Geochemical Modeling for the Contamination Risk Assessment of Mineral Deposits and Mines

Győző Jordán

Geological Institute of Hungary (MAFI)
Stefánia út 14, Budapest 1143
Hungary
http://www.mafi.hu

United States Geological Survey (USGS)
Denver CO 80225 USA

http://www.usgs.gov

Since most of the elements used by society come from mineral extraction (76 out of 90 frequently used elements), the mining of mineral resources provides essential raw material for sustainable development. Through mineral extraction and subsequent mineral processing, huge amounts of waste are produced. Metals and metal compounds exposed by mining or discarded in mine waste tend to become chemically more available, which can result in the generation of acid mine drainage and the release of toxic metals. The objective of this three-month research project was to study geochemical reactions and analyze the transport modeling of acid mine drainage and then to try to develop a risk-assessment method for mine effluents. The project was also a follow-up to a previous collaboration between the Geological Institute of Hungary and the U.S. Geological Survey, and it opened new opportunities for further joint research activities.

1. Introduction

Mining for resources to satisfy energy and raw material requirements can seriously alter the composition of the landscape, disrupting land use and drainage patterns, contaminating soil and water resources, and removing habitats for wildlife. Contamination by heavy metals and acid mine drainage effluents is a serious problem because it has direct toxic effects on humans and ecosystems. Younger et al. (2002) estimate that in the EU alone the total length of watercourses polluted by mine drainage may exceed 5,000 km. Toxic mine effluents are a major source of environmental contamination in the USA, as well. The magnitude of the problem is also demonstrated by the fact that Hungary has 15,008 mine pits registered, while there are 1,289 mineral deposits registered in Romania and 17,260 mine sites inventoried in Slovakia, both of which are neighboring countries of Hungary with many shared mining-impacted catchments (Odor et al., 1997). The Baia Mare accident in Romania in 2000 released over 100,000 m^3 of process wastewater with cyanide compounds and heavy metals into the major Tisza River in Hungary, triggering a Europe-wide realization of the environmental problems posed by mining. A water reservoir dam failure at the Recsk-Lahóca Mines in Hungary in 1999 led to the release of sediments containing significant amounts of historic heavy metal pollution that was re-suspended by the turbulent 200,000 m^3 of flood water and was then deposited downstream on the agricultural floodplain. Abandoned mines in areas with historic mining sites such as Europe, Hungary, and Colorado in the USA pose a particular environmental danger because they can be large in number and are characterized by a lack of control and a lack of data and information (Jordan and D'Alessandro, 2004).

Mining-related environmental contamination is a global problem. The extraction of non-renewable mineral resources feeds a wide range of minerals and metals into the world's economies, and mineral resources represent about 30% of the global world trade. The associated mining waste is known to be among the largest waste streams in, for example, the European Union, where it is estimated to be 400 Mt, which amounts to about 29% of total waste generated in the EU (Jordan, 2004a). Parallel to global material and waste flows, contamination flows are also on a global scale. For example, lead mining and smelting activities by the ancient Greeks and Romans have led to measurable increases in the lead concentrations of ice cores in Greenland (Boutron et al., 1994; Hong et al., 1994).

Despite the significance of the problem, the environmental effects of mineral deposits on watersheds are poorly understood, especially with regard to local and regional geology and hydrology, deposit type, climate, and the ecosystem of the watershed. If the deposits are developed, then mining techniques, methods of ore processing and tailings disposal, and metal mobility from dumps and tailings also contribute to the complexity of the environmental impacts. This complexity raises the important problem of understanding the natural background pollution due to the mineral deposits versus pollution due to mining. Jordan (2004b) reviewed the main international and national efforts and geo-scientific research methodologies for environmental assessment of deposits and mining. He concluded that the assessment must be risk-based for efficient decision-making, and it must be based on solid geological knowledge such as the USGS geoenvironmental models. It is also widely accepted (Puura et al., 2002) that due to the complexity of acting environmental factors, detailed assessment must be site-specific and based on detailed and specific geochemical modeling. Spatial and temporal geochemical modeling (Wanty et al., 2002) thus should be used for (1) parameterization of risk assessment procedures (such as transport parameters, element mobility factors, etc.) and for (2) the analysis of site-specific characteristics. The objective of this report is to present some of the results of my three-month Fulbright research project on contamination assessment and modeling in mining areas.

2. Study Area

Two test sites have been selected because there is high-density and high quality spatial and geochemical data available already for both sites, together with results from previous studies. One test site is the Redwell basin, central Colorado, with a sub-surface porphyry molybdenum and a near-surface polymetallic vein mineralization at abandoned historic mines (Verplanck et al., 2004). The other test site is the Recsk-Lahóca Mines in Hungary, where the near-surface Lahóca epithermal Au-Cu mineralization and the Recsk Deep porphyry copper deposits and associated waste dumps and tailings are the sources of the wide-spread heavy metal contamination in the catchment. Previous studies have investigated the geochemistry of water-rock interaction in the Recsk Deep (Somody and Jordan, 2005) and geochemistry of water and sediment pollution along the surface water courses (Jordan et al., 2003). Since modeling has started for the Recsk study area, this site is presented here only.

Convergent motion between the African and European plates in the Alpine period (Late Jurassic-Neogene-Quaternary) resulted in the southerly subduction of the European plate under the Pannonian continental fragment between the two continents. This led to the formation calc-alkaline volcanic arcs (Inner-Carpathian Volcanic Belt) in the Miocene, including the Matra Mts. volcano complex of the Middle Miocene age (Badenian age, 13.7 to 16.0 Ma), consisting of diorite intrusions and an associated volcano-sedimentary series of stratovolcanic andesite pyroclastics and sediments. The earliest sequence from the Eocene age in the Matra Mts. is found in the Recsk Mining Area with a shallow seated Eocene sub-volcanic hornblende diorite porphyry body intruded into the Triassic carbonate host rock and with the overlying Eocene andesite strato-volcanic sequence composing the Lahóca and other hills (Fig. 1). Oligocene transgressive marine sediments of claystone, marl and reworked tuff form the immediate cover.

The Recsk Deep Mines' (Fig. 1) mesothermal mineralisation associated with the N-S striking Eocene diorite porphyry intrusion comprises porphyry copper deposits, Cu-Zn skarn, base metal veins and replacement mineralization hosted by Eocene intrusives, Triassic limestones and dolomite, and stratovolcanic sequences, respectively. The 900 m deep mines were developed for the porphyry copper and skarn deposits between 1985 and 1998. Associated waste rock dumps were remediated between 2000 and 2003, so these do not act as contamination sources for waters, as confirmed by some studies (Mendikas, 1998; 2000). The genetically related Lahóca Mines (Fig. 1) near-surface high-sulfidation Cu-Au epithermal deposit is characterized by ore minerals of disseminated pyrite, enargite and luzonite ore minerals, with accessories of tennantite and tetrahedrite, galena, sphalerite and chalcopyrite. The Lahóca hill was mined for copper between 1852 and 1979. The Lahóca Mines have deposited more than 2.1Mt waste as unprocessed ore rock, waste rock and as tailings, covering a

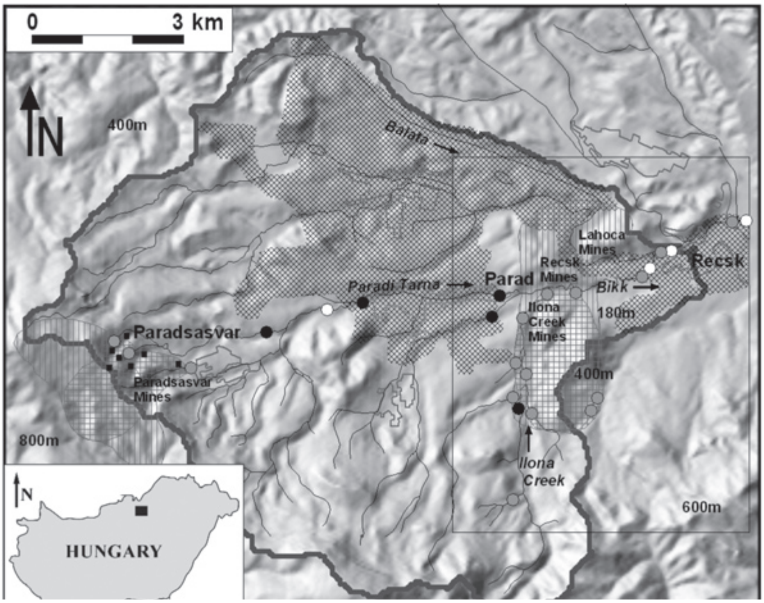

Figure 1. Location of the Recsk Mining Area. Topography: drainage lines (light lines) and the boundary of the Recsk catchment (heavy line) are shown overlaid on a shaded relief model. Arrows indicate stream flow directions. Figures show elevation above sea level (m). Landuse: cross-hatched area is agricultural lands; the rest is forest. Mines: the Lahóca, Recsk, Ilona Creek and Parádsasvár mines are indicated; solid black boxes show exploration adits at Parádsasvár. Geochemical anomalies: vertical lines and checked areas show metal anomalies identified by soil and stream water surveys, respectively. Geochemical survey samples: solid grey circles: stream water samples (Gedeon 1962); solid black circles: stream sediment samples (national survey) (Odor et al. 1997a); hatched circles: stream sediment samples (regional survey); solid white circles: torrential sediment samples (Odor et al. 2000). Light rectangle: study area shown in subsequent figures. Inset: Location of the study area in NE Hungary.

total area of about 140,000m^2 in eleven dumps. According to previous studies, soil and groundwater are polluted with metals below and around the waste dumps (VITUKI 1996) and the Baláta Creek has become enriched with polluted water and sediments near the dumps (Gedeon 1962; Rukezo 2003). The Ilona Creek Mines ('old mines') (Fig. 1) low-sulfidation epithermal deposit was mined for silver and copper between 1769-1852. Small-scale mining from narrow but high-grade veinlet zones left old adits and small waste rock dumps. Most of the old mine adits and dumps discharge highly acidic water (pH~2) with high sulphate and heavy metal content (Szebenyi 2002).

The Recsk Mining Area is located in the Parád-Recsk basin with the Lahóca Hill in the middle of the basin between the villages of Parád and Recsk in the Mátra Mts., in NE Hungary (Fig. 1). The topography of the Mátra Mts. is determined by late Middle Miocene volcanic edifices. Elevation in the catchment ranges from 180m in the basin interior to 1000 m a.s.l. in the surrounding mountains. Like the rest of the Mátra Mts., landforms of the Recsk Mining Area include primary volcanic features and landforms shaped by erosion. Mountain domes of mono and parasitic cones and elongated ridges reminiscent of fissure volcanic origin can be seen in the area. The highest mountain peak in Hungary, Mount Kékes (1,014m) lies in the south in this area (Fig. 1). The slope is gentle at the foot of the mountain domes, giving rise to undulating plains. Flat areas are limited to floodplains. Most of the catchment is vegetated by mixed beech and evergreen forest. Various oak forests interspaced with bushy grassy areas cover the low-lying regions. The flat basin area upstream of the Recsk Mines is occupied by agricultural lands (pasture and arable land) (Fig. 1). In floodplain areas, there are thick reed grasses, especially in areas around the mines. The upper part of the catchment is a part of the Mátra Natural Reserves and the whole area is rich in wildlife.

Climatic conditions are typically of temperate continental character with a mean annual temperature of 9,6°C and with a mean annual precipitation of 530-580mm. The wettest part of the year is at the end of autumn and the beginning of winter. Evaporation is important: according to estimates, two-thirds of the rainfall is lost to evapotranspiration. The dominant wind direction is westerly. The catchment of the Recsk Mining Area is 87,2km^2 characterised by numerous small streams originating from the mountain slopes forming a dendritic pattern (Fig. 1). On the foot of the mountain domes, where the slope is gentler and the terrain is undulating, the streams show a meandering pattern. In some areas, the streams follow tectonic lines, like faults, and tectonic lithological boundaries, resulting in elongated drainage patterns. The main stream Parádi-Tarna (0.040m^3/s) collects the waters of Ilona Creek at village Parád and of the Baláta Creek (0.035m^3/s) (called the Bikk Creek around the Lahóca Hill) at the Recsk and Lahóca Mines. Typically, the 10% probability flood is 51.7m^3/s for the Bikk Creek at the mines close to the village of Recsk. The Baláta Creek is a non-perennial water flow with dry channels in summer. The streams flood after winter snow melt and in summer rainstorms. The Parádi-Tarna Creek, leaving the watershed outlet, discharges eventually into the Tisza River further downstream. The Parádi-Tarna Creek is used for irrigation purposes in the farming season some 15-20km downstream of the mines. Stream flow is recharged by groundwater from the surrounding hills. Groundwater from the Recsk Deep Mines does not recharge surface waters within the catchment.

3. Materials and Methods

79 stream water and sediment samples were collected at about 250m intervals along the stream courses in the study area. Temperature, pH, and electric conductivity (EC) were measured in the field. Element concentrations of As, Cd, Cu, Fe, Mn, Ni, Pb, Sb and Zn in filtered (45µm) and acidified water samples were determined using the ICP-AES analyser. SO_4 concentration in unacidified water samples was determined using the spectrophotometry method. Stream sediment samples sieved to 2mm were digested with hot aqua-regia and analysed for total metal contents by ICP-MS.

Detailed quantitative geochemical modeling of contamination fate in mine sites for site-specific risk assessment was carried out. Detailed geochemical modeling was performed to develop case studies for the testing and verification of contamination risk assessment procedures in mine sites. In this study, geochemical modeling included univariate exploratory data analysis (EDA) techniques followed by multivariate statistical analyses, including cluster analysis (CA) and principal component analysis (PCA) in order to investigate element distribution patterns and gradients in stream water and sediments. Chemical reaction modeling was then performed by the thermodynamic reaction model PHREEQ developed by USGS to describe prevailing processes controlling contamination in the mine-impacted environment. This study was limited to the porphyry, epithermal and vein deposit types because these are the occurrences in the two test sites. For these deposit types key parameters such as acid generation potential, metal mobility, fracturedness, size, average concentration, etc. were selected based on geo-environmental deposit classification, and they were input as factors into the applied models.

4. Results and Discussion

Distribution analysis showed polymodal behaviour with less than 20% outlying values in stream water samples. Outliers (forming the ‘anomalous population’) were located either at mine sites around the Lahóca Hill or at the hydrothermal alteration area in the Ilona Creek upper reach (see Figs 1 and 2). All dissolved metals apart from Fe and Zn were below detection limits in samples from the other locations (forming the 'background population'). Strong Spearman correlation (r>0.7) of pH with SO_4, EC and metals in the 'anomalous population' at the Lahóca Mines is charactersitic to AMD. Correlations in the 'background population' in stream water showed strong relationships (r>0.7) between SO_4, EC, Mn and to a lesser extent with Fe. Lack of correlation with pH indicates that pH is determined most probably by the abundant bi-carbonates in related areas. Strong correlations (r>0.8) for sediments among As, Cu, Zn and to a lesser extent with Pb for the 'anomalous' samples are typical for the mineralization of the area. All sedimentary metals correlate with Fe, in particular those close to waste dumps where yellow boy sediments were observed, indicating that dissolved metals are attenuated by adsorption and co-precipitation with Fe oxy-hydroxides. CA and PCA were able to resolve the 'background population' into further groups corresponding to well-defined geochemical regimes in the study area. In Fig. 2a the CA dendogram makes an obvious distinction between the 'anomalous' (group A) and 'background' populations, but it also reveals the geochemical background (group B) as a strong group along the Baláta Creek draining carbonate-rich Tertiary sedimentary rocks. The group of samples in the Ilona Creek upper reach (group C) is the most distinct from the other 'background' samples, and it corresponds to the area of the surface hydrothermal alteration zone (Fig. 2b). Further downstream at the Ilona Creek lower reach samples represent stream water draining Eocene strato-volcanic andesite (group D). After the confluence with the Parádi-Tarna Creek, of more alkaline character, the Ilona Creek has a mixed character (group E) that dominates even after the confluence with the Bikk Creek draining the Lahóca and Recsk Mines (Fig. 2b). Samples found in group F-G are found in areas where gas (CO_2 primarily) exhalation is known, often manifested as bubbling water in the streams. Finally, stream sediment samples showed the same groups as stream water samples but with less well-defined boundaries, indicating mixing due to sediment transport.

Thermodynamic modeling could describe the prevailing process in the identified geochemical units. The geochemical background is characterized by carbonate equilibrium, and carbonate (calcite, aragonite) precipitation, together with metal carbonates, controls metal concentrations in the stream water upstream to the mine waste dumps. Where AMD discharges into the streams iron and manganese oxy-hydroxide precipitation has a high thermodynamic probability, and measured high metal concentrations within stream sediments at these locations confirm that metals adsorb to the oxy-hydroxides and co-precipitate with them. Similar processes occur at the hydrothermal alteration zone in the Ilona Upper reach (Fig. 2) but with less pronounced intensity.

This study concluded that drainage water has dynamic contact with the surroundings in the study area and thus the water chemistry reflects accurately AMD pollution locations. This is consistent with the findings of previous studies (Gedeon 1962). The analyzed stream sediments also correlate with ambient geology and pollution sources, but frequent flooding events in the catchment render them a mixture of up-stream drainage area contributions. Based on univariate and multivariate analyses of stream chemistry, the main geochemical regimes of the catchment could be delineated (Fig. 2). This study has also shown locations of elevated metal concentrations in the natural background due to a hydrothermal

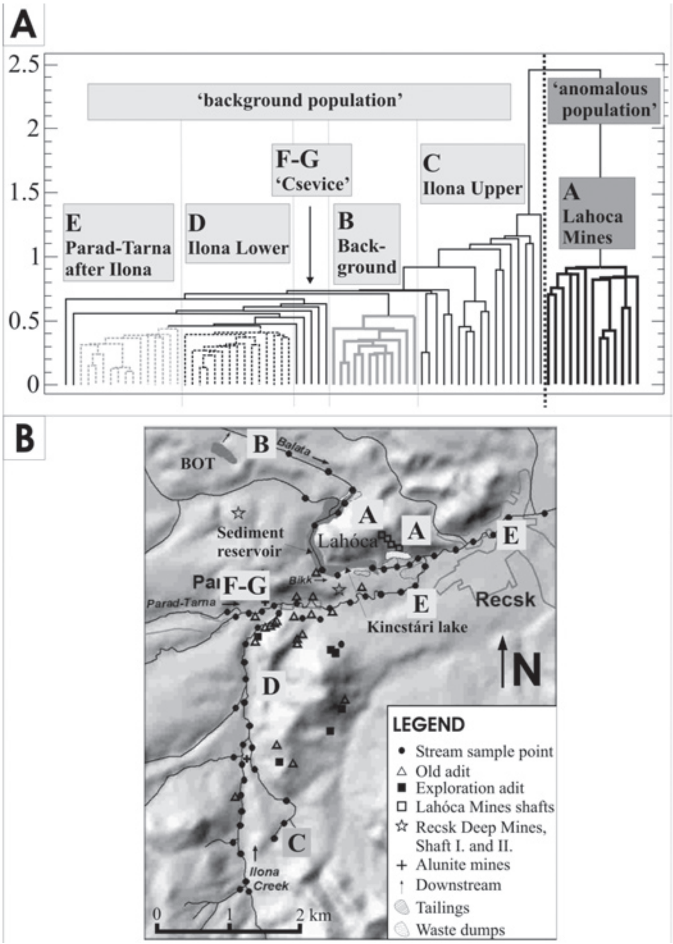

Figure 2. A. Cluster Analysis (CA) dendogram showing association of stream water samples. B. Map of study area showing location of stream water sample groups identified by CA. Stream water and sediment sample locations are also shown. Shaded relief model shows topography in the background, and black lines are drainage lines in the study area.

alteration zone in the Ilona Upper reach. On-going chemical reaction modeling is trying to describe the processes linking surface water and sediment chemistry, with special focus on the impact of mine closure on water chemistry. During active mining, water in the Baláta and Bikk Creeks around the Lahóca and Recsk Mines had pH=8.2 due to saline groundwater extraction and release from the underground mines. Water samples from this period are oversaturated with respect to carbonates (calcite and dolomite), and carbonate precipitation is confirmed by field observation of associated sediments.

These carbonate-rich sediments have significant metal (primarily As, Pb and Zn) content suggesting metal co-precipitation with carbonates. Stream water samples taken from the same location after mine closure have pH=4.0 on average. Water samples from this period are oversaturated with respect to Fe and Mn oxy-hydroxides, Al hydroxides and jarosite. Field observation of associated yellow boy sediments confirms active precipitation of these components. These secondary minerals cause significant metal concentrations in the analyzed stream sediments, suggesting that metals are efficiently scavenged from the stream water by adsorption and co-precipitation with secondary minerals.

Contaminated sediment transport was also modeled in the Recsk mining catchment in order to study the fate of metals bound to soils and sediments and to assess the impact of land use scenarios that minimize the export of contaminated waste in the catchments (van Romaey et al. 2005). In this study the WATEM/SEDEM distributed erosion and sediment transport model was used to assess the mean annual export of sediment-bound heavy metals (Cu in this study) from a catchment, taking into account the spatial pattern of metal concentrations and the topological relations between sediment sources and sinks. The model used metal enrichment factors to take into account metal adsorption to soil and sediment particles. Sediment deposition data in the Sediment Reservoir and Kincstári-tó Reservoir at the outlet of the catchment were used to calibrate the model for erosion and sediment transport. The model predicted about 9,000 t sediment and about 1.3 t of particulate copper exported from the catchment per year on average. The calculated copper export from the catchment shows good agreement with measured particulate Cu data taken from a previous study. The impact of land use on metal export was evaluated for land use scenarios taken from aerial photographs in 1987 and a field survey in 2003 (Bats 2004). Further scenarios tested a number of extreme cases: Scenario 3 assumed fallow for all agricultural lands (highest erosion and sediment transport capacity), Scenario 4 assumed 100% protection of waste dumps, and Scenario 5 applied forest cover everywhere plus protection of waste dumps (lowest erosion and sediment transport capacity, and no point sources of metals). The results show that land use configurations that minimize the total sediment export from the catchment are not necessarily the same as those that minimize the volume of exported polluted waste (Van Romaey et al. 2005). Land cover interferes with Cu-export from (1) non-point sources because of the volume of total exported sediment and because of enrichment processes induced by sediment deposition, and (2) point sources because of buffers such as forested stream banks on the flowpath between point source and river channel. This means that sedimentary metal export from the catchment cannot be modeled without a spatially distributed approach, and for future estimations spatially explicit land cover change models are necessary.

5. Conclusions

Historic mining in the Recsk catchment has made a strong impact on the surface environment. Old mines and associated waste dumps still release AMD and hinders natural re-vegetation. Industrial mining at the Lahóca Mines produced large waste rock and tailings dumps that release AMD and contaminated sediments, impacting the water and sediment chemistry of nearby streams. However, metal contamination in stream water is attenuated within a short distance (200-250m). Detailed geochemical investigation of stream water and sediment has shown that naturally high background values of metals at a hydrothermal alteration zone do occur in the area. Mine activities in the Recsk Deep Mines did not cause significant surface pollution because groundwater does not recharge surface waters and groundwater chemistry is buffered by Triassic carbonate rocks.

6. The Fulbright Project and My Personal Experience in the USA

The scientific significance of the project lies in the unique combination of geochemical models with formal risk assessment methods (the latter was not presented in this paper). The project is unique in that these are also calibrated and verified by detailed geochemical transport and reaction modeling. The social significance of the project lies in the fact that such models provide a scientifically sound and efficient inventory and ranking method for contaminated mining sites, thus supporting environmental decisions about site remediation and land development planning.

The Fulbright Project provided an opportunity to collaborate with Dr. Richard Wanty and his excellent research group at USGS. Collaboration between MAFI and USGS goes back to a US-Hungarian Joint Fund project between 1994-1998 in the field of deposit modeling and the environmental assessment of mining (Odor and McCammon, 1999). A NATO Advanced Studies Institute workshop on deposit and geoenvironmental models was held in Matrahaza, Hungary in 1998 with Richard Wanty's USGS research group as lecturers and with myself as a student and the Secretary of the Local Organizing Committee of MAFI (Fabbri et al, 2002). During these two weeks I made personal contacts with USGS researchers. Since that time most of the Hungarian participants of the US-Hungarian Joint

Fund project have retired from MAFI, causing a pause in research collaboration until today. The next contact was at my personal initiative during my stay at the Joint Research Centre of the European Commission when I organized and chaired the 'Mine Environment Session' on the 4th European Congress on Regional Geoscientific Cartography and Information Systems, in 2003 in Bologna, Italy, to which I invited Richard Wanty to present his group's results. Together with the American colleagues we carried out a joint field campaign at the Recsk study area in 2005 (Fig. 3).

The present project both renewed and improved collaboration between the world-leading USGS and MAFI as, together, a leading regional geoscientific research centre. My most interesting field-work was done together with my host USGS fellows. This was a major professional training opportunity and a lifetime experience for me (Fig. 4). Most importantly, with the personal involvement of the project participants, this project opened the opportunity to higher-level collaboration between USGS of USA and EuroGeoSurvey of the EU in the field of mine environment

Figure 3. Field work with USGS colleagues in the Recsk mining catchment study area, Matra Mts., Hungary. Above: waste rock dumps. Below: USGS and MAFI personnel discussing steam water sampling.

assessment. Results are disseminated in joint scientific papers and conference presentations, in communications on international workshops, and through expert group meetings and on the internet (Jordan 2008). Despite my international experience I had not been to the USA before, not even as a tourist. I found my trip exciting and challenging from a cultural point of view since it was my long-time plan/dream to see this wonderful country, experience a different life and work style, and to meet the friendly and open American people. It is my intention to share my personal and professional experiences with my Hungarian colleagues and with my fellow European researchers in various European international expert groups and organizations.

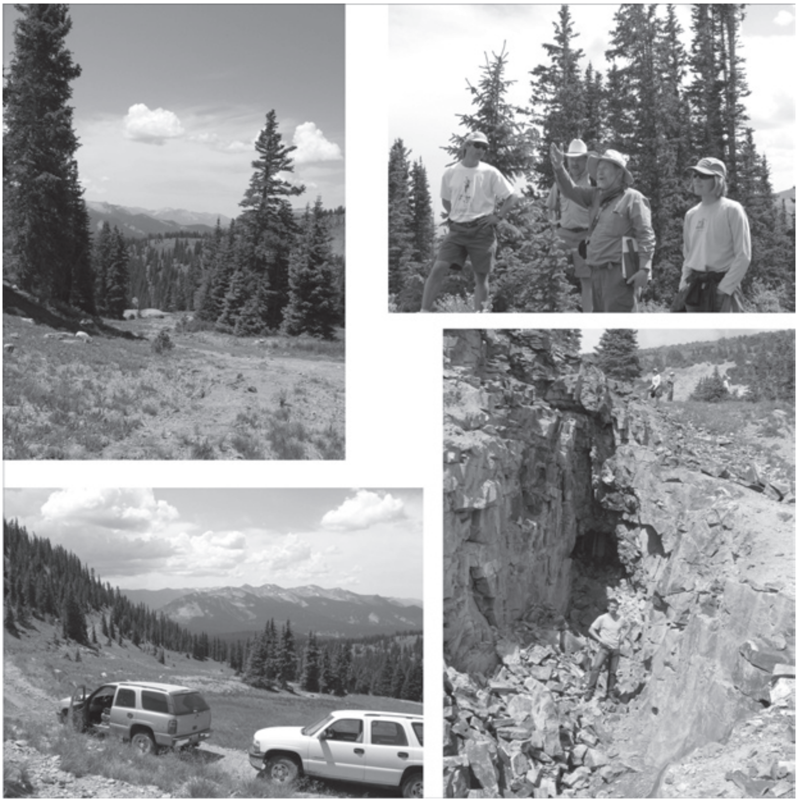

Figure 4. Field work with USGS colleagues in the Rocky Mts. old mining areas, Colorado. Above: scenery in an old mining area, field guide with USGS colleagues. Below: field trip with USGS and National Park colleagues and myself standing in on old mineral exploration adit.

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