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PRELIMINARY CONTAMINATION RISK ASSESSMENT OF MINING WASTE USING SPATIAL ANALYSIS AND GEOCHEMICAL CHARACTERIZATION OF ROCK FORMATIONS. CASE STUDY IN HUNGARY

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Abstract

The Mine Waste Directive (2006/21/EC) requires the risk-based inventory of all mine waste sites in Europe. The geochemical documentation concerning inert classification and ranking of the mine wastes requires specific field study and laboratory testing and analyses of waste material to assess the Acid Mine Drainage potential and toxic element mobility. The procedure applied in this study used a multi-level decision support scheme for the inert classification of waste rock material including: 1) expert judgment, 2) data review, 3) representative field sampling and laboratory analysis and testing of rock formations listed in the National Inert Mining Waste List, and 4) requesting available laboratory analysis data from selected operating mines. Based on a preliminary expert judgment, the listed formations were classified into three categories. A: inert B: probably inert, but has to be checked, C: probably not inert, has to be examined. This paper discusses the heavy metal contamination risk assessment (RA) in the Hungarian quarry-mine waste sites. In total 30 waste sites (including both abandoned mines and active quarries) were selected for scientific testing using the EU Pre-selection Protocol. Altogether 93 field samples were collected from the waste sites including andesite, rhyolite, coal (lignite and black coals), peat, alginite, bauxite, clay and limestone. Laboratory analyses of the total toxic element content (aqua regia extraction), the mobile toxic element content (deionized water leaching) carried out according to the Hungarian GKM Decree No. 14/2008. (IV.3) concerning mining waste management. A detailed geochemical study together with spatial analysis and GIS were performed to derive a geochemically sound contamination RA of the mine waste sites. Key parameters such as heavy metals, in addition to the landscape metric parameter such as the distance to the nearest surface and ground water bodies, or to sensitive receptors such as settlements and protected areas calculated and statistically evaluated in order to calibrate the RA methods. Results show that some of the waste rock materials, assumed to be inert, were found non-inert. Thus, regional RA needs more spatial and petrological examination with special care to rock and mineral deposit genetics.

Keywords: risk assessment, pre-selection, rock formations, spatial analysis, geochemistry, inert, mining waste

INTRODUCTION

Mining has severe impacts on the environment, including contamination by toxic metals. In this context, Europe-wide survey identified wide-spread pollution problems caused by mining, abandoned mines in particular (COM, 2003). Since most of the elements used by the society come from mineral extraction (76 out of 90 frequently used elements), mining of mineral resources provide essential raw material for economic development (COM, 2005). Abandoned mines are more of a problem in areas with long historic mining like Europe, because mine closure practices have changed with time and environmental protection has not been considered for closed mines until recently (Jordan, 2004; Navarro et al., 2008). Apart from that abandoned mines are the same as active mines in terms of types of hazard and potential impact on the environment, their major problems are uncertainty in information and lack of control. Direct exposure to acid mine drainage (AMD) and sediments discharged from abandoned metal mines poses a serious hazard to aquatic biota and to humans (Peplow and Edmonds, 2005; Panagopoulos et al., 2009; Lei et al., 2010; Sarmiento et al., 2011). Younger et al. (2002) estimated that about 1,000 to 1,500 km of watercourses are polluted by metal mine discharges in the EU (estimate is for the former EU 15). There are an estimated 3 million potentially contaminated sites in the whole European Union, of which about 250,000 are actually contaminated and in need of remediation (EEA, 2007). Due to great volumes and slow chemical processes, mineralised rock in mine workings and in mine waste can release toxic compounds for a very long time on the scale of centuries and thousands of years (BAT, 2003). Thus, remediation of mine sites, including abandoned mines, has to consider long-term solutions and remediation technologies have to be sustainable for a long time (Sinding, 1999; Panagopoulos et al., 2009). Around the mine site, soils and surface water in the receiving environment are often contaminated with harmful elements or compounds (Puura et al., 2002; Sarmiento et al., 2011). These contaminated sites act as secondary sources for pollution, especially for historic sites (Jordan and D’Alessandro, 2004).

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Significance of contamination risk posed by mining is highlighted by mine accidents (Jordan and D’Alessandro, 2004). Examples of such accidents are Wales, UK, in 1966, Stava, Italy, in1985, Aznalcollar, Spain, in 1998, Baia Mare, Romania, in 2000 and most recently the catastrophic release of 850 million cubic meters of alkaline (pH >13) caustic red mud through the failed dam of the Ajka alumina plant depository on October 4, 2010 in Kolontar, Hungary, resulting in loss of lives and contamination of agricultural lands (Jordan et al., 2011). Limited financial resources restrict remediation of sites at regional scale, therefore, there is a strong need to develop methodologies that rank sites based on risk magnitude, rather than to produce absolute estimates of health/ecological impacts, or to prioritize the remediation actions (Long and Fischhoff, 2000; Marcomini et al., 2009). U.S. EPA (2001) gives a detailed description of risk-based assessment of mine sites. The effort required to identify and prioritize contaminated sites in Europe is considerable (EEA, 2005). Moreover, as for the prioritization process, the Soil Thematic Strategy for soil protection (COM, 2006) and the EU Mine Waste Directive (2006/21/EC), point out the need to develop spatial risk-based methodologies for sustainable management of contaminated sites and mining waste sites at regional scale.

The EU MWD Pre-selection Protocol (Stanley et al., 2011) is applied for contamination risk assessment of mine waste sites (Abdaal et al., 2013). The protocol has a ‘YES-or-NO’ questionnaire and consists of 18 questions using simple criteria available in existing databases readily enabling the preliminary screening of the mine waste sites for environmental risk (Fig. 1). This screening should result in the elimination of those sites which do not cause or have the potential to cause a serious threat to human health and the environment from the inventory of waste sites. Since the pre-selection protocol meant not to involve field sampling or laboratory analysis, any level will be sufficient to pass the test and select the site for further investigation as a precautionary measure. In case of lack of knowledge or information, i.e. in the presence of uncertainty, an ‘UNKNOWN’ response is entered for the particular parameter which is the same as a YES response and the site is selected for further examination which is a precautionary position. In this study the mine waste sites included inside the rock formations and delineated as polygons’ maps.

The geochemical documentation concerning inert classification and ranking of the wastes listed in the Inert Mining Waste List of the Hungarian Office of Mining and Geology involves the following procedures: 1) expert judgment, 2) data review, 3) representative field sampling and laboratory analysis of formations listed in the Inert Mining Waste List, and 4) requesting available laboratory analysis data from selected operating mines. Based on a preliminary expert judgment the listed formations classified into three categories. A: inert B: probably inert, but has to be checked C: probably not inert, has to be examined (Table 1). According to the Hungarian GKM Decree No. 14/2008 (IV.3) the mining waste classified to inert as if the content of substances potentially harmful to the environment or human health in the waste and in particular As, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, V and Zn, including in any fine particles alone in the waste, is sufficiently low to be insignificant human and ecological risk, in both the short and long term, in order to be considered as sufficiently low to be of insignificant human and ecological risk, the content of these substances shall not exceed the thresholds values for geological medium and underground waters identified as not contaminated in relevant legal rules.

Table 1 The inert-not inert classification of the listed rock formations based on preliminary expert judgment. A: inert B: probably inert, but has to be checked C: probably not inert, has to be examined. Number of waste sites and field samples for each rock group are shown.

Rock group	Rock type	Number of waste sites	Number of samples	Inert-Not Inert ranking
Coal	Lignite	2	10	C
	Black Coal	2	7	C
Peat		4	9	C
Alginite		2	5	B
Bauxite		2	6	B
Rhyolite tuffs		2	6	B
Clay	Clay	4	8	A-B
	Bentonite clay	1	1	A
Andesite		10	37	B
Limestone		1	4	A

A detailed geochemical study together with spatial analysis and GIS performed to derive a geochemically sound contamination RA of the mine waste sites in order to identify the current geochemical status of sampled rock materials from the 30 mine waste sites and to answer the question, if these waste rock materials (i.e. coal, peat, alginate, bauxite etc.) are still inert or non-inert. Distribution analysis applied to the median values of the elements As, Cd, Co, Cr, Cu, Mo, Ni, Pb, and Zn contents in order to find if there are any significant correlations between these elements to each other and to be compared to the local (country-specific) thresholds values for geological medium and underground waters in Hungary, in addition to the environmental limit values in Europe. The key landscape parameter, the distance from the waste sites (as centroid point of the formation polygon) to the nearest surface and ground water bodies, or to sensitive receptors (such as settlements and protected areas) was statistically calculated in order to evaluate the RA method (MWD Pre-selection Protocol, Fig. 1) and to identify local thresholds (median-based values of the measured distances from waste sites to the nearest pathways and sensitive receptors) more adopted the local conditions in Hungary.

The objective of this paper is to perform a pre-selection RA for selected rock types using the Geological map of Hungary as polygons, to evaluate the EU MWD Pre-selection Protocol (Stanley et al., 2011, Fig. 1) by applying it to real-life cases of 34 mines waste sites in Hungary. Three tests are carried out.

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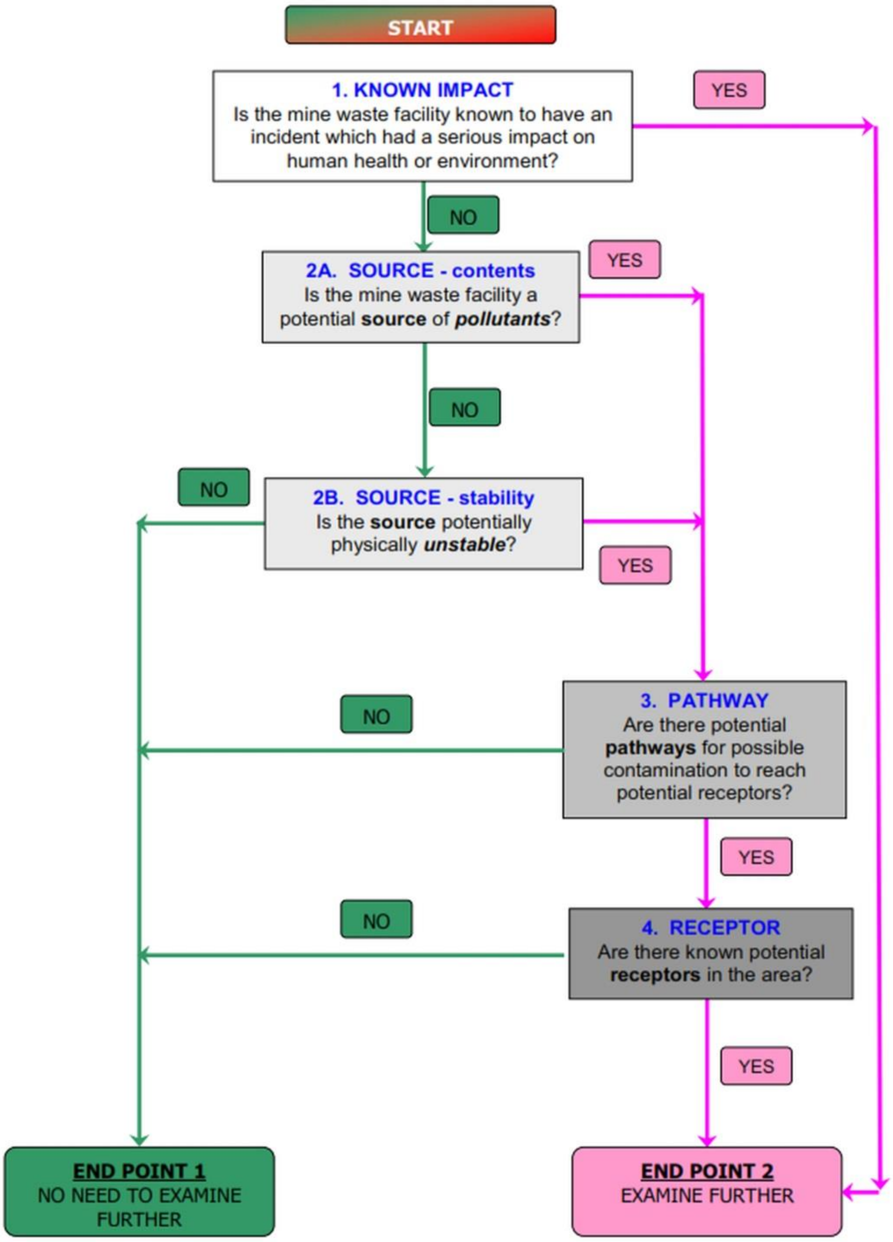

Fig. 1 The EU MWD Pre-selection Protocol Flowchart (Stanley et al. 2011)

First, a detailed statistical and landscape metric analyses are carried out for the Protocol threshold values (e.g. stream line density inside the polygons, number of patches (settlement, lake, Natura 2000 sites and agricultural areas inside each polygon) and counting the polygon overlap areas between Natura 2000 sites and the rock formation polygons.

Second, altogether 93 field samples of different rock types, collected from the waste sites, were analysed for the total toxic element content (aqua regia extraction), the mobile toxic element content (deionized water leaching) on the base of Hungarian GKM Decree No. 14/2008. (IV.3) concerning mining waste management. This detailed geochemical study together with spatial analysis and GIS were performed to derive a geochemically sound contamination RA of the mine waste sites.

Third, A, B and C inert ranking system based on the expert judgment, has been applied for the rock types in the waste sites, which were compared to a simple risk-based ranking of the mine waste sites based on the specific geochemical analysis results of the waste samples.

STUDY AREA

Altogether 30 waste sites of both abandoned mines and active quarries have been selected for scientific testing using the EU MWD Pre-selection Protocol (Fig. 2). 93 field samples have been collected from the waste sites including different rock samples such as; andesite, rhyolite tuffs, coal (lignite and black coals), peat, alginite, bauxite, clay and limestone according to the EuroGeoSurveys Geochemistry Expert Group Sampling Protocol (Fig. 3). Various wall rock and waste heap samples were collected for a detailed geochemical characterization.

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The alginite mined in Pula site (NW Hungary, Fig. 2) originated from biomass of fossil algae during several millions of years in volcanic craters. Its organic material content is about 5-50% (Szabo, 2004). Gömöryová et al. (2009) reported that tests of alginite from the deposits in Pula and Gerce showed that it can be used in agriculture and forestry to improve soil quality, soil water dynamics and nutrient content, to increase organic matter content, colloid content and to protect soil against acidification, desiccation and leakage of nutrients (Vass et al., 2003).

In the power generation sector, coal is playing a dominant role in the EU with 25% share of the total installed capacity and almost one-third of the power generation (Kavouridis and Koukouzas, 2008). Coal resources in Hungary are in total 3,300 million tons (Mt) with annual production between 9-10 Mt (of which 8 Mt is lignite) (Perger, 2009). At this rate of use the reserves could last for centuries. Three types of coal in Hungary were sampled: 1) black coal in southern Mecsek Mountains (Lower Jurassic- Lias) is Hungary's only black coal reserve, calculated to be 198.8 Mt. Due to the complicated geological circumstances and the high cost of exploitation, production was stopped in 2004. 2) Brown coal was widely mined throughout recent decades through the Transdanubian Mountains with good quality Eocene and Oligocene coal, supplying a significant amount of Hungary's energy needs. Mining has virtually stopped due to economic reasons, with remaining reserves calculated to be 170 Mt. There is only one mine operating and supplying the Vértes Power Plant. Cretaceous coal exploitation in the region ended in 2004, after resources ran out. Poor quality Miocene reserves can be found in Northern Hungary. While all underground mining were ceased, small open-pit mines are still operating and exploitation can be extended. 3) Lignite represents about 90% of the Hungarian coal reserves, which means that lignite is first on the Hungarian conventional energy sources. While some Miocene lignite reserves ran out in the Transdanubian Mountains in 1996, about 3000 Mt of Miocene-Pliocene lignite can be found in Visonta, Bükkábrány (Northern Hungary) and Torony (Western Hungary) (Fig. 2). Recently, the Visonta and Bükkábrány sites were subject to vast open-pit mining supplying the Mátra Power Plant, while the Torony site remains practically untouched by any mining activity (Hamor-Vido, 2004). Peat was used as a fuel from early times in Europe. It was exploited intensively in agriculture and currently there is a renewed interest in the material because of its potential as a general source of hydrocarbons and other more particular organic raw materials used industrially. Peat was invariably found with significant moisture content at the surface of the ground, within a depth of 2-15m (Spedding, 1988). Number of significant articles were published on different aspects of peat and its use (e.g. Del-Rio et al., 1992; Steinmann and Shotyk, 1997; Charman, 2002).

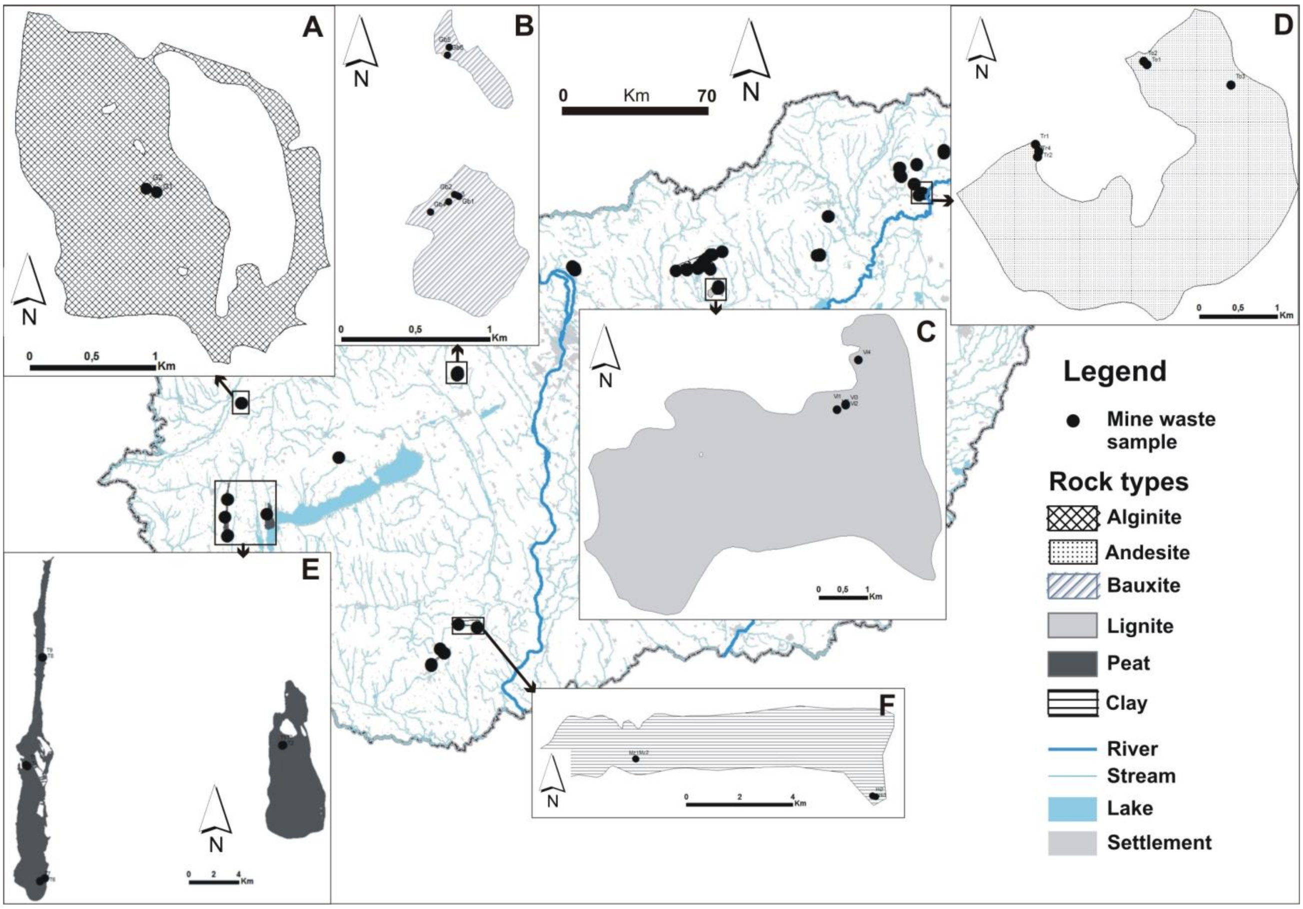

Fig.2 Examples of rock formations (as polygons) and locations of field sampling from abandoned mines and active quarries in Hungary. A) Pula Alginite Formation, B) Gant Bauxite Formation, C) Lignite Formation at Visonta, D) Andesite Formation in the TokajMts., E) Peat formation at Pölöske, F) Clay Formation at Maza

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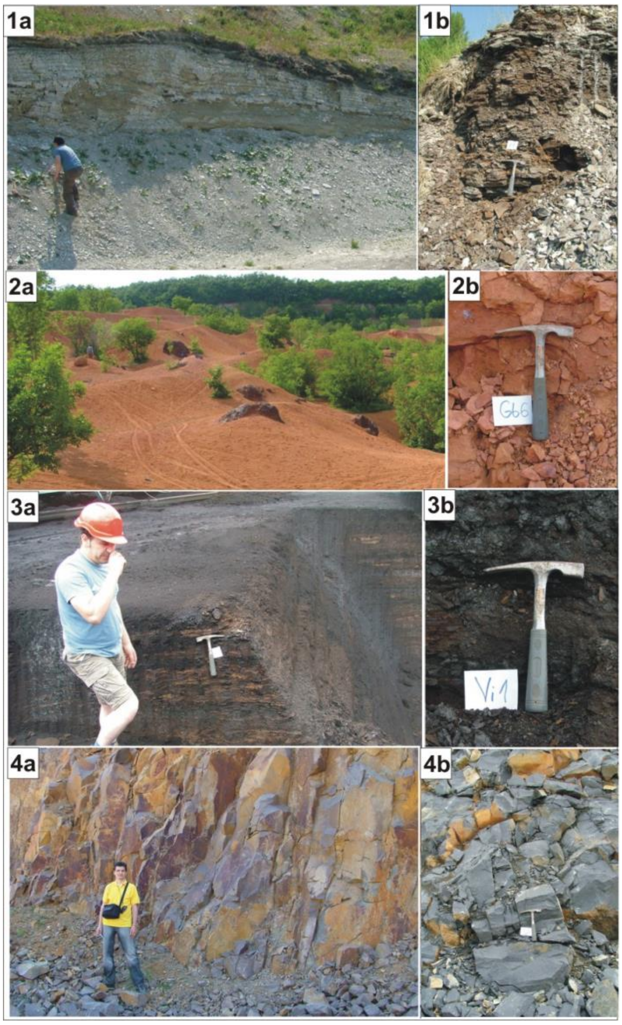

Fig.3 Field sampling for the EU Mine Waste Directive Inert waste testing and characterization in Hungary
1a and b. Alginite sampling in Pulla; 2a and b. Bauxite sampling in Gant; 3a and b. Lignite sampling in Visonta; 4a and b. Andesite sampling in Tokaj. See Fig.1 for sampling locations

MATERIALS AND METHODS

Sampling

This study used a multi-level decision support scheme including a representative field sampling and laboratory analysis of formations listed in the Inert Mining Waste List and requesting available laboratory analysis data from selected operating mines. Altogether 93 samples have been collected according to the EuroGeoSurveys Geochemistry Expert Group Sampling Protocol from 30 mine-quarry waste sites along Hungary (Fig. 2). Rock types and locations of samples are as follow: coal (10 lignite samples from Visonta and Bükkábrány sites and 7 black coal samples from Pécs-Vasas mine sites); 9 peat samples from Pölöske, Hahót and Alsopatak sites; 5 alginite samples from Pula and Gérce sites; 6 bauxite samples from Gánt site; 8 clay samples from Máza, Miskolc and Vác sites and one bentonite clay sample from Mád site; 37 andesite samples from Recsk, Tokaj, Komló, Tállya, Sárospatak and Tarcal mine sites; 6 rhyolite tuffs samples from Gyöngyöslymos and Felsoabasár sites and 4 limestone samples from Vác mine site (Fig. 2, Table 1). Selection of the samples at the site depends on the location of each sample, (e.g. lignite includes wall, overburden and waste samples), and on the rock type (mineral composition), (e.g. oxi-andesite and pyrite andesite samples were collected). The collected two kilograms of samples were always composed of three sub-samples located at a minimum of 10m distance and at any sudden change in the color of waste rock, a new sample was collected (Fig. 3).

Laboratory analysis

Laboratory analysis of the collected 93 field samples is carried out for the total toxic element content (aqua regia extraction) and the mobile toxic element content (deionized water leaching) at the Geological and Geophysical Institute of Hungary (MFGI) and 70 samples were selected for the analysis of different forms of sulfur (sulfuric acid potential) are carried on the ISD DUNAFERR laboratory at Dunaujvaros on the base of Hungarian GKM Decree No. 14/2008. (IV.3) concerning mining waste management. Samples were analyzed for total toxic element content (aqua regia extraction), the mobile toxic element content (deionized water leaching) with ICP-OES. Samples were air-dried, crushed in an agate mortar, passed through disposable sieve of 100 mesh, and digested by aqua regia with HNO_3, HCL and H_2O_2 under the ISO 11466 procedure (International Organization for Standardization 1995). All materials used during analytical determinations were kept in Teflon or other metal-free containers. To check the quality of preparation and analysis, replicate determinations were performed on approximately 25% of samples. Total element content of As, Cd, Co, Cr, Cu, Mo, Ni, Pb, V and Zn was defined by a mixed acid microwave unit digestion while deionized water leaching (pH=7) was performed to estimate the mobility of toxic elements in relative percent of total concentration.

Spatial data

Two types of data were used in this study. Waste site data includes (1) location of mines waste sites as polygons (*Fig. 2*), (2) composition of the mine waste including sulphides, toxic metals, and dangerous processing substances (Q2-Q3), (3) geometry of the waste site area (Q8) and slope of foundation (Q10), and (4) other data such as presence of impermeable layer beneath the waste site (Q12), and if the site is uncovered and thus the waste is exposed to wind or direct contact (Q13-Q14). Information on the mine waste site engineering design was obtained from mine archives, aerial photos and field studies. Spatial data include topographic data of location of settlements as polygons, surface water courses (streams and lakes).

Slope data calculated from the Hungarian national contour based military DDM 50m grid using ArcGIS $10^{®}$ software (Fig. 4). Then polygons of the rock formations added as overlay layer to the slope map in raster format using Spatial analysis tool in ArcGIS $10^{®}$. The slope value for each rock formation polygon (in degrees)

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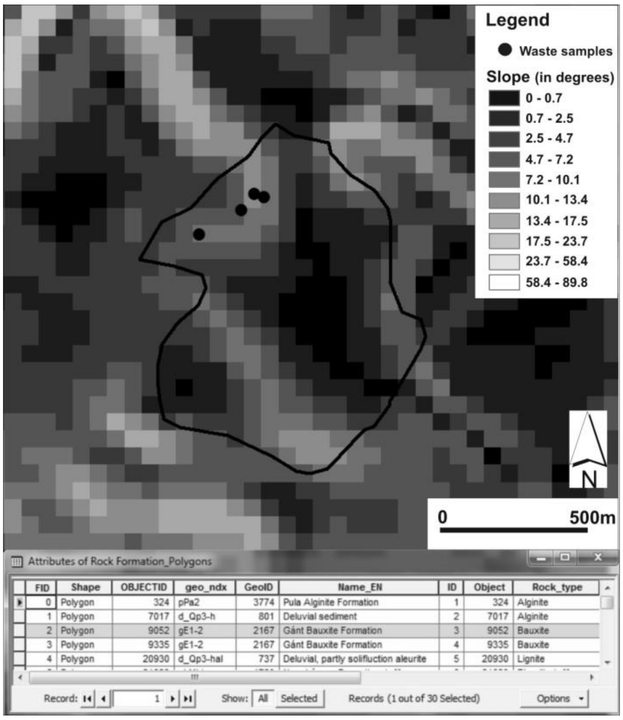
was counted as an average value from all pixels inside the polygon. Census data for 2009 is available from the Hungarian Central Statistical Office. Data on the national protected areas (Natura 2000 sites) and the location and status classification of groundwater bodies in Hungary under the Water Framework Directive (WFD) were obtained from the Hungarian Central Directorate of Water and Environment (VKKI) and from EEA website (Waterbase-Groundwater datasets). Land use/land cover data (LULC) maps at 1:100,000 scale were obtained from the European CORINE Land Cover website.



FID	Shape	OBJECTID	geo_ndx	GeoID	Name_EN	ID	Object	Rock_type
0	Polygon	324	pPa2	3774	Pula Alginite Formation	1	324	Alginite
1	Polygon	7017	d_Qp3-h	801	Deluvial sediment	2	7017	Alginite
2	Polygon	9052	gE1-2	2167	Gánt Bauxite Formation	3	9052	Bauxite
3	Polygon	9335	gE1-2	2167	Gánt Bauxite Formation	4	9335	Bauxite
4	Polygon	20930	d_Qp3-hal	737	Deluvial, partly solifluction aleurite	5	20930	Lignite

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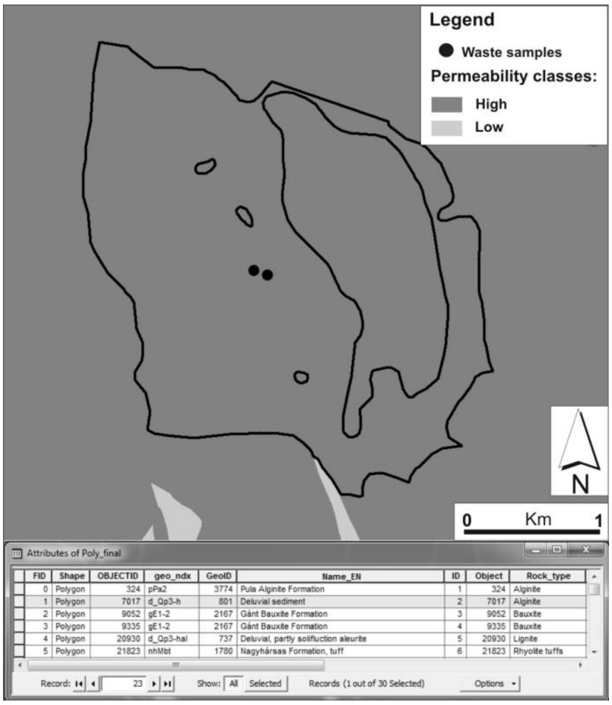
Fig.4 Calculation of the topographic slope for the sampled rock formations (as polygons) using the national contour-based spline-interpolated military 50m grid DEM. The same DEM is used for question Q10 of the EU Pre-selection protocol on the topographic slope below the mine waste site. Polygon highlighted is this example delineates the Gant Bauxite Formation including Bauxite samples from Gant bauxite mine

In order to identify if there is a high permeable layer beneath the mine waste site (Q12), a surface permeability map for the geological formations of the 1:100,000 surface geological map of Hungary has been constructed using ArcINFO® 10, on the basis of the physical and geochemical characteristics of the uppermost rock units. Three classes were distinguished (Fig. 5). Low-permeability formations (clay and other impermeable rocks), formations with medium-permeability (loess, sand-gravel and fractured metamorphic and volcanic rocks) and with high-permeability (karstified limestones and dolomites belong to this group). An example for the high permeability rock class is the Alginite Formation in Gérce Mining Area, NW Hungary (Fig.5). Polygons of the mines waste sites derived from the CORINE land cover 1:50,000 map (2000) were overlaid by Google Earth® aerial photographs (2013), in order to identify if the material within the mine waste sites is exposed to wind or not (Q13) or covered or not (Q14), (Fig. 6).



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3	Polygon	9335	gE1-2	2167	Gánt Bauxite Formation	4	9335	Bauxite
4	Polygon	20930	d_Qp3-hal	737	Deluvial, partly solifluction aleurite	5	20930	Lignite
5	Polygon	21823	nhMbt	1780	Nagyhársas Formation, tuff	6	21823	Rhyolite tuffs

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Fig.5 Surface permeability map developed to answer question Q12 of the EU MWD Pre-selection Protocol if there is a high permeable layer beneath the mine waste site. Ploygon highlighted is an example for the Alginite Formation at the Gérce Mining Area, NW Hungary. See text for details.

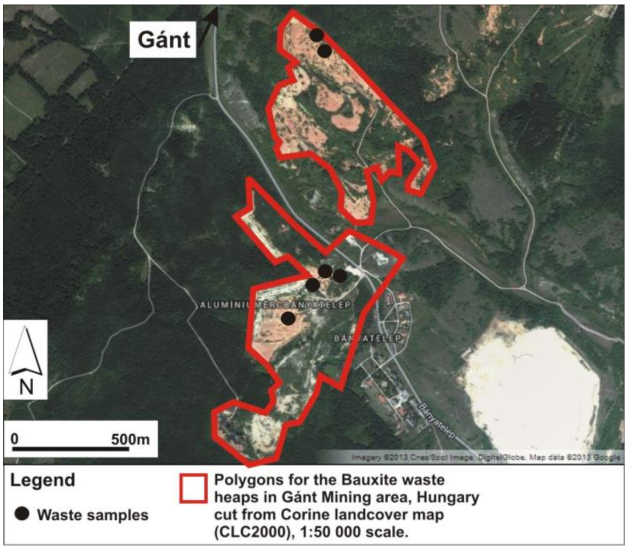

Fig.6 Polygons of the mine waste sites defined from the CORINE land cover map (CLC 2000) overlaid by Google Earth® aerial photographs (2013) to answer EU Pre-selection Protocol questions Q13 and Q14 on the air and direct contact pathways related to the cover of waste heaps, respectively. An example shows the Bauxite waste heap in Gánt Mining Area, Hungary.

In this study the mine waste sites were included inside the rock formations and delineated as polygons using ArcGIS 10® software (Fig.2). Altogether 30 mine-quarry waste sites both abandoned mines and active quarries, were selected for scientific testing using the EU MWD Pre-selection Protocol (Stanley et al., 2011; Fig. 1). Then, by running the protocol, the number of YES, NO and UNKNOWN responses are registered for each site.

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The proportion of the certain to uncertain responses for a site and for the total number of sites may give an insight of specific and overall uncertainty in the data we use (Table 2). The distance from mine-quarry waste sites to the nearest receptors such as human settlements (Q15) is measured using Proximity Analysis tools (Point Distance and Generate Near Table) in ArcINFO® 10 (Fig.2).

Table 2 Summary statistics of the EU Pre-selection Protocol responses of questions Q1-Q18, showing the number of YES, NO and UNKNOWN responses (U) based on the EU thresholds

EU Pre-selection Protocol		Number of sampled sites	EU thresholds		
			YES	NO	U
Impact	Q1	30	0	30	0
Source	Q2	30	12	18	0
	Q3	30	14	16	0
	Q5	30	0	30	0
	Q8	30	30	0	0
	Q10	30	21	9	0
Pathway	Q11	30	16	14	0
	Q12	30	21	9	0
	Q13	30	18	12	0
	Q14	30	18	12	0
Receptor	Q15	30	26	4	0
	Q16	30	22	8	0
	Q17	30	19	11	0
	Q18	30	28	2	0

Statistical analyses were carried out using STATGRAPHICS Centurion XV.II® software (*Table 3*), such as the topographic slope (Q10) and the measured distance to the nearest surface water courses (Q11), settlements (Q15), ground water bodies (poor status) (Q16), protected areas (Natura 2000 sites, Q17) and agricultural areas (Q18). Summary statistics of the analyzed heavy metal concentrations from the mine waste sites (Aqua regia leaching analysis) were compared to the local (country-specific) thresholds values for geological medium and underground waters in Hungary, in addition to the environmental limit values in Europe. Spearman's rank correlation is performed on the total concentrations of the analysed elements to determine the relationships and variance between elements in the studied rock samples. Moreover, the Ficklin Diagram is constructed for the sum of heavy metals Zn, Cr, Cd, Pb, Co and Ni against pH in the deionized water leaching (DW, Fig. 7).

RESULTS AND DISCUSSION

The contamination RA according to the EU MWD Pre-selection Protocol is carried out using the EU thresholds (slope ≤ 5^o and 1 km distance and number of people in the nearest settlement ≥ 100). The YES, NO and UNKNOWN responses of the EU MWD Pre-selection Protocol (Fig. 1) were registered and calculated for each question in Table 2. In this study each rock formation was treated as a waste site and projected in the map as one or more polygons (Fig. 2). Questions describe if the mine uses any dangerous chemicals in processing minerals (Q4), the geometry of the tailings lagoon height and area (Q6-Q7) and for the waste heap height (Q9) of the Pre-selection Protocol are not fit to the rock waste sites and were skipped in this study (Table 3). Out of 30 mine waste sites, none of sites have a documented incident (Q1, Jordan et al. 2011). In Q2, 12 sites with YES responses were producing waste with sulphide minerals, 18 sites have NO responses. While in Q3, 14 sites were producing minerals with toxic heavy metals. In Q5, all sites are waste heaps and none of sites are tailings la-

Table 3 Class boundaries of the EU MWD Pre-selection Protocol parameters based on the natural-breaks found in the corresponding cummulative histograms. Class boundaries were used to define thresholds adapted to local conditions in Hungary

Question	Class boundaries	Class-Range	Median of class	Median of all sites	Number of sites
Q10		Topographic slope below waste site (degree)			
	1.-20	1.-20	10	10	30
Q11		Distance to the nearest surface water course (m)			
	<1300	0-1280	188		22
	>1300	2219-5376	2861	631	8
Q15		Distance to the nearest settlement (m)			
	0	0	0		14
	>0<=1000	82-838	548		12
	>1000	1585-3319	2350	150	4
Q16		Distance to the groundwater bodies of 'poor status' (m)			
	0	0	0		18
	>=36	36-14717	5229	0	12
Q17		Distance to the nearest Natura 2000 sites (m)			
	0	0	0		12
	>0<=1000	158-713	286		6
	>1000	1072-5548	2416	224	11
Q18		Distance to the nearest agricultural areas (m)			
	0	0-861	0		24
	59-2092	3688-3976	359	0	6

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goon. In Q8, all 30 waste heap sites with YES responses are greater than 10,000 m^2 in surface area. The slope of the foundation upon which the waste heap rests is of concern with respect to stability. The greater the slope angle the greater the risk of waste heap failure. The EU threshold chosen is 1:12 which equates to 8.3% or a slope angle of almost 5°. Based on the slope values derived from the 50m DEM, 16 waste heap sites with YES responses are greater than or equal 1:12 (5^o) in slope (Q10). This shows that most of the sites were located in hilly areas. The use of the surface permeability map (Fig.5) developed to generate answers for Q12, resulted in 21 waste sites with YES responses and underlain by medium and high permeable layer, while 9 sites underlain by low permeable layers. When the mine waste site is covered and the original material is not accessible this means there is no direct contact with receptors. In Q13, 18 sites were exposed to the wind and 12 sites were not. While in Q14, 18 sites were uncovered and 12 sites were covered with water, vegetation, soil and forest (Fig.6).

For Q11, 16 sites are within 1 km distance to the nearest surface water bodies (streams and lakes). In Q15, 26 mine waste sites are within 1 km distance to nearest human settlements with >100 people, indicating that these sites require prime attention if settlement protection is the concern. In Q16, 22 sites are within 1 km distance to the groundwater bodies of less than good status. For Q17, 19 waste sites are within 1 km distance to the national protected Natura 2000 sites. 12 waste sites were located completely inside the Natura 2000 sites), this calls for immediate special attention if landscape protection is a priority. Moreover, in Q18, 28 waste sites are within 1 km distance to the agricultural areas including arable lands, pastures, heterogeneous and permanent crops, 24 sites are completely located inside the agricultural lands (Table 2).

Distribution analysis performed on the heavy metals (Table 3) identified various sub-groups in the parameter thresholds of the EU Pre-selection Protocol. For example, in Q10, altogether 30 waste sites have one class of topographic slope ranges from 1-20^o. This result suggests the median slope value of all waste sites 10^o as a natural threshold reflecting the local Hungarian conditions, instead of the original 5^o slope threshold. In Q11, 22 waste sites were located within distance 0-1280m to the nearest surface water bodies and 8 sites are within distance 2,219–5,376m. This shows that almost 73% of the mine waste sites are significantly (at the 90% confidence) closer (≤1280m) to receiving streams than the other sites, thus the 631m (medial value of all sites) threshold may better reflect the local topographic conditions for this question. In Q15, 14 waste sites were located directly inside the nearest settlement (distance=0), indicating that these sites require prime attention if settlement protection is the concern, 12 sites are within distance 82-838m to the nearest settlement and 2 sites are within distance 1,585-3,319m to the nearest settlement. This result suggests the distance 150m (medial value of all sites) as a local threshold for this question in Hungary. It is interesting that 18 waste sites lie directly above the groundwater bodies with ‘poor status’ (Q16) and 12 sites are located inside the protected Natura 2000 sites (Q17). While in Q18, 24 waste sites are located inside the agricultural areas (Table 3).

A preliminary risk-based site ranking is possible based on the EU thresholds (slope of almost 5^o and 1km distance) by counting and ranking the YES responses of the Pre-selection Protocol, and ranging in scores from 5 to 10. Obviously, if there is more than one hazardous material at the source or there are multiple contamination pathways and receptors the site has a higher risk. A simple risk ranking of the rock formations based on the YES responses in descending order as follows: black coal and peat (10 YES), alginite (9 YES), lignite and clay (8 YES), bauxite (7 YES), bentonite-clay (6 YES) and andesite and rhyolite tuffs (5 YES). In summary, after the existing pre-screening risk assessment of the mine waste sites in Hungary, 28 sites were directed to EXAMINE FURTHER based on the EU thresholds and two sites with no risk (one Bauxite site has no pathway and one Andesite site has no sensitive receptor).

Table 4 summarizes the estimated heavy metal concentrations from the mine waste sites (aqua regia extraction) with respect to the environmental limit values in Hungary and Europe. In case of central tendency expressed by the Median, the analyzed heavy metals are in descending order; Zn>V>Cu>Cr>Pb>Co>Ni>As>Mo>Cd. This result shows that Zn has the highest median (24.6 mg/kg) and Cd has the lowest Median (0.11 mg/kg). In case of spread expressed by IQR/Med (Interquartile range/Median), the heavy metals are in descending order; Ni>As>Cr>V>Pb>Co>Cd>Zn>Cu. It is obvious that Ni has the highest spread (5.11) and Cu has the lowest (1.11). While spread expressed by Range/Median, the heavy metals are in descending order; Ni>Cr>Mo>Co>Zn>Pb>As>Cd>Cu>V. Ni still has the highest spread (327.6) but in this case V has the lowest spread (8.42).

Total concentrations of heavy metals as defined by aqua regia extraction were compared to the environmental limit values in Hungary and to the European environmental geochemical background values based on the FOREGS European Geochemical Atlas (Table 4) as follow: the Mean of As (18.17 mg/kg) exceeds the tolerated limit in Hungarian soils (15 mg/kg) and exceeds the Mean value of EU FOREGS geochemical background value (10 mg/kg). At the same time, the Mean of Cd (0.33 mg/kg) is less than the tolerated limit in Hungarian soils (1 mg/kg) and exceeds the Mean of EU FOREGS (0.3 mg/kg). The Mean of Ni (61) exceeds the tolerated limit in Hungarian soils (40) and exceeds Mean of EU FOREGS (31). Moreover, the median of Cu (12.3) exceeds the median of EU FOREGS (12).

The Spearman’s rank correlations depicted in Tables 5 and 6 were performed between each pair of the analysed heavy metals from the waste sites by aqua regia and deionized water leaching analyses, respectively. In contrast to the more common Pearson correlations, the Spearman coefficients are computed from the ranks of the data values rather than from the values themselves.

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Table 4 Summary statistics of heavy metal concentrations from the mine waste sites (aqua regia extraction in mg/kg) in respect to the environmental limit values in Hungary and the European Top Soil Baseline Values. Minimum (MIN), maximum (MAX), median (MED) and spread expressed as median absolute deviation (MAD), lower quartile (LQ), upper quartile (UQ), Interquartile range (IQR), Standard deviation (SD). Bold figures show those heavy metal concentrations higher than the environmental standard limits (i.e. the tolerated limit in Hungarian soils or EU FOREGS Geochemical Atlas baseline value for top soils).

	As	Cd	Co	Cr	Cu	Mo	Ni	Pb	V	Zn
Min	**0.6**	**0.06**	0.018	0.537	0.766	**0.2**	0.4	1.15	3	0.1
LQ	1.54	0.073	2.92	2.58	6.8	0.2	1.88	4.56	5.48	14.4
Med	3.93	0.117	5.12	8.11	**12.3**	0.2	4.79	7.08	18.4	24.6
UQ	14.3	0.22	9.98	21	20.5	0.2	26.4	14.3	38	46.1
IQR	12.76	0.152	7.06	18.42	13.7	0	24.52	9.74	32.52	31.7
Max	**247**	6.07	**416**	1185	**573**	**24.3**	1570	468	158	1690
Mean	**18.17**	**0.33**	**19.92**	**56.24**	**34.16**	**1.08**	**60.89**	23.4	28.91	**84.28**
Range	246.4	6.01	415.9	1184.4	572.2	24.1	1569.6	466.8	155	1689.9
SD	43.31	0.87	63.67	170.09	92.44	2.96	223.3	68.72	31.64	255.83
MAD	3.07	0.057	3.52	6.34	5.7	0	4.25	3.84	13.94	15.8
Mode	0.6	0.06	11.5		13.9	0.2	0.4		3	0.1
Range/Med	62.69	51.36	81.24	146.04	46.52	120.5	327.68	65.93	8.42	68.69
IQR/Med	3.24	1.29	1.37	2.27	1.11	0	5.119	1.37	1.76	1.28
MAD/Med	0.78	0.48	0.68	0.78	0.46	0	0.88	0.54	0.75	0.64
Environmental standard values in Hungary and the European Top Soil Baseline Values (FOREGS Atlas)										
Tolerated limit in Soils, Hungary	15	1	30	75	75	7	40	100		200
EU FOREGS Min	<0.5	<0.01	<1	1	1	<0.1	<2	<3		4
EU FOREGS Max	220	14.1	255	2340	239	21.3	2560	886		2270
EU FOREGS Med	6	0.145	7	22	12	0.62	14	15		48
EU FOREGS Mean	9.88	0.28	8.91	32.6	16.4	0.94	30.7	23.9		60.9

Thus they are less sensitive to outliers than the Pearson coefficients. Table 5 shows that all the elemental pairs of Aqua regia leaching (with bold figures) have strong correlations with each other which P < 0.05, for example Pb and Zn(r = 0.63, df = 93, P < 0.05), and Ni and Pb (r = 0.71, df= 93, P < 0.05) etc. However, pairs such as Co/Mo, Cr/Mo, Cu/Mo, Mo/Ni, Mo/Pb and Mo/Zn show a weak correlation with each other (P>0.05). Table 6 shows that all the elemental pairs of deionized water leaching (with bold figures) have strong correlations with each other, for example between Co and Ni (r= 0.8). Moreover, pairs such as As and Cd, As and Pb, Cd and Cu, Cd and Pb, Co and Mo, Cr and Zn, Cu and Pb, Mo and Pb and Mo and Zn show a weak but significant correlation with each other (p>0.05). Strong correlations signify that each pair of elements may have common contamination sources. Further detailed studies of physico-chemical properties and metal associations are needed to ascertain these results.

Table 5 The Spearman's rank correlation coefficients between concentrations of heavy metals from the waste sites (aqua regia extraction). Significant correlation coefficients are in bold; $\rho < 0.05$.

	As	Cd	Co	Cr	Cu	Mo	Ni	Pb	Zn
As									
Cd	**0.45**								
Co	**0.41**	**0.34**							
Cr	**0.37**	**0.39**	**0.72**						
Cu	**0.42**	**0.42**	**0.77**	**0.66**					
Mo	**0.35**	**0.22**	-0.13	-0.12	0.06				
Ni	**0.57**	**0.5**	**0.72**	**0.81**	**0.7**	0.19			
Pb	**0.5**	**0.58**	**0.61**	**0.57**	**0.6**	0.09	**0.71**		
Zn	**0.31**	**0.39**	**0.86**	**0.61**	**0.71**	-0.17	**0.57**	**0.63**	

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The Ficklin Diagram (adapted after Plumlee et al., 1999) showing the sum of heavy metals Zn, Cr, Cd, Pb, Co and Ni from deionized water leaching (DW) analysis is plotted against pH (*Fig.7*). Differences in the sum of the previous base metals have proven the most diagnostic in differentiating between different geologic controls. This diagram shows two groups as follow.(1) Coal, Lignite, Peat and Bauxite samples are distributed from acid-high acid to near-neutral environments, with low to extreme concentrations of dissolved metals. (2) Alginate, Andesite, Clay, Rhyolite tuffs and Limestone samples are distributed in near-neutral environments, with low to high concentrations of dissolved metals.

Multivariate analysis such as CA and PCA using the analysed trace elements could not identify significant groups of samples. This is not unexpected due to the heterogeneity of the sampled rock types. It seems that specific rock formations with ore minerals content, including pyrite with acid generation potential, such as some andesites and coals are distinct from the non-mineralised as shown by the Ficklin Diagram (Fig.7).

The relative mobility of heavy metals in the various sampled rock formations was calculated as the percentage of the mobile element content (deionized water leaching) to the total element content (Aqua regia extraction) for the 93 samples. Then the median value of these mobility percentages was calculated for each rock type (*Fig.8*). Results show in Black Coal samples, the relative mobility of the heavy metals reduced in the following order: Zn (30.7) > Co (29.5) > Ni (26) > V (11.2) > Cd (4.6) > Cu (2.3) > Pb (0.3) > As (0.27) > Mo (0.26). In Lignite samples, Mo (5) > V (4.6) > As (1.4) > Cd (1.2) > Zn (0.8) > Pb (0.5) > Co (0.3) > Ni (0.2) > Cu (0.16) > Cr (0.1). In Peat samples, Zn (31) > V (16) > Mo (6) > Cd (3) > As (2.5) > Co (1.3) > Pb (0.8) > Cu (0.7) > Cr (0.4) > Ni (0.3). In Bauxite samples, Mo (5) > Cd (0.7) > V (0.4) > As (0.3) > Co (0.11) > Pb (0.1) > Zn (0.06) > Cu (0.05) > Ni (0.03) > Cr (0.01). In Alginite samples, Mo (175) > V (2.1) > Cd (0.6) > As (0.2) > Pb (0.08) > Cu (0.04) > Ni (0.03) > Zn (0.025) > Co (0.02) > Cr (0.01). In Clay samples, Mo (8.7) > V (2.3) > Cd (1.8) > Zn (0.5) > As (0.4) > Pb (0.2) > Co (0.1) > Ni (0.07) > Cu (0.05) > Cr (0.04). In Andesite samples, Mo (5) > Cd

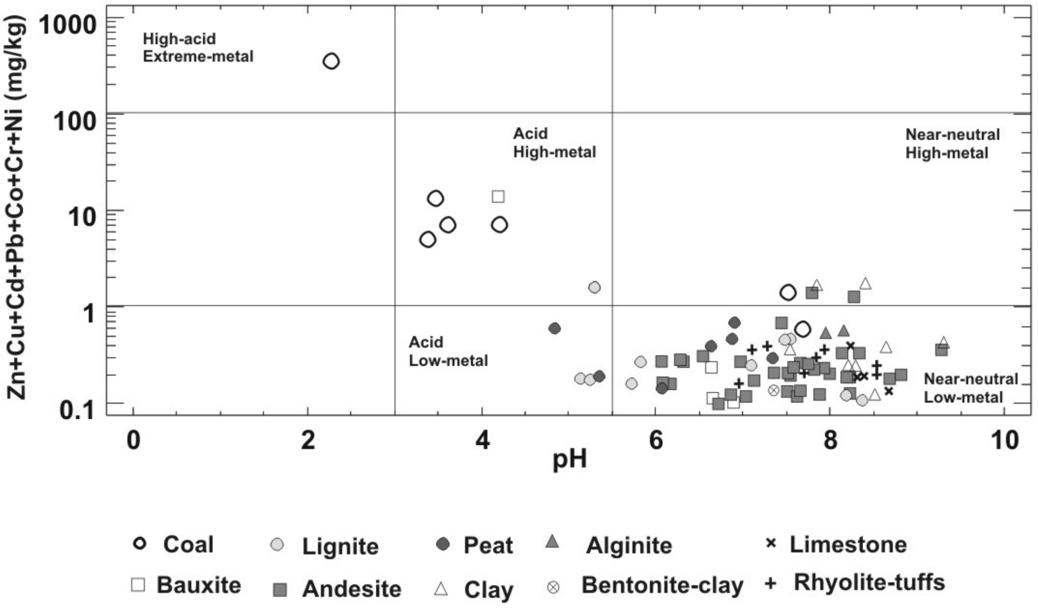

Fig.7 Ficklin Diagram showing the sum of heavy metals Zn, Cr, Cd, Pb, Co and Ni plotted against pH in the deionized water leaching (DW). Note that acid generation potential (pH<5.5) is for coal, lignite and peat rocks, in addition to a bauxite sample. Elevated mobile heavy metal content is associated with coal, andesite and some clay and a bauxite samples. See text for details.

Table 6 The Spearman's rank correlation coefficients between concentrations of heavy metals from the waste sites (deionized water leaching). Significant correlation coefficients are in bold; $\rho < 0.05$.

	As	Cd	Co	Cr	Cu	Mo	Ni	Pb	Zn
As									
Cd	0.12								
Co	**0.22**	**0.27**							
Cr	0.03	**0.25**	**0.26**						
Cu	0.17	0.16	**0.35**	0.18					
Mo	**0.28**	0.08	-0.04	0.1	**0.27**				
Ni	**0.21**	**0.3**	**0.8**	**0.28**	**0.47**	0.16			
Pb	-0.04	0.14	**0.31**	**0.24**	0.14	0.01	**0.26**		
Zn	0.14	0.02	**0.27**	-0.04	**0.47**	0.11	**0.35**	0.12	

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(4) > As (2.5) > V (1.6) > Ni (0.7) > Pb (0.6) > Zn (0.4) > Co (0.2) > Cu (0.15) > Cr (0.14). While in Rhyolite tuffs samples, V (16.6) > Mo (5) > Ni (4) > Cd (3) > As (2.3) > Co (2) > Zn (1.2) > Cr (0.8) > Cu (0.7) > Pb (0.2). It is obvious that Mo had the highest mobility in Lignite, Bauxite, Alginite, Clay and Andesite rock samples and Zn had the highest mobility in Black coal and Peat samples. While, V had the highest mobility in Rhyolite tuffs samples (Fig. 8).

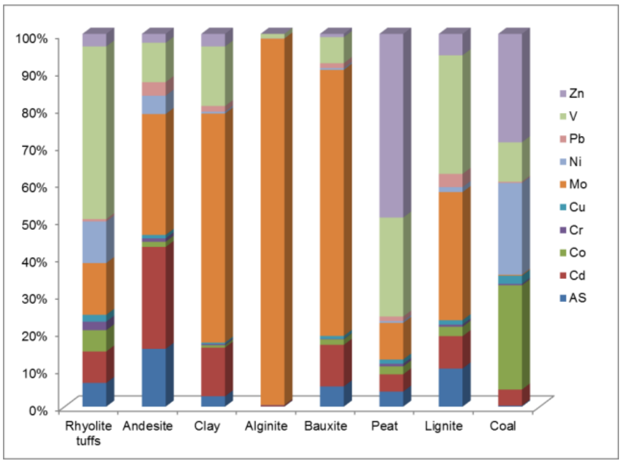

Fig. 8 Distribution of the relative mobility (%) of heavy metals in the various sampled rock formations

Based on the expert judgment, the listed rock formations were classified into three preliminary categories. A: inert B: probably inert, but has to be checked C: probably not inert, has to be examined (Table 1). According to the geochemical analysis results in this study, coal (black coal and lignite) and peat samples are not inert and classified into group C which matches with the preliminary expert judgment. While alginite, bauxite, rhyolite tuffs and clay samples are probably inert and classified into B group which also matches with the preliminary expert judgment. Moreover limestone and clay samples are inert (A group). It is interesting to report that andesite samples are probably inert (B group) and according to our geochemical analyses, it was found that 5 andesite samples contain higher concentrations of the heavy metals Ni, Zn Cu, Cr and Co than the minimum, median and mean values of the Hungarian standards. While As is even higher than the maximum values of the national environmental standards. These results may suggest that those 5 andesite samples with higher heavy metal concentrations could classify the andesite rock formation into the B or C groups.

CONCLUSIONS

This paper discusses the heavy metal contamination risk assessment (RA) in a selected group of the mine waste sites in Hungary. A detailed geochemical study together with spatial analysis using GIS was performed to derive a geochemically sound contamination RA of the mine waste sites. Key parameters such as heavy metals, in addition to the landscape parameter such as the distance to the nearest surface and ground water bodies, or to sensitive receptors such as settlements and protected areas are calculated and statistically evaluated in order to calibrate the RA methods.

In deionized water leaching, coal, lignite, peat and bauxite samples were located in one distinct group in the Ficklin diagram and distributed from acid to near-neutral region in the graph with low to extreme concentrations of dissolved metals. While alginate, andesite, clay, rhyolite tuffs and limestone samples were located in one group and distributed in the near-neutral region, with low to high concentrations of dissolved metals.

A simple risk ranking of the waste rock materials based on the YES responses to risk factor questions in descending order of risk resulted as follows: black coal and peat (10 YES), alginite (9 YES), lignite and clay (8 YES), bauxite (7 YES), bentonite-clay (6 YES) and andesite and rhyolite tuffs (5 YES). After the existing prescreening risk assessment of the studied waste sites in Hungary, 28 sites were directed to EXAMINE FURTHER based on the EU thresholds and two sites with no risk.

Results show that some of the waste rock materials, assumed to be inert such as the 5 andesite sites that contain higher concentrations of the heavy metals As, Ni, Zn Cu, Cr and Co than the minimum, median and mean values of the Hungarian standards. These results may suggest that those 5 andesite samples with higher heavy metal concentrations could reclassify the andesite rock formation into the B and C groups. Thus, regional RA needs further spatial and petrological examination with special care to rock and mineral deposit genetics.

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