Approach to the assessment of transport risk of inorganic pollutants based on the immobilisation capability of soil

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ABSTRACT

The objective of this paper is to elaborate a system of evaluation of potential risks of transport of inorganic elements, and to create a map of soil potential to immobilisation/transportation of potential risk elements. The categorization of these risks was realized in two layers, contamination and potential soil sorption. The level of contamination was evaluated according to the Slovak Soil Law. Potential sorption of soil (PSS) is formed by qualitative (soil reaction, pH value, optical value of soil humus), and quantitative factors (C_{org}, H-depth of humus horizon); it was evaluated according to the equation: $[PSS] = F(pH) + F(Q^4_6) + F(C_{org}) \times F(H)$. The map of soil immobilisation potential was created by fusion of contamination layer and layer of potential sorption of soil, and shows the distribution of five categories of risk elements immobilisation potential. Soils with very low immobilisation potential are reported predominantly in Košice and Banská Bystrica regions, where soils are contaminated by geochemical anomalies and anthropogenic sources. Transport is a reversible process to immobilisation; thus, the highest level of immobilisation potential is identical with the lowest transport category.

Keywords: inorganic contaminants; contamination; immobilisation; transport; soil properties

At present, a comprehensive EU strategy for soil protection is needed. This strategy should take into account all the different functions that soil can perform, their variability and complexity (Commission of the European Communities 2006). The EU Directive establishes a framework for the protection of soil and the preservation of capacity of soil to perform any of the following environmental, economic, social and cultural functions: biomass production including agriculture and forestry; storing, filtering and transforming nutrient, substances and water; biodiversity pool; physical and cultural environment for humans; source of raw material; acting as carbon pool and archive of geological and archeological heritage.

One of the environmental functions established by the EU Directive is filtering of substances. In our institute we try to evaluate this function as one of the main soil processes (filtering or immobilisation of water, nutrients and contaminants). Because of very different physical-chemical properties of contaminants, we divided the group of substances into organic and inorganic pollutants. Filtration/immobilisation of organic pollutants was already evaluated (Barančíková and Madaras 2003). At present, we attempt to assess the immobilisation of inorganic contaminants. Immobilisation of soil contaminants and its reverse process, i.e. their transport, play a very important role in the evaluation of the environmental function of soil. Immobilisation of soil contaminants means an ability of soil to keep substances and prevent their leaching and contamination of groundwater, or their entering into the food chain; transport allows leaching [a chemical may be transported through the soil by solvents (water) or with soil movement] and runoff (a movement to contaminate air, water, soil, plants and animals, chemical moving across a surface with a solvent or with the soil).

Inorganic contaminants, mainly potential risk elements, present a serious problem in all environmental issues. Potential risk elements include

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essential important elements (in optimal concentration range): Cu, Fe, Mn, Zn, Co, Se, and some non-essential elements – potentially toxic elements: Hg, Pb and Cd (Hansen et al. 2001). Toxicity of potential risk elements is different and declines in order: Hg > Cd > Ni > Pb > Cr (Yong et al. 1992).

To understand the behaviour of potential risk elements in the soil system is one of the most important tasks in the evaluation of their immobilisation and transport. An assessment of their relative binding power in soil was used as one criterion for Filter and Buffer function in Berlin Digital Environmental Atlas (2006). Blume and Brummer (1991) found a concept to judge the sensitivity against metal load. The main idea of this prognosis is that relative binding power of some metal depends on pH value of the soluble soil solution, based on the condition of sand soil with weak binding power and poor humus. Higher content of humus, clay and ironhydroxide enhance binding power and vice versa. This calculation of relative binding power of potential risk elements is valid till 1 m depth (Berlin Digital Environmental Atlas 2006). A similar method was used in our approach to estimate risks of soil inorganic pollutants transport.

Determination of potential risk elements sorption by the soil constituents is mostly accomplished through the selective sequential extraction of the potential risk elements from the soil samples (Hansen et al. 2001, Makovníková 2001, Barančíková and Makovníková 2003, Finžgar et al. 2007). It is well known that the variation in soil properties such as pH, organic matter content and quality, texture, or quantity and quality of adsorbing sites, can significantly influence the distribution of potential risk elements as well as their availability to plants and water. Soil reaction, content and quality of soil organic matter, content and quality of clay fraction, iron and manganese oxides belong among the main factors influencing mobility of the potential risk elements in soil (Hansen et al. 2001, Barančíková and Makovníková 2003, Borůvka and Drábek 2004).

Slovak Soil Law classifies total amounts of particular group of inorganic contaminants: Hg, Cd, Pb, Ni, Cr, As, Cu, Zn, Co. The subject of our interest was the group of risk contaminants in total. In this paper we try to work up a system of evaluation of potential transport risks of these group of inorganic contaminants based on the capability of soil compartment to immobilisation of inorganic contaminants and to create a map of soil potential to immobilisation/transportation of risk elements. The main aim of our work was to estimate immobilisation of risk elements as a whole. We realize that to fulfil our objectives, i.e. to present an estimation of filtering function with respect to inorganic contaminants, a certain simplification while solving such a complex problem is necessary.

MATERIAL AND METHODS

The categorization of potential transport risk of risk elements was realized only for agricultural soil in two layers, a contamination layer and a layer of potential soil sorption.

Soil potential to immobilisation of risk elements

↗ ↖

Contamination layer ⟷ **Layer of potential soil sorption**

Source of data. Available data sources for the categorization and mapping consist of primary (spatial information on the soil bodies) and secondary (data of relevant soil properties) geo-referenced data.

Contamination layer. Primary geo-referenced data belong into the Digital Database of Soil Profiles of Geochemical Atlas of Slovakia (GchA – 2 965 localities of agricultural soil) and the Digital Soil Map of Slovakia in the scale of 1:400 000 (PM 400). The Geochemical Atlas Database contains the data of risk elements concentration of the samples collected during the national project "Geochemical Atlas of Soils of Slovakia" (Čurlík and Šefčík 1999). Data from the Geochemical Atlas of Soils of Slovakia and data from the Digital Soil Map of Slovakia were used to generate the layer of contamination.

Layer of potential soil sorption. Secondary georeferenced data are used as a source of additional information (data of relevant soil properties) to the primary geo-referenced data. As secondary georeferenced data we used the Digital Database of Soil Monitoring of Slovakia (CMSP) in the Digital Data archive of the "Partial Monitoring System-Soil", as a part of the Complex Environmental Monitoring of Slovakia. A set of 312 representative monitoring sites was localized in agricultural land and sampled in a 5-year frequency, 21 key localities in a 1-year frequency. Data on relevant soil properties were used from the last sampling (2002). Data from CMSP were utilized to generate the layer of potential soil sorption. The territorial units are soil types/subtypes in the PM 400 Database Slovakia. ArcGIS® was applied for work with input geo-referenced digital data and for implementation of final digital data layer. This layer represents the categorization of potential transport risks of potential risk elements based on the capability of soil compartment to immobilise inorganic contaminants.

RESULTS AND DISCUSSION

Potential of soil to immobilisation and transport of risk elements depends on the total amount of potential risk elements in soil and the potential of soil sorbents, which are sensitive to the risk elements sorption. A higher amount of potential risk elements in soil replace potential sorbents and consequently decrease overall potential of soil to sorb risk elements. It is well known that the total content of metals in soil may not provide the best determination of its biovailability, it is better to consider the mobile content of risk elements. However, available soil databases do not incorporate data of mobile fraction of risk elements; for that reason we can use only the data of total content of risk elements.

Contamination layer

Total content of potential risk elements is one of the main parameters (because the information about mobile fraction for the Slovak region is not available) of soil sensitivity to potential risk elements mobility. Soil contamination was estimated as a surface contamination. Data of contamination were obtained from relational soil profile database of GchA (total content of 9 high risk elements). The special description of individual elements in Slovak soils is given by Čurlík and Šefčík (1999). The level of contamination has been evaluated according to the Slovak Soil Law. Background levels of elements in soils depend both on the nature of parent materials and on the soil texture, since many elements are associated with the fine particles that have the ability to adsorb or form solid solutions with the elements of interest (Naidu et al. 1996). Hence, the limit values of potential risk elements in soil according to the Slovak Soil Law depend on clay fraction (content of size fraction < 0.01 mm) (Table 1).

To determine the appropriate categories of contamination for agricultural soil, the content of potential risk elements was transformed into a point rating. The potential risk elements were divided into two groups according to their toxicity (Yong et al. 1992).

Group 1: Hg, Cd, Pb, Ni, Cr

Group 2: As, Cu, Zn, Co

Group 1 represents the potential risk elements of higher toxicity and group 2 of lower toxicity. Therefore, score values (for each individual element) of group 1 are considerable higher as compared to group 2 (Table 2).

The score of the overall contamination risk of particular locality was estimated as a sum of evaluation score function of all 9 elements of each sample (Table 3).

The last step in the construction of contamination layer is the calculation of contamination risks for soil types polygons (separate for arable soils and pasture) presented on the map PM 400. It is estimated as arithmetic average of score contamination of separate samples (Table 3) presented in the particular polygon. Point ranking of categories is shown in Table 4.

A high level of contamination can be found in the mountain soils on grassland (Podzol, Dystric Cambisols localized south-west and east of Banská Bystrica, east of Spišská Nová Ves, and in the mountain regions of Western Carpathians), where the soil is affected predominantly by parent material (geochemical anomalies) (Čurlík and Šefčík 1999). Some recent works reported that the distribution of heavy metals is heterogeneous on the scale of aggregates. It was shown that aggregate surfaces

Table 1. Limit values of total risk elements in agricultural soils in mg/kg according to the Slovak Soil Law

Content of soil particle < 0.01 mm	As	Cd	Co	Cr	Cu	Hg	Ni	Pb	Se	Zn
Lower than 20%	10	0.4	15	50	30	0.50	40	25	0.25	100
20–45%	25	0.7	15	70	60	0.50	50	70	0.40	150
Higher than 45%	30	1.0	20	90	70	0.75	60	115	0.60	200

Table 2. Score for evaluation of individual elements

Group	Criteria	Category	Evaluation
Group 1	content of element lower than 80% of limit	A	0
	content of element equal to 80%, lower than 120%	B	1
	content of element equal to 120%, lower than 200%	C	2
	content of element equal to or higher than 200%	D	3
Group 2	content of element lower than 80% of limit	A	0
	content of element equal to 80%, lower than 120%	B	0.5
	content of element equal 120%, lower than 200%	C	1
	content of element equal to or higher than 200%	D	1.5

show higher contents of heavy metals than inner parts of aggregates (Wilcke and Kaupenjohann 1994). Thus the fractionation of aggregates and the selective analyses of their outer and inner parts enable to trace anthropogenic contamination of heavy metals. The selective analysis of aggregates is available only for a small part of Slovakia (Makovníková 2002). Measurements of magnetic susceptibility of topsoils were successfully applied for mapping of degree of anthropogenic pollution near power plants, cement and metallurgy industries only for small region of Slovakia (Ďurža 2003). Geochemical anomalies are defined on the basis of profile distribution of risk elements (Čurlík and Šefčík 1999). In some localities of arable soils (Eutric Fluvisols and Fluvi-eutric Gleyosols along the rivers Váh, Hron, Bodrog) a high contamination is connected with a higher amount of potential risk elements in sediment deposited on the flood plains as well as with locally anthropogenic sources around factories. The lowest amount of potential risk elements is characteristic mainly for arable soils (Chernozems) in the Danube lowland.

Layer of potential sorption of soil

Sorption processes play an important role in soil fixation of potential risk elements. Many authors (Hansen et al. 2001, Makovníková 2001, Finžgar et al. 2007) estimated the influence of selected soil parameters to mobile and potentially available risk elements content. It was found that the variation in soil properties such as pH, organic matter content and quality, texture and type of clay minerals, iron and manganese oxides influence quantity and quality of adsorbing sites and thus significantly influence the immobilisation of the potential risk elements. The factor analysis data of CMSP showed the main indicators (Makovníková 2004a), which potentially influence soil potential to immobilisation of risk elements. There are mobile content, total content of the potential risk elements, pH value, content of organic matter and quality of organic matter represents by optical quotient, depth of humus horizon and soil texture. However, Slovak soil limits of the potential risk elements depend on soil texture (content of clay fraction, Table 1) and thus the map of immobilisation is modelled as the sum of the soil contamination and soil potential sorption, this soil parameter is not reflected in potential soil sorption. Soil texture belongs among the soil parameters that influence soil sorption potential. However in repeatedly inclusion of this parameter in layer of potential sorption of soil, this parameter would by double evaluate.

Soil reaction

Soil pH is the most important soil parameter, influencing most of elements in soil. This phenomenon is related to soil sorbents charges depending on the value of soil reaction. At low pH, the sorption of potential risk elements is also relatively low with respect to competition of adsorbing sites by proton (H^+). This mechanism can be active in various pH regions in a different way, i.e. proton competition is the most significant at low pH values, however, changes in charges of iron and manganese oxides are the most important in the weakly acidic to neutral pH region. It is evident, that the sorption of cations with higher tendency to hydrolyse is frequent in low pH environment, whereas cations with lower tendency to hydrolyse can be found in the area with higher value of soil reaction.

Table 3. Score values for locality

Criteria	Score		Designation
Sum of scores < 0.45	0	N	no contamination
Sum of scores 0.45–7 or content of all 9 elements < category C, D	1	L	low contamination
Sum of scores 7.5–14 or content of all 9 elements < category D	3	M	medium contamination
Sum of scores > 14 or content of one or more elements > category D	5	H	high contamination

The analysis data of CMSP (Makovníková 2001, Makovníková 2002, Makovníková 2004a, b) showed a significant influence of pH in the case of Cd, Pb, Cu, Zn, Ni, which is higher than the influence of all other used parameters (content and quality of organic matter, depth of humus horizon). pH value belongs among the dynamic soil parameters, i.e. small changes of this parameter influence the risk elements mobility. The dependence of pH is not identical for all risk elements; however, this generalisation was used for all elements of high toxicity (Cd, Pb, Cr, Ni) because this work aims at estimate of the immobilisation of risk elements as a whole. The highest average pH values in CMSP were found in Chernozems (arable land) whereas the lowest average pH values were detected in Cambisols (permanent grassland). Between arable land and grassland of the same soil type and subtype differences of average pH values were observed.

Content and quality of soil organic matter

Soil organic matter (SOM) plays an important role in the formation of complexes, but also in the retention of risk elements in an exchangeable form. Organic carbon content (C_{org}) is an important parameter used to quantify the allowable content of potential risk elements in soil (Hansen et al. 2001) and was introduced as a quantitative parameter of SOM in our categorization. The highest content of C_{org} can be found in mountain soils (Podzol, Ranker, Litozem), and Andosols and Cambisols on grassland. The lowest amount of soil humus is characteristic mainly for arable land on Gleyosols and Regosols. The immobilisation of risk elements in soil compartment, mainly in agriculture arable soils (low content of C_{org}), depends also on the quality of soil organic matter (Donisa et al. 2003, Borůvka and Drábek 2004). Humic substances with aromatic structures and high amount of carboxylic groups are dominating sorbing fractions of organic matter (Preston 1996, Barančíková and Makovníková 2003). In our previous papers (Makovníková 2001, Barančíková and Makovníková 2003) significant correlations between mobile/potentially available risk elements fractions and coarse qualitative parameter reflecting the SOM structure – Q^4_6 were found. The lower value of optical parameter Q^4_6 illustrates a higher stability and humification of the soil humus. On the basis of these findings, we decided to introduce qualitative measure of SOM – spectral parameter Q^4_6 into our categorization. According to Q^4_6 values, Phaeozem, Chernozem and Luvisol represent more stable and mature soil humus contrary to Gleyosols and Cambisols (Makovníková and Barančíková 2003).

Depth of the humus horizon

Depth of the humus horizon belongs among quantitative parameters in our categorization. A higher depth of the humus horizon positively influences total amount of potential sorbents in soil. The highest depth of humus horizon is typical

Table 4. Score values for polygons

Criteria	Score		Designation
Arithmetic average of score < 0.45	0	I	no contamination
Arithmetic average of score 0.46–1, localities number of M or H < 20%	1	J	low contamination
Arithmetic average of score 1–2, localities number of H < 20%	3	K	medium contamination
Arithmetic average of score > 2, localities number H > 21%	5	L	high contamination

for Chernozems and Mollic Fluvisols, on the other hand Podzols and some subtypes of Cambisols have the humus horizon of a small depth.

Based on these findings we concluded that the **Potential Sorption of Soil** (PSS) is formed by qualitative (pH, Q^4_6) and quantitative factors (C_{org}, H-depth of humus horizon).

The importance of the parameters determining potential sorption is different, and hence, the parameters are assessed according to the results of the factor analysis (Makovníková 2004a). A higher value of soil reaction, C_{org}, depth of humus horizon and a lower value of Q^4_6 increase the immobilisation of potential risk elements in soil. Thus, the score values of soil properties influencing the soil potential sorption are opposite to the contamination score values, which means that the highest score values of soil parameters express the lowest influence to soil potential sorption.

Because of considerable differences of soil reaction and organic carbon content values in arable soils and pasture, score evaluation was determined separately for different cultivation (Table 5).

Point evaluation of the potential soil sorption (PSS) for soil polygons was calculated according to the following score function:

$$(PSS) = F(pH) + F(Q^4_6) + F(C_{ox}) \times F(H)$$

This empirical relationship was tested on 20 key localities of CMSP. Correlations between soil sorption ECEC (effective CEC) (r = –0.55) and saturation degree (ECEC × 100/CEC) (r = –0.64) and potential soil sorption (PSS) calculated according to the proposed empirical relationship were statistically significant.

According to the evaluated parameters and calculation of PSS among arable land as well as grassland,

Table 5. Score values of potential soil sorption influenced by soil properties

	Arable land			Grassland		
	category	pH/$CaCl_2$	score	category	pH/$CaCl_2$	score
pH/$CaCl_2$	1	< 5.5	4	1	< 4.50	4
	2	5.51–6.00	3.2	2	4.51–5.00	3.2
	3	6.01–6.50	2.4	3	5.01–5.50	2.4
	4	6.51–7.00	1.6	4	5.51–6.00	1.6
	5	> 7.0	0.4	5	> 6.00	0.4
	category	C_{ox}	score	category	C_{ox}	score
C_{org} content (%)	1	< 1.0	1.00	1	< 2	0.00
	2	1.0–1.5	0.80	2	2–3	0.80
	3	1.5–2.0	0.60	3	3–4	0.00
	4	2.0–2.5	0.40	4	4–5	0.40
	5	> 2.5	0.10	5	> 5	0.10
	arable land/grassland					
	category	Q^4_6	score			
Quality of organic matter – Q^4_6 (joint categories for arable soils and pasture)	1	> 6.01	1.00			
	2	5.51–6.00	0.80			
	3	5.01–5.50	0.60			
	4	4.51–5.00	0.40			
	5	< 4.5	0.10			
Depth of humus horizon (cm) (joint categories for arable soils and pasture)	category	depth	score			
	1	> 20 cm	2			
	2	21–30 cm	1			

Table 6. Criteria for categorizing

Score values for polygons	Soil categories of potential immobilisation	Soil categories of potential transport
< 2.5	very high	very low
2.6–4.5	high	low
4.6–6.5	medium	medium
6.6–8.5	low	high
> 8.5	very low	very high

Rendzic Leptosols and Calcaric Mollic Fluvisols exhibit a high sorption potential. Eutric Regosols on arable land and Podzols on grassland belong to soils with a very low sorption potential.

Soil categories of potential transport risk of the risk elements based on the capability of soil compartment to immobilise the inorganic contaminants

The overall rating is determined as a sum of soil contamination (contamination score values for soil polygon) and soil potential sorption (PSS value for soil polygon). A high soil contamination was evaluated by the high point value and presents a high risk. On the other hand, a high soil sorption potential results in a low point value and decreases potential transport risks of risk elements in soil. The overall rating is used as a criterion for categorizing the potential of immobilisation of risk elements in agriculture soils (Table 6). Transport is a reverse process to immobilisation. Consequently, the highest category of potential to immobilisation of risk elements corresponds to the lowest category of transport.

Map of soil immobilisation potential

A map of the soil potential to immobilisation of risk elements was created by fusion of two layers, the layer of contamination and the layer of potential sorption of soil. Figure 1 shows the distribution of five categories of potential immobilisation of risk elements in agricultural land of Slovakia. Very high potential to immobilisation of risk elements was recorded in 19.74% of Slovak agricultural soils, high potential in 26.06%, medium in 27.38%, low in 21.64% and very low potential to immobilisation of risk elements only in 5.18%. Categories with very high and high immobilisation potential, and thus with low risk of transport, comprise 45.80% of all agricultural soils of Slovakia.

Most of the arable soils with the high production potential belong to the categories with very high or high potential to immobilisation, i.e. with low risk of heavy metal transport. These are mainly calcare-

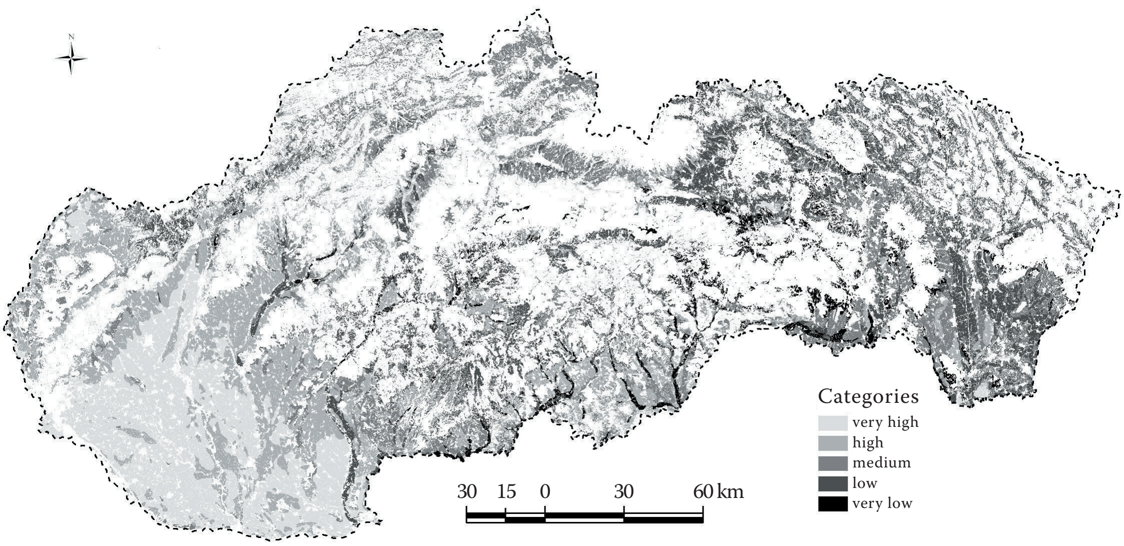

Figure 1. Soil immobilisation potential of inorganic contaminants

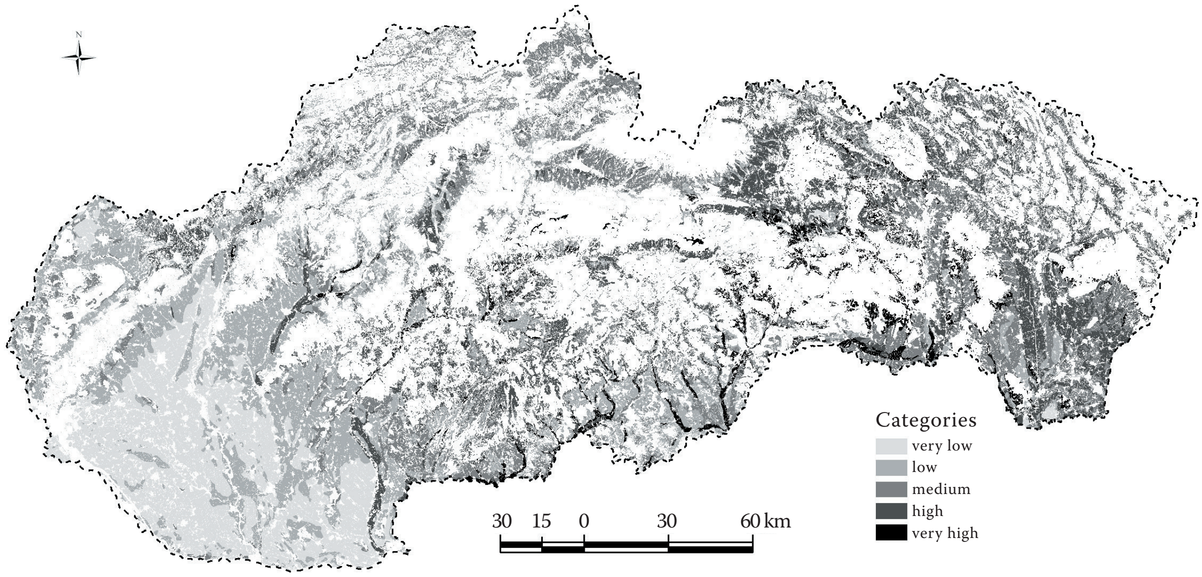

Figure 2. Soil transport potential of inorganic contaminants

ous soils on loess in the Danube lowland and the East-Slovak lowland, soils of the Záhorie lowland on sandy parent material, Chernozems and Calcaric Fluvisols without anthropogenic deposition.

Some Fluvial soils located in alluvial areas (along the rivers Váh, Hron, Bodrog) exhibit very low or low ability to immobilisation of the potential risk elements. It is caused by a higher amount of risk elements in sediments deposited on the flood plains and anthropogenic deposition; simultaneously the soil sorption potential (low pH of soils on non-calcareous parent material, low content of organic matter) of these soils is low. Soils with very low immobilisation potential are reported predominantly in Košice region and Banská Bystrica region, where soils are contaminated by geochemical anomalies and anthropogenic sources. This category comprises soils with high content of risk elements jointly with soil properties of this regions increase the risk of mobility and transfer of risk elements (Figure 1). The highest category of potential to immobilisation is identical with the lowest category of transport (Figure 2).

Potential of immobilisation and transport risks of risk elements is dependent on total amount of these elements in soil and potential of soil sorbents responsive to risk elements behavior and availability. Complex information about risk of elements allows only fusion of two information layers, the layer of contamination and the layer of potential sorption of soil. Both these layers were created using the data of specific Soil Science and Conservation Research Institute digital databases (Database of Soil Profiles of Geochemical Atlas of Slovakia and Soil Monitoring of Slovakia Database). The application of fusion of these information layers divided Slovak agricultural soil into five risk categories; the result is that more than 45.80% of agricultural soils belong to categories with very low and low risk to transport of potential risk elements, and only 5.18% of agricultural soils belong to category with high risk. This soil categorization represents the first attempt to evaluate the filtration/immobilisation function of selected inorganic risk elements as a whole and can provide primary information to improve soil management in specific agricultural localities with regard to the concentration of inorganic risk elements.

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