

ENVIRONMENTAL AND HUMAN HEALTH RISKS ASSOCIATED WITH THE CENTRAL TAILING POND IN THE EAST OF BAIA MARE CITY, ROMANIA

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Abstract

The paper highlights the risks associated with the mining waste in the Central tailing pond located in the east side of Baia Mare City. This tailing pond is made of sterile resulted from the processing of gold-bearing ores originated from the Șuior, Căvnic, Herja and Turț mining exploitations. These mining wastes contain major heavy metals concentrations (Pb, Cu, As, Cd, Mn, Fe, Zn etc) to which those from the arsenious pyrite concentrates deposited on the tailing pond surface are added. The data resulted from the chemical determination of the concentrations of heavy metals in the wastewater collected from tailing pond ditch have revealed exceedances of the values of the 4th surface water quality class.

The risks associated with the Central tailing pond are both due to the phenomena of pollutants dispersion by runoff or deflation and to the some technical-constructive features of the pond. A particularity posing a high risk degree is the presence on the tailing pond of the arsenious pyrite concentrates which were biologically reactivated by bacterial leaching.

Keywords: risk, tailing pond, mining, heavy metals, arsenious pyrite

1. Introduction

Baia Mare City is recognized as an area with significant mineral resources, especially with important polymetallic gold - silver deposits, containing Pb, Zn, Au, Ag, Cu, Sb, etc (Grancea et al., 2002). The mineralization is typically veins and belongs genetically to the tertiary volcanism. There are over 650 veins and among them, the vein Cremenea – Suior, formed inside a column of explosion, has the most important thickness, reaching up to 50 m (Tamaș, 2001). The richest gold – silver structures are represented by the metallogenesis of Săsar, Dealul Crucii, Valea Roșie sectors (Marius, 2005). Thus the metallogenetic structure of Baia Sprie and E.M. Suior mining sector are represented by epithermal deposits of polymetallic ores (Vágó and Binelly, 2006).

Development of the mining activity in Maramures County has led to the emergence of more than 300 dumps of sterile and 17 ponds, of which just one is still active however with storage activity temporarily suspended by authorities (Vasilescu et al., 2012). Three of these ponds are placed in the eastern part of Baia Mare City: the Central tailing pond, Baia Sprie tailing pond and Tăuții de Sus tailing pond. At present, the Central tailing pond poses a risk both to the environment and the human health due to the metals content and some construction issues. Furthermore, in the period 2004-2005, on this tailing pond have been

stored about 110,000 tons of arsenious pyrite concentrates which was reactivated by a bioleaching process without parameters to ensure the security of the site. Thus, the particularity of the Central tailing pond is given by the presence of the arsenious pyrite concentrations otherwise called the "Șuior pyrite". These arsenious pyrite concentrations represent a raw material which by technological considerations regarding the pretious metals extraction had not been processed and therefore was stored on the old tailing ponds area belonging to Baia Sprie and Tăuții de Sus area.

The study aims to highlight the risks for the safety of the environment and the population health generated by the presence of arsenious pyrite concentrates in the Central tailings pond area and the risk mitigation measures, respectively.

2. Materials and method

2.1 The characteristics of the Central tailing pond

The Central tailing pond is a plane pond located in the south part of the Tăuții de Sus village, at about. 6 km downstream of Baia Sprie town and cca. 3 km upstream, in the north-east of Baia Mare City. Administratively it belongs to Baia Sprie town (Romaltyn Mining Ltd., 2014b). In the north, the Central tailing pond is bordered by vacant land, some households belonging to Tăuții de Sus village and Racoș stream. In the west-north west there is the former Flotația Centrală Ores Processing Plant and in west side there is a

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penitentiary. To the south, the tailing pond is bordered by a grazing land and in the east with EM. Baia Sprie (fig. 1) (Romaltyn Mining Ltd., 2014b).

The Central tailing pond is situated between Racoș stream (on the north side at about 250 m far away from the pond) and Craica stream (on the South side at approx. 340 m far away from the pond).

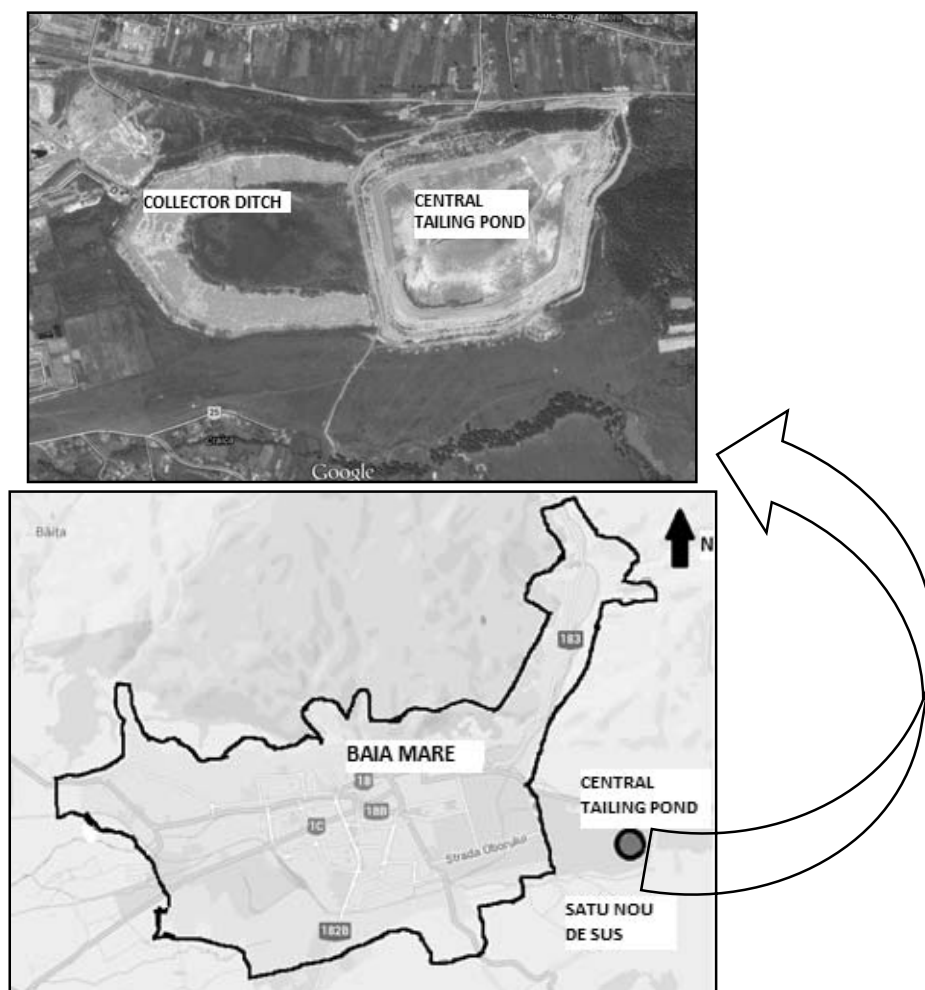


Fig. 1 The location of the Central tailing pond (<https://www.google.ro/maps>)

Geologically, the location area of the Central tailing pond belongs to the eruptive Neocene and consists of different types of andesitic formations, covering and crossing Paleocene and Neocene. Also, the area is characterized by the presence of eruptive rocks derived from Neocene volcanic activity which caused andesitic volcanic lava effusion and lower deposits of pyroclastics complex. Impermeable rocks without deep aquifers are predominant and allow sporadic movement of precipitation water (Liteanu et al., 1969).

To a better understanding of the geological structure, 8 geotechnical drillings were made before 1976 which intercepted the following formations: vegetal soil (0 to 0.2 m), clay yellowish brown with rust and gray alternate (0.2 to 1, 6 (2.3)), cobble, gravel and sandy clay mass (1.6 (2.3) - 4.2 (5));

gray marl (4.2 (5) - 5.5) (Romaltyn Mining Ltd., 2014c)

The Central tailing pond has 48 ha decanting surface at starter dam level and the current surface ones at the superior platform level of 38 ha. Currently the tailing dam altitude is between 293 and 293.8 mdMN and ground elevation at the base of the dam priming is between minimum 271 and maximum 289.6 mdMN mdMN. The pond slope angle is between 17° and 19° . Compared to the minimum level of basic land, the dam height is 22.80 m and compared to maximum level, the dam height is 4.20 m (Romaltyn Mining Ltd., 2014c).

The hydro pipelines, the pumping station etc have been dismantled over the years. Extension of successive dikes because of erosion have merged to form a single sloping embankment on the large majority of the outer surface.

The Central tailings pond is a less consolidated material such clay, sandy clay dusts, sands and clay sands. In the embankment and the canopy them material has a predominantly a sandy-clay texture which become finer and finer with the

advance to the inside of the pond (Romaltyn Mining Ltd., 2014b).

Physical and mechanical properties of flotation sterile samples are indicated in table 1.

Table 1 *Physical and mechanical properties of the Central tailings pond (Romaltyn Mining Ltd., 2014a)*

No.	Specification	unit	Value
1	Natural humidity	%	39,6 - 46,8
2	Apparent specific weight of the material in the pond	g/cm ³	1,8 - 1,9
3	Specific weight (mineral skeleton) of the material in the pond	g/cm ³	2,53 - 2,65
4	Porosity	%	50,0 - 54,0
5	Pore index		1,00 - 1,17
6	Permeability	cm/s	1,5 - 5,8 x 10 ⁻⁴
7	Angle of internal friction	grade	17 - 23
8	Cohesion	kPa	21,0 - 25,0

2.2 Methods of the risk evaluation

For risk assessment of the Central tailing pond was considered both the metal concentrations in the tailing pond and those in the arsenious pyrite concentrates which was deposited on the tailing pond. Observations were made for a period of seven years and were based both on the field visits and laboratory analyses.

The samples were taken monthly according to SR ISO 5667-10/1994 of ditches drains the water area depth allow harvesting under the water level. The effluent was taken in dry, clean and rinsed polyethylene containers. For keeping the samples safe, the containers were sealed. For the analysis of the heavy metals ions, samples were acidified at pH=1 (with 5 ml nitric acid 65% of AP/1 l sample). Determination of metals content was done with an atomic absorption in flame spectrophotometer (ContraAA 700 Analytik Jena). The method was applied as follows: for copper, zinc, cadmium and lead according to SR ISO 8288/2001 for manganese 8662 - according to SR 2/1997, for arsenic

according to EN ISO 11969/1996 and for iron, according to SR 13315/1996. Sulfates were determined by gravimetric method according to STAS 8601/70, using the calciner NABERTHERM type L 5/11.

Both sampling and chemical analysis were carried out by the Romaltyn Mining Ltd.

3. Results and discussions

3.1 Contents of heavy metals and compounds in the Central tailing pond

The Central tailing pond served to accumulate the sterile resulted from Flotația Centrală Ores Processing Plant, between 1962 -1976. Since 1976 the tailing pond has been in conservation (Romaltyn Mining Ltd., 2014c). The amount of deposited sterile on the tailing pond is approx. 10 million tons and it resulted from complex and gold ores processing from Șuior, Căvnic Herja and Turț mines (Romaltyn Mining Ltd., 2014c).

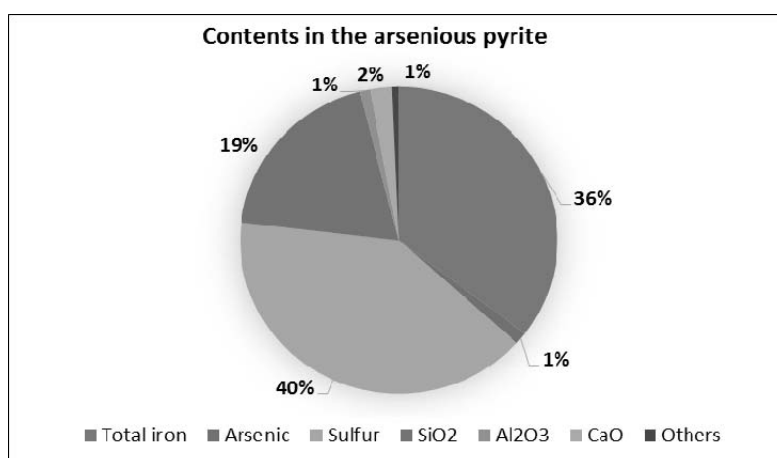


Fig. 2 *Average content of chemical elements in the arsenious pyrite concentrate (Romaltyn Mining Ltd, 2014)*

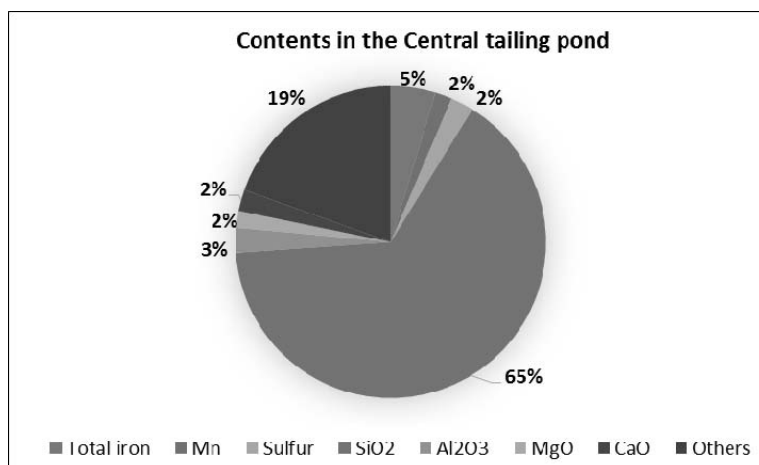


Fig. 3 Average content of chemical elements in the sterile of Central tailing pond (Romaltyn Mining Ltd, 2014)

In the period 1982-1983 the quality of Șuior exploited ores did not correspond to the requirements for pyro-metallurgical processing, the arsenious pyrite concentrates in the amount of approx. 500,000 tons were stored near Central tailing pond, in the north-west.

After the cessation of storage activity on the tailing, the period 2004-2005, a quantity of about. 110,000 tonnes of material from the arsenious pyrite deposit was transferred on the upper of the pond. Approximately 30,000 tons of this amount have been processed immediately after storage. The arsenious pyrite remained on the pond (about 80,000 tons) has been subjected to the bacterial bioleaching process (Romaltyn Mining Ltd., 2014a). The golden arsenious pyrite from Șuior has the following contents: Au 10-12 g/t; Ag 50-70 g/t; As 1,5-2,0%; 10% SiO₂; Cu 0.15-0.20%; Zn 0,60-1,20%; Pb 0.50-0.60% (Oros, 1999).

The average element contents of the Central tailing pond are presented in figures 2 and 3.

'Others' in figure 2 means the following elements content MgO 0.14%; Cu 0.035%; Pb 0.33%; Zn 0.146%; Mn 0.026%, and Cd 0.028%, respectively.

The category "others" from figure 3 includes a number of elements with a lower share, as the following: Ca 0.327%; Cu 0.029%; Pb 0.051%; Zn 0.066%; As 0.019% and Cd 0.044%, respectively. In addition the Central tailing pond may contain, in minor amounts, elements like: Ga (10-60 ppm), Cr (50-100 ppm), Sn (10-50 ppm), Ni (10-100 ppm), Mo (10-100 ppm), V (30-100 ppm) (Romaltyn Mining Ltd, 2014).

3.2 Bacterial bio treatment of arsenious pyrite

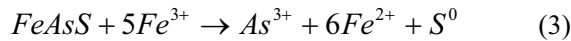
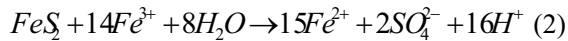
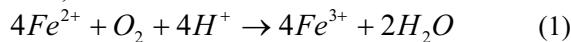
Arsenious pyrite deposited on the Central tailing pond, is a refractory sulfide requiring a special technology for separating fine gold disseminated in composition. Applying the classical

processing methods involves negative impact on the environment. For example, when roasting arsenious pyrites, the arsenic combustion compounds pollute the atmosphere (Oros, 1999). The chemistry of arsenious pyrite oxidation in acid environment has also been revealed in several scientific papers belonging to Papangelakis and Demopoulos (1990) and Fernandez (1992).

To facilitate the precious metals extraction from the arsenious pyrite concentrate deposited on the surface of the Central tailing pond, an experiment in situ was conducted. It supposed to contact the pyrite piles with a bacterial solution containing ferro-oxidizing bacteria, as: *Thiobacillus ferro-oxidans* and *Leptospirillum ferro-oxidans*. Thus, the preferential oxidation of arsenious pyrite occurred and facilitated the release of fine gold particles, leading to better contacting with cyanide reagents (Oros, 1999). At the end of the treatment by bacterial leaching, the resulting material included both unaltered arsenopyrite (sulfide) and As³⁺/As⁵⁺ soluble (Romaltyn Mining Ltd., 2014a).

The oxidation of the mineral sulfides and implicitly of the arsenopyrite may be achieved both directly when the bacteria adhere to the mineral and oxidize the sulfur, and indirectly when the iron is oxidized from the bivalent iron to the trivalent iron. (Oros, 1999). The leaching process is conditioned by several factors such as: the oxygen concentration, the carbon dioxide concentration, the physiological state and the concentration of the bacteria, the pH (2.5-3.0) or the ambient temperature (25-35°C) (Oros, 1999). Therefore, these conditions may be achieved when using the reactors or bioreactors. This method is not recommended to be applied in situ (bulk or dump) because the percolation of the fine material by the oxidizing solution is difficult and the oxygen intake not sufficient for the bacterial leaching process. (Oros, 1999).

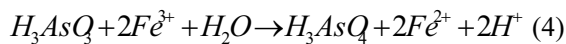
The use of bacteria for the mineral sulfide treatment contributes to continuous oxidizing of the bivalent iron to trivalent iron which in its turn has the role to oxidize the mineral sulfide. (Breed et al. 2000)



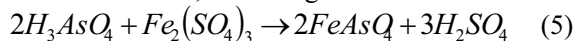
The bivalent iron resulted after the mineral sulfide oxidizing is oxidized again by the bacteria and this process is cyclical.

Poly-sulfides and sulfur or thio-sulfate produced by ferric leaching are oxidized to sulfate by the bacterial oxidizing of the sulfur. (Breed et al. 2000). By bacterial oxidizing the arsenic and iron leaches to arsenite As^{3+} and ferrous ion Fe^{2+} .

Bioleaching process may be followed by oxidation of arsenite to arsenate:



Oxidation of arsenite (AsO_2^-) to arsenate (AsO_4^{3-}) can be, in turn, followed by precipitation of ferric arsenate, according to:



4. Risk particularities of the Central tailing pond

Even though the Central tailing pond is a common tailing pond for storage wastes of non-ferrous ores industry in Maramureş County, the difference is due to the arsenious pyrite concentrates which were deposited on the tailing pond. Moreover, these concentrates were biologically activated in situ by applying a bioleaching treatment to oxidize the sulfide minerals. For some economic reasons the sterile has not been reprocessed and the concentrates remained on the deposit for more than 10 years.

The monitoring of the water quality from the collector ditch of seepage water and runoff of the pond revealed for the period 2007-2013 exceedences of the maximum admitted limits for surface waters for almost all analyzed metals and for all considered years. The low pH value of 4.3 of the collected water shows its acidic feature which facilitate the heavy metals dissolution. Thus, for copper (fig. 4) the 4th water quality value was exceeded for about 22 times by the lowest recorded value, and about 93 times by the maximum recorded value for copper during 2007-2013 (Order no. 161/2006). For zinc (fig. 5) the value related to 4th water quality class was exceeded of about 5 times by the lowest recorded value, and of about 62 times by the highest value recorded for the period 2007-2013 (Order no. 161/2006).

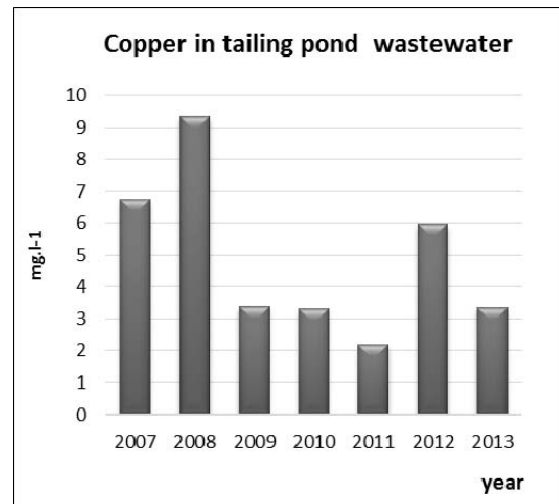


Fig. 4 Copper ions in tailing pond wastewater

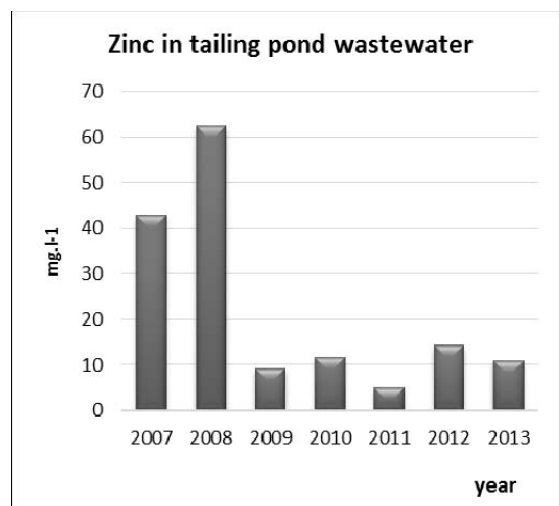


Fig. 5 Zinc ions in tailing pond wastewater

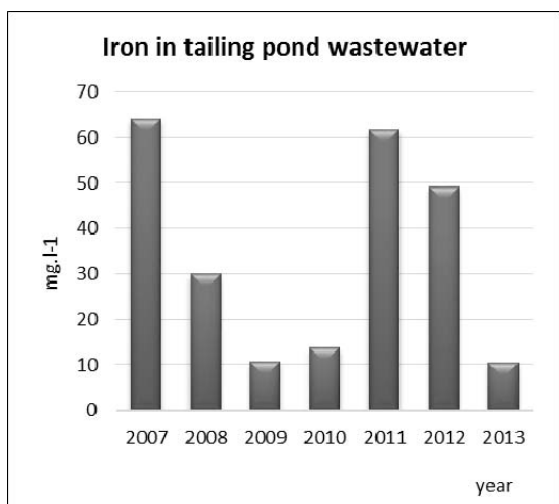


Fig. 6 Iron ions in tailing pond wastewater

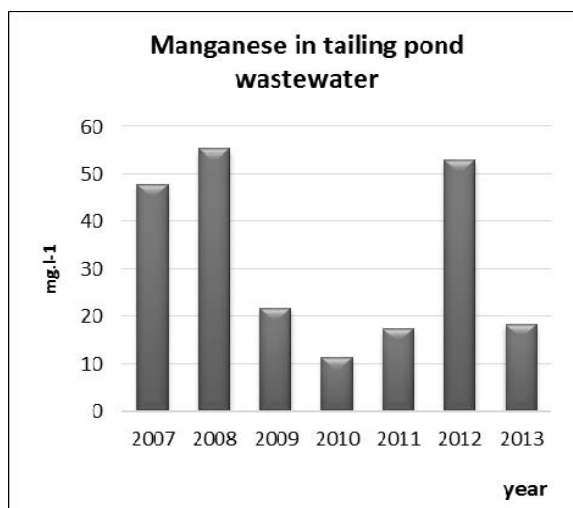


Fig. 7 Manganese ions in tailing pond wastewater

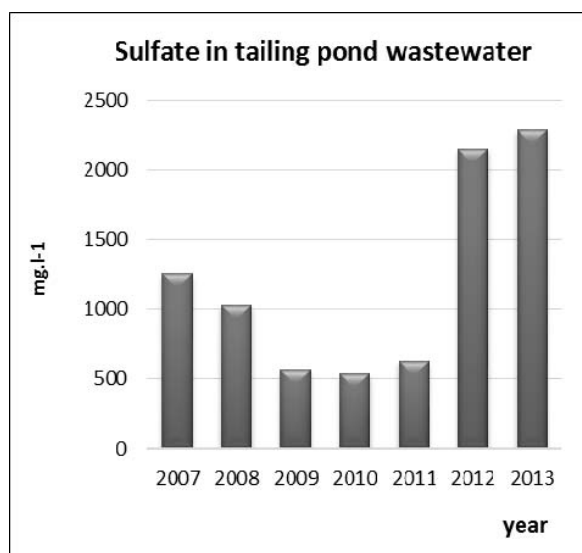


Fig. 8 Sulphate ions in tailing pond wastewater

The iron concentrations (fig. 6) in the collected effluent revealed for the period 2007-2013 exceedances of the 4th water quality class of approx. 5 times by the lowest value recorded and of approx. 32 times by the highest iron concentration value (Order no. 161/2006). The manganese concentration (fig. 7) related to 4th water quality class was exceeded in 2007-2013 of approximately 11 times by the lowest recorded value and about 55 times by the highest value of the manganese concentration (Order no. 161/2006).

The lowest concentration of sulfate ions (fig. 8) was recorded in 2010 and exceeded the 4th water quality class of approx. 2 times and the highest value exceeded this limit of approx. 8 times in 2013 (Order no. 161/2006).

The lead concentrations could not be determined because they were below the limit of detection of the atomic absorption spectrophotometer.

The annual average of arsenic concentrations of 0.0141 mg.l⁻¹ recorded for the collected wastewater include the collected water quality in the 5th water quality class (Order no. 161/2006).

In order to achieve an overall assessment of the annual amount of pollutant concentrations generated by seepage and runoff of the Central tailing pond it was considered the average of the 7 years of observation, respectively the constructive and technical characteristics of the pond (effective surface area of 38 ha). Also, to calculate the concentrations, it was taken into account the evaporation coefficient of 10%, and the average annual rainfall for the Baia Mare City of 935.5 l/m²/year.

According to the contents of the elements in the layer of pyrite concentrates or in tailings body (fig. 2, 3) resulted the amounts indicated in table 2.

Table. 2 Chemical contents in the Central tailing pond

Crt. No	Element	Sterile from Central tailing pond	Concentrate of arsenious pyrite	Total
			(tons)	
1	Total iron	463400	28320	491720
2	Arsenic	1900	1024	2924
3	Sulfur	238000	32800	270800
4	SiO ₂	6500000	15200	6515200
5	Al ₂ O ₃	258900	856	259756
6	MgO	171700	112.8	171813
7	CaO	218600	1808	220408
8	Cu	2900	28	2928
9	Pb	510000	264	510264
10	Zn	6600	116.8	6716.8
11	Mn	176700	20.8	176721
12	Cd	4400	22.4	4422.4

Thus, for an area of 38 ha, the following average amount of pollutants enter the environment annual by seepage and runoff: 1568.124 kg Cu; 7132.228 kg Zn; 10957.990 kg Fe; 4.512 kg As; 10303.992 kg Mn; 135.663 kg Cd (based on three years average) and 386593.910 kg SO₄, respectively.

5. Human health risks associated with the pollutants of the Central tailing pond

Because the slopes and the platform of the Central tailing pond are covered in a small proportion with vegetation, especially on the side oriented to Satu Nou de Sus village, the heavy rains

and deflation phenomena due to air currents are sources of risk for human health and biodiversity of the surrounding area. These solid particles have an important heavy metals content from which some are very toxic (Cd, As, Pb etc).

Because in the south south-west part of the tailing pond, at approx. 200 m, (fig. 9) exists a pasture, it poses a high risk of contamination of the human food chain with toxic elements from the tailings pond (now the pasture is a source of food for the cows from Satu Nou de Sus village). Less than 800 m in the south of the tailing pond there are private households which exposes the local population at extremely high risk of contamination with heavy metals from the tailing pond content.



Fig. 9 The pasture near the Central tailing pond (photo: Irina Smical)

For assessing the risk posed by the heavy metals to the health safe a series of investigations and toxicological test have been conducted (Martínez-Sánchez et al., 2013, Martin et al., 2013). The routes of population exposure to the heavy metal toxicity from the Central tailing pond are: inhalation (particles entrained by air currents), ingestion through consumption of contaminated food and water and dermal contact. To humans the major route for copper penetration in the body is food intake rather than inhalation. Although at normal levels the copper is an essential body element, in the case of exposure to high concentrations of copper acute symptoms appear such as nausea and metallic taste in the mouth, vomiting and digestive ulcers or lesions in the lungs by inhalation. The minimum risk level for intermediate and acute exposure by ingestion of human body for copper is 0.01 mg/kg/day (ATSDR, 2013).

As copper, zinc is an essential trace element for the organism, but when it enters the body in high concentrations then produces poisoning. Toxicity of this metal is enhanced by the presence of other metals such as copper and cadmium. Symptoms of acute exposure to high concentrations of zinc in humans are manifested especially by fever, sweating, and weakness (Oros, 2011).

The minimum concentration of zinc which poses risk by acute or chronic ingestion exposure is 0.3 mg/kg/day (ATSDR, 2013).

Regardless the route of the lead penetration into the body, it primarily affects the nervous system with effects on cognitive development, especially to children. A common disease in chronic exposure to lead poisoning is saturnine anemia or lead poisoning. Lead is deposited mainly in bones and also in the liver, kidneys and tissues. Symptoms of lead poisoning are manifested by: gastrointestinal syndrome (anorexia, nausea,

abdominal pain, etc.), nervous syndrome (headache, fatigue, irritability, encephalopathy, etc.) pseudo rheumatic syndrome (arthralgia, myalgia), anemic syndrome, renal syndrome etc (Oros, 2011; ATSDR, 2007a). The minimum lead concentration in blood that poses risk for health by ingestion is in the range of 0-1 mg/dl (Prüss-Üstün et al., 2004).

Cadmium ingested or inhaled is stored mainly in the liver and kidney in the form of metallothionein which is partly responsible for the renal toxicity (Ursinyova and Hladiova, 2000). After long term exposure to cadmium pollution, it accumulates in the pancreas and adrenal glands (ATSDR, 2012). Minimum risk level for acute exposure to cadmium is posed by inhalation of 0.00003 mg/m³, respectively 0.00001 mg/m³ for chronic exposure (ATSDR, 2013).

A feature that specifically distinguishes the Central tailing pond from other mining waste deposits in Maramures County is the arsenious pyrite concentrate placed on its surface.

This method of sulfide minerals oxidation is widely used worldwide and attracts researchers growing interest in order to improve it (Park et al., 2014). In this regard, the literature offers a wide range of concerns regarding the influence of arsenic and its associated environmental risks in relation with mining activity (Kwon et al., 2012; Yurkevich et al., 2012; Larios et al., 2012; García-Sánchez et al., 2010; Chakraborti et al., 2013). A particular attention is given to the impact of this metal in the food chain (Bundschuh et al., 2012a; Otones et al., 2011) and aquatic biocenosis (Magellan et al., 2014, Rahman et al., 2012).

Researches have revealed a higher toxicity of arsenite (AsIII) than that of arsenate (AsV) (ATSDR, 2007b).

The presence of arsenic in the human body causes a number of diseases such as bladder and lung cancer, cardiovascular problems, cognitive disorders to children, kidney, liver and skin cancer (Bundschuh et al., 2012b). The contamination of human body by inhalation of arsenate leads in the most cases to lung cancer (ATSDR, 2007b). The minimum level of risk to the human body by acute exposure to arsenic is posed by the ingestion of 0.005 mg/kg/day, respectively 0.0003 mg/kg/day for chronic exposure (ATSDR, 2013).

According to the data of Fig. 2 and 3 the Central tailing pond contains SiO₂ in a significant percentage (19% in arsenious pyrite deposit and 65% in the tailings pond). Insufficient vegetal stabilization of the Central tailing pond embankments (fig. 10) facilitated the occurrence of the runoff grooves and deflation phenomena of the stored material. These particles charged with heavy metals get on the surface water, or in the surroundings inhabitants lungs.

These fine particles made of crystalline silica are particularly dangerous to human health because their penetration into alveoli leads to atrophy of their walls and thus their inability to perform gaseous exchange with the air. The consequences are manifested by the appearance of quasi-permanent cyanosis of the extremities of the body, followed by a quasi-general cyanosis which will culminate, in case of prolonged exposure, with death. The silica entering in the lungs causes sclerosis by tissue retracting around quartz grain leading to the disease known as silicosis (Bud, 2006).



Fig. 10 The south slope of the Central tailing ponds (photo Irina Smical)

6. Other risks concerning the safety of the Central tailing pond

From seismic point of view, in the north west of Baia Mare City, were identified areas with potential of producing earthquakes at normal depths (below 60 km) and intermediate (60-180 km) (Ardelean et al., 2005). Earthquakes produced in such areas are moderate having low energy, and occurs at important intervals of time, over a century. These earthquakes are felt in some small areas of hundreds of square kilometers (Romaltyn Mining Ltd., 2014b).

In consideration of the technical and construction characteristics of the Central tailing pond, it can be said that the dynamic loads can cause landslides in the tailing pond embankment due to the liquefaction phenomenon of the submerged material found in the pond body. Also, correlated with the accident on 10th november 1971, it is possible that in case of heavy rains both runoff and overflow to occur on the slope and the starter dam.

In the frame of weather-climate events related to geo-technical structure of the tailing pond bedrock, there is a possibility of landslides as happened in 1976 when the foundation soil ceded together with partial displacement of dam (made of clayey soil) (Romaltyn Mining Ltd., 2014).

According to STAS 4273/83, the Central tailing pond fall within Class of importance IV "hydraulic structures whose damage has an impact on other socio-economic objectives" (Safety Report, 2012).

7. Conclusions

The Central tailing pond is a deposit which includes approx. 10 million tons of waste from the processing complex and gold ores from mines belonging to Baia Mare mining basin. In this tailing pond approx. 110,000 tons of arsenious pyrites were stored of which approx. 30,000 tons were processed shortly after storage. Both the sterile and especially the arsenious pyrite concentrates have high contents of pollutants in the form of oxides or heavy metal ions (Cu, Pb, Zn, Cd, As, Mn, Fe etc.).

Observations conducted during 7 years (2007-2013) on water quality in the tailing pond collector ditch revealed exceedances of 4th class values of the surface water quality for all of the investigated elements.

A feature of this tailing pond is the deposit of the arsenious pyrite treated with bacterial leaching. These pyrites have high heavy metals content and especially arsenic and represent a high risk factor for the quality of the environment and human health. The solid particles charged with heavy metals present a high risk of contamination of the

water and air. The amount of crystalline silica both in sterile and arsenious pyrite is important. This fact correlated with an insufficient vegetative stabilization of the tailing dam slopes generates significant risk to harm human health from surroundings through various pulmonary diseases such as silicosis and respiratory failure. Other risks may be caused by the technical and constructive features of the tailing pond that in certain weather-climate conditions (rainfall) can lead to cracking or sliding of the dam.

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