ENVIRONMENTAL IMPACT OF METAL MINING ON CATHCMENT DRAINAGE IN THE HISTORIC MINING AREA OF RECSK-LAHÓCA MINES, HUNGARY

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INTRODUCTION

The mining of volcanogenic sulphide ore deposits in the Recsk-Lahóca mines in the Mátra Mountains of Hungary resulted in the exposure of sulphide bearing rocks to surface water and atmospheric oxygen, which accelerate oxidation, leaching and release of metals and acidity. Tailings, mine adits and associated waste rock dumps resulting from the two-century Cu, Au and Ag mining are scattered along the streams in the catchment area (fig. 1). Environmental problems arise from Acid Mine Drainage (AMD) discharge into the drainage system, which results in metal pollution and acidification of the aquatic environment within the vicinity of the mine areas. Elevated metal content in both surface waters and sediments were reported by earlier studies.

The objective of the study was to identify and assess the impacts of AMD on the receiving stream waters and sediments. Specific objectives were to (1) identify and map metal contamination due to sulphide ore deposit mining, (2) use spatial variability to determine sources of heavy metal contamination in the studied catchment, (3) distinguish natural background contributions from mine pollution, and (4) create a geochemical risk map of catchment drainage.
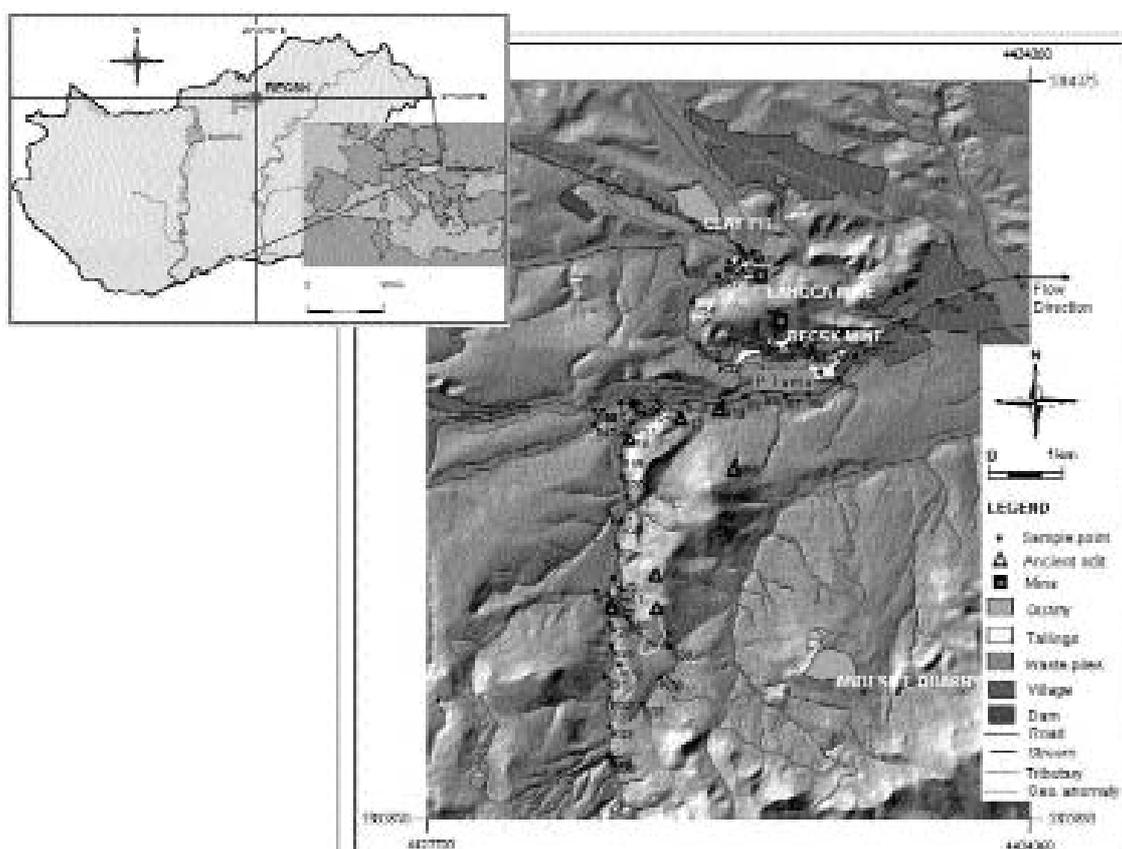

Figure 1 - Study area. Main mine features and sample locations.

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GEOLOGICAL SETTING AND MINING

Ore minerals are hosted in andesitic subvolcanic bodies, and subaerial and submarine strato-volcanic rocks that formed in the thrust-subduction zones of the inner Carpathian volcanic arc. The Eocene strato-volcanic sequence is the main host of sulphide mineralisation. Overlying the Oligocene clayey sediments, Miocene pyroxene andesite lava flows and rhyolite tuffs build-up sub-volcanic bodies and stratovolcanic sequences. Gold, silver and base metal deposits of epithermal, porphyry, vein and skarn types are associated with Eocene intrusives. The epithermal mineralisation is characterised by Au-Cu high sulphidation deposits found near surface. The porphyry copper, Cu-Zn skarn, vein and replacement mineralisations are found at deeper levels (Recsk Deeps). The near surface Au-Cu epithermal deposits are hosted by the uppermost Eocene calc-alkaline andesitic stratovolcanic sequences. In the mineralised zones, the most common ore sulphides are pyrite, chalcopyrite, sphalerite, enargite, luzonite and tetrahydrite while the gangue minerals are calcite, quartz, barite and other sulphides. The Recsk Deeps are mainly composed of three ore mineralisation types. These are galena-sphalerite vein type deposit, porphyry copper deposit and Cu-Zn skarn. Base and precious metal mining in the Recsk-Lahóca area started about 200 years ago. Several ancient adits exist in the area. The major mining activities took place mostly between the 1950s and 1998. The Lahóca hill was mined for copper between 1852 and 1979. Development of Recsk Deeps mines resulted in underground shafts extending 1200 m deep. The mine was meant to produce 3 to 5 million tons of ore per year but it never went into production since its construction in 1970.

MATERIALS AND METHODS

79 stream water samples were collected at about 250 m intervals along the stream courses in the study area (fig. 1). Duplicate samples were collected after every 10 sampling points. Samples were filtered (45µm) and acidified with HNO_3. Temperature, pH, electrical conductivity (EC) were measured in the filed. A total of 59 *stream sediment samples* were collected at the same sample sites as stream water samples where field conditions allowed. Composite samples of fine sediments collected in the active part of the stream around each water sample site were homogenised using 2-mm mesh sieve. Samples were dried in an oven at 40 ºC in lab before processing. Sample fractions of <180 µm were then sieved before subsequent chemical analysis. Metal extraction used 63% hot HNO_3 followed by shaking on a vortex shaker and shaking in a water bath at 90 ºC for 2 hours. All samples were kept on ice until analysis. Element concentrations in water samples and sediment extracts of As, Cd, Cu, Fe, Mn, Ni, Pb, Sb and Zn were determined using the ICP-AES analyser.

In this study, geochemical processes were studied as stochastic processes and statistical tools were used for data analysis. Univariate exploratory data analysis (EDA) techniques followed by multivariate statistical analyses including cluster analysis (CA) and Q-mode principal component analysis (PCA), were used to investigate element distribution patterns, geochemical abundances and gradients in the water and sediment and identify sample populations. Correlation analysis and canonical correlations were applied to investigate significant relationships among the variables measured. All correlations were visually controlled and robust-
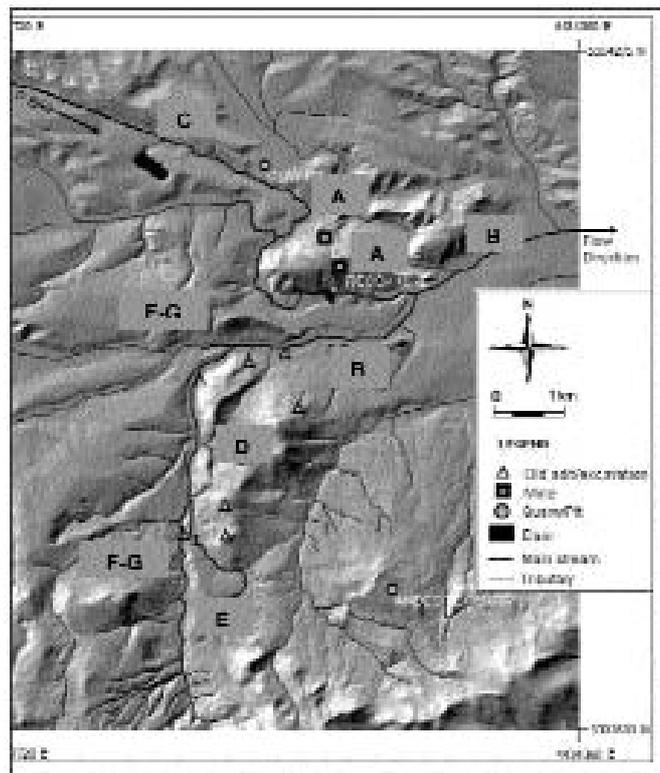

Figure 2 - Geochemical groups in stream water identified by CA and PCA. Cluster A: mines, B: biotetite-hornblende andesite, C: Oligocene clay, D: andizite with argillization, along potassium metasomatic zone, E: hydrothermal alteration zone, F-G: probable gas (CO_2, H_2S, CH_4, Radon) exhalation locations.

ness of regression analysis was assured by inter-active outlier rejection. Throughout geochemical modelling the use of robust and non-parametric techniques were preferred to classical statistical methods because geochemical datasets are often characterised by small sample sizes, multi-modal populations, outliers and non-normality. Analysis of spatial relationship between pollution source and receiving aquatic environment in the studied catchment used geographic information system (GIS) techniques. Location of mine features and sampling points were determined with GPS in the field. Digitised layers of mines, waste dumps and drainage network were overlaid on geo-referenced geological maps to study influence of the natural background and mine-induced pollution.

RESULTS AND DISCUSSION

Stem-and-leaf displays, frequency histograms and box-plots of the data sets showed poly-modal behaviour with positively skewed populations and were characterised by the presence of less than 20% outlying values, as defined by the inner-fence criteria. Outliers (forming the 'anomalous population') in water and in sediments were located either at mine sites or at the hydrothermal alteration area. All dissolved metals apart from Fe and Zn were below detection limits for other locations which form the 'background population'. Associations among metals were studied by means of the Spearman correlation analysis of log-transformed data. Where it was geochemically justified, correlations were subjected to partial correlation analysis in order to correct for possible induced correlations caused by a common variable. All correlations were checked for significance at the 0.05 significance level and were graphically examined to avoid spurious correlations. Correlations in the 'background population' in stream water showed strong correlation (r>0.7) between SO_4, EC, Mn and to lesser extent with Fe. Lack of correlation with pH indicates that pH is determined most probably by the abundant bi-carbontes in related areas. Strong correlation (r>0.7) of pH with SO_4, EC and metals in

mining areas is charactersitic to AMD. Strong correlation (r>0.8) for sediments among As, Cu, Zn, and to a lesser extent with Pb is typical to the mineralisation of the area. All sedimentary metals correlate with Fe, in particular close to mine sites where yellow boy sediments were observed, indicating metal control by co-precipitation with Fe-oxyhydroxides. Using the nearest neigbour linkage method and the Euclidean-distance as a measure of similarity in CA, water samples separated into 6 groups corresponding to well-defined geochemical regimes in the cathment (fig. 2). PCA confirmed the existence of the same groups. Sediment samples showed the same grouping but with less well-defined boundaries, indicating mixing due to sediment transport.

CONCLUSIONS

Drainage water has dynamic contact with the surroundings and thus water chemistry reflects accurately AMD pollution locations. The analysed fine stream sediments also correlate with ambient geology and pollution sources but frequent flooding events in the catchment renders them a mixture of up-stream drainage area contributions. Based on univariate and multivariate analysis of drainge chemistry, the main geochemical regimes of the catchment could be delineated. This information can be used to optimise drainage monitoring system, which aims at monitoring spatial and temporal variability due to pollution sources and natural background. This study has shown locations of elevated metal contamination in the drainage system due to natural background pollution. Finally, geochemical risk map of the drainage was created for surface waters and sediments using (1) the obtained spatial geochemical results and (2) comparison to international standards (fig. 3). The map shows that surface water is contaminated almost all along its course, partly due to natural background. Sediment pollution is concentrated in areas where physico-chemical conditions facilitate metal accumulation.
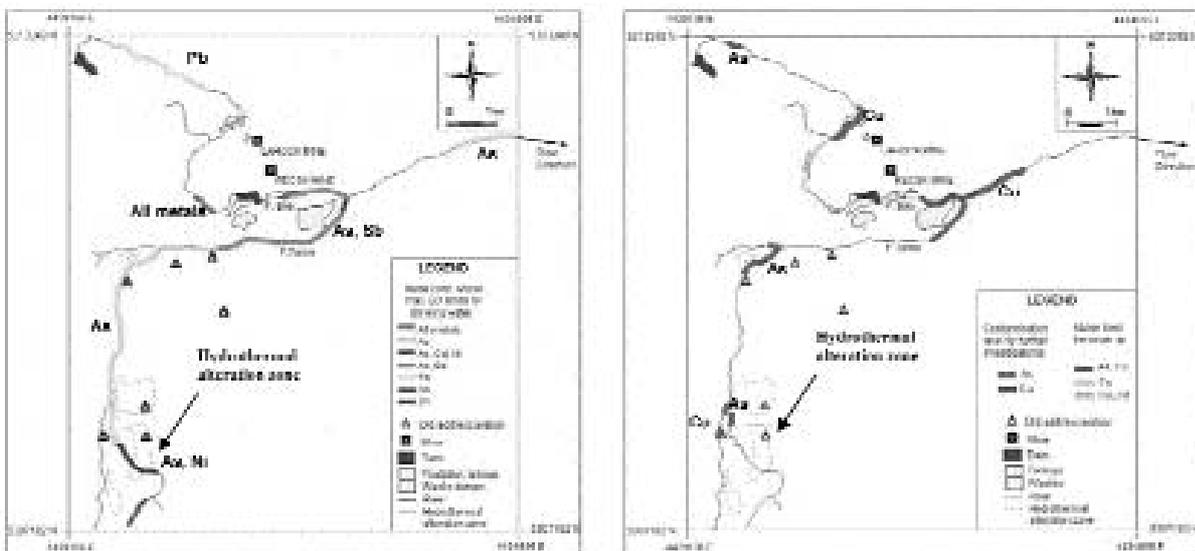

Figure 3 - Geochemical Risk Maps. A. Map showing stream water with base metal concentrations above the maximum permissible limits for EU drinking water standards. B. Map of stream sediments showing base metal concentrations requiring additional investigations and cleaning-up according to the NEN standards for soils.