

**Wet Barriers on Pyritic Uranium  
Tailings Laboratory Diffusion  
Lysimeter Studies of Uranium  
Tailings Deposited Under a Shallow  
Water Cover  
MEND Project 2.13.1b**

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**Wet Barriers on Pyrite Uranium Tailings**  
**Part III**  
**Laboratory Diffusion Lysimeter Studies of Uranium**  
**Tailings Deposited Under a Shallow Water Cover**

Nand. K. Davé

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## WET BARRIERS ON PYRITIC URANIUM TAILINGS

### PART III

#### LABORATORY DIFFUSION LYSIMETER STUDIES OF URANIUM TAILINGS DEPOSITED UNDER A SHALLOW WATER COVER

Nand K. Davé\*

#### EXECUTIVE SUMMARY

This report describes the results of laboratory diffusion lysimeter studies that were conducted as a part of a broad research program titled “Development of Wet Barriers on Pyritic Uranium Tailings for Controlling Acid Generation”. The research was undertaken jointly by CANMET, Elliot Lake Laboratory and Rio Algom Limited, under the Canadian Mine Environmental Neutral Drainage (MEND) Program.

#### **Purpose:**

Diffusion lysimeter studies were undertaken to determine the surface oxidation, leaching and mass release characteristics of underwater deposited pyritic uranium tailings for two different cases: 1) un-oxidized tailings that have been kept underwater for more than 12 years and 2) weathered (partially oxidized) tailings. The studies, conducted for shallow water covered tailings having a well mixed (circulated) water column without surface or porewater flows, determined the mass transfer and diffusion related transfer parameters (transfer flux and coefficients) for metals and radionuclides from underwater deposited tailings to the upper lying water column.

The results would provide suitable inputs for modeling the long-term surface water chemistry of water covered tailings.

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**Methods:**

The studies were conducted using aquarium type Plexiglas™ lysimeters for un-oxidized tailings, obtained from the Quirke mill and kept underwater in the laboratory since 1982, and weathered tailings, obtained from the Quirke waste management area and deposited underwater in the laboratory in 1990 . A shallow water cover, approximately 0.2 m in depth, was provided using distilled water. The water column above the tailings was continuously mixed and maintained at a constant height in each lysimeter. The mass transfer parameters for various ionic species such as Ca, Mg, Fe, Mn,  $\text{SO}_4^{-2}$  and Ra-226 etc. were determined, in the absence of any surface and/or porewater flows, by measuring their concentrations in the surface as well as pore waters as a function of time.

**Duration of the Study:**

The experiments, conducted at the CANMET, Elliot Lake laboratory, were started in November 1993 and completed in August 1994.

**Results:****Un-oxidized Tailings:**

- Un-oxidized tailings, deposited underwater in 1982, oxidized very slowly and a narrow oxidized and iron hydroxide precipitate zone, 2-3 cm in thickness, was formed at the surface of the tailings at the water-tailings interface. Mobilization and release of iron to the surface water, under oxidizing conditions, resulted in its precipitation and covering of tailings with a layer of ferric hydroxide.
- The oxidized zone at the tailings surface released low acidity (5 - 20 mg  $\text{CaCO}_3/\text{l}$ ), low concentrations of Mn and Pb, and high concentrations of Ca,

Mg and Ra-226 to the surface water. Gypsum dissolution contributed to increased release of Ca and to a certain extent that of Mg.

- Dissolution of Ra from the tailings surface, and most significantly under acidic conditions, was a dominant factor in increased Ra mobilization and its release to the surface water. Solubility of gypsum, when present, and hence the sulphate ion concentration controlled the release of Ra and its surface water concentration that decreased with increasing sulphate concentration. Diffusion of Ra from the tailings porewater was low (less than 10%) compared to mass dissolution from the surface of the tailings.
- Concentration of other metals in the surface water were low as a result of iron hydrolysis and precipitation.
- Tailings porewater contained mainly dissolved gypsum, Mg, and Ra, and low concentrations of Fe, Mn and Pb. Long-term exposure of the tailings to natural and fluorescent room light in the lysimeter resulted in the formation of an algae layer on the surface of the tailings which slowly contributed to oxygenation of the tailings substrate and decrease in the previously established anoxic conditions as well as sulphate reduction.

#### **Weathered Tailings:**

- Similar to un-oxidized tailings, further oxidation of weathered and partially oxidized tailings underwater was very slow and limited to near surface zone of the tailings.
- The weathered tailings released high acidity (~ 1700 mg CaCO<sub>3</sub>/l) and high metal concentrations (e.g. Fe ~ 550 mg/l and Al ~ 110 mg/l) to the porewater

but their diffusion to the surface water was moderate to low (acidity  $\sim 10 - 20$  mg  $\text{CaCO}_3/\text{l}$ , Fe  $\sim 0.5$  mg/l and Al  $\sim 2.0$  mg/l).

- Iron was dissolved from weathered tailings but its transfer to the surface water was also low, as it hydrolyzed and precipitated forming an iron hydroxide sink layer at the tailings surface. Mass dissolution of gypsum and Ra from the tailings surface resulted in increased concentrations of Ca, Mg and Ra (10,000 - 32,000 mBq/l) in the surface water. Transfer of other metals was low.
- For weathered tailings under high acidic conditions, the transfer flux of Ra was significantly higher (approximately 50 times) and those of Ca,  $\text{SO}_4$  and Mg were significantly lower, (e.g. decrease in transfer coefficients by 2 to 3 orders of magnitude), than those for un-oxidized tailings.

In both cases, the slow oxidation of underwater deposited tailings at the surface as well as the dissolution and release of metals and radionuclides from the oxidized surface have been important factors in determining the surface water quality. The data suggest a need to examine the use of diffusion or oxygen barriers above the tailings surface for further controlling the surface oxidation as well as the release of metals and radionuclides to the surface water column.

## DETAILED SUMMARY OF FINDINGS

Laboratory diffusion lysimeter studies were conducted for the CANMET - Rio Algom joint 'Wet Barriers' research program for determining diffusion related transfer parameters for metals and radionuclides from underwater deposited pyritic uranium tailings.

The studies were undertaken in Plexiglas™ aquarium type lysimeters for un-oxidized and weathered tailings, deposited under a shallow water cover (approximately 0.2 m), in the laboratory, in 1982 and 1990, respectively. The water cover was established and maintained at a constant height using distilled water, and was continuously mixed. The tests were conducted under a no-flow circulated water column condition without surface or porewater flows. For a given ionic or radionuclide species, the initial transfer flux  $J_0$  ( $\text{mg}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ ) at time  $t = 0$ , and its transfer coefficient  $K$  ( $\text{m}\cdot\text{month}^{-1}$ ), from underwater deposited tailings to the upper lying water column were determined by measuring the surface and porewater concentrations of the given species as a function of time. The experiments were conducted at the Elliot Lake Laboratory, CANMET, during November, 1993 - August, 1994. The results are summarized as follows:

- For un-oxidized tailings under a continuous water cover for the past 12 years or so, the water column above the tailings surface (surface water) was slightly acidic with pH approximately 4.0 and acidity in the range 5 - 20 mg/l. The surface water was characterized by significant diffusional or mass transport fluxes of gypsum ( $\text{Ca}$  and  $\text{SO}_4^{-2}$ ), magnesium and Ra-226.
- Initially when the tailings were placed underwater, the dissolved oxygen was rapidly consumed in the tailings substrate at shallow depths, and the tailings were anoxic below. With time, the un-oxidized tailings were slowly oxidizing at the tailings-water interfacing surface, mobilizing and releasing acidity, Mn and Pb in low concentrations to the upper lying water column. Iron was also mobilized in low concentrations, but it was rapidly oxidized to ferric ion, hydrolyzed and precipitated on the surface of the

tailings. The dissolved iron concentrations in the surface water were low ( 0.02 mg/l). Mobilization and release of other metals were not observed.

- A green filamentous algae had also covered the tailings on the surface and on all sides, oxygenating the tailings substrate, and gradually decreasing the anoxia and sulphate reduction which had existed previously.
- The porewater in un-oxidized tailings was also characterized mainly by dissolved gypsum, magnesium and Ra-226. It was unsaturated with respect to gypsum, having dissolved Ca in the range of 120 - 220 mg/l,  $\text{SO}_4^{-2}$  at 300 - 600 mg/l, Mg at 6 - 8 mg/l and Ra-226 at 2000 - 2500 mBq/l. The dissolved iron concentrations were higher, at ~ 1 - 2 mg/l, in the upper part of the tailings than near the bottom at ~ 0.5 mg/l, indicating oxidation and iron production at the surface. The porewater was only slightly acidic with pH approximately 5 - 6 and acidities 5 - 10 mg  $\text{CaCO}_3$ /l.
- For un-oxidized tailings, the calculated initial mass transfer flux ( $J_0$ ), and transfer coefficient (K), were, respectively, 11.9 mg/m<sup>2</sup>/h and 0.032 m/month for Ca, 29.7 mg/m<sup>2</sup>/h and 0.029 m/month for  $\text{SO}_4^{-2}$  and 1209 mBq/m<sup>2</sup>/h and 0.49 m/month for Ra-226. The transfer coefficients for Mg, Mn and Pb were, respectively, 0.042, 0.043 and 0.13 m/month. For Pb and Ra-226 the transfer coefficients were 4 and 16 times greater than those for Ca and  $\text{SO}_4^{-2}$ .
- In comparison to earlier diffusion studies of un-oxidized tailings, both the initial transfer flux and transfer coefficient for Ra-226 were higher, with  $J_0$  increasing from 650 to 1209 mBq/m<sup>2</sup>/h and K from 0.35 to 0.49 m/month. The surface oxidation of underwater deposited tailings and subsequent dissolution of Ra-226 from the oxidized surface layer, were significant contributors to the increased Ra-226 mobilization and its release to the water column.



- The mobilization and transfer of Ra-226 from underwater deposited tailings was controlled by gypsum dissolution when present.
- For weathered tailings underwater, the surface water quality was characterized by decreasing pH, from 4.9 to 2.9, and increasing acidity with time from 7 to 90 mg CaCO<sub>3</sub>/l. Concentrations of Ca, SO<sub>4</sub><sup>-2</sup> and Al increased moderately with time, and slow increases were observed in concentrations of total Fe, Mg and Mn in the water column above the tailings. Dissolved Pb concentrations increased significantly with time, from below detection to 2.5 mg/l. Concentrations of other metals, e.g. Ce, Cu, Ni, Th, U and Zn, etc., were below detection to very low in the water cover.
- Ra-226 concentrations increased very rapidly in the water cover above the weathered tailings, from initial low concentrations of approximately 670 mBq/l at the beginning of the experiment to peak values of approximately 32,000 mBq/l after three weeks, afterwards decreasing gradually to approximately 10,000 mBq/l as the SO<sub>4</sub><sup>-2</sup> level in the water cover increased. Ra-226 concentrations in the surface water were also significantly higher than those in the tailings porewater at intermediate and bottom depths, where low pH and high SO<sub>4</sub><sup>-2</sup> concentrations prevailed.
- The tailings porewater for weathered tailings was characterized by a low and acidic pH of ~ 3.0. It contained significant acidity at ~ 1700 mg CaCO<sub>3</sub>/l, and dissolved concentrations of SO<sub>4</sub><sup>-2</sup> at 3000 mg/l, Ca at 550 mg/l, Fe at 550 mg/l, Al at 110 mg/l, Mg at 15 mg/l Pb at 2 mg/l, Mn at 1.0 mg/l and Ra-226 in the range of 3000 - 10,000 mBq/l. The tailings porewater also contained trace amounts of Ce, Cu, Ni, Th and Zn. Uranium concentrations were below detection level at <0.2 mg/l.
- Except for Ca and Ra-226, the porewater concentrations of all other metals and acidity decreased gradually with time in the bottom part of the tailings.

- The calculated initial transfer flux,  $J_0$ , and transfer coefficient,  $K$ , for the weathered tailings were, respectively, 2.90 mg  $\text{CaCO}_3/\text{m}^2/\text{h}$  and  $1.11 \times 10^{-3}$  m/month for acidity, 0.34 mg/ $\text{m}^2/\text{h}$  and  $4.38 \times 10^{-4}$  m/month for Ca,  $1.35 \times 10^{-3}$  mg/ $\text{m}^2/\text{h}$  and  $1.73 \times 10^{-5}$  m/month for Fe,  $5.35 \times 10^{-4}$  mg/ $\text{m}^2/\text{h}$  and  $2.57 \times 10^{-5}$  m/month for Mg, 0.41 mg/ $\text{m}^2/\text{h}$  and 0.12 m/month for Pb, 3.72 mg/ $\text{m}^2/\text{h}$  and  $8.24 \times 10^{-4}$  m/month for  $\text{SO}_4^{2-}$  and  $5.68 \times 10^{-4}$  mBq/ $\text{m}^2/\text{h}$  and 1.92 m/month for Ra-226. The flux was primarily the result of mass dissolution at the tailings surface and the diffusion or mass transfer component from the tailings porewater was low.
- The transfer parameters for Ca, Mg, and  $\text{SO}_4^{2-}$  were all significantly lower (i.e. the transfer coefficients decreased by 2 to 3 orders of magnitude) than the corresponding values for the un-oxidized tailings. The transfer parameters for Pb were comparable for the two types of tailings.
- The iron transfer parameters for weathered tailings were low in well oxygenated and moderately acidic surface water. Upon entering the water cover, iron was readily oxidized, precipitated as hydroxide and removed at the solid - liquid interface boundary. An iron hydroxide and algae layer was formed at the surface covering the tailings, which was a sink for iron and perhaps for other ionic species as well. An iron sink layer was also forming on the surface of the un-oxidized tailings but its attenuating effects on other ionic species were not observed.
- In comparison to the un-oxidized tailings underwater, the Ra-226 transfer flux and coefficient increased by factors of approximately 50 and 4, respectively, for the weathered tailings. Initially, Ra-226 was released at a faster rate from the surface of the weathered tailings to the water column than diffusion from porewater, resulting in higher Ra-226 concentrations in the surface water than in the porewater. Afterwards, Ra-226 concentrations in the surface water decreased indicating a back, re-precipitation and/or adsorption.

- From porewater iron, sulphate and Ra-226 profiles, it was readily evident that any further oxidation of the previously weathered tailings underwater was very slow and limited to the near surface of the tailings. A similar situation also existed in the unoxidized tailings. In both cases, a sink layer of iron hydroxide precipitate was forming at the surface of the tailings but it had no attenuating or retarding effects on Ra and Pb transfer fluxes.
- Based on both the laboratory column leaching and diffusion lysimeter studies, it is concluded that the oxidation of pyritic uranium tailings underwater enhances Ra-226 release to the surface water. The study has confirmed the findings from part I and II that Ra-226 mobility is greatly enhanced when acidic conditions develop. Under acidic condition Ra-226 can be dissolved and mobilized together with its parent, Th-230 isotope, greatly increasing the transfer of Ra-226 from tailings to the water column. This further enforces the need to ensure submerged tailings basins do not become acidic.

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## 1. INTRODUCTION

For the CANMET - Rio Algom joint "Wet Barriers" research program, laboratory diffusion lysimeter studies were undertaken to determine diffusion related parameters for pyritic uranium tailings deposited under a shallow water cover. The studies consisted of measuring diffusion flux of dissolved minerals, oxidation reaction products and radionuclides across the solid - liquid interface, from un-oxidized and weathered tailings underwater.

The un-oxidized tailings were deposited fresh underwater in 1982, in the laboratory lysimeter, and have been under a 0.2 m water cover since. The weathered tailings were predominantly coarse and freshly deposited underwater in 1990 in a separate experimental lysimeter. For both tailings, the experiments were conducted in aquarium type lysimeters with well oxygenated surface water, and in the absence of both surface and pore water discharge flows. Previously in 1982-83, detailed diffusion related studies were undertaken for the un-oxidized tailings and the results have been reported by Davé and Lim (1983a and 1983b). These experiments were revisited, along with those for weathered tailings, in 1993-94, for measuring changes, if any, in the diffusion characteristics of the tailings with time, and compare them with those for the weathered tailings underwater.

When reactive tailings and/or waste rock are deposited underwater, or an insitu water cover is established on existing above grade and surface deposited wastes, mobilization of soluble minerals and oxidation reaction products, if present, occurs via both advective and diffusive processes to surface and groundwater regimes. An advection is caused by a hydraulic gradient resulting in porewater flow and the associated mass transport. A diffusion is caused by a concentration gradient resulting in diffusive mass flux along the gradient. Part II of the "Wet Barriers" study report dealt with the leaching, migration and advective aspects of pyritic uranium tailings under a shallow water cover and high porewater flow conditions. The present diffusion studies were conducted separately in an

experimental arrangement designed to provide an increased surface area of contact at the solid - liquid interface and simulations of surface and/or porewater flows. This report describes detailed diffusion results in the absence of a porewater flow.

## **2. EXPERIMENTAL METHODOLOGY**

The diffusion studies were conducted in clear Plexiglas™ aquarium lysimeters described in detail in the earlier study (Davé and Lim, 1983a and 1983b). The experimental arrangement is briefly described below.

### **2.1 Experimental Arrangement**

Figure 1 shows the experimental arrangement for the diffusion lysimeter. It consisted of a clear Plexiglas™ chamber, 1.22 m long, 0.3 m wide and 0.61 m high with a nominal wall thickness of 6.3 mm. A rigid 50 mm x 50 mm x 6.3 mm aluminum angle frame supported the Plexiglas™ side walls and bottom completing the chamber. The lower half of the chamber was filled with tailings to a height of 0.3 m and the top half with water providing the required water cover above the tailings, as shown in the figure. The height of the water column above the tailings was adjustable with an outflow height control. The lysimeter also had two side chambers with buffer plates in the upper part for mixing of the water column and providing a stream-line flow along the tailings - water interface. The water column was continuously mixed during the experimental period to prevent stratification and concentration gradient in the water column, and thermal eddy currents. The mixing of the water column provided a single concentration gradient normal to the interface. The baffle plates prevented turbulent water flow in other steady state experiments where a constant surface water flow above the tailings was required.

In the lower tailings section of the lysimeter, nine mini-piezometers and temperature probes were installed at three different depths along three vertical planes, one along the center and the other two along equally spaced left and right vertical planes. The top,

middle and bottom installations were approximately 0.025m, 0.15 m and 0.275 m below the tailings surface, respectively. The mini-piezometers were narrow diameter CPVC pipes, approximately 0.1 m long and 12.5 mm nominal diameter, perforated and covered with a filter material. They were placed horizontally in the middle, along the width of the lysimeter, and connected by Tygon™ tubing to appropriate fittings that were installed in the front face plate of the lysimeter for porewater sampling. The temperature probes were sealed copper - constantan thermocouple junctions, placed along the length of the piezometers for measuring temperature gradients in tailings, if any.

The lysimeters were designed for measuring mass diffusion or transport fluxes under two different experimental conditions of water flow: a steady-state constant flow and a no flow circulated water column. In the steady-state, a constant non-turbulent flow of water is maintained in the water column above the tailings. The total mass flux originating from the tailings to the water cover is transported by the flowing water, maintaining a constant gradient along the tailings water interface, and thus for a given flow, a steady state equilibrium condition is reached. For a given height of the water column above the tailings, measurements are taken for several steady-state equilibrium conditions by altering the rate of surface water flow.

In the no-flow circulated water column condition, the water column above the tailings is first completely flushed, then replaced with a fresh water column up to the desired height and the system is closed at time  $t = 0$ . The water column is then continuously homogenized by pumping water from the bottom of one mixing chamber to the top of other with a multi-head peristaltic or other pump, thereby mixing the water column and maintaining a horizontal cross-flow along the length of the water column. This provided minimal disturbance to the tailings surface, and at the same time, maintained a constant upwards concentration gradient between tailings porewater and the upper lying water column. The mass flux of a given species in the tailings porewater is determined by measuring its concentration increase as a function of time. These experiments are repeated for several water column heights.

In the present study, diffusion experiments were conducted with a no-flow circulated water column condition and a single water column height above the tailings surface. The height of the water column varied and was, respectively, 0.2 m and 0.15 m for lysimeters containing un-oxidized and weathered tailings.

## **2.2 Sampling and Analytical Procedures**

### **2.2.1 Tailings Sampling and Loading of Lysimeters**

As mentioned previously, the diffusion experiments were performed for two types of tailings, namely un-oxidized and weathered tailings. The un-oxidized tailings lysimeter (Aquarium #1) was initially set up in May, 1982, at the former location of the Elliot Lake Laboratory near the Nordic Mine Site, when the earlier diffusion studies were first performed. It was moved to the present new laboratory facility in January, 1986, and maintained in an operational condition since.

The un-oxidized tailings were obtained from Rio Algom's Quirke Mill tailings sampling line attached to the main tailings discharge pipeline. The tailings slurry, at 20 % solids, was allowed to settle in a sampling barrel and the supernatant liquid decanted off as much as possible prior to transferring the high solids tailings slurry to the lysimeter. In the lysimeter, the tailings were allowed to settle, the supernatant decanted and more tailings added to achieve the desired height of the settled tailings, and the supernatant liquid was further decanted and the tailings were leveled. The tailings were then covered with distilled water to the desired height of the water cover.

In the previous study, the experiments were performed with both steady-state flow and no-flow circulated water column conditions for a period of one year. Since then, the lysimeter has been maintained with a constant water cover and monitored periodically for its surface and porewater quality. During the initial period of 2 - 3 years, the tailings were anoxic throughout and below 1 - 2 cm of the top surface, and supported sulphate reducing

microbes evidenced by the presence of hydrogen sulphide gas in the porewater. Gradually, a green filamentous algae started to grow onto the tailings surface, on the sides and slowly covered the entire tailings surface, resulting in oxygenation of the tailings substrate which negatively impacted the previously established anoxic and sulphate reducing conditions.

During the past fourteen years or so, the tailings have oxidized on the surface and up to a depth ranging from 1 to 4 cm. As a result, and with the mobilization and hydrolysis of iron, an orange band of ferric hydroxide has formed onto the tailings surface. The porewater in these tailings has never been changed except its withdrawal and replacement during porewater sampling campaigns, where up to 500 ml of sample at a time is withdrawn from each port. Over the years, the tailings in the Aquarium #1 lysimeter have settled by 2 - 3 cm exposing the shallow, and immediately below the tailings surface, mini-piezometers (#1, 4 and 7). It has not impacted upon the lysimeter operation in any way except disfunctioning of the top porewater sampling ports.

The weathered coarse tailings were obtained from the West Arm tailings basin of the Quirke Waste Management Area during the summer of 1990. The tailings were collected from an exposed and weathered area in the vicinity of a former discharge point where significant oxidation had occurred. They were first homogenized by mixing in a clean cement mixer and then placed in a separate lysimeter (Aquarium #2) up to the desired height. Distilled water was gradually added to the lysimeter from the bottom sampling ports to displace the trapped air, the tailings allowed to settle, additional tailings added when required and leveled to the required elevation. After further settling and leveling, the lysimeter was filled with distilled water providing the desired height of the water cover. This lysimeter has been in operation since October, 1990, and several preliminary diffusion studies were undertaken in the beginning, but it took almost one year for the porewater to attain an equilibrium condition for its chemical constituents in relation to those in the tailings solid phase.

Initially, the surface water in Aquarium #2 was very acidic (pH approximately 2), when the water cover was first established, and contained ~ 300 - 400 mg/l of iron. Upon replacement of the water column with fresh distilled water for diffusion experiments, most of the iron entering the surface water cover hydrolyzed and precipitated forming an orange iron hydroxide coating layer onto the tailings surface. Since then, the iron concentration of the surface water had been very low. In the fall of 1993, a small sod of sphagnum peat moss was placed in the aquarium on a Styrofoam float for assessing its survival potential in the acidic surface water. This inadvertently introduced some organic matter and nutrients to the system promoting the growth of filamentous algae on top of the tailings. The final diffusion experiments were conducted under these active conditions of algae growth and iron deposition.

### **2.2.2 Water Sampling for Flux Measurements**

For the no-flow circulated water column condition, the following experimental procedure was followed for diffusion flux measurements. The lysimeter was first filled with distilled water to the desired water column height and its fill volume was determined. The water column was then homogenized by mixing for an hour and a pre-trial composite surface water sample was collected by sampling at various locations and depths. Porewater samples were also collected from all the nine ports and composited individually for each top, middle and bottom horizons by combining porewaters from ports in the same plane e.g. ports #1, 4, and 7 for the top horizon etc.

The surface water column was then decanted and refilled with fresh distilled water which was added slowly in the side chamber avoiding turbulence and erosion of the tailings surface. Upon filling to the desired height of the water cover, the mixing pump was immediately turned on and the system was isolated starting the experiment at time  $t = 0$  hour, designated as the starting time. At this time, a composite surface water sample was collected as above. The water column was again sampled at times:  $t = 1$  h, 5 h, 10 h, 24 h, 2 d, 5 d, 15 d, 30 d and at monthly intervals thereafter for approximately 8 months. The water column was always maintained at a constant height by adding distilled water,

when required, and prior to sampling. Porewater samples were also collected at regular intervals of one week initially and then once a month.

### **2.2.3 Analytical Procedures**

The water samples were analyzed for pH, redox potential (Eh), electrical conductance (Ec), total acidity, total alkalinity and dissolved concentrations of aluminum, calcium, cerium, copper, ferrous iron ( $\text{Fe}^{+2}$ ), total iron, magnesium, manganese, nickel, lead, thorium, uranium, zinc and Ra-226. The water samples were pre-filtered with 0.45  $\mu\text{m}$  membrane filters and stabilized with dilute nitric acid for dissolved metals and hydrochloric acid for sulphate and Ra-226 determinations. The acid was added to the sample in the amount of 30 ml per litre. The analytical procedures followed for primary parameters, dissolved metals, sulphate and Ra-226 were similar to those described in Part I and II of the accompanied Wet Barriers report.

## **3. THEORETICAL CONSIDERATIONS**

The underlying principles and theory of diffusion experiments for both steady-state and no-flow conditions have been discussed in detail in the earlier publications by Davé and Lim (1983a and b). The theory is briefly reviewed again here for continuity.

### **3.1 Continuous Flow - Steady-State Condition**

Assuming that there exists an equilibrium between tailings solid phase and porewater concentrations of a certain constituent such as Ra-226, Ca, total dissolved solids, etc., the mass transfer flux,  $J$ , from tailings porewater to the overlying water column above the tailings, in the absence of any porewater flow, for a given species is given by:

$$J = K(C_0 - C) \quad \text{Eq. 1}$$



where,

$J$  = mass transfer flux, mass per unit area per unit time, e.g. for Ra, Bq/m<sup>2</sup>/h,

$C_0$  = concentration of a given species in tailings porewater, e. g. for Ra, Bq/m<sup>3</sup>

and

$C$  = concentration of a given species in the surface water cover, e. g. for Bq/m<sup>3</sup>.

$K$  = transfer coefficient, m/h.

For a flow volume of  $Q$  m<sup>3</sup>/h, the concentration of a given chemical constituent in the surface water column above the tailings is give by the Equation:

$$J = \frac{CQ}{A} \quad \text{Eq. 2}$$

where,

$A$  = surface area of the tailings/water interface layer, m<sup>2</sup>.

Hence, by measuring concentrations in the tailings porewater,  $C_0$ , and the surface water cover,  $C$ , for a given flow  $Q$  in a steady-state condition, both  $J$  and  $K$  can be determined experimentally.

### 3.2 No-Flow Circulated Water Column Condition

In the case of a circulated water column of height 'h' m, above the tailings surface with no flow, the mass transfer flux,  $J(t)$ , is time-dependent given by:

$$J(t) = K[C_0 - C(t)] \quad \text{Eq. 3}$$

where,

$C(t)$  = time-dependent concentration of a given chemical species in the surface water cover above the tailings, e. g. for Ra, Bq/m<sup>3</sup>.

The rate of change of concentration  $C(t)$  from mass conservation is:

$$\frac{dC(t)}{dt} = \frac{\text{Flux} \times \text{Area}}{\text{Volume of water column}} = \frac{\text{Flux}}{\text{Effective height of water column}}$$

$$\text{i.e., } \frac{dC(t)}{dt} = \frac{J(t)}{h} = \frac{K}{h}(C_0 - C(t))$$

$$\text{i.e., } \frac{dC(t)}{dt} + \frac{K}{h}C(t) - \frac{K}{h}C_0 = 0 \quad \text{Eq. 4}$$

which is a first order differential equation with solution of the form:

$$C(t) = A + Be^{-\alpha t} \quad \text{Eq. 5}$$

Applying the boundary conditions:

$$C(t) = 0 \text{ at } t = 0$$

$$\text{and } C(t) \rightarrow C_0 \text{ at } t \rightarrow \infty$$

the time-dependent solutions for  $C(t)$  and  $J(t)$  are given by:

$$C(t) = C_0 \left[ 1 - \exp\left(-\frac{K}{h} \times t\right) \right] \quad \text{Eq. 6}$$

$$J(t) = J_0 \exp\left(-\frac{K}{h} \times t\right) \quad \text{Eq. 7}$$

where,

$$J_0 = KC_0, \text{ is the flux at time, } t = 0 \quad \text{Eq. 8}$$

From measurements of  $C(t)$  as a function of time,  $K$ ,  $C_0$  and  $J_0$  can be determined experimentally.

The molecular diffusion flux from tailings to the upper lying water columns can also be calculated from the observed concentration gradient at the interface as:

$$J_{\text{diff}} = -\Phi D \left. \frac{dC}{dZ} \right|_{Z=0} \quad \text{Eq. 9}$$

where,

$\Phi$  = effective porosity of tailings, taken as a product of porosity,  $\eta$ , and tortuosity,  $\tau$ ,

$J_{\text{diff}}$  = diffusion flux in the direction opposite to the increasing depth  $Z$ ,  $\text{mg}/\text{m}^2/\text{s}$

$\frac{dC}{dZ}$  = concentration gradient at the interface,  $\text{mg}/\text{m}^4$

and,  $D$  = molecular diffusion coefficient for a given ionic species in dilute solutions,  $\text{m}^2/\text{s}$

Experimentally, the transfer flux and coefficient of a given chemical species are determined by fitting its observed surface water concentration,  $C(t)$ , as a function of time to a non-linear function of the form:

$$C(t) = C_{01} - C_{02} \exp\left(-\frac{K}{h} \times t\right) \quad \text{Eq. 10}$$

Using a non-linear least square fit regression technique, the best fitted values for the parameters,  $C_{01}$ ,  $C_{02}$  and  $\frac{K}{h}$  are determined. The initial flux,  $J_0$  at time  $t = 0$  is calculated as:

$$J_0 = \frac{K(C_{01} + C_{02})}{2} \quad \text{Eq. 11}$$

These expressions were used in this study to calculate both the mass transfer flux and coefficient, as well as the diffusional component of the molecular flux for the various ionic species monitored.

#### 4. RESULTS

The surface and porewater quality results for un-oxidized tailings underwater in the laboratory since 1982 (Aquarium #1) are given in Tables 1 and 2, respectively. Figures 2 - 13, show the variations in surface water pH, Eh, Ec, total acidity, total alkalinity and concentrations of dissolved  $\text{SO}_4^{-2}$ , total Fe, Ca, Mg, Mn, Pb, and Ra-226 with time, respectively. The corresponding parameters for tailings porewater are shown in Figures 14 - 25.

The surface water above the tailings was slightly acidic with a pH of  $\sim 4$ , acidity in the range of 5 - 20 mg/l, and well oxygenated having Eh in the range of 600 - 650 mV. It was characterized by significant diffusional or mass transport fluxes of gypsum, magnesium and Ra-226, as seen from Figures 4, 7, 9, 10 and 13 for Ec,  $\text{SO}_4^{-2}$ , Ca, Mg and Ra-226, respectively. The results further indicated a slow oxidation at the tailings surface and release of Mn and Pb to the upper lying water column. Because of hydrolysis and precipitation of ferric ion, the dissolved iron concentration in the surface water was low at  $< 0.02$  mg/l, but it contributed to slightly increased acidity and depressed pH.

Similar to the surface water, the porewater in the previously un-oxidized tailings was mainly characterized by dissolved gypsum, magnesium and Ra-226. It was also undersaturated with respect to gypsum having dissolved concentrations in the ranges of 120 - 220 mg/l for Ca, 300 - 600 mg/l for  $\text{SO}_4^{-2}$ , 6 - 8 mg/l for Mg and 2000 - 2500 mBq/l for Ra-226. The dissolved iron concentrations were higher, at 1 - 2 mg/l, in the upper part of the tailings than near the bottom, at  $\sim 0.5$  mg/l, indicating iron production below the tailings surface as a result of slow oxidation (Figure 20). Similarly, porewater manganese concentrations were slightly elevated in the middle part of the tailings (Figure 23). Lead concentrations were elevated near the surface of the tailings.

The tailings porewaters were only slightly acidic in the middle and bottom zones with pH's of ~ 5 - 6, acidities ~ 5 - 10 mg/l and Eh's ~ 400 mV.

The porewater electrical conductance and concentrations of Ca, Mg, Mn and  $\text{SO}_4^{-2}$  also increased slowly with time. Ra-226 concentrations were practically constant, in the range 1500 - 2000 mBq/l, throughout the experimental period except in the very beginning where they were slightly low in the bottom half of the tailings.

The average height of the water column above the un-oxidized tailings in Aquarium #1 was approximately 0.22 m. Including the 33 % additional volume of water contained in the mixing side-chambers, the effective total water column height was estimated as 0.28 m. The results of a non-linear regression analysis of surface water concentration for Ca, Mg, Mn, Pb,  $\text{SO}_4^{-2}$  and Ra-226 as a function of time, are given in Table 3. The dilute solution diffusion coefficients for these parameters were also computed using the observed concentration gradients in the tailings and assuming an effective porosity of approximately  $\phi = 0.45$  for underwater deposited and somewhat compacted tailings. The effective porosity included a tortuosity ( $\tau$ ) factor of approximately 0.9 and bulk porosity factor of 0.5 for underwater disposed tailings. Because of low acidity and total dissolved iron concentrations in the surface water column, no attempt was made in further analyzing the data for these parameters.

For weathered tailings underwater (Aquarium #2), the surface and porewater quality results are given in Tables 4 and 5, respectively. Figures 26 - 36 show, respectively, the variations in surface water pH, Eh, Ec, total acidity, and concentrations of dissolved  $\text{SO}_4^{-2}$ , total Fe, Ca, Mg, Mn, Pb, and Ra-226 with time. The corresponding parameters for tailings porewater are shown in Figures 37 - 47, respectively.

The surface water quality above the weathered tailings, in a shallow water scenario, was characterized by a decreasing pH from 4.9 to 2.9, oxidative Eh's in the range, 600 - 770 mV and increasing acidity with time, from ~ 7 to 90 mg  $\text{CaCO}_3$ /l (Figures 26, 27 and 29).

There was moderate increase with time in the electrical conductance (Ec), and concentrations of Ca,  $\text{SO}_4^{-2}$  and aluminum (Figures 28, 30 and 32). Total Fe, Mg, and Mn concentrations increased slowly with time in the surface water column. Dissolved Pb concentrations increased significantly (Figure 35) with time from below detection to an equilibrium value of 2.5 mg/l.

The most significant change was observed in Ra-226 concentration, which increased very rapidly from an initial low concentration of  $\sim 670$  mBq/l at the beginning of the experiment to a peak value of approximately 32,000 mBq/l after three weeks, and decreased gradually to 10,000 mBq/l near the end of the seven month experimental period. The surface water Ra-226 concentrations were also significantly higher than those for tailings porewaters at intermediate and bottom depths (Figures 36 and 47).

For the weathered tailings, the porewater was characterized by a low acidic pH of  $\sim 3.0$  throughout the tailings depth and with a decreasing Eh, from 600 mV near the surface to 400 mV at the bottom of the tailings (Figures 37 and 38). It contained significant acidity at  $\sim 1700$  mg  $\text{CaCO}_3$ /l, and dissolved concentrations of  $\text{SO}_4^{-2}$  at 3000 mg/l, Ca at 550 mg/l, Fe at 550 mg/l, Al at 110 mg/l, Mg at 15 mg/l, Pb at 2 mg/l, Mn at 1 mg/l, and Ra-226 at 3000 mBq/l at the beginning, which decreased with the diffusional flux from bottom to the surface of the tailings. Except Ca and Ra-226, the porewater concentrations of all other metals and acidity decreased gradually with time near the bottom of the tailings (see Figures 40 - 47 for examples). Because of previous oxidation, the tailings porewater also contained trace amounts of Ce, Cu, Ni, Th and Zn. Uranium concentrations were below detection limit, at  $< 0.2$  mg/l.

The average height of the water column above the weathered tailings in Aquarium #2 was approximately 0.15 m, which corresponded to an effective water column height of approximately 0.19 m. Similar to the un-oxidized tailings, the non-linear regression analysis results for acidity, Ca, Fe, Mg, Pb,  $\text{SO}_4^{-2}$  and Ra-226 for the surface water

column above the weathered tailings are given in Table 6. Because of low surface water concentration of Manganese, its data were not further analyzed.

The surface of the weathered tailings was also characterized by a yellowish, orange layer, 0.5 cm in thickness, comprised of ferric hydroxide precipitates and overlain by a 1.0 - 1.5 cm thick organic cover containing filamentous algae. With time, the algal matter had also been covered by the iron hydroxide precipitate.

## 5. DISCUSSION

The un-oxidized tailings were deposited underwater in 1982, in the laboratory lysimeter, and have been under a continuous shallow water cover since. In all experiments to date, only distilled water was used for water cover purposes. This represented enhanced chemical activity condition as the distilled water was always well aerated, had slightly depressed pH in the range of ~ 5 - 5.5, because of dissolved carbon dioxide, and very low buffering capacity. The tailings porewater contained the original mill process water, saturated with respect to gypsum, which had never been completely replaced. Over the years, it had been mixed and diluted somewhat with the distilled water from the above lying water cover whenever porewater samples were withdrawn.

At the completion of the initial experiments in 1983, the surface water cover had a pH of 5.5 and it contained mostly dissolved Ca,  $\text{SO}_4^{-2}$  (gypsum) and Ra-226. The porewater was anoxic and saturated with respect to gypsum containing Ca,  $\text{SO}_4^{-2}$  and Ra-226 on the order of 570 mg/l, 2680 mg/l and 3900 mBq/l, respectively. The experimentally determined values of the initial transfer flux  $J_0$  at  $t = 0$  and transfer coefficient  $K$  were, respectively 26  $\text{mg/m}^2/\text{h}$  and 0.186 m/months for Ca, 96  $\text{mg/m}^2/\text{h}$  and 0.035 m/month for  $\text{SO}_4^{-2}$  and 650  $\text{mBq/m}^2/\text{h}$  and 0.34 m/month for Ra-226. It was also observed that the Ra-226 transfer flux was approximately one order of magnitude higher than the calculated molecular diffusional flux obtained by the experimentally determined concentration

gradient at the tailings-water interface using the literature reported diffusion coefficient ( $D_0 = 8.89 \times 10^{-10} \text{ m}^2/\text{s}$ ) for Ra-226 in dilute solutions.

With time, the previously un-oxidized tailings in Aquarium #1 had slowly oxidized at the top forming an orange colour oxidation and iron precipitation band, 1 - 4 cm in height, at the interface. A green filamentous algae had covered the tailings at the top and on all sides of the aquarium including its walls. It had also oxygenated the tailings substrate and significantly decreased the anoxia and sulphate reduction which had existed previously.

Although the tailings were oxidizing slowly underwater, the oxidation rate was too low to adversely affect the surface water quality except for a slight increase in its acidity in the range of  $\sim 10 - 12 \text{ mg/l}$  and corresponding decrease in pH to  $\sim 4.4$ . As a result, the near surface porewater iron and manganese concentrations were also slightly elevated and were in the ranges of  $1 - 2 \text{ mg/l}$  and  $0.2 - 0.4 \text{ mg/l}$ , respectively. The hydrolysis and precipitation of iron at the interface prevented further migration of iron to the upper lying water column, but some diffusion of manganese was observed. The surface water quality was still dominated by porewater gypsum and Ra-226 concentrations and their diffusion across the solid - liquid interface.

The average porewater concentrations of Ca,  $\text{SO}_4^{-2}$  and Ra-226 at the beginning of these experiments were respectively,  $250 \text{ mg/l}$ ,  $680 \text{ mg/l}$  and  $2300 \text{ mBq/l}$ . The computed values in the present experiment for initial transfer flux,  $J_0$ , and transfer coefficient,  $K$ , were  $11.9 \text{ mg/m}^2/\text{h}$  and  $0.032 \text{ m/month}$  for Ca,  $29.7 \text{ mg/m}^2/\text{h}$  and  $0.029 \text{ m/month}$  for  $\text{SO}_4^{-2}$  and  $1209 \text{ mBq/m}^2/\text{h}$  and  $0.49 \text{ m/month}$  for Ra-226. Both Ca and  $\text{SO}_4^{-2}$  ions in the water cover were gypsum derived as seen from their initial flux correspondence and comparable transfer coefficients. For Mg and Mn, the transfer coefficients were, respectively,  $0.042$  and  $0.043 \text{ m/month}$ , which were higher than those for Ca and  $\text{SO}_4^{-2}$ , but similar order of magnitude. The transfer coefficients for Pb and Ra-226 were  $0.13$  and  $0.49 \text{ m/month}$ , respectively, which were approximately 4 and 16 times greater than those for Ca and



SO<sub>4</sub><sup>-2</sup>. The initial flux for these species, however, depended upon their porewater concentration.

The diffusion or mass transfer flux from underwater deposited tailings differed from that of a simple ionic solution contained in a non-interactive porous media. In the former case, soluble minerals and oxidation reaction products, if any, present in the tailings contribute to additional source terms in the diffusion equation maintaining higher concentration gradients and, hence, increased mass transfers. The tailings porewater is in a dynamic state of equilibrium with its solute matrix, and depending upon the mineral dissolution rate versus diffusion, a constant or lower equilibrium concentration is maintained. For a simple ionic solution contained in a non-interactive pore media, the source term is typically absent and the porewater concentration gradients are controlled by first order molecular diffusion.

For un-oxidized tailings, the porewater Ca, Mg, Mn and SO<sub>4</sub><sup>-2</sup> concentrations profiles differed significantly from those of Fe, Pb and Ra-226. For the first group, the porewater concentration increased with depth as well as with time (Figures 19, 21 - 23) indicating lower mineral solubility and dissolution rates than diffusion. The porewater iron concentrations were higher in the upper part of the tailings and increased with time than near the bottom where they decreased as shown in Figure 20. This indicated iron production as a result of oxidation in the upper part of the tailings and its outward diffusion. The porewater lead profiles were similar to those of iron except lead concentrations were higher in the water very close to the tailings surface indicating near surface solute dissolution and, hence, increased transfer coefficient (Figure 24).

The porewater Ra-226 concentration profiles exhibited a classic case of high Ra-226 solubility and dissolution rate from the solid phase than molecular diffusion in the aqueous phase, contributing to constant equilibrium porewater concentrations as a function of time and hence increased transfer flux across the boundary (Figure 25). Similar to the finding in the earlier study, the dissolution of Ra-226 from the near surface

layer of the tailings was a significant contributor to Ra-226 transfer flux and coefficient. In comparison to the earlier study, both the Ra-226 transfer coefficient and initial transfer flux were higher, with K increasing from 0.35 to 0.49 m/month and  $J_0$  from 650 to 1209 mBq/m<sup>2</sup>/h, although the porewater Ra-226 concentration had decreased from approximately 3900 mBq/l to 2300 mBq/l in this study. Perhaps, the slow oxidation of tailings underwater was a contributor to the increased Ra-226 mobilization and its release to the upper lying water column. Similar results were obtained for weathered tailings underwater, as discussed below, and in leaching studies of un-oxidized coarse tailings underwater reported in Part II of the Wet Barriers Study, where increased mobilization of Ra-226 was observed with the oxidation and development of acidic conditions at the surface of the tailings.

For Ca and SO<sub>4</sub><sup>-2</sup>, the transfer parameters, K and  $J_0$ , from tailings porewater to the water column have decreased in the present study. The transfer coefficient and flux for Ca decreased from 0.186 m/month in the earlier study to 0.032 m/month and from 26 to 11.9 mg/m<sup>2</sup>/h, respectively. The corresponding decrease in SO<sub>4</sub><sup>-2</sup> parameters was 0.035 to 0.029 m/month for the transfer coefficient, although small, and from 96 to 29.7 mg/m<sup>2</sup>/h for the transfer flux. In both cases, the reduction in the transfer flux was caused by their decreasing porewater concentrations. In the earlier study, the initial transfer parameters were measured by the steady state flow method, where, in the presence of gypsum saturation, the Ca transfer coefficient was high.

Similar to column leaching results for unsaturated and submerged uranium tailings reported in Part I and Part II of the Wet Barriers study, the mobilization and transfer of Ra-226 from underwater deposited tailings was controlled by gypsum dissolution when present. It is also believed that the calculated Ra-226 transfer parameters were realistic equilibrium values for such tailings that have been under a continuous water cover for a long time. These results would be equally applicable to an actual field scenario of water covered tailings when gypsum saturation existed.

For weathered tailings underwater, the surface water was acidic with a pH of  $\sim 3.0$ . The porewater was moderately acidic containing approximately 1800 mg  $\text{CaCO}_3/\text{l}$  acidity, but the measured acidity flux to the upper lying water column and its transfer coefficient were low at  $J_0 = 2.90 \text{ mg CaCO}_3/\text{m}^2/\text{h}$  and  $K = 1.11 \times 10^{-3} \text{ m/month}$ . The latter value was lower than those obtained for various ionic species and Ra-226 for the un-oxidized tailings underwater. It was less than 4 % of the transfer coefficients for Ca and  $\text{SO}_4^{-2}$  in Aquarium #1. The transfer coefficient (K) and initial flux ( $J_0$ ) for other ionic species were;  $4.38 \times 10^{-4} \text{ m/month}$  and  $0.34 \text{ mg/m}^2/\text{h}$ , respectively, for Ca,  $1.73 \times 10^{-5} \text{ m/month}$  and  $1.35 \times 10^{-3} \text{ mg/m}^2/\text{h}$  for Fe,  $2.57 \times 10^{-5} \text{ m/month}$  and  $5.35 \times 10^{-4} \text{ mg/m}^2/\text{h}$  for Mg,  $0.121 \text{ m/month}$  and  $0.413 \text{ mg/m}^2/\text{h}$  for Pb,  $8.24 \times 10^{-4} \text{ m/month}$  and  $3.72 \text{ mg/m}^2/\text{h}$  for  $\text{SO}_4^{-2}$  and  $1.92 \text{ m/month}$  and  $5.68 \times 10^{+4} \text{ mBq/m}^2/\text{h}$  for Ra-226 for the weathered tailings (Table 6). The transfer parameters for Ca, Mg and  $\text{SO}_4^{-2}$  were all significantly lower (e.g. the transfer coefficient decreased by 2 to 3 orders of magnitude) than the corresponding values for the un-oxidized tailings.

Although the dissolved total iron concentrations in the weathered tailings porewater were high, at approximately 560 mg/l, upon entering the surface water, iron was rapidly hydrolyzed, precipitated and removed at the solid - liquid interface boundary. Thus, the iron transfer parameters were low in well oxygenated and moderately acidic surface water. The interface boundary layer was a sink for iron and, perhaps, for other ionic species as well, which needed further investigation. An iron sink layer was also forming on the surface of the un-oxidized tailings but its attenuating effects on other ionic species was not observed.

The transfer parameters for Pb were comparable for the two types of tailings, where again the dissolved Pb concentrations were higher at shallow depths in the tailings than those at the bottom (Figure 46). For weathered tailings, the Ra-226 concentrations were the highest in the surface water and decreased in the tailings porewater with depth (Figures 36 and 47). In comparison to the un-oxidized tailings, the Ra-226 transfer coefficient and flux increased by factors of approximately 4 and 50, respectively, for the weathered case.

Initially, Ra-226 was released at a faster rate from the oxidized surface layer to the water column than molecular diffusion from the porewater, but gradually a new solid - liquid equilibrium was established where, perhaps, a back-diffusion and re-precipitation or adsorption of Ra-226 were taking place. The growth of algae could have been also a factor in decreasing the surface water Ra-226 concentration with time, but this aspect was not further evaluated. Also, because of the altering nature of the surface water Ra-226 profiles, first increasing to a peak concentration and then decreasing, the model was inappropriate in fitting the observed data. Consequently, the calculated transfer parameters at  $t = 0$  have been underestimated, and further refinement of the model is needed.

From porewater iron, sulphate and Ra-226 profiles, it was also readily evident that any further oxidation of previously weathered tailings underwater was very slow and limited to the near surface of the tailings, as was the case for the un-oxidized tailings. In both cases, an iron hydroxide precipitation and sink layer was forming at the interface boundary, but it had no attenuating or retarding effect on Ra and Pb transfer fluxes.

It is also believed that for weathered tailings deposited underwater, the surface water Ra-226 concentrations would eventually decrease with time to their equilibrium porewater concentrations of approximately 10,000 mBq/l.

Similar to observations made in the column leaching studies of un-oxidized coarse tailings underwater and reported in Part II of the Wet Barriers study, it is necessary for long term management of underwater deposited pyritic uranium tailings to ensure that an adequate water cover is maintained and the tailings surface is protected from developing acidic conditions for controlling increased mobility and release of Ra-226.

## 6. SUMMARY AND CONCLUSIONS

Laboratory diffusion studies were undertaken for determining diffusion related transfer parameters for un-oxidized and weathered pyritic uranium tailings underwater. The results are summarized as follows:

- The water cover on the un-oxidized tailings was characterized by slightly acidic pH, low acidity and significant diffusion of gypsum, magnesium and Ra-226.
- The tailings were slowly oxidizing underwater at the surface, releasing acidity, Mn and Pb in low concentrations to the water column. Iron was also mobilized in low concentration, but it was rapidly oxidized to ferric ion, hydrolyzed and precipitated on the surface of the tailings. An iron sink layer was forming at the tailings - water interface.
- The porewater in un-oxidized tailings was also characterized mainly by dissolved gypsum, magnesium and Ra-226. It was only slightly acidic and unsaturated with respect to gypsum.
- The transfer coefficients for Pb and Ra-226 were, respectively, 4 and 16 times greater than those for Ca and  $\text{SO}_4^{-2}$ . The dissolution of Ra-226 from the near surface layer of the tailings and its oxidation, were significant contributors to the increased Ra-226 mobilization and its release to the water column.
- The mobilization and transfer of Ra-226 from underwater deposited tailings were controlled by gypsum dissolution when present, and hence by  $\text{SO}_4^{-2}$  ion concentration.
- The surface water on weathered tailings was characterized by a low pH, moderate acidity and moderate concentrations of Ca,  $\text{SO}_4^{-2}$  and Al, and low concentrations of Fe, Mg and Mn. Pb concentrations increased significantly in the water cover. The

concentrations of other metals, e.g. Ce, Cu, Ni, Th, U and Zn, etc., were below detection levels to very low in the water cover.

- Ra-226 concentrations in the water cover increased significantly and were higher than those in the tailings porewater at intermediate and bottom depths where low pH and high  $\text{SO}_4^{-2}$  concentrations prevailed.
- The tailings porewater in weathered tailings was characterized by low and acidic pH, high acidity and high concentrations of Ca,  $\text{SO}_4^{-2}$ , Al, Fe, Al, Mg, Mn, Pb and Ra-226. It also contained trace amounts of Ce, Cu, Ni, Th and Zn. U concentrations were below detection.
- The transfer parameters for Ca, Mg and  $\text{SO}_4^{-2}$  were all significantly lower than the corresponding values for the un-oxidized tailings. The transfer parameters for Pb were comparable for the two tailings.
- The iron transfer parameters for the weathered tailings were also low in the well oxygenated and moderately acidic surface water. Upon entering the water cover, it was precipitated at the interface boundary forming a sink layer which may be attenuating the transfer of other ionic species as well. The sink layer had no attenuating or retarding effects on Pb and Ra-226 transfer parameters.
- In comparison to the un-oxidized tailings underwater, the Ra-226 transfer flux and coefficients increased by factors of approximately 50 and 4, respectively, for the weathered tailings.
- Similar to un-oxidized tailings, further oxidation of previously weathered tailings underwater was very slow and limited to the near surface of the tailings.

Based on both the laboratory column leaching and diffusion lysimeter studies, it is concluded that a shallow water cover on pyritic uranium tailings controls oxidation to a slow rate at the tailings surface. Development of acidic conditions within the tailings substrate contributes to increased mobility and release of Ra-226, and should be avoided.

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- Davé, N. K. and Lim, T. P. 1983b. Migration of Contaminants from Underwater Buried Uranium Tailings - Laboratory Investigations of Ra-226, Total Dissolved Solids,  $\text{Ca}^{+2}$  and  $\text{SO}_4^{-2}$  Transfer Flux. Division Report MRP/MRL 83-105 (OP) (J). Also in the Proceedings, Sixth Annual Symposium on Management of Uranium Mill Tailings, Low Level Waste and Hazardous Waste, Fort Collins, Colorado, USA, February, 1984, pp. 83 - 92.

## **FIGURES**

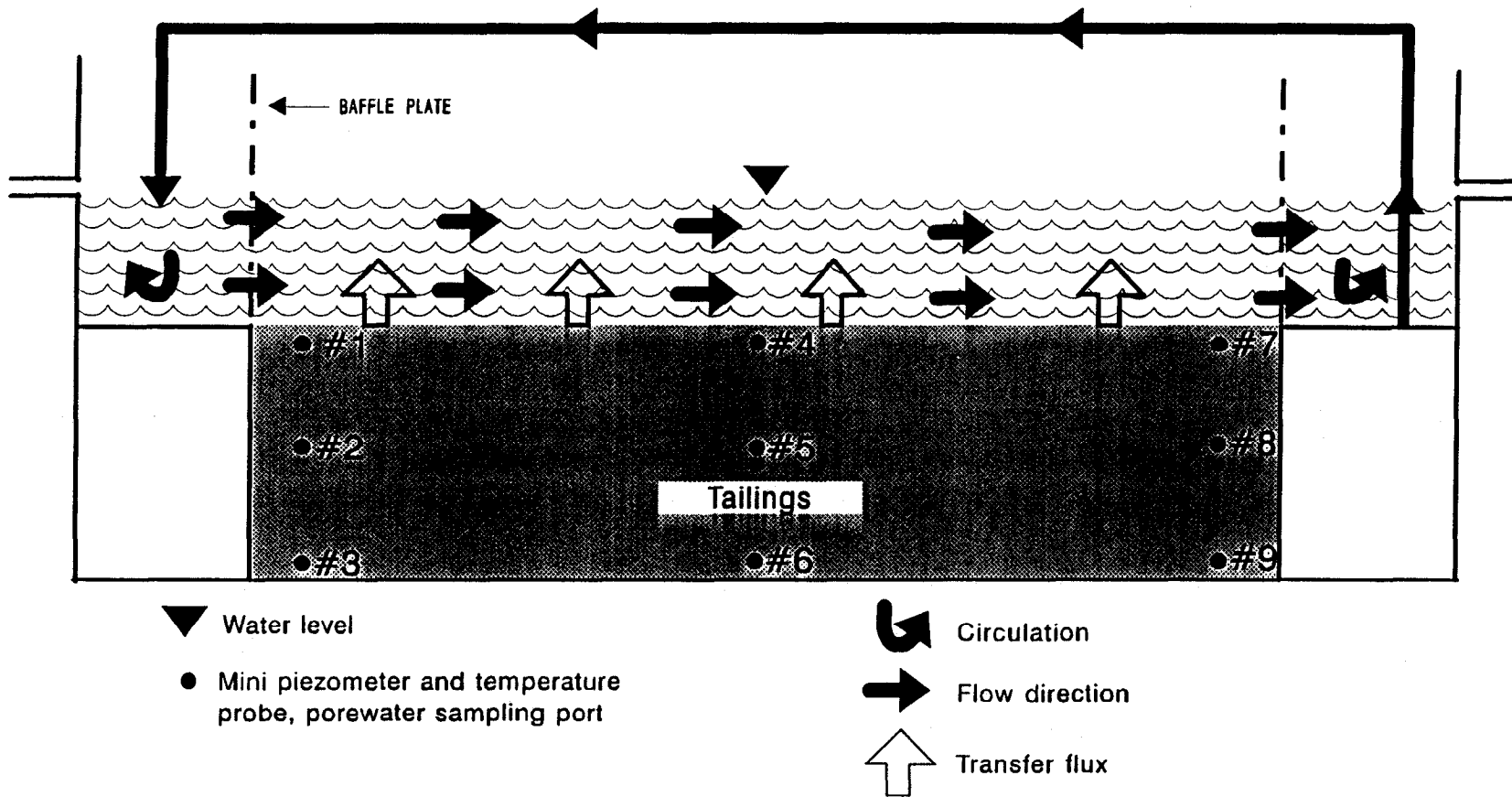


Fig. 1 Experimental arrangement of the diffusion lysimeter under a circulated water column condition in the absence of any water flow through the system.

Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Surface Water pH vs Time

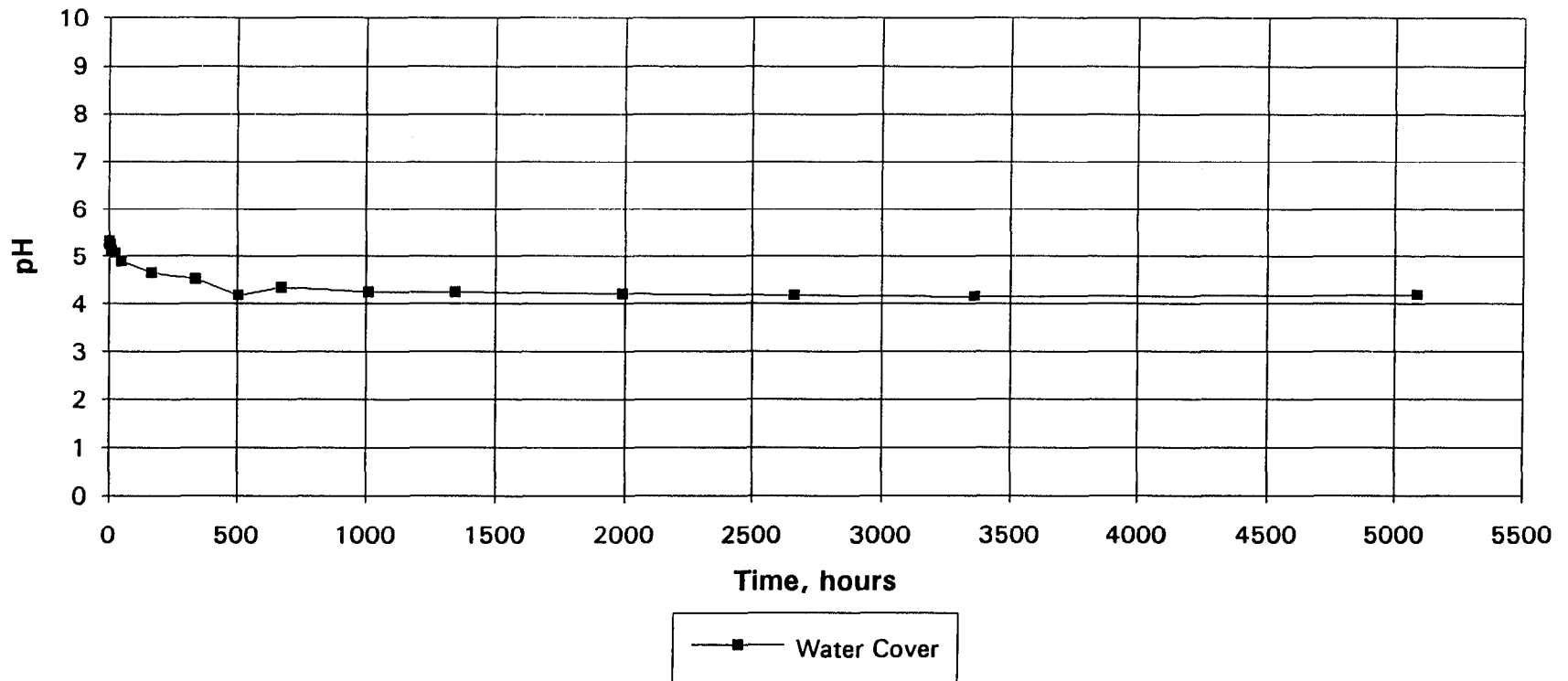


Fig. 2 Variation of pH with time in the surface water cover of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Surface Water Eh(NHE) vs Time**

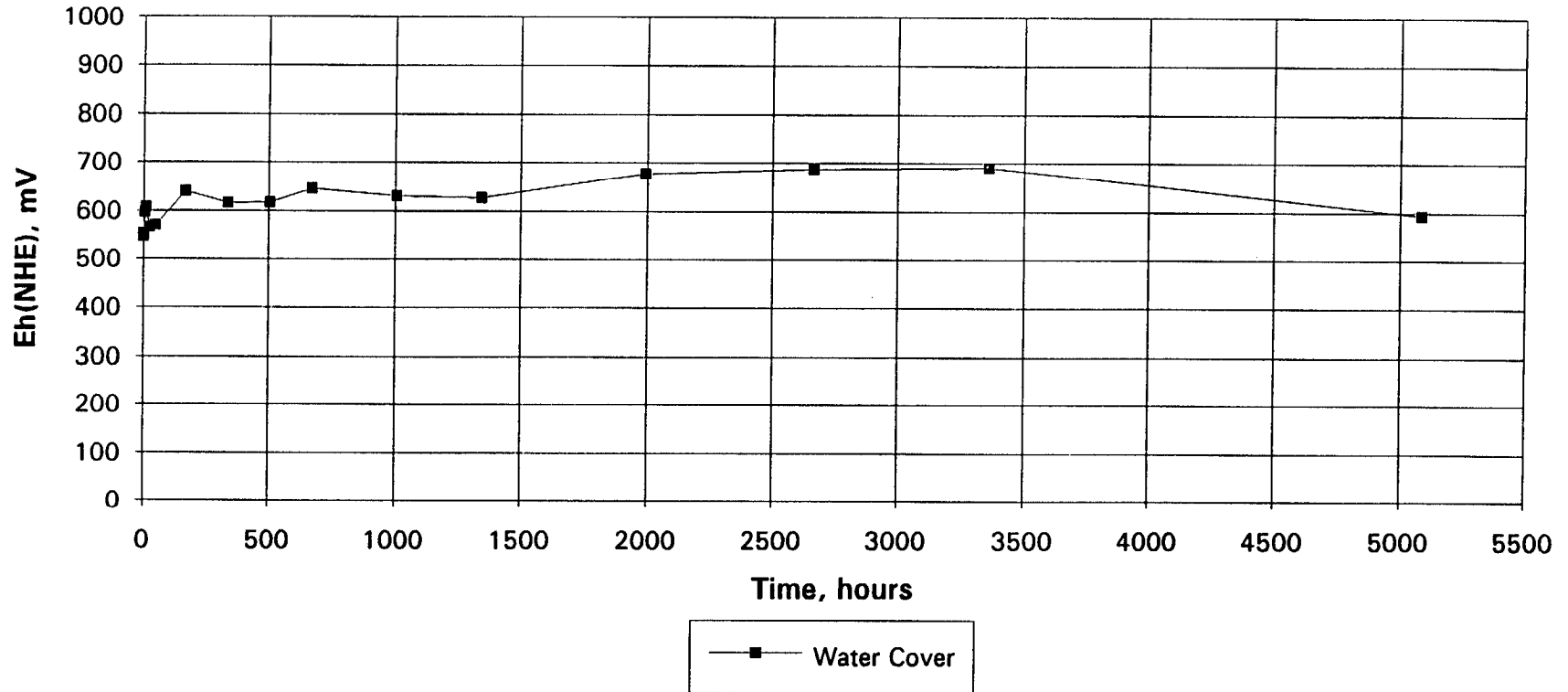
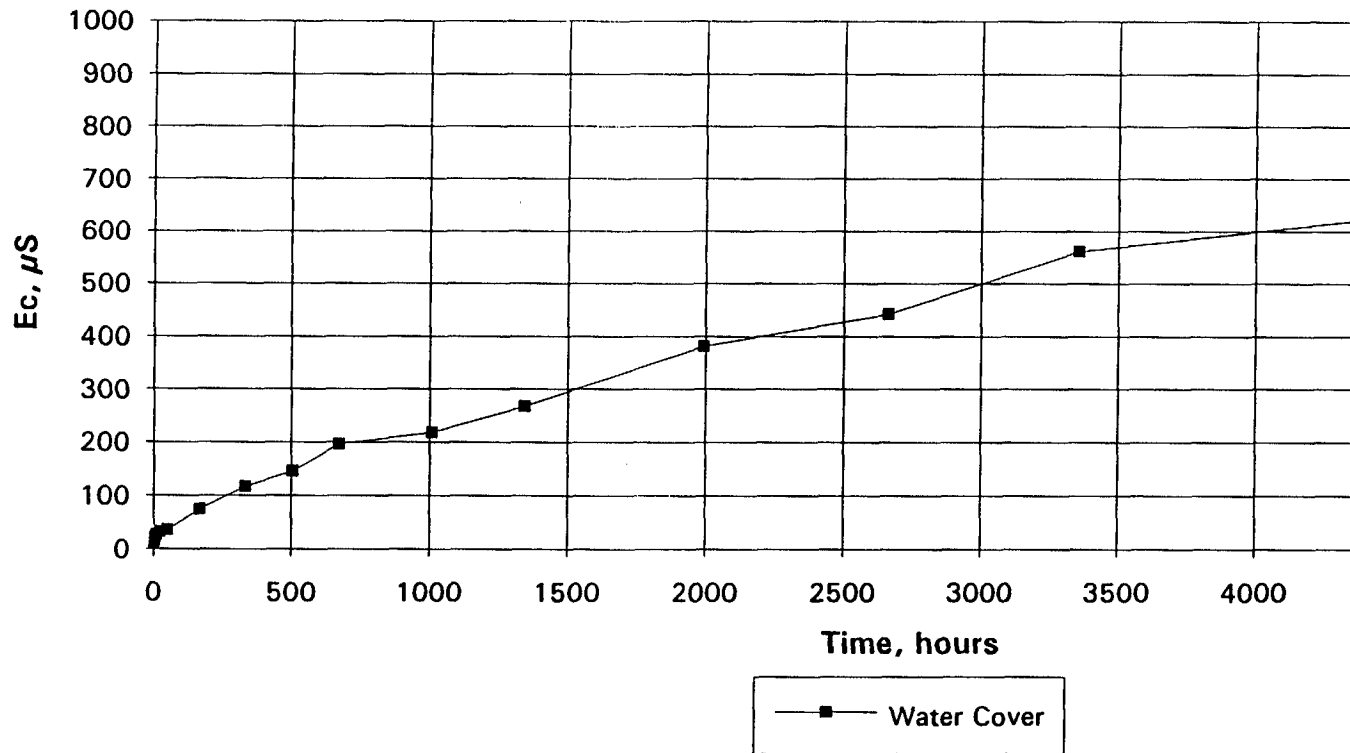


Fig. 3 Variation of Eh with time in the surface water cover of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Surface Water Electrical Conductivity vs Time**



**Fig. 4** Variation of Ec with time in the surface water cover of un-oxidized tailings und

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Surface Water Acidity vs Time**

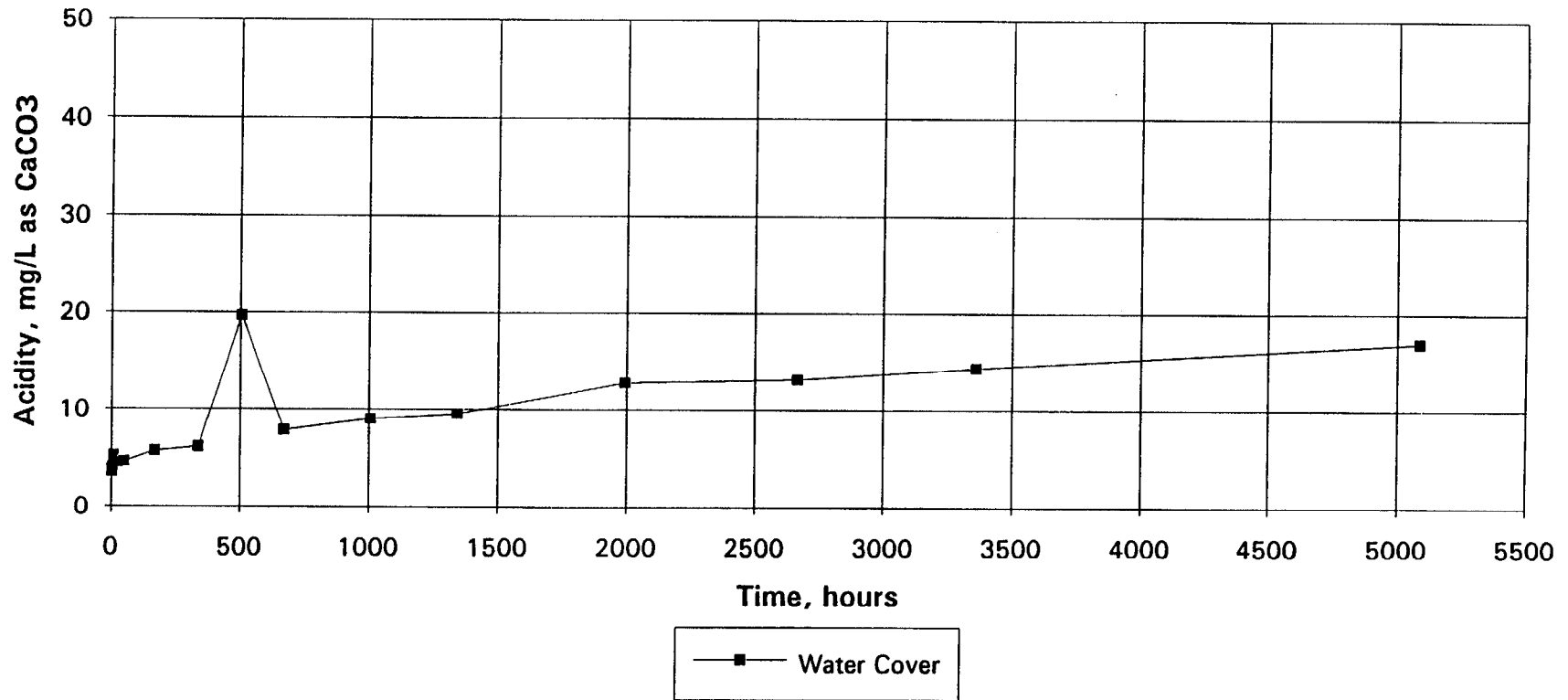


Fig. 5 Variation of acidity with time in the surface water cover of un-oxidized tailings underwater.

Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Surface Water Alkalinity vs Time

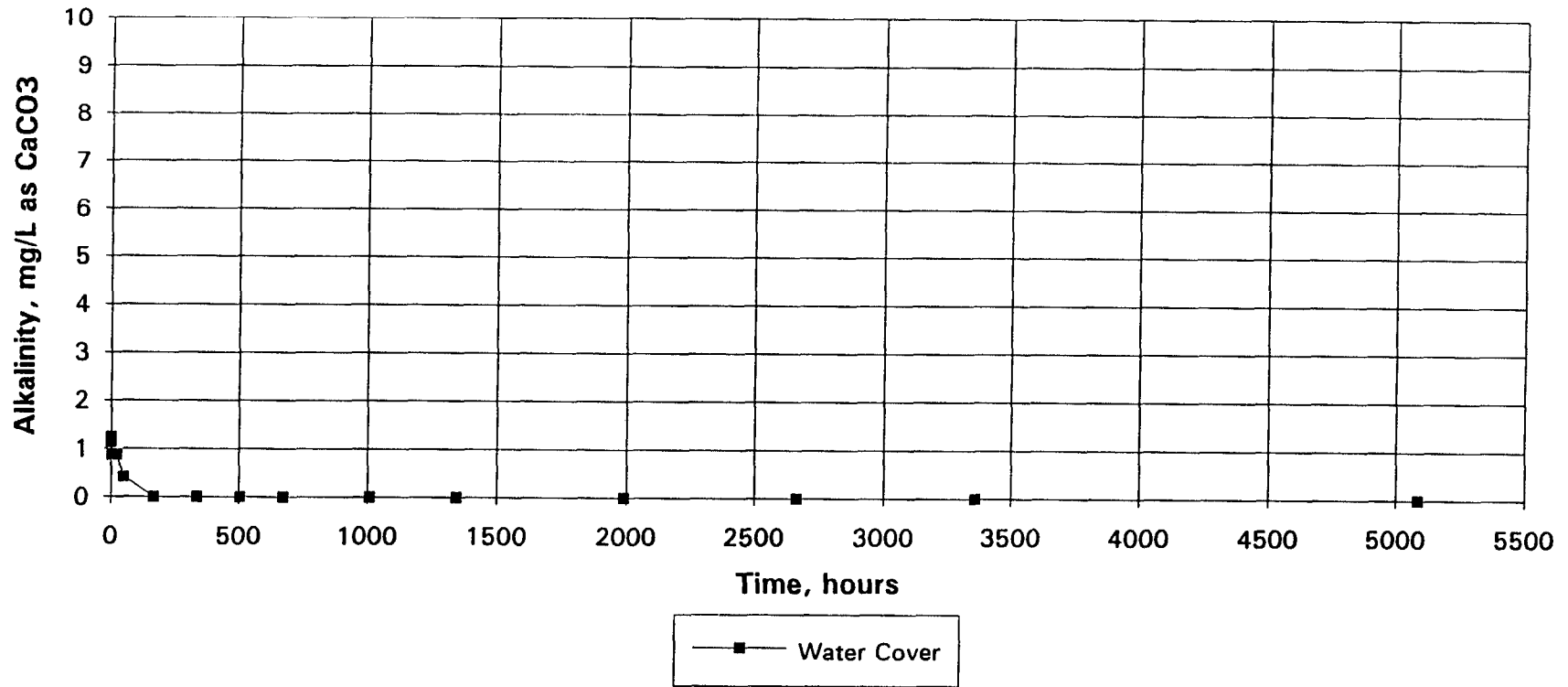
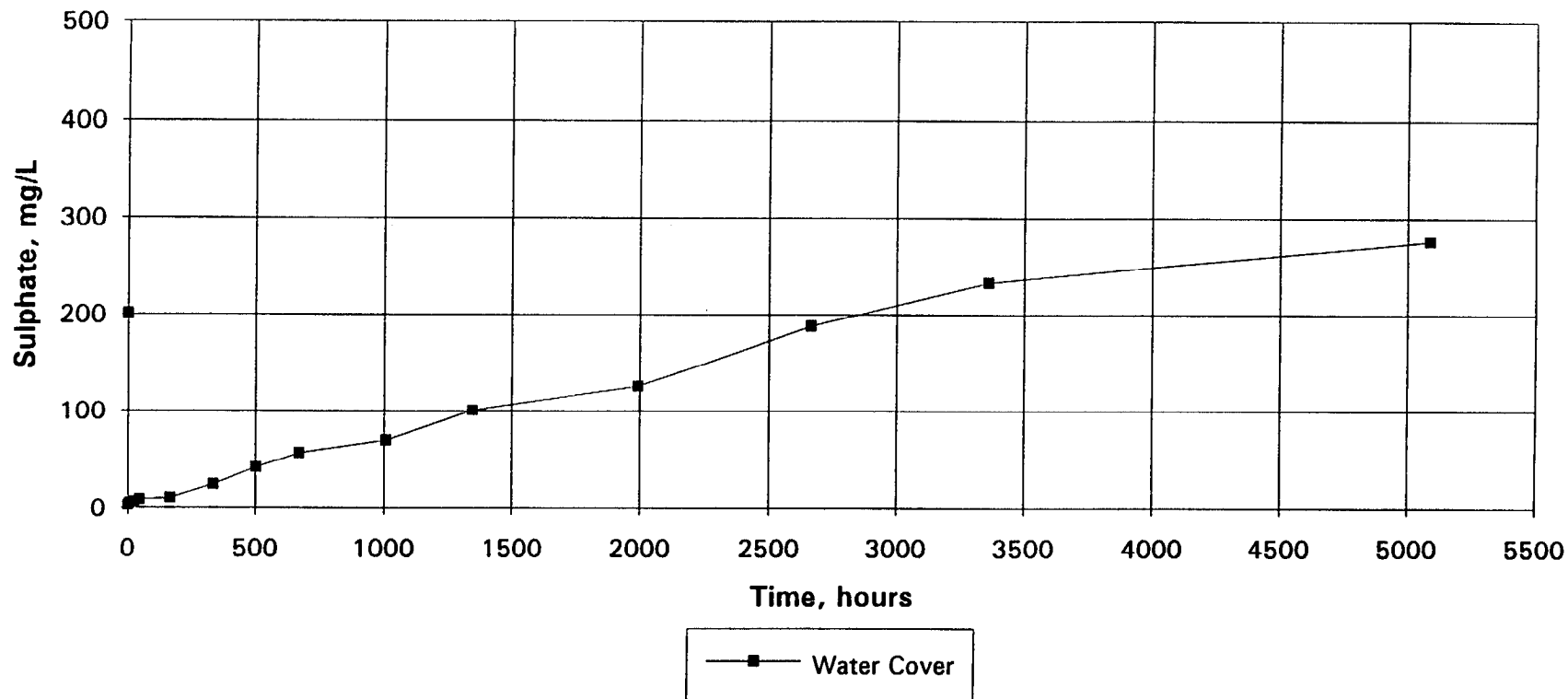


Fig. 6 Variation of alkalinity with time in the surface water cover of un-oxidized tailings underwater.

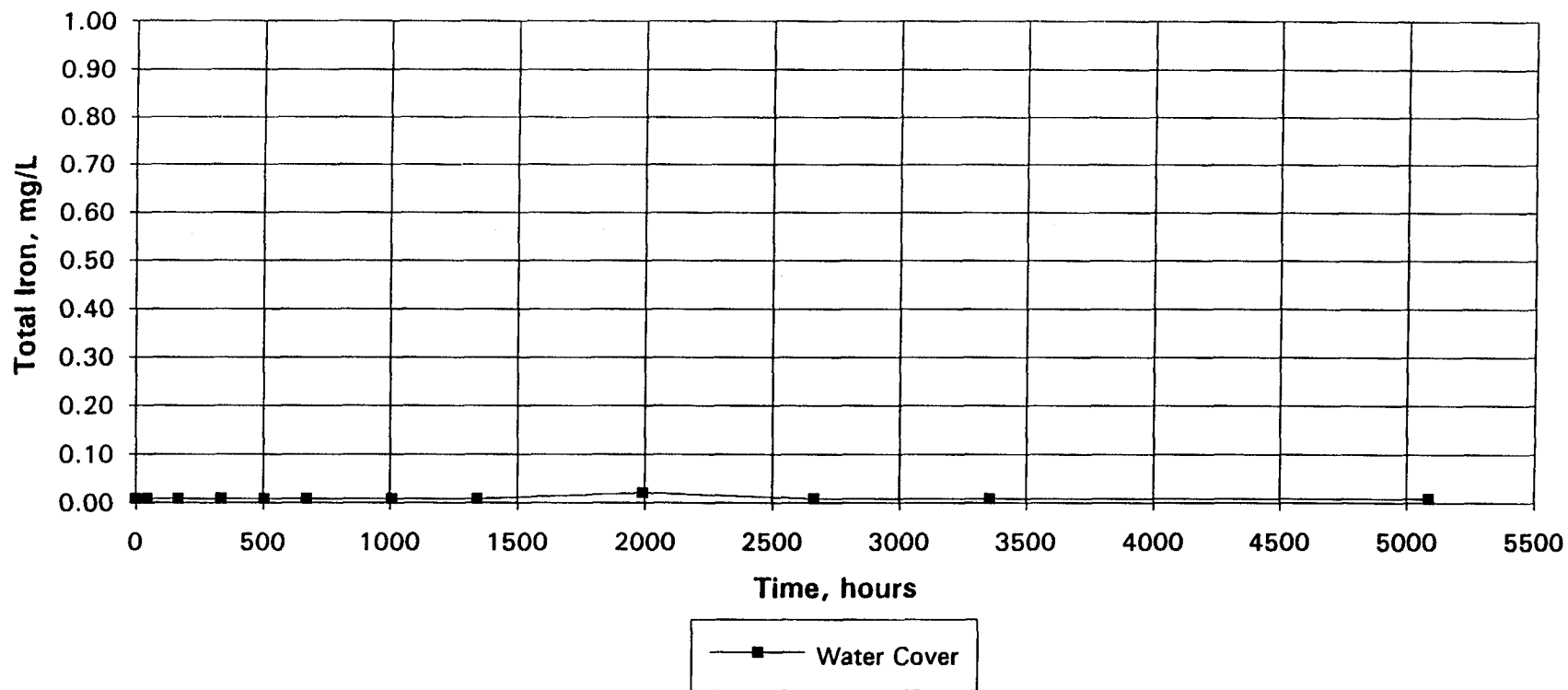


**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Surface Water Sulphate Concentration vs Time**



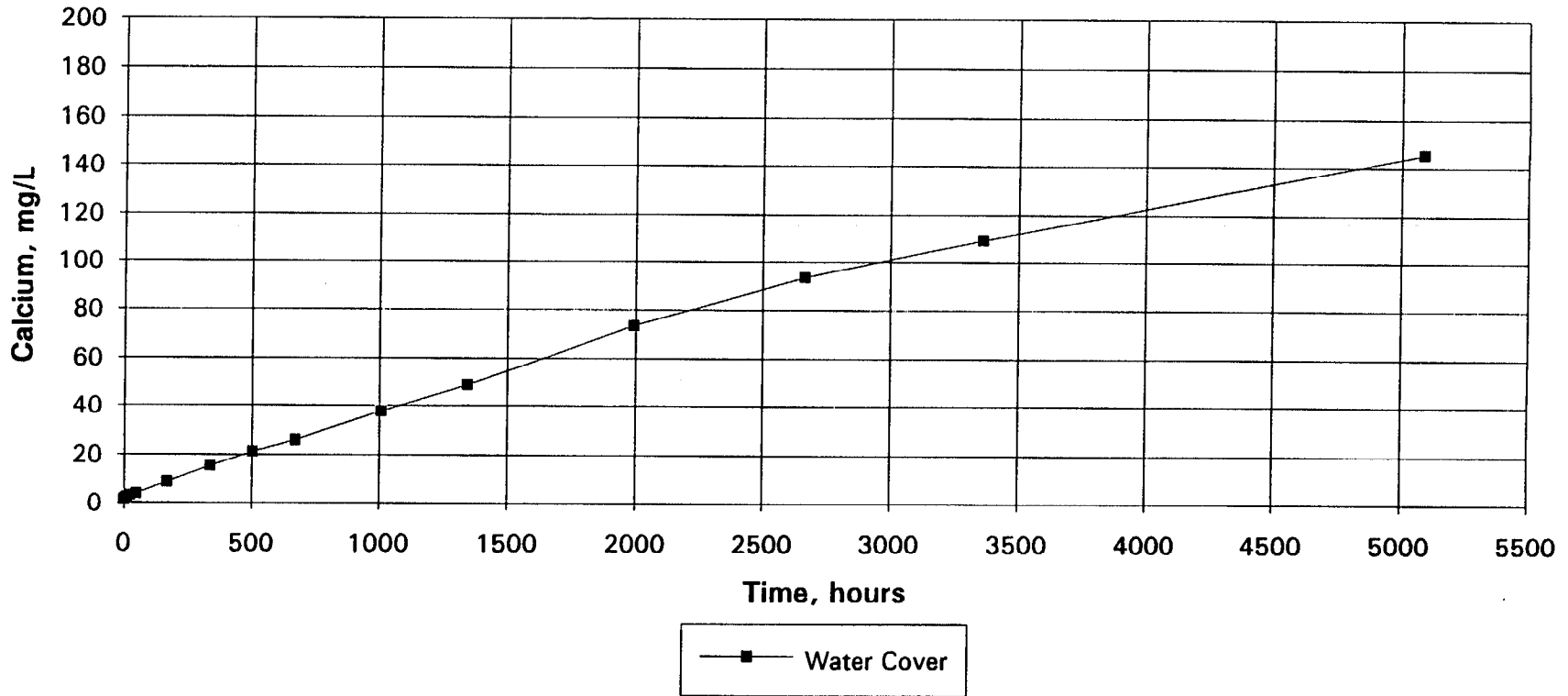
**Fig. 7** Variation of  $\text{SO}_4^{2-}$  concentration with time in the surface water cover of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Surface Water Total Iron Concentration vs Time**



**Fig. 8** Variation of total Fe concentration with time in the surface water cover of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Surface Water Calcium Concentration vs Time**



**Fig. 9** Variation of Ca concentration with time in the surface water cover of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Surface Water Magnesium Concentration vs Time**

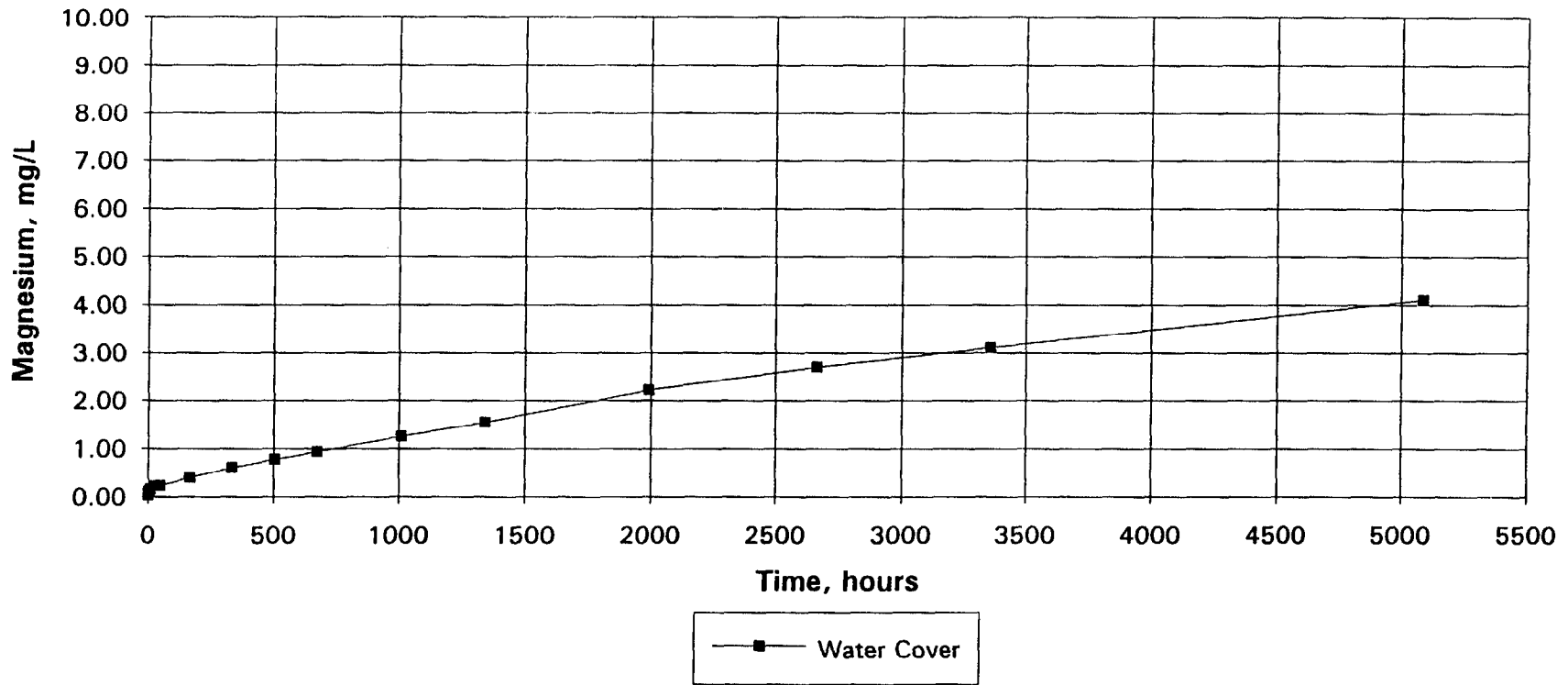


Fig. 10 Variation of Mg concentration with time in the surface water cover of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Surface Water Manganese Concentration vs Time**

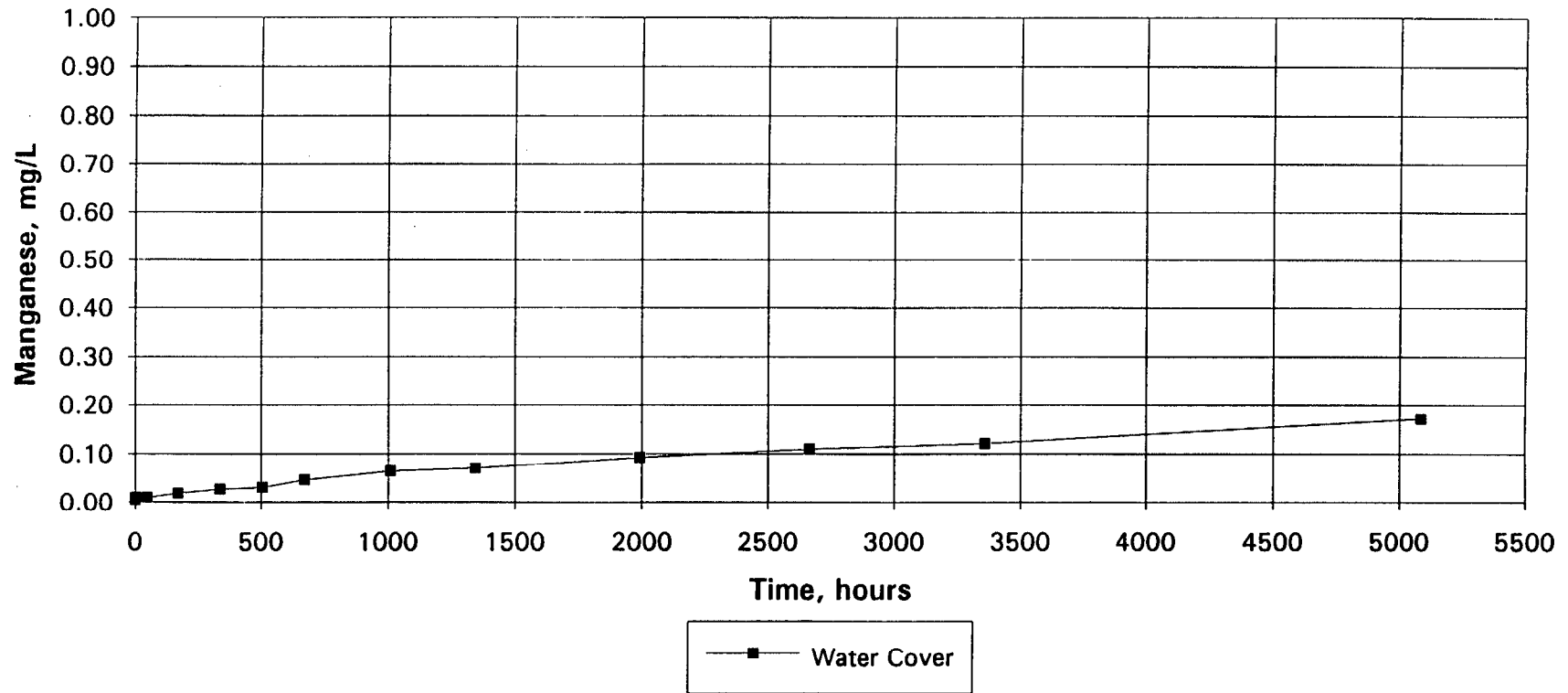


Fig. 11 Variation of Mn concentration with time in the surface water cover of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Surface Water Lead Concentration vs Time**

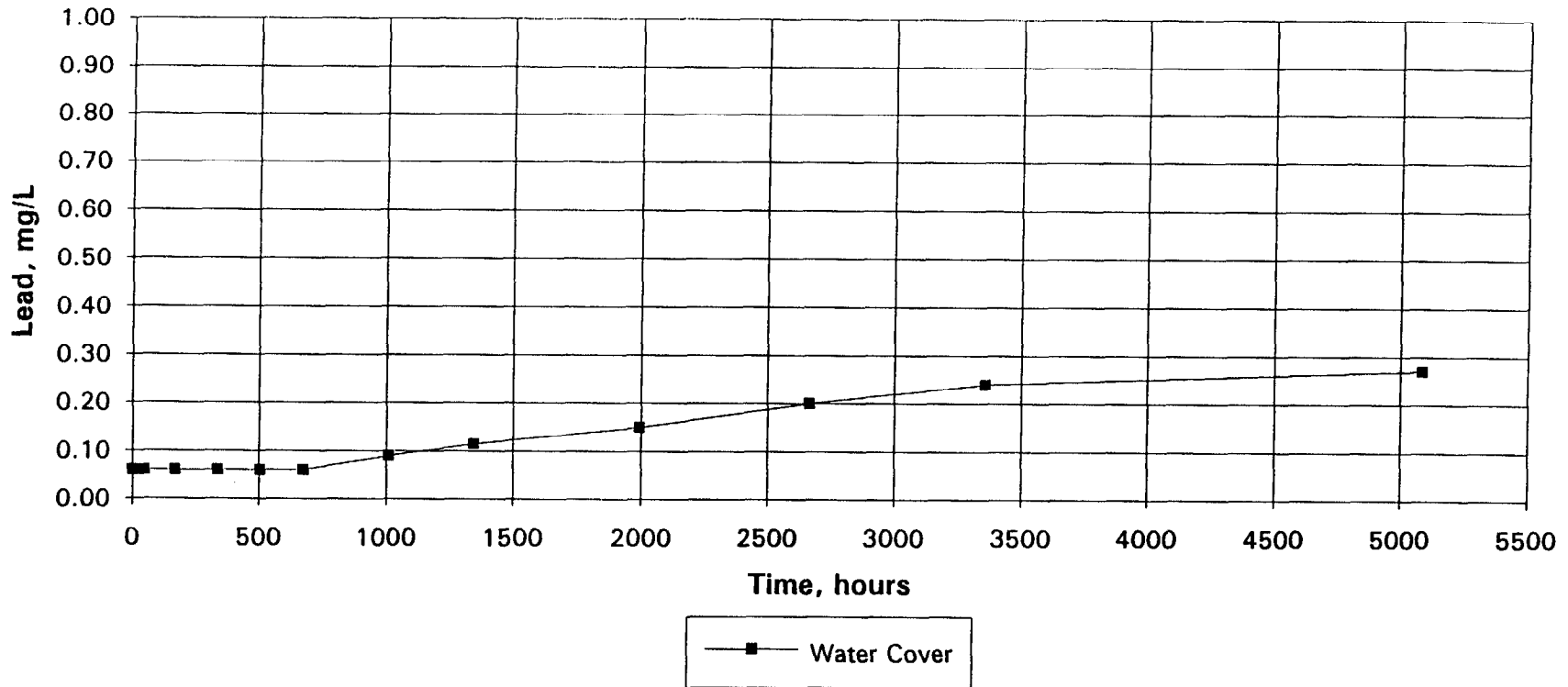
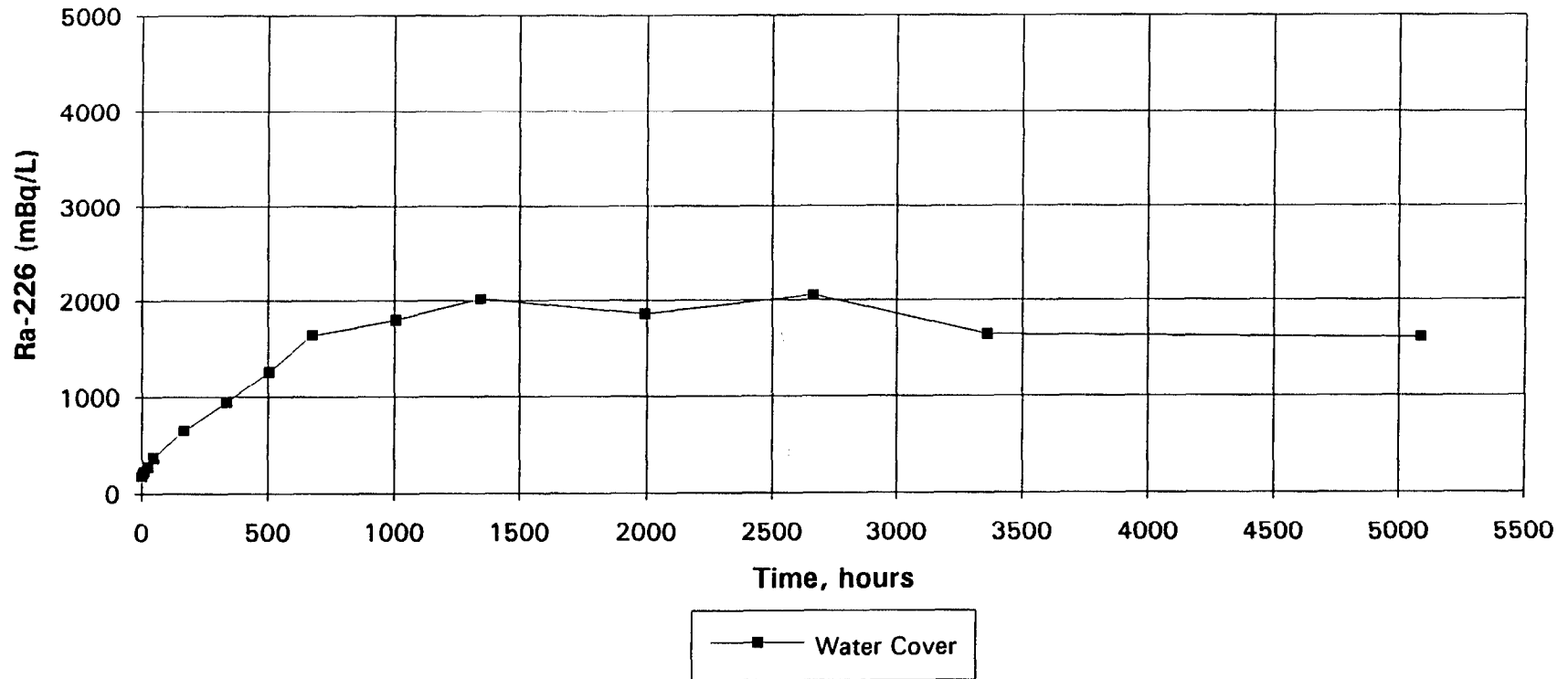


Fig. 12 Variation of Pb concentration with time in the surface water cover of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Surface Water Ra-226 Concentration vs Time**



**Fig. 13** Variation of Ra-226 concentration with time in the surface water cover of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Aquarium #1  
Porewater pH vs Time**

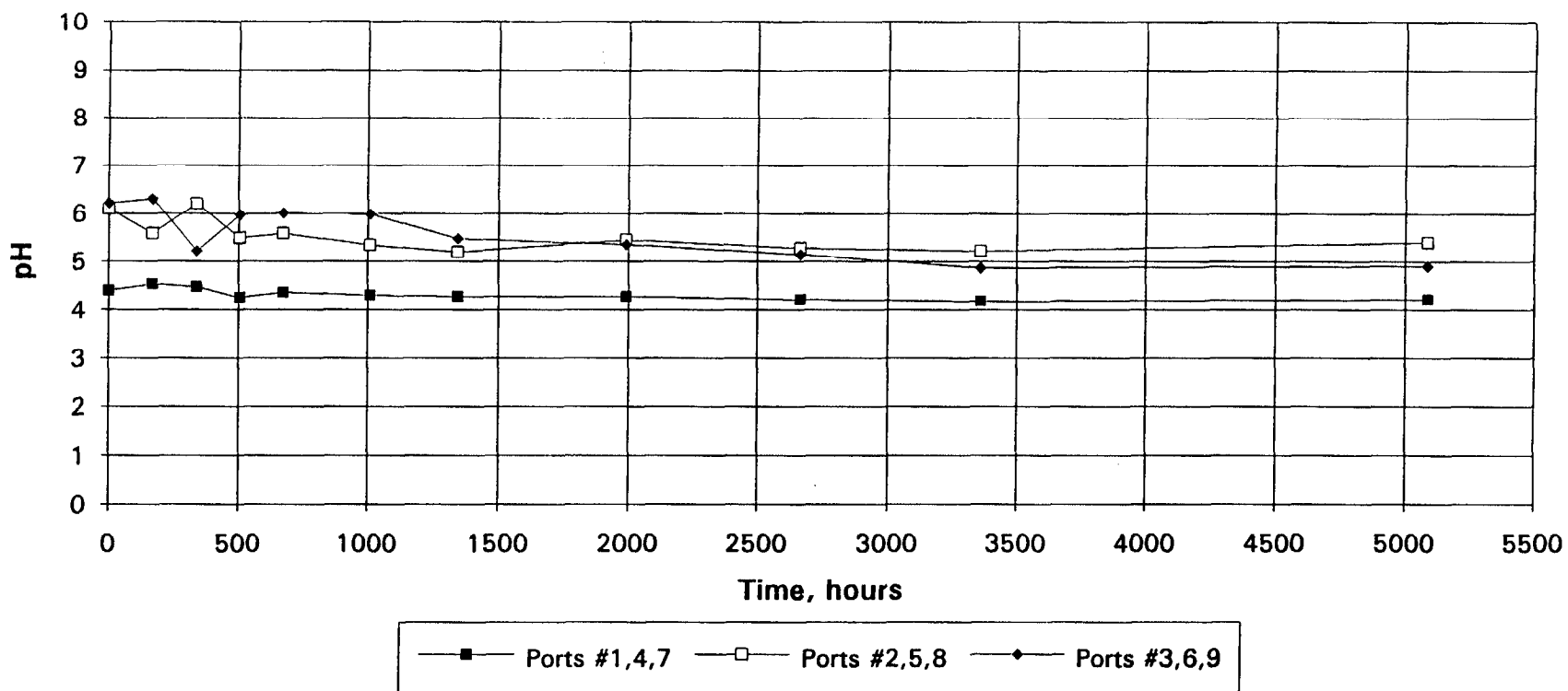


Fig. 14 Variation of pH with time in the porewater of un-oxidized tailings underwater.



**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Porewater Eh(NHE) vs Time**

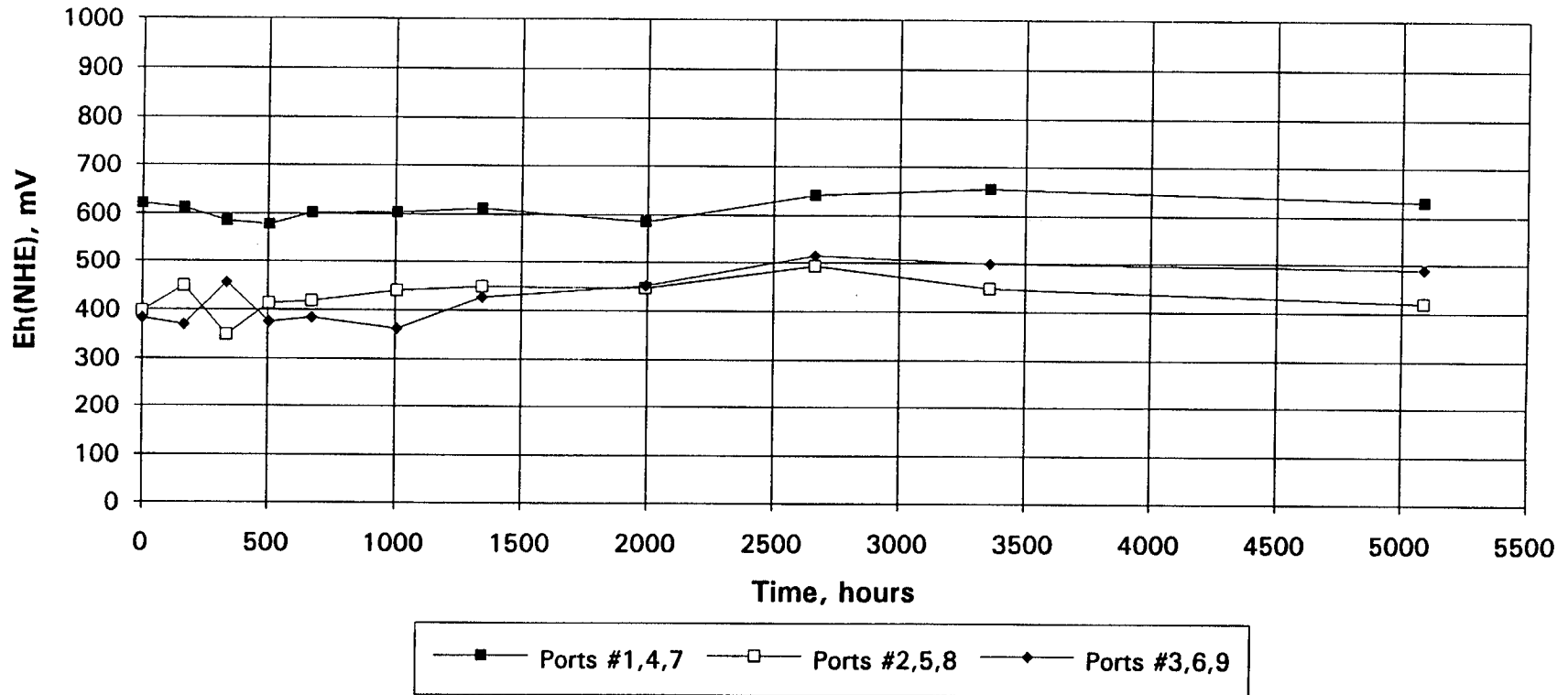


Fig. 15 Variation of Eh with time in the porewater of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Porewater Electrical Conductivity vs Time**

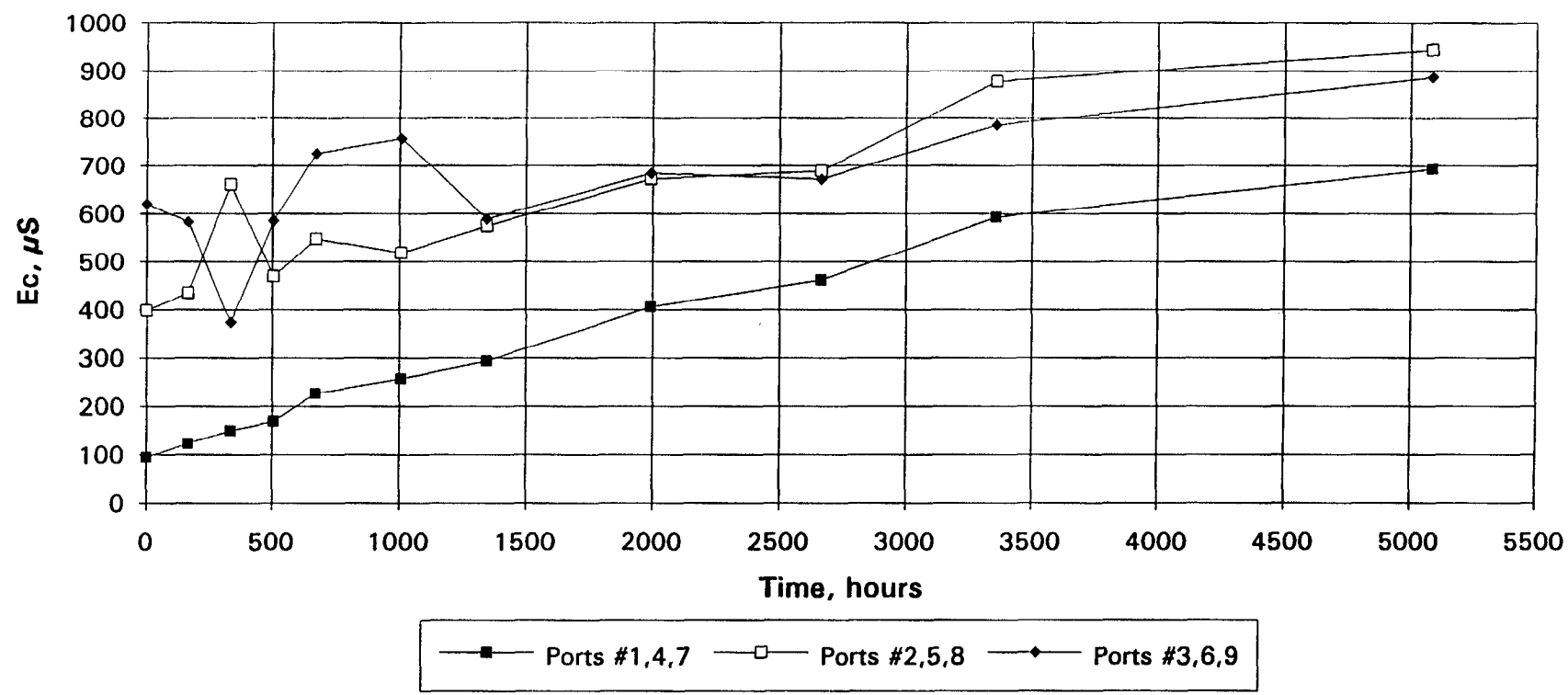


Fig. 16 Variation of Ec with time in the porewater of un-oxidized tailings underwater.

Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Porewater Acidity vs Time

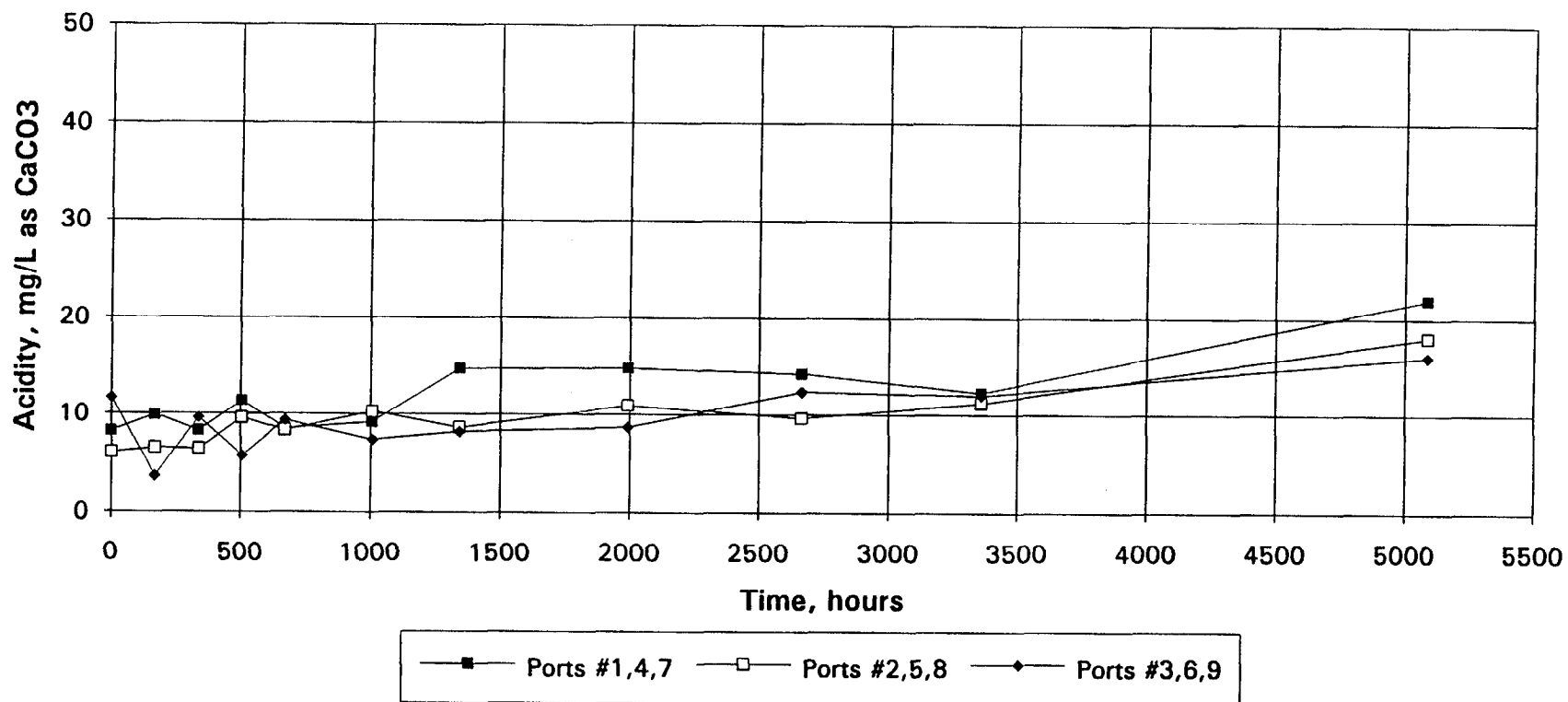


Fig. 17 Variation of acidity with time in the porewater of un-oxidized tailings underwater.

Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Porewater Alkalinity vs Time

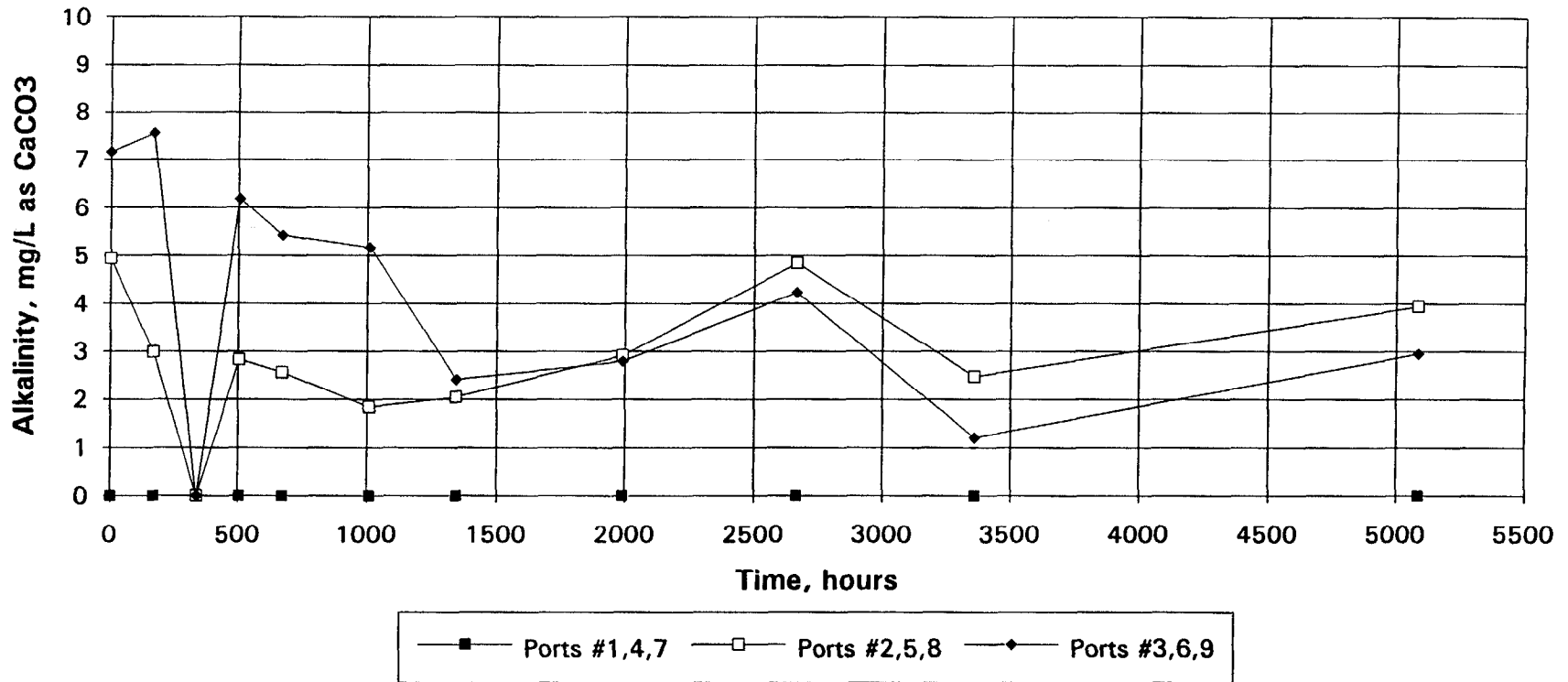


Fig. 18 Variation of alkalinity with time in the porewater of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Porewater Sulphate Concentration vs Time**

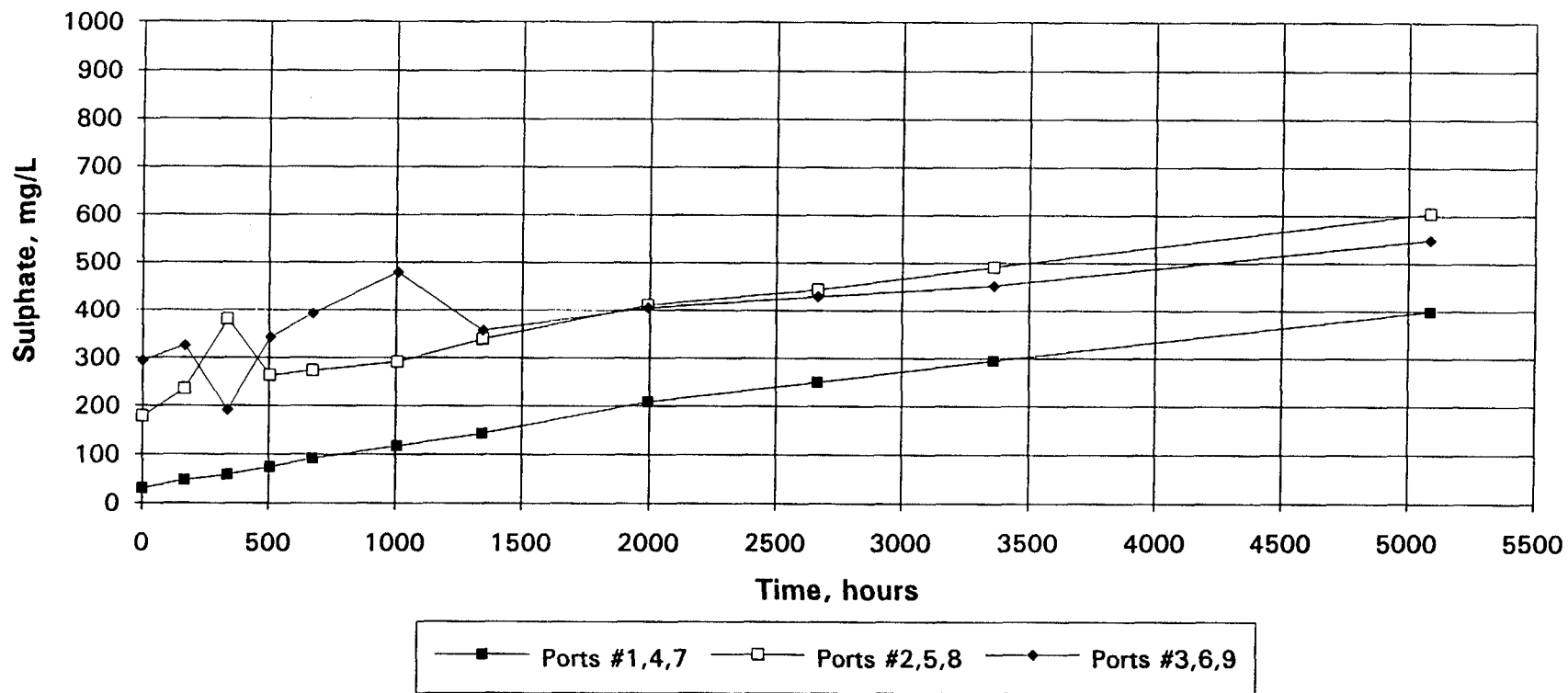


Fig. 19 Variation of  $\text{SO}_4^{2-}$  concentration with time in the porewater of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Porewater Iron Concentration vs Time**

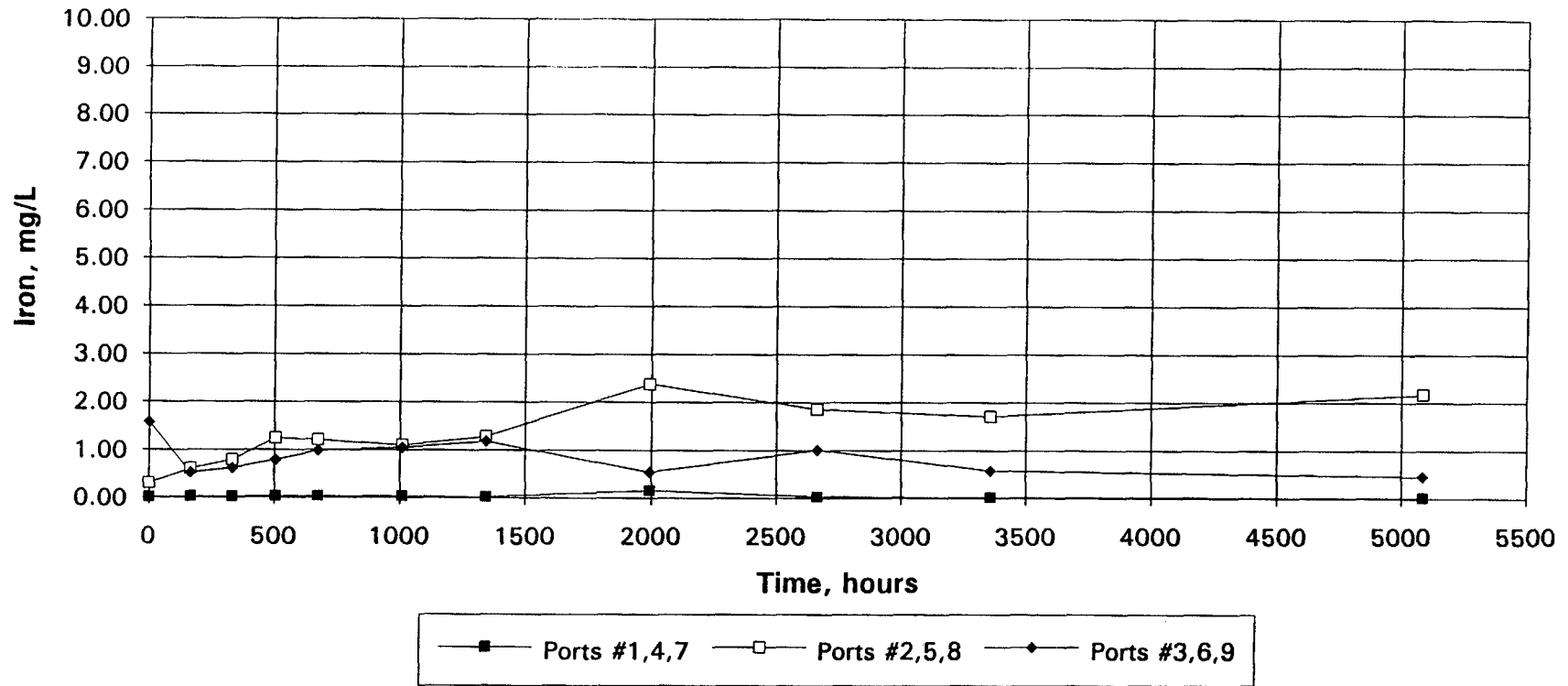


Fig. 20 Variation of total Fe concentration with time in the porewater of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Porewater Calcium Concentration vs Time**

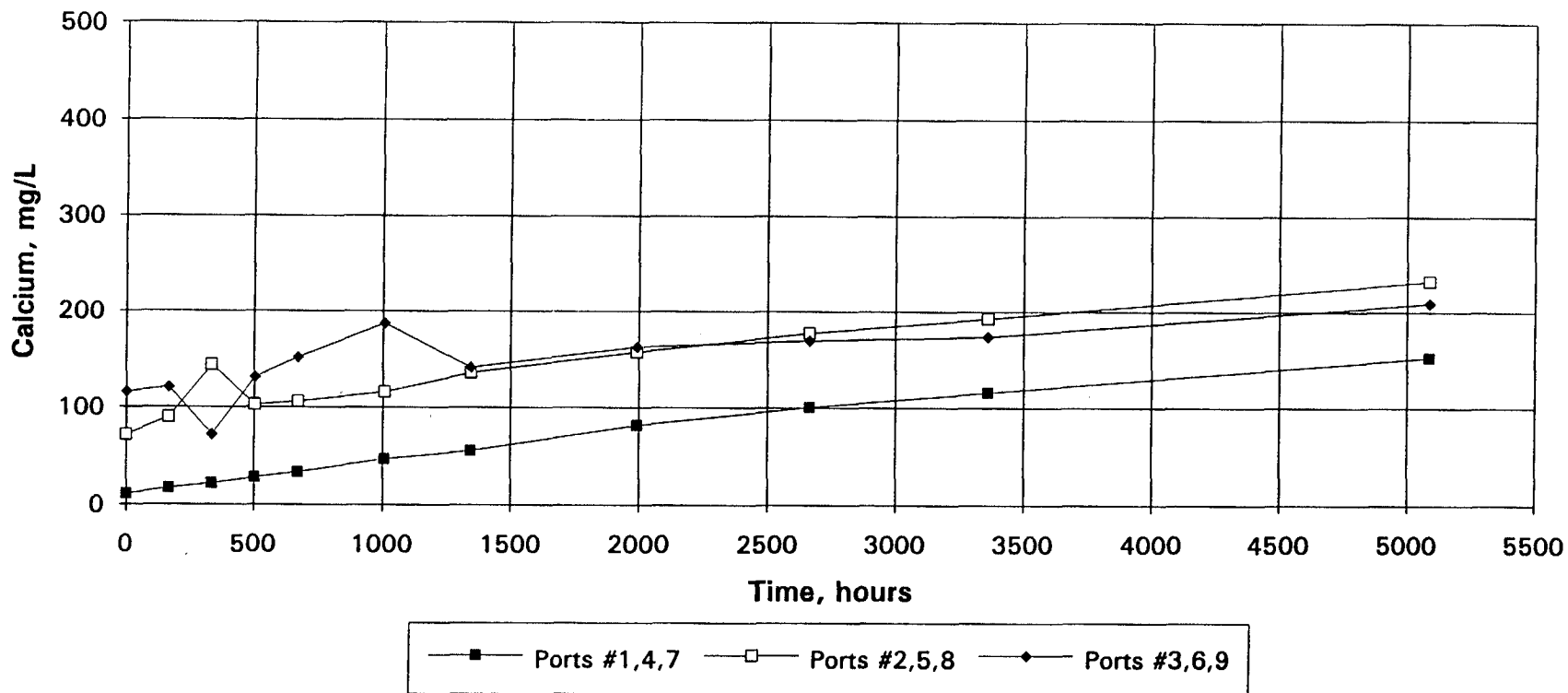


Fig. 21 Variation of Ca concentration with time in the porewater of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Porewater Magnesium Concentration vs Time**

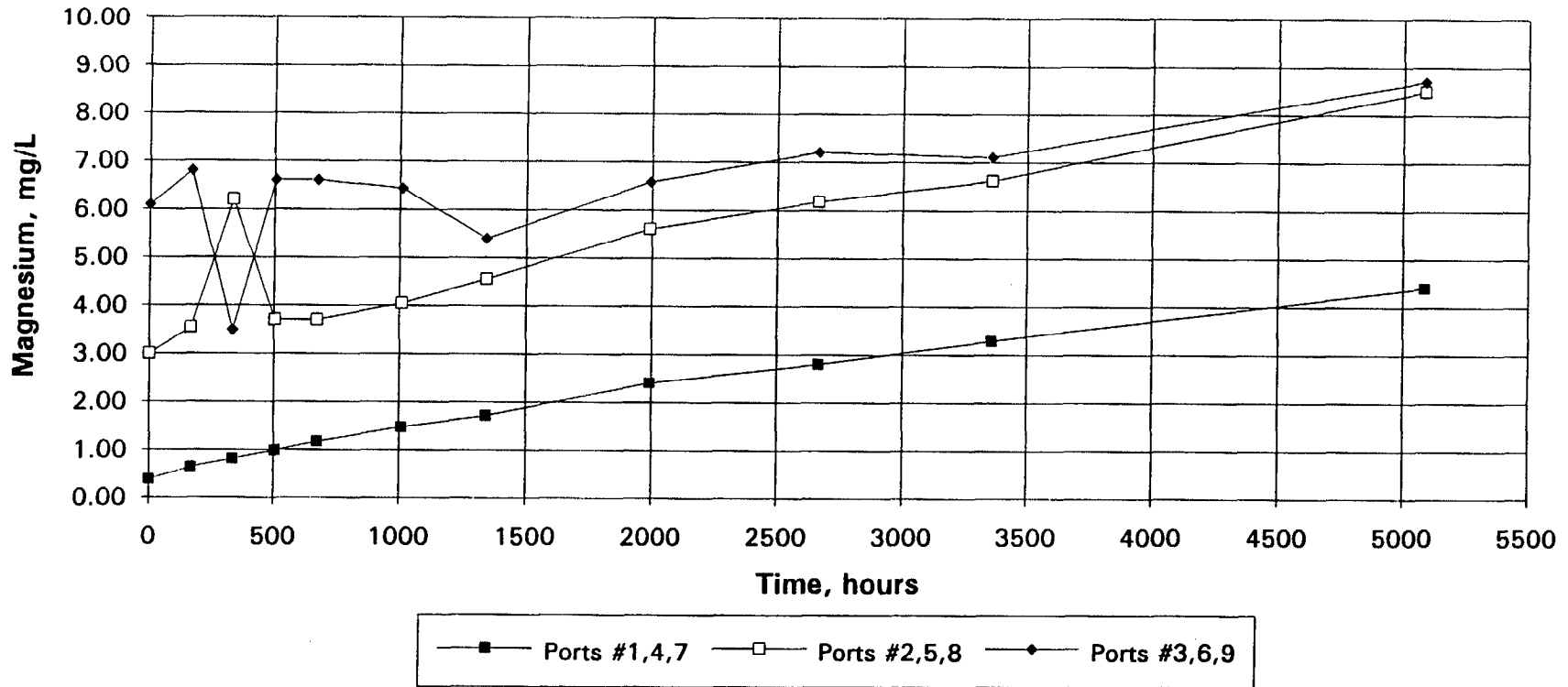


Fig. 22 Variation of Mg concentration with time in the porewater of un-oxidized tailings underwater.



**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Porewater Manganese Concentration vs Time**

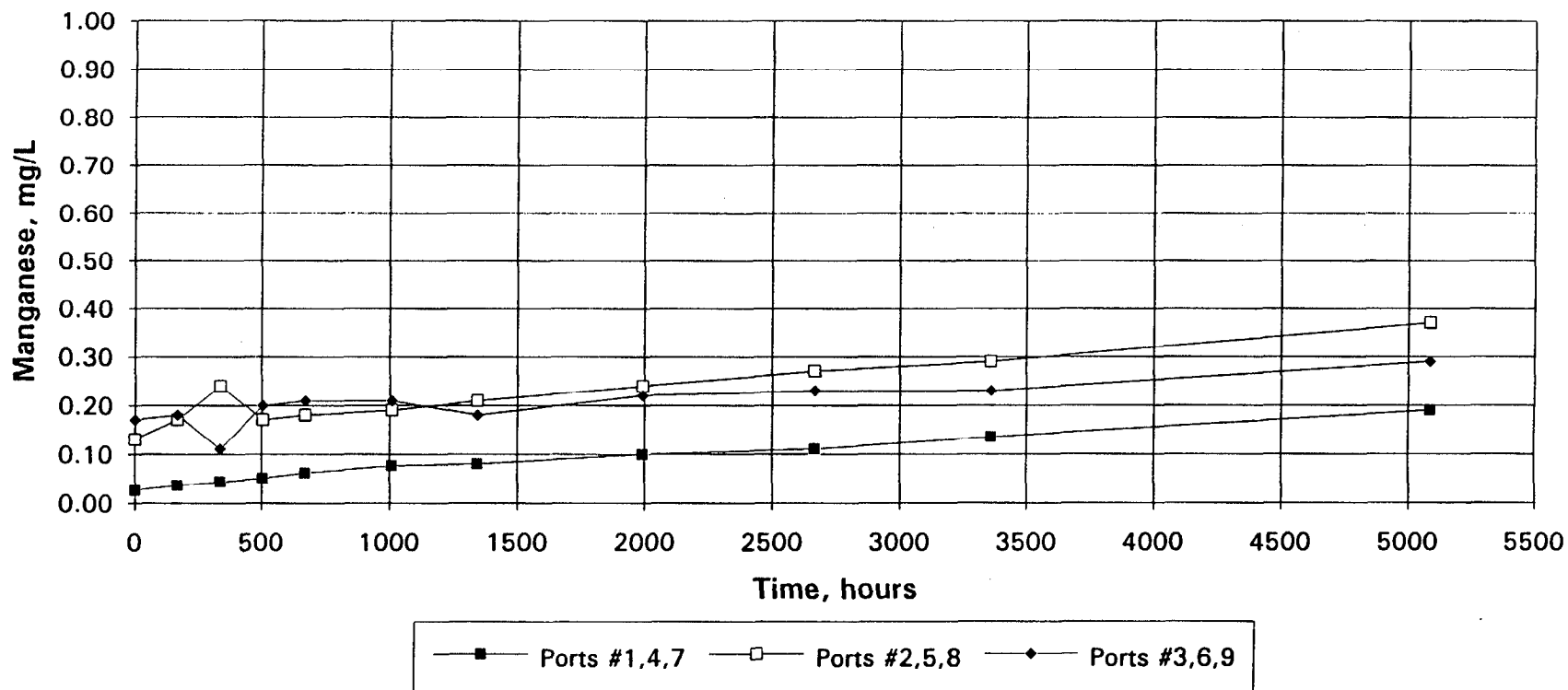


Fig. 23 Variation of Mn concentration with time in the porewater of un-oxidized tailings underwater.

Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Porewater Lead Concentration vs Time

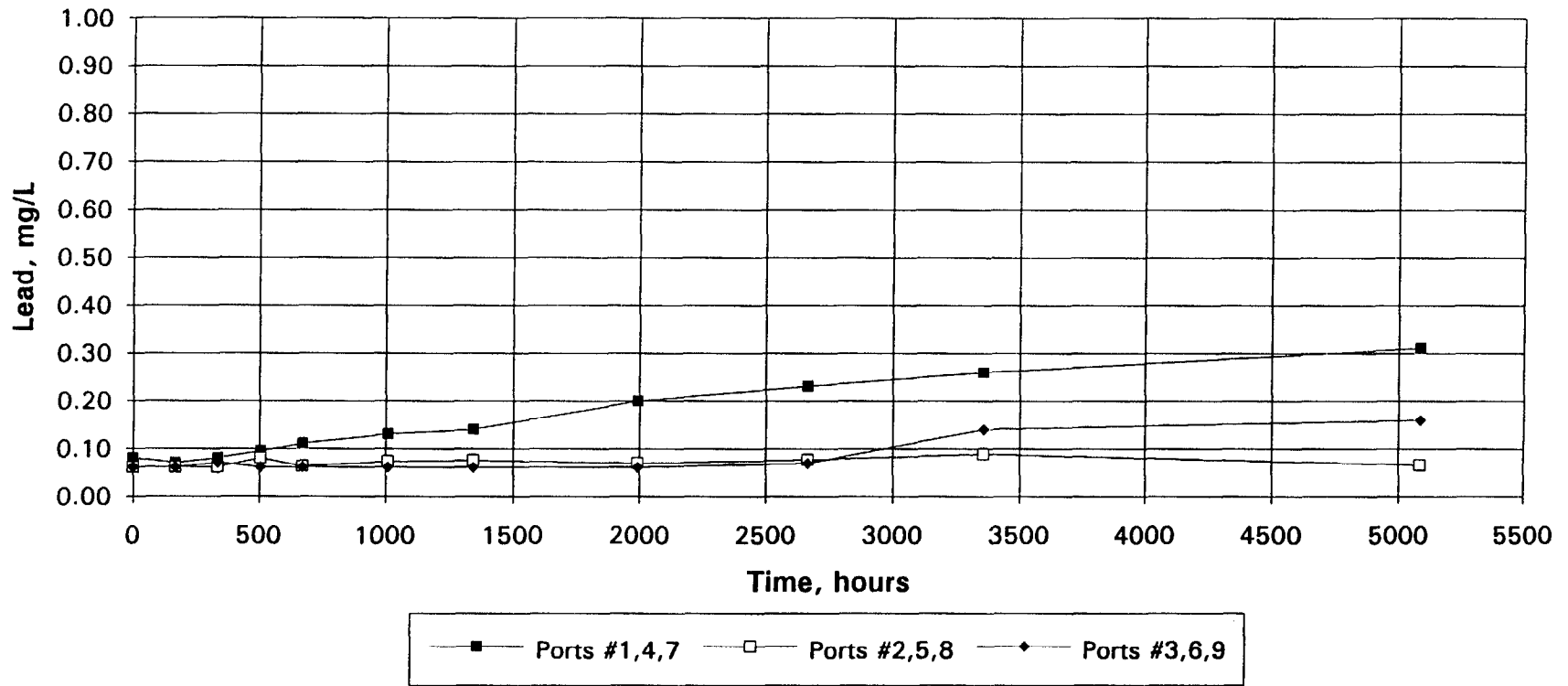


Fig. 24 Variation of Pb concentration with time in the porewater of un-oxidized tailings underwater.

**Shallow Water Cover on Un-oxidized Tailings (Aquarium #1)  
Porewater Ra-226 Concentration vs Time**

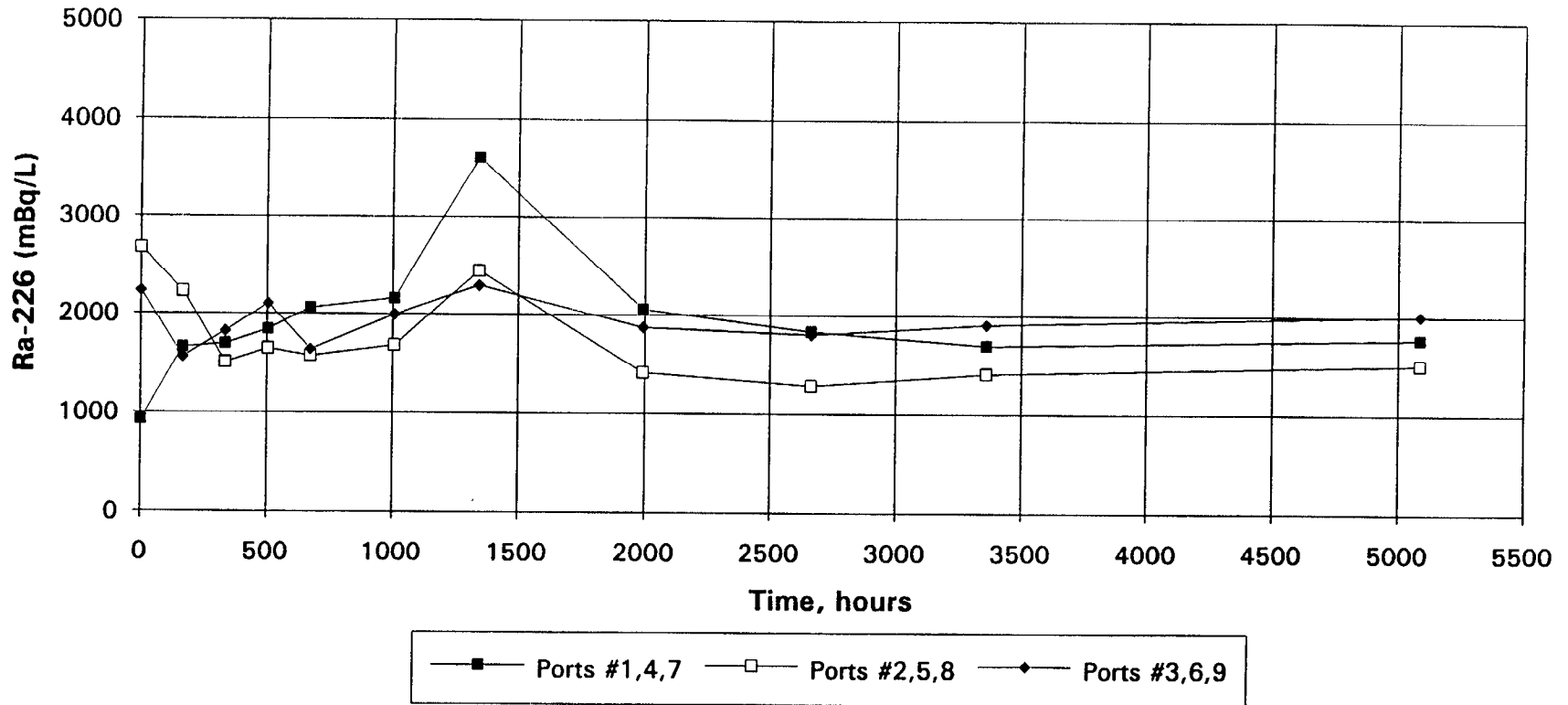
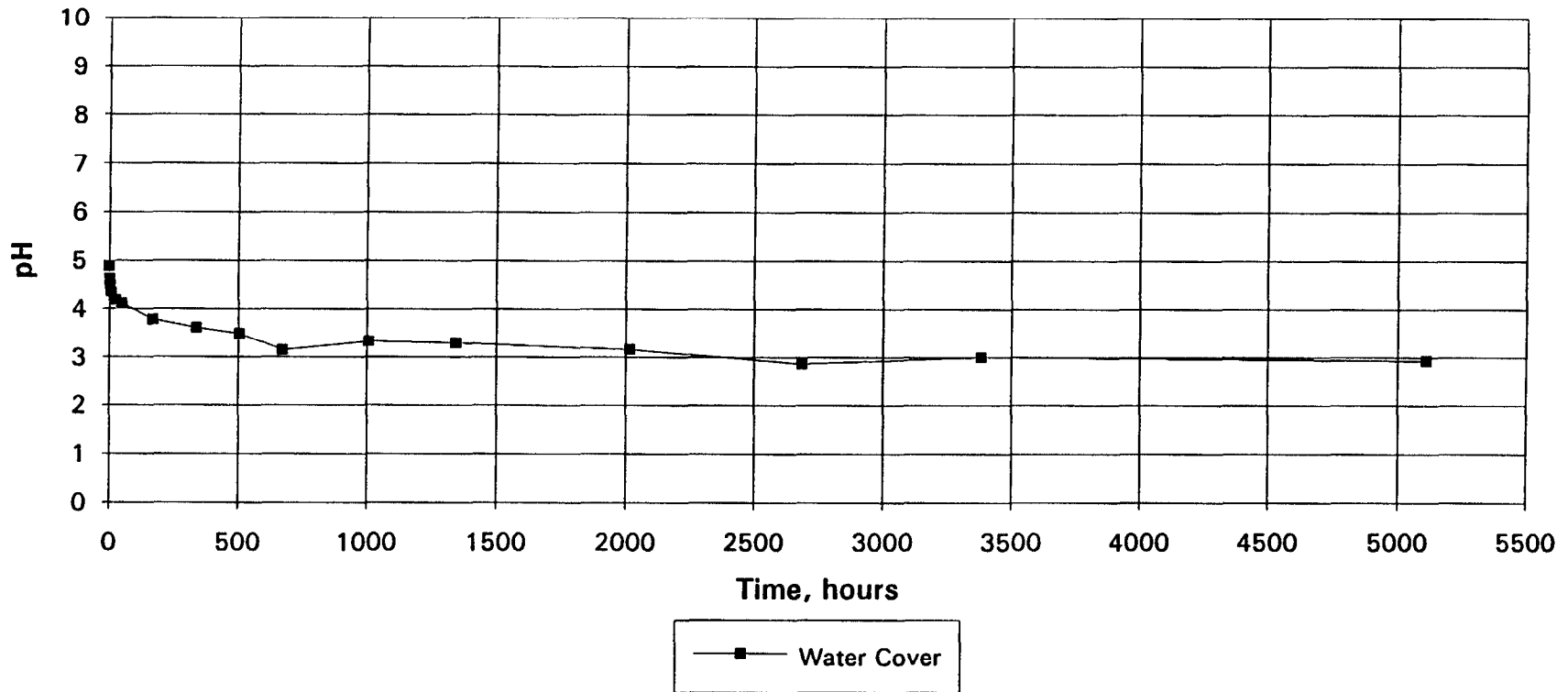


Fig. 25 Variation of Ra-226 concentration with time in the porewater of un-oxidized tailings underwater.

**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Surface Water pH vs Time**



**Fig. 26** Variation of pH with time in the surface water cover of partially oxidized and weathered tailings underwater.

Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Surface Water Eh(NHE) vs Time

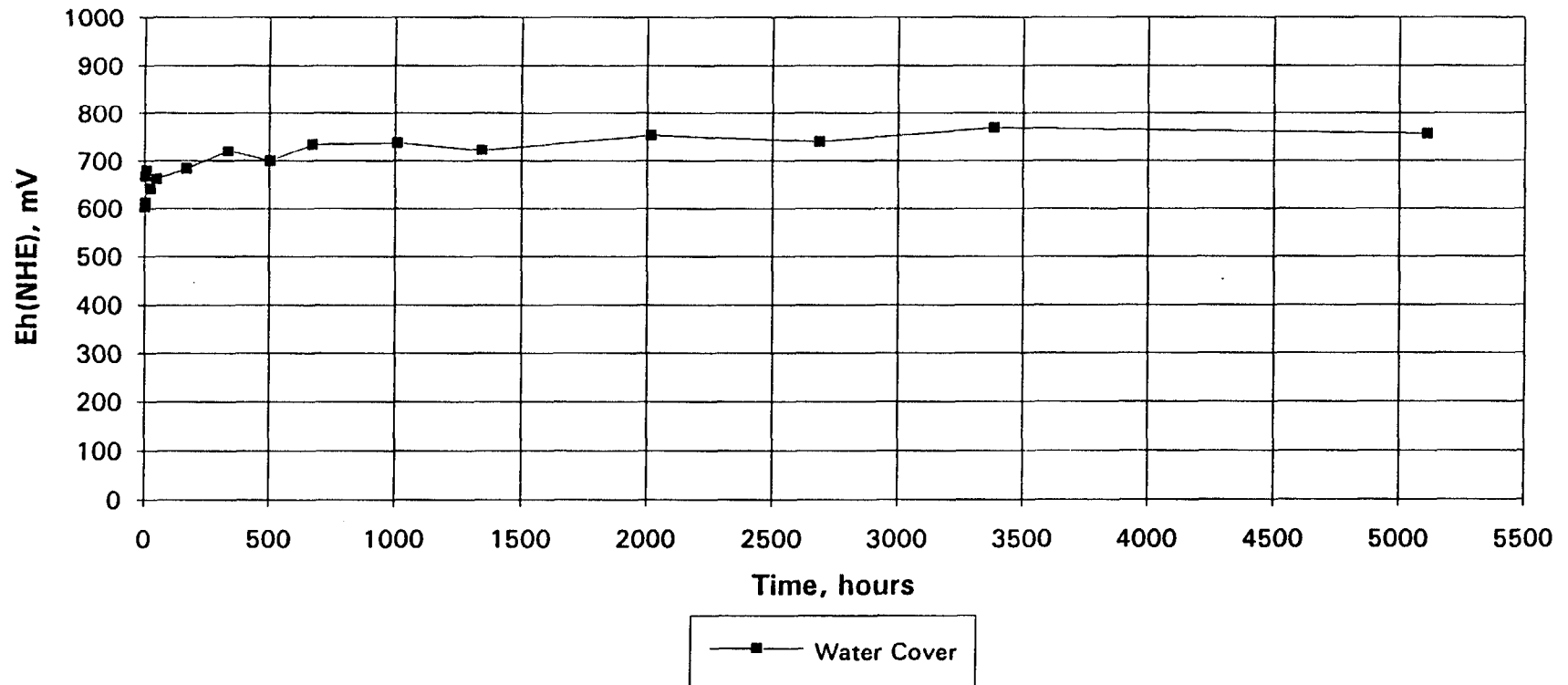


Fig. 27 Variation of Eh with time in the surface water cover of partially oxidized and weathered tailings underwater.

Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Surface Water Electrical Conductivity vs Time

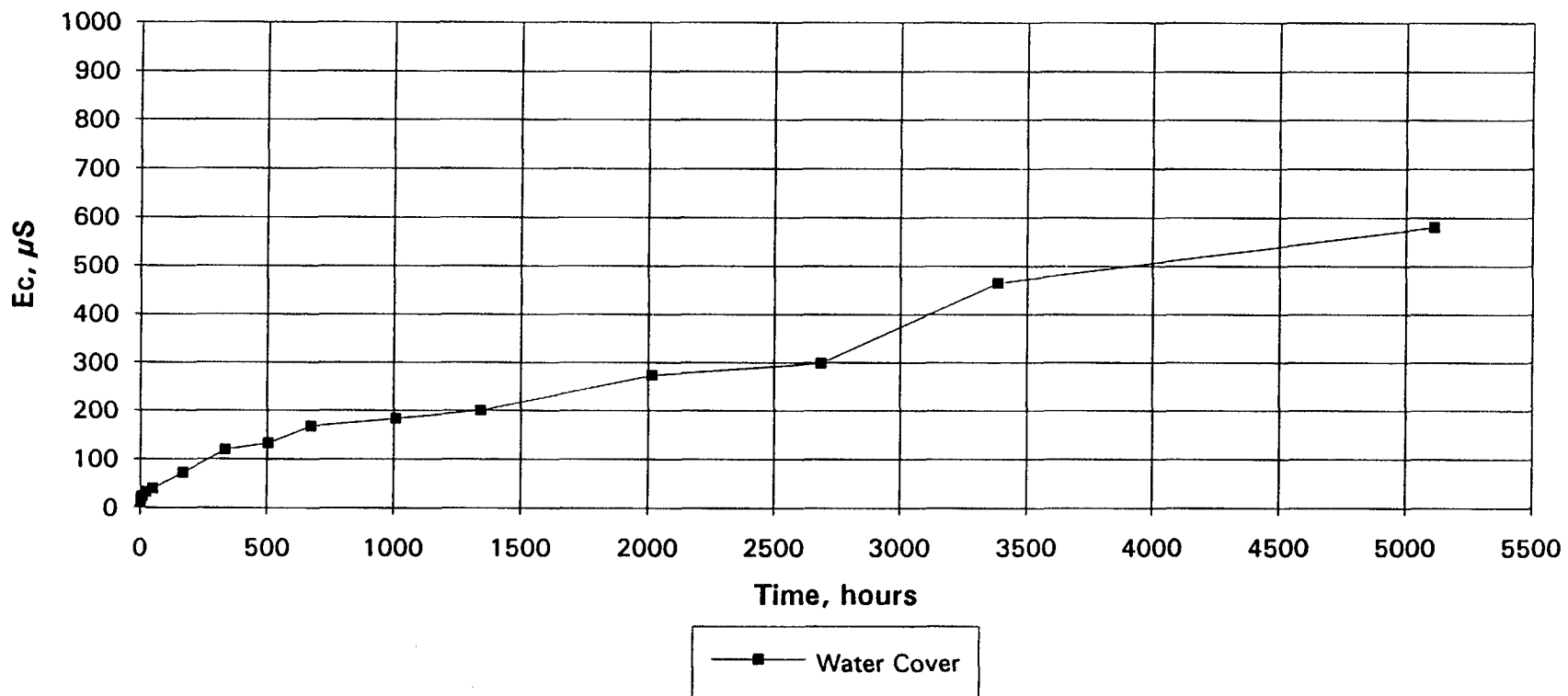
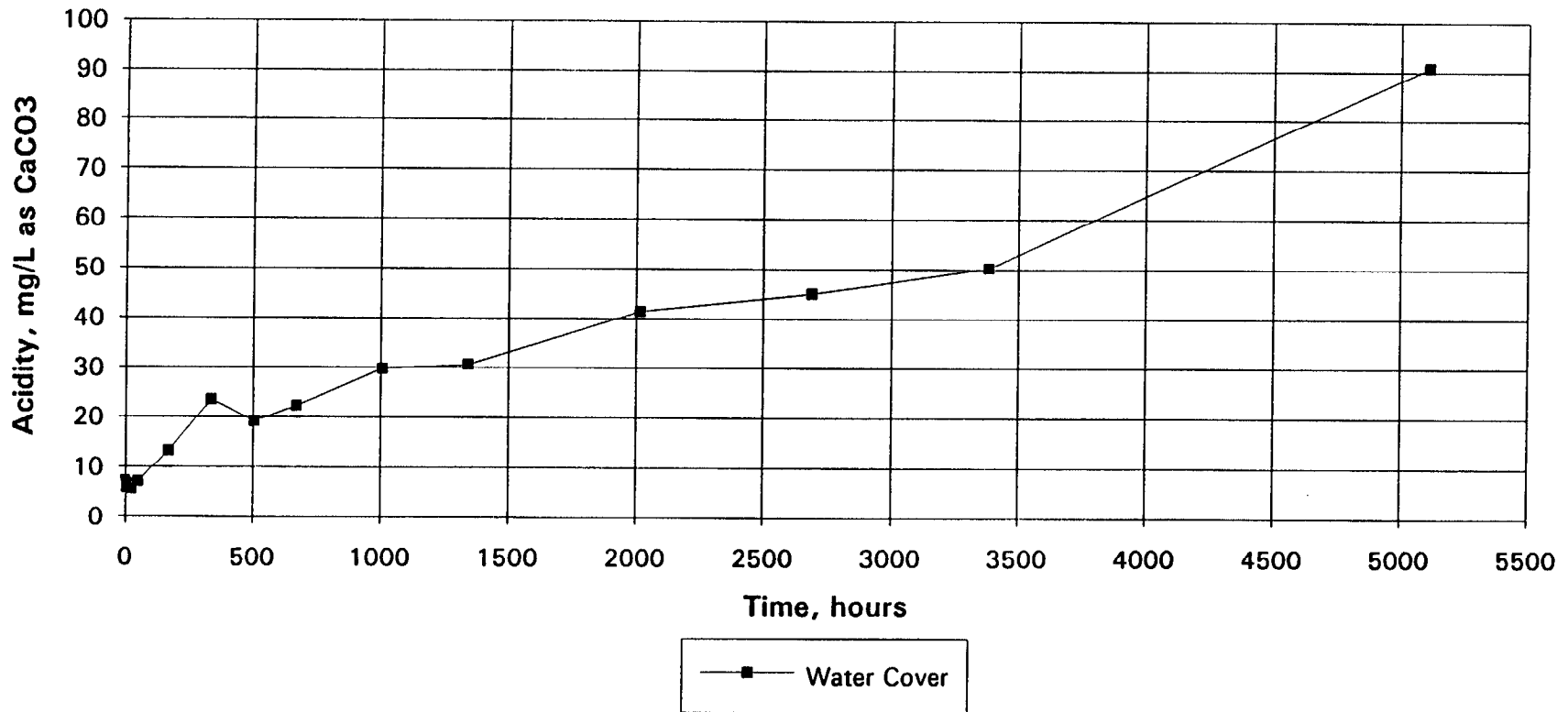


Fig. 28 Variation of Ec with time in the surface water cover of partially oxidized and weathered tailings underwater.

**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Surface Water Acidity vs Time**



**Fig. 29** Variation of acidity with time in the surface water cover of partially oxidized and weathered tailings underwater.

**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)**  
**Surface Water Sulphate Concentration vs Time**

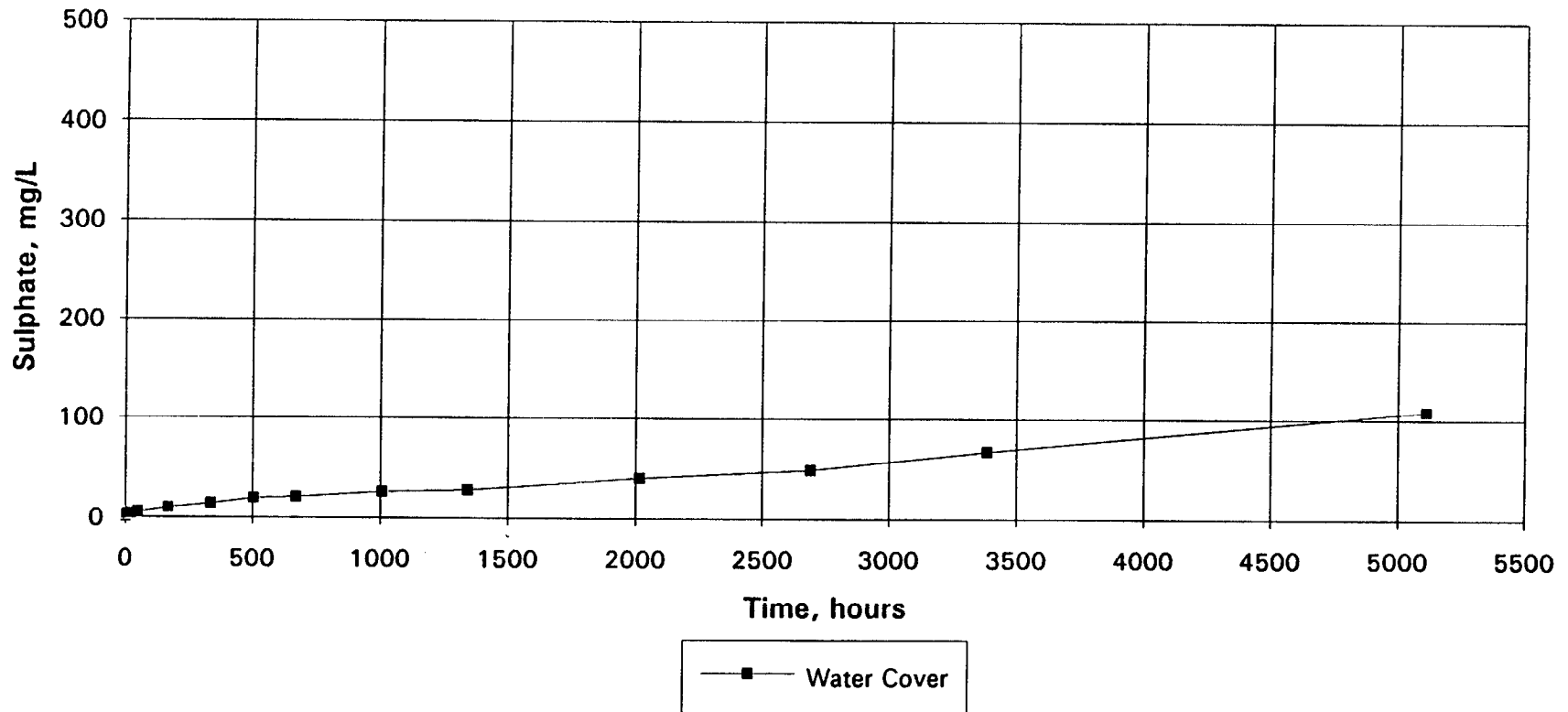
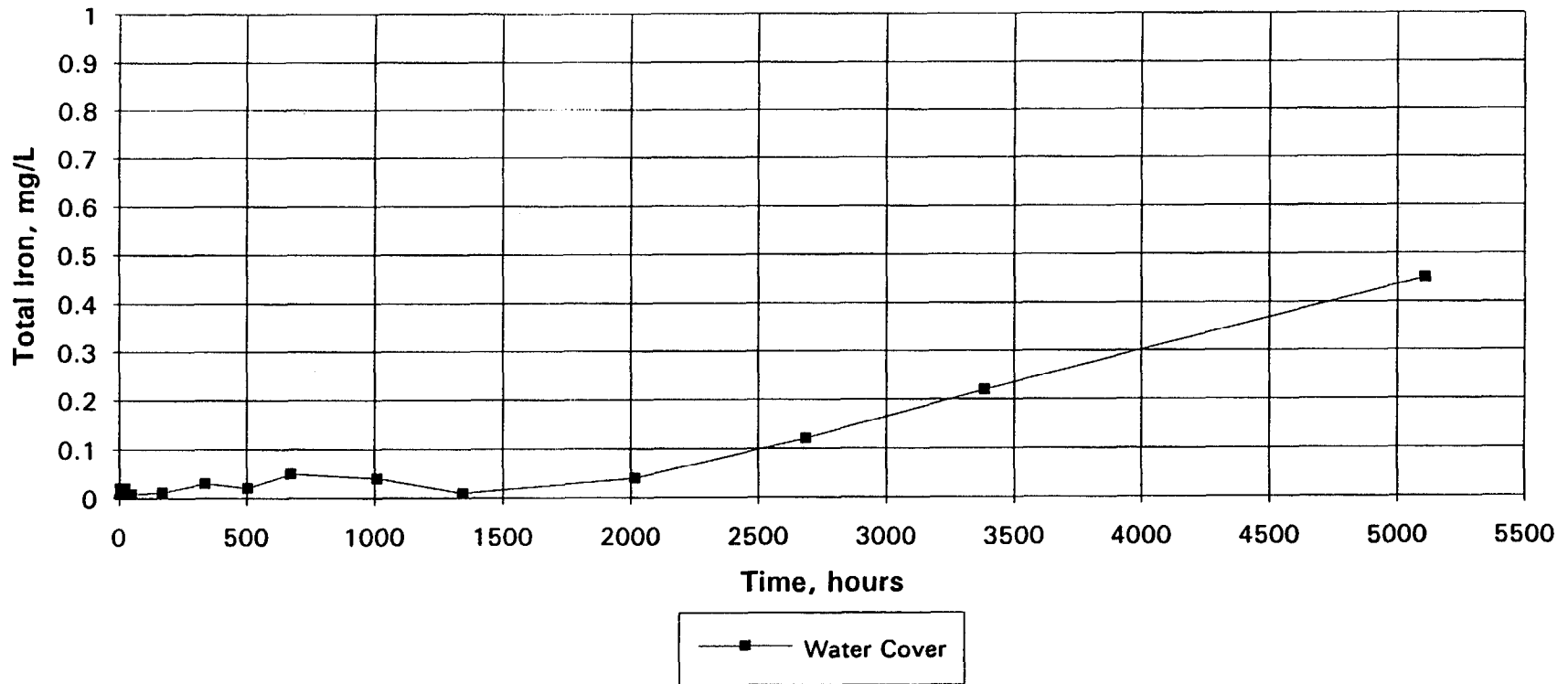


Fig. 30 Variation of  $\text{SO}_4^{2-}$  concentration with time in the surface water cover of partially oxidized and weathered tailings underwater.

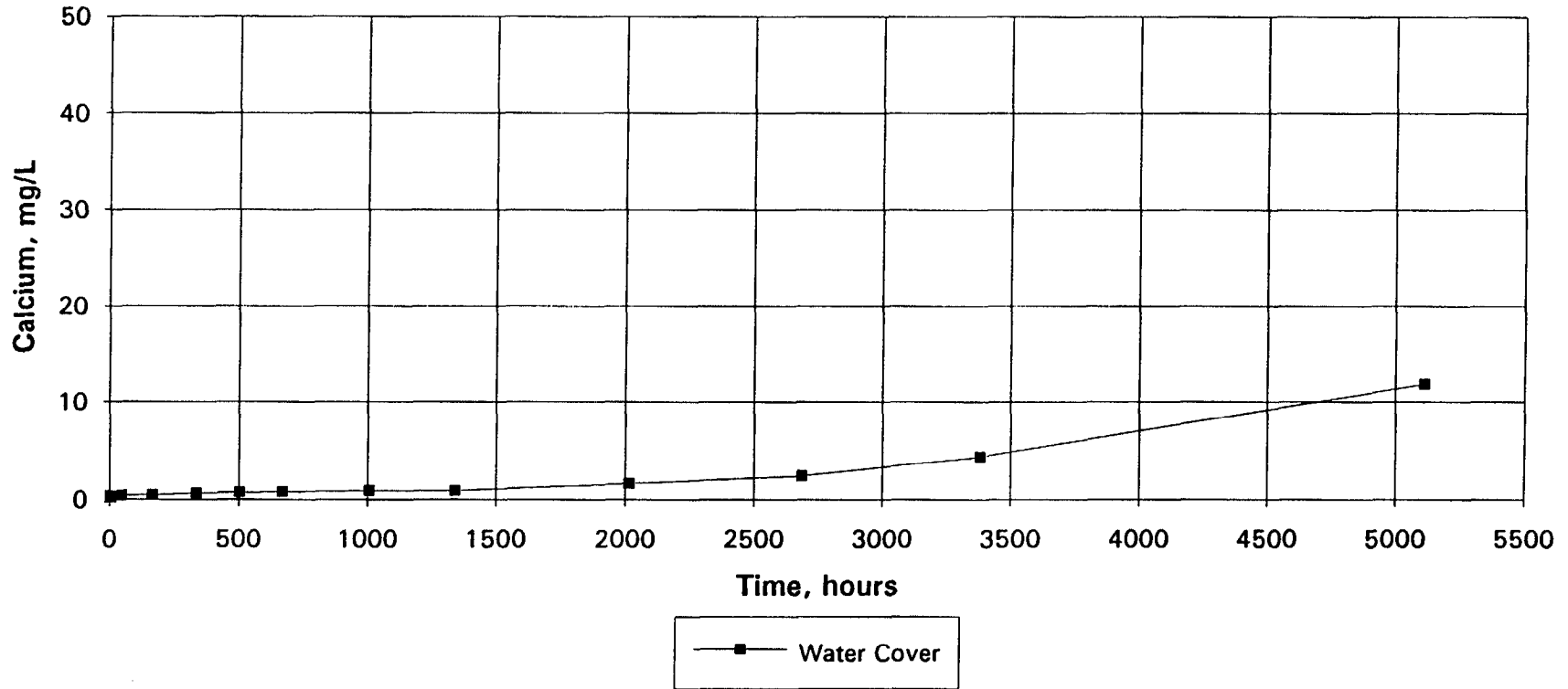


**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Surface Water Total Iron Concentration vs Time**



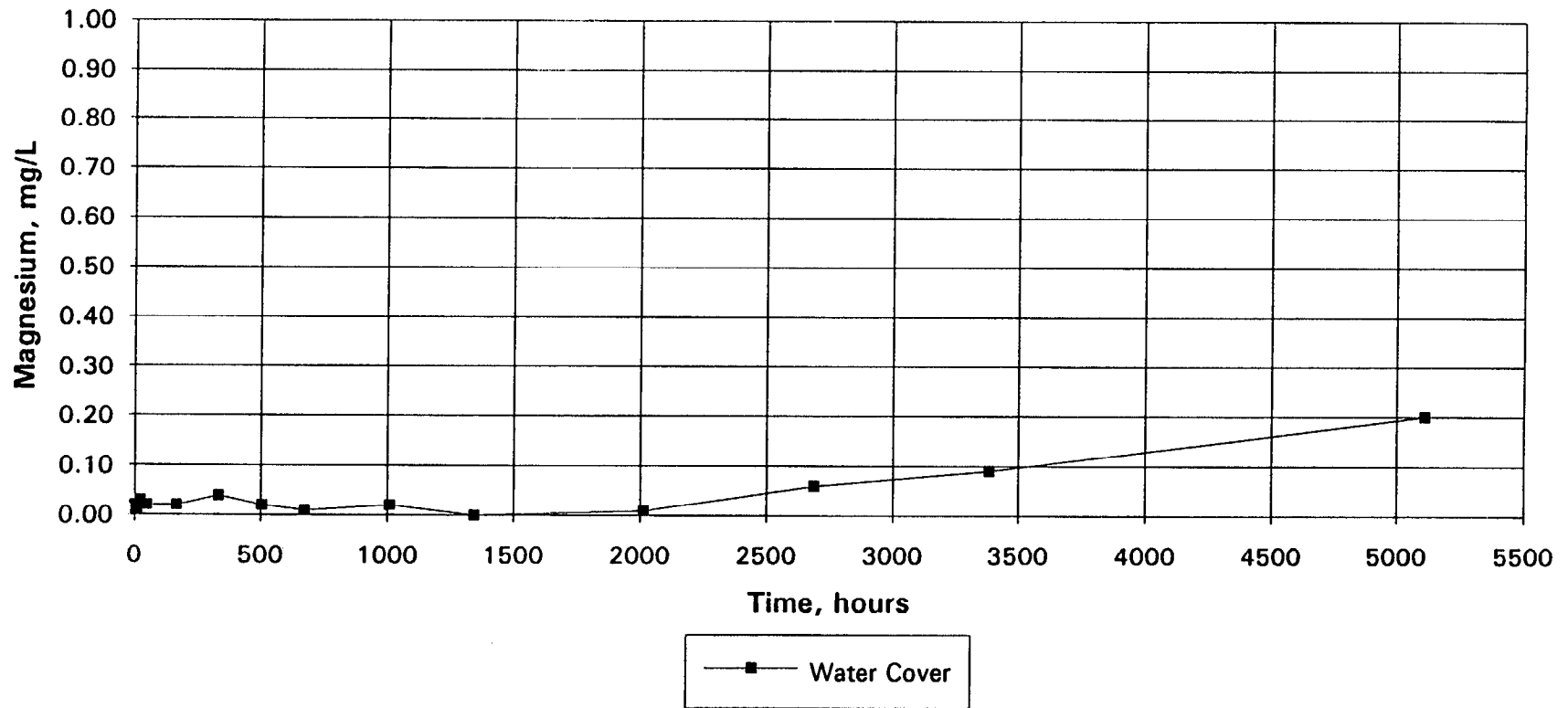
**Fig. 31** Variation of total Fe concentration with time in the surface water cover of partially oxidized and weathered tailings underwater.

**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Surface Water Calcium Concentration vs Time**



**Fig. 32** Variation of Ca concentration with time in the surface water cover of partially oxidized and weathered tailings underwater.

**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Surface Water Magnesium Concentration vs Time**



**Fig. 33** Variation of Mg concentration with time in the surface water cover of partially oxidized and weathered tailings underwater.

**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Surface Water Manganese Concentration vs Time**

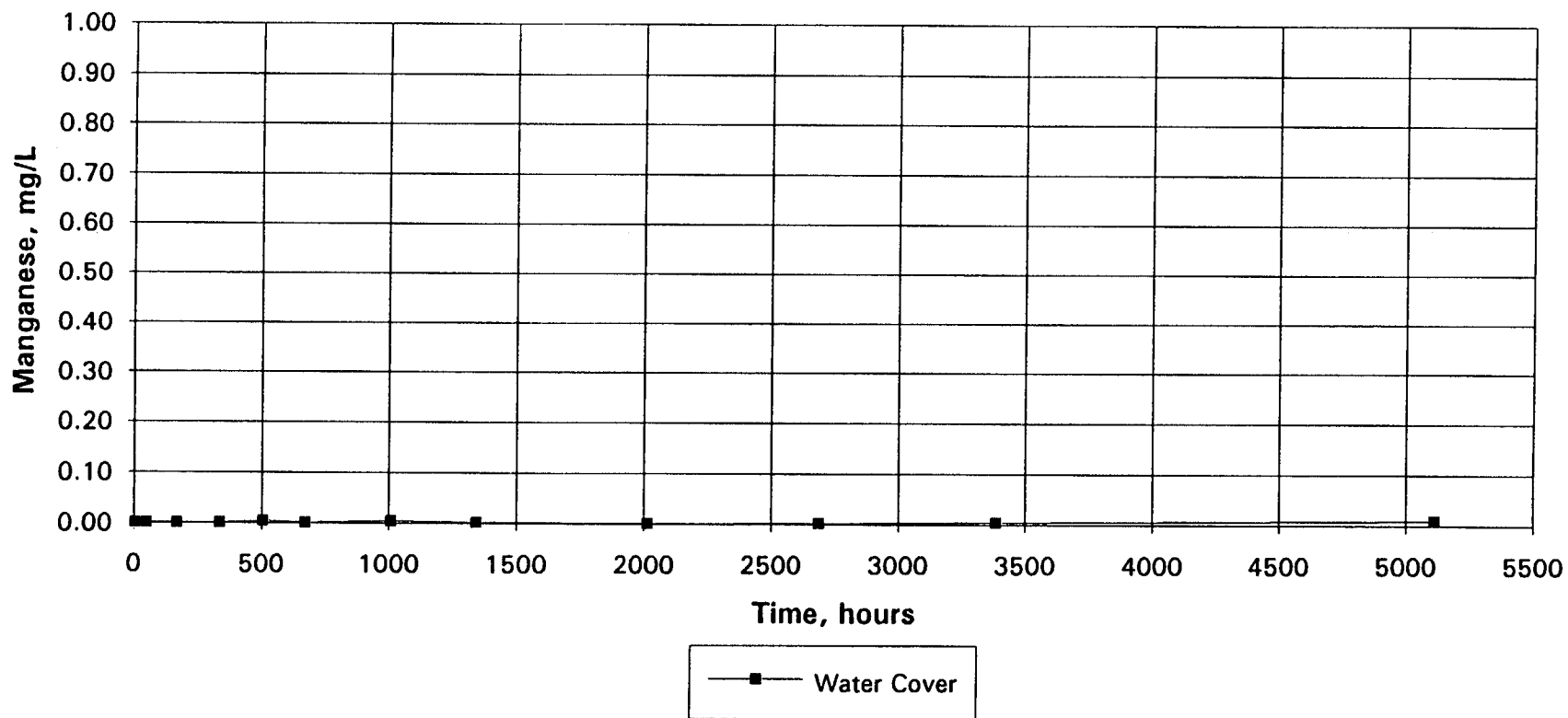


Fig. 34 Variation of Mn concentration with time in the surface water cover of partially oxidized and weathered tailings underwater.

Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Surface Water Lead Concentration vs Time

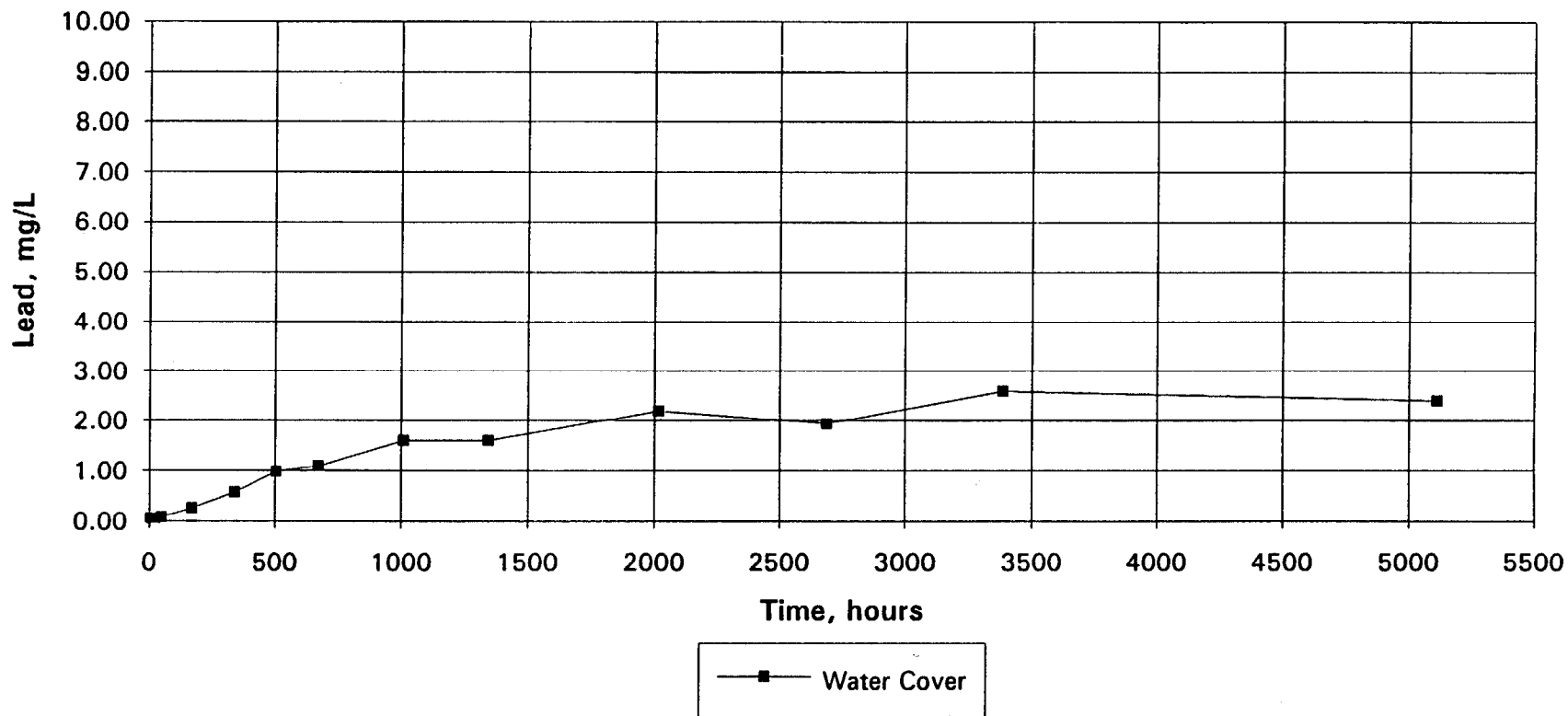


Fig. 35 Variation of Pb concentration with time in the surface water cover of partially oxidized and weathered tailings underwater.

**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Surface Water Ra-226 Concentration vs Time**

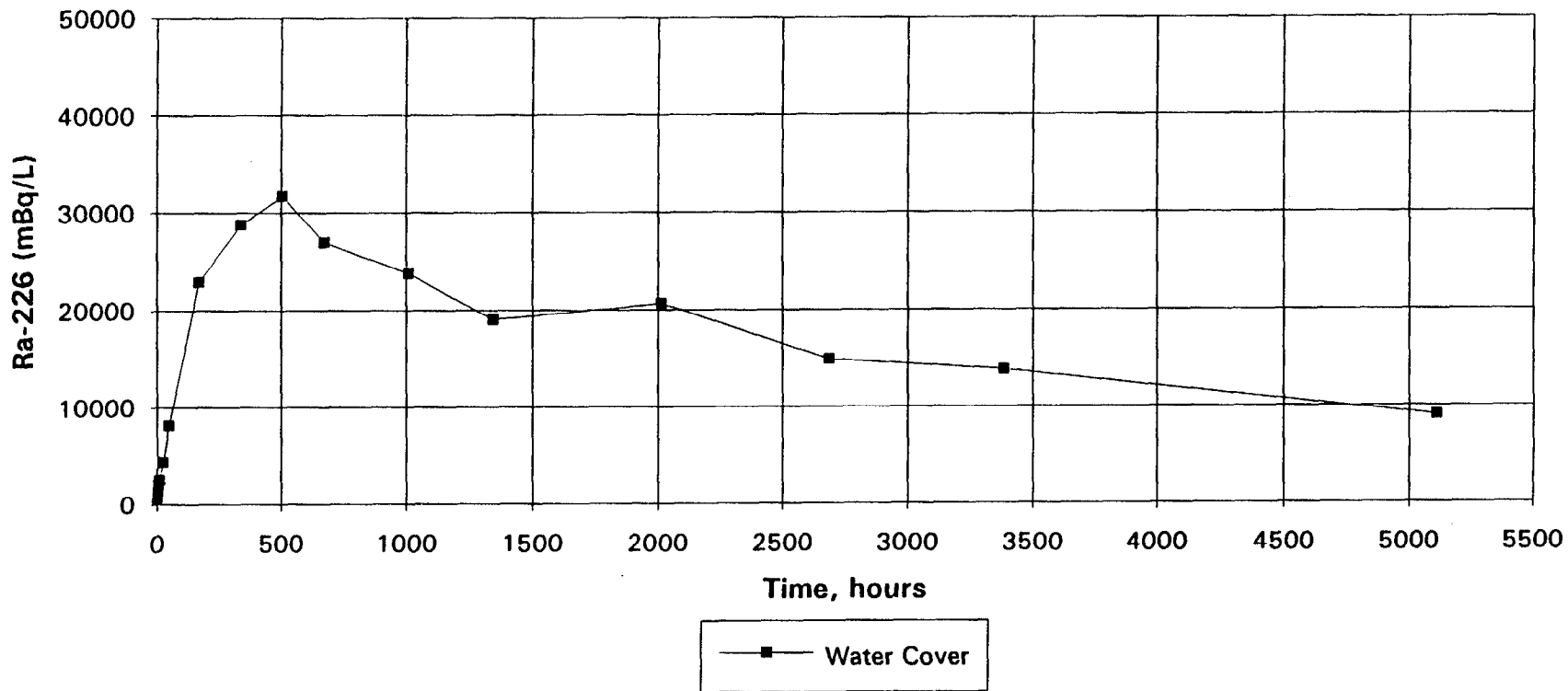


Fig. 36 Variation of Ra-226 concentration with time in the surface water cover of partially oxidized and weathered tailings underwater.

Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Porewater pH vs Time

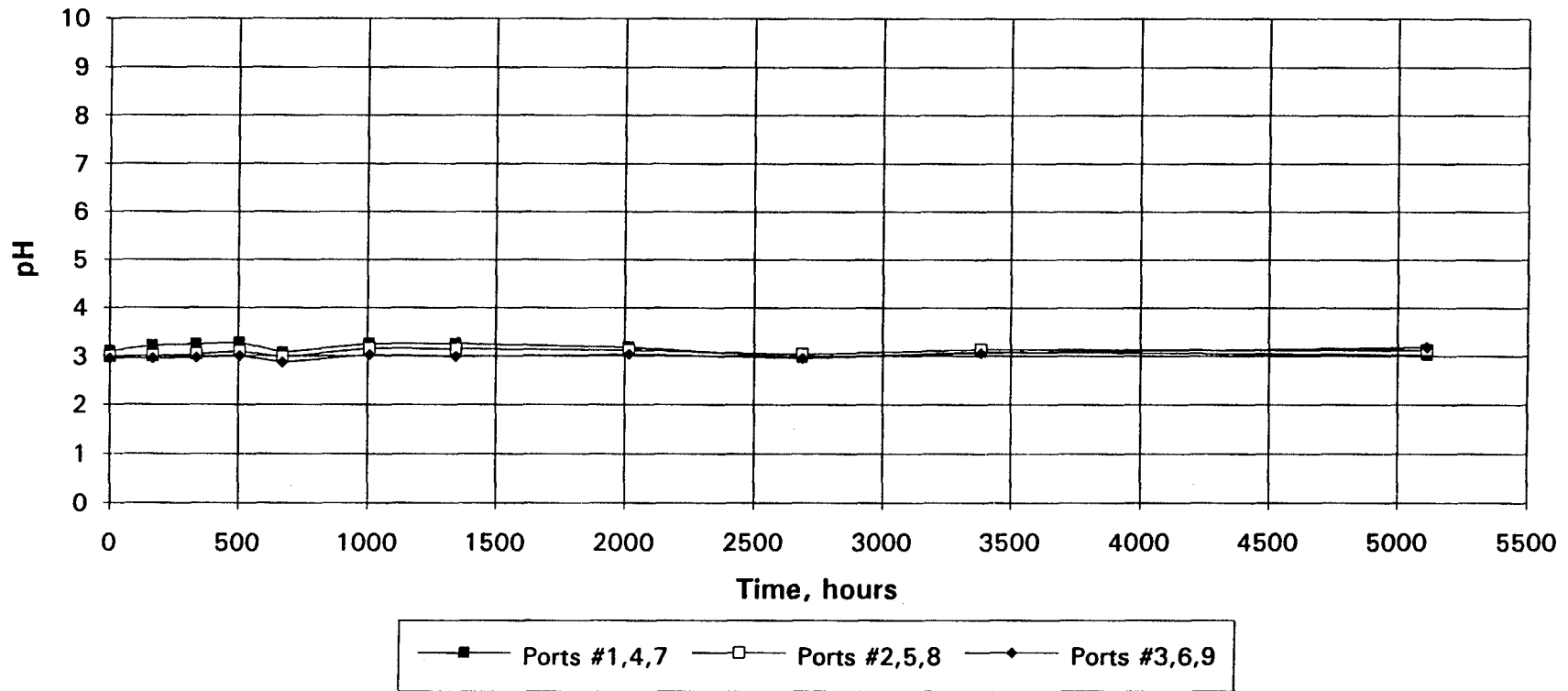


Fig. 37 Variation of pH with time in the porewater of partially oxidized and weathered tailings underwater.

Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Porewater Eh(NHE) vs Time

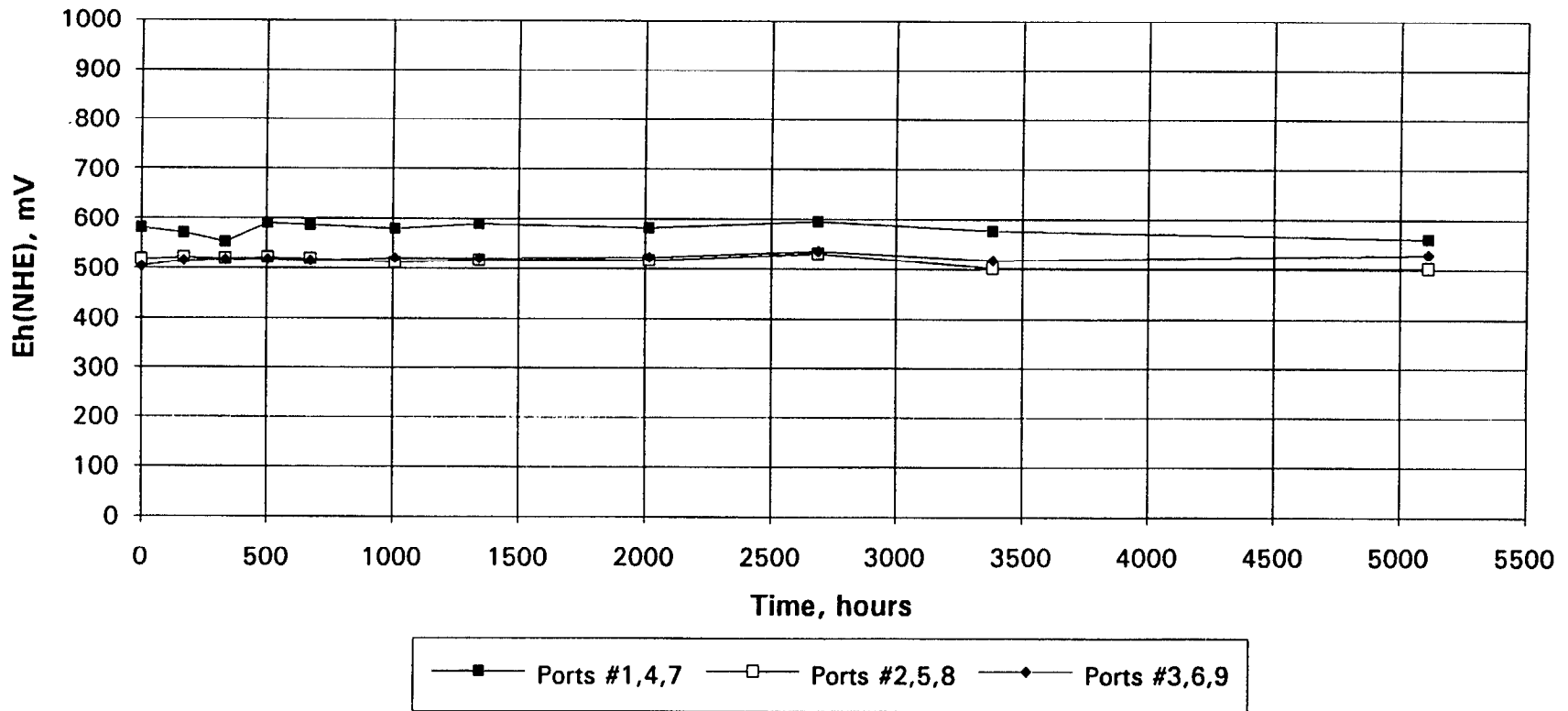


Fig. 38 Variation of Eh with time in the porewater of partially oxidized and weathered tailings underwater.



**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Porewater Electrical Conductivity vs Time**

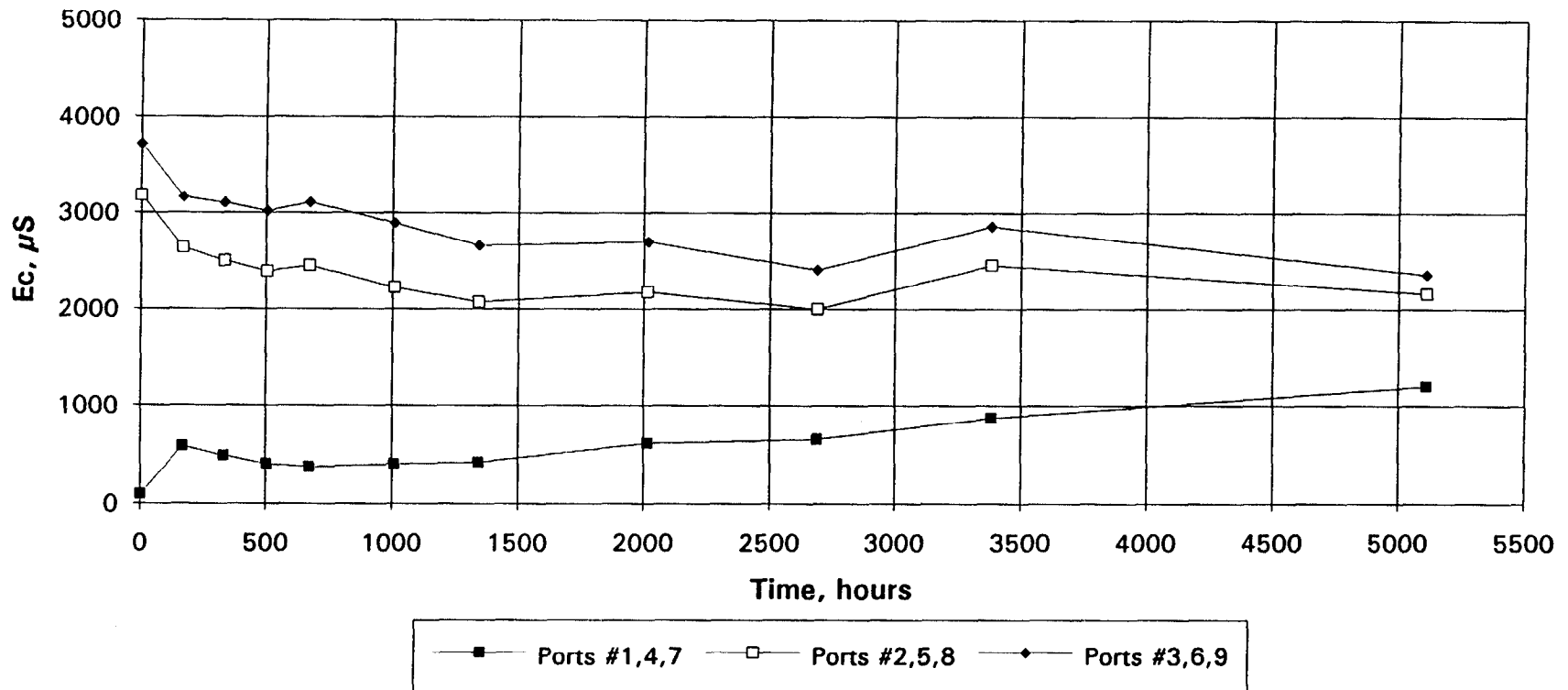
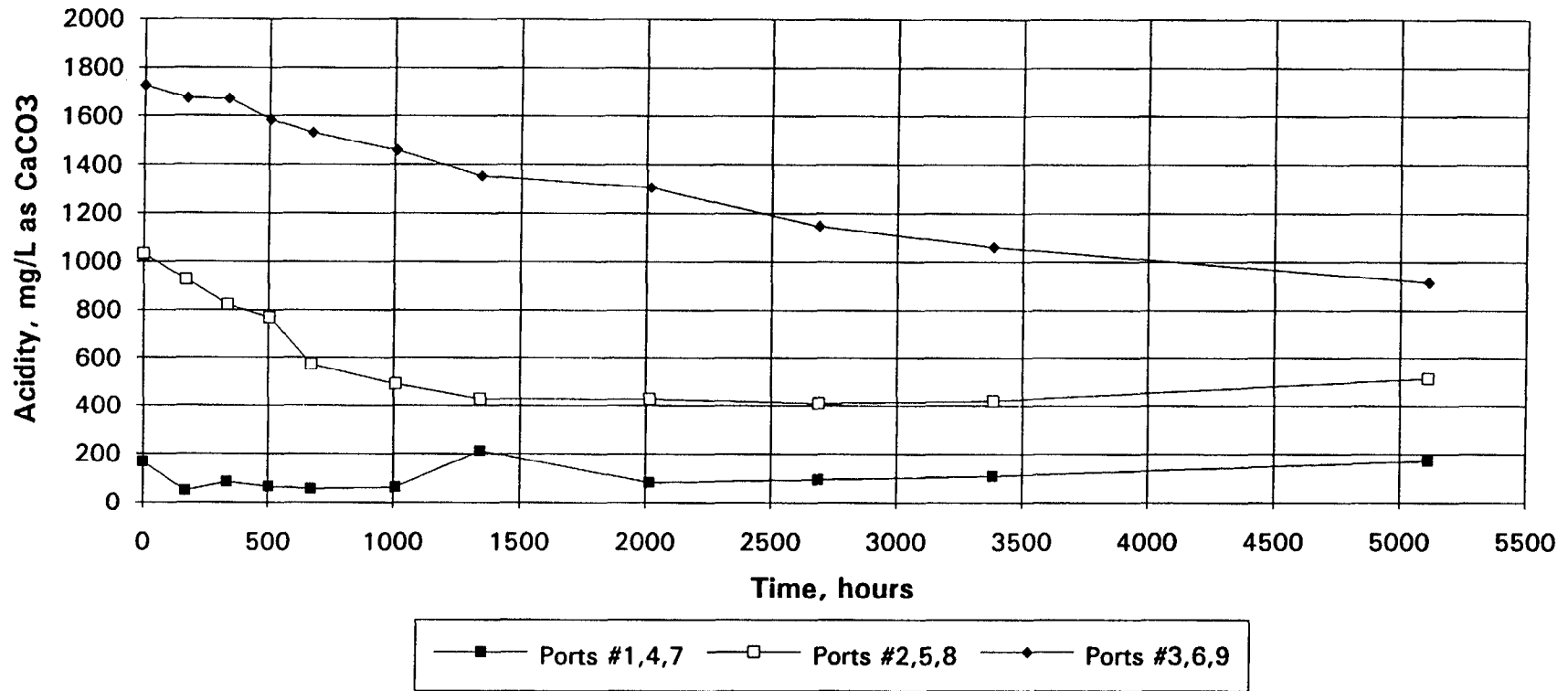


Fig. 39 Variation of Ec with time in the porewater of partially oxidized and weathered tailings underwater.

**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Porewater Acidity vs Time**



**Fig. 40** Variation of acidity with time in the porewater of partially oxidized and weathered tailings underwater.

Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Porewater Sulphate Concentration vs Time

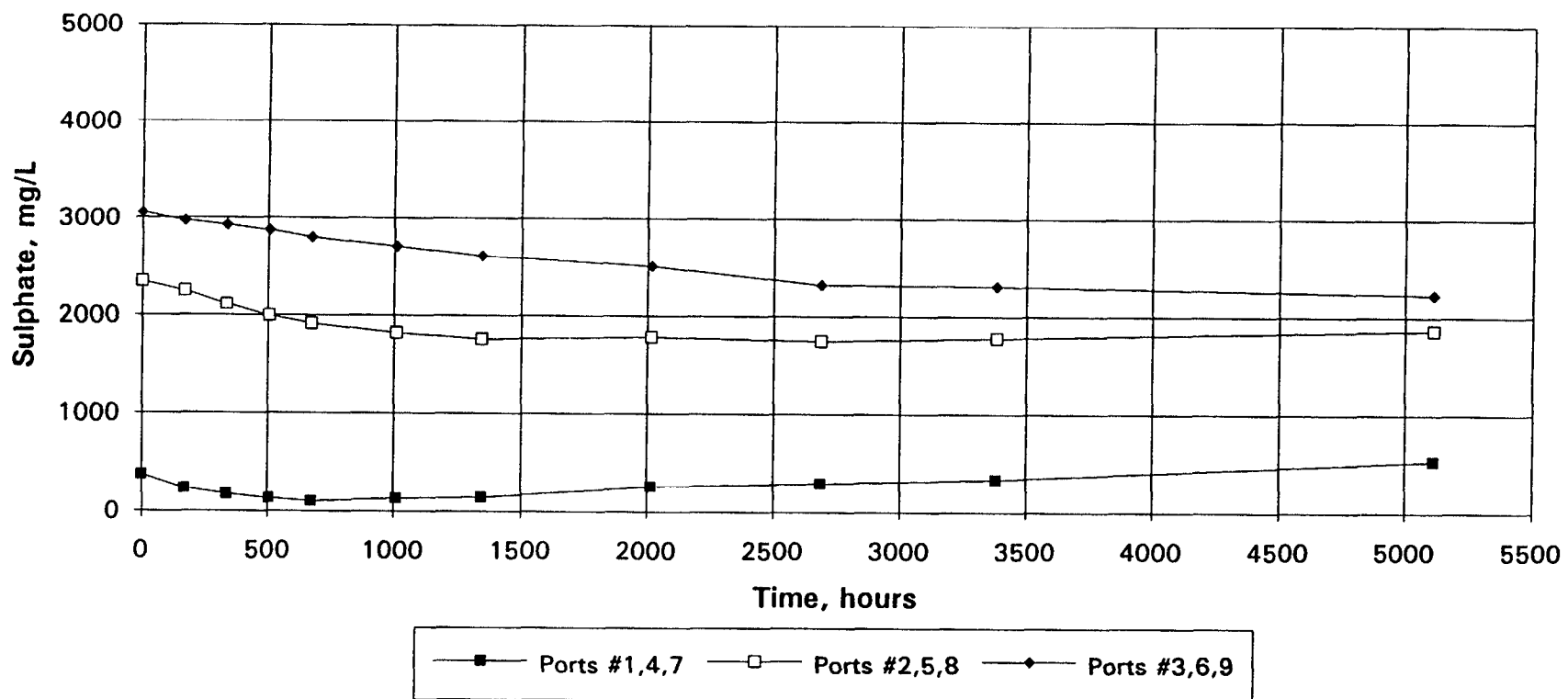


Fig. 41 Variation of  $\text{SO}_4^{-2}$  concentration with time in the porewater of partially oxidized and weathered tailings underwater.

Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Porewater Iron Concentration vs Time

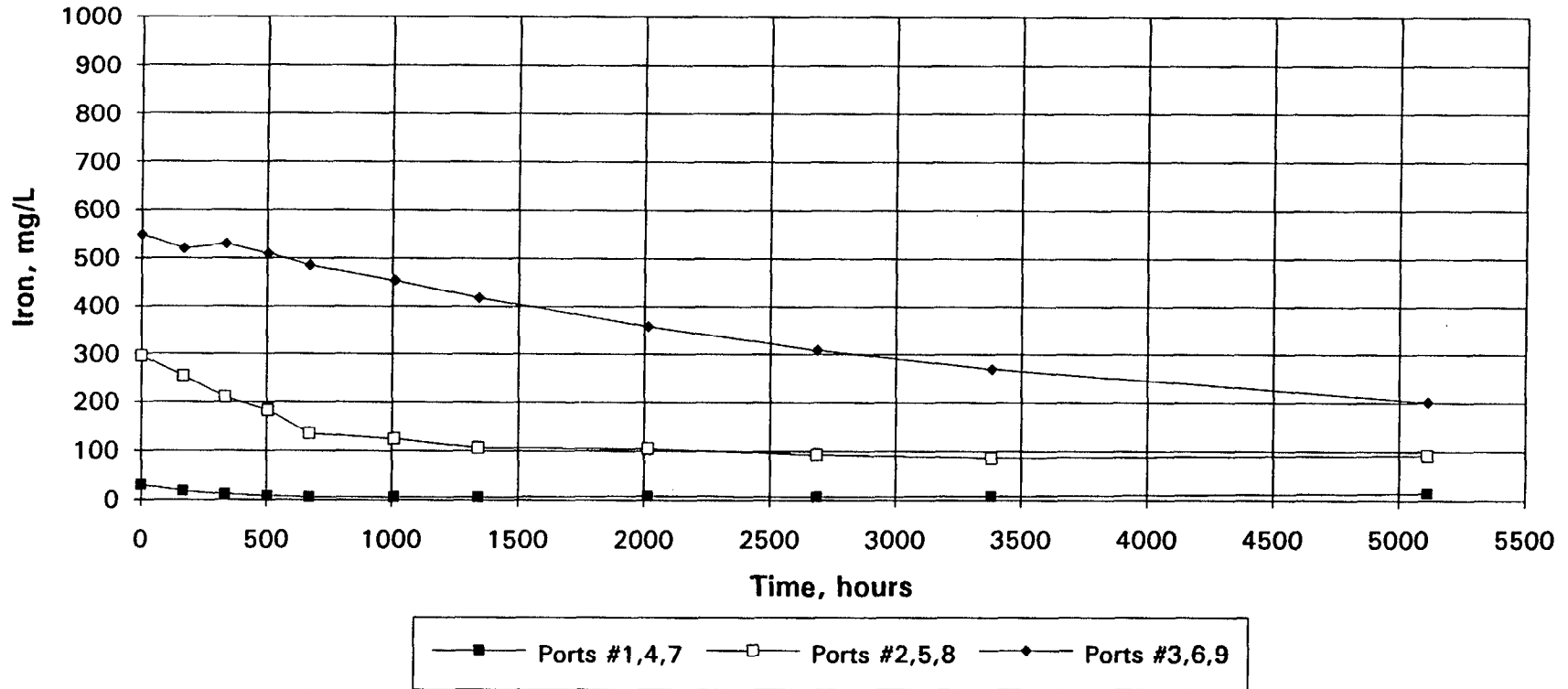


Fig. 42 Variation of total Fe concentration with time in the porewater of partially oxidized and weathered tailings underwater.

**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Porewater Calcium Concentration vs Time**

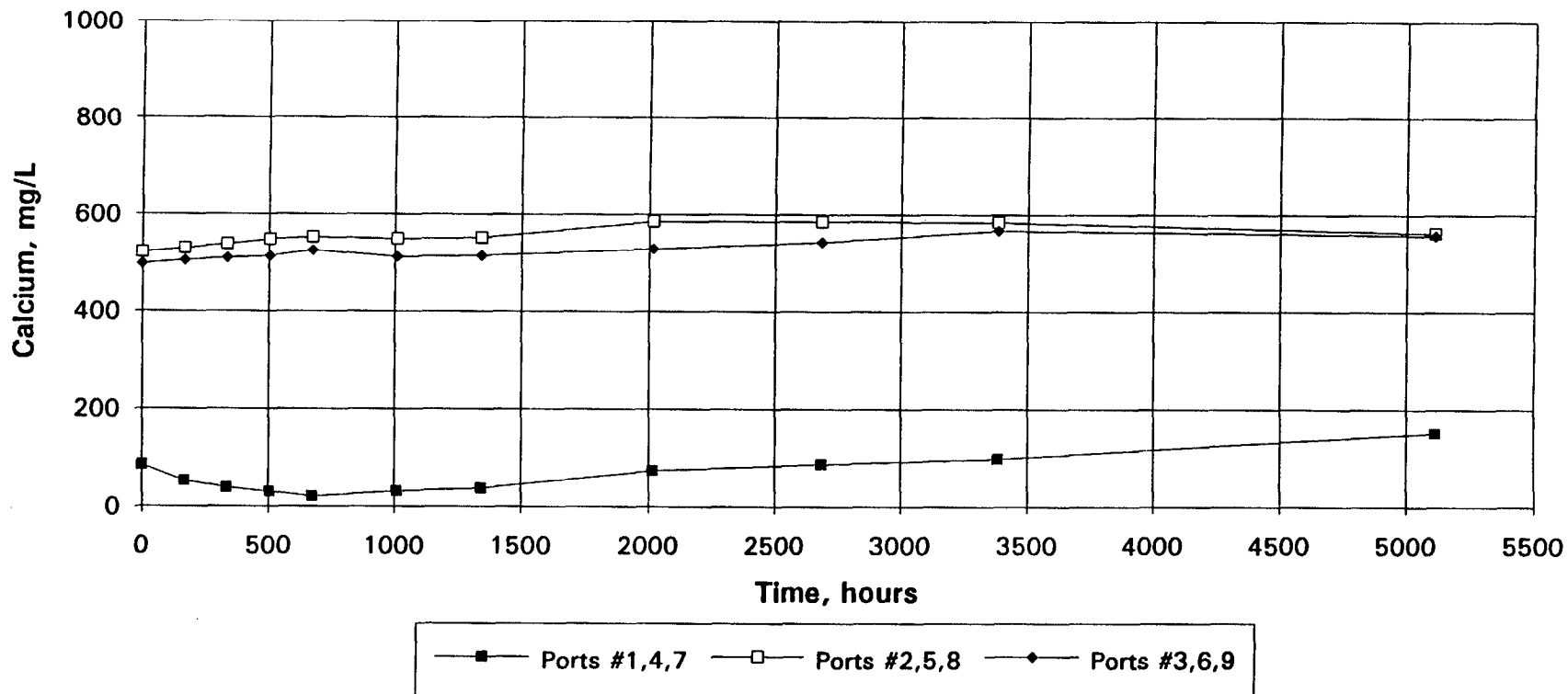


Fig. 43 Variation of Ca concentration with time in the porewater of partially oxidized and weathered tailings underwater.

Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Porewater Magnesium Concentration vs Time

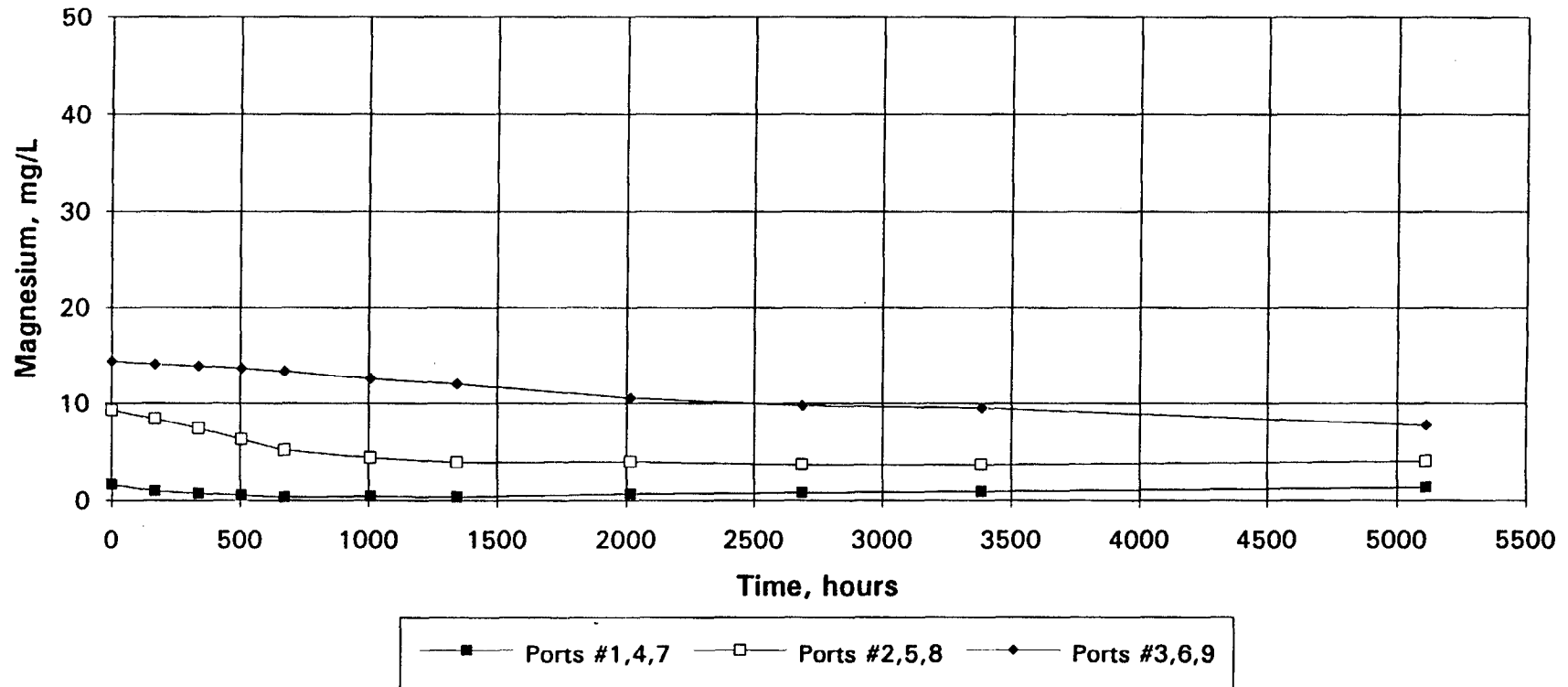
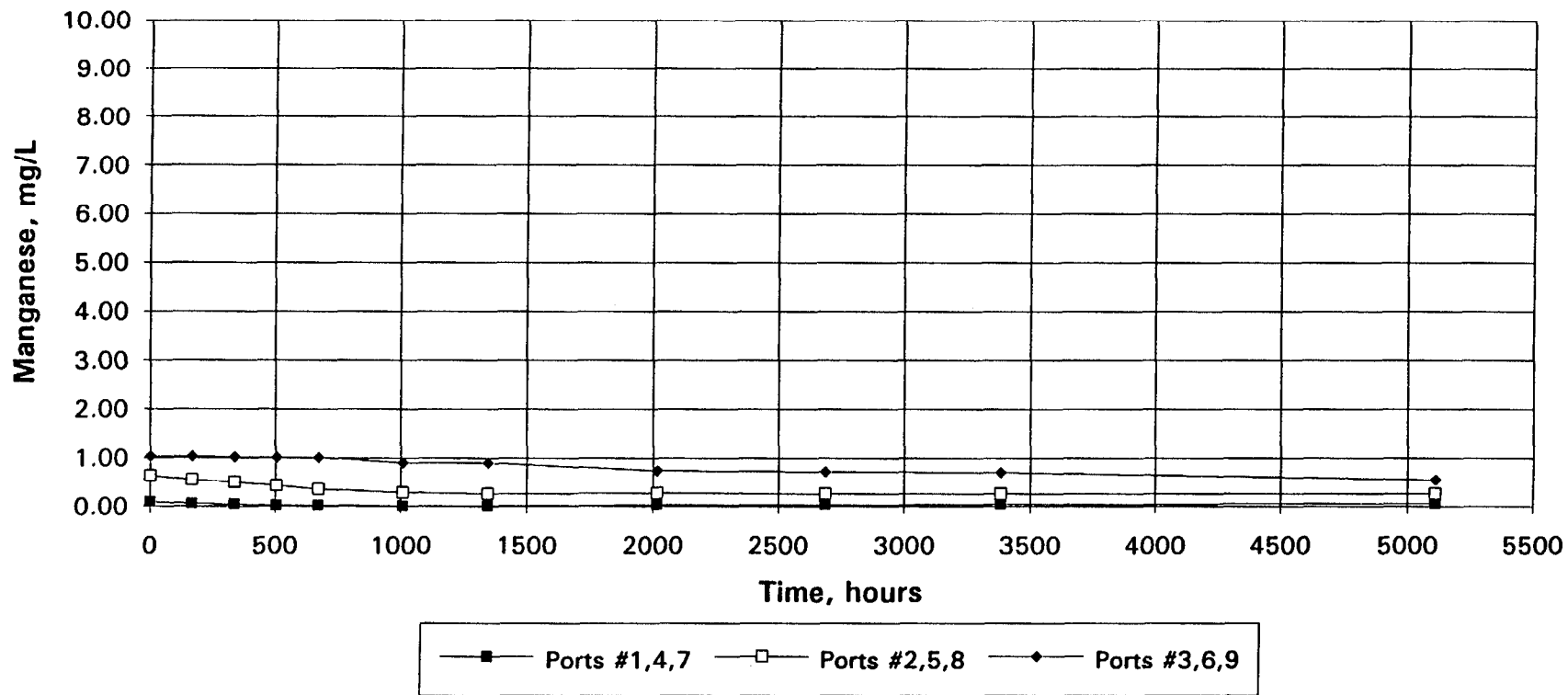


Fig. 44 Variation of Mg concentration with time in the porewater of partially oxidized and weathered tailings underwater.

**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Porewater Manganese Concentration vs Time**



**Fig. 45** Variation of Mn concentration with time in the porewater of partially oxidized and weathered tailings underwater.

**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Porewater Lead Concentration vs Time**

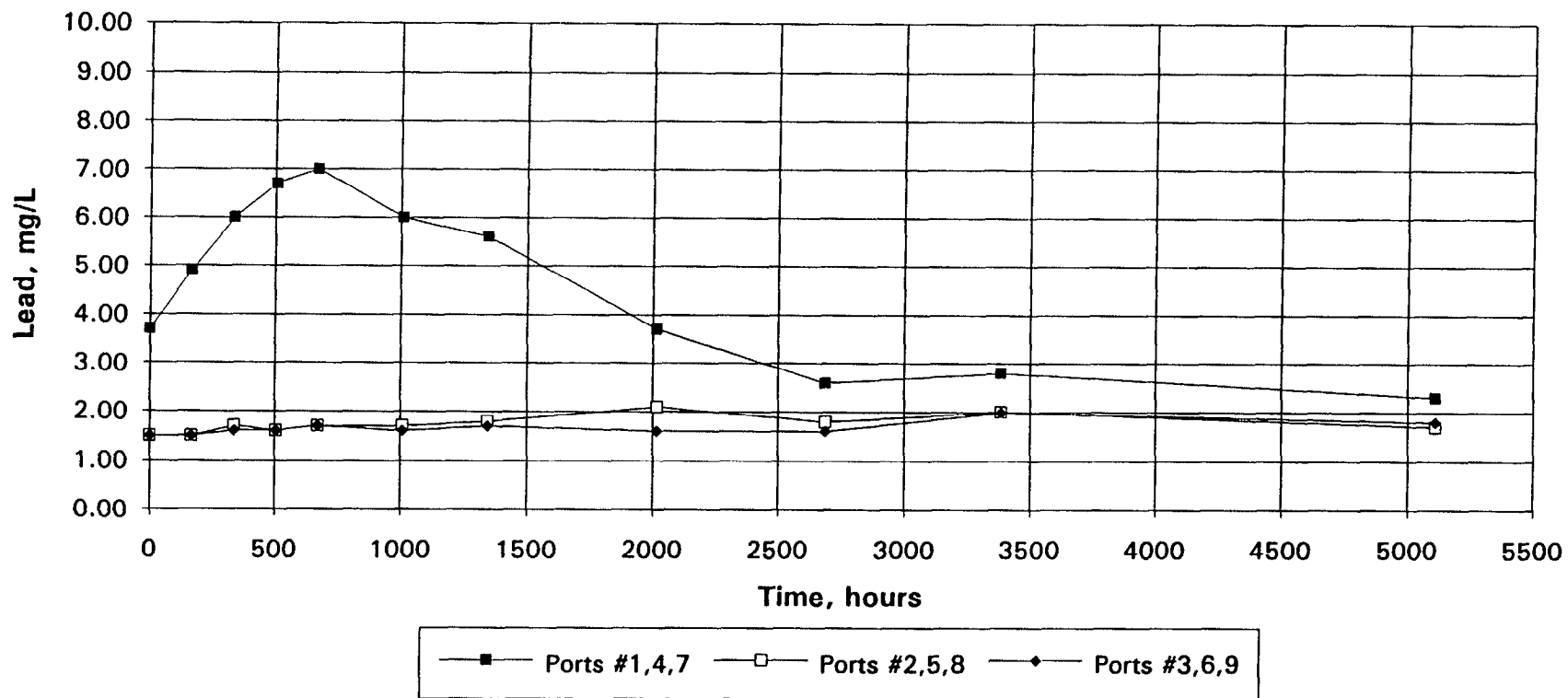
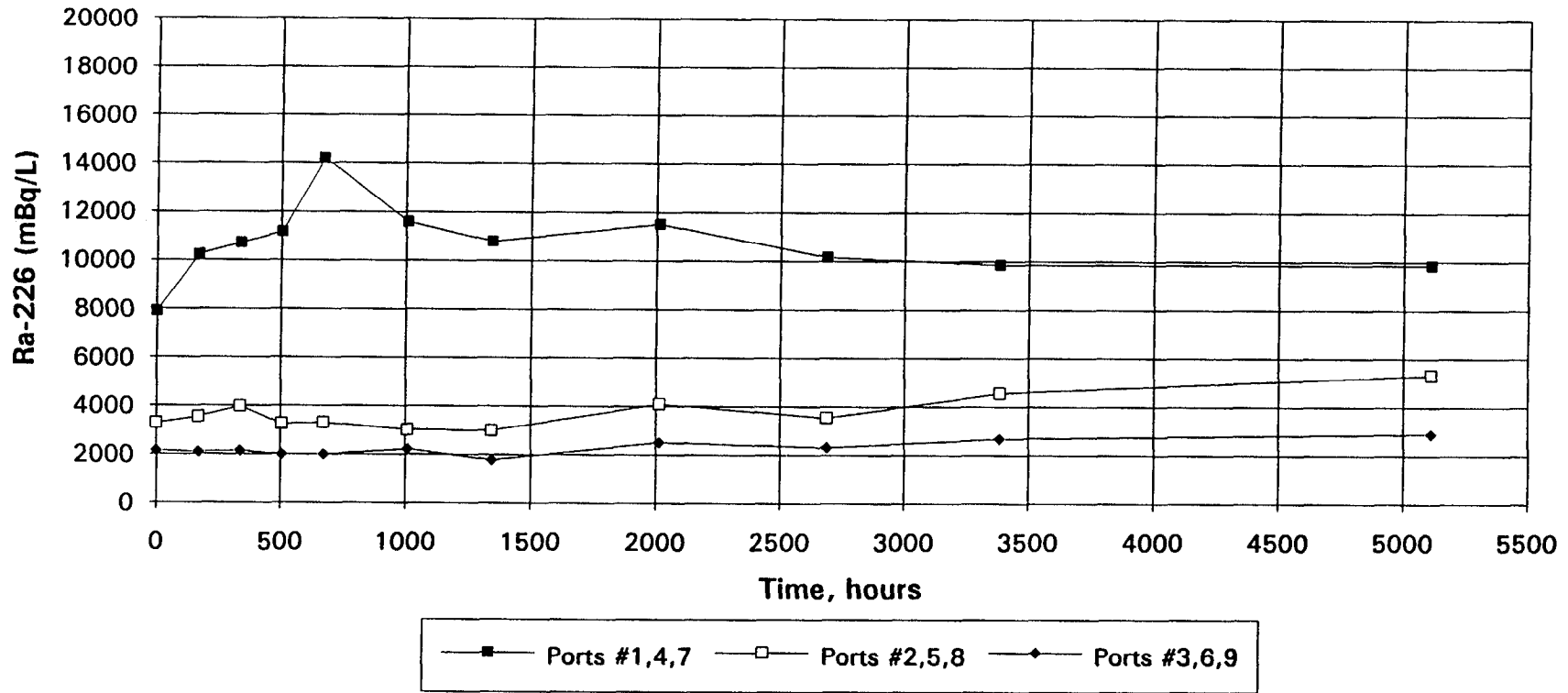


Fig. 46 Variation of Pb concentration with time in the porewater of partially oxidized and weathered tailings underwater.



**Shallow Water Cover on Partially Oxidized and Weathered Tailings (Aquarium #2)  
Porewater Ra-226 Concentration vs Time**



**Fig. 47** Variation of Ra-226 concentration with time in the porewater of partially oxidized and weathered tailings underwater.

## **TABLES**

Table 1 - Surface water quality results for un-oxidized tailings underwater (Aquarium #1).

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Volume (ml)</b>	<b>Temp °C</b>	<b>pH</b>	<b>Eh (mV)</b>	<b>Eh(NHE)</b>	<b>Ec (µS/cm)</b>	<b>Acidity (mg/l) CaCO<sub>3</sub></b>	<b>Alkalinity (mg/l) CaCO<sub>3</sub></b>
<b>Pretrial</b>	<b>WATER COVER</b>	1000	20.3	4.39	393	637	486	12	0
<b>0</b>	<b>WATER COVER</b>	1000	20.5	5.32	309	553	14	4	1
<b>1</b>	<b>WATER COVER</b>	1000	22.7	5.27	302	546	16	4	1
<b>5</b>	<b>WATER COVER</b>	1000	18.3	5.25	353	597	22	4	1
<b>10</b>	<b>WATER COVER</b>	1000	21.3	5.13	363	607	26	5	1
<b>24</b>	<b>WATER COVER</b>	1000	21.4	5.07	321	565	33	5	1
<b>48</b>	<b>WATER COVER</b>	1000	21.7	4.88	326	570	36	5	0
<b>168</b>	<b>WATER COVER</b>	1000	21.2	4.65	394	638	75	6	0
<b>336</b>	<b>WATER COVER</b>	1000	21.5	4.53	372	616	117	6	0
<b>504</b>	<b>WATER COVER</b>	1000	19.6	4.18	372	616	146	20	0
<b>672</b>	<b>WATER COVER</b>	1000	18.7	4.35	400	644	197	8	0
<b>1008</b>	<b>WATER COVER</b>	1000	23.5	4.25	386	630	219	9	0
<b>1344</b>	<b>WATER COVER</b>	1000	20.1	4.25	382	626	268	10	0
<b>1992</b>	<b>WATER COVER</b>	1000	23.4	4.22	433	677	381	13	0
<b>2664</b>	<b>WATER COVER</b>	1000	20.6	4.18	442	686	442	13	0
<b>3360</b>	<b>WATER COVER</b>	1000	22.9	4.15	446	690	561	14	0
<b>5088</b>	<b>WATER COVER</b>	1000	21.9	4.19	349	593	664	17	0

Table 1 - Surface water quality results for un-oxidized tailings underwater (Aquarium #1).

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Al (mg/l)</b>	<b>Ca (mg/l)</b>	<b>Ce (mg/l)</b>	<b>Cu (mg/l)</b>	<b>Fe Total (mg/l)</b>	<b>Mg (mg/l)</b>	<b>Mn (mg/l)</b>
<b>Pretrial</b>	<b>WATER COVER</b>	0.63	77.58	<0.08	<0.004	0.01	2.70	0.13
<b>0</b>	<b>WATER COVER</b>	<0.03	1.18	<0.08	<0.004	0.01	0.03	0.01
<b>1</b>	<b>WATER COVER</b>	<0.03	1.40	<0.08	<0.004	0.01	0.11	0.01
<b>5</b>	<b>WATER COVER</b>	<0.03	1.90	<0.08	<0.004	0.01	0.13	0.01
<b>10</b>	<b>WATER COVER</b>	<0.03	2.29	<0.08	<0.004	0.01	0.16	0.01
<b>24</b>	<b>WATER COVER</b>	<0.03	3.03	<0.08	<0.004	0.01	0.21	0.01
<b>48</b>	<b>WATER COVER</b>	<0.03	3.90	<0.08	<0.004	0.01	0.23	0.01
<b>168</b>	<b>WATER COVER</b>	<0.03	8.70	<0.08	<0.004	0.01	0.40	0.02
<b>336</b>	<b>WATER COVER</b>	0.12	15.23	<0.08	<0.004	0.01	0.61	0.03
<b>504</b>	<b>WATER COVER</b>	0.15	21.05	<0.08	<0.004	0.01	0.78	0.03
<b>672</b>	<b>WATER COVER</b>	0.23	25.87	<0.08	<0.004	0.01	0.93	0.05
<b>1008</b>	<b>WATER COVER</b>	0.34	37.47	<0.08	<0.004	0.01	1.25	0.06
<b>1344</b>	<b>WATER COVER</b>	0.45	48.58	<0.08	<0.004	0.01	1.54	0.07
<b>1992</b>	<b>WATER COVER</b>	0.72	73.40	0.09	<0.004	0.02	2.22	0.09
<b>2664</b>	<b>WATER COVER</b>	0.93	93.24	0.13	<0.004	0.01	2.70	0.11
<b>3360</b>	<b>WATER COVER</b>	1.04	109.20	0.15	<0.004	0.01	3.11	0.12
<b>5088</b>	<b>WATER COVER</b>	1.40	145.20	0.22	0.06	0.01	4.10	0.17

Table 1 - Surface water quality results for un-oxidized tailings underwater (Aquarium #1).

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Ni (mg/l)</b>	<b>Pb (mg/l)</b>	<b>Th (mg/l)</b>	<b>U (mg/l)</b>	<b>Zn (mg/l)</b>	<b>Sulphate (mg/l)</b>	<b>Ra-226 mBq/l</b>
<b>Pretrial</b>	<b>WATER COVER</b>	0.03	0.12	<0.05	<0.2	0.02	201	1014
<b>0</b>	<b>WATER COVER</b>	<0.02	<0.06	<0.05	<0.2	<0.005	3	175
<b>1</b>	<b>WATER COVER</b>	<0.02	<0.06	<0.05	<0.2	<0.005	4	183
<b>5</b>	<b>WATER COVER</b>	<0.02	<0.06	<0.05	<0.2	<0.005	5	210
<b>10</b>	<b>WATER COVER</b>	<0.02	<0.06	<0.05	<0.2	<0.005	6	233
<b>24</b>	<b>WATER COVER</b>	<0.02	<0.06	<0.05	<0.2	<0.005	9	279
<b>48</b>	<b>WATER COVER</b>	<0.02	<0.06	<0.05	<0.2	<0.005	11	367
<b>168</b>	<b>WATER COVER</b>	<0.02	<0.06	<0.05	<0.2	0.01	25	648
<b>336</b>	<b>WATER COVER</b>	0.08	<0.06	<0.05	<0.2	0.02	43	945
<b>504</b>	<b>WATER COVER</b>	<0.02	<0.06	<0.05	<0.2	0.01	57	1260
<b>672</b>	<b>WATER COVER</b>	<0.02	<0.06	<0.05	<0.2	0.02	70	1641
<b>1008</b>	<b>WATER COVER</b>	0.03	0.09	<0.05	<0.2	0.02	101	1799
<b>1344</b>	<b>WATER COVER</b>	0.07	0.11	<0.05	<0.2	0.03	126	2018
<b>1992</b>	<b>WATER COVER</b>	0.06	0.15	<0.05	<0.2	0.04	188	1857
<b>2664</b>	<b>WATER COVER</b>	0.08	0.20	<0.05	<0.2	0.04	232	2051
<b>3360</b>	<b>WATER COVER</b>	0.10	0.24	<0.05	<0.2	0.04	275	1642
<b>5088</b>	<b>WATER COVER</b>	0.17	0.27	<0.05	<0.2	0.07	377	1609

Table 2a - Porewater quality results for un-oxidized tailings underwater (Aquarium #1) at shallow, near surface, depths.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Volume (ml)</b>	<b>Temp °C</b>	<b>pH</b>	<b>Eh (mV)</b>	<b>Eh(NHE)</b>	<b>Ec (µS/cm)</b>	<b>Acidity (mg/l) CaCO<sub>3</sub></b>	<b>Alkalinity (mg/l) CaCO<sub>3</sub></b>
<b>Pretrial</b>	<b>PORTS #1, 4, 7</b>	1000	19.6	4.35	381	625	584	15	0
<b>0</b>	<b>PORTS #1, 4, 7</b>	1000	21.2	4.40	377	621	95	8	0
<b>168</b>	<b>PORTS #1, 4, 7</b>	1000	20.5	4.53	368	612	122	10	0
<b>336</b>	<b>PORTS #1, 4, 7</b>	1000	21.0	4.48	341	585	150	8	0
<b>504</b>	<b>PORTS #1, 4, 7</b>	1000	19.8	4.25	333	577	169	11	0
<b>672</b>	<b>PORTS #1, 4, 7</b>	1000	19.8	4.37	358	602	227	8	0
<b>1008</b>	<b>PORTS #1, 4, 7</b>	1000	21.1	4.30	359	603	257	9	0
<b>1344</b>	<b>PORTS #1, 4, 7</b>	1000	20.2	4.27	368	612	293	15	0
<b>1992</b>	<b>PORTS #1, 4, 7</b>	1000	23.0	4.28	341	585	406	15	0
<b>2664</b>	<b>PORTS #1, 4, 7</b>	1000	21.2	4.21	397	641	462	14	0
<b>3360</b>	<b>PORTS #1, 4, 7</b>	1000	24.7	4.18	411	655	592	12	0
<b>5088</b>	<b>PORTS #1, 4, 7</b>	1000	23.1	4.21	386	630	692	22	0

Table 2a - Porewater quality results for un-oxidized tailings underwater (Aquarium #1) at shallow, near surface, depths.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Al (mg/l)</b>	<b>Ca (mg/l)</b>	<b>Ce (mg/l)</b>	<b>Cu (mg/l)</b>	<b>Fe Total (mg/l)</b>	<b>Mg (mg/l)</b>	<b>Mn (mg/l)</b>
<b>Pretrial</b>	<b>PORTS #1, 4, 7</b>	1.05	97.84	<0.08	<0.04	0.02	3.50	0.17
<b>0</b>	<b>PORTS #1, 4, 7</b>	0.23	10.93	<0.08	<0.04	0.01	0.38	0.03
<b>168</b>	<b>PORTS #1, 4, 7</b>	0.21	16.83	<0.08	<0.04	0.02	0.65	0.04
<b>336</b>	<b>PORTS #1, 4, 7</b>	0.26	21.47	<0.08	<0.04	0.02	0.82	0.04
<b>504</b>	<b>PORTS #1, 4, 7</b>	0.29	27.86	<0.08	<0.04	0.04	0.99	0.05
<b>672</b>	<b>PORTS #1, 4, 7</b>	0.38	33.36	<0.08	<0.04	0.03	1.17	0.06
<b>1008</b>	<b>PORTS #1, 4, 7</b>	0.47	46.52	<0.08	<0.04	0.04	1.47	0.08
<b>1344</b>	<b>PORTS #1, 4, 7</b>	0.63	55.38	<0.08	<0.04	0.02	1.71	0.08
<b>1992</b>	<b>PORTS #1, 4, 7</b>	0.90	81.57	0.11	<0.04	0.15	2.40	0.10
<b>2664</b>	<b>PORTS #1, 4, 7</b>	1.17	100.10	0.10	0.05	0.04	2.80	0.11
<b>3360</b>	<b>PORTS #1, 4, 7</b>	1.30	115.40	0.16	<0.04	0.02	3.29	0.14
<b>5088</b>	<b>PORTS #1, 4, 7</b>	1.60	152.40	0.20	0.05	<0.008	4.40	0.19

Table 2a - Porewater quality results for un-oxidized tailings underwater (Aquarium #1) at shallow, near surface, depths.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Ni (mg/l)</b>	<b>Pb (mg/l)</b>	<b>Th (mg/l)</b>	<b>U (mg/l)</b>	<b>Zn (mg/l)</b>	<b>Sulphate (mg/l)</b>	<b>Ra-226 mBq/l</b>
<b>Pretrial</b>	<b>PORTS #1, 4, 7</b>	<0.02	0.22	<0.05	<0.2	0.03	254	1437
<b>0</b>	<b>PORTS #1, 4, 7</b>	<0.02	<0.08	<0.05	<0.2	0.04	30	928
<b>168</b>	<b>PORTS #1, 4, 7</b>	0.01	0.07	<0.05	<0.2	0.01	47	1655
<b>336</b>	<b>PORTS #1, 4, 7</b>	<0.02	<0.08	<0.05	<0.2	0.01	58	1693
<b>504</b>	<b>PORTS #1, 4, 7</b>	0.02	0.10	<0.05	<0.2	0.02	75	1842
<b>672</b>	<b>PORTS #1, 4, 7</b>	0.03	0.11	<0.05	<0.2	0.02	90	2059
<b>1008</b>	<b>PORTS #1, 4, 7</b>	0.06	0.13	<0.05	<0.2	0.08	117	2156
<b>1344</b>	<b>PORTS #1, 4, 7</b>	0.03	0.14	<0.05	<0.2	0.04	143	3599
<b>1992</b>	<b>PORTS #1, 4, 7</b>	0.06	0.20	<0.05	<0.2	0.04	208	2056
<b>2664</b>	<b>PORTS #1, 4, 7</b>	0.07	0.23	<0.05	<0.2	0.04	249	1834
<b>3360</b>	<b>PORTS #1, 4, 7</b>	0.10	0.26	<0.05	<0.2	0.04	295	1695
<b>5088</b>	<b>PORTS #1, 4, 7</b>	0.17	0.31	<0.05	<0.2	0.07	398	1764



Table 2b - Porewater quality results for un-oxidized tailings underwater (Aquarium #1), at intermediate depths.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Volume (ml)</b>	<b>Temp °C</b>	<b>pH</b>	<b>Eh (mV)</b>	<b>Eh(NHE)</b>	<b>Ec (µS/cm)</b>	<b>Acidity (mg/l) CaCO<sub>3</sub></b>	<b>Alkalinity (mg/l) CaCO<sub>3</sub></b>
<b>Pretrial</b>	<b>PORTS #2, 5, 8</b>	700	20.2	6.40	227	471	840	4	0
<b>0</b>	<b>PORTS #2, 5, 8</b>	1000	21.5	6.10	153	397	399	6	5
<b>168</b>	<b>PORTS #2, 5, 8</b>	1000	21.1	5.59	204	448	435	6	3
<b>336</b>	<b>PORTS #2, 5, 8</b>	1000	21.6	6.20	105	349	660	6	0
<b>504</b>	<b>PORTS #2, 5, 8</b>	1000	19.7	5.48	169	413	469	10	3
<b>672</b>	<b>PORTS #2, 5, 8</b>	1000	20.1	5.58	174	418	547	8	3
<b>1008</b>	<b>PORTS #2, 5, 8</b>	1000	21.2	5.34	195	439	518	10	2
<b>1344</b>	<b>PORTS #2, 5, 8</b>	1000	20.4	5.19	205	449	574	9	2
<b>1992</b>	<b>PORTS #2, 6, 9</b>	1000	23.2	5.45	201	445	671	11	3
<b>2664</b>	<b>PORTS #2, 5, 8</b>	1000	20.4	5.27	248	492	689	10	5
<b>3360</b>	<b>PORTS #2, 5, 8</b>	1000	24.6	5.21	203	447	878	11	2
<b>5088</b>	<b>PORTS #2, 5, 8</b>	1000	23.4	5.40	175	419	943	18	4

Table 2b - Porewater quality results for un-oxidized tailings underwater (Aquarium #1), at intermediate depths.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Al (mg/l)</b>	<b>Ca (mg/l)</b>	<b>Ce (mg/l)</b>	<b>Cu (mg/l)</b>	<b>Fe Total (mg/l)</b>	<b>Mg (mg/l)</b>	<b>Mn (mg/l)</b>
<b>Pretrial</b>	<b>PORTS #2, 5, 8</b>	0.04	190.50	<0.08	<0.004	0.89	10.01	0.39
<b>0</b>	<b>PORTS #2, 5, 8</b>	<0.03	71.36	<0.08	<0.004	0.30	2.99	0.13
<b>168</b>	<b>PORTS #2, 5, 8</b>	0.10	89.78	<0.08	<0.004	0.61	3.55	0.17
<b>336</b>	<b>PORTS #2, 5, 8</b>	<0.03	143.80	<0.08	<0.004	0.79	6.20	0.24
<b>504</b>	<b>PORTS #2, 5, 8</b>	0.04	102.00	<0.08	<0.004	1.24	3.70	0.17
<b>672</b>	<b>PORTS #2, 5, 8</b>	0.13	105.90	<0.08	<0.004	1.20	3.70	0.18
<b>1008</b>	<b>PORTS #2, 5, 8</b>	0.21	115.90	<0.08	<0.004	1.09	4.04	0.19
<b>1344</b>	<b>PORTS #2, 5, 8</b>	0.26	135.60	<0.08	<0.004	1.28	4.56	0.21
<b>1992</b>	<b>PORTS #2, 6, 9</b>	0.36	156.90	<0.08	<0.004	2.37	5.60	0.24
<b>2664</b>	<b>PORTS #2, 5, 8</b>	0.41	177.40	<0.08	<0.004	1.85	6.18	0.27
<b>3360</b>	<b>PORTS #2, 5, 8</b>	0.48	192.70	<0.08	<0.004	1.70	6.62	0.29
<b>5088</b>	<b>PORTS #2, 5, 8</b>	0.44	231.90	<0.08	<0.004	2.17	8.50	0.37

Table 2b - Porewater quality results for un-oxidized tailings underwater (Aquarium #1), at intermediate depths.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Ni (mg/l)</b>	<b>Pb (mg/l)</b>	<b>Th (mg/l)</b>	<b>U (mg/l)</b>	<b>Zn (mg/l)</b>	<b>Sulphate (mg/l)</b>	<b>Ra-226 mBq/l</b>
<b>Pretrial</b>	<b>PORTS #2, 5, 8</b>	<0.02	<0.06	<0.05	<0.2	0.02	506	869
<b>0</b>	<b>PORTS #2, 5, 8</b>	<0.02	<0.06	<0.05	<0.2	0.02	177	2667
<b>168</b>	<b>PORTS #2, 5, 8</b>	<0.02	<0.06	<0.05	<0.2	0.01	235	2224
<b>336</b>	<b>PORTS #2, 5, 8</b>	0.14	<0.06	<0.05	<0.2	0.02	380	1503
<b>504</b>	<b>PORTS #2, 5, 8</b>	<0.02	0.08	<0.05	<0.2	0.03	263	1646
<b>672</b>	<b>PORTS #2, 5, 8</b>	<0.02	0.06	<0.05	<0.2	0.01	273	1570
<b>1008</b>	<b>PORTS #2, 5, 8</b>	<0.02	0.07	<0.05	<0.2	0.02	291	1678
<b>1344</b>	<b>PORTS #2, 5, 8</b>	0.03	0.08	<0.05	<0.2	0.05	338	2439
<b>1992</b>	<b>PORTS #2, 6, 9</b>	0.04	0.07	<0.05	<0.2	0.03	410	1410
<b>2664</b>	<b>PORTS #2, 5, 8</b>	0.05	0.08	<0.05	<0.2	0.02	443	1282
<b>3360</b>	<b>PORTS #2, 5, 8</b>	0.06	0.09	<0.05	<0.2	0.03	491	1404
<b>5088</b>	<b>PORTS #2, 5, 8</b>	0.10	0.07	<0.05	<0.2	0.04	605	1505

Table 2c - Porewater quality results for un-oxidized tailings underwater (Aquarium #1), at depths near tailings bottom.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Volume (ml)</b>	<b>Temp °C</b>	<b>pH</b>	<b>Eh (mV)</b>	<b>Eh(NHE)</b>	<b>Ec (µS/cm)</b>	<b>Acidity (mg/l) CaCO<sub>3</sub></b>	<b>Alkalinity (mg/l) CaCO<sub>3</sub></b>
<b>Pretrial</b>	<b>PORTS #3, 6, 8</b>	850	20.6	5.22	262	506	925	12	15
<b>0</b>	<b>PORTS #3, 6, 8</b>	1000	22.7	6.21	138	382	620	12	7
<b>168</b>	<b>PORTS #3, 6, 8</b>	1000	21.4	6.30	126	370	583	4	8
<b>336</b>	<b>PORTS #3, 6, 8</b>	1000	20.8	5.21	212	456	374	10	0
<b>504</b>	<b>PORTS #3, 6, 8</b>	1000	21.4	5.98	132	376	585	6	6
<b>672</b>	<b>PORTS #3, 6, 8</b>	1000	21.0	6.00	140	384	725	9	5
<b>1008</b>	<b>PORTS #3, 6, 8</b>	1000	21.4	5.99	118	362	757	7	5
<b>1344</b>	<b>PORTS #3, 6, 8</b>	1000	21.5	5.47	182	426	589	8	2
<b>1992</b>	<b>PORTS #3, 6, 8</b>	1000	22.9	5.35	206	450	684	9	3
<b>2664</b>	<b>PORTS #3, 6, 8</b>	1000	20.6	5.15	270	514	672	12	4
<b>3360</b>	<b>PORTS #3, 6, 8</b>	1000	21.4	4.86	255	499	785	12	1
<b>5088</b>	<b>PORTS #3, 6, 8</b>	1000	22.6	4.90	245	489	886	16	3

Table 2c - Porewater quality results for un-oxidized tailings underwater (Aquarium #1), at depths near tailings bottom.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Al (mg/l)</b>	<b>Ca (mg/l)</b>	<b>Ce (mg/l)</b>	<b>Cu (mg/l)</b>	<b>Fe Total (mg/l)</b>	<b>Mg (mg/l)</b>	<b>Mn (mg/l)</b>
<b>Pretrial</b>	<b>PORTS #3, 6, 8</b>	0.69	219.70	0.10	<0.004	1.57	11.78	0.38
<b>0</b>	<b>PORTS #3, 6, 8</b>	<0.03	115.70	<0.08	<0.004	0.52	6.09	0.17
<b>168</b>	<b>PORTS #3, 6, 8</b>	0.06	120.60	<0.08	<0.004	0.62	6.80	0.18
<b>336</b>	<b>PORTS #3, 6, 8</b>	0.20	71.57	<0.08	<0.004	0.79	3.50	0.11
<b>504</b>	<b>PORTS #3, 6, 8</b>	0.08	130.90	<0.08	<0.004	0.99	6.60	0.20
<b>672</b>	<b>PORTS #3, 6, 8</b>	0.04	151.70	<0.08	<0.004	1.05	6.60	0.21
<b>1008</b>	<b>PORTS #3, 6, 8</b>	<0.03	186.90	<0.08	<0.004	1.18	6.44	0.21
<b>1344</b>	<b>PORTS #3, 6, 8</b>	0.19	141.00	<0.08	<0.004	0.55	5.40	0.18
<b>1992</b>	<b>PORTS #3, 6, 8</b>	0.31	162.50	<0.08	<0.004	1.01	6.58	0.22
<b>2664</b>	<b>PORTS #3, 6, 8</b>	0.57	169.40	0.09	<0.004	0.58	7.20	0.23
<b>3360</b>	<b>PORTS #3, 6, 8</b>	0.77	173.30	0.12	<0.004	0.47	7.10	0.23
<b>5088</b>	<b>PORTS #3, 6, 8</b>	1.02	209.00	0.20	<0.004	0.54	8.70	0.29

Table 2c - Porewater quality results for un-oxidized tailings underwater (Aquarium #1), at depths near tailings bottom.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Ni (mg/l)</b>	<b>Pb (mg/l)</b>	<b>Th (mg/l)</b>	<b>U (mg/l)</b>	<b>Zn (mg/l)</b>	<b>Sulphate (mg/l)</b>	<b>Ra-226 mBq/l</b>
<b>Pretrial</b>	<b>PORTS #3, 6, 8</b>	0.02	<0.06	<0.05	<0.2	0.03	581	2206
<b>0</b>	<b>PORTS #3, 6, 8</b>	0.02	<0.06	<0.05	<0.2	0.01	294	2228
<b>168</b>	<b>PORTS #3, 6, 8</b>	0.02	<0.06	<0.05	<0.2	0.01	325	1559
<b>336</b>	<b>PORTS #3, 6, 8</b>	0.02	0.07	<0.05	<0.2	0.06	191	1820
<b>504</b>	<b>PORTS #3, 6, 8</b>	0.02	<0.06	<0.05	<0.2	0.02	342	2104
<b>672</b>	<b>PORTS #3, 6, 8</b>	0.02	<0.06	<0.05	<0.2	0.11	392	1636
<b>1008</b>	<b>PORTS #3, 6, 8</b>	0.02	<0.06	<0.05	<0.2	0.02	479	1995
<b>1344</b>	<b>PORTS #3, 6, 8</b>	0.03	<0.06	<0.05	<0.2	0.04	356	2292
<b>1992</b>	<b>PORTS #3, 6, 8</b>	0.05	<0.06	<0.05	<0.2	0.03	404	1869
<b>2664</b>	<b>PORTS #3, 6, 8</b>	0.07	0.07	<0.05	<0.2	0.03	428	1800
<b>3360</b>	<b>PORTS #3, 6, 8</b>	0.08	0.14	<0.05	<0.2	0.04	452	1908
<b>5088</b>	<b>PORTS #3, 6, 8</b>	0.13	0.16	<0.05	<0.2	0.08	548	2007

Table 3 - Non-linear regression analysis results for Ca, Mg, Mn, Pb, SO<sub>4</sub><sup>-2</sup>, and Ra-226 in the surface water column of un-oxidized tailings underwater. Computed values are given for porewater concentration (C<sub>o</sub>), transfer rate constant ( $\frac{K}{h}$ ), transfer coefficient (K), initial flux (J<sub>o</sub>) at time t = 0 and bulk molecular diffusion coefficient in water (D<sub>o</sub>). For Ra-226, the parametric values are given in mBq.

Parameter	Computed porewater concentration C <sub>o</sub> , mg/l	Transfer rate constant $\frac{K}{h}$ , h <sup>-1</sup>	Transfer coefficient K, m/month	Initial flux (J <sub>o</sub> ), at t = 0, mg/m <sup>2</sup> /h	Bulk molecular diffusion coefficient in water (D <sub>o</sub> ), m <sup>2</sup> /s
Ca	270	1.52 x 10 <sup>-4</sup>	0.032	11.9	6.8 x 10 <sup>-9</sup>
Fe	6.2	2.01 x 10 <sup>-4</sup>	0.042	0.36	9.0 x 10 <sup>-9</sup>
Mg	0.25	2.06 x 10 <sup>-4</sup>	0.043	0.015	9.5 x 10 <sup>-9</sup>
Pb	0.90	6.05 x 10 <sup>-5</sup>	0.126	0.016	2.7 x 10 <sup>-9</sup>
SO <sub>4</sub> <sup>-2</sup>	727	1.41 x 10 <sup>-4</sup>	0.029	29.7	6.9 x 10 <sup>-9</sup>
Ra-226 (mBq)	1775	2.35 x 10 <sup>-3</sup>	0.49	1209	1.05 x 10 <sup>-7</sup>

Table 4 - Surface water quality results for partially weathered and oxidized tailings underwater (Aquarium #2).

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Volume (ml)</b>	<b>Temp °C</b>	<b>pH</b>	<b>Eh (mV)</b>	<b>Eh(NHE)</b>	<b>Ec (µS/cm)</b>	<b>Acidity (mg/l) CaCO<sub>3</sub></b>	<b>Alkalinity (mg/l) CaCO<sub>3</sub></b>
<b>Pretrial</b>	<b>WATER COVER</b>	1000	19.6	2.97	525	769	617	95	0
<b>0</b>	<b>WATER COVER</b>	1000	22.9	4.88	360	604	11	7	0
<b>1</b>	<b>WATER COVER</b>	1000	20.3	4.63	369	613	17	7	0
<b>5</b>	<b>WATER COVER</b>	1000	20.1	4.50	425	669	21	6	0
<b>10</b>	<b>WATER COVER</b>	1000	20.8	4.35	436	680	24	7	0
<b>24</b>	<b>WATER COVER</b>	1000	20.5	4.18	398	642	32	6	0
<b>48</b>	<b>WATER COVER</b>	1000	23.5	4.11	419	663	40	7	0
<b>168</b>	<b>WATER COVER</b>	1000	20.6	3.78	441	685	72	13	0
<b>336</b>	<b>WATER COVER</b>	1000	21.3	3.60	477	721	119	24	0
<b>504</b>	<b>WATER COVER</b>	1000	21.0	3.49	456	700	133	19	0
<b>672</b>	<b>WATER COVER</b>	1000	18.2	3.15	491	735	167	22	0
<b>1008</b>	<b>WATER COVER</b>	1000	20.4	3.33	495	739	184	30	0
<b>1344</b>	<b>WATER COVER</b>	1000	20.1	3.29	479	723	200	31	0
<b>2016</b>	<b>WATER COVER</b>	1000	22.7	3.16	511	755	274	41	0
<b>2688</b>	<b>WATER COVER</b>	1000	21.0	2.87	498	742	299	45	0
<b>3384</b>	<b>WATER COVER</b>	1000	22.4	3.00	526	770	465	50	0
<b>5112</b>	<b>WATER COVER</b>	1000	22.1	2.93	514	758	581	91	0



Table 4 - Surface water quality results for partially weathered and oxidized tailings underwater (Aquarium #2).

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Al (mg/l)</b>	<b>Ca (mg/l)</b>	<b>Ce (mg/l)</b>	<b>Cu (mg/l)</b>	<b>Fe Total (mg/l)</b>	<b>Mg (mg/l)</b>	<b>Mn (mg/l)</b>	<b>Ni (mg/l)</b>
<b>Pretrial</b>	<b>WATER COVER</b>	3.20	29.75	0.09	<0.04	0.94	0.52	0.030	0.02
<b>0</b>	<b>WATER COVER</b>	<0.03	0.19	<0.08	<0.04	0.01	0.02	<0.001	<0.02
<b>1</b>	<b>WATER COVER</b>	<0.03	0.33	<0.08	<0.04	0.02	0.01	<0.001	<0.02
<b>5</b>	<b>WATER COVER</b>	<0.03	0.22	<0.08	0.04	0.01	0.01	<0.001	<0.02
<b>10</b>	<b>WATER COVER</b>	<0.03	0.23	<0.08	<0.04	0.01	0.01	<0.001	<0.02
<b>24</b>	<b>WATER COVER</b>	<0.03	0.34	<0.08	<0.04	0.02	0.03	<0.001	<0.02
<b>48</b>	<b>WATER COVER</b>	<0.03	0.42	<0.08	<0.04	0.01	0.02	<0.001	<0.02
<b>168</b>	<b>WATER COVER</b>	<0.03	0.48	<0.08	<0.04	0.01	0.02	<0.001	<0.02
<b>336</b>	<b>WATER COVER</b>	0.04	0.69	<0.08	<0.04	0.03	0.04	<0.001	<0.02
<b>504</b>	<b>WATER COVER</b>	0.08	0.72	<0.08	<0.04	0.02	0.02	0.005	0.04
<b>672</b>	<b>WATER COVER</b>	0.08	0.80	<0.08	<0.04	0.05	0.01	<0.001	<0.02
<b>1008</b>	<b>WATER COVER</b>	0.14	0.88	<0.08	<0.04	0.04	0.02	0.005	<0.02
<b>1344</b>	<b>WATER COVER</b>	0.15	0.92	<0.08	<0.04	0.01	0.00	0.002	<0.02
<b>2016</b>	<b>WATER COVER</b>	0.31	1.60	<0.08	<0.04	0.04	0.01	0.001	0.14
<b>2688</b>	<b>WATER COVER</b>	0.52	2.40	<0.08	<0.04	0.12	0.06	0.003	0.05
<b>3384</b>	<b>WATER COVER</b>	0.84	4.33	<0.08	<0.04	0.22	0.09	0.005	<0.02
<b>5112</b>	<b>WATER COVER</b>	1.90	11.82	<0.08	<0.04	0.45	0.20	0.010	<0.02

Table 4 - Surface water quality results for partially weathered and oxidized tailings underwater (Aquarium #2).

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Pb (mg/l)</b>	<b>Th (mg/l)</b>	<b>U (mg/l)</b>	<b>Zn (mg/l)</b>	<b>Sulphate (mg/l)</b>	<b>Ra-226 mBq/l</b>
<b>Pretrial</b>	<b>WATER COVER</b>	1.20	<0.05	<0.2	0.030	159	8260
<b>0</b>	<b>WATER COVER</b>	<0.06	<0.05	<0.2	<0.005	1	672
<b>1</b>	<b>WATER COVER</b>	<0.06	<0.05	<0.2	<0.005	3	1357
<b>5</b>	<b>WATER COVER</b>	<0.06	<0.05	<0.2	<0.005	3	2064
<b>10</b>	<b>WATER COVER</b>	<0.06	<0.05	<0.2	0.008	3	2528
<b>24</b>	<b>WATER COVER</b>	<0.06	<0.05	<0.2	<0.005	4	4326
<b>48</b>	<b>WATER COVER</b>	0.08	<0.05	<0.2	<0.005	6	8149
<b>168</b>	<b>WATER COVER</b>	0.26	<0.05	<0.2	0.007	10	22874
<b>336</b>	<b>WATER COVER</b>	0.58	<0.05	<0.2	0.008	14	28733
<b>504</b>	<b>WATER COVER</b>	0.99	<0.05	<0.2	0.010	20	31663
<b>672</b>	<b>WATER COVER</b>	1.10	<0.05	<0.2	0.009	21	26944
<b>1008</b>	<b>WATER COVER</b>	1.60	<0.05	<0.2	0.007	27	23706
<b>1344</b>	<b>WATER COVER</b>	1.60	<0.05	<0.2	0.006	29	18996
<b>2016</b>	<b>WATER COVER</b>	2.20	<0.05	<0.2	<0.005	41	20672
<b>2688</b>	<b>WATER COVER</b>	1.94	<0.05	<0.2	0.007	49	14923
<b>3384</b>	<b>WATER COVER</b>	2.60	<0.05	<0.2	0.010	68	13822
<b>5112</b>	<b>WATER COVER</b>	2.40	0.07	<0.2	0.009	108	9041

Table 5a - Porewater quality results for partially weathered and oxidized tailings underwater (Aquarium #2), at shallow, near surface, depths.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Volume (ml)</b>	<b>Temp °C</b>	<b>pH</b>	<b>Eh (mV)</b>	<b>Eh(NHE)</b>	<b>Ec (µS/cm)</b>	<b>Acidity (mg/l) CaCO<sub>3</sub></b>	<b>Alkalinity (mg/l) CaCO<sub>3</sub></b>
<b>Pretrial</b>	<b>PORTS #1, 4, 7</b>	1000	21.5	2.97	290	534	1661	515	0
<b>0</b>	<b>PORTS #1, 4, 7</b>	1000	23.4	3.10	337	581	94	166	0
<b>168</b>	<b>PORTS #1, 4, 7</b>	1000	21.3	3.22	327	571	588	49	0
<b>336</b>	<b>PORTS #1, 4, 7</b>	1000	23.0	3.24	309	553	486	84	0
<b>504</b>	<b>PORTS #1, 4, 7</b>	1000	23.0	3.29	346	590	404	66	0
<b>672</b>	<b>PORTS #1, 4, 7</b>	1000	19.9	3.08	343	587	376	56	0
<b>1008</b>	<b>PORTS #1, 4, 7</b>	1000	21.0	3.26	335	579	406	64	0
<b>1344</b>	<b>PORTS #1, 4, 7</b>	1000	20.2	3.26	345	589	424	212	0
<b>2016</b>	<b>PORTS #1, 4, 7</b>	1000	23.6	3.18	338	582	618	83	0
<b>2688</b>	<b>PORTS #1, 4, 7</b>	1000	21.1	2.97	351	595	659	93	0
<b>3384</b>	<b>PORTS #1, 4, 7</b>	1000	24.7	3.09	333	577	875	108	0
<b>5112</b>	<b>PORTS #1, 4, 7</b>	1000	23.3	3.03	317	561	1203	173	0

Table 5a - Porewater quality results for partially weathered and oxidized tailings underwater (Aquarium #2), at shallow, near surface, depths.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Al (mg/l)</b>	<b>Ca (mg/l)</b>	<b>Ce (mg/l)</b>	<b>Cu (mg/l)</b>	<b>Fe Total (mg/l)</b>	<b>Mg (mg/l)</b>	<b>Mn (mg/l)</b>	<b>Ni (mg/l)</b>
<b>Pretrial</b>	<b>PORTS #1, 4, 7</b>	35.30	270	0.83	0.06	117.80	4.90	0.310	0.17
<b>0</b>	<b>PORTS #1, 4, 7</b>	10.06	86	0.23	<0.04	30.04	1.60	0.100	0.06
<b>168</b>	<b>PORTS #1, 4, 7</b>	5.55	53	0.12	<0.04	17.93	0.98	0.060	0.12
<b>336</b>	<b>PORTS #1, 4, 7</b>	3.40	39	0.09	<0.04	12.24	0.72	0.043	0.03
<b>504</b>	<b>PORTS #1, 4, 7</b>	2.40	30	<0.08	<0.04	8.50	0.53	0.028	0.03
<b>672</b>	<b>PORTS #1, 4, 7</b>	1.72	21	<0.08	<0.04	5.81	0.37	0.020	0.38
<b>1008</b>	<b>PORTS #1, 4, 7</b>	2.20	32	<0.08	<0.04	5.80	0.39	0.010	<0.02
<b>1344</b>	<b>PORTS #1, 4, 7</b>	2.40	37	0.08	<0.04	5.40	0.39	0.016	0.04
<b>2016</b>	<b>PORTS #1, 4, 7</b>	3.80	75	0.13	<0.04	7.80	0.66	0.036	0.05
<b>2688</b>	<b>PORTS #1, 4, 7</b>	5.10	88	0.15	<0.04	7.70	0.76	0.035	0.03
<b>3384</b>	<b>PORTS #1, 4, 7</b>	5.90	99	0.20	<0.04	8.80	0.87	0.041	0.06
<b>5112</b>	<b>PORTS #1, 4, 7</b>	10.30	152	0.35	<0.04	14.18	1.30	0.060	0.03

Table 5a - Porewater quality results for partially weathered and oxidized tailings underwater (Aquarium #2), at shallow, near surface, depths.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Pb (mg/l)</b>	<b>Th (mg/l)</b>	<b>U (mg/l)</b>	<b>Zn (mg/l)</b>	<b>Sulphate (mg/l)</b>	<b>Ra-226 mBq/l</b>
<b>Pretrial</b>	<b>PORTS #1, 4, 7</b>	1.70	<0.05	<0.2	0.260	1186	6852
<b>0</b>	<b>PORTS #1, 4, 7</b>	3.70	<0.05	<0.2	0.093	371	7897
<b>168</b>	<b>PORTS #1, 4, 7</b>	4.90	<0.05	<0.2	0.050	236	10232
<b>336</b>	<b>PORTS #1, 4, 7</b>	6.00	<0.05	<0.2	0.045	173	10707
<b>504</b>	<b>PORTS #1, 4, 7</b>	6.70	<0.05	<0.2	0.030	135	11172
<b>672</b>	<b>PORTS #1, 4, 7</b>	7.00	<0.05	<0.2	0.030	105	14204
<b>1008</b>	<b>PORTS #1, 4, 7</b>	6.00	<0.05	<0.2	0.030	132	11601
<b>1344</b>	<b>PORTS #1, 4, 7</b>	5.60	<0.05	<0.2	0.030	144	10787
<b>2016</b>	<b>PORTS #1, 4, 7</b>	3.70	<0.05	<0.2	0.060	257	11508
<b>2688</b>	<b>PORTS #1, 4, 7</b>	2.60	<0.05	<0.2	0.057	294	10157
<b>3384</b>	<b>PORTS #1, 4, 7</b>	2.80	<0.05	<0.2	0.060	335	9848
<b>5112</b>	<b>PORTS #1, 4, 7</b>	2.30	0.10	<0.2	0.070	530	9857

Table 5b - Porewater quality results for partially weathered and oxidized tailings underwater (Aquarium #2), at intermediate depths.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Volume (ml)</b>	<b>Temp °C</b>	<b>pH</b>	<b>Eh (mV)</b>	<b>Eh(NHE)</b>	<b>Ec (µS/cm)</b>	<b>Acidity (mg/l) CaCO<sub>3</sub></b>	<b>Alkalinity (mg/l) CaCO<sub>3</sub></b>
<b>Pretrial</b>	<b>PORTS #2, 5, 8</b>	1000	20.2	2.99	286	530	2590	1357	0
<b>0</b>	<b>PORTS #2, 5, 8</b>	1000	24.3	2.99	274	518	3180	1031	0
<b>168</b>	<b>PORTS #2, 5, 8</b>	1000	22.2	3.01	278	522	2640	924	0
<b>336</b>	<b>PORTS #2, 5, 8</b>	1000	22.1	3.04	275	519	2500	822	0
<b>504</b>	<b>PORTS #2, 5, 8</b>	1000	22.1	3.11	278	522	2390	767	0
<b>672</b>	<b>PORTS #2, 5, 8</b>	1000	19.7	2.99	275	519	2450	571	0
<b>1008</b>	<b>PORTS #2, 5, 8</b>	1000	21.3	3.16	268	512	2220	490	0
<b>1344</b>	<b>PORTS #2, 5, 8</b>	1000	20.3	3.16	273	517	2070	428	0
<b>2016</b>	<b>PORTS #2, 5, 8</b>	1000	22.8	3.12	273	517	2170	428	0
<b>2688</b>	<b>PORTS #2, 5, 8</b>	1000	21.9	3.04	287	531	2000	411	0
<b>3384</b>	<b>PORTS #2, 5, 8</b>	1000	23.5	3.14	258	502	2460	420	0
<b>5112</b>	<b>PORTS #2, 5, 8</b>	1000	22.8	3.12	258	502	2160	515	0

Table 5b - Porewater quality results for partially weathered and oxidized tailings underwater (Aquarium #2), at intermediate depths.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Al (mg/l)</b>	<b>Ca (mg/l)</b>	<b>Ce (mg/l)</b>	<b>Cu (mg/l)</b>	<b>Fe Total (mg/l)</b>	<b>Mg (mg/l)</b>	<b>Mn (mg/l)</b>	<b>Ni (mg/l)</b>
<b>Pretrial</b>	<b>PORTS #2, 5, 8</b>	91.20	507	1.50	<0.04	406	11.64	0.820	0.47
<b>0</b>	<b>PORTS #2, 5, 8</b>	69.99	521	1.30	0.11	296	9.30	0.640	0.35
<b>168</b>	<b>PORTS #2, 5, 8</b>	63.69	530	1.20	0.05	255	8.40	0.560	0.31
<b>336</b>	<b>PORTS #2, 5, 8</b>	52.50	538	1.20	<0.04	210	7.40	0.510	0.29
<b>504</b>	<b>PORTS #2, 5, 8</b>	46.88	548	1.13	<0.04	184	6.30	0.430	0.24
<b>672</b>	<b>PORTS #2, 5, 8</b>	34.88	553	0.99	<0.04	136	5.20	0.360	0.20
<b>1008</b>	<b>PORTS #2, 5, 8</b>	31.95	549	0.94	<0.04	124	4.40	0.290	0.15
<b>1344</b>	<b>PORTS #2, 5, 8</b>	26.67	552	0.85	<0.04	106	3.90	0.260	0.15
<b>2016</b>	<b>PORTS #2, 5, 8</b>	24.40	586	0.82	<0.04	105	4.00	0.280	0.17
<b>2688</b>	<b>PORTS #2, 5, 8</b>	23.11	585	0.80	<0.04	93	3.70	0.260	0.12
<b>3384</b>	<b>PORTS #2, 5, 8</b>	22.94	585	0.80	<0.04	87	3.70	0.260	0.12
<b>5112</b>	<b>PORTS #2, 5, 8</b>	25.47	563	0.98	<0.04	92	4.03	0.260	0.13

Table 5b - Porewater quality results for partially weathered and oxidized tailings underwater (Aquarium #2), at intermediate depths.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Pb (mg/l)</b>	<b>Th (mg/l)</b>	<b>U (mg/l)</b>	<b>Zn (mg/l)</b>	<b>Sulphate (mg/l)</b>	<b>Ra-226 mBq/l</b>
<b>Pretrial</b>	<b>PORTS #2, 5, 8</b>	1.40	0.10	<0.2	0.650	2648	2975
<b>0</b>	<b>PORTS #2, 5, 8</b>	1.50	0.06	<0.2	0.461	2348	3290
<b>168</b>	<b>PORTS #2, 5, 8</b>	1.50	<0.05	<0.2	0.390	2252	3540
<b>336</b>	<b>PORTS #2, 5, 8</b>	1.70	0.07	<0.2	0.340	2111	3981
<b>504</b>	<b>PORTS #2, 5, 8</b>	1.60	0.06	<0.2	0.290	1992	3265
<b>672</b>	<b>PORTS #2, 5, 8</b>	1.70	<0.05	<0.2	0.230	1911	3309
<b>1008</b>	<b>PORTS #2, 5, 8</b>	1.70	<0.05	<0.2	0.190	1821	3022
<b>1344</b>	<b>PORTS #2, 5, 8</b>	1.80	<0.05	<0.2	0.170	1761	2993
<b>2016</b>	<b>PORTS #2, 5, 8</b>	2.10	<0.05	<0.2	0.210	1788	4093
<b>2688</b>	<b>PORTS #2, 5, 8</b>	1.80	<0.05	<0.2	0.190	1752	3543
<b>3384</b>	<b>PORTS #2, 5, 8</b>	2.00	<0.05	<0.2	0.190	1779	4573
<b>5112</b>	<b>PORTS #2, 5, 8</b>	1.70	0.06	<0.2	0.200	1866	5323



Table 5c - Porewater quality results for partially weathered and oxidized tailings underwater (Aquarium #2), at depths near tailings bottom.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Volume (ml)</b>	<b>Temp °C</b>	<b>pH</b>	<b>Eh (mV)</b>	<b>Eh(NHE)</b>	<b>Ec (µS/cm)</b>	<b>Acidity (mg/l) CaCO<sub>3</sub></b>	<b>Alkalinity (mg/l) CaCO<sub>3</sub></b>
<b>Pretrial</b>	<b>PORTS #3, 6, 9</b>	1000	20.1	2.94	286	530	2840	1819	0
<b>0</b>	<b>PORTS #3, 6, 9</b>	1000	24.4	2.96	259	503	3720	1726	0
<b>168</b>	<b>PORTS #3, 6, 9</b>	1000	21.2	2.96	272	516	3160	1673	0
<b>336</b>	<b>PORTS #3, 6, 9</b>	1000	22.1	2.97	273	517	3100	1672	0
<b>504</b>	<b>PORTS #3, 6, 9</b>	1000	22.2	3.01	274	518	3020	1584	0
<b>672</b>	<b>PORTS #3, 6, 9</b>	1000	18.6	2.87	272	516	3110	1530	0
<b>1008</b>	<b>PORTS #3, 6, 9</b>	1000	21.5	3.03	276	520	2890	1459	0
<b>1344</b>	<b>PORTS #3, 6, 9</b>	1000	20.7	3.00	276	520	2660	1355	0
<b>2016</b>	<b>PORTS #3, 6, 9</b>	1000	23.7	3.04	279	523	2700	1307	0
<b>2688</b>	<b>PORTS #3, 6, 9</b>	1000	21.6	2.96	292	536	2410	1147	0
<b>3384</b>	<b>PORTS #3, 6, 9</b>	1000	22.4	3.06	274	518	2860	1061	0
<b>5112</b>	<b>PORTS #3, 6, 9</b>	1000	23.7	3.19	286	530	2360	916	0

Table 5c - Porewater quality results for partially weathered and oxidized tailings underwater (Aquarium #2), at depths near tailings bottom.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Al (mg/l)</b>	<b>Ca (mg/l)</b>	<b>Ce (mg/l)</b>	<b>Cu (mg/l)</b>	<b>Fe Total (mg/l)</b>	<b>Mg (mg/l)</b>	<b>Mn (mg/l)</b>	<b>Ni (mg/l)</b>
<b>Pretrial</b>	<b>PORTS #3, 6, 9</b>	111.90	493	1.50	0.12	577	14.57	1.060	0.62
<b>0</b>	<b>PORTS #3, 6, 9</b>	111.20	498	1.60	0.09	550	14.34	1.040	0.61
<b>168</b>	<b>PORTS #3, 6, 9</b>	105.70	505	1.50	0.11	521	14.05	1.040	0.62
<b>336</b>	<b>PORTS #3, 6, 9</b>	110.00	509	1.50	0.06	530	13.85	1.020	0.59
<b>504</b>	<b>PORTS #3, 6, 9</b>	103.40	513	1.50	0.07	509	13.66	1.010	0.56
<b>672</b>	<b>PORTS #3, 6, 9</b>	100.60	525	1.40	<0.04	486	13.32	1.010	0.56
<b>1008</b>	<b>PORTS #3, 6, 9</b>	93.72	512	1.50	<0.04	454	12.55	0.910	0.49
<b>1344</b>	<b>PORTS #3, 6, 9</b>	86.09	514	1.40	<0.04	417	11.99	0.910	0.50
<b>2016</b>	<b>PORTS #3, 6, 9</b>	76.56	529	1.44	0.07	359	10.54	0.750	0.40
<b>2688</b>	<b>PORTS #3, 6, 9</b>	67.89	543	1.30	<0.04	309	9.80	0.720	0.37
<b>3384</b>	<b>PORTS #3, 6, 9</b>	62.17	568	1.30	<0.04	270	9.55	0.710	0.36
<b>5112</b>	<b>PORTS #3, 6, 9</b>	54.38	558	1.20	<0.04	202	7.70	0.560	0.27

Table 5c - Porewater quality results for partially weathered and oxidized tailings underwater (Aquarium #2), at depths near tailings bottom.

<b>TIME (t) From Start (Hr)</b>	<b>SITE</b>	<b>Pb (mg/l)</b>	<b>Th (mg/l)</b>	<b>U (mg/l)</b>	<b>Zn (mg/l)</b>	<b>Sulphate (mg/l)</b>	<b>Ra-226 mBq/l</b>
<b>Pretrial</b>	<b>PORTS #3, 6, 9</b>	1.48	0.12	<0.2	0.830	3055	1795
<b>0</b>	<b>PORTS #3, 6, 9</b>	1.50	0.10	<0.2	0.750	3055	2135
<b>168</b>	<b>PORTS #3, 6, 9</b>	1.50	0.11	<0.2	0.730	2971	2097
<b>336</b>	<b>PORTS #3, 6, 9</b>	1.60	0.10	<0.2	0.710	2929	2140
<b>504</b>	<b>PORTS #3, 6, 9</b>	1.60	0.10	<0.2	0.710	2872	1996
<b>672</b>	<b>PORTS #3, 6, 9</b>	1.70	0.11	<0.2	0.670	2803	2001
<b>1008</b>	<b>PORTS #3, 6, 9</b>	1.60	0.07	<0.2	0.620	2710	2230
<b>1344</b>	<b>PORTS #3, 6, 9</b>	1.70	0.05	<0.2	0.600	2612	1777
<b>2016</b>	<b>PORTS #3, 6, 9</b>	1.60	0.09	<0.2	0.540	2513	2518
<b>2688</b>	<b>PORTS #3, 6, 9</b>	1.60	0.08	<0.2	0.500	2324	2314
<b>3384</b>	<b>PORTS #3, 6, 9</b>	1.99	0.08	<0.2	0.480	2312	2676
<b>5112</b>	<b>PORTS #3, 6, 9</b>	1.80	0.09	<0.2	0.400	2234	2908

Table 6 - Non-linear regression analysis results for acidity, Ca, Fe, Mg, Pb, SO<sub>4</sub><sup>-2</sup>, and Ra-226 in the surface water column of partially oxidized and weathered tailings underwater. Computed values are given for porewater concentration (C<sub>o</sub>), transfer rate constant ( $\frac{K}{h}$ ), transfer coefficient (K), initial flux (J<sub>o</sub>) at time t = 0 and bulk molecular diffusion coefficient in water (D<sub>o</sub>). For Ra-226, the parametric values are given in mBq.

Parameter	Computed porewater concentration C <sub>o</sub> , mg/l	Transfer rate constant $\frac{K}{h}$ , h <sup>-1</sup>	Transfer coefficient K, m/month	Initial flux (J <sub>o</sub> ), at t = 0, mg/m <sup>2</sup> /h	Bulk molecular diffusion coefficient in water (D <sub>o</sub> ), m <sup>2</sup> /s
Acidity (CaCO <sub>3</sub> equivalent)	1873	8.15 x 10 <sup>-6</sup>	1.11 x 10 <sup>-3</sup>	2.90	3.07 x 10 <sup>-10</sup>
Ca	553	3.20 x 10 <sup>-6</sup>	4.38 x 10 <sup>-4</sup>	3.36 x 10 <sup>-1</sup>	1.20 x 10 <sup>-10</sup>
Fe	561	1.27 x 10 <sup>-7</sup>	1.73 x 10 <sup>-5</sup>	1.35 x 10 <sup>-2</sup>	4.76 x 10 <sup>-12</sup>
Mg	15.0	1.88 x 10 <sup>-7</sup>	2.57 x 10 <sup>-5</sup>	5.35 x 10 <sup>-4</sup>	7.04 x 10 <sup>-12</sup>
Pb	2.5	8.81 x 10 <sup>-4</sup>	1.21 x 10 <sup>-1</sup>	4.13 x 10 <sup>-1</sup>	3.31 x 10 <sup>-8</sup>
SO <sub>4</sub> <sup>-2</sup>	3251	6.02 x 10 <sup>-6</sup>	8.24 x 10 <sup>-4</sup>	3.72	2.42 x 10 <sup>-10</sup>
Ra-226 (mBq)	21150	1.41 x 10 <sup>-2</sup>	1.92	5.68 x 10 <sup>+4</sup>	5.30 x 10 <sup>-7</sup>