

# Heavy Metal Contamination of Groundwater and Surrounding Soils by Tailing Leachates from a Gold Mine in Thailand

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**ABSTRACT:** Mining sites generally release substantial amounts of potentially toxic metals that are problematic to both the environment and human health. This study evaluates the discharge of potentially toxic metals from the mill tailings of a gold mine in Thailand. Metal analysis of the mill tailings were performed according to EPA method 3051. A series of batch and column desorption experiments were conducted to investigate the sorption/desorption and mobility of various metals from the tailings under three different pH conditions (pH 4, 7 and 10). The concentrations of potentially toxic metals in the tailings were not over the Thailand Soil Standards for Habitat and Agriculture, except for Mn. Batch desorption experiments showed that the release of potentially toxic metals was pH dependent with Mn more easily released from the tailings than other metals.

Using column experiments, the amount of metals released in decreasing order from the tailings were: Mn >> Zn > Ni ~ Pb although the rate of release with gradually decreasing pH was Mn > Ni > Zn~Pb. The maximum concentrations in the leachate were 40, 30, 20 and 18 mg/L for Mn, Zn, Ni and Pb, respectively, which were all over the Thailand Industrial Effluent Standards.

The metal concentrations in the groundwater collected from shallow and deep wells at the mining site over a two-year period typically showed a relatively high concentration of Mn supporting the laboratory results of the batch and column desorption experiments.

**Key words:** Tailings, gold mine, heavy metals, pH effect, desorption

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## INTRODUCTION AND BACKGROUND

Mining activities generally release substantial amounts of waste that can subsequently become problematic for the environment and human health. Their release of potentially toxic metals at mining sites is generally caused by poorly designed and maintained storage facilities, poor environmental management of mining wastes and the consequences of acid mine drainage.<sup>(1)</sup> Many previous studies have shown that potentially toxic metals released from

mine wastes can lead to adverse health effects in animals and humans.<sup>(1-7)</sup> These metals include lead, zinc, arsenic, nickel and copper which, at trace quantities, are essential for maintenance of metabolism in the organisms and humans but which at high concentrations are toxic.<sup>(8-11)</sup> In addition, these potentially toxic metals are persistent and can accumulate in the environment for decades to millennia.

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Vast masses of mill tailings are generated by the mining industry and the current amount of tailings is forecast to double over the next 20 to 30 years.<sup>(12)</sup> Leaching of toxic metals from tailings is expected to occur but when acid mine drainage conditions are present, the transportation and impact of potentially toxic metals at the site will increase considerably and at many sites mine tailings may then be a major source of pollutants. The severity of the negative environmental effects associated with mine tailings or tailings in an impoundment, may not be evident initially but become apparent long after the mine's closure and decommissioning of the tailings impoundment.<sup>(13)</sup>

Therefore, it is very important to evaluate the amount of potentially toxic metals that can be leached from the mine tailings, along with the impact of the contamination on the environment, in order to initiate adequate management practices to prevent and control pollution of the surrounding areas and surface and ground water systems, in the vicinity of the mine.

The Akara Mine, which is the biggest gold mine in Thailand, was chosen as a representative site for this study because it is an active and accessible mine. It is located at the border between Pichit and Petchbun Provinces, about 280 km north of Bangkok (Figure 1). The mine has been in operation for fourteen years. Waste rocks from the mining process are used in the construction of a tailings storage facility (TSF) or deposited in two mine waste

emplacements along with the groundwater monitoring wells (Figure 1). Approximately 750,000 dry tons per year of tailings in the form of particulate suspensions are generated from ore processing (referred to as mill tailings) and deposited in the TSF. Results from an environmental impact assessment study conducted in 1999 revealed that shallow municipal wells in nearby villages intersected the shallow, unconfined, alluvial aquifers at a depth of between 1.5-7 m below the ground surface and the groundwater table was approximately 6 m below the ground surface.<sup>(14)</sup>

Gold ore in the area is embedded in quartz and carbonate rocks in the sulphide boundaries, in fractures and as 20-micron belbs within the sulphides. In several potential ore zones, substantial amounts of pyrite (FeS<sub>2</sub>) with subordinate sphalerite and chalcopyrite as well as sulphides are present which, when exposed to oxygen and water, may generate acidic water by oxidation<sup>(15)</sup> leading to acidic mine conditions with enhanced pollution.

The main objective of this study was to determine the leaching potential of toxic metals from mill tailings in the TSF of the gold mine. To achieve the objective, metal analysis of mill tailings were employed according to EPA method 3051<sup>(16)</sup> and a series of batch and column desorption experiments were conducted to investigate the sorption/desorption and mobility of various metals from the tailings under different pH conditions. The experimental results were then compared with the groundwater quality data collected from monitoring wells in this area.

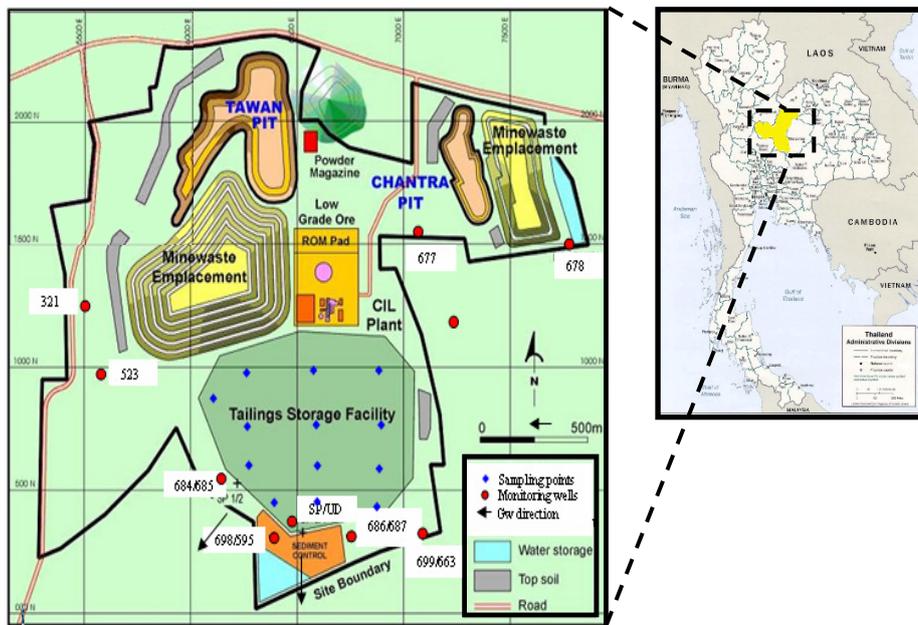


Figure 1. Map showing location of Akara Mine and sampling points.

### HEAVY METALS IN MILL TAILINGS

The mill tailing samples were collected, about 1 kg per sample, using a hand auger at 13 randomly selected different locations distributed over the TSF area at depths from 0-2 m below the ground surface. The samples were kept in clean plastic bags and transported back to the laboratory. Each sample was air-dried and sieved through a 0.075 mm mesh prior to being used in the experiments. Sieve analysis revealed that most tailing particles were very fine with particle sizes between 0.075 mm and 0.002 mm.

The metal contents in the tailing samples were analyzed according to the EPA Method 3051.<sup>(16)</sup> A tailing sample of 0.5 g was transferred into a fluorocarbon microwave digestion vessel with 10 mL nitric acid and digested for 10 minutes. The sample was cooled, allowed to settle and the supernatant diluted with 50 mL of deionized water. The slurry was then filtered through Whatman filter paper (1.2 µm) and the aqueous phase analyzed using an inductively coupled plasma optical emission spectroscopy (ICP-OES, model Varian Vista MPX, Varian, Palo Alto, CA).

The metals analyses obtained by ICP-OES for mill tailings and topsoil, summarized from the 13 sites together, are presented in Table 1. The results show that the metal concentrations in the tailings were generally higher than those found in the topsoil, except for the amount of iron. In the mining process, valuable metals are extracted from the ores and only the unwanted metals are left and these are mostly toxic. Most of the metals found in the tailings did not exceed the Thailand Soil Standards for Habitat and Agriculture.<sup>(17)</sup> However, the Mn concentration in the tailings exceeded (~1.4-fold) the soil standards for habitat and agriculture (the Mn standard is 1.8 mg/g).

### BATCH DESORPTION EXPERIMENTS

Several buffer solutions were initially tested to select the most suitable extraction solution. The optimal buffers for pH 4, 7 and 10 were 0.14 M of NaAc (pK = 4.76), 0.124 M of PIPES (Piperazine-1,4-bis(2-ethanesulfonic acid), pK<sub>2</sub> = 6.80) and 0.0075 M of CAPs (3-(Cyclohexylamino)-propan-1-sulfonic acid, pK<sub>1</sub> = 10.40), respectively.

**Table 1. Chemical analysis of tailing solid samples at the Akara Mine.**

Metal	Concentrations of metals (mg/g) <sup>+</sup>				Thailand Soil Standards for Habitat and Agriculture <sup>(17)</sup>
	Top soil		Tailings		
	Range	Mean. ± S.D.	Range	Mean. ± S.D.	
Aluminium	5.98 - 8.48	7.28 ± 1.07	7.28 - 13.67	10.82 ± 1.73	-
Arsenic	ND	ND	ND	ND	0.0039
Barium	0.01 - 0.09	0.05 ± 0.03	0.04 - 0.29	0.147 ± 0.06	-
Bismuth	ND	ND	ND	ND	-
Boron	0.02 - 0.17	0.09 ± 0.06	0.04 - 0.17	0.103 ± 0.02	-
Cadmium	ND	ND	ND	ND	< 0.037
Calcium	0.59 - 3.47	1.69 ± 1.10	24.21 - 51.76	41.80 ± 7.73	-
Chromium	ND	ND	ND	ND	< 0.3
Cobalt	ND	ND	ND	ND	-
Copper	0.02 - 0.06	0.05 ± 0.01	0.02 - 0.09	0.07 ± 0.02	-
Iron	4.83 - 28.04	19.05 ± 8.79	11.41 - 22.90	19.71 ± 2.38	-
Lead	ND	ND	0 - 0.13	0.04 ± 0.02	< 0.4
Magnesium	0.38 - 2.01	1.48 ± 0.65	3.86 - 5.36	4.87 ± 0.36	-
Manganese	0.03 - 0.18	0.12 ± 0.06	1.67 - 2.84	2.49 ± 0.26	< 1.8
Mercury*	0.028 - 0.381	0.127 ± 0.147	0.12 - 1.73	0.40 ± 0.46	< 23
Nickel	ND	ND	0.05 - 0.11	0.07 ± 0.01	< 1.6
Potassium	0.67 - 2.15	1.04 ± 0.58	2.37 - 2.79	2.53 ± 0.01	-
Silver	ND	ND	ND	ND	-
Sodium	ND	ND	0.33 - 0.45	0.41 ± 0.01	-
Strontium	0.01 - 0.03	0.02 ± 0.01	0.02 - 0.05	0.04 ± 0.01	-
Zinc	0.06 - 0.09	0.06 ± 0.03	0.05 - 0.28	0.17 ± 0.06	-

+ number of samples analyzed: top soil = 5, mill tailings = 22

\*Unit: µg/g of tailing

ND: not detected

To study the desorption of heavy metals from mill tailings, 0.5 g tailings were added into 60 ml plastic bottles and shaken with 50 ml of the optimal buffer solutions at 180 rpm and sampled periodically over 24 hours to ensure equilibrium was attained. For each sampling the suspension was centrifuged, filtered and subsequently the metal concentrations in the aqueous solution were analyzed using ICP-OES. Desorption of Mn from the tailings under each condition is presented in Figure 2 while desorption of Pb, Zn and Ni at pH 4 are presented in Figure 3. The results show that at pH 4, the tailings leached out higher amounts of heavy metals (Mn, Pb and Zn) than that at pH 7 or 10. For example, the total amount leached at steady state conditions, for pH 4, 7 and 10 were 1.20, 0.38 and 0.03 mg/g of Mn, respectively. The amount of Pb desorbed at pH 4 (0.10 mg/g of tailing) was more than that at pH 7 or

10 (not detected). Moreover, at pH 4, the amounts of Mn leached were much higher than Pb and Zn. In other words, Mn was more easily leached than Pb and Zn. For pH 4 the desorption of Mn appeared to reach steady state conditions after 72 hours, whilst at pH 7 this may have been attained by 24 hours but the rate of release is too slight for unequivocal determination. Insufficient Mn was released at pH 10 for any such analysis. The leaching of Pb persisted for longer requiring at least 312 and maybe up to 408 hours to reach the local steady state condition at pH 4. This could be due to the lower solubility of the lead species as compared to the other metal species.<sup>(18)</sup>

From Figures 2 and 3, it is obvious that the leaching of heavy metals from tailings was pH dependent with heavy metals desorbing more under acidic conditions.

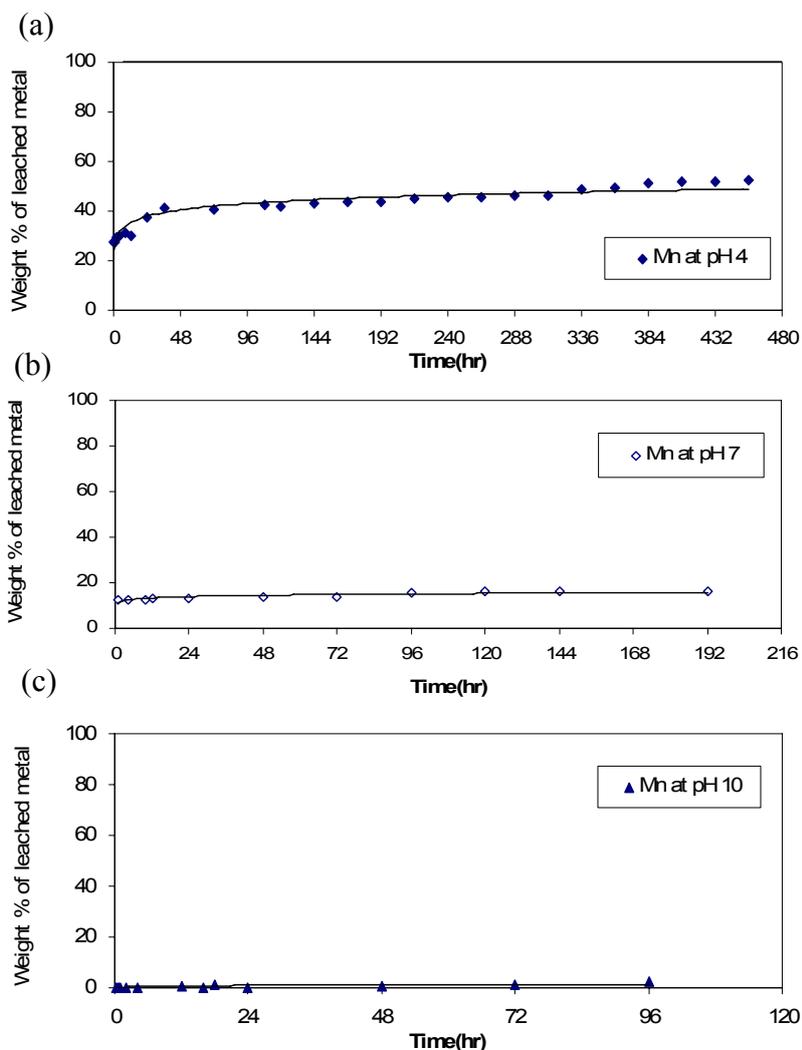


Figure 2. Leached Mn levels, as a % of the total initial concentration in the tailings, under three different pH conditions at various times.

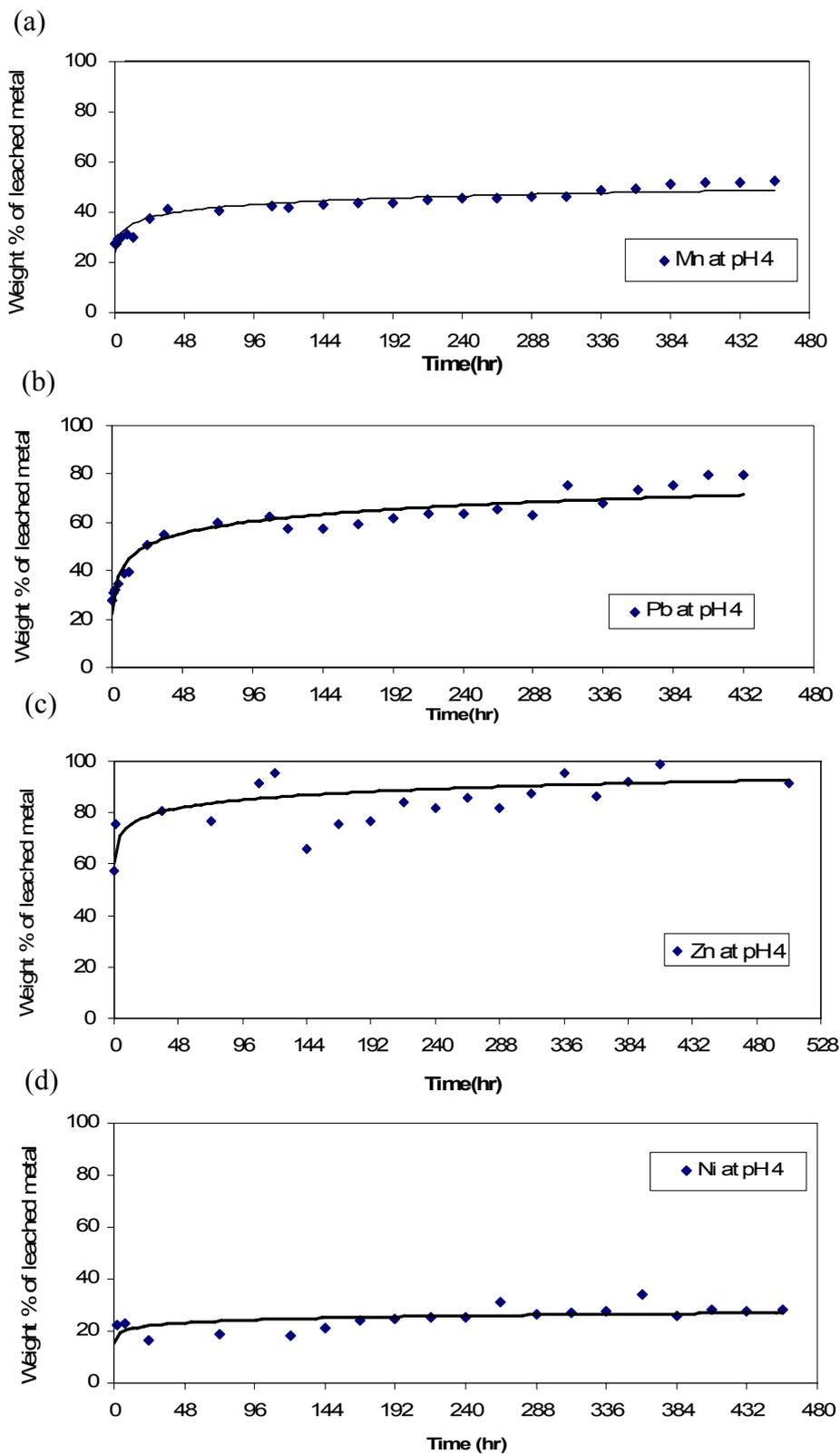
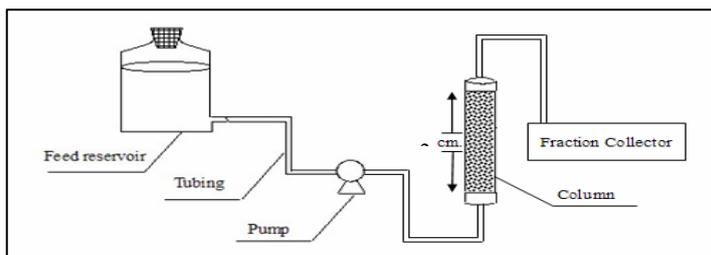


Figure 3. Leached Mn, Pb, Zn and Ni levels, as a % of the total initial concentrations in the tailings, at pH 4 at various times.

### COLUMN DESORPTION EXPERIMENTS

The column desorption setup consisted of a reservoir, a peristaltic pump, a column and sample collection (Figure 4). The column desorption experiments were conducted using an aqueous solution of 0.01 mol/L of NaNO<sub>3</sub> at pH 4 and 7. An acrylic column with a 2.5 cm inner diameter and 20 cm length was uniformly packed with 110 grams of air-dried tailings with a pore volume of 40 mL.

The column was kept under saturated flow conditions with flow entering at the bottom and exiting at the top. The flow in each column was 0.4 mL/min, which was provided by the peristaltic pump and a fraction collector was used to collect the leachate from the column every hour for ten days. Ten mL of each sample was analyzed for metals using ICP-OES.



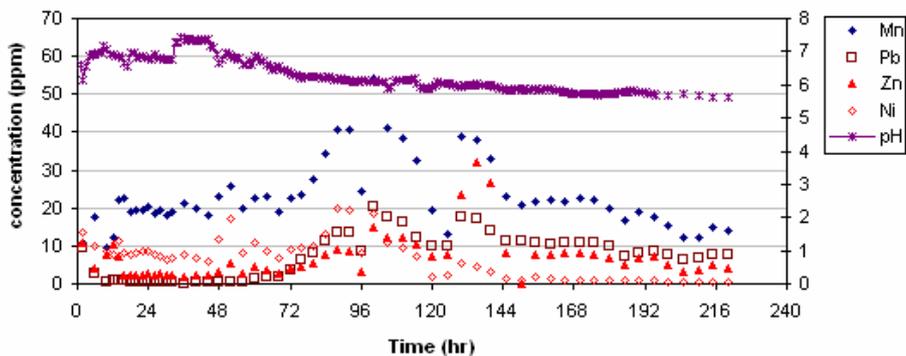
**Figure 4. Desorption Column experiment setup.**

The column experiment results (Figure 5) show that Mn, Ni, Zn and Pb were leached at relatively high concentrations and certainly exceeded the Thailand Industrial Effluent Standards<sup>(19)</sup>, especially under the weakly acidic conditions, with significantly smaller amounts leached under neutral conditions (Figure 5). For example, the maximum concentration for Pb was 18 mg/L under acidic conditions compared to none under neutral conditions, and was higher than the industrial effluent standard (0.2 mg/L). Even for Zn, Ni and Mn, desorption from tailings under this weakly acidic condition was higher than the industrial effluent standards (5 mg/L for Zn, 1 mg/L for Ni and 5 mg/L for Mn). Overall, these results are in accordance with those from the batch desorption, where the desorption of heavy metals from tailings collected from the Akara mining area was found to also be pH dependent. Figure 6 reveals that the total amount of released metals from the tailings was in the following order: Mn >> Zn > Ni ~ Pb. The cumulative curves showed that Ni reached a ‘plateau’ with no new metal release after 120 - 144 hours when the pH reached ~5.7 and that the leaching behavior of Zn was quite similar to Pb with both metals almost reaching a plateau at 204 – 216 hours but with a persistent low level release of new metal ions thereafter. In contrast, Mn showed no plateau up to the longest assayed point (216 hours) although the rate of release of metal ions decreased significantly from 204 hours to a relatively low level. With respect to pH, Mn and Ni were mobilized early on, when the pH of the pore water was about 7, whilst Pb and Zn were mobilized when the pH decreased to 5.7 or less.

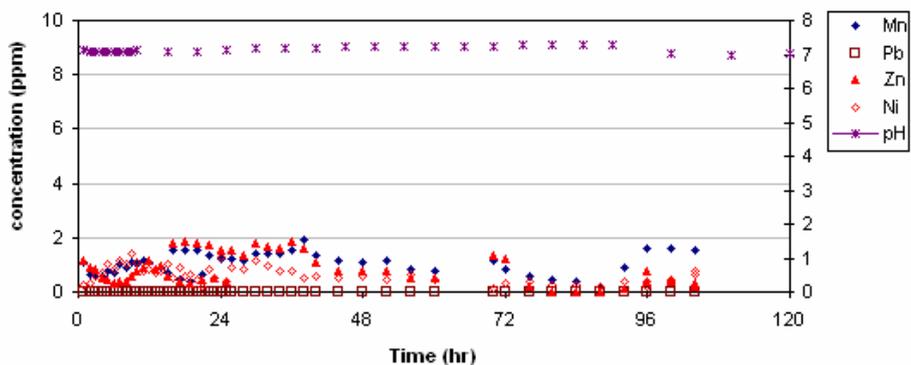
### DISCUSSION AND CONCLUSIONS

In several potential ore zones, substantial amounts of pyrite (FeS<sub>2</sub>) are present which when exposed to oxygen and water may generate acidic water by oxidation to sulphurous and sulphuric acids<sup>(15,20)</sup> leading to acidic mine conditions. This acidification, especially if coupled with further oxidation, will result in the release of heavy metals. Once the release occurs, water containing heavy metals can migrate downwards and eventually contaminate the groundwater. The concentrations of Mn, Fe and SO<sub>4</sub><sup>2-</sup> found at the underdrained (UD) and the seepage (SP) wells over a two-year period at Akara mine were higher than those found at the downstream ambient monitoring wells (Figure 7). The monitoring wells are divided according to the groundwater depth: shallow and deep. Shallow groundwater wells (10 m depth) included well no. 684, 698, 686 and 699; deep groundwater wells consisted of well no. 685, 595, 687 and 663 and their locations are shown in Figure 1.

There is clear evidence that the Mn, Fe and SO<sub>4</sub><sup>2-</sup> concentrations exceeded the industrial effluent standards (5 mg/L for Mn) and groundwater standards for drinking purposes<sup>(21)</sup> (0.5 mg/L and 250 mg/L for Fe and SO<sub>4</sub><sup>2-</sup>, respectively) at the TSF monitor sites. In addition excess levels were noted at the downstream sites for iron in late 2006 (663, deep) and Mn (595, deep) and SO<sub>4</sub><sup>2-</sup> (595, deep; 698, shallow) in early 2004. These results, including the SO<sub>4</sub><sup>2-</sup> levels, support the occurrence of oxidation of sulfide (S<sub>2</sub><sup>2-</sup>) minerals in the TSF and movement out into the nearby downstream water systems.<sup>(3,12)</sup>



(a) Acidic pH condition



(b) Neutral pH condition

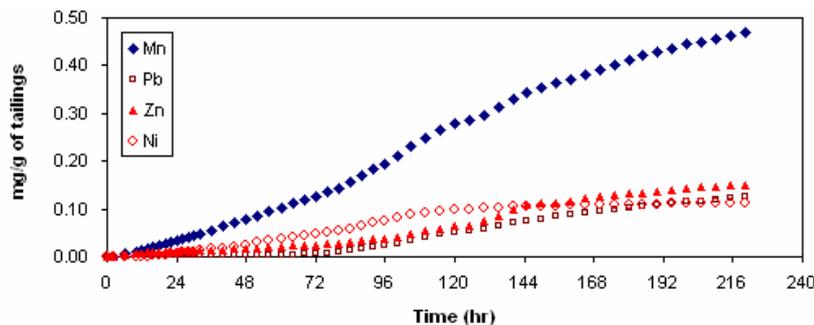
**Figure 5. Metal leaching from the column under weakly acidic and near neutral pH conditions.**

Furthermore, as shown in Figure 7d, the concentrations of total dissolved solids (TDS) found at the UD and the SP wells were at the levels of 3,000-4,000 mg/L, which were higher than the TDS concentrations found at the ambient monitor wells. The increased TDS concentrations in the groundwater might also have been generated from the oxidation of sulfide in the TSF.<sup>(3)</sup>

Metal analysis, according to EPA Method 3051, in the Akara mining area revealed that most of the metals found in the tailings did not exceed the soil standards for habitat and agriculture<sup>(17)</sup>, but Mn

was the clear exception. These results are in accordance with the high amounts of Fe and Mn in leachates from the tailings in UD and SP wells as shown in Figure 7.

The batch desorption data (Figures 2 and 3) are crucial in the investigation of the leaching behavior of potentially toxic metals from TSF and revealed that at about pH 5 – 6 a significant leaching of metals occurred, indicating that under acid mine drainage the mobility of metals increased and that Mn is more easily leached than Pb and Zn.

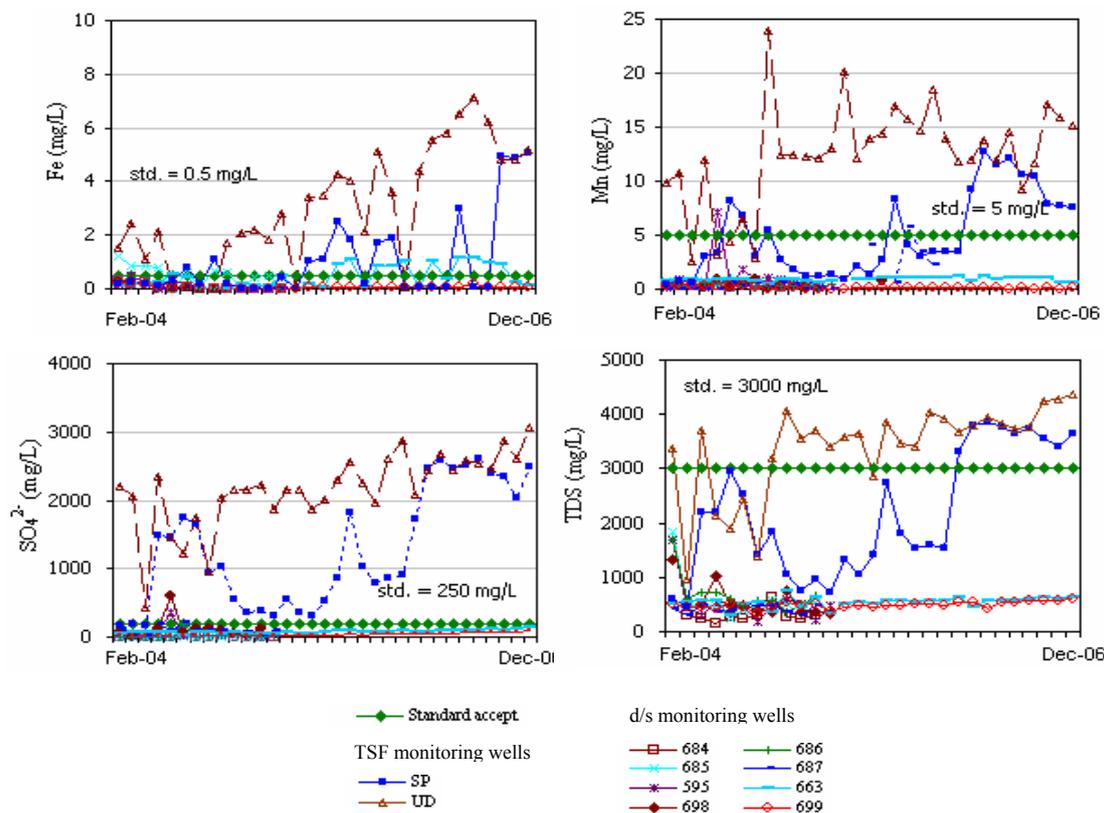


**Figure 6. Cumulative amounts of released Mn, Pb, Zn and Ni versus time under the weakly acidic conditions.**

This agrees with results from UD and SP wells (Figure 7), where Mn was released more than other metals. It may be concluded that the desorption behavior from tailings depended on the metal species (Figure 8) which might result from the difference in the solubility of each metal.

That the potentially toxic metals (i.e., Mn,

Ni, Zn and Pb) that leached from the tailing column exceeded the industrial effluent standards<sup>(19)</sup>, especially those metals leached under mildly acidic conditions serves as a warning of mining tailings as a potential long term reservoir of toxic heavy metals and ease of release into the environment following mild acidification.



**Figure 7. Monthly groundwater quality (Fe, Mn, SO<sub>4</sub><sup>2-</sup> and TDS) of TSF and d/s (downstream) monitoring wells from February 2004 to December 2006. SP = seepage wells; UD = underdrained wells.**

The mobility of metals released from tailings could be described in the following order: Mn > Ni > Zn ~ Pb which might reflect the competition between metals and protons in the aqueous solution and each metal's solubility characteristics under different pH conditions.

These findings support the batch desorption experiments in that the desorption behavior from tailings depended on the pH and on the metals species and are also in agreement with the order reported by Dubrovsky<sup>(22)</sup> for the Nordic Main tailings impoundment near Elliot Lake, Ontario, where the metal mobility was Co ~ Ni > Zn > Pb > Cu. Moreover, Jurjovec et al.<sup>(23)</sup> using a tailings column based evaluation reported that the metal concentrations in the effluent water were controlled by pH and could be divided into three groups on the

basis of their mobility. Zn, Ni and Co became mobile at pH 5.7 and Cr, V, Pb and Cd became mobile at pH 4.0. Cu concentrations remained unaffected by pH changes.

Interestingly, the amount of Fe was approximately eight times higher than Mn (Table 1) in the tailings, but the amount of Fe leached from the column was much lower than Mn, at both the acidic and neutral conditions (data not shown). These results support the observed data at the UD well (Figure 4), where more Mn leached from the tailings than Fe despite the latter's greater abundance. Thus, understanding the leachability of each metal could provide a more reliable evaluation of the transport of metals in the groundwater and the extent of environmental contamination from mine tailings.

The results of this research highlights the need for necessary attention by responsible parties because metal leaching problems may arise long after the mine's closure and/or abandonment. In addition, in order to reduce the release of these potentially toxic metals into the environment, methods for reclaiming mining sites must be implemented. Even though some methods for the reclamation of a mining site, such as revegetation approaches can reduce soil erosion and restrict leaching to groundwater,<sup>(23)</sup> they can lead to other problems such as release of organic acids by plant roots and microbes in the rhizosphere which may combine with metals to form water soluble complexes or colloids.<sup>(24-25)</sup>

This study illustrates the possibility of environmental contamination from the mining site, particularly after the mine's closure, as the metals in the tailings may be leached for decades or even centuries after a mine's closure.<sup>(26-28)</sup> Groundwater monitoring wells should be carefully installed to monitor and evaluate the contaminated elements released from mining activities, even after the mine site has been closed. Proper preventive efforts should be considered and implemented to prevent future possible contamination.

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