

ENVIRONMENTAL RISK FROM THE CONTAMINATION OF GROUNDWATER WITH TOXIC ELEMENTS IN THE SLOVAK REPUBLIC AND BRATISLAVA REGION

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Abstract: The presented research aims for identification of environmental risk resulting from the contamination of groundwater with toxic elements in Slovakia, and with special focus on the region of the capital of Slovakia, Bratislava. Analyses were carried out at the level of basic settlement units for the chemical elements arsenic, lead, mercury, and cadmium. Every investigated element in groundwater exceeds the legislatively given limit value in certain locations. The highest identified values of investigated elements are as follows: arsenic 4.9 mg/l, lead 0.27 mg/l, mercury 0.164 mg/l and cadmium 4.0 mg/l. We carried out analysis of spatial patterns of distribution of the individual elements and calculated the overall environmental risk of all elements combined expressed by an environmental risk index. Only slightly more than 1% of the area of the Slovak Republic lies within the zones of very high environmental risk index, with additional less than 2% within the zones of high environmental risk index. Although the area is relatively small, we cannot underestimate the possible risk to human health, especially when the high and very high risk zones are in populated areas.

Keywords: environmental risk, toxic element, groundwater, Slovakia

1. INTRODUCTION

State and quality of environment have always been of concern to the general public and researchers because of its impact on health and well-being of humans as well as other living organisms. WHO estimates that environmental hazards account for 25% of the global burden of disease. Bacteria in drinking water or food are considered to be biological factors, toxic chemical elements in drinking water, soil, housing, and chemicals released into the environment by the industry are considered to be chemical factors. Physical factors are usually related to the transportation system and deployment of buildings used for housing. WHO Regional Office for Europe (2013) lists the

following major environmental factors influencing population health and well-being:

- access to clean water and good sanitation services,
- poor housing conditions (such as dampness, poor indoor air quality and overcrowding),
- road safety (such as road and vehicle conditions, use of protective equipment and speed limits),
- poor air quality (such as pollution with particulate matter, gases, toxic fumes and moulds),
- work environments (including employment conditions and occupational risks),
- extreme climate conditions (such as extreme heat or cold).

Contamination of groundwater and soils with

heavy metals is one of the major concerns in environmental health (Herman & Michael 2003). Therefore, we are focusing in the presented paper on locating and assessing the areas where groundwater contains toxic elements arsenic (As), lead (Pb), mercury (Hg), and cadmium (Cd), as well as on identifying the areas where concentrations of the elements in question exceed the limit values given by the Slovak legislation for drinking water and can thus pose significant human health risk by calculating overall environmental risk from these elements.

Spatial distribution and impact of environmental factors can be studied at various scales. All in all, three levels can be distinguished: global, regional, and local (Frumkin 2010). At all levels, geographical information systems (GIS) can serve as a helpful tool to discover spatial patterns of both health and environmental data as well as their relationships.

2. THEORETICAL BACKGROUND

Groundwater represents only a small part of water sources on Earth. Due to the industrial and other mostly human activities in the last decades the amount of toxic substances in water sources has been increasing; this contributes to reduction of drinking water sources (Mukherjee et al., 2011). The US Agency for Toxic Substances and Disease Registry maintains a list of 275 water pollutants with inorganic or organic nature. According to their Priority List of Hazardous Substances, considering only inorganic chemical elements, the first four places belong to arsenic, lead, mercury, and cadmium, respectively with their sources and health effects differing.

The occurrence of arsenic in nature is related to ores containing sulphide, along with metals. The major source of arsenic in soils and natural waters is usually caused by weathering of rocks and minerals. A study from West Bengal (India) (Islam et al., 2004) reported metal-reducing bacteria might also be responsible as they play a significant role in the release of arsenic from sediments.

Lead is present in the environment naturally, but its spatial distribution is also affected by human activities; automobile fuels, for example, contain organic lead which is turned into inorganic salts during combustion. Lead occurs in nature primarily as sulphides (galena, PbS), oxides (anglesite, PbSO₄) and carbonates (cerussite, PbCO₃) (Zimdahl et al., 1973).

Mercury is present in nature as a metal in several forms. The metallic form of mercury is a silver-white, odourless liquid. Metallic mercury and inorganic mercury compounds can enter water and soils from natural deposits, volcanic activity or during waste disposal, as well as from air from manufacturing

plants, burning coal, waste and mining activities. Methylmercury may be formed in soils and water by bacteria and can be built into the tissues of fish (Agency for Toxic Substances and Disease Registry 1999).

Cadmium represents a rare but widely dispersed element which can be found naturally in the environment as cadmium ore. It is also released into environment through human activities like smelting, mining and releasing contaminated water used in various industrial processes (Agency for Toxic Substances and Disease Registry 2008).

When the water is used for consumption in households, elevated levels of arsenic, lead, mercury, and cadmium might pose health risk to the consumers. Because of the strong toxicity of these elements, groundwater contamination therewith is one of the most important environmental issues nowadays (Ahmad et al., 2015). The toxicity of these elements has been recognized for many years and has been causing health problems in several parts of the world. The health effects caused by exposure to toxic elements of arsenic, lead, mercury, and cadmium are very extensive, hence we will not summarize them. For their review we refer readers to (Järup 2003).

Increased amounts of arsenic in groundwater prevail in whole south-east and east Asia, besides Bangladesh and India (Mukherjee et al., 2011) also in Cambodia (Phan et al., 2013), China (Sun 2004), Pakistan (Bhowmik et al., 2015), and Nepal (Thakur et al. 2010), and in some parts of South America (e.g. Argentina, Mexico) (Navoni et al., 2014; Smedley 2006). Arsenic concentrations in groundwater in Europe have been reported, too. Largest areas affected by arsenic are located in countries like Hungary, Romania (Pannonian basin), Serbia, Croatia, Greece, Italy, and Spain (van Halem et al., 2009). Around half a million people use drinking water with arsenic concentrations exceeding 0.01 mg/l in Hungary; in Serbia and Croatia the numbers are approximately 600 000 and 200 000, respectively (Katsoyiannis et al., 2015).

Most of the literature on groundwater contamination is concerned with the arsenic analyses. This might be because arsenic is considered the most formidable groundwater pollutant, has strong carcinogenic effect, and is present in varied environments worldwide (Mukherjee et al., 2011).

Risk assessment represents a systematic procedure for the identification of potential risks to the environment or to the human health (DANTES 2006). Within the environmental health field, risk assessment focuses on negative health impacts resulting from exposure to hazards. The main goal of environmental health risk assessment is to provide a basis for decision

making (Frumkin 2010). For the methodological approaches and guidelines for risk assessment of the EU the following definition has been adopted: The environmental risk assessment approach “attempts to address the concern for the potential impact of individual substances on the environment by examining both exposures resulting from discharges and/or releases of chemicals and the effects of such emissions on the structure and function of the ecosystem” (Joint Research Centre 2003). One of the approaches used in these examinations is a quantitative estimation by comparing predicted environmental concentrations with a concentration below which unacceptable effects on organisms will most likely not occur (i.e. predicted no-effect concentration).

The data of environmental-geochemical mapping (predicted environmental concentrations) are appropriate for examination at regional scale. Regional scale is the simplest way to apply the limit concentrations defined in the valid environmental technical standards as predicted no-effect concentrations. In the case of groundwater in Slovakia, it is possible to use limit values set in the Regulation of the Government of the Slovak Republic No. 354/2006 Coll. as later amended that defines the requirements for water intended for human consumption and quality control of this water.

Although Slovakia does not belong to countries that are most severely affected by the negative effects of groundwater contaminated by heavy metals and toxic elements, it was reported as one of 12 European countries where groundwater contamination by heavy metals is a serious issue (EEA 1999). There are areas where limit values given by the legislation are exceeded, rendering risk assessment necessary. In this study, spatial distribution and spatial patterns of the concentrations of four toxic elements (arsenic and heavy metals lead, mercury, and cadmium) in groundwater in the Slovak Republic, with focus on the Bratislava region, are evaluated. Adverse effects might be combined and become more serious when water is contaminated by several contaminants simultaneously. Therefore, a calculation of combined environmental risk index based on the four analysed toxic elements in groundwater is carried out at the level of basic settlement units (BSU), which are the smallest statistical units in Slovakia.

3. DATA AND METHODS

3.1 Data and the Integrated Geographical Base of Environmental Health Data of the Slovak Republic

Data used in the presented paper are stored in the spatial database developed for the purposes of

the project *Comenius University in Bratislava Science Park* under the name “Integrated Geographical Base of Environmental Health Data of the Slovak Republic” (IGBEHD). IGBEHD is based on the PostgreSQL database technology (version 9.3.9) with the PostGIS spatial extension (version 2.1.5). PostgreSQL is one of the most reliable open source RDBMS systems and together with the PostGIS spatial extension, this system enables building of a robust spatial database capable of storing and analysing geographical objects. A geographical object represents a special type of data containing a geographical position defined by coordinates of a selected coordinate reference system. Geographical data can be analysed using spatial functions of PostGIS. This system enables us to manipulate complex geographical and environmental datasets as at the level of GIS. However, PostgreSQL system does not feature a suitable graphical environment for displaying spatial data, making external graphical tools such as QGIS necessary (Marquez 2015).

In our analyses, we used spatial data representing distribution of the toxic chemical elements arsenic, lead, mercury, and cadmium in groundwater. The original data coming from environmental-geochemical mapping for the purposes of the Geochemical Atlas of Slovakia (Ministry of Environment of the Slovak Republic 1996) form an irregular vector point network covering the whole area of the Slovak Republic. During the mapping between 1991 and 1994, 16 359 groundwater samples were collected (approximately one sample per 3 square kilometres). The concentrations of 32 chemical elements and compounds were determined by a chemical analysis (Ministry of Environment of the Slovak Republic 1996). The data might seem quite old; however, this is the only existing consistent source of information of this kind for groundwater and for the area of whole Slovakia.

The point data were subsequently processed by the interpolation function *Regularized spline with tension* in the GRASS GIS software into the form of raster coverages with spatial resolution of 100 meters, reclassified, and converted to vector polygons. In data reclassification, the limit values given by the *Regulation of the Government of the Slovak Republic No. 354/2006 Coll. as later amended* were considered where available. The resulting data were imported in this form to the spatial database IGBEHD. For the four elements analysed (arsenic, lead, mercury, and cadmium) the following limit values were applied: arsenic – 0.01mg/l, lead – 0.01mg/l, mercury – 0.001mg/l, and cadmium – 0.005mg/l.

3.2 Descriptive Statistics of Concentrations of Toxic Elements

To provide a summary of the average concentrations of the selected toxic elements in groundwater in BSUs, basic descriptive statistics were calculated – mean and percentile values (25th, 50th, 75th). To describe spatial patterns of the concentrations, local Moran's I (Anselin 1995) was calculated using the GeoDa 1.6.6 software (GeoDa Center for Geospatial Analysis and Computation 2014) and tested using 9999 permutations. Spatial patterns (clusters and outliers) were considered statistically significant at the level of $p < 0.05$. The weight matrix for the index calculation was determined by a distance band of 10 000 m. Since there is no rule to determine the optimal distance band size (Zhang et al., 2008), the value was chosen as the desired size of smallest identified clusters.

3.3 Calculation of Environmental Risk Index

According to Rapant et al., (2004) environmental risk can be expressed numerically by the environmental risk quotient (QER) (1). QER represents the ratio between real (measured) and limit (risk) concentration values. Environmental risk index (IER) can be used to express the overall effect of several elements (in our case QER of arsenic, lead, mercury, and cadmium in groundwater) if at least one concentration exceeds the respective limit value. It is a sum of risks posed by the individual elements (2).

$$Q_{ERi} = \frac{AC_i}{RC_i} - 1 \quad (1)$$

$$I_{ER} = \sum_{i=1}^n Q_{ERi} \quad (2)$$

where:

Q_{ERi} – environmental risk quotient of the i^{th} element with concentration exceeding the limit

AC_i – i^{th} element's measured concentration

RC_i – i^{th} element's limit concentration

I_{ER} – environmental risk index of the tested sample

Contaminants with concentrations not exceeding limit values cause no environmental risk, thus their $QER = 0$ and they are omitted from the calculation. By subtracting the number one in the QER formula only the concentrations that exceed the limit values should be considered. Subsequently, IER will provide us with an expression of the potential cumulative risk from all considered contaminants that exceed the limit content in groundwater.

The spatial distribution of QER is expressed by raster coverage maps, computed from coverage maps of distribution of the toxic chemical elements of arsenic, lead, mercury, and cadmium in groundwater. IER is expressed by a raster coverage map; this was

generated as the sum of four QER coverage raster maps by simple map algebra. Subsequently, the average value of IER for each BSU was calculated. To assess the risk range, five-level scale based on the nationwide results for individual geological compartments of the environment, as adapted by Rapant et al., (2004), is used:

- $I_{ER} \leq 0.5$ very low risk
- $0.5 < I_{ER} \leq 1$ low risk
- $1 < I_{ER} \leq 3$ medium risk
- $3 < I_{ER} \leq 5$ high risk
- $I_{ER} > 5$ very high risk

Thanks to the database structural query language (SQL) it is possible to query data in a way enabling us to identify all BSUs where environmental risk is high and/or very high.

4. RESULTS

4.1 Descriptive Statistics of Concentrations of Toxic Elements

Basic descriptive statistics of concentrations of the analysed toxic elements (arsenic, lead, mercury, and cadmium) in groundwater in the Slovak BSUs are presented in table 1. Apart from mean and selected percentiles, limit values and numbers of BSUs exceeding the limit values are stated. In all cases, the mean value is slightly higher than the median value, suggesting positive skewness of the concentration distributions.

As shown in table 1, every investigated element in groundwater exceeds the limit value in certain locations. Limit values for lead and mercury are exceeded only in several BSUs, whereas arsenic and especially cadmium concentrations are above the legislatively given limits in a considerable number of BSUs (81 and 541, respectively). In the Bratislava region, the limit values are not met for arsenic and cadmium in 6 and 27 BSUs, respectively.

The results of spatial pattern analysis (Fig. 1) suggest strong clustering behaviour of the concentrations of the four toxic elements in groundwater, especially of low values (coldspots), but more importantly of high values (hotspots). These two spatial patterns of the analysed elements dominate within the study area. The clearest spatial pattern structure can be identified for cadmium. On the other hand, spatial distribution of lead has the most complicated spatial structure, with numerous high-low as well as low-high spatial outliers. In most cases the locations of hotspots differ for the elements. In the Bratislava region, hotspots were identified for arsenic and cadmium. For lead and mercury, with regard to high values, only several high-low outliers were found.

Table 1. Summary statistics of the average concentrations of toxic elements in groundwater (mg/l) in basic settlement units (BSUs). SK stands for the territory of Slovakia; BA stands for the Bratislava region.

Statistic	Area	Arsenic	Lead	Mercury	Cadmium
Limit value		0.01	0.01	0.001	0.005
Min	SK	0.0005	0.0005	0.0001	0.00025
	BA	0.000597	0.000558	0.0001	0.00025
25 th	SK	0.000584	0.000718	0.0001	0.000254
	BA	0.000931	0.000723	0.000101	0.000276
50 th	SK	0.000828	0.000917	0.000103	0.000271
	BA	0.001156	0.000853	0.000104	0.000346
75 th	SK	0.001384	0.001204	0.000119	0.000339
	BA	0.001887	0.001043	0.000112	0.001161
Max	SK	4.9	0.27	0.164	4.0
	BA	0.025588	0.004026	0.00037	0.241584
Mean \pm SD	SK	0.001742 \pm 0.013934	0.001145 \pm 0.001249	0.000145 \pm 0.000669	0.001181 \pm 0.010615
	BA	0.001819 \pm 0.002258	0.000942 \pm 0.000354	0.000112 \pm 0.000024	0.002661 \pm 0.014392
Number of BSUs above limit concentration	SK	81	13	19	541
	BA	6	0	0	27

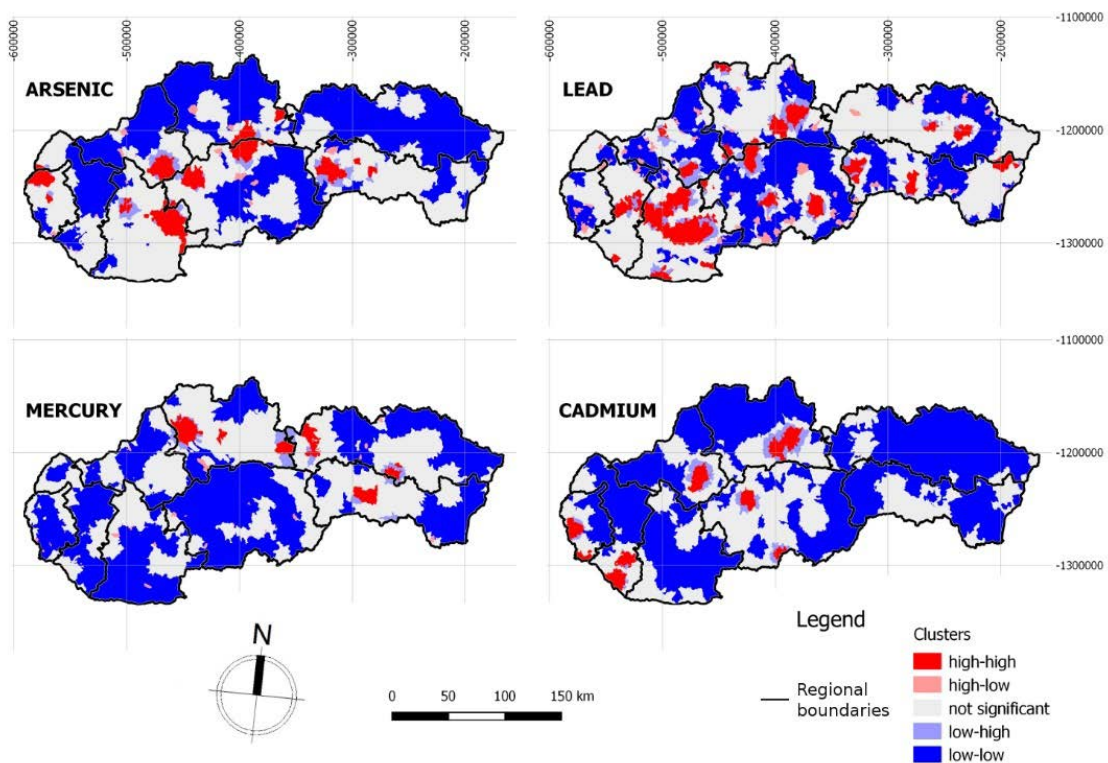


Figure 1. Spatial patterns of arsenic, lead, mercury, and cadmium concentrations in groundwater at the level of basic settlement units.

4.2 Environmental Risk

To get an image of the overall groundwater contamination from the four investigated elements, an environmental risk index was calculated at the BSU level (Fig. 2). The largest areas with high and

very high IER are located in the western part of the Prievidza district (area between the towns of Partizánske and Prievidza), border area located south of Dunajská Streda, eastern part of the Ružomberok district, north-western part of the Liptovský Mikuláš district, western part of the Žilina district (south of

the town of Žilina) and southern part of the Bratislava district. Smaller zones of high and very high environmental risk are located in the north-western part of the Zvolen district, in the north-western part of the Bratislava district (area between the Bratislava IV district and town of Stupava), and in the eastern part of the Veľký Krtíš district. In total, 1.80% (885 km²) of the area of the Slovak Republic lies within high environmental risk zone and 1.06% (520 km²) in the very high environmental risk zone (see table 2 for the values of each of the four elements). The high-risk areas are mostly located in the northern part of central Slovakia and in the south-east. On the other hand, the eastern part of Slovakia barely has any BSUs with high or very high environmental risk from the groundwater contamination with arsenic, lead, mercury and/or cadmium.

Table 2. Area taken by risk zones of the selected toxic elements from the overall area of the Slovak Republic (49 035 km²).

Indicator	Area
Arsenic risk zones	1.42 % / 694 km ²
Cadmium risk zones	2.15 % / 1055 km ²
Mercury risk zones	0.26 % / 126 km ²
Lead risk zones	0.23 % / 113 km ²

A very high environmental risk is present in the Liptovské Behárovce BSU (and a municipality at the same time), located three kilometres north of the

Liptovská Mara water dam. This village includes permanent settlements and the proportion of houses not connected to public water supply is 11.9%. Toxic elements that exceed the maximum allowed values in groundwater are cadmium and lead.

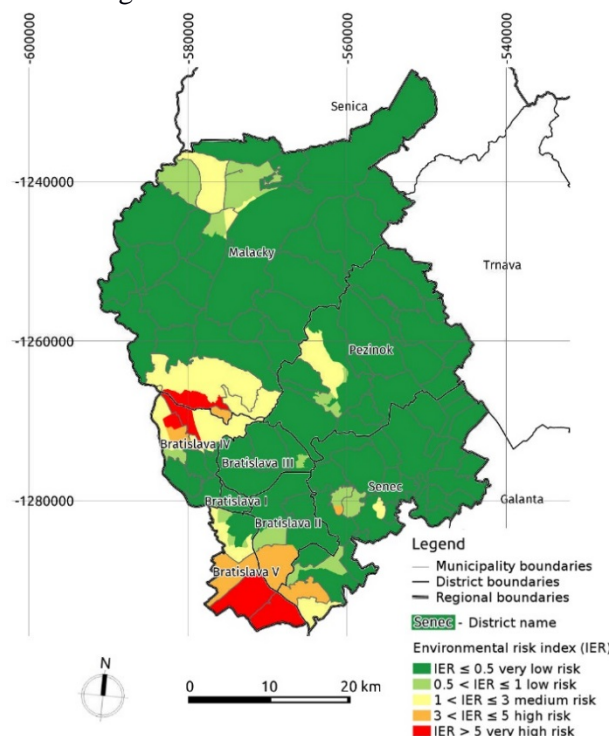


Figure 3. Environmental risk index (IER) from the contamination of groundwater with arsenic, cadmium, mercury, and lead at the level of basic settlement units in the Bratislava region.

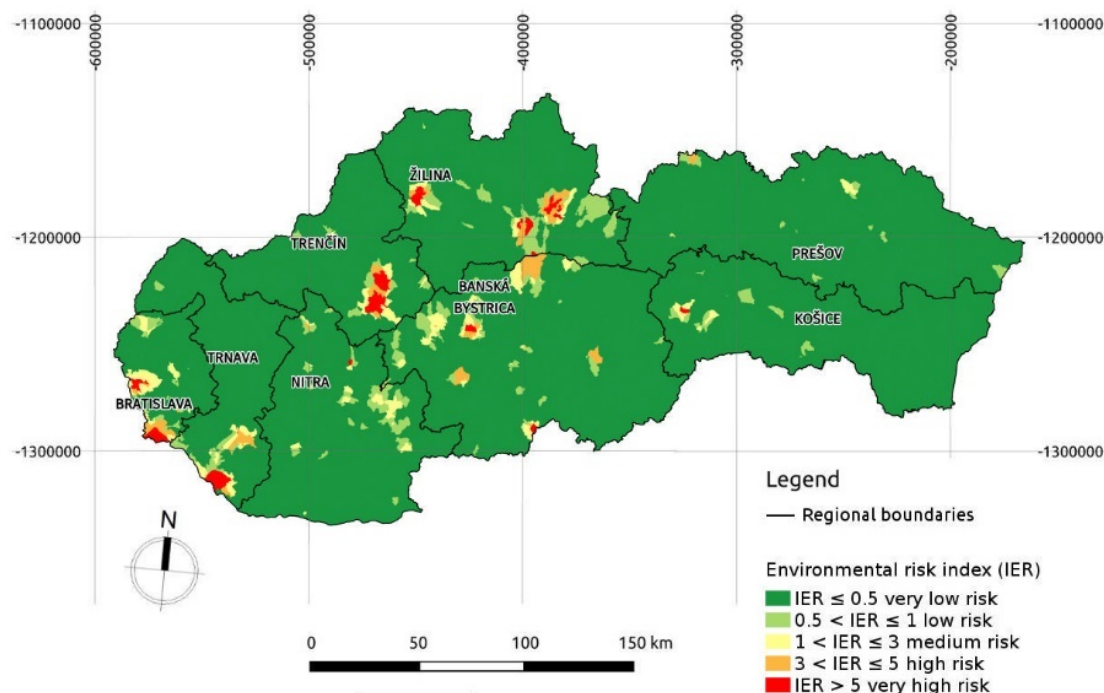


Figure 2. Environmental risk index (IER) from the contamination of groundwater with arsenic, cadmium, mercury, and lead at the level of basic settlement units.

High environmental risk applies also to the Tehelne BSU, a part of the Drienovská Nová Ves municipality. The proportion of houses not connected to public water supply here is 10.53%. Groundwater is contaminated with above-limit values of mercury, most likely from the brickfield located in this BSU.

By comparing the results obtained for the whole Slovak Republic to those for the Bratislava region, it can be concluded that there are two main zones with high or very high environmental risk (Fig. 3), both crossing the area of the capital. The environmental risk index reaches values above 3 (high and very high risk) in the southern and central western part mostly because of the presence of cadmium, which exceeds the maximum allowed values in groundwater.

5. DISCUSSION

The results suggest that the highest environmental risk is on the periphery of several large industrial towns. For example, in the Bratislava region, heavy industry is located in these areas, specifically the Volkswagen (German car manufacturer) plant in the central western part and the Slovnaft oil refining company in the south; this could explain why cadmium is dispersed in the surrounding environment. Although cadmium is usually dispersed into environment through air by its smelting and mining or burning fossil fuels, it can also enter the ground and its compounds move easily through soil layers and can reach groundwater. Groundwater could also be contaminated by industrial wastewater or seepage from hazardous waste sites (Agency for Toxic Substances and Disease Registry 2008). On the other hand, large industrial areas exist where no significant environmental risk was identified, *e. g.* U.S. Steel Košice. Unfortunately, our data alone do not make it possible to differentiate between toxic elements originating from human activities and those of natural origin, hence we only can speculate about it according to the local geological and industrial situation.

The environmental risk from groundwater contamination represents a health risk mainly for people who directly consume the contaminated water. Presumably, these are usually those who reside in houses without connection to public water supply. If people residing in such zones use their own wells as a drinking water source in a long term with no water treatment, health problems may occur. Another factor that ought to be considered in the interpretation of the results is that people living in houses without connection to public water supply and with their own drinking water sources might be using special filters for cleaning water or buying drinking water from grocery stores.

There are several limitations to this research. The first of them is related to data quality; it is important to mention that the data coming from the environmental-geochemical mapping were collected in the time period of 1991 – 1994, rendering it more than 21 years old at the moment. Thus, they might be already out of date in some locations; mostly due to the major economic and industrial changes in the 1990's in the Central-Eastern European region some of the pollution sources might have been eliminated and new ones might have appeared. On the other hand, toxic elements in groundwater might also occur naturally, and in such case the age of the data is not expected to influence data quality significantly. Nevertheless, this is the only consistent data source of the content of toxic elements in groundwater for the area of the whole Slovak Republic.

Another factor that may influence the quality and reliability of results of the applied methodology is the interpolation method used to compute raster surfaces. Irrespective of the interpolation method, the final raster surface will always represent approximated values only, rather than the real values of the observed phenomenon. Talking about raster surfaces we must also consider the spatial resolution settings, usually set according to the density of the input vector data. In our case, the spatial resolution of 100 meters was set given the relatively dense irregular vector point network (one sample per 3 square kilometres).

6. CONCLUSIONS

The presented research dedicated to the identification and description of environmental risk of toxic elements arsenic, lead, mercury, and cadmium in groundwater has brought an insight into the spatial distribution of environmental risk in the Slovak Republic and the Bratislava region at the level of the smallest statistical units in Slovakia (*i.e.* BSUs). Several locations in the Slovak Republic where these toxic elements exceed the maximum allowed values in groundwater given by the *Regulation of the Government of the Slovak Republic No. 354/2006 Coll. as later amended* were identified. The affected area is quite small (it is largest for cadmium, 2.15% of the overall area of the Slovak Republic, and smallest for lead, 0.23%). To assess the overall effects of the four analysed elements together, environmental risk index was calculated. Only slightly more than 1% of the area of the Slovak Republic lies within the zones of very high environmental risk index, with additional less than 2% within the zones of high environmental risk index. Although the area is relatively small, we cannot underestimate the possible risk to human health, especially when the high and very high risk zones are

in populated areas. The results obtained in our experiment can help not only professionals but also the general public, *e.g.* when planning to build a drinking water well.

Acknowledgment

This publication is the result of the project implementation: Comenius University Science Park - 2. phase ITMS 2014+ 313021D075 supported by the Research & Innovation Operational Programme funded by the European Regional Development Fund.

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Received at: 14. 10. 2016
 Revised at: 25. 02. 2017
 Accepted for publication at: 06. 03. 2017
 Published online at: 28. 03. 2017