

AN INTEGRATED APPROACH TOWARDS THE DEPOLLUTION OF THE APUSENI MOUNTAINS

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Abstract

Romania has a long-standing tradition of mining, particularly within the Apuseni mountains area. Unfortunately, there are various negative consequences which occur as a direct and indirect result of this process, such as acid mine drainage and heavy metal pollution of air, soils, surface and underground waters. This paper proposes an integrative and interdisciplinary strategy for the depollution of acid mine drainage and heavy metal contaminated areas in the Apuseni Mountains, Romania. We aim to view the reported problems within the designated area, which occur as a result of ore exploitation, from various perspectives (environmental science and engineering, medicine, veterinary medicine). Secondly we aim to provide a plan of action for depollution. Specifically we will focus on: the treatment of tailing ponds and surface waters, soil remediation, greening of heaps of debris, mine closure, stabilization and rehabilitation. We wish to focus on efficient, technologically easy to implement solutions, which preferably use natural, indigenous materials.

Key words: heavy-metals; acid mine drainage; toxicity, bioremediation, natural materials.

INTRODUCTION

Acid mine drainage (AMD) is recognized as the foremost significant problem associated with mining worldwide. Romania is a country with a long history of mining. A study estimates that more than half of the total assessed ore deposits of Romania occur in the East Carpathians and the Apuseni Mountains. Currently, Romania is still struggling with mine-related pollution, namely AMD, heavy metal and SO₂ pollution of air, soil and waters, which in turn are toxic for plants, animals and people (Toth and Quiquerez, 2006; Sima et al., 2011; European Commission, 2014).

This study focused on one of Romania's most polluted areas by the mining industry, the Apuseni Mountain range, with particular emphasis on Zlatna. The area affected by mine related pollution around Zlatna alone is around 55660 hectares (Lacatusu et al., 2009). Zlatna soils were found to be highly acidic as they are watered by acidic leachates (pH=3.7-4), and have a high heavy metal concentration (Williamson et al., 1998).

The mean annual emissions of SO₂ are 150450 tonnes and almost 3500 tonnes of

heavy metal filled dust (Lacatusu et al., 2009).

OBJECTIVES

The purpose of this review paper is to provide an integrative approach to the problem of AMD in Zlatna, and is divided as follows: short description of AMD, sulphur dioxide and heavy metal effects on soils, plants, animals and people, species that can be used for biomonitoring in order to assess the pollution levels over time, potential ways to treat AMD problems.

INTRODUCTION TO AMD

AMD is formed when material containing sulphide (generally FeS₂ containing rocks) is exposed to oxygen and water. Sulphur is released as sulphate (SO₄²⁻) which can dissolve to produce weak sulphuric acid, which in turn solubilizes heavy metals. This gives usually acidic, sulphur rich waters that greatly increase the solubility of heavy metals. Acidic water with heavy metal contaminants has serious negative effects on the plants, animals and soils in the contaminated

areas, which shall be discussed in the subsequent sections.

MAIN AMD POLLUTANTS

This section aims to introduce the main pollutants associated with AMD, namely SO_2 and heavy metals. General characteristics, the source of occurrence and the movement of these elements through the environment will be addressed, as well as how they are toxic to soils, plants, animals and people.

Anthropic SO_2 is produced as a result of ore mining, alongside heavy metals. Many species of plants are very sensitive to SO_2 concentrations. The main effect of the compound is the interference with the process of photosynthesis; it destroys the chlorophyll in the leaves (Malschi, 2014).

SO_2 particles combine with particles which are in suspension in the atmosphere, and form complexes which can transform this compound in H_2SO_4 and as such can

contribute to the formation of acid rain. When inhaled, a significant part remains in the respiratory system and is excreted or absorbed in the organism very slowly.

In people, in the case of short term exposure, the compound produces irritation of the respiratory system, blocking of the bronchi, increases the secretion of mucous and determines the constriction of the airways.

A high incidence of chronic bronchitis as a result of SO_2 has been noticed (Bartók and Crisan, 2007).

The increased mucus as well as the constriction phenomenon prevents the elimination of the solid particles from the lungs, which in turn determines chronic bronchitis. In the case of long term exposure, this compound has been linked to lung cancer (Ludusan, 2002).

Heavy metals naturally exist within the environment and are redistributed following various biological and geological cycles.

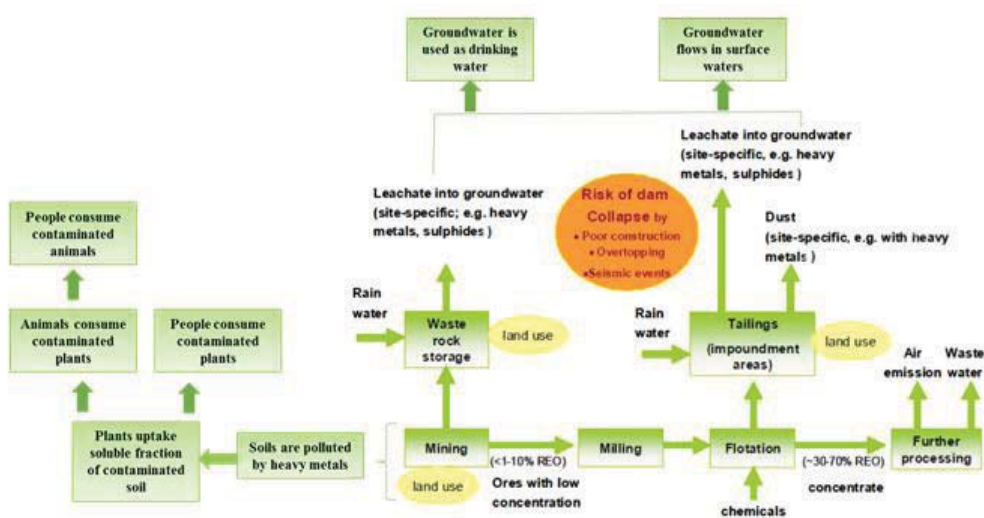


Figure 1. Heavy metals and sulphide movement within the environment in the context of mine exploitation (reproduced and altered from Massachusetts Institute of Technology, 2013)

One of the main anthropogenic sources for heavy metal pollution is mining. The accumulation of heavy metals in plants is influenced by the type of plant, soil conditions as well as the microorganisms present in the soil. These last two elements can shield the plants from the polluting agents, and thus be biological neutralization agents. The most crucial soil characteristics that are relevant in this context are: the thickness of the soil layer, the cationic and anionic exchange capacity (present especially in soils with high quantities of clay and organic matter), the biological activity of the soil, the precipitation of the ions from the soil solution

Some of the most commonly occurring heavy metals in the study area are presented in table 1.

Table 1

Table 1. Selected heavy metals from area of interest, their most common form in the environment and the Romanian maximum admitted limits

Heavy metal	Maximum admitted limits in Romania (HG 325/2005 NTPA 001)	Almasu Mare surface river samples	U.M.
Al	5.0	57	mg/L
Pb	200	15	µg/L
Cd	200	65	µg/L
Cu	100	190	µg/L
Fe	5.0	127	mg/L
Mn	1.0	106	mg/L
Zn	0.5	47	mg/L

* The samples were collected by the author of this paper in June 2013.

Lead (Pb) is toxic to the soil as it inhibits the enzymatic processes, it reduces the intensity of eliminating the carbon dioxide and it decreases the number of microorganisms as well as their metabolism (Ludusan, 2002). Lead's phytotoxicity consists of diminishing the uptake of micro and macronutrients, decreasing the transpiration rate and water content of the tissues and inducing oxidative stress in growing plant tissues (Sharma and Dubey, 2005). No biological need for Pb was found. In mammals, the most significant risk is posed to the nervous system. Lead can accumulate both in bones and

in soft tissues. Within the soft tissue, Pb tends to mostly accumulate in the liver and kidneys. Lead's main excretion pathway is through the kidney. Pb can also cross the placenta, hence can be transmitted from the mother to the foetus. Effects of exposure include peripheral or chronic neuropathy, hypertension, lead-induced anaemia, lead nephropathy (Goyer and Clarkson, 1996).

Cadmium (Cd) is a particularly dangerous heavy metal as it has high mobility and affects plants even when present in small amounts. Cd toxicity in plants is generally shown by a decreased growth and chlorosis (loss of the normal green leaf colour), due to its interference with Fe uptake interferes with the uptake, transport and use of Ca, Mg, P and K. Physiological and genetic damage were observed in onions, beans, peas and barely (Das et al., 1997). In people, chronic toxicity gives chronic obstructive pulmonary disease and emphysema as well as chronic renal tubular disease. Calcium metabolism is affected by Cd toxicity, thus inducing skeletal effects. These may include bone pain, osteoporosis and osteomalacia. Studies also linked Cd toxicity to essential hypertension, lung cancer and possibly prostate cancer (Goyer and Clarkson, 1996). Cd is excreted in urine.

Copper (Cu) polluted soils have a smaller aggregate number and a lower hydraulic stability which leads to increasing susceptibility to erosion and compaction. The biological activity from the soil is also weakened because an increased Cu concentration in the soil increases the mobile fraction of the humus, which in turn increases the hydrolytic acidity and decreases from the concentration of the basic cations. In plants, a high Cu concentration reduces the intensity of the respiration and slows down the process of producing chlorophyll. Plants which present high uptakes of Cu are herbaceous plants, vegetables, grape vines, trees and shrubs (Ludusan, 2002). Cu is an essential element for the body. Homeostatic mechanisms rule its gastrointestinal absorption. Ingestion of drinking water which has more than 3mg/L Cu produce gastrointestinal symptoms such as abdominal pain, diarrhoea, nausea and vomiting.

Symptoms of iron (Fe) toxicity in plants are brown roots and the appearance of brown spots on the surface of leaves. In the human body, Fe is regulated by homeostatic mechanisms, with 2-15% being absorbed from the GI tract. The greatest concentration of iron in the body, which occurs from chronic exposure, accumulates in cells of the liver and pancreas, in addition to the heart and the endocrine organs (Goyer and Clarkson, 1996).

In plants, symptoms of manganese (Mn) toxicity include brown spotting of a necrotic nature which occurs on stems, petioles and leaves (Reichman, 2002). In people, gastrointestinal absorption is less than 5% and occurs in the small intestine. Mn is mostly contained in the kidneys, liver, blood and fat. Chronic exposure gives high uric acid levels in urine and serum, slow growth, anaemia and diarrhoea (Goyer and Clarkson, 1996).

Zinc (Zn) reduces biological activity in the soil and tempers with the enzymatic activity of the microorganisms in the soil. It is easily absorbed by plants and mostly accumulates in the green parts of the plant. Symptoms of zinc toxicity are chlorosis and the reddening of young leaves (Reichman, 2002). Zn has relatively low toxicity to animals. Zn is an essential metal for biological systems. Acute zinc toxicity due to ingestion is not common and there is no hepatic, renal or hematologic toxicity that was found (Goyer and Clarkson, 1996).

In plants, aluminium (Al) interferes with the uptake as well as transport and efficiency of the utilisation of essential minerals (Rout et al., 2001). Very sensitive species to Al include beet, lettuce, mustard, cucumber, medium-sensitive species include pea, sunflower, potato, oat and less sensitive species are turnip, currant, cranberry, tea, corn and rye. Al from acidified waters is especially toxic for invertebrates as it replaces Ca ions from their bodies. In humans, chronic exposure to Al targets the lungs, the bones and the nervous system. Al competes with Ca metabolism in the body (Goyer and Clarkson, 1996). Its toxicity results mainly from replacing Mg^{2+} and Fe^{3+} by Al^{3+} , hence the afflictions have a cellular basis; cellular growth, intercellular communication and secretory functions are affected. Al is neurotoxic, inducing lesions in neurones. Intoxication with Al is reflected in the osseous

system by an unusual softness of the bones and a predisposition to bone fractures, while in the blood system anaemia appears (Barabasz et al., 2002.).

BIOACCUMULATION AND BIOMONITORING

Bioaccumulation is defined as “the biological sequestering of a substance at a higher concentration than that at which it occurs in the surrounding environment or medium” (US Geological Survey, 2007). Understanding this dynamic process of bioaccumulation is imperative for identifying and using biomonitors for pollutant assessment. There are several species which have been identified as biomonitors for a long time and are well adapted to a wide range of geographical conditions. Identifying these in the Zlatna area is of particular importance as they provide a means of comparing our levels of pollution to other countries’.

Biomonitoring uses biological responses to determine the changes in the environment. This process can use indicator species, such as macro invertebrates, fish, algae, protozoa, lichens, plants (Bailey and Stokes, 1985; Berkman et al., 1986; Caçadora et al., 2012). In the context of our study area, we are concerned mainly with fish, benthic macro invertebrates lichens and plants. Fish are good indicators of chronic effects spread over several years. Fish communities are indicative of a number of trophic levels, and are consumed by humans (North Carolina State University, 1995). They are easy to raise, have a long life cycle and are easy to handle. Biomonitoring using fish can be done in different ways; the most common one several decades ago was lethal testing of fish to measure the pollutants they contains, however studies are now shifting towards observing the behaviour response of fish, growth, metabolism, reproduction and fertility (Zhou et al., 2008). In Romania, the fish species *Chondrostoma nasus*, *Leuciscus cephalus* and *Phoxinus phoxinu* were used to monitor heavy metal pollution the Mures, Crisul Negru and Crisul Repede rivers respectively. Various pathologies and elevated levels of a number of heavy metals were found (Triebkorn et al., 2008; Petrovici and Pacioglu, 2010). Benthic

macro invertebrates have also been used as biomonitors for pollution, in various environments and geographical locations, producing a significant number of papers since the mid 1900's (Cairns Jr and Pratt, 1993). A study used insects, beetle larvae, and amphipods along with sediment samples to determine the heavy metal concentration in streams within the Abrud river catchments (Mates). Various plants which are susceptible to significant metal ion uptake can be used as biomonitors. In Romania, a study used four moss types to assess heavy metal pollution on more than 60% of the country's territory; target metals included Cd, Cu, Fe and Zn. The study found that Romania's median values were greater than most of the other European regions which had also done similar studies and that all the collection points showed concentration ranges above toxic level for humans. Various studies used lichens as biomonitors (Richardson et al., 1982; Haas et al., 1998). Lichens have also been used as monitors in the area of Zlatna, for SO₂, Pb, Zn, Cu and Cd levels. To be noted is that Zlatna was found to be so polluted that only some very resilient species of lichens were able to survive in that environment.

WATER TREATMENT

There are several methods to water treatment, which do not use a water treatment plant. For low quantities of water, a biosand filter can be used. These filters are cheap and technologically easy to build, and as such they are a viable solution for small, rural communities to procure the water they need for each day. Biosand filters reduce turbidity and colour of the wastewater and remove chemical contaminants and microorganisms through a single filtration process (Muhammad, 1997). The downside is that this process cannot clean up an entire river; a typical flow rate is limited to 15-20 L hr⁻¹ but can be slower depending on the height of the water column on the filter top and the size and type of the filter components. Treatment usually involves diverting a part of the river to the filter, or manually pouring water on the filter top, which is why this treatment is more suitable for individual households. A South African study has developed a cost-

effective biosand filter (total cost of < USD 20) using the natural zeolite clinoptilolite (usually found in volcanic ash). The filter was comprised of four areas which played a part in filtration; the first layer after the standing water zone was the biological layer (sediments, slime and micro-organisms), the second one was the biological zone, which occurred in a fine sand layer (5-10 cm from the surface of sand and removed Fe and microbial contaminants), the third one was the zeolite layer which removed the majority of the heavy metals and the last one was the gravel zone which prevented the zeolite layer from being washed down (Figure 2). Results obtained after 1 hour of filtration showed a removal of 80% Ca, 89% Mg, 99% Fe, 56% As (Mahlangu et al., 2011).

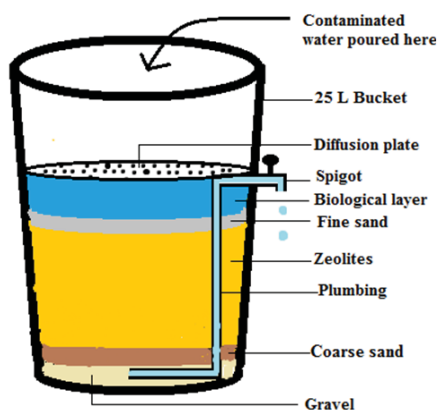


Figure 2. General sketch of bucket zeolite-enriched biosand filter (Mahlangu et al., 2011)

Other studies using simple biosand filters (no added zeolites) confirmed its potential of heavy metal removal (Tang et al., 2010).

Various other low-cost materials have been explored for the purpose of AMD remediation. Zeolites are naturally occurring aluminosilicates with a cage-like structure which contains loosely bound non-toxic metallic cations (usually Na⁺, K⁺ and Ca²⁺) which can be exchanged with heavy metal ions from the environment and as such have been successfully used for the treatment of acid mine drainage.

Romania has important natural volcanic tuff resources. (Bedelean, 2010). Extensive research has been done on various heavy metal removals with natural zeolite; tests included various pH,

zeolite type, experimental procedures (mostly batch or columns) and all of them proved the efficiency of the material in this context (Ouki and Kavannagh, 1999; Erdem et al., 2004; Al-Anber and Al-Anber, 2008). Bentonite clay is a type of aluminium phyllosilicate which is an adsorbent. It has a similar sorption capacity to zeolites, it attracts metals because it has various cations and anions on its surface and as such can ion-exchange them with the metal ions or adsorbs the ions onto its structure, and as such has been used in the context of AMD remediation.

Biomass fly ash is a ferro-alumino silicate with a variable fraction of oxides; for AMD treatment, calcareous FA (10%CaO) is needed, which has a typical pH=9-12.

Biomass ash represents about 19% of Romania's primary energetic potential hence biomass ash is a cheap, readily available resource Romania. Numerous authors have studied coal ash as an adsorbent for heavy metals, in various types of contaminated water, at a range of different pH, temperature and contact time (Panday et al., 1984; Barakat, 2011; Vadapalli et al., 2012).

For accumulation ponds, biosorption by various plants can also be used. Types of brown marine algae, fungi, biomass of order Mucorales fungi, chitin and chitosan (from fishery wastes) represent good biosorbents (Volesky and Holan, 1995; Ng et al., 2002).

SOIL TREATMENT

Soil treatment is more difficult to achieve than water treatment and more difficult to implement technologically. In the case of the Zlatna area, the best approach would probably be a passive type of treatment, using a combination of biosorption and phytoremediation methods, especially designed for the geophysical conditions of the area, as opposed to active treatments. Phytoremediation is a long-term way of remediation, but represents a more sturdy and technologically easily to implement and to maintain way of soil treatment. Phytoremediation involves the usage of plants, algae, microorganisms from the soil as well as biomass for the remediation process; this includes phytostabilisation and phytoextraction. Phytostabilisation involves

decreasing from the mobility or immobilising the contaminants from the soil by using plants for hydraulic control. Addition of high zeolite content volcanic ash has also been used (Damian and Damian, 2007; Malschi, 2014). Phytoextraction involves the accumulation of heavy metals within plants or algae which have a high tolerance to these pollutants.

Plants with high potential that have been studied are: forest products (wood, logging, shrubs and wood residues, sawdust etc.), energy crops (grasses, starch, forage, herbaceous woody or oilseed crops), aquatic plants (water weeds or hyacinth, algae, reed and rushes), wetland plants (brass buttons, duckweed, umbrella plant, smartweed, water lettuce) (Goodrich-Mahoney, 2001; Ciubotaru-Rosie et al., 2008).

There are also certain types of metal-binding algae and lichens which effectively accumulate heavy metals and which have been addressed in the bioaccumulation and biomonitoring section.

Biosorption of heavy metals can also be done on various types of waste by-products from agricultural production and processing, crop residues, urban wood and organic wastes. Phytoremediation is usually done by constructing a wetland (Brenner, 2001).

TREATMENT FOR PEOPLE

In people, blood, urine and hair are used as indicator tissues for the measurement of heavy metal exposure. Urine and blood show recent exposure. For certain metals, such as Hg, hair analysis can provide a good long term indication of exposure by comparing samples from different portions of the hair segment. The valence state and the ligand binding of the metals are also very important factors which relate to toxicity. In terms of chemical bonding, organometallic and inorganic forms of the metals behave very differently. The most popular method of chronic exposure to heavy metals' treatment in people and animals is chelation therapy.

Chelators are agents which bind to toxic metal ions, thus forming metal complexes which can be excreted by the body from intra or extracellular spaces, primarily through urine.

In terms of the characteristics it exhibits, a good chelator should have a high affinity for

the toxic metal ions, low toxicity, a high capacity to compete with endogenous chelating agents, the capacity to penetrate cell membranes, high solubility in aqueous media, the capability of forming non-toxic complex and it should have the same distribution as the metals it is aimed for. Another aspect of chelating agents that should be regarded is the type of complexes they form.

The structure of the ligand may reduce toxicity in the local *in vivo* environment by forming a closed complex, thus shielding biological targets. Similarly, there are certain chelators that can expose the metal more to the biological environment, thus increasing its toxicity. Care should be taken when administering chelating ligands since they can bind to useful metals in the body as well and as such can have very serious side effects such as kidney damage, stomach and intestinal bleeding, cardiac problems, seizures and can even result in death (Lowry, 2010; Medline Plus, 2010).

An alternative to chelating agents, with significantly less adverse effects are zeolites.

Several studies have shown their efficiency in the removal of heavy metals from the body through urinary excretion (Papaioannou et al., 2005; Karampahtsis, 2012).

CONCLUSIONS

Zlatna is still a particularly AMD affected area of Romania with very severe levels of SO₂ and heavy metal pollution. Of even more concern is the fact that it is an example of many former or current mining sites in Romania, particularly in the Apuseni Mountains. Even though there have been some attempts at mine closures over the years, as well as a multitude of both national and international fundamental research and even pilot studies, action has not been taken to treat the affected soils and water and alleviate people and animals which have been affected by chronic exposure to these pollutants. The author recognizes the limitations imposed by government spending and funding allocation on these types of environmental issues and by the large surface area and number of geographic locations which need to be treated, and as such the author proposes that the Zlatna area treatment strategy should focus on low-cost, technologically easy

to implement, long-term, *in situ* treatments of river and creek waters, tailings ponds and soils. Regarding the people living in pollution hot-zones, a medical examination should be given to each resident and a database should be constructed with the assessment of each patient. Seeing as conventional therapies which involve chelating ligands are costly and can be dangerous for the patients, a decision should be made for each patient regarding the severity of the heavy metal intoxication. Domestic animals, particularly animals which are consumed or which give products which are consumed by people should be tested for heavy metal contamination. Even if there aren't a high number of studies attesting the efficiency of zeolites in treating heavy metal, there is enough evidence to suggest that it represents a feasible treatment, and as such a pilot study concerning heavy metal treatment with zeolites should be conducted in Zlatna as well.

Treatment of water and soils should be done simultaneously with the above-mentioned treatments in order to prevent further exposure of people and animals to the same high pollutant levels.

The author proposes an environmental assessment study be done in the Zlatna area, and preferably include updates of previous studies in order to potentially find a pattern of pollution evolution with time or cross-reference the results with previous data. Based on the particularities of the surveyed area, small scale *in situ* treatment facilities should be designed and implemented along AMD affected creeks, rivers and in tailings ponds, based on low-cost, indigenous materials such as zeolites, fly ash, clay, waste biomass.

Based on literature studies, plant species with a high potential of heavy metal uptake should be identified in the area and a wetland containing those indigenous species should be designed and constructed.

A pilot study should be done by using zeolites volcanic tuff, betonies and other natural absorbing materials and mixing it with soil on patches of land which can be used for agricultural purposes.

Biomonitoring species, including fish, lichens and benthic macro invertebrates should be collected from the affected area in order to build a database for the assessment of pollutant

levels. This is essential for monitoring the efficiency of the treatments, both on a short term and a long term.

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