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Physical modeling of subaqueous high sulphide mine tailings disposal: Impact of hydrodynamic conditions on water cover quality

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ABSTRACT

The objective of this study was to simulate the impact of various hydrodynamic conditions (from normal to extreme) on changes in water quality when high sulphide tailings are deposited under a water cover. A 35 to 38 cm thick high sulphide tailings layer was placed into three laboratory columns and covered by 80 cm of deionized water. The water cover was stagnant in Column 1, continuously mixed with low tailings resuspension in Column 2, and continuously mixed with high tailings resuspension in Column 3. Water was regularly sampled from both tailings and water covers to determine changes in geochemical parameters. Results obtained after an eight-month monitoring show that pH remains near neutral in the water cover in Column 1. However, pH in water covers in columns 2 and 3 becomes acidic. Oxidation is present in the tailings, with sulphate concentrations from 100 to 1500 mg/l in all columns. Concentrations of heavy metals in the water cover in Column 1 remains generally low over the entire testing period. In columns 2 and 3, metal concentrations increase with time, sometimes reaching high concentration, especially in Column 3 with high resuspension where Zn reaches 78 mg/l.

RÉSUMÉ

L'objectif de cette étude était de simuler l'impact de diverses conditions hydrodynamiques (normales à extrêmes) sur l'évolution de la qualité de l'eau lorsque des résidus miniers très sulfureux sont déposés sous un recouvrement en eau. 35 et 38 cm de résidus miniers recouverts par 80 cm d'eau déionisé ont été montés dans trois colonnes au laboratoire. L'eau de recouvrement était stagnante pour la colonne 1, continuellement agitée avec une faible remise en suspension des résidus pour la colonne 2, et avec une forte remise en suspension des résidus dans la colonne 3. Des échantillons d'eau étaient prélevés régulièrement dans les résidus et dans les couvertures d'eau afin de déterminer l'évolution des paramètres géochimiques. Les résultats obtenus après un suivi de 8 mois montrent que le pH demeure autour de la neutralité dans la couverture d'eau pour la colonne 1, alors que dans les colonnes 2 et 3, le pH devient acide dans les couvertures d'eau. Pour toutes les colonnes, il y a production de sulfate dans l'eau. Les concentrations des métaux dans la couverture d'eau de la colonne 1 restent très faibles dans le temps. Dans les colonnes 2 et 3 par contre, les concentrations des métaux augmentent dans le temps atteignant parfois des concentrations très élevées surtout dans la colonne 3 où le Zn atteint 78 mg/l.

1 INTRODUCTION

The main environmental problem associated with surface storage of mine tailings is acid mine drainage (AMD), which causes water contamination that can seriously affect the environment. This phenomenon occurs naturally when sulphide minerals in tailings oxidize on exposure to air and water. Chemical equations describing these oxidation reactions are provided in the literature (e.g., Jambor and Blowes 1994; Evangelou, 1995; Aubertin et al. 2002). Reducing the oxygen availability to tailings appears to be the most effective way to prevent AMD production in wet climates. One such method is to create an oxygen barrier by maintaining a layer of water (i.e., a water cover) over potentially AMD generating tailings (MEND 2001). This technique can be applied by subaqueous disposal of fresh mine tailings or by flooding mine tailings initially stored in a tailings impoundment. In that case, the mine tailings can be previously oxidized. The advantage of the water cover in controlling sulphide oxidation, and hence acid generation, lies in its ability to limit the availability of dissolved oxygen, because molecular oxygen has low solubility and diffusivity in water (~ 8.6 mg/L at 25 ºC and 2×10^{-9} m^2/s, respectively) compared to air (~ 285 mg/L and 1.82×10^{-5} m^2/s, respectively) (Yanful and Verma 1999; Aubertin et al. 2002; Romano et al. 2003; Davé et al. 2003).

Many studies have examined water covers, and several authors have demonstrated their efficiency in both laboratory (Aubé 1995; Rescan 1996; Davé et al. 1997; Li, et al. 1997; Holmström and Öhlander 1999; Catalan 2000; MEND 2001; Li et al. 2000) and field tests (MEND 1989; MEND 1993; Yanful and Simms 1997; Vigneault et al. 2000).

Although this method has proven efficient and beneficial, some uncertainty remains. A water cover is a complex and dynamic system subjected to several interdependent factors such as tailings erosion and resuspension, which are generally induced by wind action. Wind-induced waves and currents can remove mine tailings in suspension and increase their exposure to dissolved oxygen. The hydrodynamic conditions of tailings erosion and resuspension can be estimated from the wind speed and the thickness and fetch of the water cover (Adu-Wusu et al. 2001; Catalan and Yanful 2002; Mian and Yanful 2004; Mian et al. 2007). Particles in suspension can oxidize, causing increased acidity of the cover water and the liberation of metals. This phenomenon can be significant if the mine tailings are highly sulphidic.

The objective of this study was to simulate, using laboratory column tests, changes in cover water quality when high sulphide mine tailings are deposited underwater and subjected to hydrodynamic conditions. Three columns simulating a stagnant water cover, a continuously mixed water cover with low resuspension, and a continuously mixed water cover with high resuspension of mine tailings were tested. Column configurations and the water sampling procedure above and within the tailings are first described in the following. The physico-chemical and chemical analyses performed on the water samples are then presented. Only results related to the water cover after an eight-month monitoring are presented and discussed in this paper.

2 MATERIALS AND TESTING PROCEDURES

2.1 Mine tailings characterization

Tailings were sampled at four locations beneath a water cover from a barque using a dredge. Tailings were kept in buckets and under a water layer to minimize surface oxidation. Once in the laboratory, tailings were placed into barrels and kept under water prior to testing. Tailings were mixed and homogenized in an industrial mixer for the physical, mineralogical and chemical characterization.

Two samples were taken from the homogenized tailings for characterization. Grain-size distribution was determined using a Malvern Mastersizer laser granulometer to obtain grain-size volume distributions for diameters from 0.05 to 900 µm. Solid grain density was determined using an Accupyc 1330 Helium pycnometer (Micromeritics). Tailings were digested by the addition of concentrated acid nitric, liquid bromine, hydrofluoric acid and concentrated hydrochloric, all heated. The solution was then evaporated and salts were dissolved in hydrochloric acid at 10%. An elementary analysis for 22 elements was then performed using inductively coupled plasma-atomic emission spectroscopy (ICP-AES). Mineralogy of the solid materials was also analyzed by X-ray diffraction (XRD) using a Bruker AXS D8 Advance θ/2θ for diffraction angles of 5° to 70°. Mineral phases were identified using the software EVA and quantified using TOPAS (Bruker AXS).

2.2 Laboratory column test simulations

2.2.1 Description of columns

The experimental setup consists of three PVC columns of 13 cm internal diameter and 128 cm height. A perforated plate covered with a geotextile filter is placed at the bottom of each column. Five sampling ports consisting of ¼ inch NPT holes are placed along the column height. Three sampling ports are installed in the tailings to sample pore water and two sampling ports are located in the cover water to collect water above the tailings (see Figure 1; plexiglass columns are presented to better illustrate the sampling ports). The tailings–water interface is placed at approximately 30 cm from the bottom of the column. The three sampling ports in the tailings were placed initially at 0, 10 and 20 cm depths under the tailings–water interface. The sampling ports in the cover water were placed 10 cm and 40 cm above the tailings surface.

The pore water sampling system consists of three horizontal perforated PVC tubes covered with a geotextile to prevent tailings from entering the tube (Figure 1). The internal volume of the tube is about 40 ml. Each sampling tube is connected to two valves used for water sampling, as described below. The end of each plastic vial into the tailings was prevousily covered with a fine geotextile. The two sampling ports in the water cover are also connected to valves.

Each column is provided with a cap to support the stirrer motor in the columns with mixed water covers.

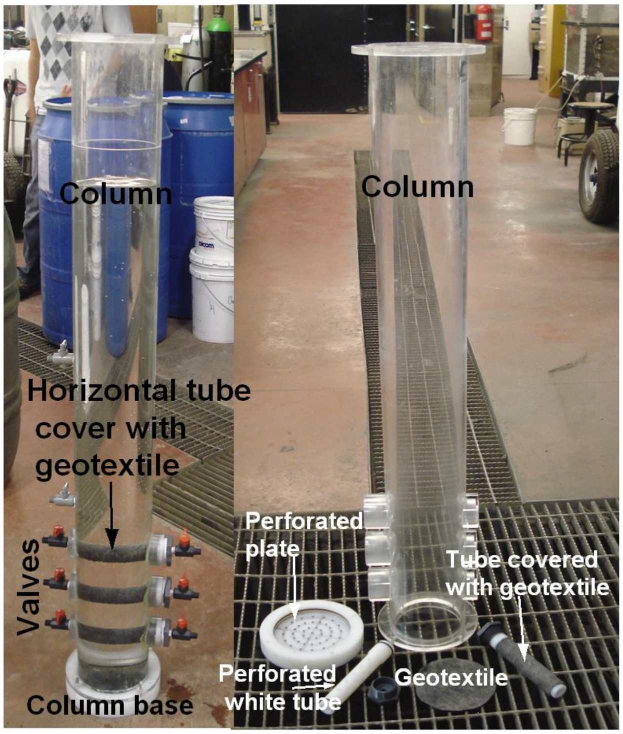

Figure 1: Column components

2.2.2 Experimental setup

Once all the column components were in place and column watertightness was tested, each column was weighed, then filled with deionized and deoxygenated water (DDW) up to the top limit of the geotextile placed in the bottom of the column to eliminate airlock below the geotextile. Tailings were rinsed with DDW to eliminate any sulphates that might have formed. The completely saturated tailings were mixed and homogenized in a barrel using an industrial mixer. The tailings slurry was then poured gradually into each column using a funnel.

Once the level of the slurried tailings reached the lower limit of the first horizontal tube, it was filled with DDW to prevent an airlock from forming in the tube. This procedure was repeated until all three tubes were filled. The tailings were then allowed to settle for at least four days until the overlying water was clear. The overlying water was then emptied by pumping at a low flow rate (using a peristaltic pump). As the PVC columns were not transparent, the thickness of tailings in the columns was deducted from the total height of the column and the free height over tailings after the water was emptied. An 80 cm thick water cover was placed above the tailings by pumping fresh deionized water at the lowest possible flow rate to avoid removing tailings in suspension.

The targeted tailings thickness (after settling and consolidation) was 30 cm in each column. As shown in Table 1, tailings thickness in columns 1, 2 and 3 is 38 cm, 35 cm and 38 cm, respectively. Consequently, the positions of the sampling ports with respect to the tailings–water interface changed slightly. The hydrodynamic conditions applied to the different columns are also given in Table 1: cover water is stagnant in Column 1, continuously mixed with low tailings resuspension in Column 2, and continuously mixed with high tailings resuspension in Column 3. The initial objective was to continuously mix the water without tailings resuspension in Column 2. Subsequent checks revealed suspended tailings in this column as well, albeit in lower proportion than for Column 3.

Table 1. Column configurations and applied hydrodynamic conditions

Column No.	Description
1	38 cm tailings + 80 cm stagnant water
2	35 cm tailings + 80 cm continuously mixed water with low resuspension
3	38 cm tailings + 80 cm continuously mixed water with high resuspension

Once the columns were filled, they were left to settle for up to two days. Each column was then weighed and water samples were taken via each sampling port in the cover water and analyzed to determine the initial geochemical profile. Electric stirrers (Model 103A, Talboys Laboratory) were placed in columns 2 and 3. The stirrer rod was 33 cm long in Column 2 and 48 cm long in Column 3. Each stirrer had a propeller with three 2.5 cm diameter wings. Column 2 was mixed at about 53 RPM to fully oxygenate the water overlying the tailings without removing them in suspension. However, some tailings (about 200 to 300 mg/l) were seen to resuspend. Column 3 was mixed at a maximum speed of about 100 RPM to remove solid particles in suspension. Mixing speeds were obtained from tests using a transparent Plexiglas cylinder. Figure 2 illustrates the laboratory column test. PVC columns were finally used because the initially used Plexiglas columns (Figure 1) cracked at the sampling tube connections three months after column testing began.

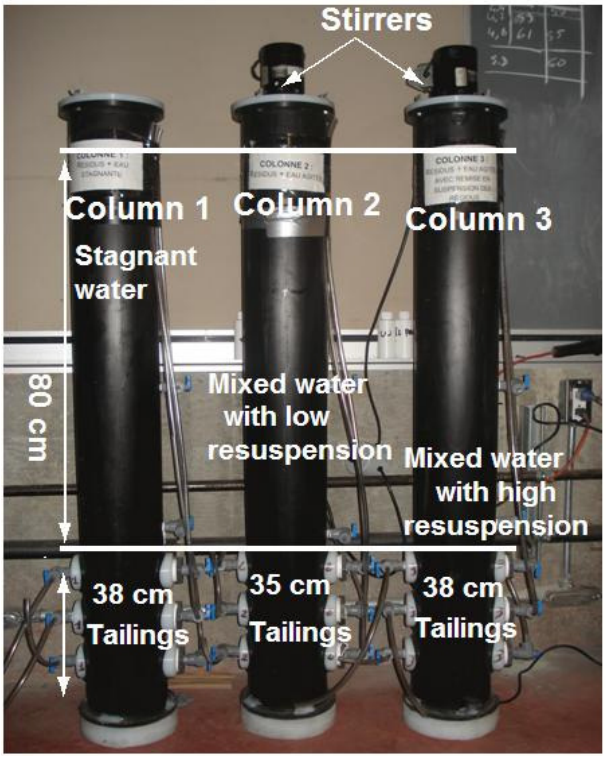

Figure 2. Experimental setup in the laboratory

2.2.3 Water Sampling

For each column, vertical dissolved oxygen (DO) concentrations were measured at 20 cm vertical intervals using a PSt3 oxygen sensor and an OXY-10 oxygen meter (PreSens GmbH, Germany). The sensor consists of a polymer optical fibre (POF) with a polished distal tip coated with a planar oxygen-sensitive foil. The end of the polymer optical fibre is covered with a high-grade steel tube (OD 4 mm) to protect both the sensor material and the POF. These sensors can measure oxygen concentration in the gas phase or dissolved in water. They provide a DO reading in less than 60 seconds. They must be calibrated prior to each test. Water samples were then taken from the top to the bottom of each column. Water samples taken via each of the two sampling ports above the tailings were stored in a 150 ml HDPE bottle. Part of the sampled cover water was returned to the columns. A small quantity of deionized water (about 20 ml) was then added to retain the 80 cm water cover above the tailings. The dilution effect of this added water was assumed to be negligible.

Pore water was taken via the three horizontal tubes using pipes connected to the sampling valves. For each tube, the pipe on one side was filled with DDW, and the other pipe on the opposite side was used to sample the amount of water needed to perform an ICP-AES analysis (15 ml). The column was slightly inclined towards the sampling pipe. Both valves of the same sampling tube were opened gradually, and as the water flowed into the 20 ml sampling bottle on one side, the DDW penetrated the horizontal tube on the other side.

This procedure allowed sampling and replacing the water in the pipe without disturbing the system. It was assumed that ion exchange occurred by molecular diffusion between the pore water and the water contained in the tubes, and that a balance was established before sampling. The time required to reach equilibrium (less than one month) was previously determined using pore fluids with various ion concentrations (results not presented here). Thus, the water in the tubes was assumed to be representative of the pore water in the tailings in the tube at the beginning of the test (April, 2009). The sampling frequency interval was subsequently set at at least one month.

2.2.4 Water analysis

The following physico-chemical parameters were determined in the water samples: hydrogen potential (pH), redox potential (Eh), electric conductivity, dissolved oxygen (DO) and temperature (T). pH was measured using an Orion Triode™ pH probe with automatic temperature compensation. It was coupled with an Orion 920A Benchtop pH/ISE Meter with an accuracy of ± 0.002 units. Eh was determined with a CPS12 platinum electrode (Endress+Hauser). Values obtained in mV were corrected with respect to the normal hydrogen electrode by adding 204 mV to the obtained values. The normal hydrogen electrode is the universal reference. Electric conductivity was determined with a conductometer (OAKTON) equipped with a probe including two platinum electrodes to measure dumping properties. The result is given in µS/cm to 1% accuracy. Dissolved oxygen was measured with a VWR Symphony probe. Temperature was measured with a combined oxygen–temperature probe.

About 15 ml of each cover water and pore water sample was filtered using a 0.45 µm diameter nylon filter (Fisher Scientific). About 9 ml was then acidified with 2% v/v HNO_3 for elementary ICP-AES analysis for the following 21 elements: aluminum (Al), arsenic (As), barium (Ba), beryllium (Be), bismuth (Bi), calcium (Ca), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), total iron (Fe/total), magnesium (Mg), manganese (Mn), molybdenum (Mo), nickel (Ni), lead (Pb), sulphur (S), Antimony (Sb), selenium (Se), silicon (Si) and zinc (Zn). Sodium (Na) was not analyzed. The device was calibrated according to standard guidelines. Results given in mg/l are compensated for the dilution that occurred during acidification. Note that the ICP-AES analyses of water show a relative error of about 6%. Sulphate content was calculated from the total sulphide concentration, using molar mass ratios, and based on the following hypotheses: 1) the most stable form of sulphide in solution in the oxidizing conditions typically found in DMA is the sulphate form; and 2) the concentrations of other sulphide species (S^{2-}, SO_3^{2-}, etc.) are negligible.

For two selected sampling times, the concentrations of suspended solids were determined in columns 2 and 3 by filtration of a known volume of cover water using a 0.45 µm nylon filter. The concentration was obtained by the mass difference between filter weight before filtration and after filtration and drying. The obtained samples were also analyzed for grain size distribution, elementary analysis and mineralogy of the solid materials.

3 RESULTS AND DISCUSSION

3.1 Grain size distribution and solid grain density of tailings

Volumetric grain-size distributions of the two analyzed samples are presented in Figure 3 and specific diameters d_{10}, d_{50}, d_{60} and d_{90} are given in Table 2, where d_x corresponds to x% finer volume than on the cumulative grain-size distribution curve. This table also presents the relative solid grain density (D_r) of tailings. The grain-size distribution curves show significant similarity between tailings samples (with consistently close diameters d_{10}, d_{50}, d_{60} and d_{90}). The coefficients of uniformity (C_u) for both samples range from 5 to 20, which indicates that the tailings are poorly graded. The coefficients of curvature (C_c) are close to 1, which suggests that these curves are quasi-linear between D_{60} and D_{10}.

The relative solid grain density of tailings also shows significant similarity between the two samples, with an average of 4.58. These results are close to the relative solid grain density of 5.01 for pure pyrite (Landry et al., 1995), indicating a large proportion of pyrite in the tailings.

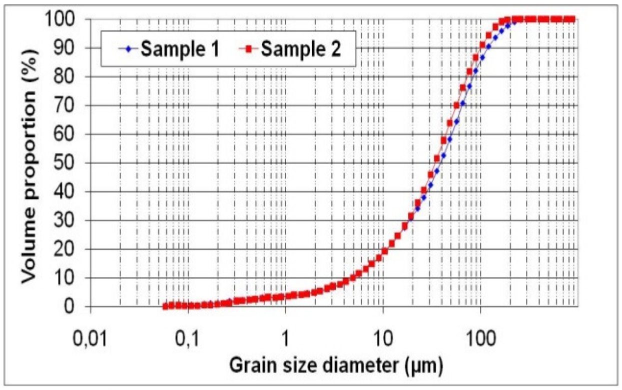

Figure 3. Grain-size distribution curves of tailings

Table 2. Tailings grain-size parameters and relative solid grain density D_r

	Sample		
Parameter	1	2	Mean
Passing Sieve 200 (%)	85	78	81.5
d_{10} [µm]	4.90	4.98	4.94
d_{50} [µm]	34.22	38.68	36.45
d_{60} [µm]	44.10	50.53	47.32
d_{90} [µm]	100.19	118.44	109.32
C_u (-)	8.99	10.16	9.57
C_c (-)	1.53	1.40	1.47
D_r (-)	4.59	4.57	4.58

3.2 X-ray diffraction

The main mineralogical components are presented in Table 3. Pyrite content varies from 82% to 84% for the two samples, with an average of 83%. The high pyrite content can also be observed visually in dried tailings samples. Results show a high dominance of pyrite, which is an acid generating mineral, with a low proportion acid neutralizing minerals. Considering the minimum and maximum relative density for each mineral (2.7 for quartz, 5.0 for pyrite, 2.6 to 3.3 for chlorite and 2.8 to 3 for muscovite) and using the average content in Table 3, the calculated relative density of the tailings is between 4.34 and 4.49. These values fall well within measured values (Table 2), indicating that the DRX characterization is relatively accurate.

Table 3. Mineralogical analysis of tailings by X-ray powder diffraction (XRD)

Sample	Quartz	Pyrite	Chlorite	Muscovite
1	6.93	81.98	10.92	0.16
2	5.88	83.66	6.90	3.56
Mean	6.41	82.82	8.91	1.86

3.3 Elementary analysis of tailings

Table 4 shows the results on the two samples for four main elements (Fe, S, Al and Zn), clearly indicating the strong dominance of Fe and S with an average content of 48.9 and 38.7% respectively (Table 4). Elementary analysis of tailings also shows the presence of Al and Zn, but their average content is lower than 1%, and other elements are present in trace amounts. The presence of quartz (Si) (see Table 3) was not quantified in this analysis (this explains why the sum of the element contents in Table 4 is about 89%).

Table 4. Mineralogical analysis of tailings (%)

Sample	Fe	S	Al	Zn
1	49.3	39.3	0.91	0.38
2	48.5	38.0	0.91	0.38
Mean	48.9	38.7	0.91	0.38

3.4 Water quality in the columns

Of the results on physico-chemical properties, only those on dissolved oxygen (DO) and pH are presented, and the elementary analysis results are limited to sulphate and Zn. In the following, only the results on the cover water parameters above the tailings from April to December 2009 are presented.

3.4.1 Dissolved oxygen profile

Typical vertical DO profiles are given in Figure 4, where depth 0 corresponds to the tailings–water cover interface. In columns 2 and 3, dissolved oxygen concentrations range from 8 to 9 mg/l. It can be seen that OD values are generally homogeneous from surface to bottom of the water cover, indicating that the water cover is well mixed. Water temperature during measurements ranges from 19.2 to 20.4 °C. Saturating concentration of dissolved oxygen at these temperatures ranges from 9.1 to 9.3 mg/l (Benson and Krause, 1984). The dissolved oxygen values in the water cover in columns 2 and 3 are then close to the saturation value of dissolved oxygen. In Column 1, dissolved oxygen concentration falls from 7.2 mg/l at the water surface to 6.0 mg/l close to the tailings–water cover interface for temperatures from 19 to 19.6 °C. According to Benson and Krause (1984), the saturated concentration of dissolved oxygen at these temperatures is 9.1 to 9.3 mg/l. Thus, DO concentration in the water cover in Column 1 is lower than the saturating concentrations.

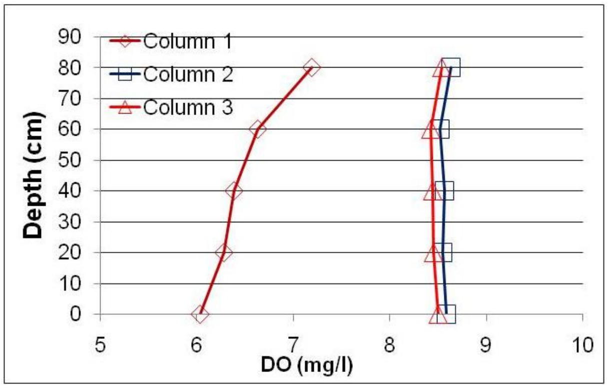

Figure 4. Vertical DO profiles in columns 1, 2 and 3

3.4.2 pH

Figure 5 shows the vertical pH profiles for the three columns, where depth 0 corresponds to the tailings–water cover interface. It can be seen that for each column, there is no significant variation in pH between the two depths for a given period. Figure 6 shows the temporal change in pH at the bottom sampling port in the water cover (2 cm above the tailings–water interface for columns 1 and 3, and 5 cm for Column 2). In Column 1, pH ranges from 6 to 8 in the cover water, whereas in columns 2 and 3 pH varies from 7.5 to 4 and from 7.5 to 2.5, respectively, in the cover water. These results suggest that pH remained about neutral throughout Column 1, whereas pH decreased in columns 2 and 3 where tailings were mixed and resuspended. In columns 2 and 3, there is almost complete saturation of dissolved oxygen throughout the water cover above the tailings, unlike in Column 1.

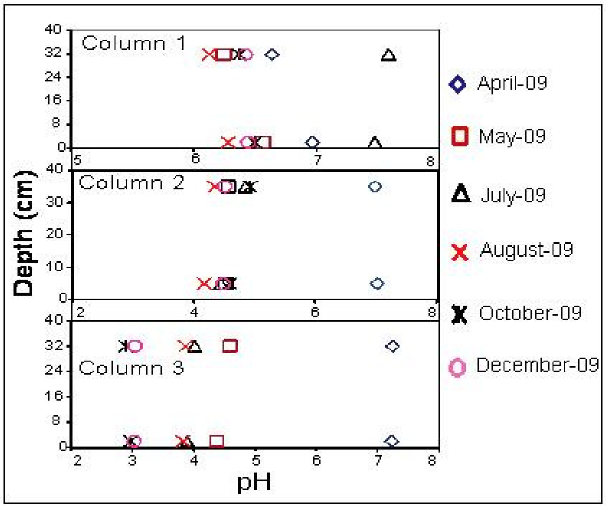

Figure 5. pH profiles in columns 1, 2 and 3

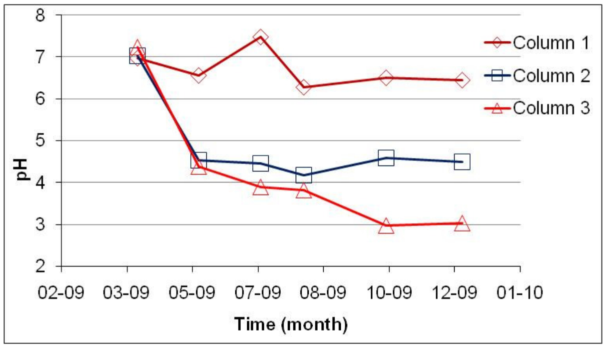

Figure 6. Temporal change in pH in columns 1, 2 and 3

3.4.3 Sulphate

Figure 7 shows the vertical sulphate (SO_4^{2-}) concentration profiles in columns 1 to 3, respectively. Figure 8 presents the temporal change in sulphate concentration at the bottom sampling port in the water cover, showing increased sulphate contents in the cover water over time in all columns, going from 10 to 200 mg/l in Column 1, from 4 to 400 mg/l in Column 2 and from 11 to 1500 mg/l in Column 3. If the maximum sulphate concentration in each column is considered, one can see that the sulphate concentration in Column 3 is 7.5 times higher than in Column 1 and 3.5 times higher than in Column 2. This indicates sulphate production in all columns, but a much higher production rate in Column 3, where the cover water is continuously mixed to induce tailings resuspension. The concentration of suspended solid is about 3000 mg/l compared to Column 2, with only 300 mg/l. In fact, mixing promotes oxygenation and resuspension of tailings in the water layer by facilitating contact between the particles in suspension and DO, which in leads in turn to more tailings oxidation and sulphate production. The concentrations of resuspended tailings in the columns 2 and 3 are hovewer very high compared to concentration encountered in typical field conditions.

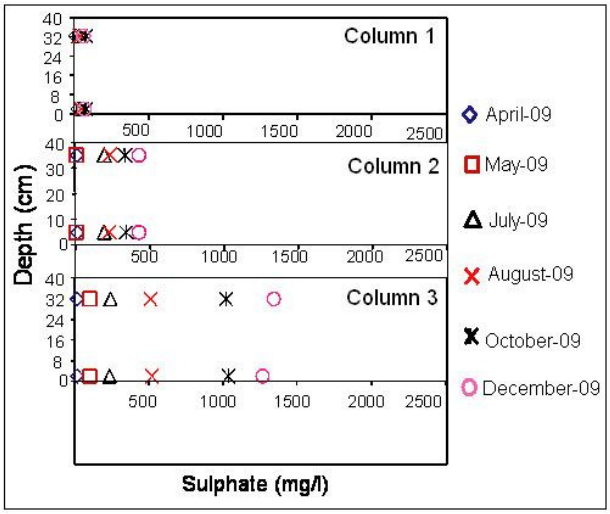

Figure 7. Sulphate profiles in columns 1, 2 and 3

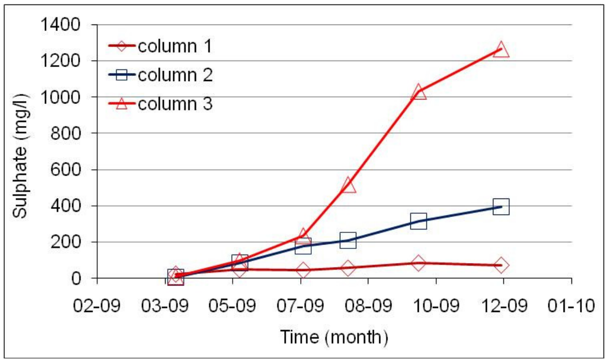

Figure 8. Temporal change in sulphate in columns 1, 2 and 3

3.4.4 Zinc

The vertical Zn profiles in the water cover and the temporal change in Zn at the sampling port near the middle of the water cover are presented in figures 9 and 10 for the three columns, respectively. From April to December 2009, Zn concentrations fell from 0.18 to 0.07 mg/l in Column 1 and increased in columns 2 and 3 from 0.16 to 8.89 mg/l and from 0.2 to 78 mg/l, respectively. When Zn concentrations measured in December 2009 are considered, the concentration in Column 3 is about 9 times higher than that in Column 2 and about 433 times higher than in Column 1. As explained above, the water cover in columns 2 and 3 was fully saturated in dissolved oxygen, unlike in Column 1. Moreover, due to the continuous mixing of the cover water, tailings were resuspended in columns 2 and 3, exposing them to dissolved oxygen. As a result, tailings oxidation increased, pH decreased, and the metals, including Zn, were released into the water cover.

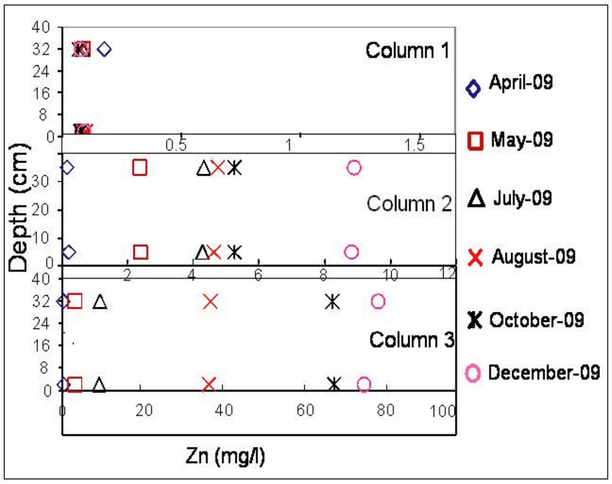

Figure 9. Zn profiles in columns 1, 2 and 3

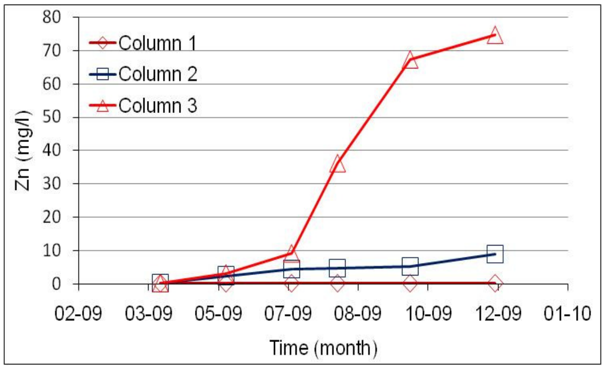

Figure 10. Temporal change in Zn in columns 1, 2 and 3

Results presented in this paper for columns 2 and 3 were obtained for extreme hydrodynamic conditions with suspended solids of 300 mg/L and 3000 mg/L applied over a lengthy period, respectively. The stirrers used did not allow good control of the mixing speed, even after shortening rod length. Furthermore, it is difficult to predict the rotational speed required to induce a given amount of suspended tailings particles. In the field, high-speed winds can induce significant resuspension, albeit of short duration. Thus, these column results are non-transferable to the field. However, they may be used to validate a geochemical model that could eventually be used to predict the long-term geochemical behaviour of a water cover subjected to more realistic hydrodynamic conditions.

4 CONCLUSION

The objective of this study was to simulate, using laboratory column tests, changes in water quality when very high sulphide tailings are deposited underwater and subjected to different hydrodynamic conditions. Tailings containing about 82% pyrite were used.

Tailings layers of about 35 cm were placed in columns and covered with an 80 cm water cover. The water cover was submitted to various hydrodynamics and the following results were obtained after an eight-month monitoring:

1) In columns 2 and 3, where the water cover was mixed with resuspended tailings, pH decreased and reached acidic values, whereas in Column 1, with a stagnant water cover, pH remained near neutral.
2) High sulphate concentrations were produced in the column that was mixed with low resuspension, and even higher concentrations in the column mixed with high resuspension, compared to Column 1 without mixing.
3) Zn concentrations were higher in the mixed column with high resuspension than in the mixed column with low resuspension. Both showed higher Zn concentrations than those in Column 1 without mixing.

In conclusion, tailings resuspension has a significant impact on water chemistry. As mentioned above, some of the results presented here (Column 2 and 3) were obtained under normal to extreme hydrodynamic conditions. Work is underway to continuously monitor resuspension in the field, as well as wind velocity, which induces waves that could cause resuspension.

5 ACKNOWLEDGEMENTS

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