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MIHAELA SERBAN (BUCHAREST), MARK G. MACKLIN (ABERYSTWYTH), PAUL A. BREWER (ABERYSTWYTH), DAN BALTEANU (BUCHAREST), GRAHAM BIRD (ABERYSTWYTH)

THE IMPACT OF METAL MINING ACTIVITIES ON THE UPPER TISA RIVER BASIN, ROMANIA AND TRANSBOUNDARY RIVER POLLUTION

Abstract. Mining is an old activity in Romania, dating from the Daco-Roman times in the Apuseni Mountains, and its impact on the environment is as long-standing. The most important metal-bearing areas in this country are found in the north-west, in the Apuseni Mountains and in the northern part of the Eastern Carpathians (Maramureş area). In January and March 2000 two tailings dam failures in Maramureş County, northwest Romania, resulted in the contamination with cyanide and heavy metals of the Tisa River, a major tributary of the Danube. Following these accidents, a research programme was initiated in northwest Romania to establish metal levels in rivers affected by tailings dam failures and by historical mining activity. One of the main findings of this study is that nearly all of the major pollution hotspots in north-western Romania relate to inputs of mine waste from currently active mines whose minewater treatment plants are not functioning properly. Contamination from mine waste, particularly in river and groundwater, has been shown to extend no more than 5 km from the point of effluent discharge and affects a corridor approximately 1 km width central on the present river channel.

Key words: metal mining, Tisa River, tailings dam failures, transboundary pollution, Romania

INTRODUCTION

Mining is an old activity, dating from the Daco-Roman times in the Apuseni Mountains, and its impact on the environment is as long-standing. The most important metal-bearing areas in this country are found in the north-west, in the Apuseni Mountains and in the northern part of the Eastern Carpathians (Maramureş area). Nonferrous metals are widespread in the volcanic mountains, where their genesis is related to magmatic processes. Among these, polymetallic sulphur ores (copper, lead and zinc) are the most numerous ones, especially in the Oaş, Gutâi and Ţibleş volcanic chain, as well as in the southern part of the Apuseni Mountains (more precisely, the Metalliferous Mountains). The presence of gold-silver ores is due to the Neogene eruptions and are concentrated in the Maramureş County at Săsar-Valea Roşie, Şuior and Baia Sprie, and in the Metalliferous Mountains, where they are deposited in four layers forming the Apuseni Mountains' golden quadrilateral: Brad–Săcărâmb, Almaş-Stanija, Bucium and Baia de Arieş (Fig. 1). Recent interpretations consider that the spatial distribution of mineral substances at regional level is related to the main faults and structures according to the plate tectonics concept (Radulescu and Sandulescu 1973; Popescu 2003).

The studied area numbers 20 mining exploitation, belonging to the two companies: Minvest Deva (for the Apuseni Mountains area) and Remin Baia Mare (for the Maramureş area). Their activity produced 168 waste dumps, 73 in Apuseni Mountains and 95 in Maramureş County, totaling 7 million m³ and covering 308 ha. At the same time, large quantities of waste are being spilled into the 92 tailings dams (511 million tonnes) extending over 2,025 ha, of which 40 are active (40%), 38 under conservation and 14 are held in reserve (Fodor and Baican 2001).

Waste dumps and tailings dams are large positive landforms which totally modify terrain morphology. They might be affected by deflation, landslides, sheet and gully erosion. The drainage network around the pits is disorganized through

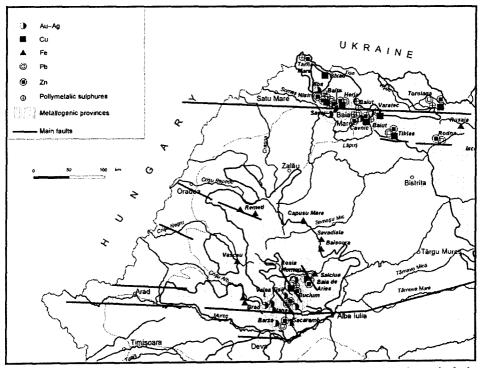


Fig. 1. Distribution of mineral substances in north-west Romania in relation with the main faults

the deviation and even disappearance of watercourses; they collect a large quantity of suspensions and toxic elements, particularly metal ions (Cu, Pb, Zn, Cd, etc.), increasing acidity and implicitly their contamination.

ENVIRONMENTAL IMPACTS. A HISTORICAL PERSPECTIVE

There are several stages in Romanian mining history, which differ in what regards the extractive techniques and the quantity of extracted ores. A first phase is known in the Daco-Roman antiquity, when mines were exploited in the Apuseni Mountains. The methods of collecting the precious metals from the rivers were quite rudimentary (by pestling or stamping). The Romans brought new technologies of extracting and processing metals straight from the ores through galleries. This practice continued up to 16th–17th centuries. Reservoirs were built in the upper part of the river basins to store the water needed for the exploitation process. The mines of Maramureş County were little known by the Romans, in exploitation beginning only in the Middle Ages. Historical documents say that during Roman times 720,000 kg of silver and 1,345,000 kg of gold were extracted from the Apuseni Mountains mines alone (H a i d u c 1940).

After the Romans' withdrawal from Dacia, the mining activity slowed down, yet without ceasing altogether. In the Middle Ages, German miners were brought into Transylvania and colonized in a few gold-rich localities (e.g. Baia Mare, Abrud). As from the year 1000, the mines of the Apuseni Mountains, as well as those in Maramureş County became prosperous, being systematically operated. Apart from gold and silver, complex ores were exploited as well as copper, lead and zinc. Also, the extraction techniques were improved. In the 18th–19th centuries, mines were licenced to some foreign mining companies and operated with modern technologies.

The capitalist phase (mid-19th century–mid-20th century) is characterized by a strong development of the mining industry due to the improvement of exploitation and processing techniques (the invention of the steam machine, electric energy), to the emergence of big metallurgical plants and to a favorable legislation (The Mining Law adopted in 1924). Ores were exploited both by private and state companies.

The socialist period (mid-20th century–1989) witnessed an intensive ore exploitation with high extraction and production rates defying rational use. Geological R&D activities gained importance and new pits were opened (Roşia Poieni, the largest copper mine in Europe). The quantitative development implied major losses and negative impact on the environment. The wastes resulted from primary metal extraction was no longer spilled into the rivers, but deposited in dumps and tailings dams, which thus became the main sources of pollution.

At present (since 1990), the mining industry has been declining considerably: under the current technical and technological conditions mines were no longer economically viable, many deposits having been exhausted in the previous period. The closed mines are not benefiting from ecological rehabilitation and conservation programmes, continuing to have a direct impact on the environment. In spite of these, some companies are still operational, new pits are going

to be opened (Rosia Montana, in the Aries river basin). The main problem of mining affected areas is related to dumps stability. So far now, 75 accidents related to tailings dam failures have been recorded world-wide, 7 of them in the last 4 years alone, many resulting in casualties and great damage for the environment (Diehl 2003). In Romania three such accidents occurred, too: a very severe one in 1971, on the Certej River (one of the River Mures tributaries) causing more than 90 deaths and two in 2000, in Maramureş County. The tailings dam failures that occured in January and March 2000 in the upper Tisa basin, Maramures county, northwest Romania attracted enormous media interest, largely because the spill resulted in the pollution and fish deaths downstream in Hungary, Serbia and Bulgaria. The first accident took place at Bozânta tailings pond belonging to "Aurul" Baia Mare mining company, used for the processing old tailings pond material just outside city of Baia Mare using cyanide to recover Ag and Au. As a result of very high snowfall, followed by rapid thawing and heavy rainfall, the tailings dam wall was breached on the 30 January 2000, releasing nearly 100,000 m³ of waste water and sediment containing high concentrations of cyanide and contaminant metals into the Lăpuş and Somes rivers. The second major tailings pond failure in the Novăţ valley, 10 km north of Baia Borsa occured two months later on the 20 March. Similar to the earlier spill, the Novăt tailings dam wall was breached following rapid snowmelt and heavy rain, discharging around 100,000 m³ of contaminated water and 40,000 tonnes of solid waste into the Vaser and Viseu rivers, the latter joining the River Tisa at the Ukrainian border.

Mine tailings in the affected river systems were dispersed under very high flow conditions, during wich sediment-associated metals were deposited in both within-channel and overbank environments. The volume of solid waste in relation to the size of both drainage basins was, however, relatively small, and as a consequence it did not disrupt river dynamics except immediately downstream of the dam failure sites.

These accidents were investigated in order to assess their long-term impact (Brewer et al. 2003; Macklin et al. 2003; Bird et al. 2003a, b). This paper is a synthesis of the results obtained in a four-year survey (2000–2003) in Romania, carried out by researchers from the University of Wales, UK and the Institute of Geography, Romanian Academy. Initially, investigation were conducted in Maramureş and Satu Mare counties, being subsequently extended to Hungary, on the Tisa River and its tributaries on Romanian territory (2001). Over the 2002–2003 period, part from the investigations performed in Maramureş and Satu Mare counties, they assessed environmental impact of mining activities in the Apuseni Mountains on the Arieş–Mureş, Ampoi, Crişuri and Certej river basins (Fig. 2).

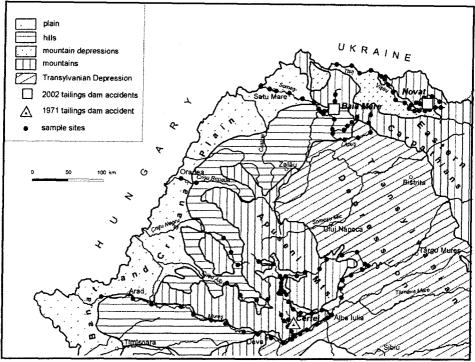


Fig. 2. The main geomorphic units and sample site locations

Depending on the mining impact, the Romanian river systems can be classified into several types: 1) with historical mining activity, presently inactive; 2) with historical and present mining activity; 3) with opportunities for the opening of new exploitations and 4) with tailings dam accidents.

FIELD SAMPLING AND LABORATORY METAL ANALYSIS

During the 2000–2003 period, over 1,000 surface and groundwater, as well as floodplain and channel sediment samples were collected from the main mining-affected Romanian rivers. Water samples were filtered through 0.45 m filter papers and acidified with three drops of 50% nitric acid in the field, before multi-element analysis using an ICP-MS (VG Elemental Plasma Quad II+). River channel and floodplain sediment samples were collected using a stainless steel trowel from bar surfaces and river banks, respectively. Sediment samples were air dried, sieved through a 2 mm plastic mesh, digested in nitric acid and metal levels determined using AAS (Perkin-Elmer 2380). Channel sediment samples (<63 μ m fraction) were subjected to a sequential extraction procedure (SEP) based on a method described by A. M. Ure et al. (1993).

Metal concentrations in surface water were assessed against target and imperative values in the EC directive (75/440/EEC) required of surface water intended to be used for the abstraction of drinking water (Table 1). Metal levels in river and floodplain sediment were compared with the latest (4 February 2000) Dutch target and soil remediation intervention values (Table 2). The Dutch intervention values for soil/sediment remediation are considered to be numeric manifestations of the concentrations above which there can be said to be a case of serious contamination. These values indicate the concentration levels of metals above which the functionality of the soil for human, plant, and/or animal life may be seriously compromised or impaired. Target values indicate the level at which there is a sustainable soil quality and gives an indication of the benchmark for environmental quality in the long term on the assumption of negligible risk to the ecosystem.

Table 1

	Target value [μg · L ⁻¹]	Imperative value [µg · L ⁻¹]
Pb	_	50
Zn	500	3,000
Cu	20	50
Cd	1	5

EC target and imperative values for the abstraction of surface water for drinking (75/440/EEC)

Table 2

Target values and soil remediation intervention values for selected metals from the Dutch Ministry of Housing, Spatial Planning and Environment (VROM, 2001). Values have been expressed as the concentration in a standard soil (10% organic matter, 25% clay)

	Target value [mg · kg ⁻¹]	Intervention value [mg · kg ⁻¹]
Pb	85	530
Zn	140	720
Cu	36	190
Cd	0.8	12

TAILINGS DAM FAILURES AND HEAVY METAL POLLUTION

1. Zn concentrations in surface water and river channel sediment samples collected during 2000, 2001, 2002 and 2003 in the river Sasar are plotted against distance downstream from the Săsar's source (Fig. 3). In 2000, metal concentrations in surface waters in the upper Săsar were relatively low but rose sharply downstream of the Esmeralda Baia Sprie mine. Zn concentrations remained above imperative

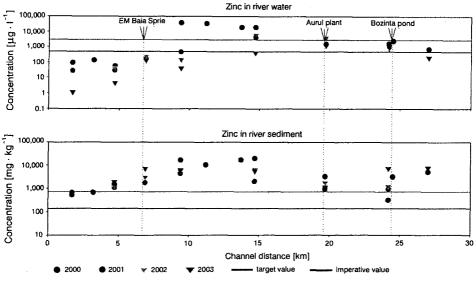


Fig. 3. Săsar River - Zn concentration in river water and sediments

values at Baia Mare than reduced to near imperative values immediately upstream of the Aurul tailings pond. Metal concentrations in river water are generally lower in the Săsar in 2003 than in previous years. Solute Cd and Zn in 2003 still exceed EU target values downstream of the Aurul plant and Bozânta pond. In 2003 Cd, Cu and Zn concentrations in river water are 5–80% lower than in 2000.

The pattern of metal contamination in Săsar River has remained largely stable over the 4 year sampling period and matches the patterns described above for surface water. Pb and Zn concentrations exceed Dutch target values at headwater sample sites, but then increase to exceed intervention values between Baia Sprie and Baia Mare. Metal concentrations then reduce downstream of Baia Mare (adjacent to the Aurul tailings pond) to levels either just above intervention (Zn) or above target (Pb) values. However, Pb and Zn concentrations exceed Dutch target values at all sites in all 4 years, and 60% of all samples exceed Dutch intervention values. In general, metal concentrations in river channel sediments were lowest in 2001, with Zn concentrations tending to be highest in 2003, and Pb concentrations tending to be highest in 2002.

Water and sediment quality data from the last three years show that the Săsar is persistently polluted by heavy metals, however, Pb concentrations did comply with EU directive 75/440/EEC at all but two sites in the River Săsar between 2000 and 2003. Although it appears that the Esmeralda Baia Sprie mine was not contaminating the River Săsar to the same extent in 2002 and 2003 as it was in 2000, Zn levels in river water flowing through Baia Mare did exceed imperative values in all four years. In summary, surface water quality in Baia Sprie and Baia Mare

poses a potentially serious hazard to human health and it is interesting to note that metal concentrations adjacent to the Aurul tailings pond, 10 km downstream of Baia Mare, are consistently lower than in the town itself.

2. The second recent mine tailings dam failure at the Novăţ-Roşu tailings pond (March 2000) affected the Novăţ-Vişeu-Tisa river system. Surface water and river channel sediment metal concentrations (Cu) measured during July 2000–July 2004 in the Novaţ-Vaser-Vişeu river system are plotted against distance downstream form the Novăţ's source in Figure 4.

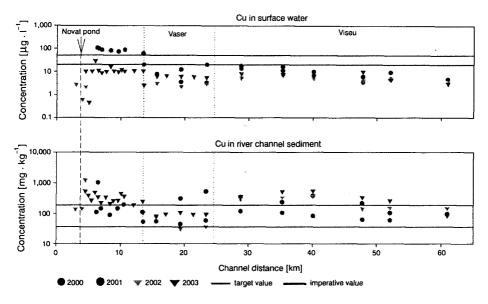
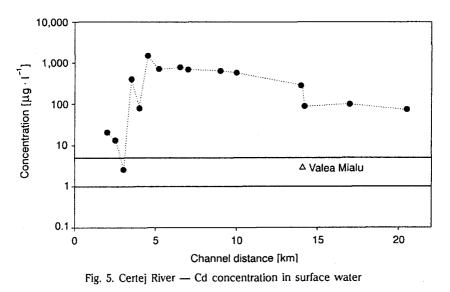


Fig. 4. Novăţ-Vaser-Vişeu river system - Cu concentration in river water and sediments

All surface water samples in the Novăţ-Vaser-Vişeu rivers have dissolved Pb and Zn concentration that are below EC target values, including river water samples immediately downstream of the Novat tailings pond that failed in 2000. Zn concentrations generally decrease downstream in the Novăţ-Vaser-Vişeu system, but Pb concentrations increase downstream of the Vaser-Vişeu confluence. In general, surface water metal concentrations in the Vaser and Vişeu rivers were higher in 2003 than in 2002, but lower than 2000 and 2001, suggesting a possible recovery of surface water quality in these rivers following the March tailings dam failures. Although Pb concentrations are similar in the Săsar and Novăţ-Vaser-Vişeu river systems, Zn concentrations are tipically 10 to 100 times higher in the Săsar. The principal source of contamination in the Novăţ-Vaser-Vişeu river system is from the Novăţ-Rosu tailings pond, and it may be that additional Zn is being supplied to the Săsar from ore processing activities or from municipal waste produced from Baia Mare and Baia Sprie. Over the four year sampling period metal concentrations in Vaser and Vişeu rivers sediments were at their highest in July 2003 (3 years after the Novăţ–Roşu tailings dam failures), recent floods (July–August 2002) may have activated metal sources. Although metal levels in the Novăţ in 2001 were similar to levels in the Vaser and Vişeu, the highest concentrations, found close to the Novăţ–Roşu tailings pond, did exceed Dutch intervention values and indicate that the channel sediments downstream of the tailings pond are still significant potential source of sediment bound metals. In this respect, system recovery is being significantly slowed by metal inputs from the Novăţ tailings dam and mining in the Borşa area.

3. The Certej catchment, a tributary of the River Mureş, lies within the Metalliferous Mountains (southern part of the Apuseni Mountains) and contain open-cast base and precious metal mines and tailings ponds. Today, ore processing waste is stored in the Mialu tailings pond which is impounded by a 64 m tailings dam. The tailings are enriched in As (370 mg \cdot kg⁻¹), Pb (3,400 mg \cdot kg⁻¹) and Zn (2,200 mg \cdot kg⁻¹). In 1971, a pond which was located on the hillside above the town of Certeju de Sus failed and resulted in death of more than 90 people. An investigation is presently being undertaken which is examining the long-term effects of 1971 dam failure, particularly on downstream floodplain sediment quality in the Certej valley. Initial data suggest that highest metal concentration (550 mg \cdot kg⁻¹ Pb and 1,000 mg \cdot kg⁻¹ Zn) in surface floodplain soils (0–15 m) are found within 20 metres of the river channel.

In July 2002, Cd concentrations in both surface water and river channel sediment significantly increase immediately downstream of the Coranda mine. However, Cd concentrations in water and sediments exhibit very different downstream patterns. Whilst Cd concentrations in surface water reduce with distance down-





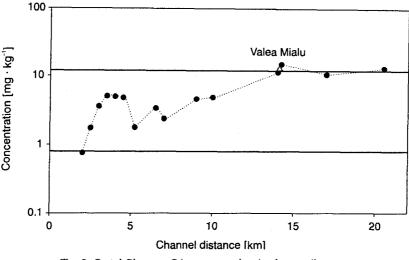
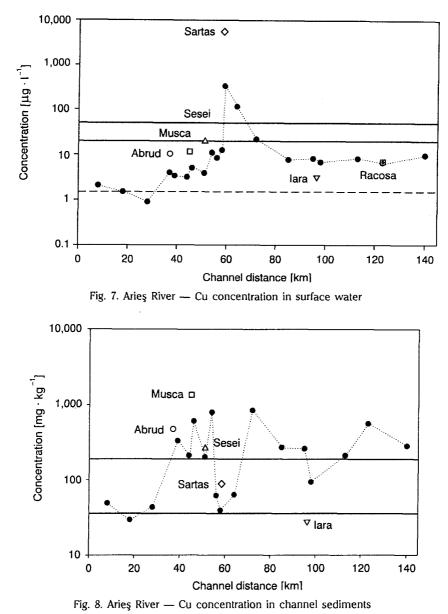


Fig. 6. Certej River - Cd concentration in river sediments

stream in the Certej river from a peak concentration at 3,400 μ g · l⁻¹ at 5.2 km, concentrations of Cd in river sediment increase, peaking at 22 km downstream (Fig. 5 and Fig. 6). This suggests that there may be an exchange of metals between the solute and sediment-bound phases, which is probably related to downstream changes in the physico-chemical environment.

PRESENT-DAY MINING ACTIVITY AND HEAVY METAL POLLUTION

Aries catchment has the highest metal potential from the Apuseni Mountains. There are about 20 ore deposits, the most important ones being in exploitation at Rosia Montană and Roșia Poieni. Downstream variations in surface water and river channel sediment Cu and Zn concentrations in the River Aries are plotted against distance downstream in Figure 7 and Figure 8. Despite the presence of long-established mining activity in the Aries system, particularly at Baia de Aries and Rosia Poieni, solute metal concentrations are relatively low compared to the River Săsar, and with the exception of three sites downstream of the Sartăş Valley confluence, do not exceed target values at any of the sample sites. This phenomenon can be due to three factors: first, there may not be a direct hydrological coupling between mine sites and river channels in the Aries catchment; second, the large number of unmined tributary catchments in the Aries basin may deliver uncontaminated water to the river, diluting contaminants resulted from active mine sites; third, the natural buffering capacity of the local, limestone rich, geology, which creates high pH in the River Aries system (7.3-8.7), will promote dissolution of solute metals, thus giving rise to low solute metal levels in the River Aries.



Channel sediments are relatively more contaminated than surface waters in the Arieş river, particularly for Cu and Zn wich exceed intervention values at 61 and 16 percent of sample sites. Cu and Zn concentrations increase downstream of mining-affected tributaries, suggesting their importnace as a source of metals to channel sediments in the Arieş river. The presence of large-scale Cu mining at Roşia Poieni may in part account for the number of sites where Cu concentrations exceed the Dutch intervention value. Under periods of flood discharge on the

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River Arieş, the contaminated channel sediments could be remobilised and deposited on local floodplains, ultimately being incorporated into agricultural soils and thus available for uptake by both crops and livestock.

The onset of large-scale ore extraction from the Roşia Montana gold deposit could pose potentially serious problems with respect to metal contamination in the Arieş–Mureş system. Data collected from the River Arieş has indicated that the decoupling of mining activity from river systems, in addition to a well-buffered natural environment, can help to keep solute metal levels relatively low, despite the presence of large-scale mining activity and polluted tributary streams. However, it is also apparent that channel sediments in the River Arieş are often highly polluted, and highlights the fact that even now attention needs to be paid to the potentially deleterious effects of existing metal mining on local river systems. If large scale mining operations are initiated at Roşia Montană, there is still an opportunity to adopt more stringent environmental quality controls to ensure the Arieş and Mureş river systems are not degraded to the same extent as many of the mining affected rivers in Maramureş County.

TRANSBOUNDARY IMPACT OF MINE TAILINGS DAM FAILURES

In July 2001, surface water and sediment samples were collected from 16 sites along the Tisa River on the Hungarian territory, as well as from the principal tributaries coming from Romania, affected by industrial, mining and munic-

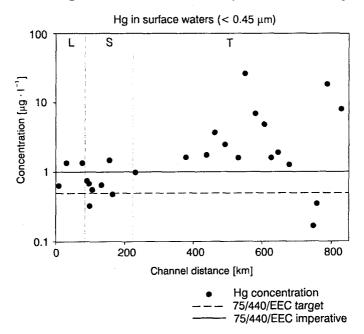


Fig. 9. Solute Hg concentration in the rivers Lăpuş, Someş and Tisa

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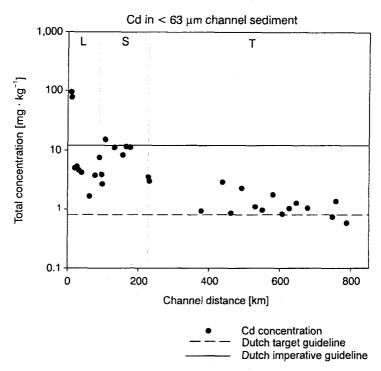


Fig. 10. Concentration of Cd in river channel sediments in the rivers Lăpuş, Someş and Tisa (Bird et al. 2003)

ipal metal pollution (Someş, Mureş, Criş) for assessing the transboundary impact of 2000 tailings dam accidents and of mining activities. The Tisa was the only river sampled in July 2001 where no surface water metal concentrations exceeded intervention values for Pb, Zn, Cu or Cd. Mercury was the only element that exceeded intervention values on the Lăpuş, Someş and Tisa rivers (14 out of 16 sites on the Tisa) (Fig. 9). But, concentrations on the Tisa are generally much higher than on the Lăpuş and Someş, indicating that there must be sources of Hg within the Ukraine or Hungary. Hg concentrations on the Tisa are a cause of concern.

River sediments metal concentrations in the Tisa, downstream of its confluence with the River Someş, are below intervention thresholds at all of the 16 sample sites. The highest metal concentrations in the River Tisa sediments are found 3 km downstream of the River Somes confluence. This would suggest that metals are being introduced into the River Tisa adsorbed to sediment supplied from the River Someş. In general, Cd, Cu, Zn and Pb concentrations in the Tisa fall between Dutch target and imperative guidelines (Fig. 10).

CONCLUSIONS

One of the main findings of this study is that nearly all of the major pollution hotspots in north-western Romania relate to inputs of mine waste from currently active mines whose minewater treatment plants are not functioning properly. Contamination from mine waste, particularly in river and groundwater, has been shown to extend no more than 5 km from the point of effluent discharge and affects a corridor approximately 1 km width central on the present river channel. Between these relatively localised pollution hotspots, river and growndwater generally comply with EU guidelines, although metal concentrations in river sediment and floodplain soils can be elevated as a result of historical contamination.

River systems still affected by long-term mining activity (e.g. Săsar–Someş) are characterised by very high metal concentrations often due to inadequate waste treatment facilities. Data collected from the Arieş River has indicated that the degree of pollution is dependent upon the nature of mine waste, the hydrological link between mines and local rivers, and the local physico-chemical environment.

Catchments affected by tailings dam failures (e.g. Novăţ-Vaser-Vişeu and Certej) seem to have recovered in terms of surface water quality. Channel sediments are still contaminated with mine tailings, having a long-term effect on the river system. Identifying contaminant sources, the location of temporary contaminant stores (e.g. floodplains) and characterising the physical and chemical controls on the mobilisation and dispersal of contaminant metals is critical to the sustainable development of future mining activity in Romania.

In these areas research is urgently required on controlling mine-waste generation and preventing its release into rivers that should include: event-based sampling and use of isotope fingerprinting techniques to identify contaminant sources and monitoring the efficiency of mine-waste treatment plants associated with both tailings dams and ore processing plants, facilitating steps towards compliance with the EU Water Framework Directive.

Institute of Geography the Romanian Academy 12 Dimitrie Racovita, Sector 2 RO-70307, Bucharest, Romania email: geoinst@rnc.ro.

Institute of Geography and Earth Sciences University of Wales, Aberystwyth, Ceredigion, SY23 3DB, UK

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STRESZCZENIE

M. Serban, M. G. Macklin, P. A. Brewer, D. Balteanu, G. Bird

WPŁYW GÓRNICTWA METALI KOLOROWYCH NA GÓRNĄ CZĘŚĆ DORZECZA TISY W RUMUNII I TRANSGRANICZNE ZANIECZYSZCZENIE RZEK

Górnictwo metali kolorowych należy do najstarszych przejawów działalności gospodarczej w Górach Apuseni (Karpaty Południowo-Wschodnie). Są to góry wulkaniczne, w których występują złoża miedzi ołowiu i cynku. Obecność złóż złota i srebra jest związana z neogeńskimi erupcjami. Przestrzenne rozmieszczenie substancji mineralnych w skali prowincji Maramureş jest związane z głównymi strukturami uskokowymi uformowanymi pod wpływem tektoniki płytowej. Rzeźba terenu jest w znacznym stopniu przekształcona przez człowieka. Powstały liczne wały i hałdy kopalniane, na których działają procesy deflacji, osuwania, spłukiwania i erozji żłobinowej. W wielu miejscach zanikła naturalna sieć wodna, a wody powierzchniowe zawierają wielką ilość toksycznych zawiesin w postaci jonów Cu, Pb, Zn, Cd i innych.

W styczniu i marcu 2000 roku uległy zniszczeniu dwa sztuczne zbiorniki wodne w prowincji Maramureş, a rzeka Tisa — główny dopływ Dunaju, została skażona cyjanidami i metalami ciężkimi. Po tych wydarzeniach podjęto badania terenowe, których celem było określenie poziomu skażeń spowodowanych eksploatacją złóż. Jednym z głównych wniosków tego międzynarodowego projektu jest stwierdzenie, że prawie wszystkie źródła zanieczyszczeń w północno-zachodniej Rumunii, są związane z odpadami górniczymi wokół kopalń współcześnie prowadzących eksploatację i posiadających niewłaściwy system odprowadzania wód kopalnianych. Zanieczyszczenia są stwierdzane w wodach powierzchniowych i gruntowych, na odległość nie większą niż 5 km od źródła skażenia i w pasie o szerokości około 1 km w stosunku do współczesnego koryta.