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A Geoszférák időszaki kiadvány köteteinek grafikai terve Jacob Péter és Pál-Molnár Elemér munkája

Címlapfotó: Az aljzati magaslat és környezetében számított hőmérsékleti mező keresztmetszeti képe
(Vass et al., 250. oldal)

GEOSZFÉRÁK
2014

A Szegedi Tudományegyetem Földtudományok Doktori Iskola
és a Környezettudományi Doktori Iskola (Környezeti geográfia program)
eredményei

Szerkesztette
Unger János – Pál-Molnár Elemér

GeoLitera
SZTE TTIK Földrajzi és Földtani Tanszékcsoport
Szeged, 2015

környezettörténeti és régészeti kutatások eredményeit, megvizsgálta a környezetben történt változásokat, a területen élt régészeti kultúrák lakosságának települési helyszíneit, valamint a környezettörténeti vizsgálat eredményeit hozzárendelte a megfelelő régészeti korszakokhoz.

Pap Ági a kulturális örökség globális felértékelődése kapcsán elemezte az örökség újszerű megítélését a magyarországi gyakorlatban, illetve az azokra vonatkozó hasznosítási és kezelési elképzeléseket, melyek gyakran a város fejlődési irányvonalainak meghatározásában részt vevő aktorok eltérő nézőpontjainak ütközésével jár.

Sándor Renáta a talaj–növény–légkör rendszer folyamatait modellezte, melynek során leírta a talajban történő folyamatok léptékfüggését a növényzet figyelembevételével. Mivel a víz- és tápanyagtranszport folyamatok fő mozgatóereje a talajnedvesség potenciálkülönbsége, ezért több vizsgálatot is végzett a talaj nedvességállapotának minél pontosabb megállapítására, illetve a nedvesség talajszelvényen belüli eloszlására.

Sümeghy Borbála vizsgálata az Alföld harmadik legnagyobb hordalékkúpjára irányult, amelyen fluviális formák dominálnak és különlegesen szabályos legyezőszerű alakja van. Munkájában feltárta a Maros hordalékkúpi rendszerét és meghatározta a külső hatásokra adott válaszreakcióit is, valamint feltárta, hogy az elmúlt 20 ezer évben a Maros rendkívül dinamikusan változó rendszert alkotott, mindig az egyensúlyi állapot elérésére törekedve.

Szalontai Csaba vízrajzi és domborzati adottságok alapján arra a következtetésre jutott, hogy Szeged középkori lakott részei kevéssé voltak alkalmasak a tartós megtelepedésre, ugyanis a rendelkezésre álló kis szigetek kevés védelmet nyújtottak az árvízi fenyegetettséggel szemben és eltartó képessége is igen alacsony volt. Ugyanezen adottságok elemzése alapján viszont sikerült meghatároznia olyan területeket, amelyek jó és kedvező életföldrajzi körülményeket teremtettek az itt megtelepedni szándékozó népek számára.

Szolnoki Zsuzsanna Szeged példáján vizsgálta az antropogén tevékenységek együttes hatását a városi „pufferzónában" elhelyezkedő, növénytermesztési funkcióval rendelkező, művelt kerti talajok tulajdonságaira és nehézfémterheltségére. Elkülönítette a kerti talajokban antropogén forrásból dúsuló fémek körét a kizárólag geogén eredetű fémektől, továbbá az elemek mobilitási sajátságait is elemezte a vizsgált talaj–növény rendszerben.

Vass István munkája az aljzati fluidum-tárolók komplex repedéshálózat vizsgálati módszeren alapuló hidrodinamikai és hőtranszport modellezésére irányult. Régóta ismert, hogy regionális földtani okok miatt a Pannon-medence mélyebb üledékes részmedencéiben sok helyen magas túlnyomás alakult ki. A numerikus modell azt szemlélteti, hogy bizonyos geológiai szituációkban a metamorf kőzetegyüttes rossz szivárgási tulajdonságai ellenére a fluidum beszivárog és felfelé migrál a nyomáskompenzáció által létrehozott hidrodinamikai „kéményen" keresztül. A kapcsolt hőterjedés modellezés és analitikus számítások eredményei azt mutatták, hogy a hidraulikai „kémény" középpontjában jelentkező maximális hőanomália mértéke megközelíti a 20 °C-ot, s ebben mind a konduktív, mind a konvektív hőterjedés közel azonos szerepet játszott.

Az előző évek tapasztalatai alapján biztos vagyok benne, hogy változatos témájú kötetünk most is tetszést arat a Kedves Olvasók körében.

Szeged, 2015 márciusa

Unger János
koordinátor
SZTE Földtudományok Doktori Iskola

ENVIRONMENTAL MODELLING AND SPATIAL LANDSCAPE ANALYSIS FOR THE CONTAMINATION RISK ASSESSMENT OF SENSITIVE AREAS

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ABSTRACT

This study evaluates the EU MWD Pre-selection Protocol (Stanley et al., 2011) by applying it to real-life cases and adopting it to country-specific conditions (Abdaal et al., 2013). Altogether 145 ore mine waste sites in Hungary were selected for scientific testing and evaluation using the EU MWD Pre-selection Protocol. Key parameters, formulated as questions in the EU MWD Pre-selection Protocol, are linked to a GIS system and key parameters such as the topographic slope and distance to the nearest surface and groundwater bodies, to settlements and the Natura 2000 protected areas were calculated and statistically evaluated in order to adjust the RA models to country-specific conditions in Hungary. In order to assess the sensitivity of mine waste site risk assessment in response to various methods the EU MWD Pre-selection Protocol was compared to the European Environmental Agency (EEA) Preliminary Risk Assessment Model for Soil contamination in Europe (PRAMS). As the second component of the research project, the heavy metal contamination risk assessment (RA) based on actual laboratory analysis of collected samples was performed for selected 30 min-quarry waste sites in order to study the inert characteristics of the potentially generated mine wastes, in accordance with the EU MWD legislation. In addition to detailed geochemical study, spatial analysis using ArcGIS was performed to derive a geochemically sound contamination RA of these mine waste sites. As the third component of this research, the relationship between selected water quality variables (e.g. Ni, Mn, Cr, Zn and conductivity) in streams nearby the studied 33 mining waste sites and the landscape metrics of watersheds of these mining sites was investigated and analysed. It is concluded that the Mean Shape Index (MSI) and the Main Fractal Dimension Index (MFRACT) are the most important "key" landscape indcies in years 2000 and 2006 respectively, from the stream water quality heavy metal contamination point of view.

1. Introduction

Major incidents involving mine waste facilities and poor environmental management practices have left the legacy of thousands of contaminated sites in historic mining areas like the Carpathian Basin. These mining-specific problems require special tools to address the complexity of the environmental problems of the mining-related contamination. Significance of contamination risk posed by mining is also highlighted by large mine accidents such as those in Baia Mare, Romania in 2000 and in Aznalcollar, Spain in 1998 (Jordan, D'Alessandro, 2004) 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 10 lives and injuring 150 persons and contamination of agricultural lands (Jordan et al., 2011). Numerous 'country specific' and regional studies were adopted for hazard (e.g. Sun Hong-fei et al., 2010), impact (e.g. Horvath, Gruiz, 1996; Sommer et al., 2003; Hansen et al., 2008; Ramsey, 2009; Zobrist et al., 2009; González et al., 2011), and risk assessment of mining sites (e.g. Veliciu, Stratulat, 2004; Komnitsas, Modis, 2006; Lim et al., 2008; Broadhurst, Petrie, 2010; Yenilmez et al., 2011). Also, many spatial methods for environmental RA have been developed (e.g. Slowanska, 1997; deLemos et al., 2009; Sollitto et al., 2010; Pizzol et al., 2011).

The Mine Waste Directive (2006/21/EC) requiress the risk-based inventory of all mine waste sites in Europe. In order to address the problem a standard risk-based pre-selection protocol has been developed by the EU Commission consisting of 18 simple questions (Q1–Q18) about contamination source, pathway and receptor, for example, if the mine waste contains sulphide minerals (Q2) or heavy metals (Q3) for the contamination source, or if there is a high permeability layer beneath the mine waste site (Q12) for the pathways, and, for the sensitive receptor, if a settlement with >100 inhabitants is located within 1 km of a waste site (Q15).

The complex problem of mining contamination impacts requires methods that should be (1) holistic, i.e. address the problem in its integrated complexity in the total human ecosystem, and (2) direct decision support tools, i.e. environmental decisions can be directly based on their results. The main approaches that meet these criteria are described and compared in this study. In order to evaluate some of the most important decision support methods that were developed and applied to mining contamination a thorough review has been published (Jordan, Abdaal, 2013) that compares the 'holistic' approaches including (1) landscape ecology (LE), (2) industrial ecology (IE), (3) landscape geochemistry (LG), (4) geo-environmental models (GEM), (5) environmental impact assessment (EIA), (6) environmental risk assessment (RA), (7) material flow analysis (MFA), and (8) life cycle assessment (LCA). This study concluded that none of the methods alone can address all of the environmental problems of mining. Methods of LE, IE, LG and GEM put the emphasis on the study of natural systems while EIA, RA, MFA and LCA study more the decision making process within the human socio-economic systems. The common in all of these methods is that they try to bridge the gap between socio-economic and natural sciences in order to support decisions on the management of the environment. Among natural science techniques an integrated use of the LG with MFA seems to be the most efficient for contamination studies of mining. Among socio-economic techniques, asset LCA may provide the broadest and the most 'holistic' framework to bring together EIA, RA and decision analysis, in general. In the European legislative context, the Strategic Environmental Assessment Directive (Directive 2001/42/EC) is the most holistic European directive that integrates many of the different methods considered in this study (Jordan, Abdaal, 2013). In the case of abandoned mines LCA and EIA have no application in making decisions on the necessary site remediation in the lack of mine site operator.

Table 1 provides a comparison of the key parameters of some 11 recognized methods for pre-screening RA of mine waste sites. For the source parameters, size (area and volume), heavy metal content, and the waste type (tailings lagoon or heap) are the most commonly included parameters. While the slope is included in four RA methods (the EU MWD Pre-selection Protocol (Stanley et al., 2011, PRAMS EEA, 2005; HMS-IRC Irish EPA, 2009; Turner et al., 2011), rehabilitation is included only in the EU MWD Pre-selection Protocol (Stanley et al., 2011) and in Di Sante et al, (2009). For the pathway parameters, surface water (lakes and streams), air, groundwater and direct contact are included in most of the RA methods. While distance to the nearest surface water bodies is included in six RA methods only (EU MWD Pre-selection Protocol (Stanley et al., 2011; AIMSS Pioneer Technical Services, 1994; PRAMS EEA, 2005; HMS-IRC Irish EPA, 2009; Turner et al., 2011; Pizzol et al., 2011). Permeability of layers below the waste site is included only in two RA methods (EU MWD Pre-selection Protocol (Stanley et al., 2011), and PRAMS EEA (2005), while erosion/wind transport is included only in Pizzol et al. (2011) (Table 1). For the receptor parameters, human (health, population), ecosystem, groundwater, surface water and toxicity analysis are included in most of the RA methods. It is interesting that vulnerability (sensitivity) is included only in three RA methods (HMS-IRC Irish EPA, 2009; Fan et al., 2010; Pizzol et al., 2011).

The first objective of this study is the evaluation of the EU MWD Pre-selection Protocol (Stanley et al., 2011) by applying it to real-life cases and adopting it to country-specific conditions. The data derived for the implementation of the Protocol such as the distance to the nearest stream or the size of the contamination source mine waste site is compared to those resulted from the 'Pre-screening of problem areas' according to the European Environmental Agency (EEA) Preliminary Risk Assessment Model for Soil contamination in Europe (PRAMS) in order to assess the sensitivity of mine waste site risk assessment in response to various methods. Altogether 145 ore mine waste sites in Hungary were selected for scientific testing and evaluation using the EU MWD Pre-selection Protocol. Questions of the EU MWD Pre-selection Protocol are linked to a GIS system and key parameters such as the topographic slope and distance to the nearest streams, lakes and groundwater bodies, to settlements and the Natura 2000 protected areas were calculated and statistically evaluated in order to adjust the RA models to country-specific conditions in Hungary.

The second objective of this study is the heavy metal contamination risk assessment (RA) for a number of selected quarries in order to study the inert characteristics of the potentially generated mine wastes, in accordance with the EU MWD legislation, using the chemical analysis results of collected samples. Altogether 30 waste sites (including both abandoned mines and active quarries) were selected for scientific testing using the Pre-selection Protocol.

Table 1 – Comparing the key parameters of some recognized pre-screening RA methods for mine waste sites.

FACTORS	RA methods Key parameters	EU Pre-selection Protocol	HRS, US EPA 1992	US AIMSS, Poineer 1994	EU PRAMS, EEA 2005	HMS, Irish EPA 2009	Di Sante et al., 2009	Braodhurst, 2010	Fan et al., 2010	Turner et al., 2011	Wang et al., 2011	Pizzol et al., 2011	Ranking
SOURCE	Size: Area	X	X	X	X	X			X	X		X	8
	Heavy metals (total)	X		X	X	X		X			X	X	7
	Size: Volume (m³)	X	X	X	X	X						X	6
	Waste type (tailings lagoon or heap)	X		X	X	X		X					5
	Soil		X		X		X		X		X		5
	Slope	X			X	X				X			4
	Mining: Years of activity				X	X						X	3
	Sulphide Minerals	X						X					2
	Chemicals (processing)	X						X					2
	Rehabilitation	X					X						2
PATHWAY	Surface water (lakes and streams)	X	X	X	X	X	X			X	X	X	9
	Air	X	X	X		X	X		X			X	7
	Groundwater	X	X	X		X			X			X	6
	Direct contact	X		X	X	X	X		X				6
	Distance to surface water bodies	X		X	X	X				X		X	6
	Distance to groundwater bodies	X		X		X							3
	Distance to the nearest settlements	X			X	X							3
	Distance to Natura 2000 sites	X			X							X	3
	Permeability of layers beneath the site	X			X								2
	Erosion/wind transport											X	1
RECEPTOR	Human (health, population)	X	X	X	X	X	X	X	X			X	9
	Ecosystem (protected)	X	X	X	X	X	X				X	X	8
	Groundwater	X	X	X			X		X			X	6
	Toxicity analysis		X	X		X	X	X	X				6
	Surface water	X					X			X	X	X	5
	Land use	X			X				X			X	4
	Vulnerability					X			X			X	3

Ninety three field samples were collected from the waste sites including andesite, rhyolite, coal (lignite and black coals), peat, alginite, bauxite, clay and limestone mines. Laboratory analyses of the total toxic element content (aqua regia extraction), the mobile toxic element content (deionized water leaching) were carried out according to the Hungarian national standards (GKM Decree No. 14/2008. IV.3) concerning mining waste management. A detailed geochemical study together with spatial analysis using ArcGIS was performed to derive a geochemically sound contamination RA of the mine waste sites. Key parameters such as heavy metal content and distance to the nearest surface and ground water bodies, or to sensitive receptors such as settlements and protected areas, were calculated and statistically evaluated in order to calibrate the RA methods.

In the third objective of this study, to analyse the linkage between the water quality variables from streams near by the mining waste sites and the landscape metrics of 33 watersheds enclosing those mining sites. The water quality variables Ni, Mn, Cr, Zn and conductivity that represent the total pollution of water in Hungary were investigated and analyzed. The hypothesis is that landscape pattern structure may have an influence on and thus a relationship with contamination transport from the mine sources to the receiving surface waters. The water quality variables were selected on the basis that 1) these point source chemical contamination variables are important in this study, and 2) other point source contamination variables were not measured by the Central Environmental Agency of Hungary, and 3) these are the most complete data series available for the stream water quality monitoring stations in Hungary concerning the studied watersheds.

2. Data and Methods

For the EU MWD Pre-selection Protocol risk assessment (Stanley et al., 2011), waste site data was used such as location of mine waste sites, composition of mine waste including sulphides, toxic metals, and dangerous substances (Q2-Q4) , geometry of the waste heap (height and area) and slope of foundation (Q6-Q10), and other data such as presence of a high permeable layer beneath the waste site (Q12), and if the facility is uncovered and thus the waste is exposed to wind or direct contact (Q13-Q14) (Fig. 1). Information on the mine waste facility 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 in addition to slope data calculated from the Hungarian DEM 50m grid using the ILWIS® 3.7 open source raster GIS software. Census data for Hungary (census 2009) was obtained from the Hungarian Central Statistical Office. Data on protected areas such as Natura 2000 protected areas was available from the Hungarian Central Directorate of Water and Environment (VKKI). Location and status classification of groundwater bodies in Hungary under the Water Framework Directive (WFD) was obtained from VKKI and from EEA website (Waterbase-Groundwater datatests). Land use/land cover data (LULC) maps at 1:100,000 scale were obtained from the European CORINE Land Cover website.

2.1. Contamination Risk Assessment Methods in landscapes

Two major methods of risk assessment of contamination at mining sites are used in this study as described below.

2.1.1. EU MWD Pre-selection Protocol

The EU MWD Pre-selection Protocol (Fig. 1) consists of four sections: 1) Known serious im-

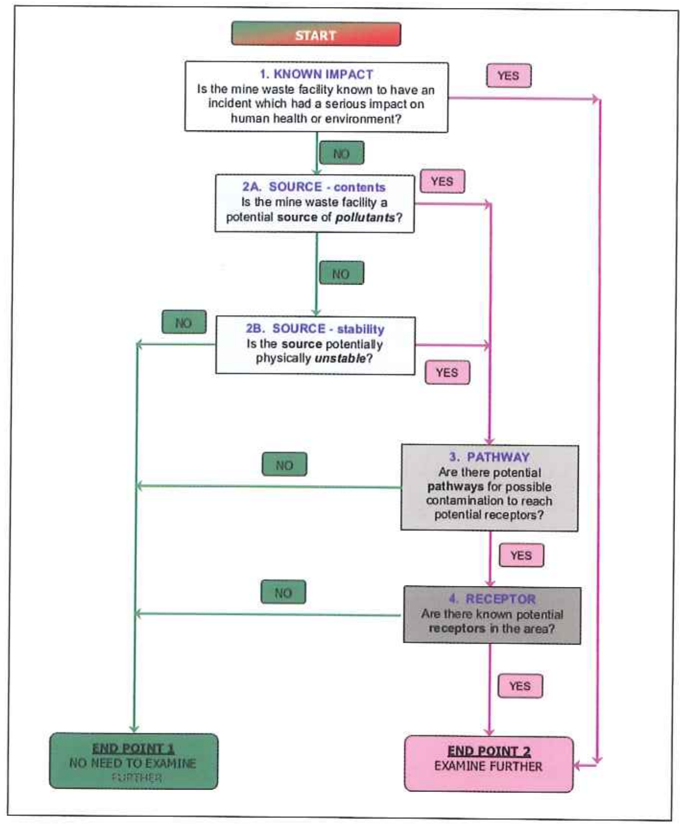

Figure 1 – The EU MWD Pre-selection Protocol flowchart (Stanley et al., 2011).

pact, 2) Source, 3) Pathways and 4) Receptors. Section 1 seeks to determine if a site has had a documented incident with a serious impact on human health or the environment (Q1). For example, 850 million cubic meters of toxic red mud spilled through the failed dam of the Ajka alumina depository in Kolontar, Hungary, in 2010, resulting in a serious impact on human health and the agricultural lands (Jordan et al., 2011). The site would directly be assigned to the EXAMINE FURTHER category. Section 2 addresses the chemical composition and physical stability of the mine waste site acting as potential contamination source (source questions Q2-Q10). Three questions address the content of the waste site, if the waste contains sulphide minerals (Q2), heavy metals (Q3) or the mine uses dangerous chemicals (Q4). This is followed by six questions that address the stability of the facility. Q5 asks if the type of the facility is either a tailings lagoon or a waste heap. If the site is a tailings lagoon there are two further questions: if the area of tailings lagoon site is >10.000 m^2 (Q6) and the height is >4 m (Q7). If the site is a waste rock heap there are three further questions: if the waste heap area is >10.000 m^2 (Q7), the height is >20 m (Q8) and the topographic slope under the waste heap site is >=5° (Q10). Section 3 considers the potential pathways by which receptors could be impacted by the mine waste source. Four pathway questions cover the four potential contamination transport routes: if a surface water course is within 1 km of a mine waste site (Q11), if there is a high permeability layer beneath the mine waste site (groundwater pathway; Q12), if the waste material is exposed to air (Q13), and if the waste site is uncovered allowing direct contact (Q14).

Section 4 seeks to identify four major sensitive human and ecosystem receptors. Question Q15 examines if a human settlement with >100 inhabitants is located within 1 km of a waste site, Q16 asks if the waste site is located within 1 km distance of groundwater body in 'poor status', Q17 asks if a Natura 2000 site is located within 1 km distance of a waste site, and Q18 inquires if a waste site is within 1 km distance of an agricultural area. The possible responses to each question are YES, NO or UNKNOWN. The YES answer means the presence of a risk factor, such as a toxic metal in the waste, the potential of transport by groundwater or a

nearby located settlement as a receptor. The UNKNOWN response indicates uncertainty in information and uncertainty implies risk. Thus, UNKNOWN follows the same route as the YES response pointing towards further examination, according to the precautionary principle.

If there is at least one YES or UNKNOWN response in each of the three Sections of source, pathway and receptor then the assessor is directed to the EXAMINE FURTHER endpoint. This case means that there possibly exists a contamination source, at least one possible pathway and a sensitive receptor. If the answers to all questions in at least one Section are NO then the source-pathway-receptor chain is broken, no risk exists for the site, and the assessor is directed to NO NEED TO EXAMINE FURTHER endpoint (Fig. 1).

The EU MWD Pre-selection Protocol sets a 1 km threshold for the distance to the nearest surface water course (Q11), settlement (Q15), groundwater body (Q16), Natura 2000 site (Q17), and agricultural area (Q18). The Protocol also sets 100 inhabitants as a limiting value for the nearest settlement (Q15) and a 5 degree threshold for facility stability in question 10 (Q10). The Protocol thresholds are based on the Irish regulation for the operation of ponds with respect to quarries (Safe Quarry, 2008). In the present study a detailed statistical analysis is carried using the 145 ore mines test cases and the original 1 km threshold value is modified to the values identified as natural breaks in the distance histograms (see Fig. 6). The lowermost break in the histogram identifies sites that are located within the closest distance and therefore these have the highest risk. In this way, the distance threshold is adopted to, for example, the settlement and stream course density conditions in Hungary. Also, the median of the calculated 145 distances is calculated for all threshold limited parameters allowing a threshold estimation representing a 50% probability of the site falling within the risk limiting distance (Median-based threshold). The same calculations are performed for the census and slope data. Therefore, each Member State can choose a different threshold which can meet their particular topographic and census conditions.

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 digitalized using ArcINFO® 10, on the basis of the physical and geochemical characteristics of the uppermost rock units. Three groups have been distinguished and aggregated (Fig. 2). 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). Polygons of the mine waste sites derived from the CORINE land cover 1:50,000 map (2000) are overlaid by the most recent Google Earth® aerial photographs, 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. 3).

The topographic slope data calculated from the Hungarian National DDM 50m grid using ArcGIS 10® software. Census data of Hungary for 2009 obtained 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, Directive 2006/118/EC) were obtained from the Hungarian Central Directorate of Water and Environment (VKKI) and from EEA website (Waterbase-Groundwater datasets). Land use/land cover data maps at 1:100,000 scale used the CORINE 2006 database. Altogether 145 ore mine waste sites (Fig. 4) are tested using the EU MWD

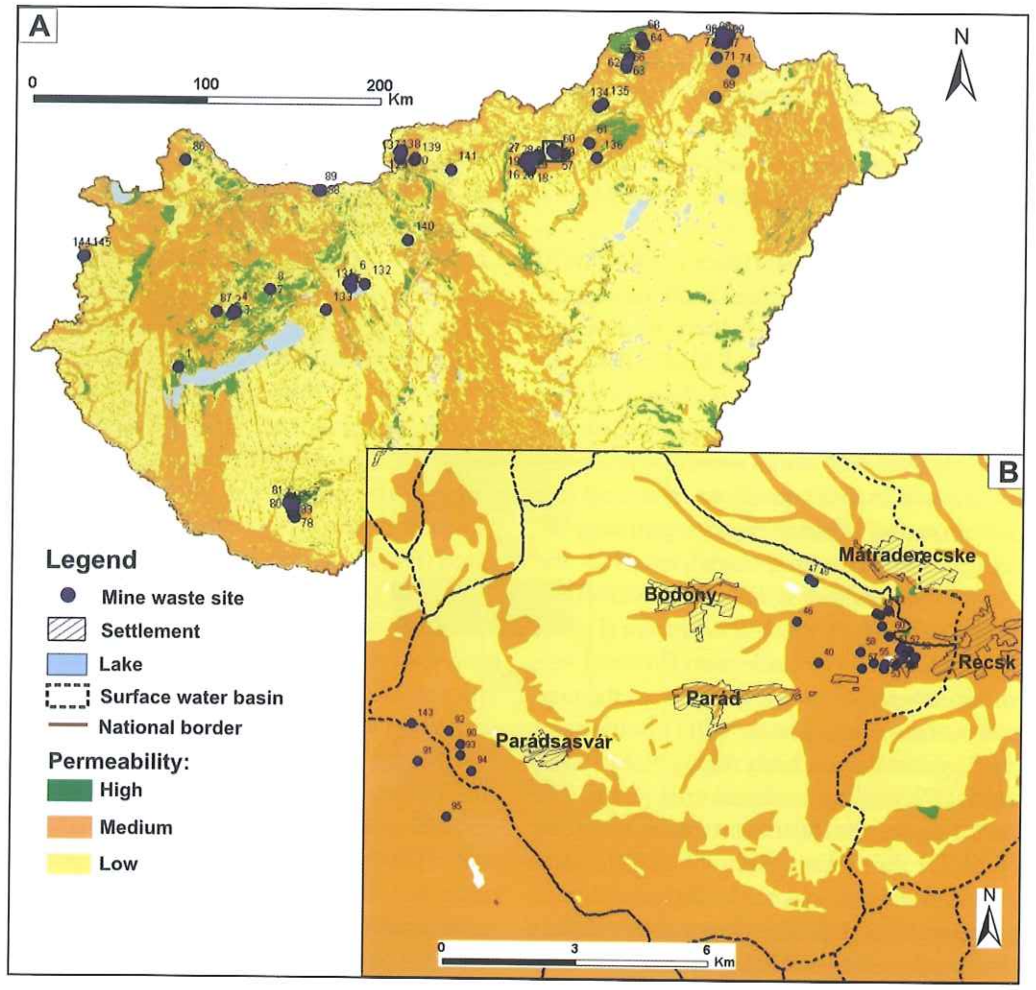

Figure 2 – Surface permeability map developed to answer question Q12 of the EU Pre-selection Protocol. A. Surface permeability map for Hungary. Solid box shows location of Figure 2B. B. An example for the Recsk Mining Area in the Parádi-Tarna Creek catchment.

Pre-selection Protocol as a case study from Hungary. Then, by running the protocol, the number of YES, NO and UNKNOWN responses are registered for each site.

2.1.2. EEA Preliminary Risk Assessment Model

The Pre-screening procedure (Tier 0) of the EEA Preliminary Risk Assessment Model (PRAMS, EEA, 2005), another international standard was applied to the 145 test sites. The results of the EU MWD Pre-selection Protocol are compared to those of PRAMS in order to provide a further means of parameter sensitivity analysis. In the PRAMS model the potentially contaminated areas of EU interest are preliminarily identified according to two sets of criteria as follows. The "A" criteria address sites with available knowledge on impact extent and the "B" criteria inquire about sites where this knowledge is not sufficiently available and surrogate information is used. "A" criteria include a YES/NO answer to one or more EU relevant policy questions. While "B" criteria include a set of questions on size in terms of surface area, waste or stored toxic materials volumes, and complexity in terms of number of sites, requiring simple information more likely to be readily available in data archives.

Figure 3 – Polygons of the mine waste sites defined from the CORINE land cover map (CLC 2000) overlaid by Google Earth® aerial photographs (2010-2011) to answer EU Pre-selection Protocol questions Q13-14 on the air and direct contact pathways related to the cover of tailings. Example shows the Ajka alumina plant tailings lagoon. Note that cells 9 and 10 are not covered while cells 1-8 have been rehabilitated with soil and plant cover

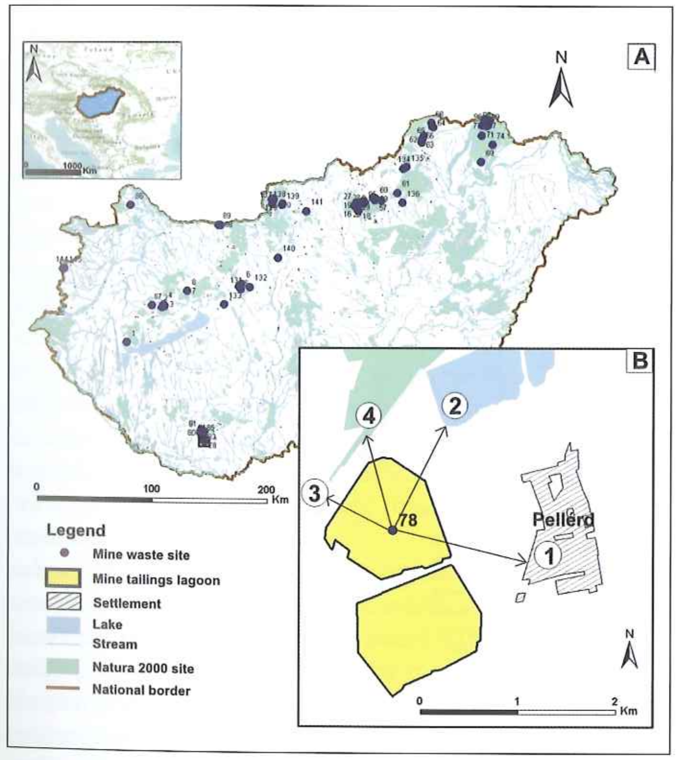

Figure 4 – A. Mine waste sites in Hungary considered in this study. Solid box shows location of Figure 4B. B. Distance measurement from the waste sites (polygon centroid) to the nearest settlement (1), surface water lake (2), stream (3) and to the nearest Natura 2000 protected area (4).

2.1.3. Risk Assessment sensitivity analysis: numerical comparison of methods

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. The distance from mine waste sites to the nearest receptors such as human settlements (Q15) was measured using proximity analysis tools (Point Distance and Generate Near Table) in ArcINFO® 10 (Fig. 4). Statistical analyses were carried out using STATGRAPHICS Centurion XV.II® software, 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). The data derived for the implementation of the EU MWD Pre-selection Protocol such as the distance to the nearest stream or the size of the contamination source mine waste site is compared to those resulted from the

PRAMS model in order to assess the sensitivity of mine waste site risk assessment in response to various methods.

2.2. Waste rock geochemical characterization and risk mapping

The EU MWD Pre-selection Protocol was applied on 30 mine-quarry waste sites in Hungary (Fig. 5) in order to study the geochemical characteristics of the potentially generated mine wastes, in accordance with the EU MWD legislation. Altogether 30 mine waste sites 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. Altogether 93 samples have been collected according to the EuroGeoSurveys Geochemistry Expert Group Sampling Protocol from 30 mine-quarry waste sites in Hungary (Fig.5). 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, Bedrog and Felsoabasár sites and 4 limestone samples from Vác mine site.(Fig. 5, Table 2).

The collected two kilograms of samples were always composed of three sub-samples located at a minimum of 10m distance from each other. Selection of the samples at the site depended 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 subsamples located at a minimum of 10m distance and at any sudden change in the color of waste rock a new sample was collected.

Laboratory analyses of the total toxic element content (aqua regia extraction) and the mobile toxic element content (deionized water leaching) were carried out with ICP-OES according to the Hungarian standards (GKM Decree No. 14/2008. (IV.3) concerning mining waste management. Altogether 70 samples were analyzed for different forms of sulfur (sulfuric acid potential) using HORIBA EMIA element analysis method. Calibration for this method is made according to the Hungarian AVKL-01-SPO-01-03 description procedure.

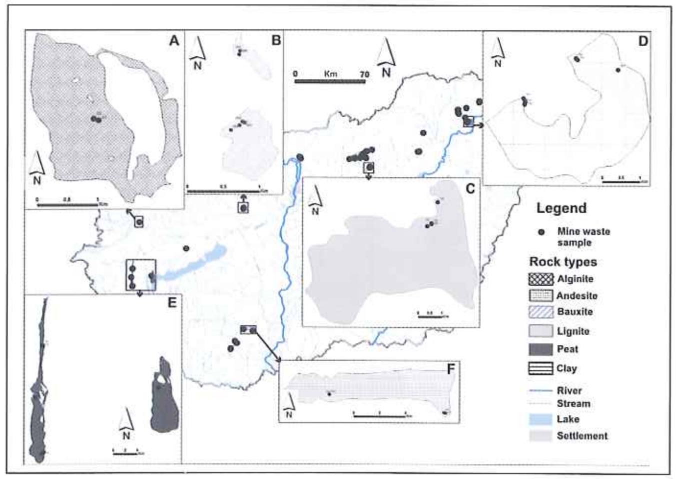

Figure 5 – Examples of rock formations (as polygons) and locations of field sampling from abandoned mines and active quarries in Hungary. A. Pula Alginite Formation, B. Gánt Bauxite Formation, C. Lignite Formation at Visonta, D. Andesite Formation in the Tokaj Mts., E. Peat formation at Pölöske, F. Clay Formation at Máza

Table 2 – Showing 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		14	37	B
Limestone		1	4	A

In this way, the sampled rock types could be characterized for toxic element content that can be extended to the whole spatial extent (polygon) of the rock type in the geological map. Thus, not only the mine waste sites as point sources can be used for the contamination risk assessment but the whole area occupied by the mined rock type acts as a spatially extent contamination source. This data, the geochemically characterised rock formation polygon, is then input into the risk assessment model. Accordingly, two types of risk assessment were then carried out: (1) a point source assessment for each mine site as shown above and (2) a spatially extended source assessment for the mined rock type polygons.

For the point source assessment for each mine site, locations of the mine waste sites derived from the CORINE land cover 1:50,000 map (CLC 2000) were overlaid by the most recent Google Earth® aerial photographs, in order to identify if the material within the mine waste sites is exposed to wind or not (Q13) or covered or not (Q14). The median slope value for each rock formation polygon (in degrees) was calculated from all pixels inside the polygon using Spatial Analysis tool in ArcGIS 10®. The distance from each rock formation polygon (as centroid point) to the nearest pathways (such surface water courses (Q11) and receptors (such as human settlements (Q15) and Natura 2000 protected areas (Q17) was measured using Proximity Analysis tools (Point Distance and Generate Near Table) in ArcINFO® 10.

3. Results

3.1. EU MWD Pre-selection Protocol Risk assesment using the EU thresholds

The contamination RA according to the EU MWD Pre-selection Protocol is carried out in two runs. The first run uses the original EU thresholds (slope ≤ 5°, 1 km distance and number of people in the nearest settlement ≥ 100, Table 3). The second run uses local thresholds defined by (1) the highest natural break in the parameter (slope (Q10) and the lowest natural break for the nearest distance (Q11, Q15-18)) cumulative distribution curves (corresponding to local minima in the frequency histog-

Table 3 – Summary statistics of the EU MWD Pre-selection Protocol responses of questions Q1-18, showing the number of YES and NO responses based on the EU MWD Pre-selection Protocol thresholds, and the local median-based thresholds and on the local highest group-based thresholds. The number (U) and percentage of certain to uncertain (U%) responses for each question are based on the number of UNKNOWN responses. Bold indicates questions and statistics depending on the thresholds.

Pre-selection Protocol		Number of Sites	EU thresholds		Local thresholds (Median-based)		Local thresholds (Highest group)		U	U %
			YES	NO	YES	NO	YES	NO		
Impact	Q1	145	19	126	19	126	19	126	0	0
Source	Q2	145	101	40	101	40	101	40	4	3
	Q3	145	126	15	126	15	126	15	4	3
	Q4	145	7	138	7	138	7	138	0	0
	Q5	145	9	136	9	136	9	136	0	0
	Q6	9	9	0	9	0	9	0	0	0
	Q7	9	4	2	4	2	4	2	3	33
	Q8	136	34	92	34	92	34	92	10	7
	Q9	136	9	115	9	115	9	115	12	9
	Q10	136	**110**	26	**74**	62	**2**	134	0	0
Pathway	**Q11**	145	**64**	81	**73**	72	**144**	1	0	0
	Q12	145	120	25	120	25	120	25	0	0
	Q13	145	17	128	17	128	17	128	0	0
	Q14	145	17	128	17	128	17	128	0	0
Receptor	**Q15**	145	**45**	100	**73**	72	**141**	4	0	0
	Q16	145	**28**	117	**73**	72	**142**	3	0	0
	Q17	145	**131**	14	**112**	33	**142**	3	0	0
	Q18	145	**84**	61	**73**	72	**142**	3	0	0

ram; see Fig. 6) (Local threshold), and by (2) the median value of these parameters (Median-based threshold, Table 4). The highest break value threshold represents the precautionary principle and tries to include the largest number of sites for further examination while adjusting to the local physiographic conditions (Hungary in this study). The Median-based threshold takes a neutral position by giving a 50% chance of relative risk. This test results altogether in three final selections of sites according to the three different thresholds (EU threshold, Local threshold and Median-based threshold).

The YES, NO and UNKNOWN responses of the EU MWD Pre-selection Protocol (Fig. 1) are registered and calculated for each question in Table 3. Out of 145 mine waste sites, only 19 sites have a documented incident (Q1), and among these is the toxic red mud spilled through the failed dam of the Ajka alumina depository in Kolontár, Hungary, in 2010, killing 10 persons, injuring more than 150 and polluting agricultural land areas. These 19 sites are immediately directed to further examination in the EU MWD Pre-selection Protocol. In Q2, 101 sites with YES responses were producing waste with sulphide minerals, 40 sites have

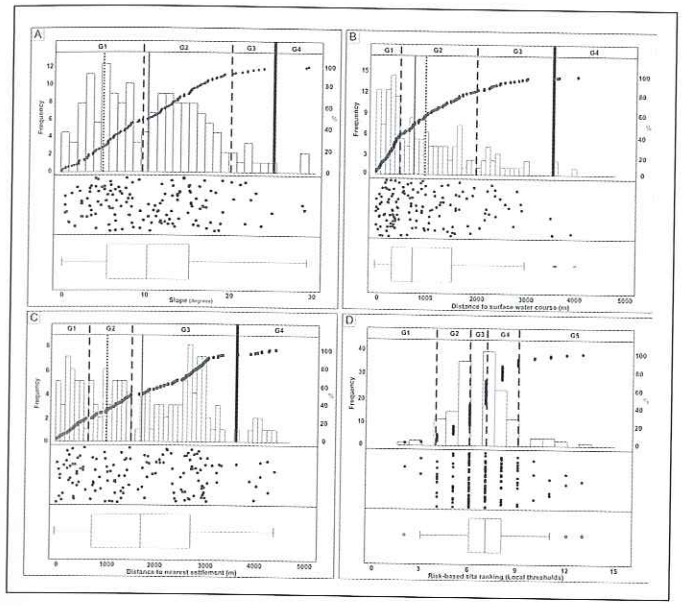

Figure 6 – Distribution analyses for the EU MWD Pre-selection Protocol parameters with histograms, scatterplots, box-whisker and cumulative probability plots. Vertical lines show sub-groups (G1, G2,...) identified by the natural-breaks found in the cumulative probability plots, corresponding to local minima in the frequency histograms. Dotted line shows the EU MWD Pre-selection Protocol threshold, dashed line shows the median, thin solid line shows the median in all sites and thick solid line indicates the highest group boundary, both used for defining thresholds for the questions in the protocol. See text for details. A. Distribution analysis for slope (question Q10). B. Distribution analysis for distance to the nearest surface water course (question Q11). C. Distribution analysis for distance to the nearest settlement (question Q15). D. Distribution analysis for the total site ranking classes based on the number of YES responses and using median-based local threshold.

NO responses, and the other 4 sites (3% of the studied sites) with UNKNOWN response. While in Q3, 126 sites were producing minerals with toxic heavy metals, 15 sites have NO responses, and 4 sites (3% of the total number of sites) have UNKNOWN response. In Q4, seven sites with YES responses have documented use of dangerous chemicals for the mineral processing, the other 138 sites have NO responses. In Q5, nine sites are tailings lagoon sites in Hungary and 136 sites are waste heaps. Still, the tailings lagoons represent a higher risk due to the fluid nature of the stored material and to the large size of these facilities. In Q6, the area of each of the 9 tailings lagoons is greater than the 10,000 m² threshold. In Q7, only four tailings lagoons with YES responses are >4 m in height of the waste site, while two sites with NO responses are <4 m and the other three sites (33% of the 9 tailings lagoons) have UNKNOWN responses. In Q8, 34 waste heap sites with YES responses are greater than 10,000 m² in surface area and 10 waste heaps area extent (7% of the 136 waste heaps) is unknown. It is interesting to have lack of information and thus uncertainty in the simple engineering properties of abandoned mine waste facilities. One would expect that mine archives of former active mines shall contain readily this information. In Q9, nine waste heap sites are >20 m in height and 12 sites (9%) have unknown heights. The height of the waste rock heap is hard to determine due to the irregular geometry of the rock mass over a sloping terrain. The slope of the foundation upon which the waste heap rests is of concern with respect to stability.

The greater the slope angle of the foundation 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, 110 waste heap sites with YES responses are greater than or equal 1:12 (5°) in slope and 26 sites with NO responses are less than 5° (Q10). This shows that most of the sites are located in hilly areas. It is interesting that the failed Ajka

Table 4 – Class boundaries of the EU MWD Pre-selection Protocol parameters based on the natural-breaks found in the parameter distribution plots (see Fig. 6). Class boundaries are used to define thresholds adapted to local conditions (in Hungary in this case). The highest class boundary and the median of all sites value local thresholds are discussed in this study. Number of sites falling within each natural class helps guiding the selection of the proper threshold. See text for details.

Question	Class boundaries (local thresholds)	Topographic slope below waste site (degree)	Median of class	Median of all sites (local thresholds)	Number of sites
Q10	>25	25–29	29	**10°**	3
	20–25	20–24	22		8
	9–20	9–19	14		64
	<9	0–9	5		70
		Distance to the nearest surface water course (m)			
Q11	<500	11–481	270	**760**	57
	500–2000	531–1997	1089		66
	2000–3604	2029–3014	2457		19
	>3604	3604–4021	3643		3
		Distance to the nearest settlement (m)			
Q15	<686	0–582	319	**1,722**	33
	686–1478	686–1462	1119		37
	1478–3604	1478–3305	2618		66
	>3604	3604–4367	4083		9
		Distance to the groundwater bodies of 'poor status' (m)			
Q16	0	0	0	**6,044**	25
	14–9541	14–9541	5687		85
	9541–11692	9545–11055	10005		28
	>11692	11692–23771	13635		7
		Distance to the nearest Natura2000 sites (m)			
Q17	0	0	0	**470**	91
	13–1299	13–1299	470		42
	1480–1725	1480–1725	1612		6
	>2294	2294–6526	2732		6
		Distance to the nearest agricultural areas (m)			
Q18	<1064	0–861	167	**612**	81
	1064–2585	1064–2272	1515		28
	2585–3688	2585–3402	3128		31
	>3688	3688–3976	3956		4

red mud tailings facility is in fact located in a flat area below the slope threshold value.

The use of the surface permeability map (Fig. 2) developed to generate answers for Q12, resulted in 120 sites with YES responses (three sites underlain by high permeable layers and 117 sites underlain by medium permeable layers), while 25 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, 17 sites are exposed to the wind and 128 sites are not due to engineered or natural re-vegetation. While in Q14, 17 sites are uncovered and 128 sites are covered with water, vegetation, soil and forest (Fig. 3). For example cells 9, 10 and 10a in the Ajka alumina tailings lagoon are not covered while cells 1-8 have been rehabilitated with soil and plant cover (Fig. 3). The recent shift from wet to dry deposition decreased the risk of catastrophic spill but it has increased dusting as confirmed by field observation.

For Q11, 64 sites are within 1 km distance to the nearest surface water bodies (streams and lakes). In Q15, 45 mine waste sites are within 1 km distance to nearest human settlements with >100 people. 28 sites are within 1 km distance to the groundwater bodies of less than good status (poor status). For Q17, 131 mine waste sites are within 1 km to the national protected 'Natura 2000' sites (91 waste sites are completely inside the Natura 2000' sites), and 14 sites are within distance >1 km. Moreover, in Q18, 84 sites are within 1 km distance to the agricultural areas including arable lands, pastures, heterogeneous and permanent crops.

3.2. EU MWD Pre-selection Protocol Risk assessment using the local thresholds

Distribution analysis identified various sub-groups in the studied parameter thresholds (topographic slope, distance and census data) (Table 4, Fig. 6). For example, in Q10 (Fig. 6A), 3 sites have a topographic slope greater than 25°, 8 sites with slope 20–25°, 64 sites with slope 9–20° and 70 sites with slope less than 9°. This result suggests the 9° slope as a natural threshold reflecting the local (Hungarian) conditions, instead of the original 5° slope threshold. Also, there are 11 (8+3) sites located on very steep slopes above 20° which may single out these sites for specific attention in terms of slope movement and facility stability. According to Figure 6B (Q11), 57 sites are within distance less than 500 m to the nearest surface water bodies, 66 sites are within distance 531–1,997 m, 19 sites within 2,029–3,014 m and three sites are within distance 3,014 – 4,021 m. This shows that almost half of the mine waste sites are significantly (at the 90% confidence) closer (≤500 m) to receiving streams than the other sites, highlighting these sites for more detailed surface transport modeling if identified for 'further examination' in the EU MWD Pre-selection Protocol. Moreover, the second group of 531–1,997 m distance contains the original 1 km threshold and thus the 2 km (1,997 m) threshold may better reflect the local topographic conditions for this question. In Q15 (Fig. 6C), 33 sites with population more than 820 inhabitants are within distance less than 680 m to the nearest settlement, indicating that these sites require prime attention if settlement protection is the concern. It is interesting that 25 sites lie directly above the groundwater bodies with 'poor status' (Q16) and 91 sites are located inside the protected Natura 2000 sites (Q17). The amazing high portion (63%) of mine waste sites lying directly in protected ecosystems calls for immediate special attention if landscape protection is a priority. While in Q18, 81 sites are within distance less than or equal to 861m to the nearest agricultural areas.

The neutral local thresholds based on median values (Median-based threshold; Table 4), selecting half of the sites for YES response, yields 10° for the slope below the waste site (Q10), 760 m for the distance to surface water bodies (Q11) and 1,722 m for the distance to settlements with 820 inhabitants (median-based) (Q15). This is all consistent with the fact that mining areas lie in forested hilly areas with high density drainage network and sparse population: sites are located on steep 10°>>5° slopes, close (760m<1 km) to abundant stream network and with settlements remote (1,722m>>1 km) from mine sites. The settlement population cut off value is much higher than the original EU value (820>>100 inhabitants), since people live in villages in Hungary unlike farm areas in Ireland. This calls for stringent catastrophe response in case of civil protection and rescue. The 6,044 m distance to the nearest groundwater bodies with 'poor status' (Q16) is however reassuring, unlike the median distances of 470 m to Natura 2000 sites (Q17) and 612 m agricultural areas (Q18).

Distribution analysis was performed on the population census data of Hungary (census 2009), to develop a population threshold number for Q15 of the EU MWD Pre-selection Protocol, resulting in 53 classes ranging from <45 to >45,000 persons bounding the two extreme groups. The analysis indicates that 1,670 of the total 3,157 settlements with less than or equal to 820 persons are representing 53% of the total number of settlements in Hungary. Therefore this number, 820 persons, is a reasonably representative choice as a local threshold (Median-based) for the population in Q15. By running the EU MWD Pre-selection Protocol using these local thresholds (Median-based), the YES, NO and UNKNOWN responses are compared to those of EU thresholds as depicted in Table 5. Table 5 shows that the number of waste sites with YES responses of the EU MWD Pre-selection Protocol varies from using the EU thresholds to local thresholds (Median-based). For example, in Q10 on underlying terrain slope, sites with YES responses are decreased from 110 (EU thresholds) to 74 (local thresholds (Median-based) and to two sites with the highest threshold group, whilst in Q11 on the distance to the nearest surface water course, the sites with YES responses are increased from 64 (EU thresholds) to 73 (Median-based local threshold) and 144 (the highest group).

Table 5 – Summary statistics of 'A' and 'B' criteria of the Pre-screening of problem areas of the EEA PRAMS (Tier 0) model, showing the number of YES, NO and UNKNOWN (U) responses and the percentage of uncertain to certain (U %) responses for each question.

PRAMS (Tier 0) Questions		Number of sites	YES	NO	U	U %
Are natural ecosystems of European concern affected?	A1	145	19	126	0	0
Is contamination impact on surface water such that reaching the target set according to the EU Water Framework Directive prevented?	A2	145	0	19	126	87
Is contamination in the "groundwater body" (working unit of the Groundwater Directive) such that "good status" (as defined in the Groundwater Directive) cannot be reached?	A3	145	122	23	0	0
Is safety of food products brought on EU markets (exported outside the area) affected?	A4	145	0	145	0	0
Is the contamination, because of impacts on human and/or environmental health, leading to use restrictions blocking* regional social and economical development (as supported by EU structural funds)?	A5	145	0	145	0	0
May the area, upon meeting at least one of the A criteria, and according to your expert judgment, be classified as a problem area of EU interest?	A6	145	19	126	0	0
Dimension of potentially affected problem area	B1	Known data				
Single site: Size of contaminated or suspected contaminated site (Surface (ha) and Waste volume (m^3)	B2					
Complexity of problem area (contaminated or suspected contaminated multiple sites/multiple ownerships)	B3					
May the area, according to your expert judgment and upon checking B criteria, be classified as a problem area of EU interest?	B4	145	88	57	0	0

The local threshold of the highest distance group boundary (Table 6) represents the worst case scenario by selecting the possible largest number of sites for YES response and therefore for further examination based on the reasonable level of risk, depicted by solid lines in Fig. 6A, B and C. Thus, this threshold selection follows the precautionary principle.

In summary, after the existing pre-screening risk assessment of the mine waste sites in Hungary, 127 mine waste sites are directed to EXAMINE FURTHER based on the EU thresholds, 18 sites with no risk (these sites have no pathway). While, 129 sites are directed to EXAMINE FURTHER based on the local thresholds (Median-based), 16 mine waste sites with no risk (these sites have no pathway). In the case of using the local threshold (lowest group boundary) (Table 4) in Q10 (5°), Q11 (270 m), Q15 (319 m), Q16 (0 m), Q17 (0 m) and Q18 (167 m), 118 sites are directed to EXAMINE FURTHER and 27 sites have no risk (19 sites with no Pathway and 8 sites with no Receptor). While by using the local threshold (highest group boundary) (Table 4) in Q10 (29°), Q11 (3,643 m), Q15 (4,083 m), Q16 (13,635 m), Q17 (2,732 m) and Q18 (3,956 m), all the 145 mine waste sites are directed to EXAMINE FURTHER. It is obvious that this threshold selection represents the worst case scenario and follows the precautionary principle.

3.3. Pre-screening (Tier 0) EEA PRAMS Risk Assessment Model

Table 5 illustrates the summary statistics of YES, NO and UNKNOWN responses of 'A' and 'B' criteria of the pre-screening of problem areas, according to the EEA PRAMS model. In question A1, 19 mine waste sites have YES responses with natural ecosystems of EU concern affected. In A2, 19 sites have NO responses with contamination impact on surface water course which is not prevented according to the EU Water Frame Directive (WFD), while 126 sites with UNKNOWN responses that represent 87% of the total 145 sites. This shows that there is little harmonization among EU directives (MWD and WFD) and there are no linked environmental databases yet. In A3, the overwhelming majority of sites (122 sites) has YES responses and have contamination in the groundwater body, so 'good status', as defined by the Groundwater (Directive 2006/118/EC), cannot be reached. In A4, none of the 145 mine waste sites have proven effect on food products brought on EU markets. According to question A5, there are no waste sites with contamination impacts on human and/or environmental health leading to use restrictions blocking regional social and economic

Table 6 – Site ranking classification based on the number of YES responses of the EU MWD Pre-selection Protocol using the original EU thresholds and the local median-based thresholds with risk classes, according to Fig. 6D. The number of waste sites in each class is also shown.

Class	EU thresholds	Number of sites	local thresholds (Median-based)	Number of sites
5	3–4	13	2–3	3
4	5	41	4–5	25
3	6–7	48	6	35
2	8–9	28	7–8	62
1	10–12	15	9–13	20
No Pathway	18		16	
Examine further	127		129	

development. In A6, 19 sites with YES responses are classified as problem areas of EU interest upon meeting at least one of the 'A' criteria questions and 126 sites with NO responses are classified as no problem areas.

Next step is running the EEA PRAMS risk assessment model by checking the 'B' criteria using available known data of the 145 mine waste sites. The necessary archive data is available from the Geological Institute of Hungary such as dimension of the area affected (ha) (question B1), size of the contaminated site (ha), waste or stored toxic materials volume in cubic meters (B2) and complexity of problem area (the number of contaminated or suspected as contaminated multiple sites/multiple ownerships) (B3). In question B4, 88 sites have YES responses and classified as a problem area of EU interest and 57 sites have NO responses and classified as no problem areas. The results of the pre-screening of problem areas by the EEA PRAMS model show that 88 sites are classified as a problem sites for further examination. It is obvious that the number of mine waste sites that classified as a potential problem areas of EU interest and directed to further examination, increased from 19 (in question A6) to 88 sites in question B4 (Table 5). This decision is based on the availability of known data to answer the questions of 'B' criteria.

3.4. Sensitivity and uncertainty analysis of the EU MWD Pre-selection Protocol

Uncertainty is inescapable in the assessment of environmental hazard, exposure and the consequent risks to human health, and it arises at every stage in these assessments (Ramesy, 2009). In this study, uncertainty assessment is limited to the UNKNOWN responses (U) in each question of the EU MWD Pre-selection Protocol due to missing of site specific data. According to number of UNKNOWN responses (U) which is ranging in the sites from 0 to 2 U responses and resulting in, 125 sites have no uncertain responses (U=0), 7 sites have one (U=1) and 13 sites have two (U=2) using the EU threshold and local Median-based threshold within the EU Pre-selection Protocol. While in case of using the Pre-screening PRAMS model, 19 sites have no uncertain responses (U=0), 123 sites have one (U=1) and 3 sites have two (U=2). Table 5 indicates that UNKNOWN (U) responses are located only in the source questions in the EU MWD Pre-selection Protocol, ranging from 3% in Q2 (presence of sulphide minerals in waste) and Q3 (toxic element potential in waste) and 7% in Q8 (size of the waste heap) to 33% in Q7 (height of dam wall of the tailings lagoon). Thus, relaxing the source questions, the percentage of uncertain responses (U%) reduces to zero. This is the most unexpected outcome of this study, because high certainty about the source, i.e. the mine waste facilities, was expected due to the assumed mine industry engineering archive documentation. An explanation is that mining flourished in the centrally directed economy period in the 50-80s when waste treatment and environmental issues were not among the priorities leading to poor documentation of related facilities. This is confirmed by the amazing fact that the overwhelming majority of mine sites have no environmental monitoring data whatsoever available.

In order to identify the key parameters and to check the sensitivity (in terms of final selection for further examination) by removal of parameters (questions of the MWD Pre-selection Protocol) from Q2 to Q18, the number of YES responses are recalculated in the other questions for all sites using the EU and local Median-based thresholds. By removal of question Q1 (if site has a known impact with documented incident) there is no change to the total source-pathway-receptor site ranking because the 19 sites with known impact are directed to 'Examine Further' in one step. For the Source Q2 to Q10, by removal of Q2, 125 sites are directed to 'Examine Further' using EU thresholds while 141 sites with 'Examine Further' using local Median-based thresholds. In Q3, 126 sites with 'Examine Further' using EU thresholds while 136 sites with 'Examine Further' using local thresholds. In Q4, Q5, Q6, Q7 and Q9, 126 sites with 'Examine Further' using EU thresholds while 142 sites with 'Examine Further' using local thresholds. In Q8, 125 sites with 'Examine Further' using EU thresholds while 141 sites with 'Examine Further' using local thresholds. In Q10, 120 sites with 'Examine Further' using EU thresholds while 139 sites with 'Examine Further' using local thresholds. For the Pathway Q11 to Q14, by removal of Q11, 127 sites with 'Examine Further' using EU thresholds while 139 sites with 'Examine Further' using local thresholds. In Q12, 69 sites with 'Examine Further' using EU thresholds while 92 sites with 'Examine Further' using local thresholds. In Q13 and Q14, 127 sites with 'Examine Further' using EU thresholds while 142 sites with 'Examine Further' using local thresholds. For the Receptor Q15–Q18, by removal of Q15 and Q16, 127 sites with 'Examine Further' using EU thresholds while 142 sites with 'Examine Further' using local thresholds. In Q17, 74 sites with 'Examine Further' using EU thresholds while 140 sites with 'Examine Further' using local thresholds. In Q18, 124 sites are directed to 'Examine Further' using EU thresholds while 128 sites with 'Examine Further' using local Median-based thresholds.

The key parameters as depicted from above are Q3 (if sites are producing minerals with toxic heavy metals) and Q10 (slope) for source questions, Q12 (presence of higher permeable layer beneath the waste site) for pathway and Q17 (distance to the nearest surface water course) and Q18 (distance to the nearest agricultural areas) for receptor questions. The final selection of sites to further examination will be sensitive to and depends most heavily on these parameters.

For the topographic slope (Q10), by increasing the slope from 1 to 5 degrees (EU threshold) the number of sites decreases from 138 to 111 sites, respectively. At 9° 78 sites will be risky while at 10° 74 sites are in risk position and at 11° 69 sites will be risky. And so on, the number of risky sites is decreasing to 39 at 15°, to 11 sites at 20° and to 3 sites at 25°.

3.5. A preliminary risk-based ranking based on the EU MWD Pre-selection Protocol

Although risk-based site ranking is a subject for Tier 1 RA for the sites selected for further examination by any pre-selection (Tier 0) procedure, a simple preliminary ranking is already enabled by the numeric evaluation of responses to the questions. The number of YES responses using the local Median-based threshold is counted for each site from the possible 0 to 13 (Table 6). Obviously, since a YES response means presence of risk, the higher number of YES responses exist for a site, the higher the risk is. The number of YES responses was also analysed for distribution by the Jenks' natural breaks analysis (Jenks, 1967) within ArcINFO® 10 as shown in Fig. 6D. The resulting five risk classes according to the number of YES responses for each site are: 2-3 YES (class V, 3 sites), 4-5 YES (class IV, 25 sites), 6 YES (class III, 35 sites), 7-8 YES (class II, 62 sites), and 9-13 YES (class I, 20 sites). It is noted that in this exercise only the YES responses are calculated supported by solid data. Although UNKNOWN is identical with a YES response in RA, in this part of the investigation sites with UNKNOWN responses are separately studied and ranked in order to have a clear and transparent picture of site ranking related to responses to the questions.

3.6. Waste geochemical characterization of the selected 30 mine – quarry waste sites

Table 7 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 V has the lowest spread (8.42).

Total concentrations of the heavy metals 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 7). Results show that the median value of Cu (12.3 mg/kg) is less than the Hungarian environmental limit (75 mg/kg) and exceeds the median of the EU FOREGS Atlas (12 mg/kg). In case of central tendency expressed by the Median, the analyzed total heavy metal concentrations are in the descending order of 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 relative variability (spread) expressed by IQR/Med (Inter-quartile range/Median), the total heavy metal concentrations follow the order: Ni>As>Cr>V>Pb>Co>Cd>Zn>Cu. It is obvious that Ni has the highest variability (5.11) and Cu has the lowest (1.11). In case of the $S_{sulphide}$, the median (0.02%) is less than the Hungarian environmental limit (0.1%) and $S_{sulphide}$ has a range from 0.003% to 3.82%.

Table 7 – 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
	Max	220	14.1	255	2340	239	21.3	2560	886		2270
	Med	6	0.145	7	22	12	0.62	14	15		48
	Mean	9.88	0.28	8.91	32.6	16.4	0.94	30.7	23.9		60.9

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 mine waste rock samples. Then the median value of these mobility percentages was calculated for each rock type (Fig. 7). Results showed 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. 7).

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

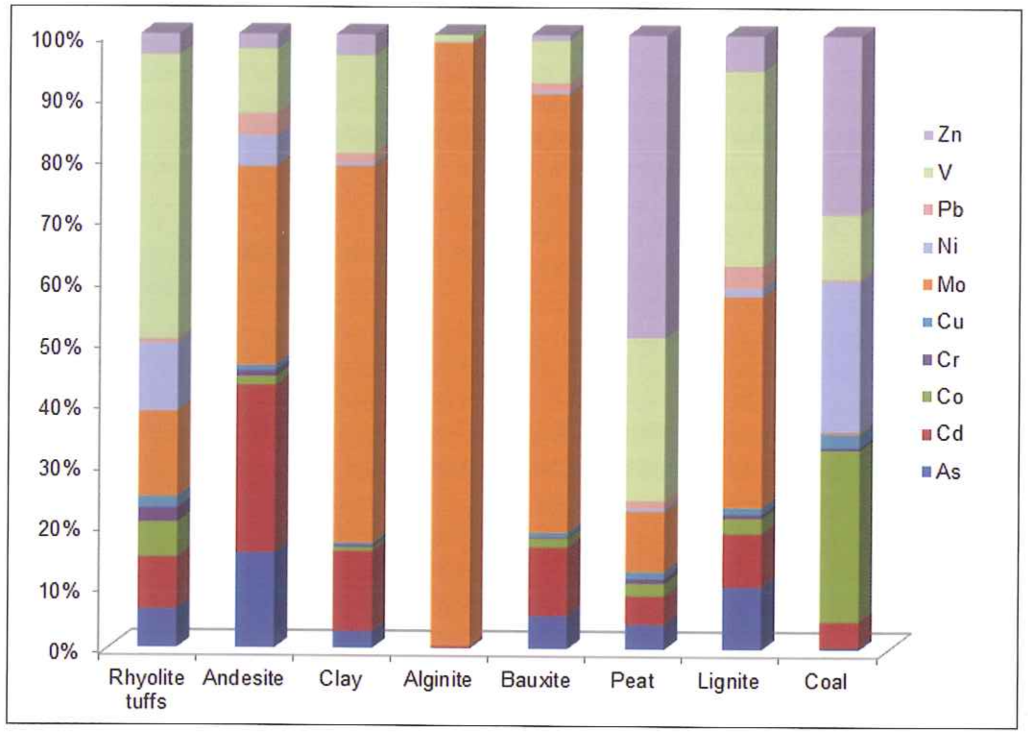

Figure 7 – Distribution of the relative mobility (%) of heavy metals in the various sampled rock formations.

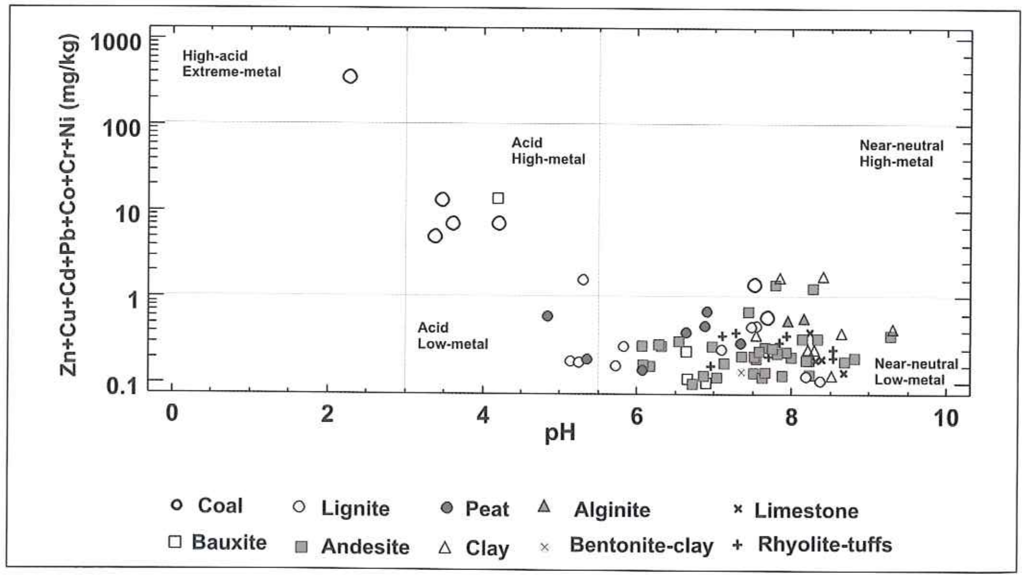

Figure 8 – Ficklin Diagram showing the sum of heavy metals Zn, Cr, Cd, Pb, Co and Ni plotted against pH in the deionized water leaching (DW). Note that acid generation potential ($\mathrm{pH}<5.5$) is for coal, lignite and peat rocks, in addition to a bauxite sample. Elevated mobile heavy metal content is associated with coal, andesite and some clay and a bauxite samples. (See text for details.)

andesites and coals are distinct from the non-mineralised rocks as shown by the Ficklin Diagram (Fig. 8).

For the deionized water leaching, the Ficklin Diagram (Fig. 8) showed that acid generation potential ($\mathrm{pH}<5.5$) 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 sample.

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, and C: probably not inert, has to be examined (Table 2). According to the geochemical analysis results in this study, coal (black coal and lignite) and peat samples are not inert and are 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 for the minimum, median and mean values than the Hungarian standards. While maximum values of As is even higher than the national environmental standard. 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.

3.7. Linkage between heavy metal contamination RA and the landscape metrics

Landscape pattern has a big influence on the sediment delivery ratio from catchments (Jordan et al., 2005; Szilassi et al., 2006). The landscape characteristics (mainly land use and land cover) of catchments have a relevant impact on the surface and subsurface water quality (Xia et al., 2012). To illustrate the role of the landscape pattern on the water quality, for example, the forest patches at the river banks (thick but long linear land use patches parallel with the streams and rivers) would be strong barriers of the sediment and contamination transport. But if there is no any riparian forest near the river or stream, and the linear land cover/land use units without vegetation cover (such as arable land parcels in Spring or Autumn) are dominant, this kind of landscape pattern has an important role on the increasing level of the sediment and contamination transport processes. In this case the long arable land parcels in the slope direction towards the river or stream charnels, can be defined from the environmental risk assessment point of view as "pathways" between point or non-point contamination sources and the receptors such as rivers, streams or settlements.

Several articles showed a strong statistical relationship between the landscape pattern and the water quality in case of the percentage cover of forests and the non-point source pollutions of water such as nitrate and nitrite contamination (e.g. Wu et al., 2012; Xiao, Ji, 2007; Uuemaa et al., 2005, 2013). This is the reason for why, beside the landscape metric parameters, the percentage of the main land cover classes (such as artificial surfaces (CLC1), agricultural areas (CLC2) and forest and semi-natural areas (CLC3)) was investigated in this study. The hypothesis is that the landscape structure may have an influence on and thus a relationship with contamination transport from the mine sources to the receiving surface waters. The water quality variables were selected on the basis that (1) these point source chemical contamination variables are important in this study, (2) other point source contamination variables were not measured by the Central Environmental Agency of Hungary, and (3) these are the most complete data series available for the stream water quality monitoring stations in Hungary concerning the studied watersheds.

The following landscape indices were considered for the watershed containing the selected 33 mine-quarries: Total Number of Patches (NP), Core Area (CA), length of Total Edge (TE) Splitting Index (SPLIT), Division Index (DIVISION), Effective Mesh Size (MESH), Main Patch Size (MPS), Patch Size Standard (PSSD), Deviation Mean Patch Ratio (MPE), Mean Shape Index (MSI) Mean Perimeter Area Ratio (MPAR) and Mean Fractal Dimension Index (MFRACT). The parameters were calculated for each of the 33 mining watersheds based on regional scale (1:100,000) CORINE land cover database from years 2000 and 2006. The percentage area of the main CORINE land cover classes was also calculated, and its role on the water quality was also investigated. The V-late (vector-based landscape analysis tools extension) within ArcGIS 10® and the STATGRAPHICS® software were used for spatial and statistical analyses.

Spearman correlation coefficients were calculated for all landscape metrics and the minimum, median, average and maximum values of stream water quality data pairs of years 2000 and 2006 (Tables 8 and 9). Results show that median dissolved Ni in stream water, minimum and maximum Zn and average stream water conductivity values were significantly correlated with MSI, while median Mn with MESH, average Mn with CA, TE, MPE and MPAR, maximum Mn with artificial surfaces (CLC1), minimum Conductivity with MPS, PSSD, MFRACT, agricultural areas (CLC2) and forest and semi-natural areas (CLC3) showed significant correlations. However, Cr showed no correlation with the landscape indices. For 2006 data, minimum and average Ni values were significantly correlated with DIVISION and SPLIT, minimum Mn with NP, PSSD, CA, TE, MPE and MESH, median Mn with CA and TE, minimum Conductivity with MFRACT, median Conductivity with MPAR, average Conductivity with MSI and MFRACT, maximum Conductivity with MSI, MPAR and MFRACT. In this case all Cr and Zn values showed no significant correlation with the landscape indices. However, no stream water quality variable had significant correlation with Main Patch Size (MPS), Artificial surfaces (CLC1), Agricultural areas (CLC2), and Forest and semi-natural areas (CLC3).

Table 8 – The Spearman's rank correlation between the water quality variables (heavy metals) and the landscape metrics data of 2000. Significant (p<0.05) correlation coefficients are in bold.

	Ni				Mn				Conductivity			
	Min	Med	Avg	Max	Min	Med	Avg	Max	Min	Med	Avg	Max
NP	0.21	0.20	0.40	0.40	0.77	0.40	–0.60	–0.80	0.00	0.40	–0.20	–0.40
CA	–0.63	–0.20	0.40	0.40	0.26	–0.40	**–1.00**	–0.80	0.80	0.40	0.20	–0.40
MPS	–0.95	–0.40	0.20	0.20	–0.26	–0.80	–0.80	–0.40	**1.00**	0.20	0.40	–0.20
PSSD	–0.95	–0.40	0.20	0.20	–0.26	–0.80	–0.80	–0.40	**1.00**	0.20	0.40	–0.20
TE	–0.63	–0.20	0.40	0.40	0.26	–0.40	**–1.00**	–0.80	0.80	0.40	0.20	–0.40
MPE	–0.63	–0.20	0.40	0.40	0.26	–0.40	**–1.00**	–0.80	0.80	0.40	0.20	–0.40
MSI	0.21	**1.00**	–0.80	–0.80	–0.26	0.00	0.20	0.40	–0.40	–0.80	**–1.00**	–0.80
MPAR	0.63	0.20	–0.40	–0.40	–0.26	0.40	**1.00**	0.80	–0.80	–0.40	–0.20	0.40
MFRACT	0.95	0.40	–0.20	–0.20	0.26	0.80	0.80	0.40	**–1.00**	–0.20	–0.40	0.20
DIVISION	0.63	–0.60	0.80	0.80	0.77	0.80	0.20	–0.40	–0.40	0.80	0.60	0.80
SPLIT	0.63	–0.60	0.80	0.80	0.77	0.80	0.20	–0.40	–0.40	0.80	0.60	0.80
MESH	–0.95	0.00	–0.40	–0.40	–0.77	**–1.00**	–0.40	0.20	0.80	–0.40	0.00	–0.40
CLC1%	0.11	0.40	–0.80	–0.80	–0.77	–0.20	0.80	**1.00**	–0.40	–0.80	–0.40	0.00
CLC2%	0.95	0.40	–0.20	–0.20	0.26	0.80	0.80	0.40	**–1.00**	–0.20	–0.40	0.20
CLC3%	–0.95	–0.40	0.20	0.20	–0.26	–0.80	–0.80	–0.40	**1.00**	0.20	0.40	–0.20

It is concluded that the Mean Shape Index (MSI) is the most important 'key' landscape index in 2000 and the Main Fractal Dimension Index (MFRACT) in 2006, in respect to the stream water quality heavy metal contamination in the studied mining watersheds. Based on the above results, in case of the further development of the RA methods, at least these two landscape indices should be taken into consideration.

It is important to note that the minimum Conductivity values are positively correlated (1.00) with the forest and semi-natural areas (CLC3) and negatively correlated (-1.00) with the arable lands (CLC2). This is an unexpected result; however, the higher percentage of arable land shows a positive correlation with the conductivity which represents the total pollution of water. The possible background of this "false" result

Table 9 – The Spearman's rank correlation between the water quality variables (heavy metals) and the landscape metrics of 2006. Significant (p<0.05) correlation coefficients are in bold.

	Ni				Mn				Conductivity			
	Min	Med	Avg	Max	Min	Med	Avg	Max	Min	Med	Avg	Max
NP	–0.42	0.31	0.00	0.25	**–0.88**	–0.72	–0.71	–0.43	0.10	0.02	0.05	–0.02
CA	–0.63	0.12	–0.26	–0.02	**–0.95**	**–0.85**	–0.55	–0.31	0.02	–0.07	–0.07	–0.05
MPS	–0.49	–0.24	–0.19	–0.13	–0.68	–0.52	–0.48	–0.38	0.05	–0.48	–0.21	–0.17
PSSD	–0.68	–0.07	–0.29	–0.14	**–0.85**	–0.73	–0.57	–0.33	0.00	–0.31	–0.24	–0.19
TE	–0.42	0.40	–0.02	0.19	**–0.85**	**–0.74**	–0.67	–0.45	0.17	0.07	0.10	0.10
MPE	–0.54	–0.14	–0.26	–0.07	**–0.85**	–0.66	–0.62	–0.45	–0.02	–0.33	–0.17	–0.10
MSI	–0.66	–0.42	–0.59	–0.42	–0.32	–0.19	0.13	0.54	–0.90	–0.52	**–0.85**	**–0.84**
MPAR	0.08	–0.68	0.19	0.05	0.25	0.54	–0.10	0.07	–0.36	**–0.85**	–0.56	**–0.79**
MFRACT	–0.38	–0.57	–0.45	–0.47	0.17	0.28	0.40	0.69	**–0.93**	–0.67	**–0.93**	**–0.93**
DIVISION	**0.92**	0.50	**0.76**	0.66	0.56	0.54	0.05	–0.24	0.52	0.50	0.71	0.55
SPLIT	**0.92**	0.50	**0.76**	0.66	0.56	0.54	0.05	–0.24	0.52	0.50	0.71	0.55
MESH	–0.68	–0.07	–0.29	–0.14	**–0.85**	–0.73	–0.57	–0.33	0.00	–0.31	–0.24	–0.19
CLC1 %	0.36	0.29	0.17	0.16	0.51	0.30	0.60	0.55	–0.02	0.57	0.26	0.12
CLC2%	0.52	0.50	0.48	0.47	0.44	0.41	0.19	0.24	0.00	0.40	0.24	0.02
CLC3 %	–0.56	–0.48	–0.50	–0.51	–0.44	–0.37	–0.21	–0.21	–0.12	–0.52	–0.38	–0.14

refers to a mistake in the measurement process, because the minimum values are very low and very difficult to be measured accurately.

It is concluded that the Mean Shape Index (MSI) is the most important "key" landscape index in 2000, and the Main Fractal Dimension Index in 2006 (Table 8), from the surface water quality heavy metal contamination point of view. Based on our statistical analyses it can be conclude that in case of the further modification of the RA methods, at least these two landscape indices should be taken into consideration, and integrate into the RA methods. The median Ni, average Mn, average Zn and minimum conductivity variables are significantly correlated the strongest with the landscape indices in 2000. While the minimum and average Ni, the minimum and median Mn, the average and maximum conductivity variables are correlated the strongest with the landscape indices in 2006 (Table 9).

4. Conclusions

The EU MWD Pre-selection Protocol provides a systematic methodology for the pre-screening contamination risk associated with mine waste sites. The method is based on a fundamental understanding of the key factors and parameters controlling the contamination fate along the source-pathway-receptor chain and the chemical behavior of wastes in the mine sites. The preliminary screening RA by the Protocol plays a key role in the initial stage decision-making. The data derived from the Protocol is compared with those resulted from the Pre-screening of problem areas of EEA PRAMS model in order to highlight the sensitivity and differences in each question in each site. It is an unexpected outcome of this study that so high unknown parameters are found for facility engineering conditions, such as the heights and size of the waste dumps. Similarly, the number of YES responses can be accumulated for each site for the source, pathway and receptor questions separately which may indicate the presence of multiple contamination source, multiple pathways or receptors. The results show that the key parameter-questions of the MWD Pre-selection Protocol are Q3, Q10, Q12, Q17 and Q18. Results of the pre-screening EEA PRAMS Model show that the number of waste sites that classified as potential problem areas of EU interest, increased from 19 (in question A6 using archive data) to 88 sites in question B4 when using actual measured parameters. This results from the availability of known data to answer the questions of 'B' risk criteria.

Altogether 30 mine-quarry waste sites were selected for scientific testing of the EU MWD Pre-selection Protocol using laboratory analysis of mine waste rock samples. In addition to detailed geochemical study spatial analysis using ArcGIS was performed to derive a geochemically sound contamination RA of these mine waste sites. Total concentrations of the heavy metals 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 7). Results show that Zn has the highest median (24.6 mg/kg) and Cd has the lowest Median (0.11 mg/kg). Moreover, Mo had the highest relative 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. 7). Results showed that 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 group B which also matches the preliminary expert judgment. Moreover, limestone and clay samples are inert (group A).

In this study the connection between the water quality datas and the landscape pattern has been investigated to improve the RA methods. The linkage between selected water quality variables (Ni, Mn, Cr, Zn and conductivity) in streams nearby the studied 33 mining waste sites and the landscape metrics of watersheds of these mining sites was investigated and analysed. It is concluded that the Mean Shape Index (MSI) is the most important "key" landscape index in 2000, and the Main Fractal Dimension Index in 2006 (Tables 8 and 9), from the surface water quality heavy metal contamination point of view. Based on our statistical analyses it can be concluded that in case of the further modification of the RA methods, at least these two landscape indices should be taken into consideration, and integrated into the RA methods. The median Ni, average Mn, average Zn and minimum conductivity variables are the most significantly correlated with the landscape indices in 2000. While the minimum and average Ni, the minimum and median Mn, the average and maximum conductivity variables are the most significantly correlated with the landscape indices in 2006.

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