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Environmental modelling and spatial landscape analysis for the contamination risk assessment of sensitive areas

Theses of Ph.D. Dissertation

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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. As a first step, some of the most important decision support methods were evaluated and compared that were developed and applied to mining-related contamination. Comparing the 'holistic' approaches, among other, landscape ecology (LE), landscape geochemistry (LG), and environmental impact assessment (EIA), environmental risk assessment (RA), material flow analysis (MFA), and life cycle assessment (LCA) (Jordan and Abdaal 2013). This study, as a part of the PhD thesis, concluded that none of the methods alone can address all of the environmental problems of mining. Among natural science techniques an integrated use of the LG with MFA seems to be the most efficient for contamination studies in mining areas. 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. RA received a specific attention and a detailed comparison of the key parameters of 11 internationally recognized pre-screening RA methods of mine waste sites was developed.

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 (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).

The **second objective** of this study is the heavy metal contamination risk assessment (RA) for a number of selected mines in order to study the inert 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 risk-based EU MWD Pre-selection Protocol (Abdaal 2014). In addition to detailed geochemical study together with spatial analysis using ArcGIS was performed to derive a geochemically sound contamination RA of these mine waste sites.

In the **third objective** of this study, 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. The hypothesis is that 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, 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. Research Methods and Materials

2.1 Contamination risk assessment in landscapes

The EU MWD Pre-selection Protocol (Stanley et al. 2011) was used and applied in order to evaluate that RA method. The EU MWD Pre-selection Protocol considers 18 parameters (Q18) for the selected 145 ore mine waste sites in Hungary formulated as binary Y/N questions along the Source-Pathway-Receptor chain such as if the waste contains sulphide minerals (Q2) or heavy metals (Q3), if a surface water course is within 1km of a mine waste site (Q11) or major sensitive human and ecosystem receptors are present close to the mine waste site (e.g. Natura 2000 protected area is located within 1km distance of a waste site).

Some of parameters have risk-based threshold such as the distance to the nearest sensitive receptor is less than 1km indicates higher risk. These thresholds are based on the Irish EPA risk assessment method (Safe Quarry, 2008). Number of YES, NO and Unknown responses were registered for each site. Spatial data were used such as distance to the nearest settlements, streams and lakes, groundwater bodies and the protected Natura 2000 areas and measured by the Proximity analysis tool within ArcGIS® 10, and the topographic slope data calculated from a 50m DEM. Land use/land cover data maps at 1:100,000 scale used the CORINE database.
For parameter Q12 (Is there an impermeable layer below the mine waste facility?), a surface permeability map based on 1:100,000 surface geological map of Hungary was constructed using ArcINFO® 10 and based on the physical and geochemical characteristics of the uppermost rock units to derive high and low permeability classes beneath the studied mine sites.

Threshold values such as the distance to pathways or sensitive receptors, topographic slope and census data are defined for some of the key parameters in the EU MWD Pre-selection Protocol. For example, if there is a stream or protected ecosystem with 1km of the site or there is a nearby settlement with more than 100 inhabitants the site potentially bears high risk. The EU MWD Pre-selection Protocol sets a 1km threshold for the distance to the nearest surface water course (Q11), settlement (Q15), groundwater body (Q16), Natura 2000 site (Q17), and agricultural area (Q18) 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 was carried using the 145 ore mines test cases. For example, the original 1 km threshold value was modified to the values identified as natural breaks in the distance histograms (see Fig.3). 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).

2.2 Waste rock geochemical characterization and risk mapping

The EU MWD Pre-selection Protocol was applied on 30 abandoned and active quarries in Hungary in order to study the geochemical characteristics of the potentially generated mine wastes, in accordance with the EU MWD legislation. Ninety three field samples have been collected from the waste sites including andesite, rhyolite, coal (lignite and black coals), peat, alginite, bauxite, clay and limestone. The collected two kilograms of samples were always composed of three sub-samples located at a minimum of 10m distance from each other. 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 national 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. 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)) is measured using Proximity Analysis tools (Point Distance and Generate Near Table) in ArcINFO® 10.

2.3 Risk assessment sensitivity analysis: numerical comparison of methods

The Pre-screening European Environmental Agency (EEA) Preliminary Risk Assessment Model (PRAMS, EEA, 2005c) was also applied to the 145 test mining sites. The PRAMS model has two sets of criteria. Criteria 'A' include YES/NO answers to one or more EU relevant policy questions such as 'Are natural ecosystems of European concern affected? (question A1)' while criteria 'B' include a set of questions such as the waste site surface potentially affected area (question B1) which requires simple information likely to be readily available in data archives. Similarly to the EU MWD Pre-selection Protocol, the mine waste site may be classified as a problem area of EU interest after checking B criteria. The data derived for the implementation of the MWD 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.

3. Results and Conclusions

3.1 The EU MWD Pre-selection Protocol risk assessment using the EU thresholds

Contamination RA according to the Pre-selection Protocol was carried out in two runs. The first run used the original EU thresholds (existing risk factor in the case of slope below the site is more than 5^{o}, distance to the nearest transport route or sensitive receptor is less than 1km). The second run used 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 histogram and by (2) the median value of these parameters (Median-based threshold). 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 and landscape structure conditions in Hungary. The Median-based threshold takes a neutral position by giving a 50% chance of relative risk. This test results altogether in three final selection of sites for further more detailed risk-based investigation (high risk sites) according to the three different thresholds (EU threshold, Local threshold and Median-based threshold).

According to the number of YES, NO and Unknown responses, out of 145 mine waste sites, 19 sites have a documented incident (Q1), and among these is 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 (Jordan et al. 2011). These 19 sites are immediately directed to further examination. In summary, results show that most of the mine sites are located in hilly areas which promotes contamination transport. It is interesting that the failed Ajka red mud tailings facility is in fact located in a flat area below the slope threshold value (5^o).

3.2 EU MWD Pre-selection Protocol with local thresholds

Distribution analysis identified various sub-groups in the studied parameter thresholds (topographic slope, distance and census data). The neutral local thresholds based on median values yields 10^o for the slope below the waste site (Q10, Fig.1A), 760 m for the distance to surface water bodies (Q11, Fig.1B) and 1,722 m for the distance to settlements with 820 inhabitants (median-based) (Q15, Fig.1C). 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 $10^o >> 5^o$ slopes, close (760m < 1km) to abundant stream network and with settlements remote (1,722m >> 1km) 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 (REF: Irish EPA Method). This calls for stringent catastrophe response in case of civil protection and rescue. The 6,044m 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 (2009) resulting in 1,670 of the total 3,157 settlements with ≤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 for the population in Q15.

Table 1. Site ranking classification based on the number of YES responses of the EU Pre-selection Protocol using the original EU thresholds and the local median-based thresholds with risk classes, according to Fig.1D. 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	

According to number of YES responses to the Protocol questions, a risk-based site ranking was performed resulting in 127 and 129 mine waste sites are directed to 'Examine Further' using the EU and local Median-based thresholds (Table 1).

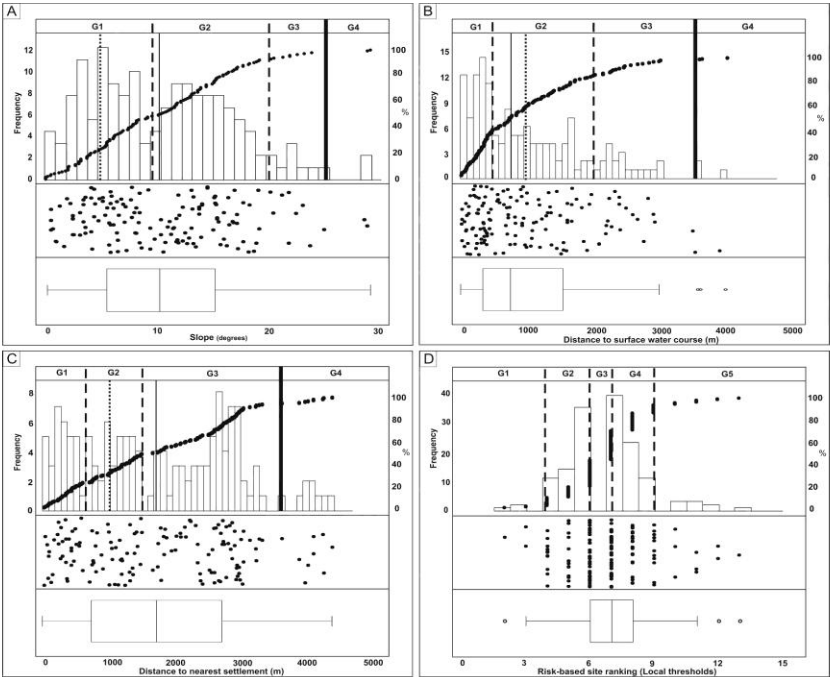

Figure 1. Distribution analyses for the EU Pre-selection Protocol parameters with histograms, scatterplots, box-whisker and cumulative probability plots. Vertical lines show sub-groups (G1, G2,.., G5) identified by the natural-breaks found in the cumulative probability plots, corresponding to local minima in the frequency histograms. Dotted line shows the EU 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 (Q10). B. Distribution analysis for distance to the nearest surface water course (Q11). C. Distribution analysis for distance to the nearest settlement (Q15). D. Distribution analysis for the total site ranking classes based on the number of YES responses and using median-based local threshold.

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

As an example for the EEA PRAMS Risk Assessment Model application, results show that according to the YES, NO and Unknown response of the PRAMS model, 19 mine waste sites have YES responses with natural ecosystems of EU concern affected (question A1), 19 sites have NO responses with contamination impact on surface water course which is not prevented according to the EU Water Frame Directive (WFD) (question A2), 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 database yet.

After checking 'B' criteria of the PRAMS RA model with available known data of the 145 mine waste sites. This archive data was collected for Hungary in this thesis research such as the 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). For example, in question B4, 88 sites have YES responses and classified as a problem area of EU interest.

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

Results show that the highest uncertainty was associated with the engineering conditions of the waste facilities and the uncertain responses were only located in the source questions, 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 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. Also, the number of YES responses can be accumulated for each site for the source, pathway and receptor questions separately which may indicate presence of multiple contamination source, multiple pathways or receptors. 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. The key parameters resulted are Q3, Q10, Q12, Q17 and Q18. For an explanation, by removal of Q3, 126 sites directed to 'Examine Further' using EU thresholds while 136 sites with 'Examine Further' using local thresholds. 'Examine Further' using local Median-based thresholds. The final selection of sites to further examination will be sensitive to and depends most heavily on these parameters.

3.5 Pre-selection RA of the 30 mine–quarry 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. 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 EU FOREGS (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%. The Spearman correlation coefficients shows significant ($p<0.05$) relationship for the aqua regia extraction in the cases of As and Ni (r=0.57), Cd and Pb (r=0.58), Co and Zn (r=0.86), Co and Cu (r=0.77), Cr and Ni (r=0.81), Cu and Zn (r=0.71), Ni and Pb (r=0.71) and Pb and Zn (r=0.63). For the deionized water leaching the elemental pair Co and Ni (r=0.8) is significantly correlated at $p<0.05$ level.

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. Results show 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.

For the deionized water leaching, the Ficklin Diagram (Fig. 2) showed that acid generation potential (pH<5.5) is for coal, lignite and peat rocks. Elevated mobile heavy metal content is associated with coal, andesite and some clay and a bauxite samples.

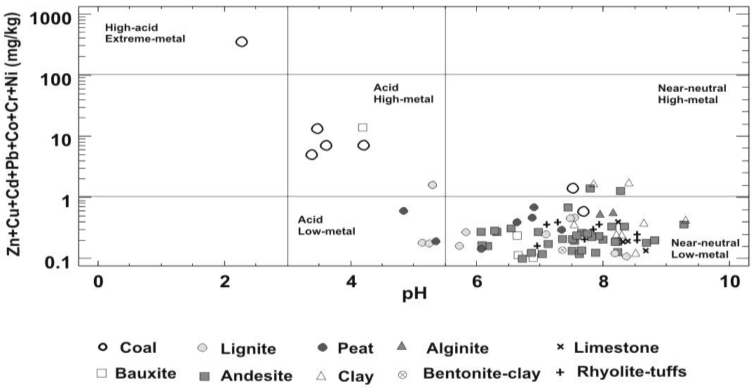

Figure 2. Ficklin Diagram showing the sum of heavy metals Zn, Cr, Cd, Pb, Co and Ni plotted against pH in the deionized water leaching (DW).

3.7 Linkage between stream water quality variables (heavy metals) and the landscape metrics

Beside the landscape metric parameters, the percentage of the main land cover classes such as artificial surfaces (CLC1) was investigated in this study (Wu et al. 2012; Xiao and Ji 2007; Romic et al. 2007; Uuemaa et al. 2005; 2013). 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. 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 MPS, Artificial surfaces (CLC1), Agricultural areas (CLC2), and Forest and semi-natural areas (CLC3). 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.

List of Publications

Abdaal A. 2014. Preliminary contamination Risk Assessment of mining waste using spatial analysis and geochemical characterization of rock formations: Key study in Hungary. Journal of Environmental Geography, in press.

Abdaal A., Jordan G. and Szilassi P., 2013. Testing contamination risk assessment methods for mine waste sites. Water, Air and Soil Pollution, 224:1416. (***IF: 1.748***).

Jordan, G., and **Abdaal A**. 2013. Decision support methods for the environmental assessment of contamination at mining sites. Environmental Monitoring and Assessment, 1859:7809-7832. (***IF: 1.4***).

Abdaal A., Jordan G., Bartha A., Fugedi U. 2013. Toxic element mobility assessment and modeling for regional geo-scientific survey to support Risk Assessment in a European Union context. The EGU General Assembly 2013, Vol. 15, EGU2013-13517, Vienna 07-12 April 2013. (Poster presentation).

Abdaal A., Jordan G., Selba I., Karapinar N. 2013. Pre-selection Methodology for the EU Mine Waste Directive Inventory of mine waste sites to support risk assessment in a European Union context. Case studies from EU Member States and Turkey. 23th International Mining Congress and Exhibition of Turkey (IMCET), Antalya 16-19 April 2013. (Oral presentation and paper in press).

Kiss J. and Jordan G., 2012. Inventory and risk ranking of closed mine waste facilities. MBFH-ELGI Co-operation Project (E7). Final Report on the National Programme, 2012. MFGI Archives, Budapest. Contributors: Detzky G., Vertesy L., **Abdaal A**., Muller T., Zsámbok I., Paszera G., Gulyas A., Ori G., Sores L., Radi K., Albert J., Hermann V. and Jerabek C. http://www.mbfh.hu/home/html/index.asp?msid=1&sid=0&hkl=547&lng=1

Abdaal A., Jordan G., Szilassi P., Kiss J., Detzky G. 2012. Ranking and testing of contamination risk assessment methods for toxic elements from mine waste sites. 9th International Symposium on Environmental Geochemistry, Aveiro, Portugal, 15-21 July 2012. (Oral presentation).

Abdaal, A., Jordan, G., Szilassi, P., Kiss, J., and Detzky, G. 2012. Testing contamination risk assessment methods for toxic elements from mine waste sites. The EGU General Assembly 2012, Vol. 14, EGU2012-6522, Vienna 22-27 April 2012. (Poster presentation).