the water catchment area located in Romania. The area has typical lowland conditions in terms of the slope of watercourses with a number of side-channels connected to Double Körös-Criș Rivers. Among them, the most important are Élővíz-channel, Vargahosszai-channel and Hosszúfok-Határér-Kölesér- main channel. The Romanian catchment area of the White Körös-Criș River (952 km²) is located in Ineu, Șicula, Zărând, Chișineu Criș and Pîlu administrative areas.

The study area is mainly used for agricultural purposes: farming sector (60%) and pasture utilization (17%). This is followed by forests and semi-natural areas (7%), villages (5%) and private gardens that belong to them (5%) and water surfaces represented by Körös-Criș Rivers and their backwaters (3%) and wetlands (3%).

Although the explicit aim was the examination of joined border area of two countries we noticed that they have a number of differences. The most important are that Romanian part of the research area is rather urbanized and more industrialized than the Hungarian.

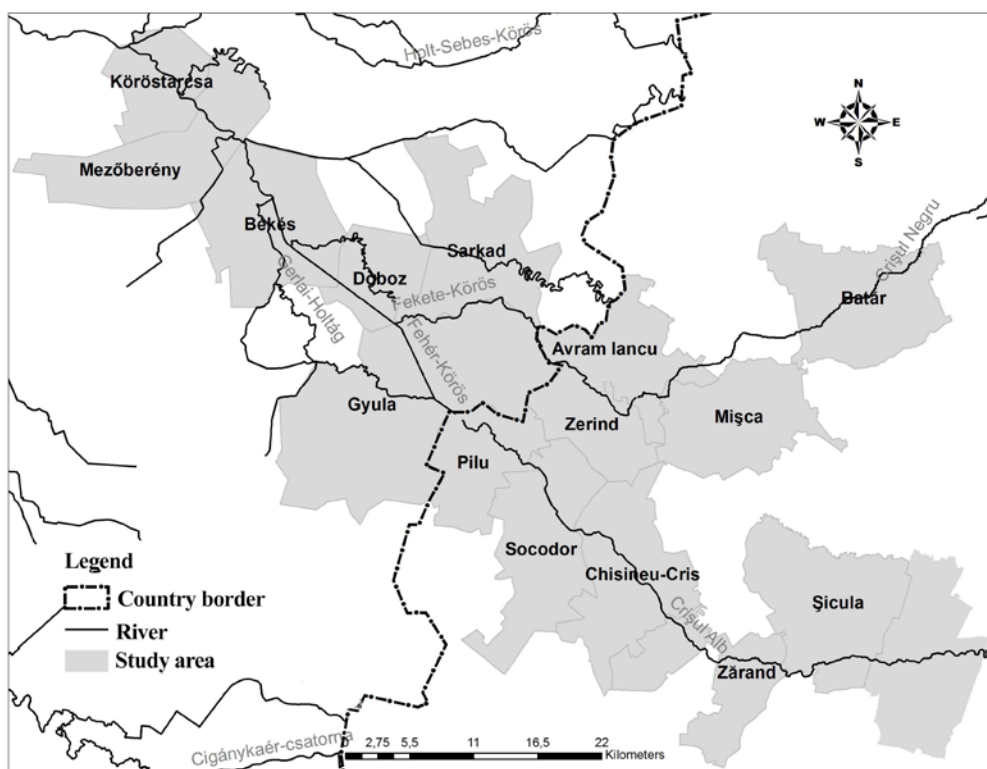


Figure 1. Study area with locations of settlements.

Nearly one-third of the research area (31.8%) is part of the Natura 2000 network, which is located in a larger share in Romania (72%) with 14% of the research area (8,193 acres) under legal protection. Comparing the two countries' Natura 2000 network, Romanian part of the network is characterized by agricultural and woodland areas with extensive cereal production and grassland management. Hungarian part of the network is characterized by salt marshes, dry prairies and wet meadows. Romanian part of the White Körös-Criș River (*Crișul Alb*, ROSCI0048) is mostly marshy, swampy with deciduous forest (41-45%), followed by extensive cereal cultivation and grassland management. The region between Black and White Körös-Criș Rivers (*Câmpia Crișului Alb* and *Crișului Negru*, ROSPA0015) is the largest part of the Natura 2000 network (70%) characterized by dominant grassland management interspersed with woods and corn-growing sites. In *Rovina-Ineu* protected areas (ROSCI0218) and in the most eastern part of the region *Câmpia Cermeiului* (ROSPA0014) are also present grassland and woodland landscapes interspersed by water bodies and marshlands. Upper part of the Romanian Black Körös-Criș River (*Crișul Negru*, ROSCI0049) is characterized by cereal growing areas, pastures and wetlands. *Nadab-Socodor-Varsand* (ROSCI0231) and *Lunca Teuzului* (ROSCI0350) are predominantly grassland areas (95% and 51%, respectively), while the *Crișul Alb* area between Gurahonț and Ineu (ROSCI0294) is mainly covered by forests (51%).

2.2. Data and Methods

As there is no single international pollution limits based on differentiated land use, both in Hungary and Romania - as functions of local conditions and planned usage – evaluation of necessity for intervention is performed under the risk analysis. Later on, these limits will help the general authority on decision-making in which the knowledge of local conditions often allow the remediation assessment.

According to Romanian legislation (Regulation of The Ministry of Environment on the environmental pollution, 1997; Emergency decree on the liability for environment, elimination and pollution prevention of

environmental damage, 2007; Government decision for soil testing methods, 2007; Government decision on soils and land ecosystem restoration, 2007) on soil contamination-sensitive land uses, soil groups of sensitive and less sensitive can be categorized (Table 1). Sensitive land use is in residential, recreational areas and agricultural land, while commercial and industrial areas can be classified among the less sensitive areas. The soil samples were mostly taken from sensitive soils, so this category was used for determining the limit values.

The Hungarian regulation (Decree of Ministry of Health on allowed chemical contamination limits of food, 1999; Decree of Ministry of Agriculture and Rural Development on mandatory specification for Hungarian feed codex, 2003; Common decree of Ministry of Environment and Water Management, Ministry of Health and Ministry of Agriculture and Rural Development on the geological and groundwater pollution limit values, 2009) of soils' limit concentration uses the B) contamination level to characterize the quality of the underground waters and soils. The B) value means hazardous concentration above that the soil or underground water is contaminated (Table 2).

Table 2. Heavy metals and (B) limit values of pollution in mg/kg dry matter.

Heavy metals	B limit values
Cu	75
Cd	1
Zn	200
Pb	100

Sources: 6/2009 (IV.14.) Common decree of Ministry of Environment and Water Management, Ministry of Health and Ministry of Agriculture and Rural Development.

Overall, Hungarian regulation of Cu, Zn and Pb alarm-pollution values are stricter, especially in the case of Pb where Romanian pollution limits are significantly lower. "Hot-spots", anthropogenic interventions in the environment, are defined on the basis of primary data sources (satellite images, maps 1:10.000 EOV, sectoral and regional development plan documents) and during site visits and interviews.

Table 1. Soil pollution alert and intervention values for Romania (mg/kg dry matter)

Elements	Sensitive soils			Less sensitive soils	
	Normal	Alarming value	Intervention value	Alarming value	Intervention value
Cu	20	100	200	250	500
Cd	1	3	5	5	10
Zn	100	300	600	700	1500
Pb	20	50	100	250	1000

Source: Regulation of the Ministry of Environment on the assessment of environmental pollution (756/1997)

The soil and vegetation sampling was performed at the same locations. The soil samples were taken manually from a depth of 5-10 cm. Plants were taken from the edges of agricultural fields and along the roads. Above ground parts of plants, stem and foliage were analyzed.

After defining the hot-spots, soil and plant samples from such regions were analyzed for the soil and vegetation contamination. The test parameters were Cu, Zn, Cd and Pb as heavy metals and phosphate (PO_4^{3-}) and nitrate (NO_3^-), because nitrogen and phosphorus content is a good indicator of the nutritional value of the soil. The extent of any undesirable over fertilization provides information that is of particular importance in protected areas. The main objectives were to investigate heavy metal content in the agricultural soils and to identify the potential environmental risks.

The heavy metal content in the samples was examined by ASA Atomic Absorption Spectrometry, (AVANTA) according to ISO 11464-98 and ISO 11047-99 standards. The determination of nitrate and phosphate content in samples was based on the STAS 7184/7-87 method (Determination of mineral salts of 1:5 aqueous extract). It was possible to correlate Romanian and Hungarian soil contamination thresholds (Pb: 6.0 mg/kg; Zn: 0.8 mg/kg; Cu: 2.5mg/kg; Cd: 0.9mg/kg).

The permitted heavy metal concentrations in vegetation in Hungary are governed by Regulation (Decree of Ministry of Health on allowed chemical contamination limits of food, 1999) and can be seen in table 3. By the Romanian regulation (Order 18/2007 of National Authority for Sanitarian Veterinary and Food Safety) the permitted Pb and Cd concentration are 30 mg/kg and 1 mg/kg of dry matter, respectively.

A total of 93 soil and plant samples were taken (Fig. 2). Based on the sources of anthropogenic pollutants, samples could be categorized into four groups: 1) Samples from

agricultural land (25), 2) Samples from industrialized area (20), 3) Samples from municipal and industrial waste landfills and sewage water treatment areas (16) and 4) Samples along the roads (15). Samples were also taken from sensitive and protected areas (17) (Table 3).

During the synthesis of the heavy metal content of the soil a simple cumulative process was derived from individual pollution parameters.

3. RESULTS AND DISCUSSION

3.1. Sources of pollution in the research area

The settlements along the Romanian part of Black Körös-Criş River have on average 2,000 inhabitants. This is mainly agri-rural area, where the major industrial activity is a paint factory in Satu Nou and the largest livestock farm in the area. The Romanian research area of the White Körös-Criş River is more urbanized (Chisineu and Crislineu) and industrialized with three industrial parks. Notable sources of pollution are two pig farms in Ineu and Mocrea settlements. The treated municipal and industrial wastewater effluent is going to the White Körös-Criş River. The two towns had a joint landfill until 2013. Wastewater and landfill sites are relatively significant sources of pollution in the region.

At the Hungarian side the primary pollution sources are the livestock farms (Köröstarcsa, Mezőberény, Békés, Sarkad), followed by the brickyard sites (Mezőberény, Gyula). The waste landfill in Doboz settlement is characterized by very bad conditions (here is measured the maximum level for Pb and other heavy metals). The results showed that one-third (34%) of the research area are least affected by human activities, of which 23% (14,000 hectares) are considered potentially threatened by the identified sources of pollution. Almost half of the sensitive areas are located along the roads and motorways (28,100 hectares).

Table 3. Maximum allowed heavy metals content in food (content of heavy metals, total weight, mg/kg)

Food groups types	Pb	Cd	Cu	Zn
Flour and other cereal flours (excluding the bran)	0.15	0.1	5	30
Rice	0.15	0.2	5	30
Eating bran (wheat, barley, rye, oats)	0.3	0.2	*	*
Pulses	0.2	0.1	*	*
Fresh and frozen fruits	0.1	0.05	*	*
Dried Fruit	1	0.5	*	*
Dried vegetable	1	0,5	*	*
Fresh cultivated mushrooms, mushroom products	0.3	0.1	10	20
Sunflower seeds, hulled (raw and fried)	0.5	0.6	*	*
Spices (except pepper)	2	0.2	*	*
Red pepper	2	0.25	*	*

Source: Decree of Ministry of Health on allowed chemical contamination limits of food (17/1999. (VI. 16.))

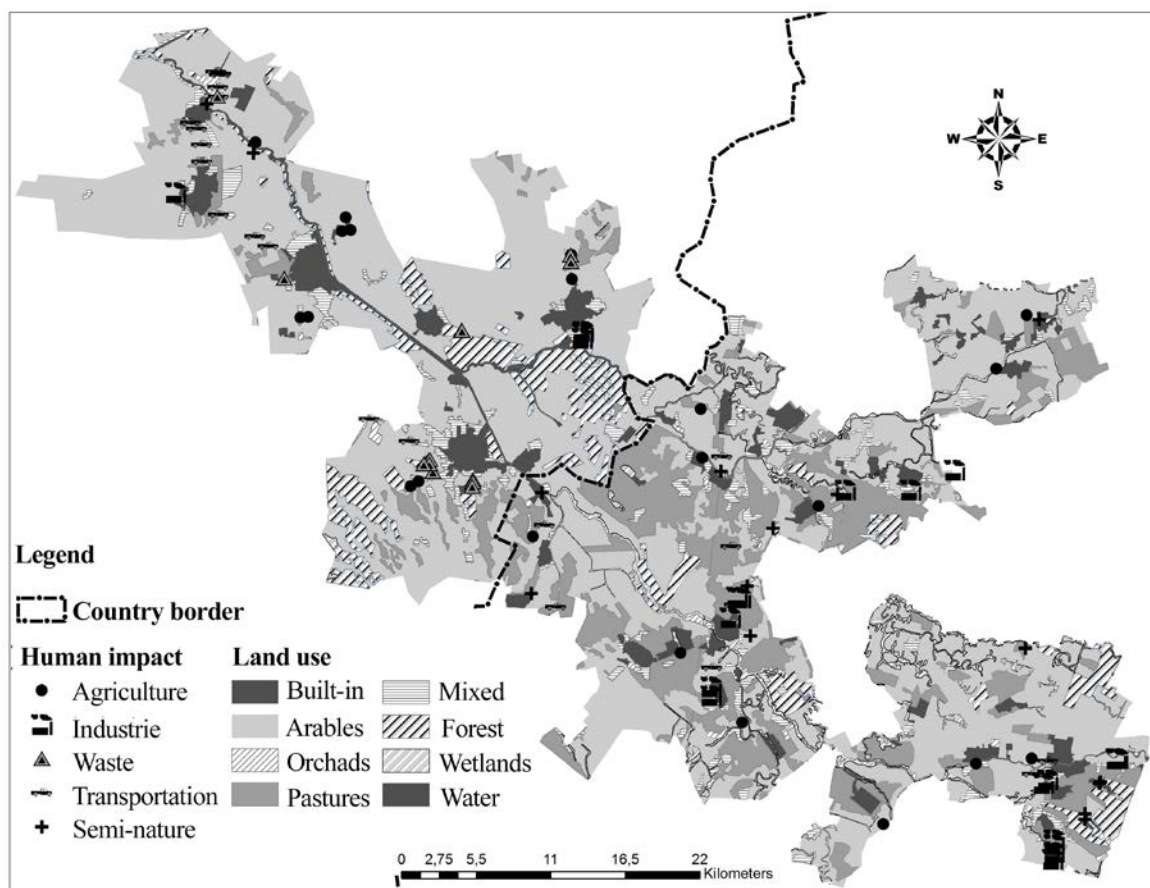


Figure 2. Sampling locations, human activities and land use types in the research area

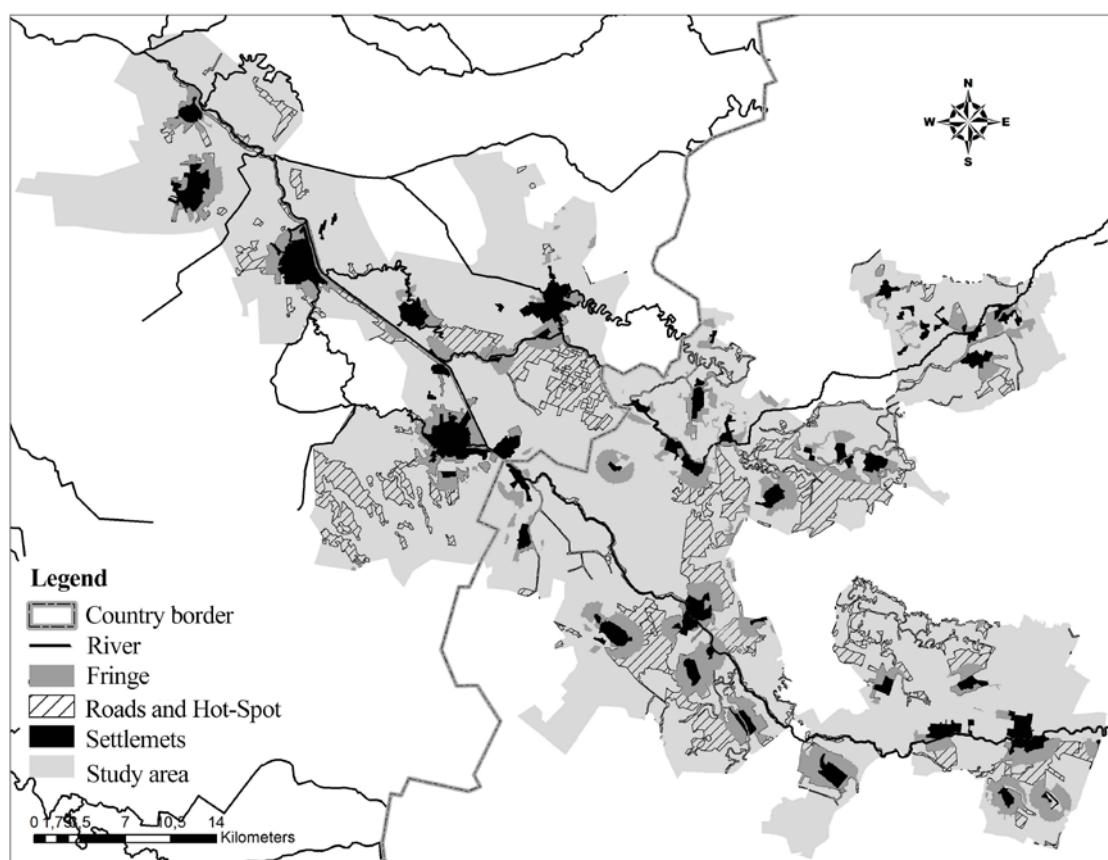


Figure 3. Semi-natural landscape affected by settlements and roads. Source: author's work based on CLC 2006

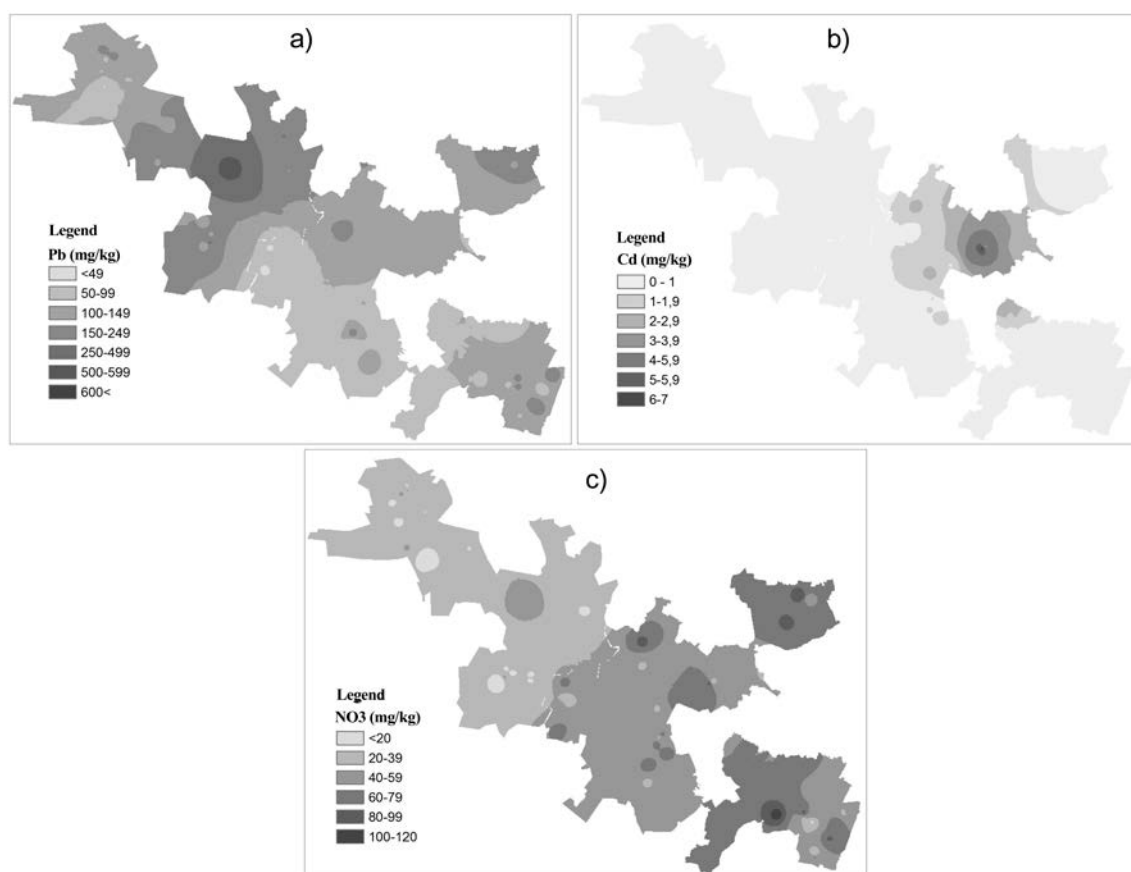


Figure 4.a) Pb b) Cd and c) NO3 concentrations in the research area

Table 4. Minimum, median and maximum level of analyzed elements (mg/kg) in soil and plant samples.

Elements	Statistics	Groups									
		Agriculture		Industry		Waste		Transportation		Semi-nature	
		Soil	Plant	Soil	Plant	Soil	Plant	Soil	Plant	Soil	Plant
Cd	Minimum	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Median	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Maximum	4.6	0.0	3.9	0.0	0.0	0.0	2.5	0.0	6.2	0.0
Cu	Minimum	8.6	0.0	9.2	0.0	18.8	0.0	11.2	0.0	6.3	0.0
	Median	20.3	0.0	17.3	0.0	22.7	0.0	20.4	0.0	21.5	0.0
	Maximum	26.8	6.2	33.5	6.1	39.9	3.4	40.5	6.2	35.1	5.8
Zn	Minimum	28.3	2.6	30.7	3.3	42.4	3.8	26.3	2.9	23.4	3.3
	Median	52.6	10.3	52.8	7.2	66.4	12.6	55.9	12.2	54.6	10.2
	Maximum	76.0	19.0	84.3	34.8	134.6	30.9	80.4	20.3	115.0	26.3
Pb	Minimum	34.1	0.0	55.5	0.0	85.5	0.0	48.3	0.0	45.9	0.0
	Median	126.4	0.0	110.8	0.0	155.1	0.0	119.1	0.0	77.5	0.0
	Maximum	276.1	11.2	178.9	9.8	7466.6	26.0	288.8	8.5	201.3	0.0
Nitrate	Minimum	-	2.5	-	5.0	-	15.0	-	15.0	-	24.0
	Median	-	35.0	-	34.0	-	25.0	-	35.0	-	65.0
	Maximum	-	110.0	-	75.0	-	53.0	-	75.0	-	86.0
Phosphate	Minimum	-	0.2	-	0.1	-	0.2	-	0.5	-	0.6
	Median	-	2.2	-	1.1	-	2.5	-	1.8	-	2.1
	Maximum	-	4.5	-	4.4	-	4.5	-	3.5	-	4.2

Another 17,000 hectares are natural landscapes located near the settlements (10% of the project area; (Fig. 3). Overall, one-third of the region is potentially vulnerable area of which 70% are natural landscapes represented mainly by meadow

and pasture landscapes (50%), followed by a complex farmland (18%). The remaining one-third of the area belongs to almost natural meadows and grasslands, forests, and water landscapes.

3.2. Soil pollution

Minimum, median and maximum level of analyzed elements (mg/kg) in soil and plant samples are presented in Table 4. One of the major pollutants is Pb. Its concentration is mainly higher than the limit value. In some regions Pb concentration exceeds the alarming value with more than 10% (Fig. 4a).

Highest heavy metals concentrations are along the main roads and their 50-meter buffer zones. In the less sensitive soils (see: Romanian regulation) samples taken from surroundings of landfills and industrial parks, the Pb concentrations are below the alarming value, but exceeds the research area (B) limit values (100 mg/kg dry matter). In this case, there is a risk of Pb contamination but still there is no need for intervention measures.

The concentration of Cu and Zn in soils of the region is below the alarming values. In more than half of the soil samples, Cd concentrations were below the alarming values, but because of the lower allowed values of Cd defined by Romanian legislation in some places it is reached the alert threshold (Fig. 4b).

3.3. Plant contamination

Plant samples mostly do not contain higher

than permitted levels of Pb and Cd concentration. The low concentrations of Cu and Zn can specifically stimulate the growth of the crops or can be used as useful microelements in animal feed. The higher nitrate concentrations (15-80 mg/kg dry materials) are present in the upper section of Körös-Criş Rivers, especially in the vegetation samples from agricultural and semi-natural areas (Fig. 4c). On the basis of aggregate indicators of soil pollution (Fig. 5) the largest pressure can be found in the vicinity of settlements Doboz, Misca and Avram Iancu, as well as along the road network.

4. CONCLUSION

Based on the results from the analysis performed in this paper, the relationship between the social and economical activities and contamination level of the soil and plant samples in the research area is evidenced.

The strongest correlation exists with the Pb concentration next to the main roads with traffic being the main source of pollution. However, the maximum level for Pb and other heavy metals was registered at waste landfill in Doboz settlement in Hungary. Most of the potential "contaminant areas" are located on arable lands and gardens.

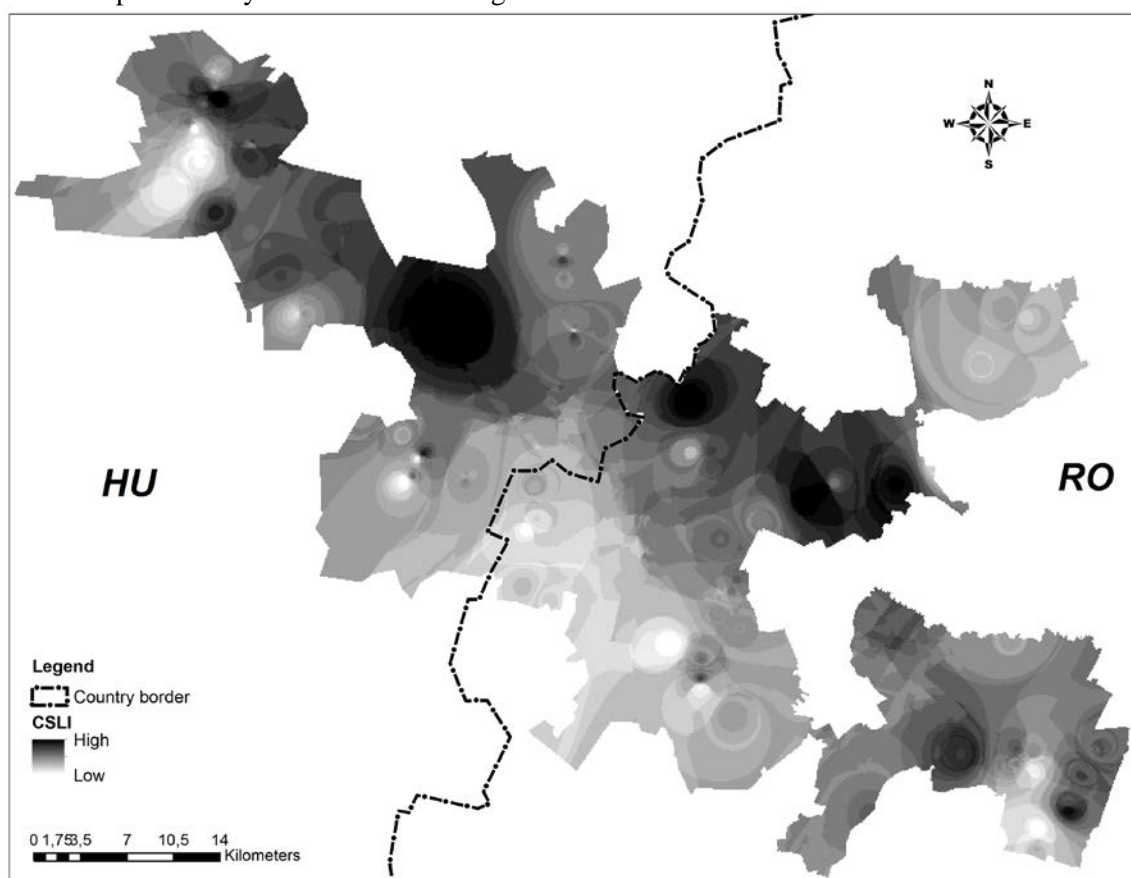


Figure 5. Cumulative soil load indicators (CSLI) of the research area

In the case of sensitive soils, the pollution intensity often exceeds the alarming values, so it is recommended to limit grazing practices along the roads. Industrial and farming activities do not cause major pollution in the environment of the research area. Heavy metals concentrations in samples taken from the vicinity of landfills are below alarming values, but are higher than normal values. In the future, it is possible that intervention measures would be needed.

The research results are a contribution to the development of local environmental programs, regional spatial development and nature conservation strategies and spatial planning, in addition to improving the quality of life in the region.

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