STUDY OF MERCURY MOBILIZATION FROM RESUSPENDED DREDGE SPOILS IN HOWE SOUND

by

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A report prepared under the terms of
DSS contract 08SS.KF832-6-1065

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ABSTRACT

The rate of release of mercury from suspended Howe Sound sediments under wide ranges of ionic strength and pH was studied. The effect of temperature, poison and storage time was also examined.

In all cases a period of rapid mercury release occurred, usually reaching a maximum value within several hours of sediment addition, followed by a slow decline of mercury levels to an equilibrim value after several days. The maximum mercury released in no case exceeded 30% of the total sediment mercury. Temperature, storage and biological effects were small. Salinity and pH had much greater effects; the maximum mercury release occurred for low pH and high salinity. The flux of mercury across the sediment-estuarine water interface under aerobic conditions was in the order of 100 ng/cm^2/day.

The variation of disolved, particulate and interstitial mercury over a tidal cycle in the tidal flat area of the Squamish River Estuary suggest that the tidal oscillation is a major mechanism for mobilization of mercury from sediments into overlying Howe Sound waters.

TABLE OF CONTENTS

Page

APPENDIX A

APPENDIX B

LIST OF FIGURES

PREFACE

This report summarizes a study performed between December 6, 1976 and March 18, 1977 under DSS contract 08SS.KF832-6-1065. It is divided into two parts. PART A deals with

(1) a laboratory investigation of the rate of release of mercury from resuspended Howe Sound dredge spoils in relation to temperature, salinity, pH, length of water storage time and biological effects; and

(2) a laboratory investigation of the flux of mercury across the sediment water interface under aerobic conditions.

PART B deals with a field study conducted December 1 to December 2, 1976 on the Squamish River tidal flats and concerned with the measurement of dissolved,particulate and interstitial mercury over a tidal cycle.

Because of the large amount of data generated in this study, only a very general treatment of the data was possible. Note also that all mercury values reported represent the sum of inorganic and methyl mercury fractions.

1. INTRODUCTION

Numerous laboratory experiments, for example those of Lee et al. and Lindberg et al., studying the release of potentially hazardous or undesirable components from resuspended dredge spoils have been conducted in recent years as a response to the anticipated large-scale ocean dumping of contaminated dredge spoils. In most cases, only qualitative results were obtained because insufficient care was exercised in experimental design. The common usage of small volume containers for reaction vessels in these studies has at least two drawbacks. Firstly, a large surface area to volume ratio increases the importance of wall effects especially at trace and ultra trace concentrations. Secondly, comparison of results from a large number of containers in time studies greatly increases the data scatter because of variations in the properties of the containers themselves. In this study, an attempt was made to minimize container effects by using specially cleaned 50-ℓ glass vessels. Such large containers also permitted serial sampling for a given experimental condition in a single vessel.

An important factor in release experiments, seldom studied yet frequently used to excuse inconsistent results, is the presence of biological communities. There are two main reasons for anticipating that this effect is important especially in the case of mercury:

(1) certain bacteria can reduce Hg(II) to Hg(0) which can then be lost through volatization. Baier et al. (1975) found that, at the sub-ppb level of dissolved mercury and at natural water pH, over 80% of the mercury in solution may be lost within days due to this effect; and

(2) the suspension of dredge spoil in water results in the release of nutrients which encourages the proliferation of bacteria.

An estimate of the magnitude of biological effects was made in this study by comparing the Hg release rate for a biologically active and biologically inactive case. The biologically inactive case was created by the use of $CuSO_4$ as a poison. Copper sulphate was chosen over other biocides because it was effective, safe and supposedly free from undesirable properties such as surface activity and mercury complexing ability.

As in any research situation there were several unexpected developments. The most severe of these was an air contamination problem. Ambient levels of mercury in the air of our laboratory increased from < 1 ng/ℓ to in excess of 10^6 ng/ℓ. The sewer system was identified as the source of the contamination. This was verified by measuring high levels of airborne mercury in buildings in the same vicinity of the laboratory. The problem was eventually solved but unfortunately samples for the static release experiment (anaerobic case) were contaminated and the experiment was therefore lost.

2. EXPERIMENTAL MATERIALS AND METHODS

2.1 Materials Used

2.1.1 Water

For the purposes of this study the salinity definitions for freshwater, estuarine water and seawater were <10°/oo, 15-25°/oo and > 28°/oo respectively. The water was collected at two different times (1) November 29 - December 2, 1976 (Cruise OC-76-IS-008) and (2) February 14, 1977 (Cruise OC-77-IS-002) at several stations in Howe Sound (for exact positions see appendix A.1)

Polyethylene carboys (220 l) with lids covered and sealed with double thickness 4 mil thick polyethylene sheeting, were used to store the water. Prior to use, each carboy was cleaned by leaching with a 0.02°/o (w/v) freshwater solution of $K_2Cr_2O_7$ in 1 N ACS grade HNO_3 for several days. Before filling with sample water, each carboy was pumped empty using a Jabsco in-line pump, rinsed twice with about 20 liters of sample water then pumped full through acid-cleaned polyethylene tubing during which time the salinity was monitored using a hand-held refractometer/salinometer. All manipulations were accomplished using plastic gloves.

2.1.2 Sediment Samples

The sediment used in the experiments was collected at a series of locations on the tidal flats in the vicinity of the FMC plant at Squamish November 18 and 19, 1976 by pounding an acrylic tube into the sediment and withdrawing it. The mercury contents of 2 cm subsamples of these cores were provided by Chemex Labs Ltd., North Vancouver, B.C. These subsamples were, in turn, subsampled after being homogenized by rolling and then used wet in the experiments.

2.1.3 Box Cores

Box cores used in the static release experiments were collected during cruise OC-76-IS-008 using a Bruland-Soutar box corer and kept frozen until used.

2.1.4 Chemicals

Sodium hydroxide was purified by adding $SnCl_2$ (5% by weight) and purging with nitrogen. Reagent grade copper sulphate and mercury-determination-grade nitric acid were used without further purification.

2.2 Cleaning Procedures

Glassware:

New 500 ml glass-stoppered pyrex bottles were cleaned by the hot oxidation method described by Bothner (1974; also see 3.1 below). As many as three oxidations were necessary before new bottles provided an acceptable blank value. Fifty-litre pyrex containers used as reaction vessels for the release experiments were cleaned prior to initial use by filling with a 0.1% (w/v) solution of $K_2Cr_2O_7$ in 1 N ACS grade HNO_3 for three days then emptying and rinsing at least three times with forty liters of tapwater. Between experiments these containers were cleaned as above but only for one day. Other glassware and TFE teflon stirring rods were cleaned by heating (at 70°C for 12 h in ACS grade nitric acid (3 N) then rinsed three times with cold tap water and stored double-bagged in polyethylene until used.

Other Equipment:

All TFE teflon and polyethylene tubing, teflon pumps and peristaltic pump tubing was cleaned by recirculating concentrated ACS grade nitric acid for 1 h, then cold tap water five times for 15 minutes or until the recirculating wash water was no longer acidic.

Nuclepore filters were cleaned prior to use by leaching in a 0.01% (w/v) $K_2Cr_2O_7$ solution in 1 N ACS grade nitric acid overnight then rinsing three times with tap water before use.

Plexiglass containers and lids were cleaned by soaking in 1 N ACS grade nitric acid at room temperature for several hours before rinsing with tap water.

2.3 Experimental Procedures

2.3.1 General Description of Laboratory Experiments

A series of nine experiments was conducted to measure the release rate of mercury from suspended sediment in water. In addition, an experiment was conducted to measure the flux of mercury across the sediment-water interface. The following is a description of these experiments.

Experiment 1:

Measurements to determine experimental precision.

Experiment 2:

A five-day experiment to measure the release rate of mercruy from mercury-rich sediment into freshwater, seawater and estuarine water, $CuSO_4$ poisoned estuarine water and the variation of dissolved mercury in estuarine water at pH 4 and pH 7.7.

Experiment 3:

A five-day experiment - repeat of experiment 2 with blanks of seawater and estuarine water at natural pH. Combined with experiment 2, this experiment provides an estimation of the effects of water storage on the rate of mercury release.

Experiment 4:

A one-day experiment to measure both dissolved and particulate phase mercury to determine whether or not the total mercury concentration changes with time.

Experiment 5: Effect of Poison

A five-day experiment to determine the effect of $CuSO_4$ (as a biocide) on the rate of mercury release from mercury-rich and Hg-poor sediment at natural pH in estuarine water. Two concentrations of $CuSO_4$ were used - 0.2 g and 2.0 g per 30 liters solution.

Experiment 6: Effect of pH

A five-day experiment measuring the rate of mercury release from mercury-rich sediment into freshwater at pH 4,6,7,8 and 9.

Experiment 7:

Continuation of experiment 6 using seawater at pH 4,6,7 and 9 and estuarine water at pH 4,6 and 9.

Experiment 8: Effect of pH

A five-day experiment similar to experiments 6 and 7 but using Hg-poor sediment in the place of mercury-rich sediment. Due to a lack of high salinity water only the freshwater and estuarine water cases were examined.

Experiment 9:

A 15-day experiment to measure the release rate of mercury into fresh, estuarine and sea waters from Hg-poor sediment at natural pH for poisoned an unpoisoned cases.

Static Release Experiment:

A thirty-day experiment to measure the migration of mercury across the sediment-estuarine water interface and within the sediment for aerobic and anaerobic conditions using a sediment of known organic carbon content, moisture content and mineralogy.

Due to contamination problems the results of the anaerobic case were lost.

A separate run was not made for the effect of temperature since a combination of experiments 2, and 7 provide the rate of mercury release from sediment at ~ 17°C and ~ 6°C

2.3.2 Experimental Apparatus

2.3.2.1 Release Experiments

The release experiments were conducted in a series of 50-ℓ rectangular Pyrex glass containers, as many as twelve being used at any one time. Each container was covered by a fitted 3/8" plexiglass lid containing two holes - one for a Teflon stirring rod and a second for a pH electrode and sampling tube. Black and Decker ¼" drills (Model # 5718) mounted above each container by means of a lab rack and wired in series with dimmer switches provided variable, easily controlled stirring. The chance of drill-generated contamination was reduced by shielding the drills with 6 mil polyethylene sheeting. The entire configuration was enclosed in a wooden box with hinged sides providing easy access to the containers at sampling times. Mechanical noise was reduced by lining the inside surface of the box with ½" foam while radio frequency electrical noise was reduced by grounding the drills to chicken wire which had been stapled over the foam.

2.3.2.2 Static Release Experiment

The static release experiments were conducted in plexiglass containers as shown in Figure 1 . The container dimensions were designed to exactly fit a 20 cm thick Bruland Soutar box core covered by approximately 10 liters of water. A gas inlet tube (A) allows the maintenance of a nitrogen atmosphere by the introduction of nitrogen gas. A slurry of $HgCl_2$ is introduced through the injection port (B).

2.3.3 Experimental Procedure and Sampling Technique

2.3.3.1 Release Experiments

Each cleaned container was filled with water of the desired salinity by pumping it directly from the storage carboys through 3/8" diameter polyethylene tubing using a Saturn teflon pump (Fluorocarbon Company). No attempt was made to resuspend the settled sediment because of the difficulty in producing an homogeneous suspension. Concentrated mercury-free nitric acid or 10 N mercury-free sodium hydroxide was then added to adjust the pH if necessary. After twelve hours of stirring, samples were drawn for dissolved mercury analysis (reported as "background" value) and salinity. Temperature and pH were measured in situ. A homogenized pre-weighed amount of wet sediment was then added

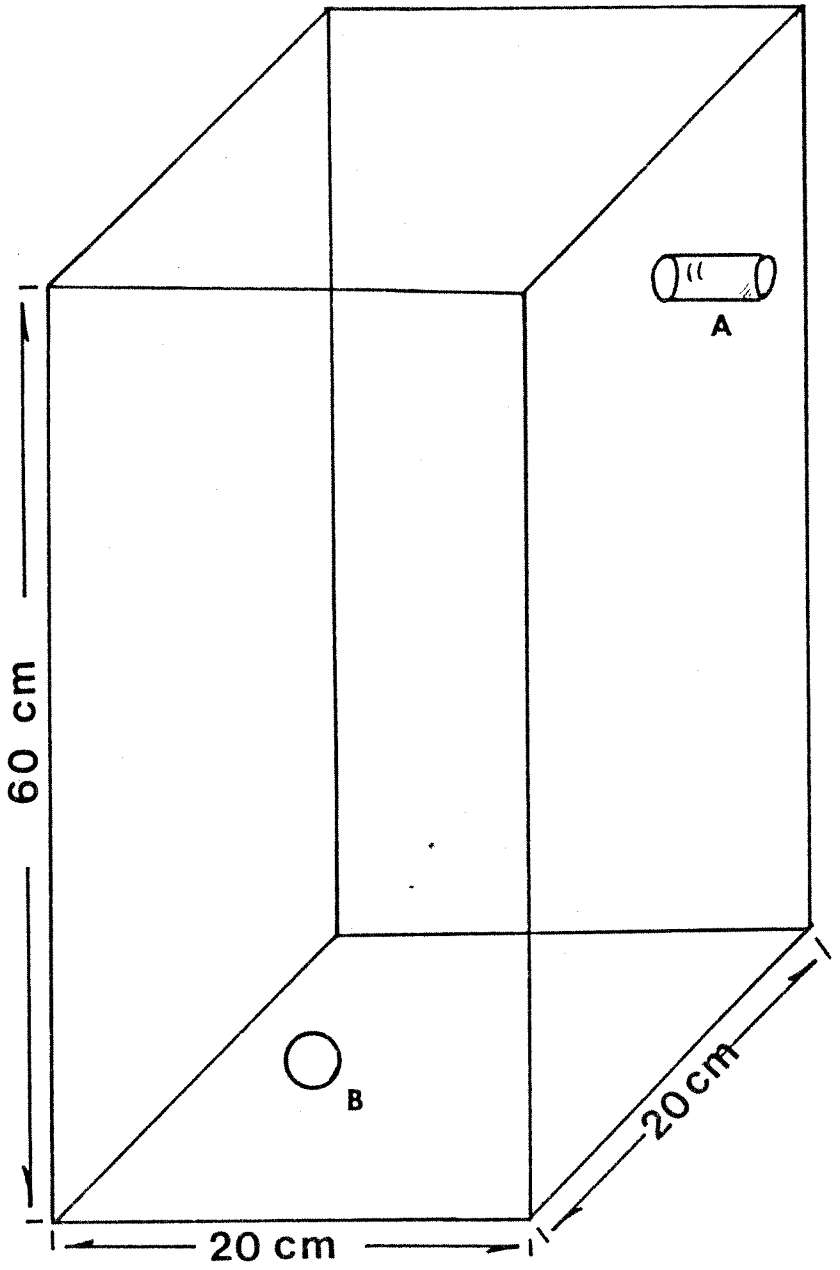

FIGURE 1. Plexiglass container used in Static Release Experiment

to each container and a sample drawn for particle size analysis and dissolved mercury. This sampling period was designated t = 0h. Routinely, the temperature and pH were measured in situ and the pH adjusted to the t = 0 value, if necessary, before drawing samples for dissolved mercury at t = 1h, 2h, 4h, 8h, 12h, 1d, 2d, 3d, 4d and 5d although these sampling times were modified on occasion to accommodate experimental problems.

At each sampling period, the technique for drawing the dissolved mercury sample was as follows: the teflon-weighted end of 1/8" i.d. teflon tubing was inserted into each container, in turn, to a position exactly six inches from the bottom and sample withdrawn by pumping through a Masterflex peristaltic pump (Horizon Ecology Company). Approximately six tubing dead volumes (150 cm^3) were discarded ensuring that it was free of traces of the previous sample. Next, 50 ml were pumped into an all-glass Millipore filtration apparatus and filtered through a 0.4 μm Nuclepore filter under reduced pressure into a 1 liter glass filter flask. This 50 ml wash was also discarded. The next 500 ml was filtered and transferred to a 500 ml ground glass stoppered sample bottle. Ten milliliters of a 0.5% (w/v) $K_2Cr_2O_7/HNO_3$ solution was then added to the filtrate to prevent adsorption of mercury onto the container walls and the filter stored in a Petri plate. The rate of filtration was variable. In some cases the rate was so slow that less than 500 ml had to be sampled in order to remain on the sampling schedule. The filtration apparatus was washed with 1 N HNO_3 and tap water between samples and with 0.5% (w/v) $K_2Cr_2O_7/HNO_3$ solution between experiments.

Beginning with experiment three this sampling technique was modified to reduce possible contamination introduced by excessive handling. Accordingly, the 1 liter filter flask was replaced by a 500 ml sample bottle fitted with an adapter that reduced the Millipore holder to a 24/29 ground glass joint. To overcome the problem of losing mercury quickly to the glass walls of the sample bottle, the 10 ml of 0.5% $K_2Cr_2O_7/HNO_3$ was added prior to the time of filtration.

At all sampling times it was assumed that the sediment was evenly distributed throughout the liquid phase. (No water was added to the containers to replace that pumped out during sampling). The validity of this assumption was tested by measuring the weight of particulates filtered from ten 500 ml samples. Turbulent mixing was provided by 18" glass propellors rotating at two speeds - one fast enough to keep all sediment in suspension, the second fast enough to keep only part of the sediment in suspension. Thirty liters of water and 0.600 g of sediment were used. Hence, if homogeneously dispersed, each 500 ml sample should contain 10 mg suspended solids. The results were as follows:

mg suspended solids per 500 ml

LOW SPEED (some sediment visible at bottom of container)	HIGH SPEED (all sediment in suspension)
3.25	9.87
1.80	10.12
1.17	9.93
2.40	9.98
0.99	10.01
2.11	9.59
2.02	9.90
0.83	9.77
1.48	10.25
2.94	9.96

The particle size distribution was also measured for each of the above ten samples (see Appendix A.3). Only for high speed stirring were the distributions reproducible. Thus, only for the condition where all sediment is kept in suspension is the above assumption valid.

Unfortunately, the mechanical strength of the glass propellors was insufficient to withstand the strain of prolonged high speed stirring. Accordingly, the glass propellors were replaced by some constructed of 3/4" solid TFE teflon rod into the bottom of which were drilled two holes for the orthogonal placement of two, four-inch ½" solid teflon rods. These propellors were then machined to thread directly onto the rotating shaft of the drill and provided excellent turbulence at low revolutions which decreased wear of the drills substantially. In addition, a series of ten samples of estuarine water were drawn to assess the experimental sampling precision which should give a better estimate to the precision of the dissolved mercury concentrations than the precision obtained by measuring replicate mercury-spiked samples. The ten samples were drawn from the same glass container using the sampling technique began in Experiment 3, as outlined above. Between the drawing of each sample, estuarine water containing in excess of 100 ng/l was pumped through the hosing to test the efficiency of 150 cm^3 of wash water in eliminating large scale mercury fluctuations. The results were as follows:

Sample	Dissolved Mercury (ng/l)
1	45.92
2	37.34
3	44.10
4	38.00
5	48.56
6	38.66
7	39.32
8	37.34
9	36.04
10	38.28
	$\bar{x}$ = 40.36

Thus, the relative standard deviation at the 40 ng/l level is 10.01% or roughly four times the r.s.d. found for replicate analyses of spiked samples.

2.3.3.2 Static Release Experiment

Two approximately 20 cm long frozen box core sections were placed in two plexiglass containers (described above) and covered with ten liters of estuarine water (salinity $\cong 16.6^{o}/oo$). Anaerobic conditions were encouraged in one container by maintaining a nitrogen atmosphere over the core while aerobic conditions were encouraged in the other by allowing the free movement of air over the core. After two days, background samples were drawn for dissolved mercury determinations and in situ pH values were measured. Then into each sediment, through the injection port 10 cm below the sediment-water interface, 0.23 g of mercury (as $HgCl_2$, 10 ml of a 0.023 gHg/ml solution) was injected using a 10 cm cannula. Sampling was then performed once a day for thirty days in the aerobic experiment while the anaerobic experiment was aborted after 15 days because the samples were lost due to a severe airborne mercury contamination problem (the experiments were not run concurrently). At each sampling period, 1000 ml of water were removed (500 ml for a sample and 500 ml for washing the tubing) and 1000 ml of estuarine water were added to maintain the 10 liter volume above the sediment. Control of the amount of water removed and added was critical because the amount of mercury in solution at any time had to be corrected for the fact that mercury rich water was being replaced by mercury poor water (estuarine) at each sampling period.

At the completion of aerobic experiment, nine 20 cm x 2 cm cores were taken in a symmetrical pattern around the original point of injection by pushing acrylic tubes into the sediment, freezing the sediment in a chest freezer then withdrawing the acrylic tubes containing the frozen cores. The cores were then extruded (frozen) and, after thawing, sampled at several depths. These sediment samples were then analyzed for mercury in order to obtain an impression of the three dimensional migration of mercury around the original point of injection.

3. ANALYTICAL PROCEDURES

3.1 Mercury in Seawater by Flameless Atomic Absorption

A modified version of the wet chemical flameless atomic absorption technique developed by Bothner (1974) was used to determine total mercury in seawater. The sample is first strongly oxidized, then the mercury is reduced to the elemental state by reduction with stannous chloride and is purged from the sample with a stream of nitrogen gas. Water vapour is removed by passing the nitrogen carrier gas through a magnesium perchlorate drying tube. The dry gas is passed through a U.V. monitor with a 30 cm path length cell, and the absorption is measured at 253.7 nm. The absorbance is proportional to the mercury concentration in the sample.

The Bothner (1974) method was modified to increase analysis rate and precision by increasing the sample volume from 100 to 250 ml, oxidizing the sample directly in the collection flask, decreasing the purge gas flow rate, and damping the recorder signal to reduce instrument noise. By using a 47 µF capacitor across the recorder input, the instrument signal to noise ratio at the 20 ng/l level was ~ 50, compared to ~ 10 without dampling. Use of larger sample volumes (250 ml) gave a flat-top peak for 20-40 seconds as opposed to a fast (2-3 sec) peak for 100 ml sample volumes. The peak response time increased because mercury was purged from a larger volume using a slower (½) rate of nitrogen gas flow.

After incorporating these changes, the precision at the 20-40 ng/l level increased from ± 20% (2σ) for 100 ml samples to ± 5% (2σ) for 250 ml samples. The detection limit (twice noise) was 0.8 ng/l.

Analysis of a 1.18 ± 0.04 ng/ml (National Bureau of Standards) mercury reference sample using our revised method gave a value of 1.19 ± 0.06 ng/ml.

The recorder sensitivity (or recorder span factor) in ng Hg (free)/mm peak height was determined 2-3 times per day for 4-5 aliquots. Differences in sensitivity between fresh and saline water, and free mercury vs total mercury (Berrang and Erickson, 1976) are within experimental error. Variations in sensitivity were ± 5% over several weeks. At the 20 ng Hg/1 level, the sensitivity was 0.043 ng/nm (at 40 µv/cm recorder and 0.02 U.V. monitor absorbance ranges) with a baseline noise level of 2-3 mm and a long term (several hours) baseline drift of 10-20 mm. For 20 ng/1 the signal/noise was ~ 50. Fluctuations in 60 Hz AC line voltages were apparently reduced during night-time hours and week-ends, with a corresponding reduction in baseline noise of 2-5. Subsequently, a voltage line regulator was used for daytime operation.

Some common problems encountered using this technique were high (oxidizing agent) blanks, sample flask contamination, "bubble-blanks", mercury absorption onto flask walls and AC input line fluctuations. Since specially purified $KMnO_4$ and $K_2S_2O_8$ were not available, ACS reagent grade purity was used to oxidize total merucyr samples. Blanks from 5 ml of 5% (w/v) solutions of $KMnO_4$ and $K_2S_2O_8$ per 250 ml sample were 1.0 ng (i.e. a 4 ng/1 correction). Cold tap water, containing 15 ng Hg/1, was used to clean, rinse, prepare solutions and standards etc., and gave a blank of ~ 0.15 ng (or ~ 10% of the total reagent blank). Whereas the $SnCl_2$ and $NH_2 \cdot OH$ HCl solutions could be purged with N_2 gas to remove mercury, and clean nitric acid purchased, $KMnO_4$ and $K_2S_2O_8$ are not readily purified, except by perhaps subliming $KMnO_4$ and recrystalizing $K_2S_2O_8$. Use of lesser amounts of both $KMnO_4$ and $K_2S_2O_8$ may be feasible, although time did not permit an investigation of this possibility. New sample flasks require very rigorous cleaning (at least two hot oxidizing steps of 2-3 hours each) to eliminate bottle blanks. The "bubble-blank", i.e. the difference in the baseline between sample by-pass and sample purging was 2-3 mm, apparently due to a 25% increase in N_2 line pressure in the by-pass mode. Initially, a very large (~ 50 mm) "bubble-blank" was encountered, and subsequently reduced by oxidizing the polyethylene drying tube and packing the Mg $(ClO_4)_2$.

Since few mercury values analysed during this study were below ~ 30 ng/ℓ, a reagent blank of ~ 4 ng/ℓ and a baseline noise level equivalent to 0.4 ng/ℓ were acceptable.

3.1.1 Procedure

3.1.1.1 Preparation of Mercury Standard Solutions

A 1,000 ppm mercury solution (Fisher Scientific Co. Ltd.) was diluted first to 1 ppm, then to 10 ppb (ng/ml) with a tap water solution* containing 2% (v/v) nitric acid (specially purified for mercury determination)[++] and 0.01% (w/v) potassium dichromate (ACS grade). The 10 ppb working standard was prepared daily.

3.1.1.2 Preparation and Purification of Reagents

Nitric acid/potassium dichromate solution

A solution containing 0.5% (w/v) potassium dichromate (ACS grade) in mercury free nitric acid[++] was added to a 1 liter digi-dispensing pipettor. The potassium dichromate was dissolved in a small volume of tap water prior to the addition of nitric acid.

Stannous chloride/sulphuric acid solution

A 20% (w/v) stannous chloride (ACS grade) solution and a 20% (v/v) sulphuric acid (ACS grade) solution were prepared with tap water and purged with N_2 gas for 4-6 hours to remove mercury. The purified solution should have < 1 ng/ℓ mercury.

Hydroxylamine hydrochloride solution

Five ml of the stannous chloride solution were added to approximately 2 liters of a 12% (w/v) hydroxylamine hydrochloride (ACS grade) solution prepared with tap water. This solution was purged for 4-6 hours with N_2 gas to remove mercury. The mercury blank should be < 1 ng/ℓ.

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* Mercury contamination from this solution represents ~ 0.2% **error** in the 10 ppb standard.

[++] Available from Canadian Laboratories Co. Ltd.

Potassium permangante solution

Two liters of a 5% (w/v) potassium permanganate (ACS grade) solution was prepared with tap water. Aliquots of this solution were measured for mercury blank before additions to samples and discarded if > 0.15 ng/ml solution.

Potassium persulphate solution

Prepared in the same way as the potassium permanganate solution above.

3.1.1.3 Sample Analysis

The 500 ml sample and 10 ml nitric acid/dichromate solution were aliquoted into two 255 ml portions. Five milliliters of persulphate and 5 ml of permanganate solution were added to each aliquot prior to heating at 80°C for two hours in a water bath. Two and one-half milliliters of hydroxylamine hydrochloride solution were added to the cooled samples and the permanganates dissolved with gentle swirling. The air space above the sample solution was purged with N_2 gas for one minute to remove traces of chlorine gas, since chlorine absorbs at 253.7 nm. Just prior to analysis, 10 ml of stannous chloride solution were added, the diffuser inserted, and the sample shaken for 30 seconds, let stand for 30 seconds and purged with N_2 gas at a flow rate of 0.4 l/min for approximately 1 minute and the peak height measured in mm. Peak heights from two 250 ml aliquots were averaged for each sample.

The instrument settings were:

U.V. Monitor (Laboratory Data Control, Riviera Beach, Florida -
30 cm path length cell)
Range - 0.02 Absorbance

Recorder (Fisher Recordall - Series 5000)
Range - 1 mv Full Scale (25 cm)
Chart speed - 2 inches/minute

Nitrogen gas (Grade G) flow rate - 0.4 ℓ/minute.

The system waspurged between samples using tap water. The 6 cm (length) x 2 cm (diameter) polyethylene drying tube was packed with fresh ACS grade magnesium perchlorate after analysis of ~50 aliquots. Glass wool is used at each end of the drying tube to prevent $Mg(ClO_4)$ from entering the U.V. gas cell.

3.1.2 Blanks and Calibration Samples

Total reagent blanks were determined as follows: To a 500 ml flask containing 250 ml tap water were added 5 ml of nitric acid/dichromate, 2.5 ml of hydroxylamine hydrochloride, 5 ml of persulphate, and 5 ml of permanganate solutions. After gentle swirling, 10 ml of stannous chloride solution were added and the mercury purged with N_2 gas. Precision of peak heights should be ± 5-10%. Peak heights should be less than 4 ng/l.

The recorder span factor (ng Hg/mm peak height) was determined by spiking each of 3-5 aliquots of 250 ml of tap water, containing 5 ml nitric acid/dichromate solution, with 5 ng Hg. The precision (2σ) of (free) mercury peaks should be ~ 5%. Standard spiked samples were done prior to every run (~ 9 samples).

3.1.3 Calculation of Results

Mercury concentrations in ng/liter are calculated as follows:

$$C\ (\text{ng/l}) = \left[\text{Average of two sample peak heights (mm)} - \text{Total Reagent Blank (mm)}\right] \cdot \left[\frac{5\cdot(\text{ng Hg spike})}{\text{spiked tap water peak height (mm)} - \text{spike reagent blank (mm)}}\right] \cdot 4$$

3.2 Mercury in Sediments

About 2 g of wet sediment was placed into a teflon decomposition bomb and 15 ml of freshly prepared aqua-regia added. After standing for1 h at room temperature, the solution was brought to boiling for an additional hour. The digest was then cooled and transferred to a 500 ml glass sample bottle and analyzed in duplicate as per the method of section 2.4.1 omitting the oxidation step.

3.3 Mercury in Particulate Phase

The Nuclepore filters and filtered particulate matter were transferred directly into 500 ml sample bottles then 10 ml of 0.5% (w/v) $K_2Cr_2O_7$ in mercury-free nitric acid and 500 ml tap water added. This volume was then digested by the procedure of 3.1 , aliquotted and mercury determined in duplicate.

3.4 Organic Carbon in Sediments

The Walkley-Black titrimetric procedure as outlined in Black et al (1972) was used to determine the organic matter in sediments. Oxidizable matter in the sample is first oxidized by $K_2Cr_2O_7$ and the reaction is facilitated by the heat generated when two volumes of H_2SO_4 are mixed with one of 1N $K_2Cr_2O_7$. The excess $Cr_2O_7^{=}$ is determined by titration with standard $FeSO_4$ solution and the quantity of substances oxidized is calculated from the amount of $Cr_2O_7^{=}$ reduced. Ferroin (o-phenanothroline ferrous sulphate) is used as the indicator. Since Cl^- interferes by producing a positive error, Ag_2SO_4 is added to precipitate out the Cl^-. The oxidizable organic matter determined by this method differs from the total carbon content by a correction factor, f equal to 1.33. Then,

$$\%\ \text{Organic Carbon} = \frac{(\text{meq } K_2Cr_2O_7 - \text{meq } FeSO_4)}{\text{g water-free sediment}} \times 1.33 \times 0.003\ \frac{\text{gC}}{\text{meq C}} \times 100\%$$

The reagent blank was less than 0.01 ml and precision at 2% organic carbon content was ± 5% (2σ).

Reagents

(1) 1 N $K_2Cr_2O_7$:	49.00 g ACS grade $K_2Cr_2O_7$ (pulverized and dried at 105°C for 3-4 h) dissolved in Milli-Q water and made up to 1 liter in a volumetric flask.
(2) H_2SO_4/Ag_2SO_4:	15.0 g Ag_2SO_4 added to 1 liter of H_2SO_4.
(3) 0.5 N $FeSO_4$:	278 g ACS $FeSO_4$-$7H_2O$ dissolved in Milli-Q water. Eighty ml conc H_2SO_4 added solution allowed to cool and made up to one liter.
(4) Indicator:	o-phenanthroline ferrous sulphate - prepared by the method outlined by Kolthoff and Sandell. Stoichiometric amount of o-phenanthroline monohydrate dissolved in a 0.025 M solution of ferrous sulphate in Milli-Q water.

Procedure

The sediment was ground to pass a 0.42 mm nylon sieve. Five ml of 1 N $K_2Cr_2O_7$ were added to 0.1 g of sediment in an Erlenmeyer flask and the soil dispersed into the solution with vigorous swirling. Next, 10 ml of conc. H_2SO_4 were added and, after swirling for about one minute, the flask placed on an asbestos sheet for 30 minutes. The solution was then diluted with about 200 ml Milli-Q water, 3-5 drops of ferroin were added and the solution titrated to a red brown end point with 0.05 N $FeSO_4$. The $FeSO_4$ was standardized daily.

3.5 Dissolved Organic Carbon

The procedure was similar to the one used for sediment except that the concentrations of the oxidizing and reducing agents were decreased by an order of magnitude. The size of sample was 25 ml.

3.6 pH

The pH ($\pm$ 0.05 unit) was measured in situ using a Horizon Ecology Digi Sense Model 5985-40 portable pH meter with combination electrode.

3.7 Salinity

The salinity ($\pm$ 0.1^{o}/oo) was measured with an Environmental Devices Corporation Type 102 Handheld Refractometer/Salinometer.

3.8 Temperature

The temperature ($\pm$ 0.05 C^{o}) was measured using a thermometer (range 0^{o}C to 50^{o}C) calibrated against an NBS thermometer.

3.9 Particle Size Spectra

Particle size distributions were measured using a Coulter Counter Model TA II adjusted to the following instrumental parameters:

Operation Mode:	Concentration Index
Scope Display:	Differential
Auto/Man:	Auto
Print/Plot:	Print
Sampling volume:	2 ml
Active Channels:	2-15
Size Calibration Channels:	10-11
Aperture tube:	200 µm

The instrument was calibrated according to the procedure outlined in the Operator's Manual for Coulter Counter Model TA II using 27.3 μm pollen grains dispersed in ethanol. Samples were run by the method as outlined in the manual.

4. RESULTS AND DISCUSSION

4.1 Release Experiments

The data for the release experiments are tabulated in Appendix A.3. In all the experiments, the relationship between released mercury and time was similar. After the addition of sediment, there was a rapid increase in dissolved mercury, usually within the first hour, followed by a decrease to an apparent equilibrium value after several days. This behaviour is similar to that observed in similar studies by Lindberg et al (1975), Lee et al (1975), and Thomas (1976). Two essential characteristics of the release-time curves (Figures 2 to 29), (a) the maximum mercury released per gram sediment and (b) the equilibrium value of released mercury per gram sediment will be used for comparing the experimental results. In no case was the amount of mercury released greater than 30% of total available in the sediment.

The effect of water storage on the rate of mercury release from suspended sediment can be assessed by comparing the approximate maximum and equilibrium mercury concentrations for seawater, estuarine water and freshwater in Experiments 2 and 3. The following summarizes the results obtained at natural pH using mercury rich sediment:

	Experiment 2		Experiment 3	
	Max. Released Hg (ng/g)	Eq^{m}Hg (ng/g)	Max. Released Hg(ng/g)	Eq^{m}Hg (ng/g)
Seawater	1100	120	1123	400
Estuarine Water	650	125	600	150
Freshwater	650	-400	650	100

The generally good agreement of maximum and equilibrium values in the two separate experiments indicates that storage of water has no significant effect on the release of mercury from suspended sediment.

That mercury was not being stripped out of solution by adsorption onto the walls of the glass reaction vessels was confirmed by measuring the dissolved mercury concentration in these containers at pH 3.95 and 7.80 over a period of five days (Figures 2 and 3). A similar result was obtained for all other blank runs. The validity of concluding that changes in the dissolved mercury concentrations at much higher levels than background were the result of sediment water interactions alone was tested by measuring the variation of total mercury in a glass reaction vessel over a 24 hour period. Figures 8 and 9 show the results for seawater and estuarine water respectively. In both cases total mercury is approximately constant with changes in dissolved mercury being compensated by equal and opposite changes in particulate mercury.

The preliminary measurement of the effect of poison (2.0 g $CuSO_4$) on the rate of mercury release from mercury-rich sediment in estuarine water (Figure 6) indicated that, compared to the unpoisoned case (Figure 5), there was little difference between release maxima but a large difference between equilibrium values (poisoned lower). Experiment 5 examined the effect of using $CuSO_4$ at 2.0 g and 0.2 g per 30 litres of estuarine water on the release rates from mercury-rich and relatively mercury poor sediments. Although the concentration of $CuSO_4$ had little effect on the mercury released, the values in Experiment 5 were much higher than those of Experiment 2. Because mercury values for the $CuSO_4$ blank were similar to those for non-poisoned blanks, it is reasonable to conclude that rather than a biological effect, the high mercury concentrations in Experiment 5 resulted from a sediment preference for Cu over Hg.

The release of mercury from both mercury-rich and mercury-poor sediment showed similar behaviour for seawater, estuarine water and freshwater (Figures 10-29). The highest release occurred at lowest pH (4) and the lowest release at highest pH (9). At high pH, the equilibrium values may be negative (Figures 16, 17, 20, 21). There is ample evidence to suggest that this could be the result of the high pH flocculation of humic substances which are very effective mercury scavengers (Eckert and Sholkovitz, 1976; Sholkovitz, 1976). For each pH, the maximum release depended on ionic strength (largest for seawater, smallest for freshwater).

The composition of the sediment used in Experiment 9 (Chemex Core #1, 0-10 cm) was as follows:

(a)	X-ray analysis	Vermiculite greater than	40%
		10A Mica	20-40%
		Kaolinite	10-20%
		Chlorite less than	5%
		Amphiboles, feldspar and quartz	trace
(b)	Organic carbon		0.15%
(c)	Sand	2 mm - 0.05 mm	91.2 %
	Silt	0.05 mm - 0.002 mm	8.3 %
	Clay	less than 0.002 mm	0.5 %

The same salinity dependence occurred in this 15-day experiment as has already been noted. The difference between the poisoned and unpoisoned cases was quantitatively unimportant for all salinities (Figures 22-29).

The effect of temperature on the release of mercury was small. For example the difference between the release maximum for the mercury-rich sediment-seawater system at 6°C (Figure 16) and 17°C (Figure 7) was smaller than experimental error.

Several authors (Kurbatov et al (1951); Posselt et al (1968), Loganathan and Burau (1973)) have, in the past, used mass-law relationships to study the adsorption and desorption of trace metals on various adsorbents. This approach can be applied to the exchange reaction,

$$\text{Hg-Sed} + n\ S^{o}/oo \xrightarrow{K} \text{Hg}^{++} + n\ \text{Sed-}S^{o}/oo$$

whence,

$$K = \frac{(Hg^{++})\ (Sed\text{-}S^{o}/oo)^{n}}{(Hg\text{-}Sed)\ (S^{o}/oo)^{n}}$$

Rearranging and taking logs

$$\log\left(\frac{\text{Hg released}}{\text{Hg sorbed}}\right) = \log K + n \log (S^{o}/oo) + n \log (Sed\text{-}S^{o}/oo)$$

Assuming constant sorbent activity which is strictly true only in the case of low exchange, and that $Hg_{total} = Hg_{dissolved} + Hg_{particulate}$.

$$\log\left(\frac{\text{Hg released}}{\text{Hg sorbed}}\right) = \log K' + n \log (S^{o}/oo)$$

Figure 30 is a plot of the logarithm of the solid-solution phase distribution coefficient vs. log (S^{o}/oo), is given for the unpoisoned sea, fresh, and estuarine water cases of Experiment 9. This relationship does not imply that the interaction between mercury and water is controlled by salinity alone but the linearity does suggest the importance of ionic strength to the overall equilibrium. (It is difficult to assign any theoretical significance to the curve because the assumption of constant sorbent activity during sorption is not true here).

4.2 Static Release Experiment

The data for the 30-day aerobic static release experiment appear in Appendix A, 4. At t = OD, the concentration of dissolved mercury was about 400 ng/l increasing steadily to over 40,000 ng/l after 19 days and then decreasing to about 3,000 ng/l by 30 days. These data, once corrected for dilution effects, were used to calculate an average daily flux of mercury across the sediment-water interface by dividing the total change in dissolved mercury by 400 cm^2. This gave fluxes as high as 470 ng/cm^2/day but most were less than 100 ng/cm^2/day.

A theoretical flux considering only diffusion transport of mercury through the interstitial waters can be calculated from

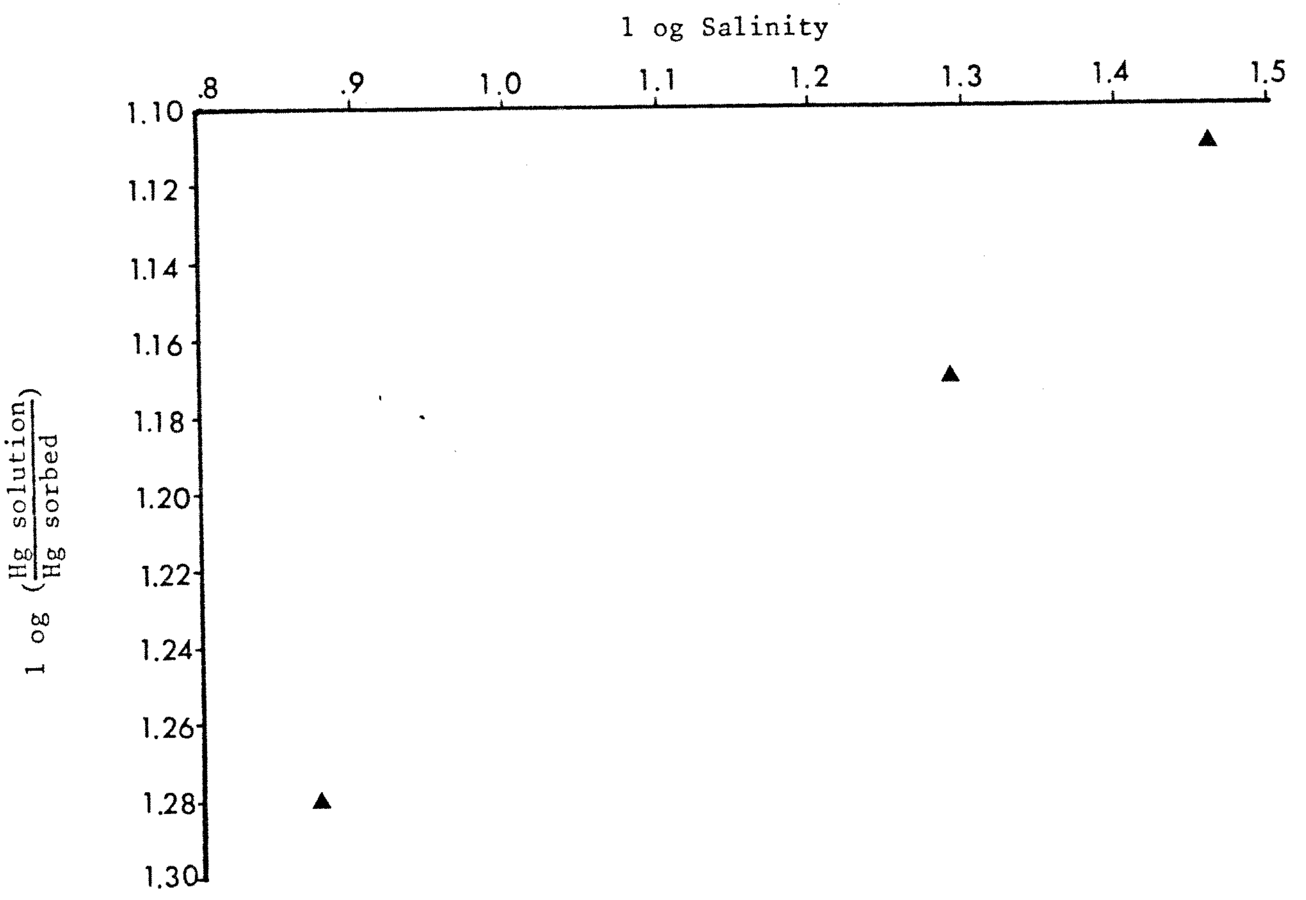


FIGURE 30. Logarithm of the distribution coefficient as a function of salinity for Experiment 9, unpoisoned case.

Fick's first law. Assuming a maximum molecular diffusion coefficient of 10^{-5} cm^2/sec and using a value of 300 ng/ℓ-7 cm for the concentration gradient (based on interstitial water analyses at t = 30 d, 6.0 ppb Hg and 6.3 ppb Hg at 10 cm and 3 cm respectively), the calculated flux is 37 ng/cm^2/sec which compares favourably with the observed fluxes. In contrast, Lu and Chen (1977) reported that for polluted mercury sediments under aerobic conditions, there was no change in interfacial mercury concentrations over a period of five months.

Analyses of sediment at the completion of the experiment indicated that most of the added mercury had been lost. Enough mercury was added to yield an average concentration of 30 ppm in the sediment and interstitial water but no value was found to exceed 1 ppm.

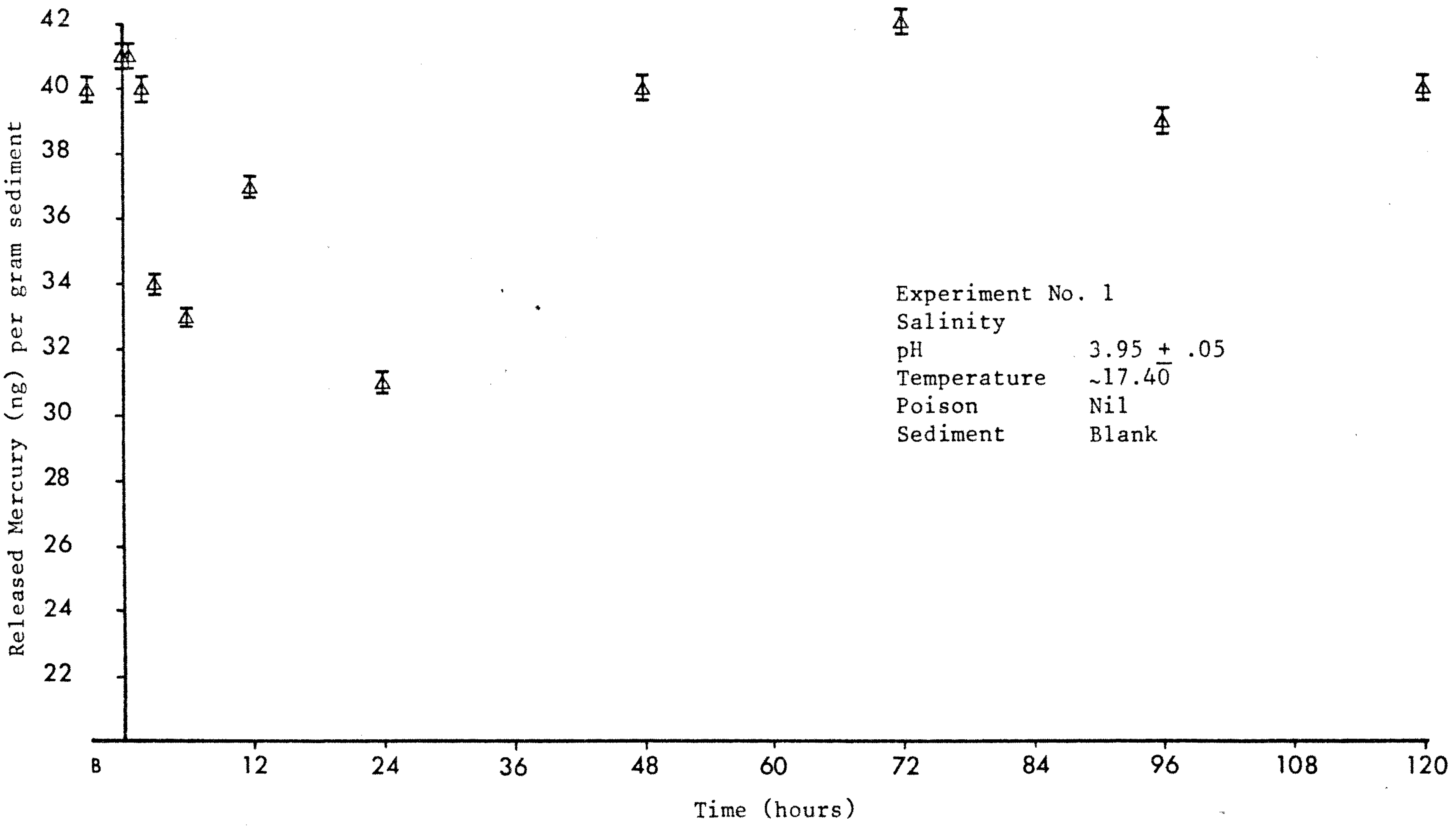

Released Mercury (ng) per gram sediment
42
40
38
36
34
32
30
28
26
24
22
B
12
24
36
48
60
72
84
96
108
120
Time (hours)
Experiment No. 1
Salinity
pH 3.95 + .05
Temperature ~17.40
Poison Nil
Sediment Blank

FIGURE 2

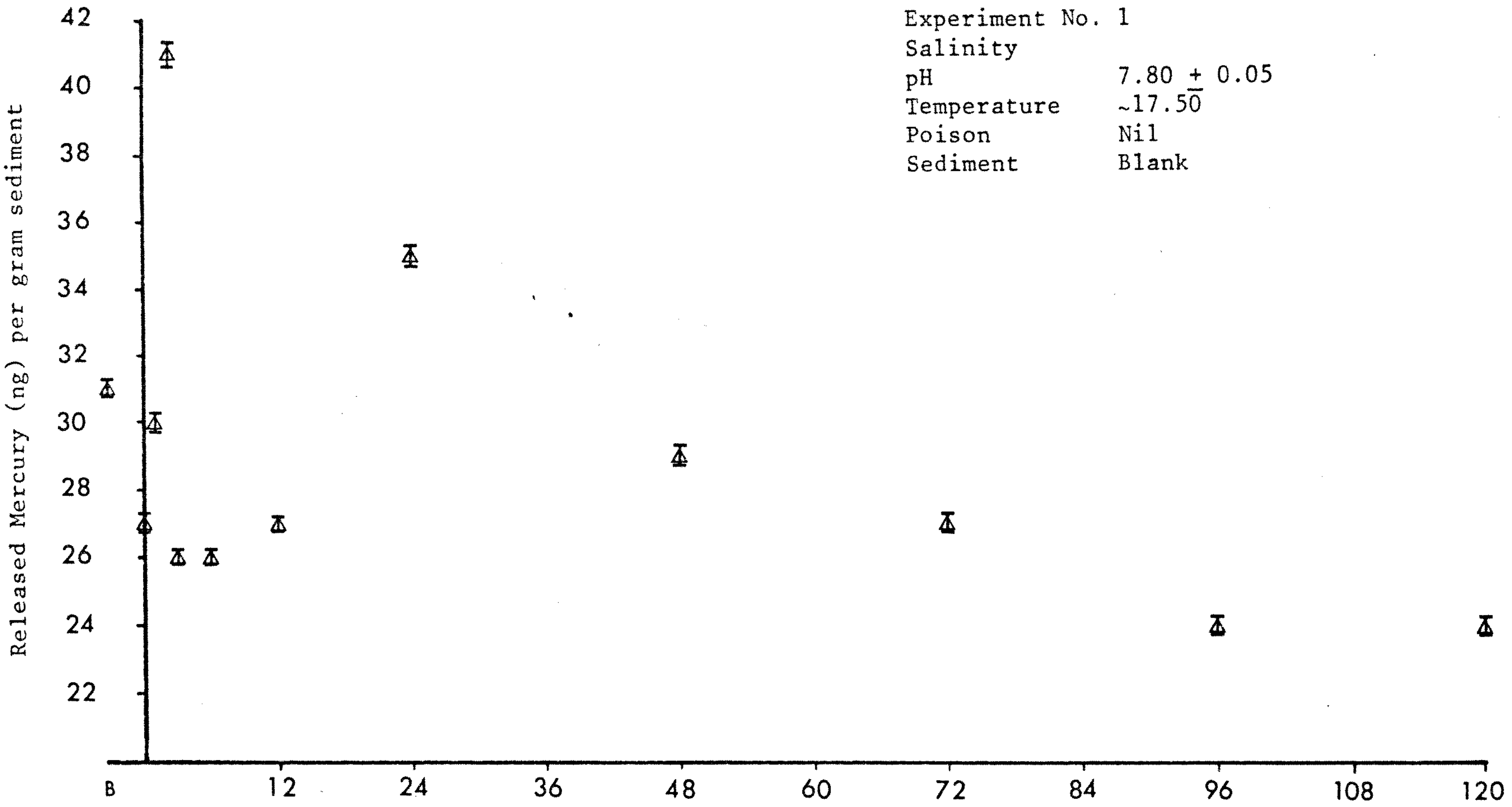

Experiment No. 1
Salinity
pH 7.80 ± 0.05
Temperature ~17.50
Poison Nil
Sediment Blank
Released Mercury (ng) per gram sediment
42
40
38
36
34
32
30
28
26
24
22
B
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 3

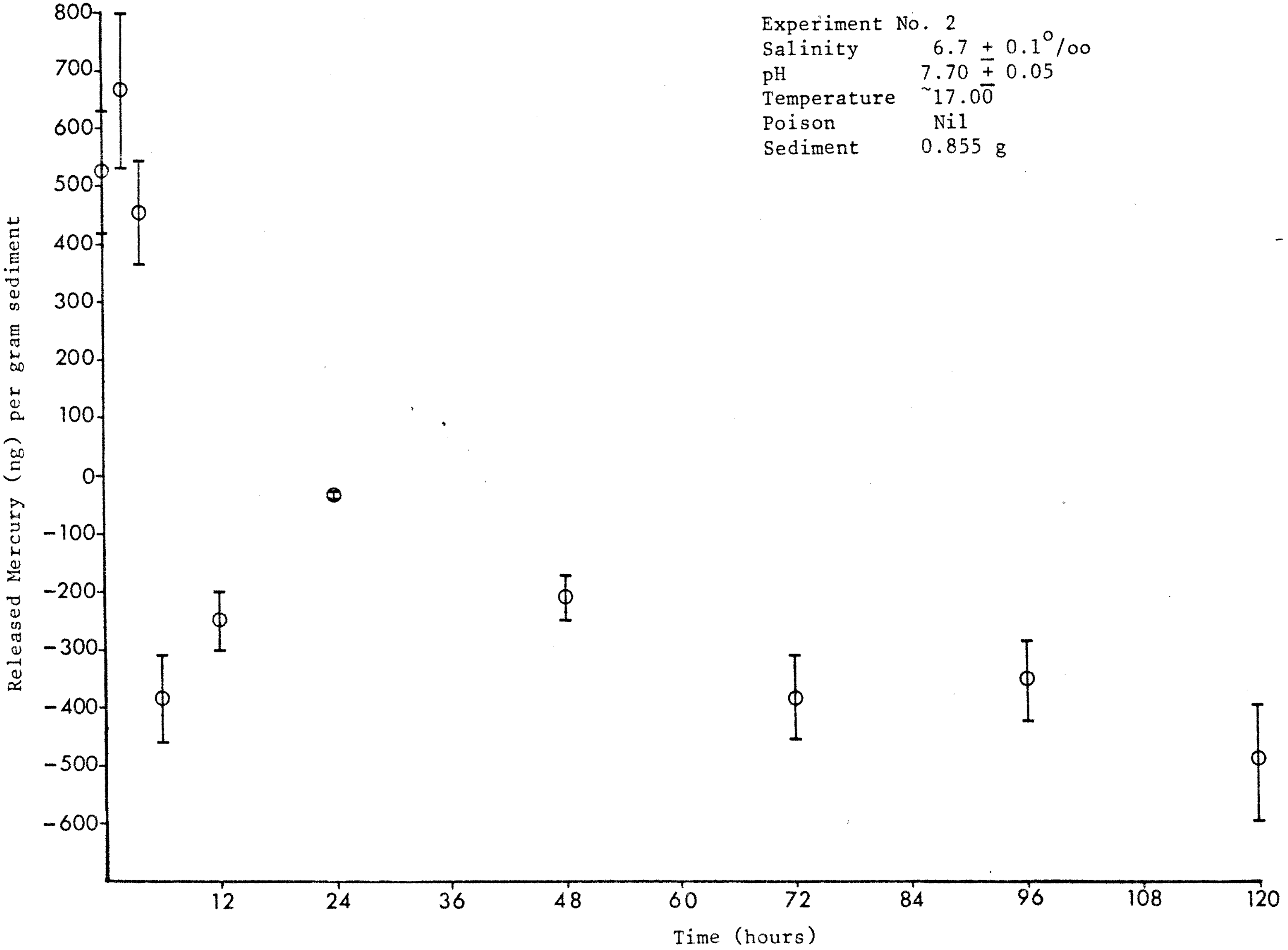

Experiment No. 2
Salinity 6.7 ± 0.1°/oo
pH 7.70 ± 0.05
Temperature ~17.00
Poison Nil
Sediment 0.855 g
Released Mercury (ng) per gram sediment
800
700
600
500
400
300
200
100
0
-100
-200
-300
-400
-500
-600
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 4

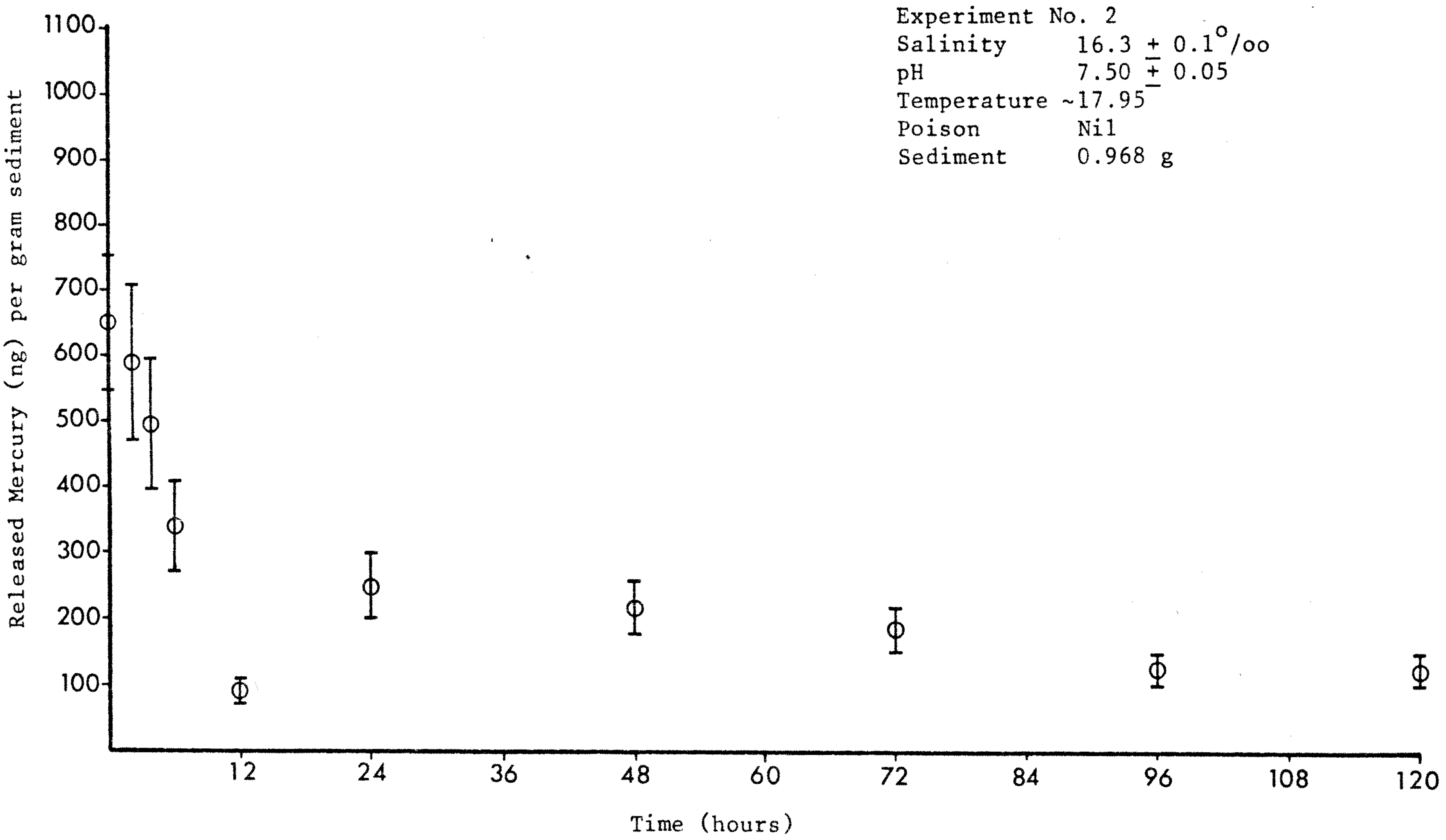

Experiment No. 2
Salinity 16.3 ± 0.1°/oo
pH 7.50 ± 0.05
Temperature ~17.95
Poison Nil
Sediment 0.968 g
Released Mercury (ng) per gram sediment
1100
1000
900
800
700
600
500
400
300
200
100
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 5

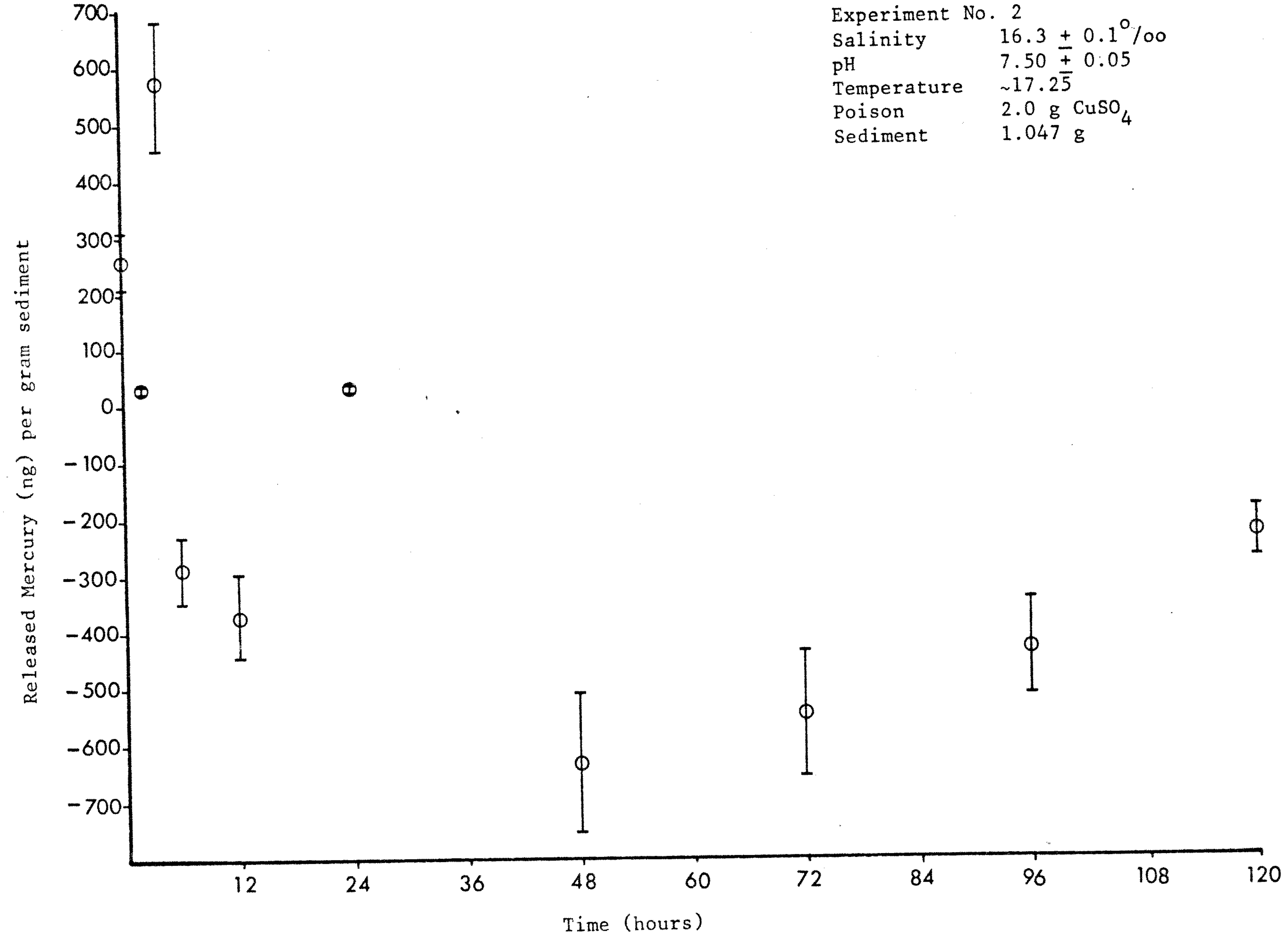

Experiment No. 2
Salinity 16.3 ± 0.1°/oo
pH 7.50 ± 0.05
Temperature ~17.25
Poison 2.0 g CuSO4
Sediment 1.047 g
700
600
500
400
300
200
100
0
-100
-200
-300
-400
-500
-600
-700
Released Mercury (ng) per gram sediment
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 6

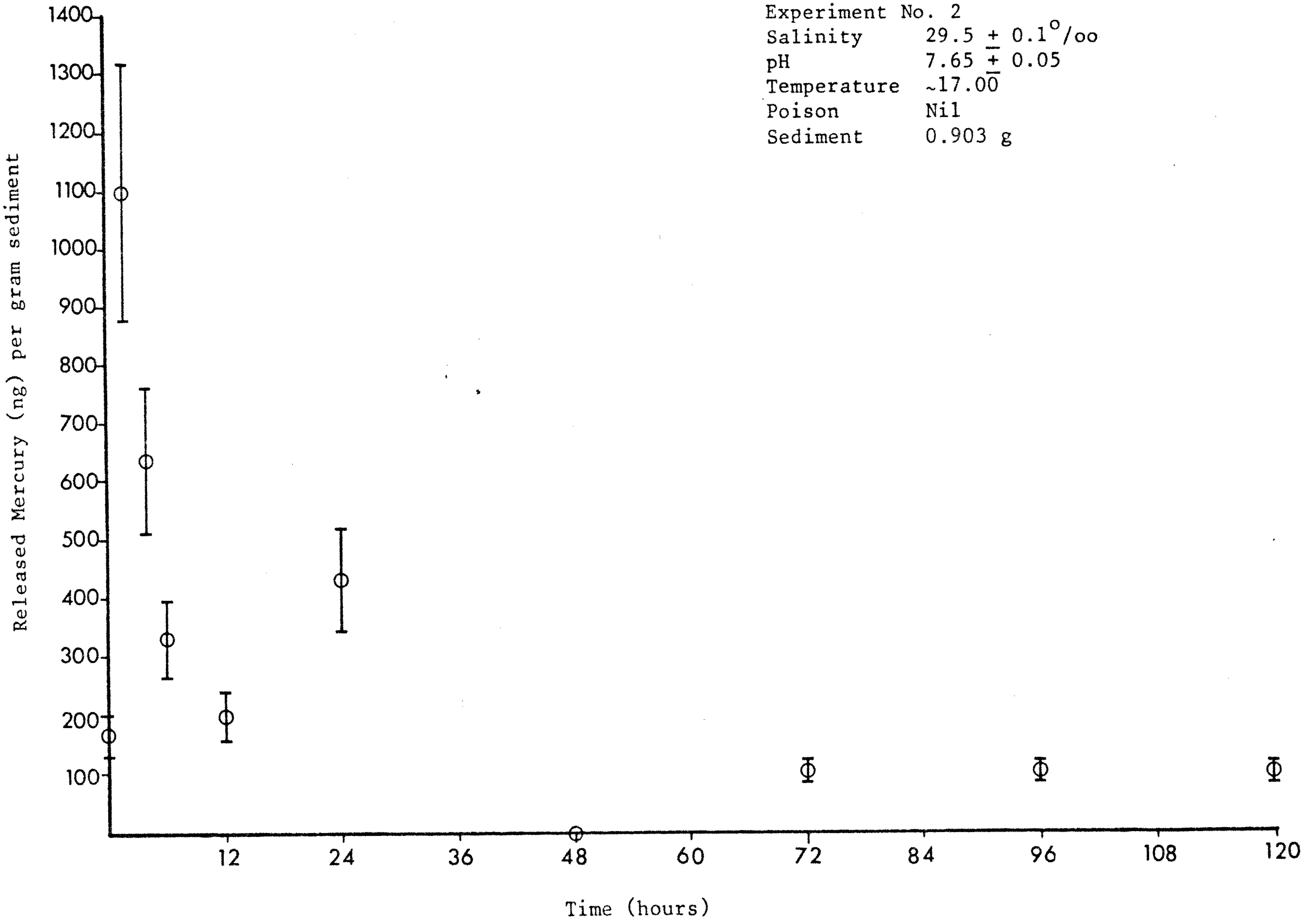

Experiment No. 2
Salinity 29.5 ± 0.1°/oo
pH 7.65 ± 0.05
Temperature ~17.00
Poison Nil
Sediment 0.903 g
Released Mercury (ng) per gram sediment
1400
1300
1200
1100
1000
900
800
700
600
500
400
300
200
100
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 7

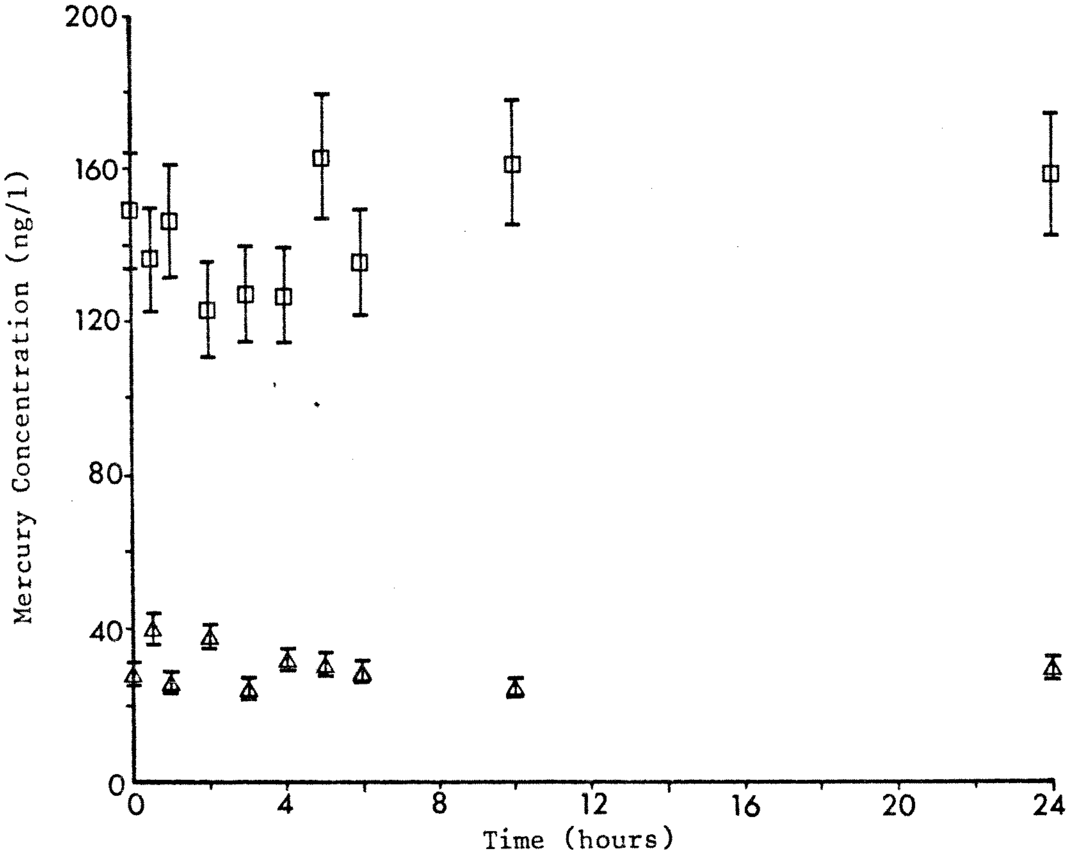


FIGURE 8. Variation of dissolved (Δ) and particulate (□) mercury over a 24 hours period for estuarine water with suspended solids concentration of 100 ppm.

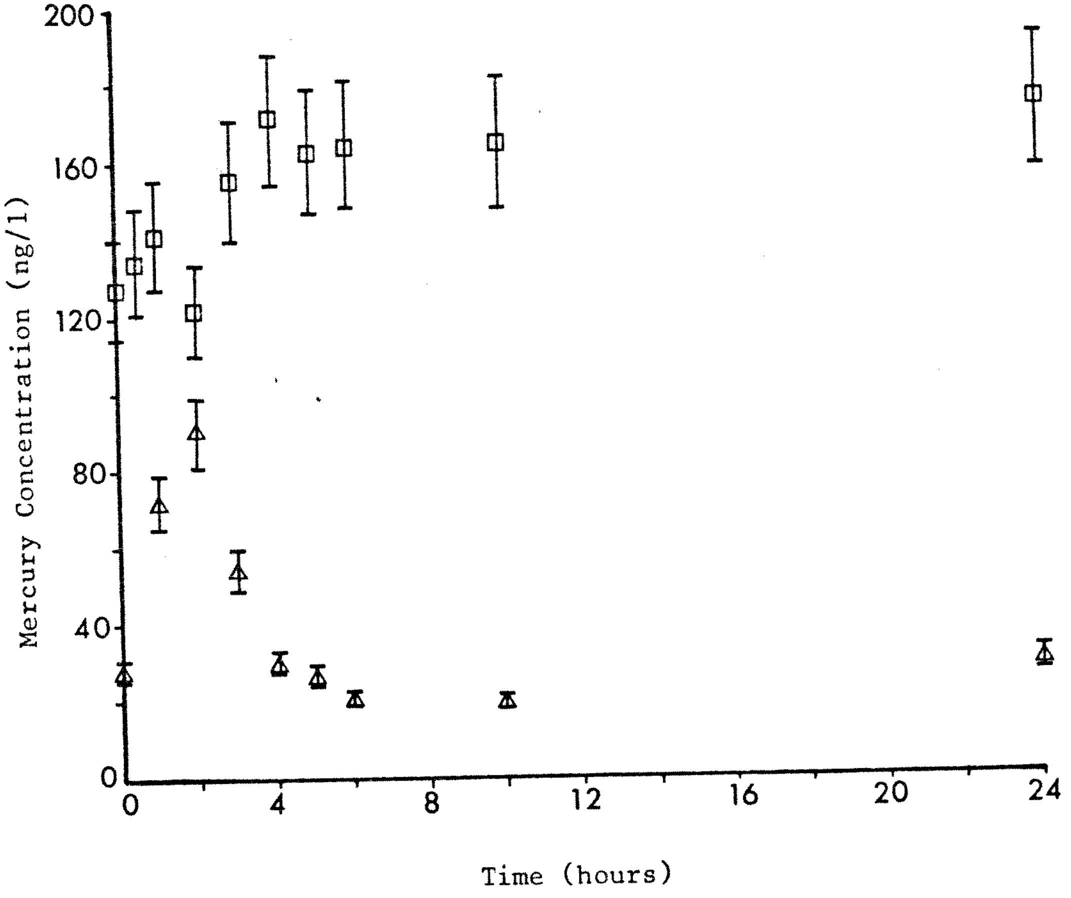


FIGURE 9. Variation of dissolved (Δ) and particulate (□) mercury over a 24 hour period for seawater with suspended solids concentration of 100 ppm.

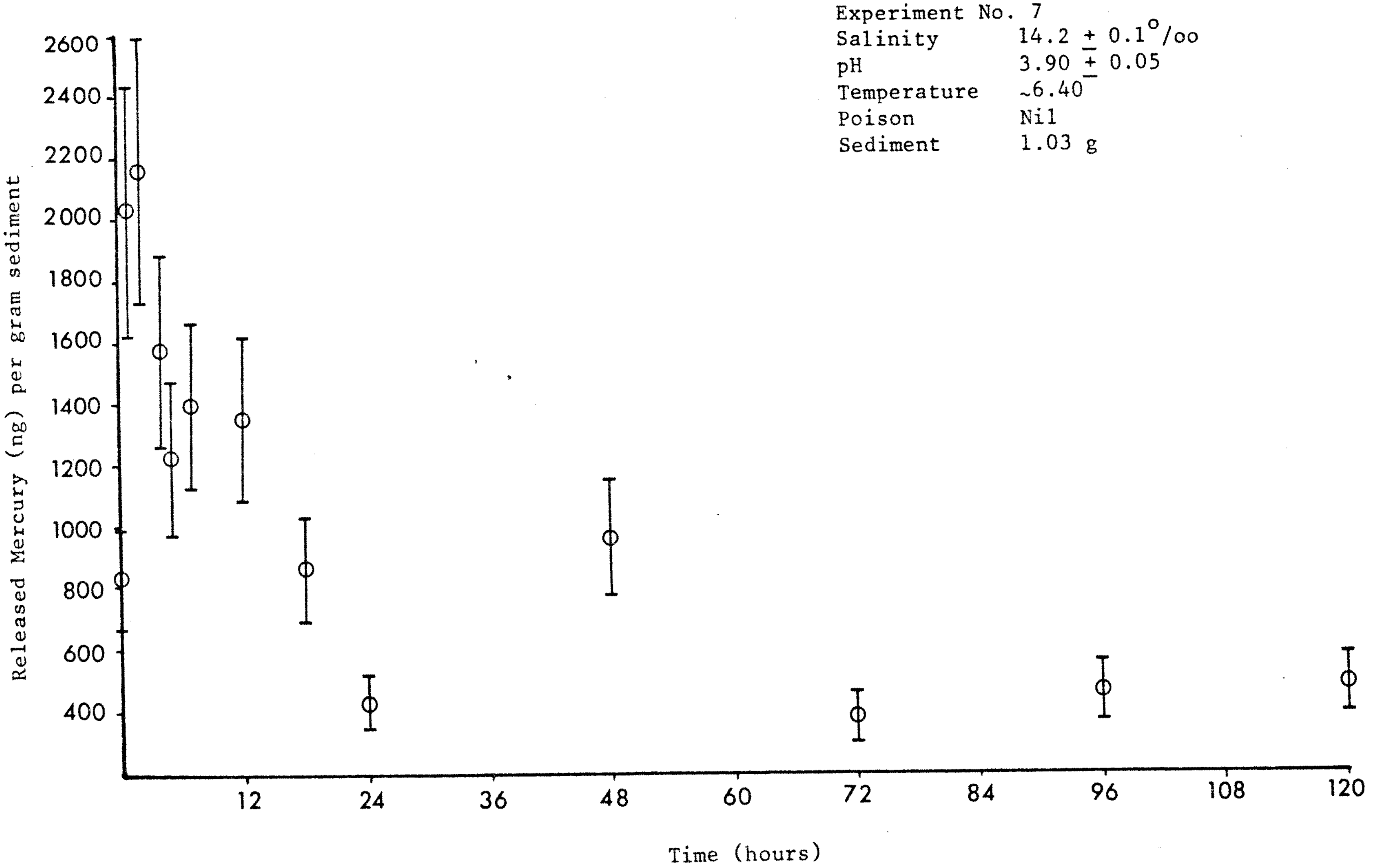

Experiment No. 7
Salinity 14.2 ± 0.1°/oo
pH 3.90 ± 0.05
Temperature ~6.40
Poison Nil
Sediment 1.03 g
Released Mercury (ng) per gram sediment
2600
2400
2200
2000
1800
1600
1400
1200
1000
800
600
400
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 10

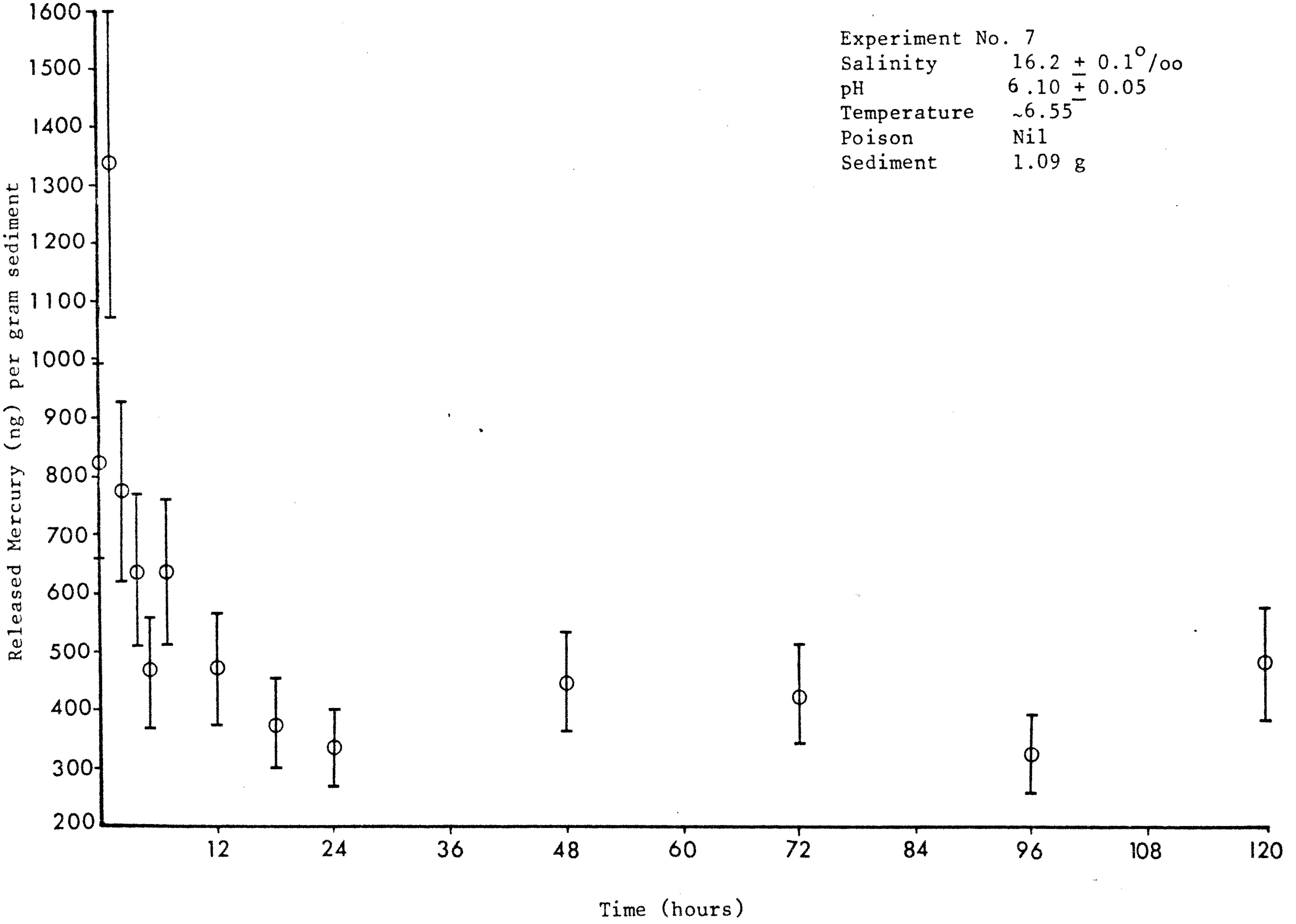

Experiment No. 7
Salinity 16.2 ± 0.1°/oo
pH 6.10 ± 0.05
Temperature ~6.55
Poison Nil
Sediment 1.09 g
Released Mercury (ng) per gram sediment
1600
1500
1400
1300
1200
1100
1000
900
800
700
600
500
400
300
200
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 11

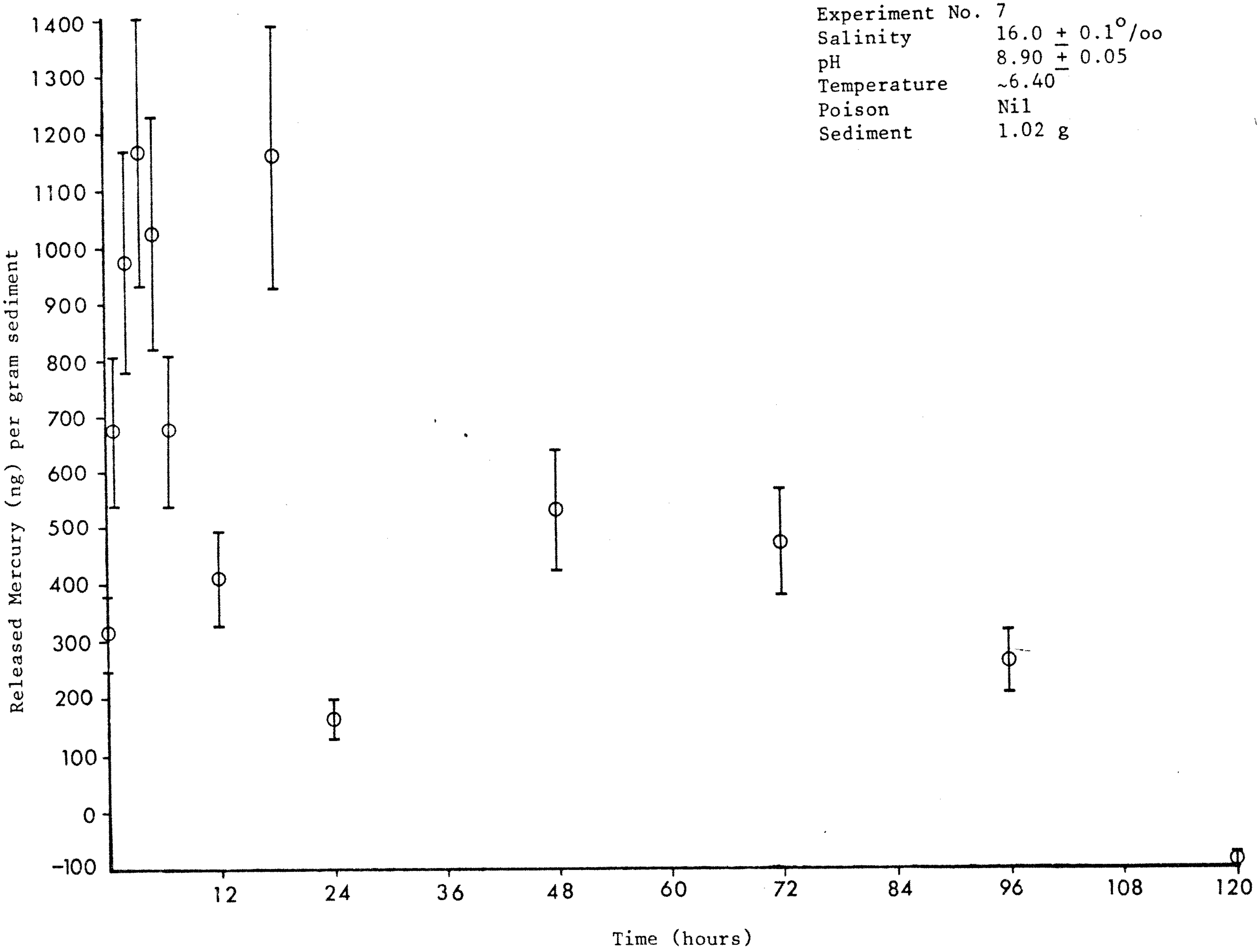

Experiment No. 7
Salinity 16.0 ± 0.1°/oo
pH 8.90 ± 0.05
Temperature ~6.40
Poison Nil
Sediment 1.02 g
Released Mercury (ng) per gram sediment
1400
1300
1200
1100
1000
900
800
700
600
500
400
300
200
100
0
-100
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 12

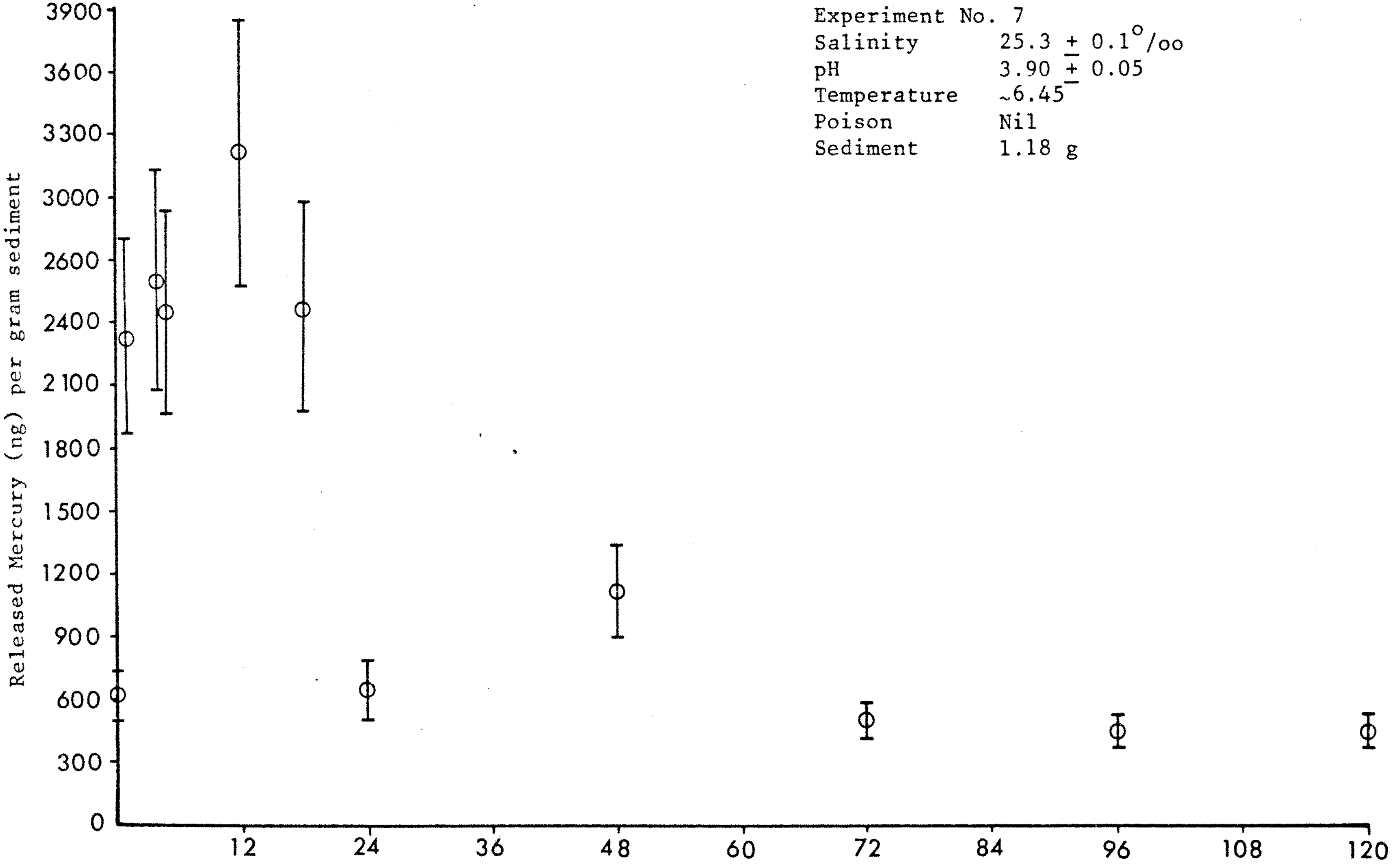

Experiment No. 7
Salinity 25.3 ± 0.1°/oo
pH 3.90 ± 0.05
Temperature ~6.45
Poison Nil
Sediment 1.18 g
Released Mercury (ng) per gram sediment
3900
3600
3300
3000
2600
2400
2100
1800
1500
1200
900
600
300
0
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 13

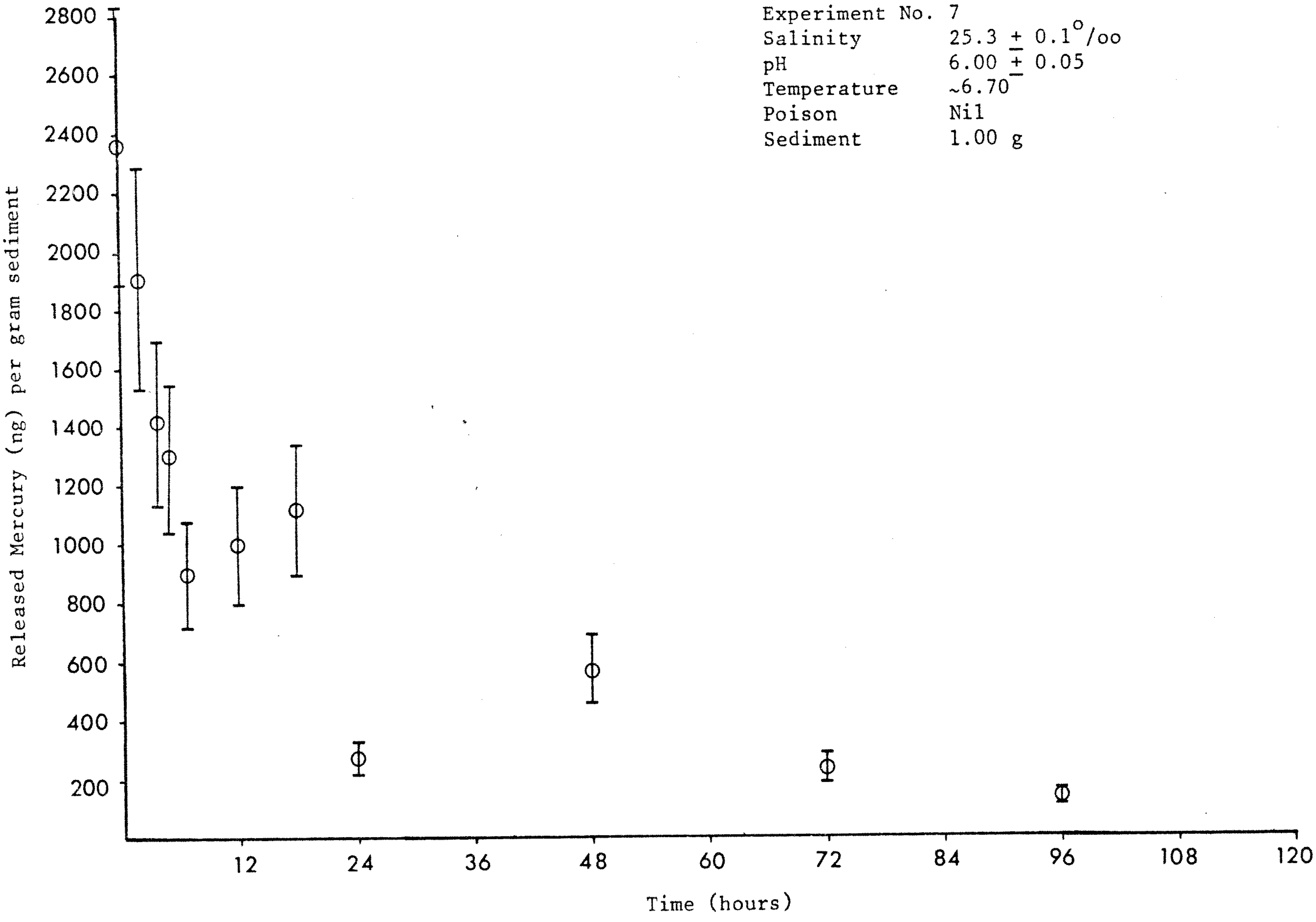
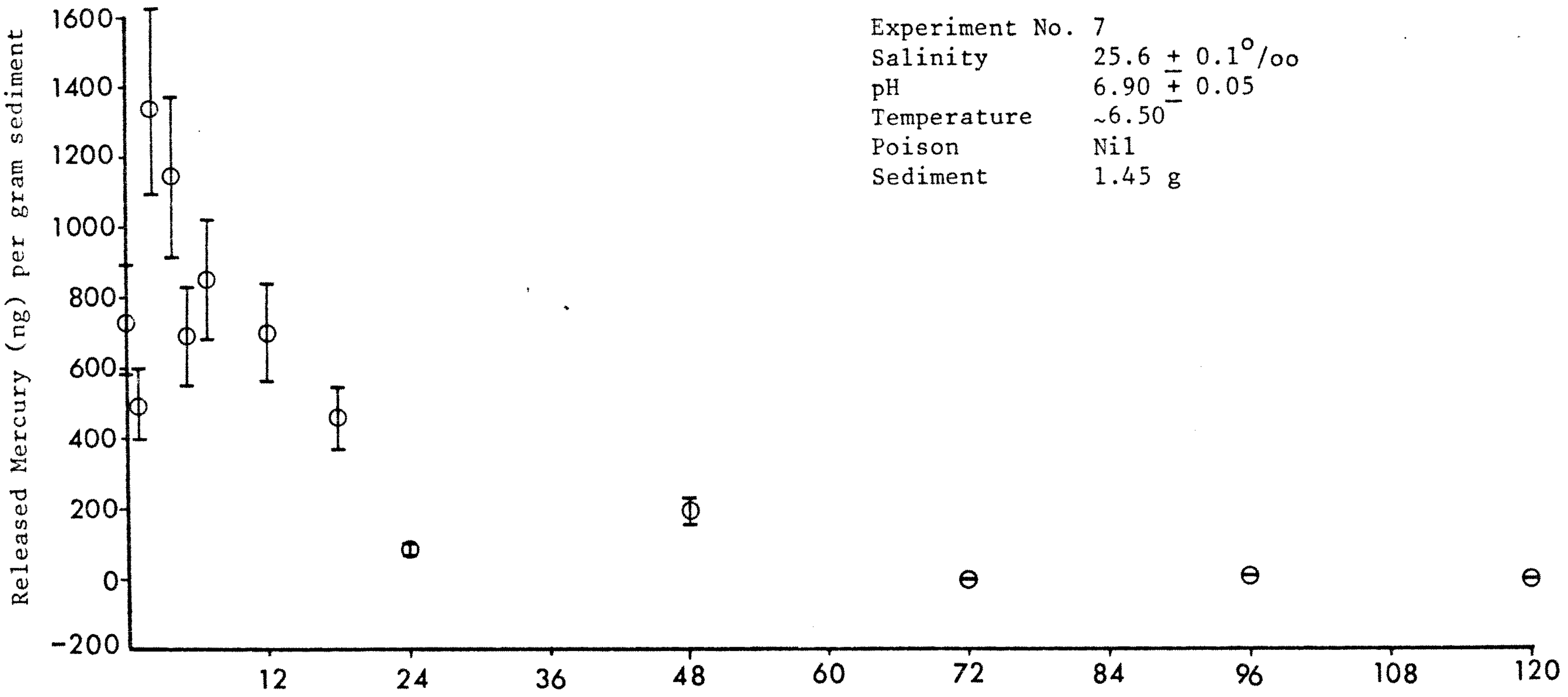

Experiment No. 7
Salinity 25.3 ± 0.1°/oo
pH 6.00 ± 0.05
Temperature ~6.70
Poison Nil
Sediment 1.00 g
Released Mercury (ng) per gram sediment
2800
2600
2400
2200
2000
1800
1600
1400
1200
1000
800
600
400
200
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 14



Time (Hours)

FIGURE 15

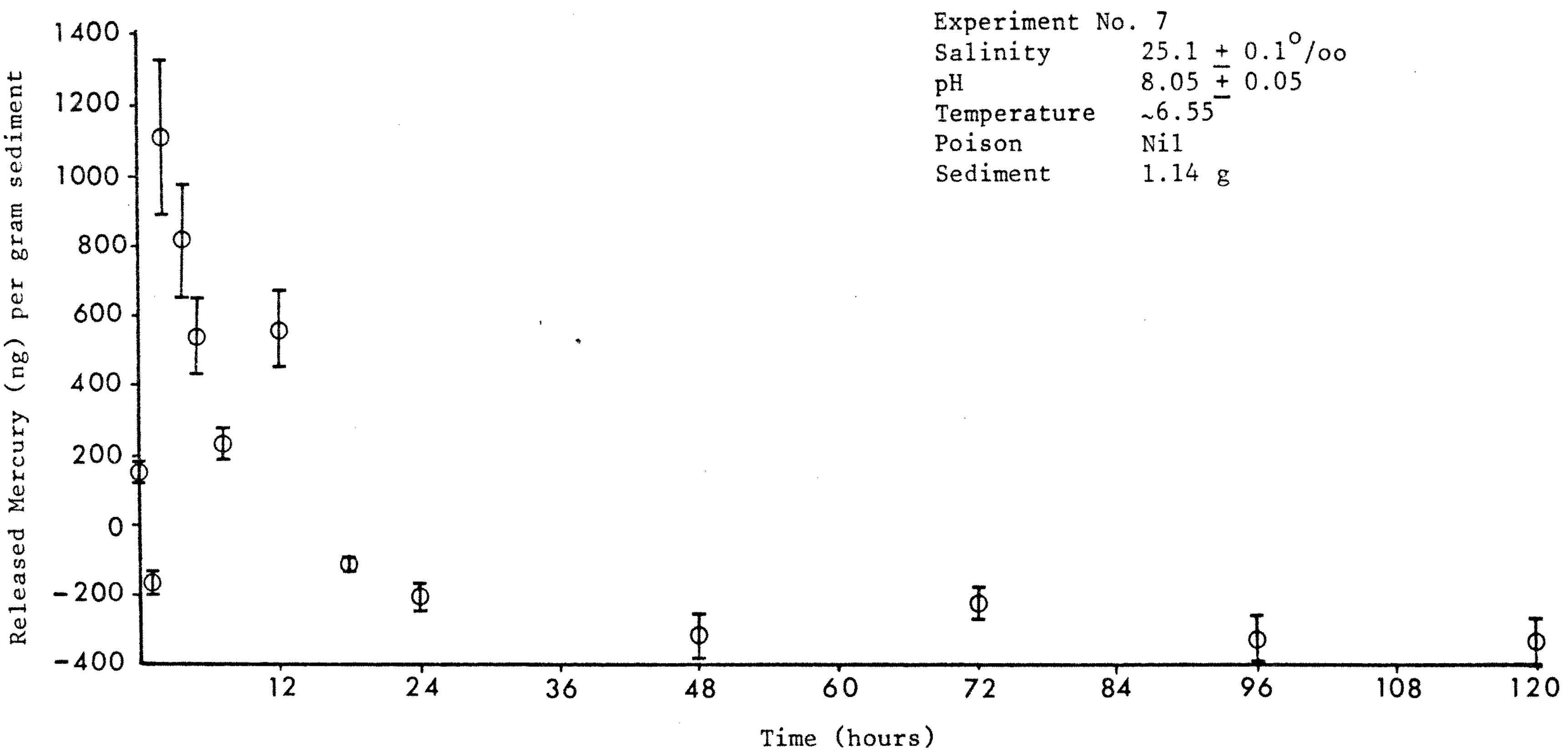

Experiment No. 7
Salinity 25.1 ± 0.1°/oo
pH 8.05 ± 0.05
Temperature ~6.55
Poison Nil
Sediment 1.14 g
Released Mercury (ng) per gram sediment
1400
1200
1000
800
600
400
200
0
-200
-400
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 16

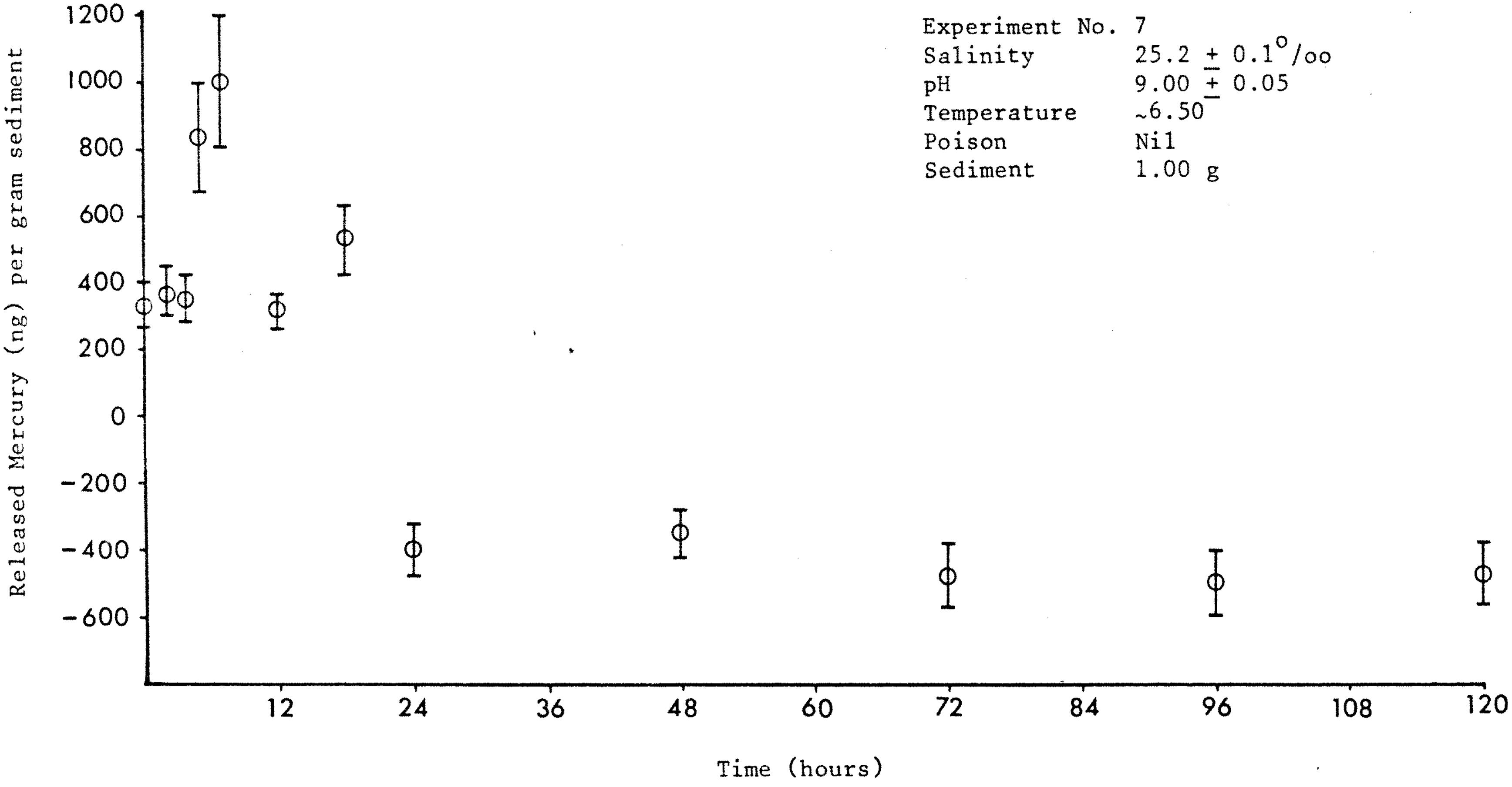

Experiment No. 7
Salinity 25.2 ± 0.1°/oo
pH 9.00 ± 0.05
Temperature ~6.50
Poison Nil
Sediment 1.00 g
Released Mercury (ng) per gram sediment
1200
1000
800
600
400
200
0
-200
-400
-600
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 17

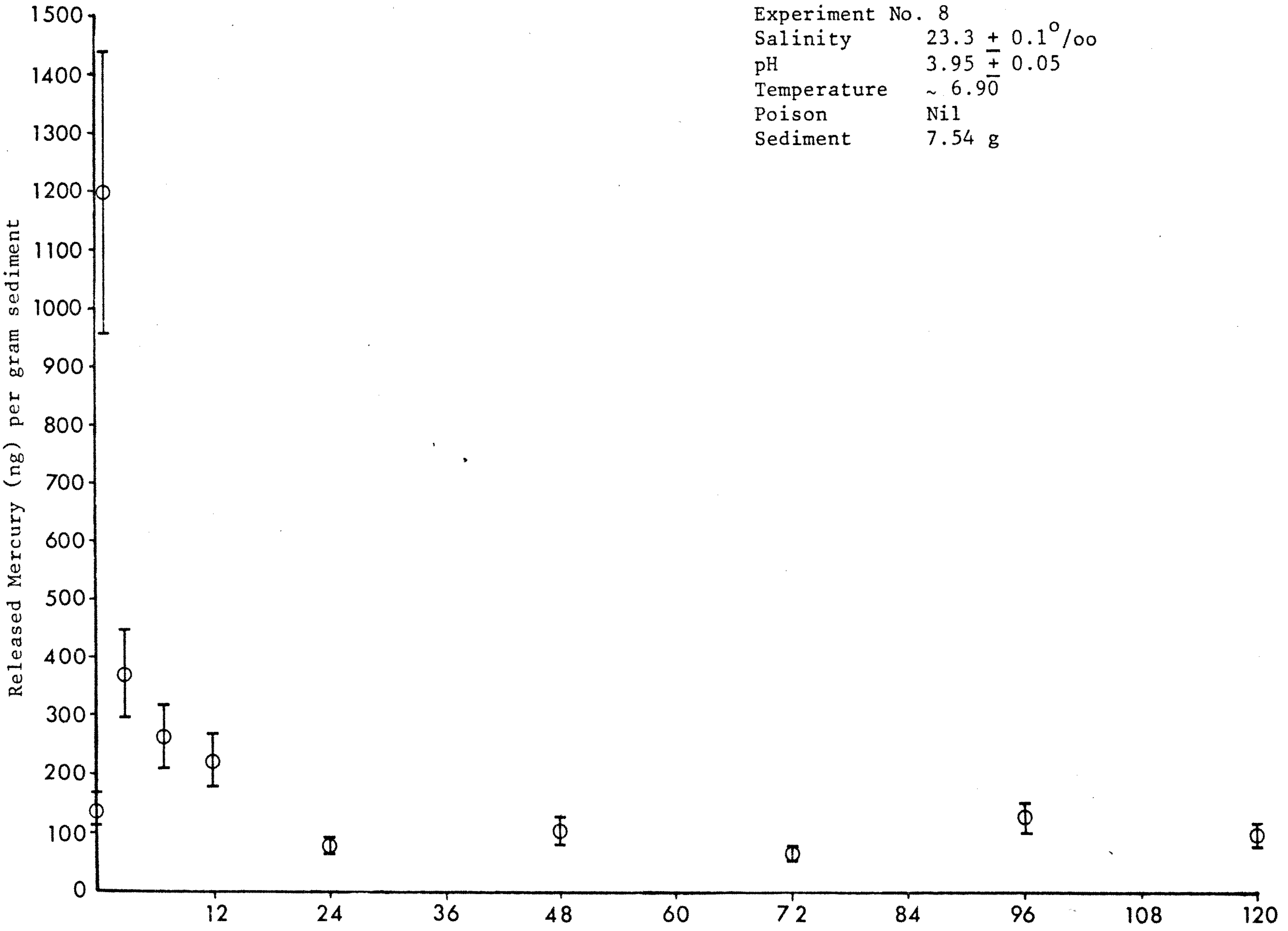

Experiment No. 8
Salinity 23.3 ± 0.1°/oo
pH 3.95 ± 0.05
Temperature ~ 6.90
Poison Nil
Sediment 7.54 g
Released Mercury (ng) per gram sediment
1500
1400
1300
1200
1100
1000
900
800
700
600
500
400
300
200
100
0
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 18

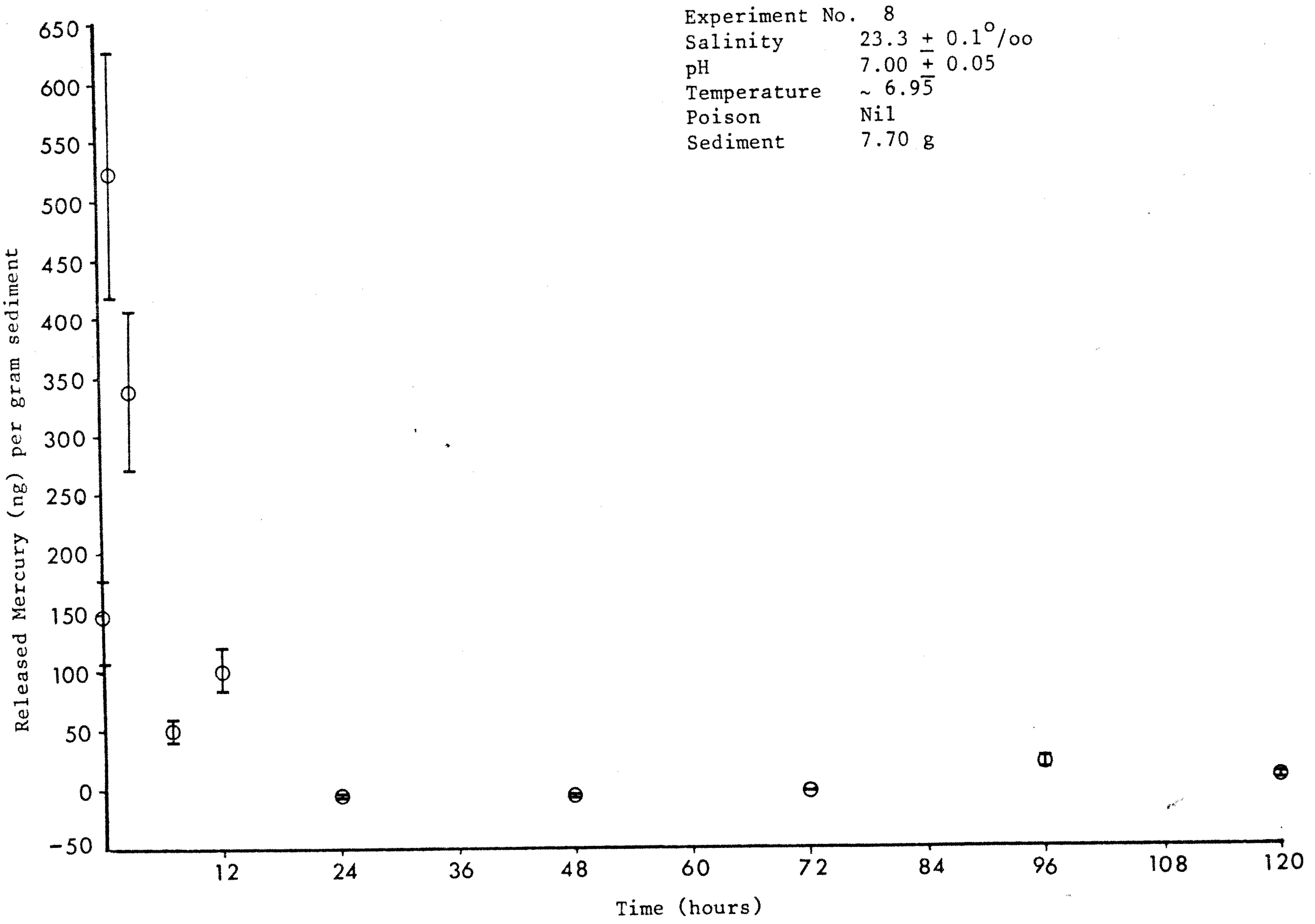

Experiment No. 8
Salinity 23.3 ± 0.1°/oo
pH 7.00 ± 0.05
Temperature ~ 6.95
Poison Nil
Sediment 7.70 g
Released Mercury (ng) per gram sediment
650
600
550
500
450
400
350
300
250
200
150
100
50
0
−50
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 19

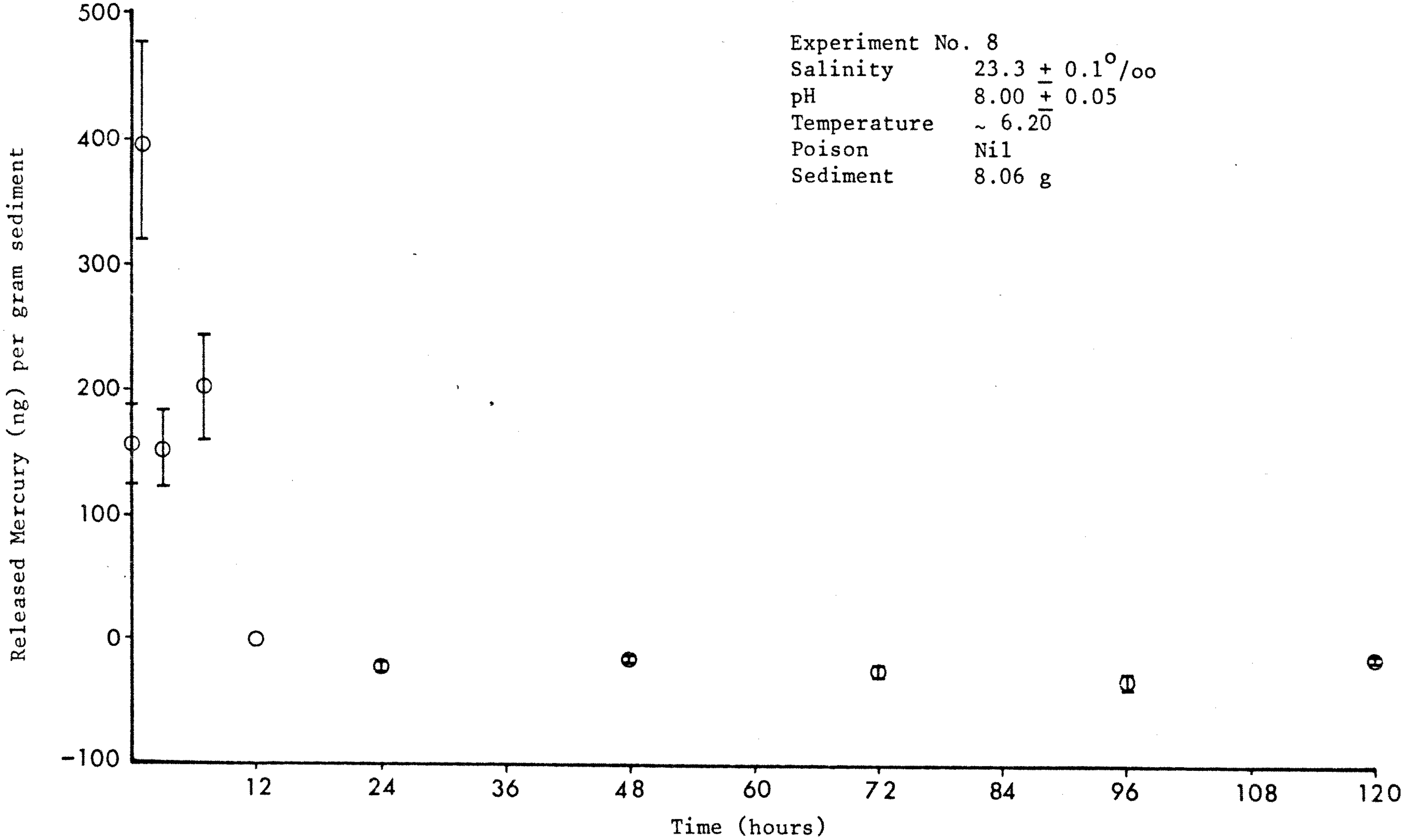
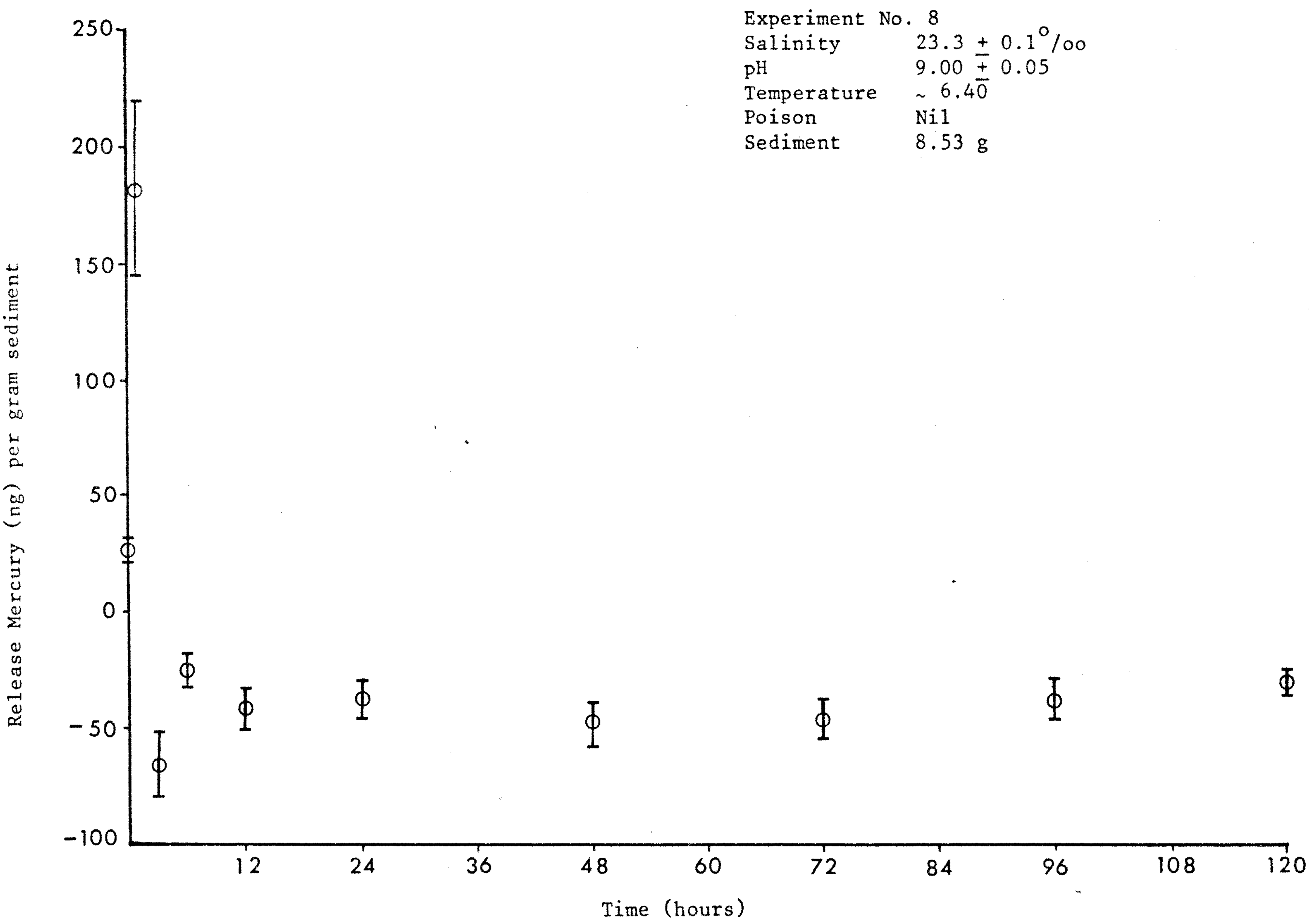

Experiment No. 8
Salinity 23.3 ± 0.1°/oo
pH 8.00 ± 0.05
Temperature ~ 6.20
Poison Nil
Sediment 8.06 g
Released Mercury (ng) per gram sediment
500
400
300
200
100
0
-100
12
24
36
48
60
72
84
96
108
120
Time (hours)

FIGURE 20



Experiment No. 8
Salinity 23.3 ± 0.1°/oo
pH 9.00 ± 0.05
Temperature ~ 6.40
Poison Nil
Sediment 8.53 g

FIGURE 21

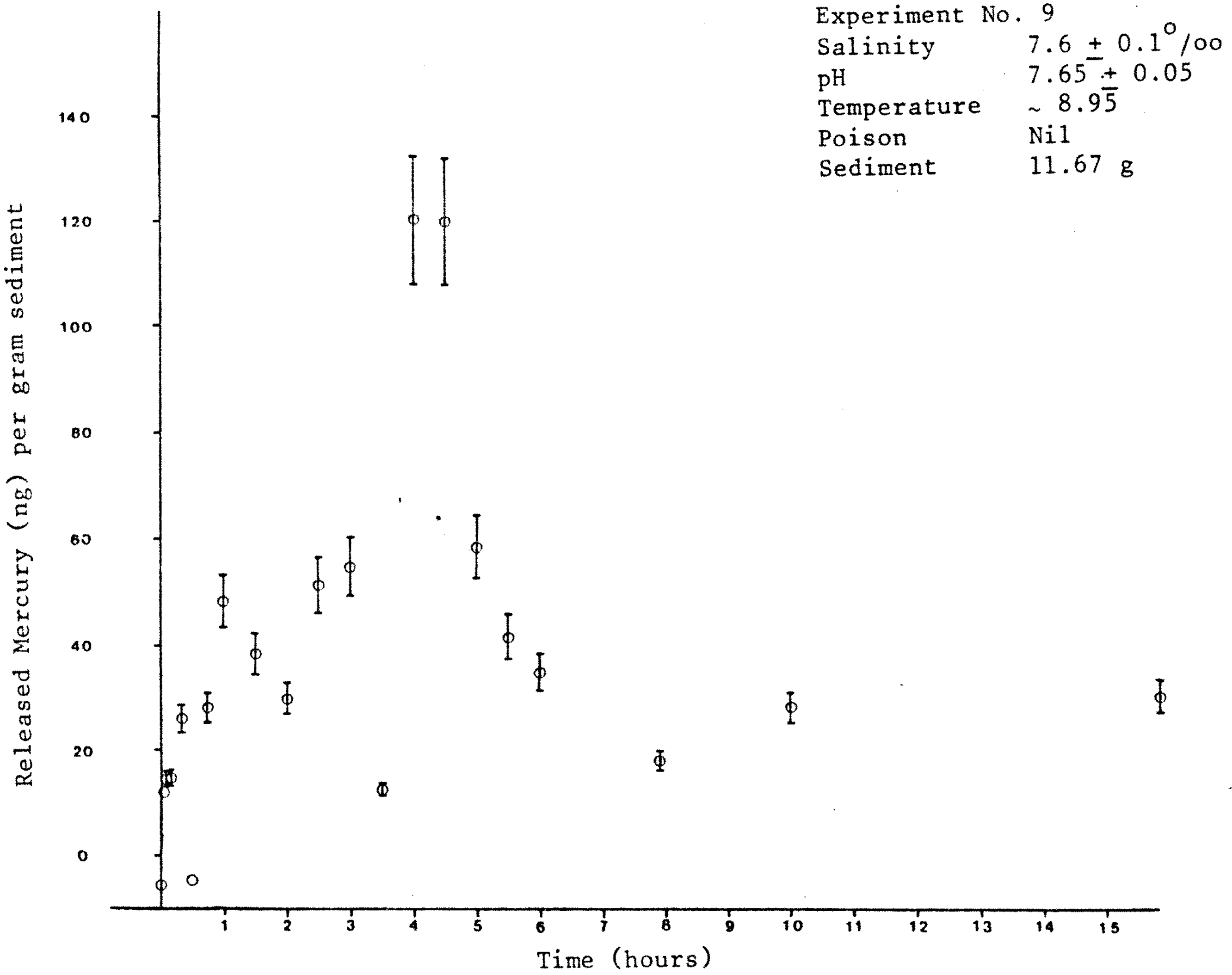

Experiment No. 9
Salinity 7.6 ± 0.1°/oo
pH 7.65 ± 0.05
Temperature ~ 8.95
Poison Nil
Sediment 11.67 g
Released Mercury (ng) per gram sediment
140
120
100
80
60
40
20
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
Time (hours)

FIGURE 22

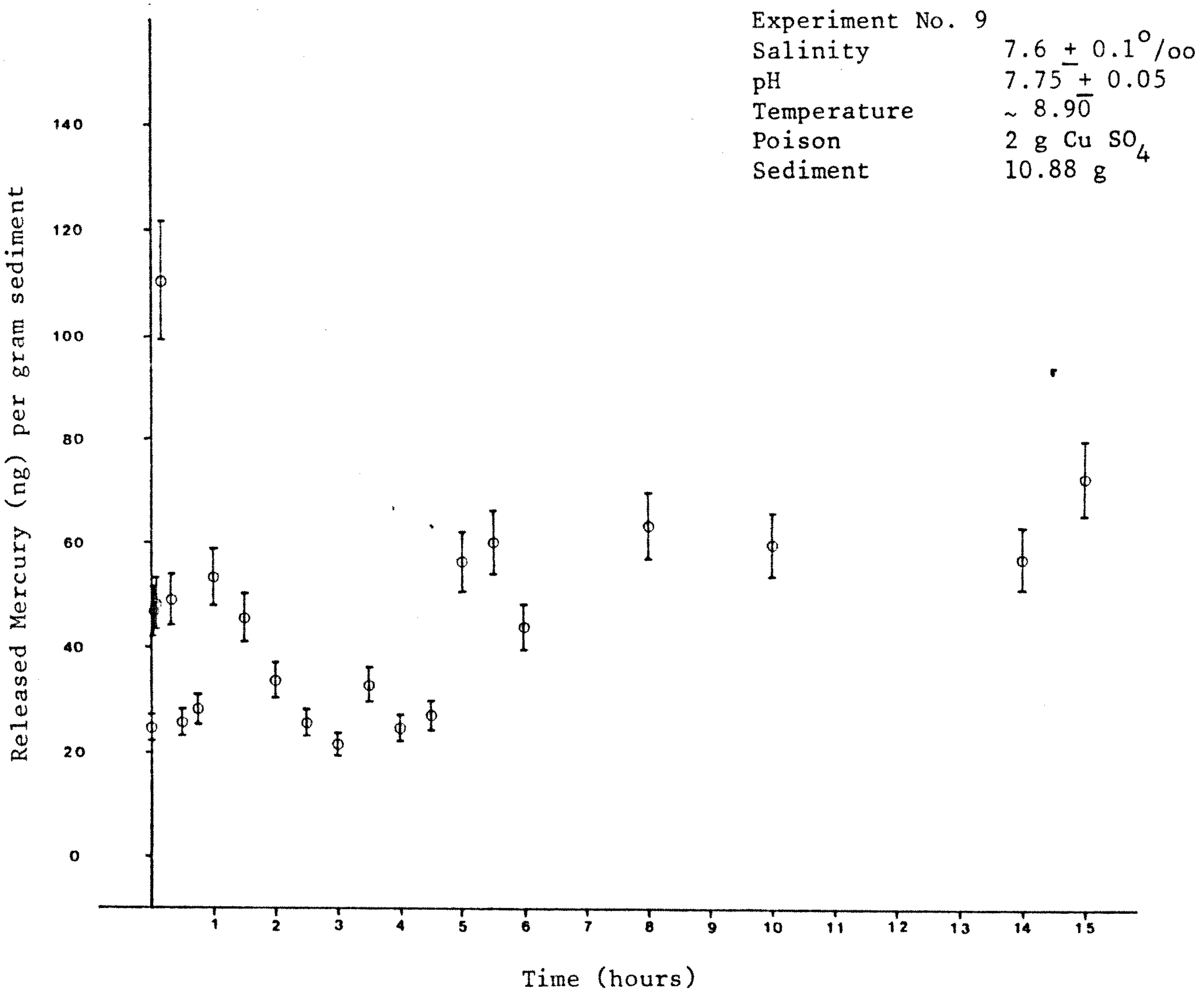
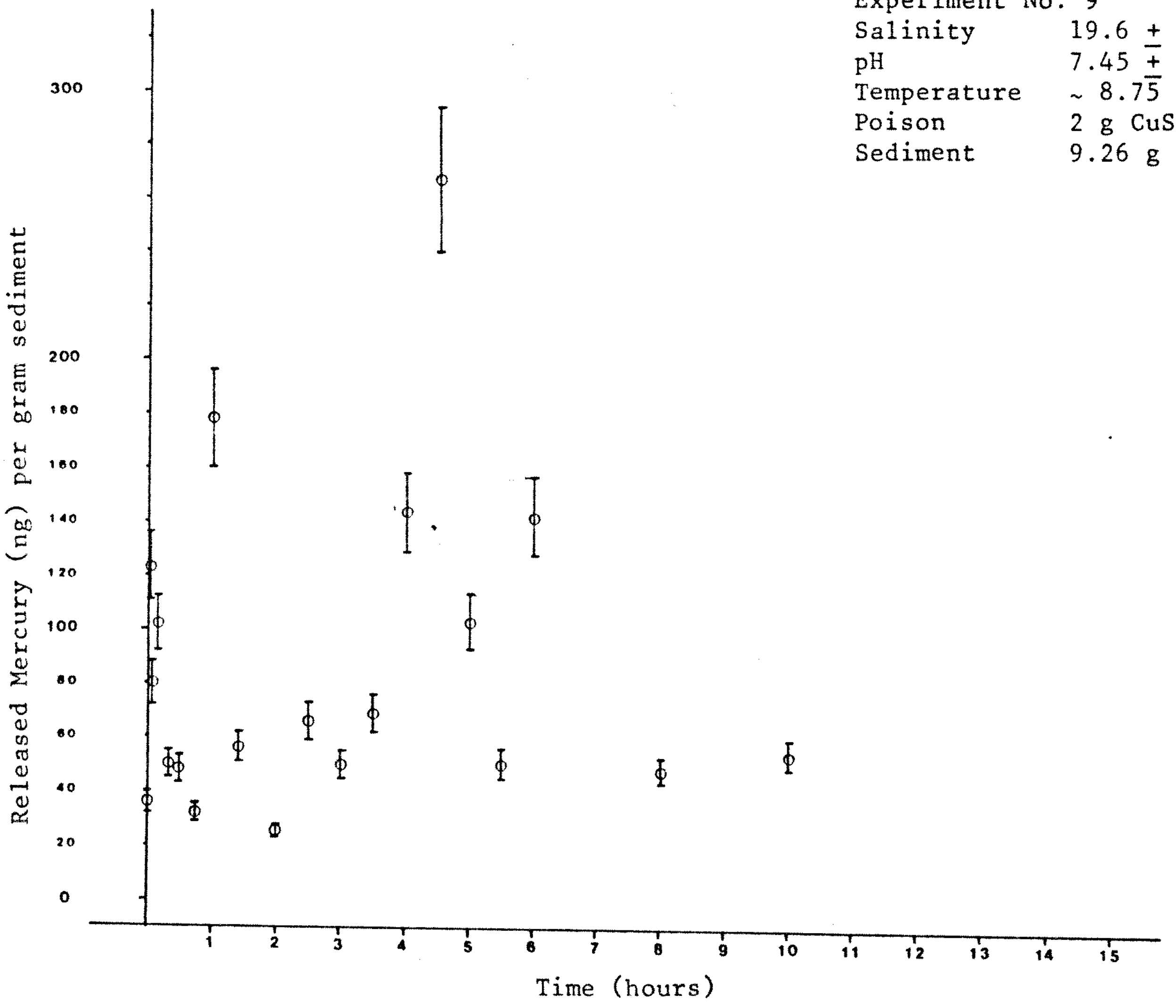

Experiment No. 9
Salinity 7.6 ± 0.1°/oo
pH 7.75 ± 0.05
Temperature ~ 8.90
Poison 2 g Cu SO_4
Sediment 10.88 g
Released Mercury (ng) per gram sediment
140
120
100
80
60
40
20
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
Time (hours)

FIGURE 23



Experiment No. 9	
Salinity	19.6 ± 0.1°/oo
pH	7.45 ± 0.05
Temperature	~ 8.75
Poison	2 g $CuSO_4$
Sediment	9.26 g

FIGURE 24

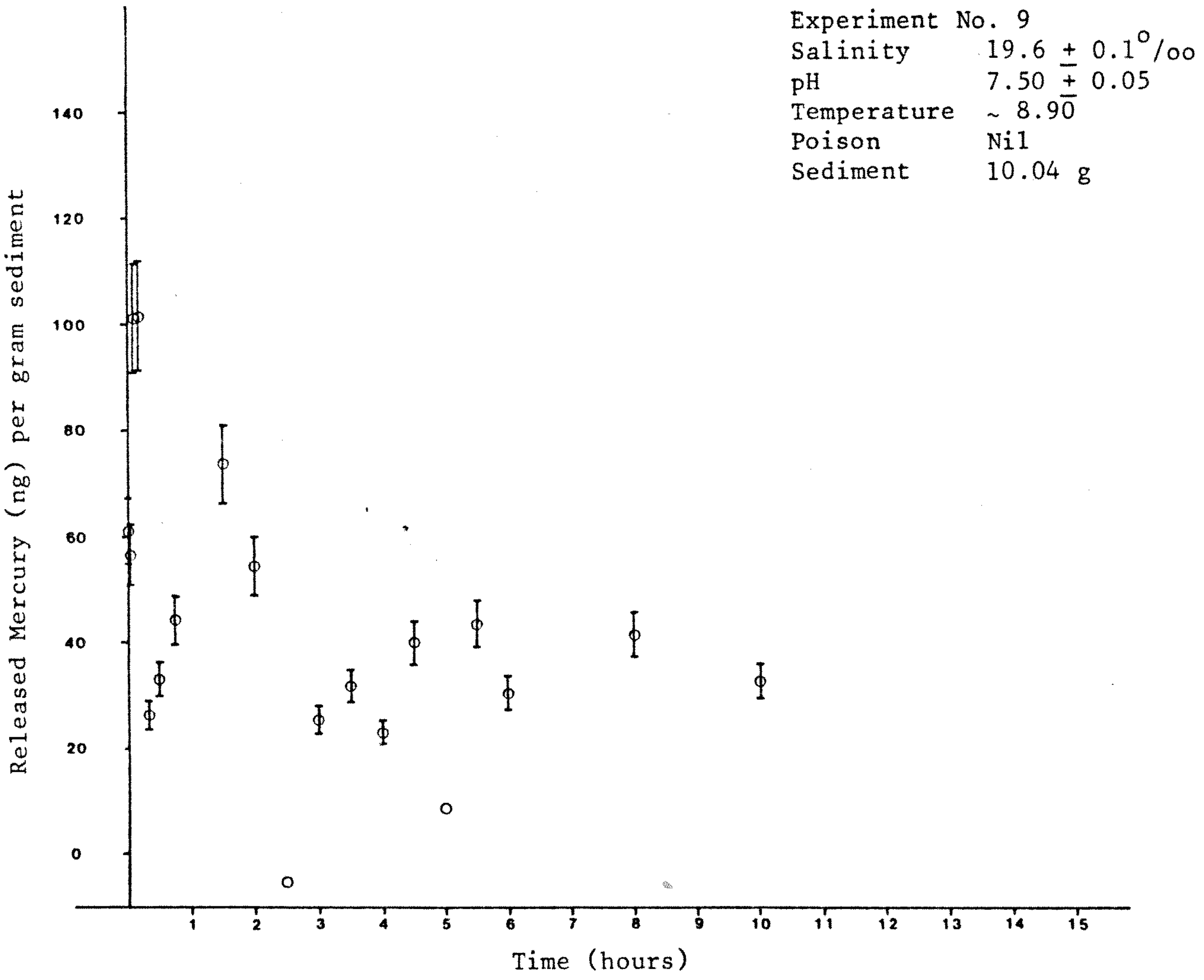

Experiment No. 9
Salinity 19.6 ± 0.1°/oo
pH 7.50 ± 0.05
Temperature ~ 8.90
Poison Nil
Sediment 10.04 g
Released Mercury (ng) per gram sediment
0
20
40
60
80
100
120
140
Time (hours)
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15

FIGURE 25

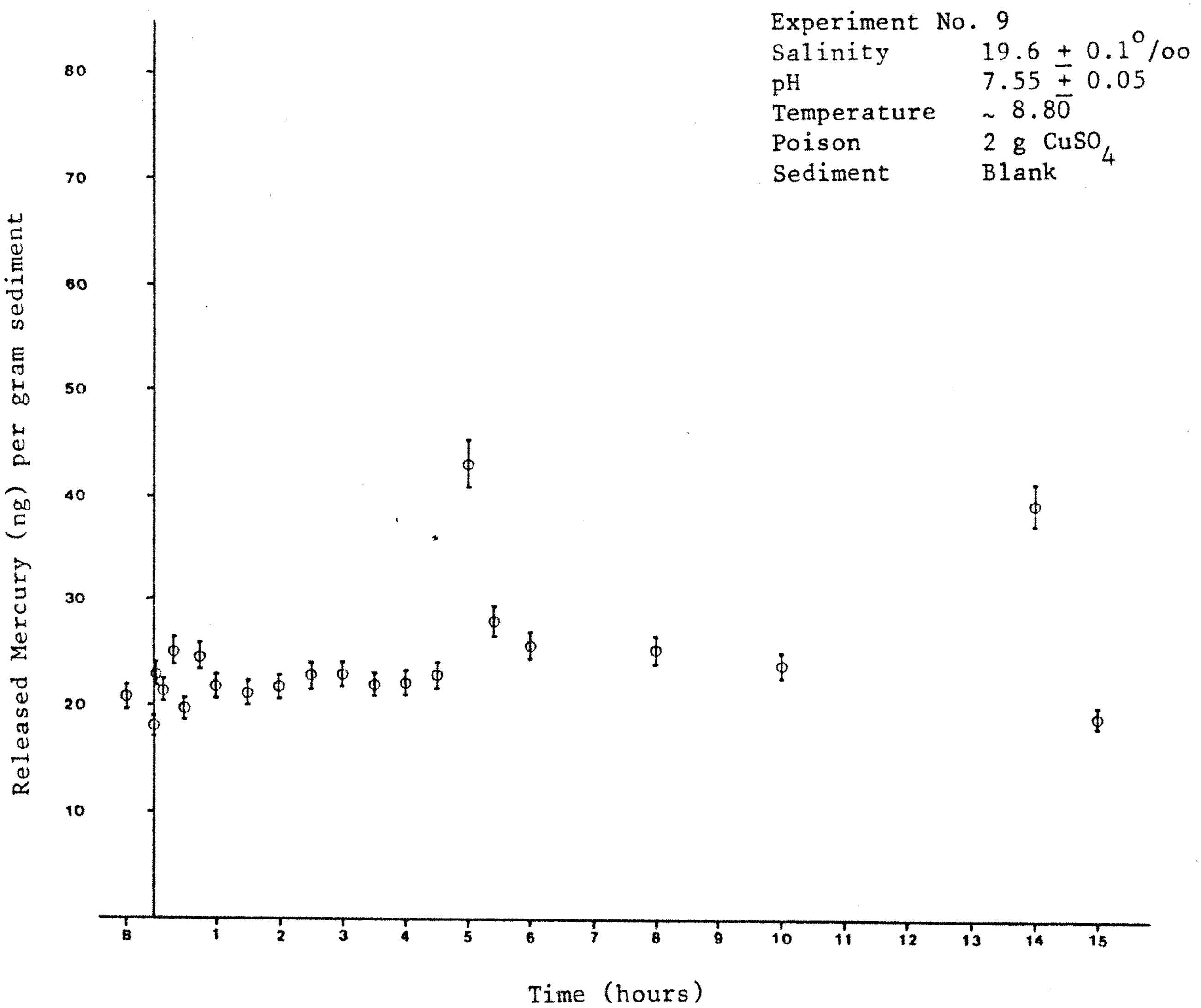

Experiment No. 9
Salinity 19.6 ± 0.1°/oo
pH 7.55 ± 0.05
Temperature ~ 8.80
Poison 2 g CuSO4
Sediment Blank
Released Mercury (ng) per gram sediment
80
70
60
50
40
30
20
10
B
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
Time (hours)

FIGURE 26

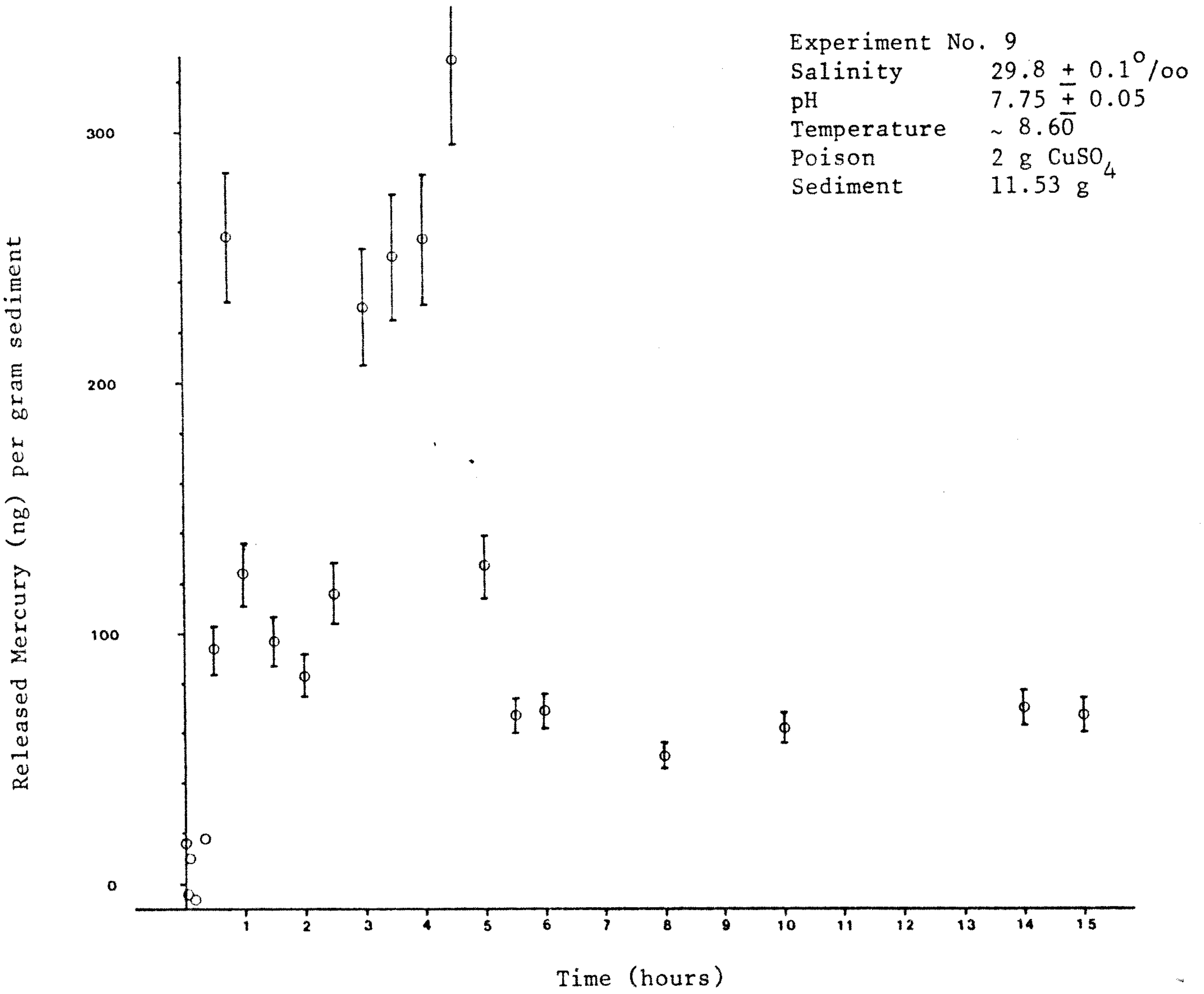

Experiment No. 9
Salinity 29.8 ± 0.1 °/oo
pH 7.75 ± 0.05
Temperature ~ 8.60
Poison 2 g $CuSO_4$
Sediment 11.53 g
Released Mercury (ng) per gram sediment
0
100
200
300
Time (hours)
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15

FIGURE 27

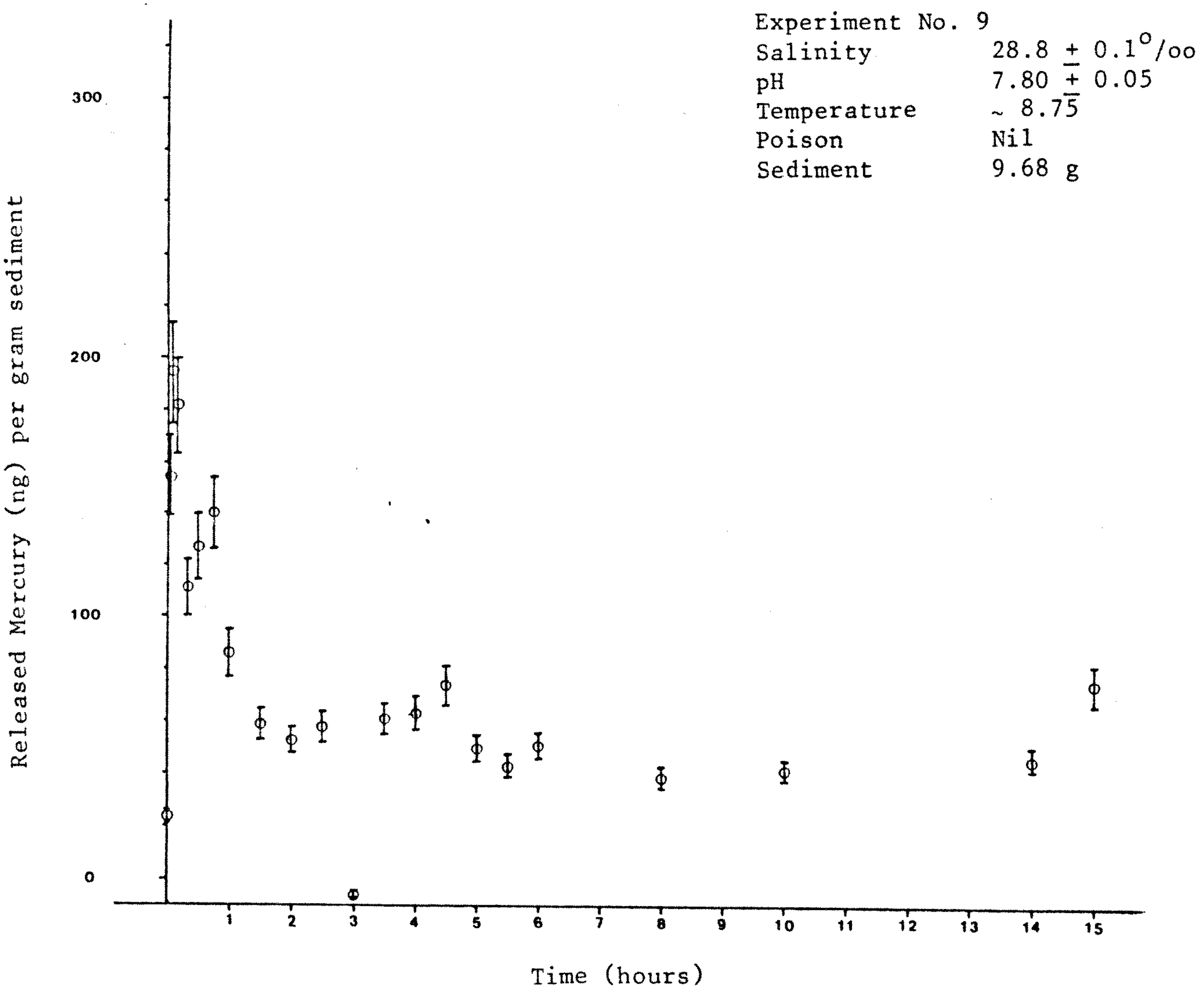

Experiment No. 9
Salinity 28.8 ± 0.1°/oo
pH 7.80 ± 0.05
Temperature ~ 8.75
Poison Nil
Sediment 9.68 g
Released Mercury (ng) per gram sediment
300
200
100
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
Time (hours)

FIGURE 28

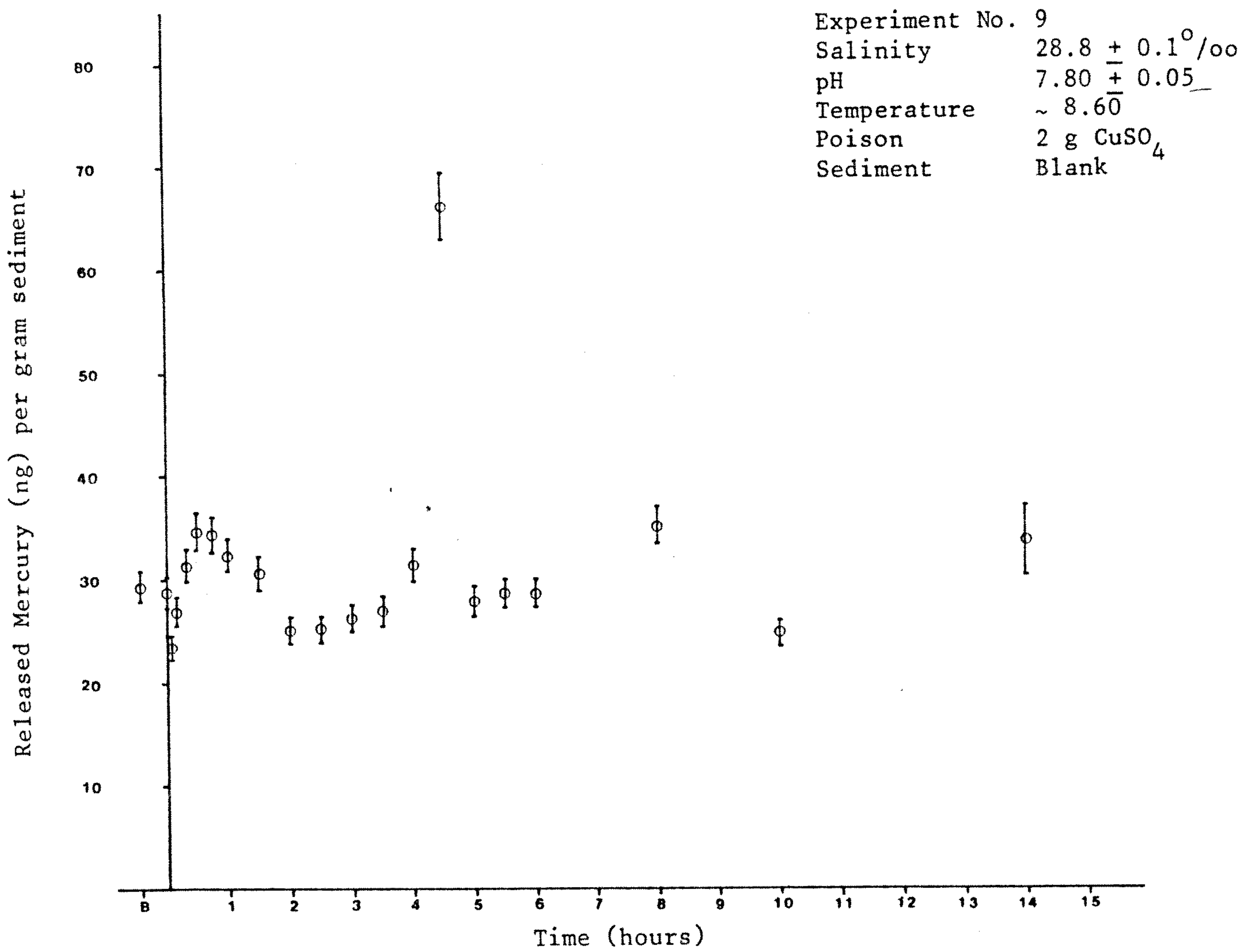

Experiment No. 9
Salinity 28.8 ± 0.1 °/oo
pH 7.80 ± 0.05
Temperature ~ 8.60
Poison 2 g $CuSO_4$
Sediment Blank
Released Mercury (ng) per gram sediment
80
70
60
50
40
30
20
10
B
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
Time (hours)

FIGURE 29

PART B: IN SITU MERCURY MOBILIZATION STUDY

1. INTRODUCTION

Studies involving trace metal distributions in the Dutch Wadden Sea (Duinker, 1974) have indicated that, in inter-tidal waters, the observed mobilization of trace metal from the sediment to the overlying waters may be related to the tidal oscillation. A conception of the process, though likely over-simplified, is that during a flooding tide, the sediment is disturbed causing solid particles to be re-suspended and interstitial waters (usually higher in dissolved constituents than overlying waters) to be mixed upward. The net result may be a sharp and significant increase in dissolved metal concentration.

Other mechanisms of metal mobilization have been proposed, for example, biological reduction followed by removal through evolution of gas bubbles (Alberts et al, 1974) biological resuspension effects (Horne, 1969), simple diffusion (Presley, et al, 1967), desorption (Rohatgi and Chen, 1975), methylation (Jensen and Jernelov, 1969) and complex formation (Linberg and Harris, 1974). However, these mechanisms, with the possible exception of complex formation involving humic acids (Nissenbaum and Swaine, 1976) are not considered capable of accounting for the major part of the mercury mobilized from sediment (Lindberg et al, 1975).

Therefore, emphasis in this study was placed on investigating in situ, the variation of dissolved, particulate and pore water mercury over a tidal cycle. An area adjoining the wastewater outfall of the FMC chlor-alkali plant on the Squamish River mudflats at Squamish, B.C. was chosen as the study site for several reasons. Firstly, because of past pollution, the sediments in this area are reported to contain high levels of mercury (Thompson and McComas, 1973). Secondly, the mercury-rich sediments of the tidal flats near the FMC outfall are easily accessible and enclosed on three sides by groins forming a relatively small and well sheltered embayment.

Thirdly, there is a continuing source of mercury rich waters at the present time.

2. SAMPLING PROGRAMME

On December 1 - December 2, 1976 during an 8.8' tide, a series of five sampling sites spaced about equally along a line extending 150 m seaward from the outfall area was occupied (Figure 31). Site 1 was located directly in the effluent stream approximately 5 metres from the outfall. Site 2 was located in a weed bed growing upon a great deal of putrifying organic debris and fine silt. Site 3 was in the main effluent stream (between a large well-sorted san bar and the weed bed. The sediment was poorly sorted sands, silts and organic debris. Site 4 was located in the centre of a large sandy depression. Site 5 was about 150 metres from the outfall in an area of highly compacted and well sorted sand.

The path of the effluent stream varied with tidal conditions. At high tide the effluent from the FMC holding pond veered left on entering the embayment and described the path shown in Figure 31 . As the water level fell with the ebbing tide the path of the water became distributed between two paths (see Figure 31) until at low tide the water effluent was carried away exclusively by apath to the right of the outfall.

Using a 12 foot Canova boat as a working platform, eight-foot wooden markers were driven into the sediment for site identification during high slack water. Water, salinity, particle size distribution and sediment core samples were collected as conditions permitted at the times shown on Figure 32 . Water samples were collected as follows: acid-cleaned teflon tubing, taped two inches from the end of a ten-foot wooden pole was placed very carefully to the water-sediment interface. This ensured that a water sample unaffected by disturbed sediment was being obtained. The water was then pumped into 500 ml glass sampling bottles using a portable peristaltic pump. To each sample for total mercury, ten millilitres of a 0.5% (w/v) $K_2Cr_2O_7$ solution in mercury-free ACS nitric acid was added. Sediment cores were obtained in duplicate by driving 12 inch long, 3 cm in diameter acrylic tubes into the sediment and

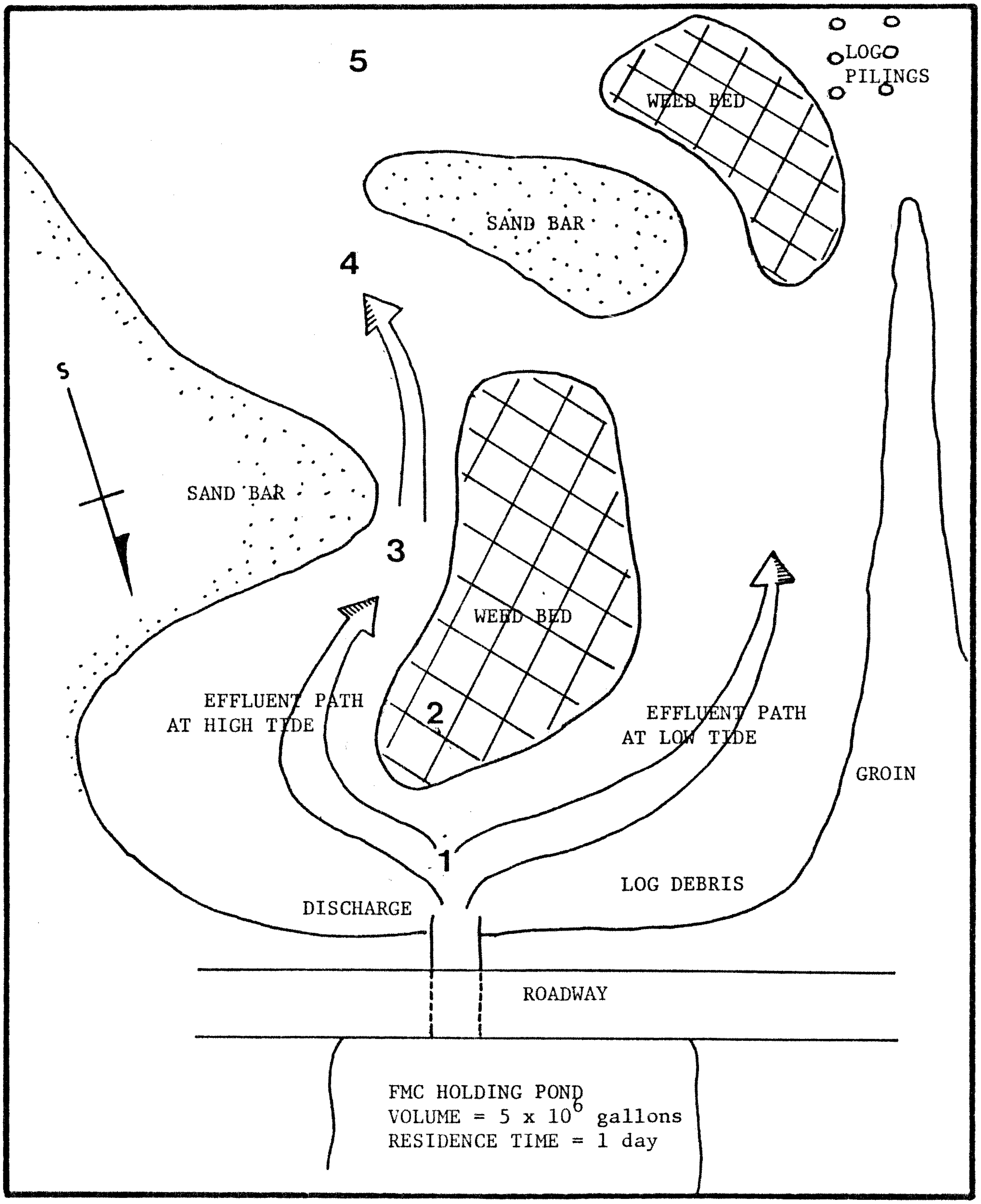


FIGURE 31. Sketch map (not to scale) showing the study area and main topographical features.

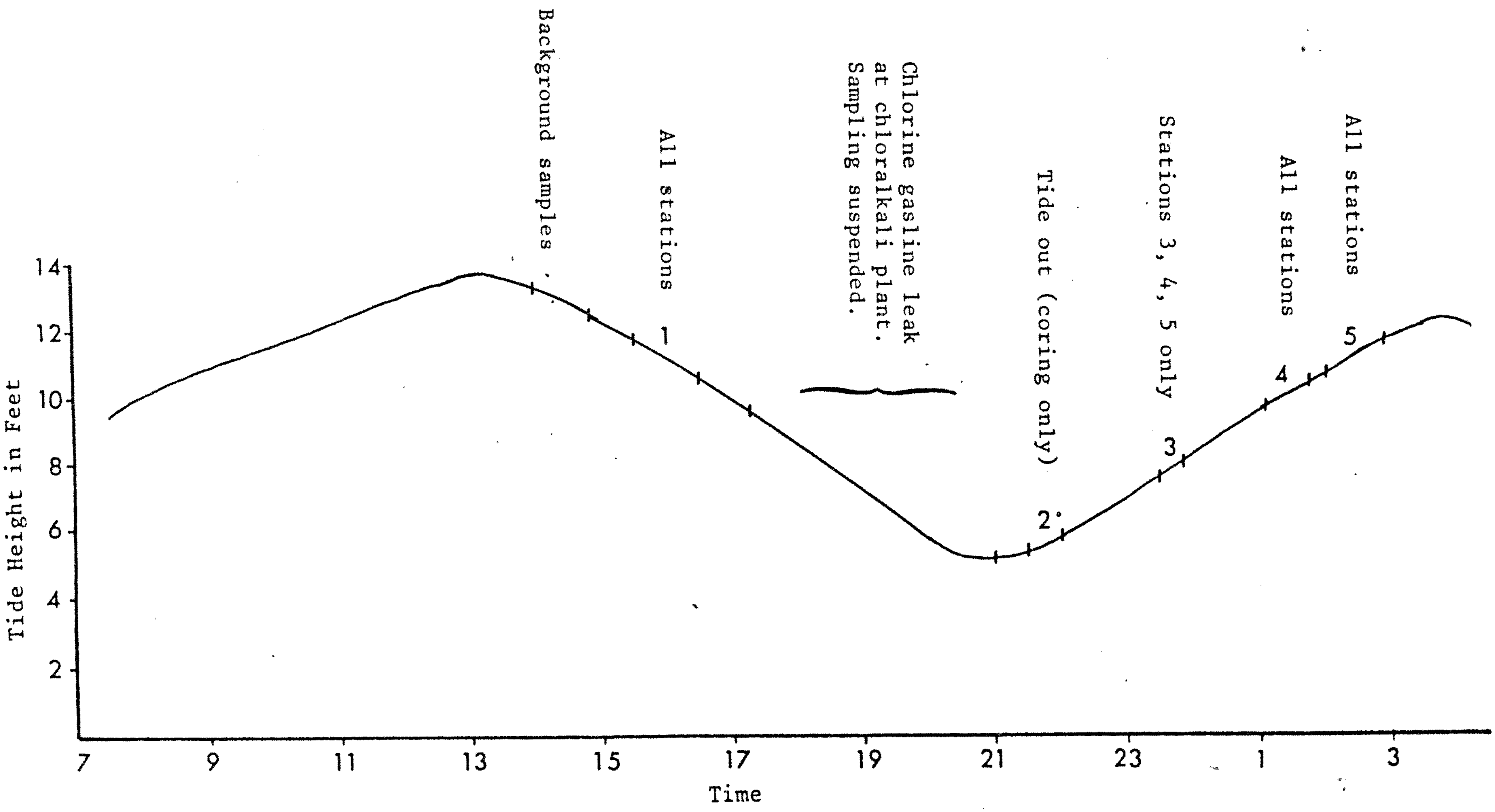


FIGURE 32. In situ mercury mobilization sampling programme in relation to tidal cycle December 1 - December 2, 1976. Numbers on curve refer to sampling periods.

withdrawing. One core was frozen immediately, the other was used for the extraction of interstitial waters.

Water samples were filtered through 0.4 µm pre-weighed Nuclepore membrane filters which had been prewashed with 0.1 N HNO_3. To the filtrate (500 ml) was added 10 ml 0.5% (w/v) $K_2Cr_2O_7/HNO_3$ solution.

Interstitial waters were extracted under a nitrogen pressure of 2 to 5 atmospheres using a nylon squeezer similar to the one described by Reeburgh (1967). About 20 minutes of squeezing yielded 50-100 ml of pore water which was stored by adding 5 ml $K_2Cr_2O_7/HNO_3$ solution to the 150 ml collection flask.

3. ANALYTICAL METHODS

Mercury in water, sediment and particulates was determined by methods described in Part A, 3.

4. RESULTS AND DISCUSSION

The data for the in situ mercury mobilization study appear in Appendix B1. Figure 33 shows the variation of dissolved mercury over a tidal cycle. At each site, the dissolved mercury levels were generally lowest at low water and, as the tide flooded there was a substantial (about 40%) short-lived increase. This trend, although based on limited data, is similar to the one observed by Duinker (1976). The interstitial waters, by contrast, behave oppositely being at a maximum during low tide and minimum during high tide which is a consequence of dilution. These data, however, may be in error because of possible mercury scavenging that occurred when iron hydroxide precipitated during the squeezing of several cores.

The dissolved mercury levels around the outfall at FMC were, at slack water, approximately ten times Howe Sound background values (~ 15 ng/l). During a flood tide this increased to about twenty times Howe Sound background. Assuming that (a) most of the mercury mobilized out of the tidal flat area is dissolved mercury (b) the sediments of the entire tidal flat

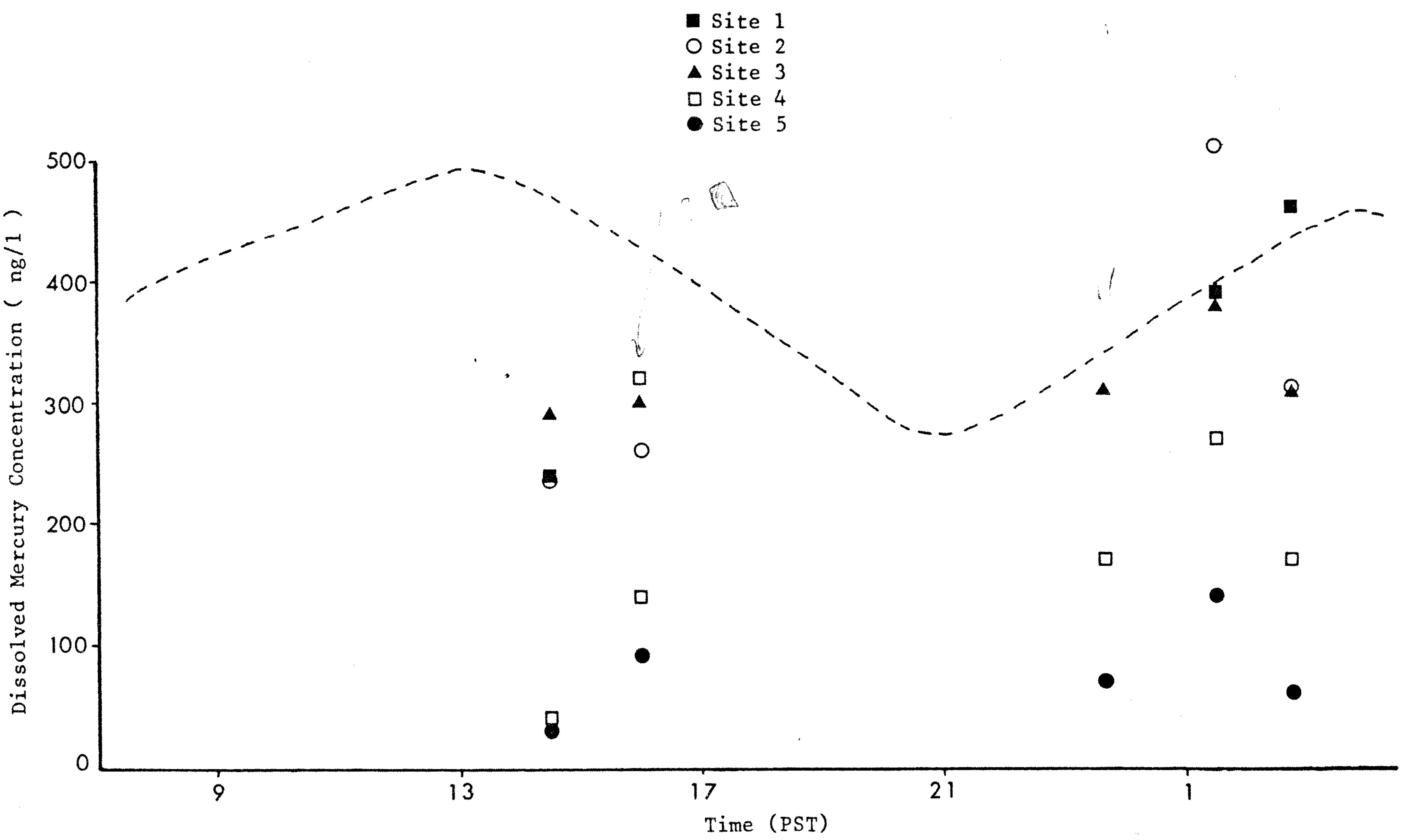


FIGURE 33. Dissolved Mercury in waters of Squamish River Tidal Flats off FMC Plant over a tidal cycle December 1 - December 2, 1976.

area contain about 1.0 ppm Hg in the top 10 cm (c) all the sediments in the tidal flats behave similarly to those for the five sites studied here and (d) on the average the sediments were covered by about 2 meters of water during each tidal cycle, then the observed released (~ 100 ng/l) corresponds to a half-life of mercury in the sediments of about 500 days. Although this is an extremely crude estimate, it does compare favourably with the half-life for mercury release from (a) Ottawa River sediments about one year (Waslenchuk, 1975) and (b) Bellingham Bay sediments of about 1.5 years (Carpenter, 1973). It is, therefore, possible that the mobilization of mercury out of the tidal flat area is driven mainly by the tidal oscillation.

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APPENDICES

Appendix A·1

POSITIONS OF WATER COLLECTION STATIONS

CRUISE	LAT (°N)	LONG (°N)	WATER COLLECTED
OC-76-IS-008	49°22.3'	123°30.6'	Seawater
	49°40.9'	123°10.8'	Freshwater, Estuarine water
OC-77-IS-002	49°26.5'	123°59.6'	Seawater
	49°40.9'	123°10.6'	Estuarine water

Appendix A.2

Dissolved Organic Carbon Content of Water's Used in Experiments

Salinity ($^{o}/oo$)	D.O.C. (mg/1)
5.2	0.7
6.7	0.9
7.6	0.9
14.9	1.5
16.0	1.8
16.3	1.9
16.4	2.0
16.9	2.3
9.6	1.6
23.3	1.2
25.2	1.0
28.8	0.6
29.5	0.5

Appendix A.3

Data for Release Experiments

Experiment No. 1

Initial Conditions			
	1. Water:	Salinity	16.0 ± 0.1^{o}/oo
		pH	3.95 ± .05
		Volume used	30 ± 0.5 1
		Poison	Nil
	2. Sediment:	type	
		amount added	Nil

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	3.85	17.40	40	
0h	3.80	17.35	41	
1h	3.80	17.75	41	
2h	3.90	17.40	40	
3h	3.95	17.65	34	
6h	4.05	17.80	33	
12h	4.10	17.80	37	
1d	3.85	17.85	31	
2d	3.90	17.95	40	
3d	4.00	18.10	42	
4d	4.00	18.30	39	
5d	3.90	18.05	40	

Experiment No. 1

Initial Conditions	1. Water:	Salinity	16.5 ± 0.1 $^{o}/oo$
		pH	7.80 ± 0.05
		Volume used	30 ± 0.5 1
		Poison	Nil
	2. Sediment:	type	
		amount added	Nil

Sampling time	pH	T(^{o}C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	7.85	17.50	31	
0h	7.80	17.55	27	
1h	7.75	17.80	30	
2h	7.75	17.55	41	
3h	7.75	17.80	26	
6h	7.85	17.85	26	
12h	7.90	17.90	27	
1d	7.75	18.10	35	
2d	7.80	18.15	29	
3d	7.80	18.40	27	
4d	7.85	18.00	24	
5d	7.75	18.10	24	

Experiment No. 2

Initial Conditions	1.	Water:	Salinity	6.7 ± 0.1^{o}/oo
			pH	7.70 ± 0.05
			Volume used	30 ± 0.5 l
			Poison	Nil
	2.	Sediment:	type	Core #8, 0-2 cm
			amount added	0.855 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	7.60	17.00	38	
0h	7.65	17.00	53	526
2h	7.75	17.10	57	667
4h	7.50	17.10	51	456
6h	7.50	17.25	27	-386
12h	7.55	17.10	31	-246
1d	7.60	17.20	37	- 35
2d	7.60	17.25	32	-211
3d	7.65	17.10	27	-386
4d	7.50	17.90	28	-351
5d	7.60	18.25	24	-491

Experiment No. 2

Initial Conditions	1. Water:	Salinity	16.3 ± 0.1^{o}/oo
		pH	7.50 ± 0.05
		Volume used	30 ± 0.5 1
		Poison	Nil
	2. Sediment:	type	Core #8, 0-2 cm
		amount added	0.968 g

Sampling time	pH	T(^{o}C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	7.45	17.95	45	
0h	7.40	18.00	66	651
2h	7.45	18.00	64	589
4h	7.45	18.05	61	496
6h	7.55	18.00	56	341
12h	7.60	18.30	48	93
1d	7.65	18.40	53	248
2d	7.50	18.20	52	217
3d	7.50	18.00	51	186
4d	7.55	18.80	49	124
5d	7.50	18.95	49	124

Experiment No. 2

Initial Conditions	1. Water:	Salinity	16.3 ± 0.1‰
		pH	7.50 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	2.0 g $CuSO_4$
	2. Sediment:	type	Core #8, 0-2 cm
		amount added	1.047 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	7.50	17.25	47	
0h	7.55	17.25	56	258
2h	7.55	17.40	48	29
4h	7.65	17.75	67	573
6h	7.40	17.70	37	-287
12h	7.40	17.50	34	-372
1d	7.45	17.80	48	29
2d	7.55	17.80	25	-630
3d	7.60	18.15	28	-544
4d	7.50	18.00	32	-430
5d	7.50	18.05	39	-229

Experiment No. 2

Initial Conditions	1. Water:	Salinity	29.5 ± 0.1°/oo
		pH	7.65 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	Nil
	2. Sediment:	type	Core #8, 0-2 cm
		amount added	0.903 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	7.55	17.00	24	
0h	7.60	17.05	39	166
2h	7.60	17.00	57	1096
4h	7.65	17.10	43	631
6h	7.70	17.15	34	332
12h	7.70	17.10	30	199
1d	7.70	17.20	37	432
2d	7.50	17.05	24	0
3d	7.55	17.00	27	100
4d	7.60	18.10	27	100
5d	7.70	18.55	27	100

Experiment No. 3

Initial Conditions	1. Water:	Salinity	6.7 ± 0.1°/oo
		pH	7.65 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	Nil
	2. Sediment:	type	Core #8, 0-2 cm
		amount added	0.909 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	7.65	17.45	25	
0h	7.75	17.50	23	-44
1h	7.50	17.50	33	267
3h	7.50	17.45	45	667
6h	7.55	17.30	29	133
12h	7.60	17.90	28	100
1d	7.65	18.35	28	100
2d	7.70	18.40	26	33
3d	7.70	17.95	29	133
4d	7.65	18.00	41*	
5d	7.65	17.80	22	-100

*Sample contaminated

Experiment No. 3

Initial Conditions	1. **Water:**	**Salinity**	16.3 ± 0.1°/oo
		pH	7.65 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	Nil
	2. **Sediment:**	**type**	Core #8, 0-2 cm
		amount added	0.951

Sampling time	**pH**	**T(°C)**	**Corrected Hg conc (ng/l)**	**Released Hg (ng) per g sediment**
Background	7.65	17.80	22	
0h	7.70	17.85	22	0
1h	7.70	17.85	38	505
3h	7.75	17.85	41	599
6h	7.55	17.80	26	126
12h	7.55	17.95	22	0
1d	7.60	18.00	25	95
2d	7.60	18.05	24	63
3d	7.60	18.50	26	126
4d	7.60	18.00	37	473
5d	7.60	18.05	27	158

Experiment No. 3

Initial Conditions	1. Water:	Salinity	16.3 ± 0.1°/oo
		pH	7.50 ± 0.05
		Volume used	30 ± 0.5 1
		Poison	2.0 g $CuSO_4$
	2. Sediment:	type	Core #8, 0-2 cm
		amount added	1.00 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	7.50	17.20	25	
0h	7.50	17.30	31	180
1h	7.50	17.40	47	660
3h	7.50	17.45	41	480
6h	7.60	17.45	34	270
12h	7.65	17.70	31	180
1d	7.45	17.80	34	270
2d	7.50	17.95	40	450
3d	7.55	18.20	32	210
4d	7.55	18.35	38	390
5d	7.50	18.25	42	510

Experiment No. 3

Initial Conditions	1.	Water:	Salinity	16.4 ± 0.1°/oo
			pH	7.45 ± 0.05
			Volume used	30 ± 0.5 l
			Poison	Nil
	2.	Sediment:	type	
			amount added	Nil

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	7.45	17.80	21	
0h	7.55	17.80	32	
1h	7.60	17.85	30	
3h	7.40	17.95	34	
6h	7.40	18.05	29	
12h	7.45	18.00	36	
1d	7.50	18.35	35	
2d	7.50	18.45	31	
3d	7.40	18.35	38	
4d	7.45	18.30	38	
5d	7.45	18.25	36	

Experiment No. 3

Initial Conditions	1. **Water:**	**Salinity**	29.5°/oo
		pH	7.60 ± 0.05
		Volume used	30 ± 0.5 1
		Poison	Nil
	2. **Sediment:**	**type**	Core #8, 0-2 cm
		amount added	0.910 g

Sampling time	**pH**	**T(°C)**	**Corrected Hg conc (ng/1)**	**Released Hg (ng) per g sediment**
Background	7.70	17.15	24	
0h	7.70	17.25	27	99
1h	7.55	17.20	47	758
3h	7.60	17.20	58	1123
6h	7.60	17.15	35	363
12h	7.60	17.00	37	429
1d	7.60	18.10	35	363
2d	7.60	16.65	36	396
3d	7.60	17.80	76*	
4d	7.65	17.75	37	429
5d	7.60	17.90	36	396

*Contaminated sample

Experiment No. 3

Initial Conditions	1.	Water:	Salinity	29.5 ± 0.1°/oo
			pH	7.70 ± 0.05
			Volume used	30 ± 0.5 1
			Poison	Nil
	2.	Sediment:	type	
			amount added	Nil

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	7.70	17.80	37	
0h	7.70	17.40	37	
1h	7.70	17.45	34	
3h	7.80	17.55	34	
6h	7.65	17.85	29	
12h	7.65	17.95	35	
1d	7.70	17.80	31	
2d	7.70	17.90	38	
3d	7.70	18.20	99*	
4d	7.70	18.15	38	
5d	7.70	18.15	34	

* Contaminated sample

Experiment No. 4

Initial Conditions:	Tap Water Blank	17 ng/1
	Nuclepore Filter Blank	2 ng/1
	Seawater Salinity	$29.4 \pm 0.1^{o}/oo$
	Estuarine Salinity	$14.9 \pm 0.1^{o}/oo$

Time	SEAWATER Dissolved Hg (ng/1)	SEAWATER Particulate Hg (ng/1)	ESTUARINE Dissolved Hg (ng/1)	ESTUARINE Particulate Hg (ng/1)	ESTUARINE BLANK Dissolved Hg (ng/1)	ESTUARINE BLANK Particulate Hg (ng/1)
Total Background			33		38	
Filtered Background	26	33	23	31	23	31
Background No Sed.	20	17	21	19	23	32
0h	28	128	28	149	29	26
0.5h	Lost	135	40	136	36	21
1h	72	142	26	146	26	17
2h	90	122	38	123	26	32
3h	54	156	24	127	23	20
4h	30	171	32	126	28	26
5h	27	163	31	163	26	19
6h	21	165	29	135	34	21
10h	20	165	25	161	26	26
24h	30	175	30	158	29	20

Experiment No. 5

Initial Conditions	1. **Water:**	**Salinity**	16.8 ± 0.1 °/oo
		pH	7.45 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	0.2 g Cu SO_4
	2. **Sediment:**	**type**	Core #6, 6-8 cm
		amount added	6.45 g

Sampling time	**pH**	**T(°C)**	**Corrected Hg conc (ng/l)**	**Released Hg (ng) per g sediment**
Background	7.45	17.45	30.2	
0h	7.35	17.85	68.6	179
1h	7.35	17.85	62.1	148
2h	7.40	18.10	87.7	267
4h	7.40	17.85	86.7	263
8h	7.40	17.60	68.1	176
12h	7.55	17.95	84.2	251
18h	7.45	17.95	73.4	201
24h	7.45	18.10	77.9	222
1.5d	7.40	18.80	72.6	197
2d	7.50	18.75	73.8	203

Experiment No. 5

Initial Conditions	1.	**Water:**	**Salinity**	16.5 ± 0.1 ^{o}/oo
			pH	7.55 ± 0.05
			Volume used	30 ± 0.5 1
			Poison	2.0 g $CuSO_4$
	2.	**Sediment:**	**type**	Core #6, 6-8 cm
			amount added	7.02

Sampling time	**pH**	**T(^{o}C)**	**Corrected Hg conc (ng/1)**	**Released Hg (ng) per g sediment**
Background	7.55	18.10	38.1	
0h	7.60	18.35	75.8	238
1h	7.45	18.20	61.3	99
2h	7.45	18.45	Lost	
8h	7.50	18.30	101.8	271
18h	7.50	17.95	77.2	167
1d	7.60	17.80	74.5	156
1.5d	7.50	18.10	82.9*	191
2.5d	7.55	18.25	161.8*	529
4d	7.60	18.40	124.6*	370
5d	7.60	18.05	126.6*	378

*Brown precipitate in samples

Experiment No. 5

Initial Conditions	1. Water:	Salinity	16.7 ± 0.1°/oo
		pH	7.45 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	0.2 g $CuSO_4$
	2. Sediment:	type	Core #8, 0-2 cm
		amount added	1.00 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	7.45	17.80	43.8	
0h	7.40	18.45	54.0	306
1h	7.45	18.40	81.3	1125
2h	7.50	18.30	95.7	1557
8h	7.55	18.35	65.9	663
18h	7.65	18.15	60.2	492
24h	7.40	18.20	46.7	87
1.5d	7.40	18.00	66.5	681
2.5d	7.40	17.65	52.5	261
4d	7.45	17.90	64.0	606
5d	7.40	18.10	66.2	672

Experiment No. 5

Initial Conditions	1. **Water:**	**Salinity**	16.6 ± 0.1°/oo
		pH	7.60 ± 0.05
		Volume used	30 ± 0.5 1
		Poison	2.0 g $CuSO_4$
	2. **Sediment:**	**type**	Core #8, 0-2 cm
		amount added	1.05 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	7.60	17.90	30.4	
0h	7.55	18.35	41.5	317
1h	7.45	18.30	75.7	1294
2h	7.55	18.20	92.9	1786
8h	7.55	18.25	91.5	1746
18h	7.60	18.45	60.9	871
1d	7.65	17.95	62.9	929
1.5d	7.55	18.10	61.1	877
2.5d	7.45	18.05	313.3*	
4d	7.60	18.35	288.8*	
5d	7.60	18.45	268.6*	

* Contaminated sample

Experiment No. 5

Initial Conditions	1. **Water:**	**Salinity**	16.9 ± 0.1°/oo
		pH	7.60 ± 0.05
		Volume used	30 ± 0.5 1
		Poison	2.0 g $CuSO_4$
	2. **Sediment:**	**type**	
		amount added	Nil

Sampling time	**pH**	**T(°C)**	**Corrected Hg conc (ng/l)**	**Released Hg (ng) per g sediment**
Background	7.60	17.65	40.7	
0h	7.55	17.65	27.8	
1h	7.55	17.95	24.2	
2h	7.60	17.70	29.3	
8h	7.65	17.80	28.4	
18h	7.70	17.80	95.1*	
1d	7.55	17.90	24.1	
1.5d	7.55	17.90	26.5	
2.5d	7.60	18.15	33.3	
4d	7.60	18.40	27.1	
5d	7.60	18.90	28.9	

* Contaminated sample

Experiment No. 5

Initial Conditions	**1. Water:**	**Salinity**	$16.9 \pm 0.1^{o}/oo$
		pH	7.65 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	0.2 g $CuSO_4$
	2. Sediment:	**type**	
		amount added	Nil

Sampling time	**pH**	**T(°C)**	**Corrected Hg conc (ng/l)**	**Released Hg (ng) per g sediment**
Background	7.65	17.10	22.5	
0h	7.75	17.80	23.5	
1h	7.80	17.85	27.3	
2h	7.50	17.85	30.2	
8h	7.50	17.60	24.1	
18h	7.55	17.70	28.0	
1d	7.60	17.80	30.6	
1.5d	7.60	17.95	27.6	
2.5d	7.65	17.65	37.7	
4d	7.55	18.10	30.5	
5d	7.65	18.60	27.5	

Experiment No. 6

Initial Conditions	1. Water:	Salinity	5.2 ± 0.1°/oo
		pH	4.00 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	Nil
	2. Sediment:	type	Core #8 0-2 cm
		amount added	1.39 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	4.05	6.65	23.0	
0h	4.00	6.20	49.0	561
1h	4.00	6.00	64.8	902
2h	3.95	6.00	53.0	648
5h	4.00	5.80	81.5	1759
8h	4.05	5.75	49.0	561
12h	4.00	5.30	36.5	291
1d	4.00	5.75	33.5	723
2d	4.05	5.65	33.1	218
3d	4.00	6.00	30.6	164
4d	4.05	5.80	29.3	136
5d	4.00	5.80	30.9	171

Experiment No. 6

Initial Conditions	1.	Water:	Salinity	5.2 ± 0.1°/oo
			pH	5.90 ± 0.05
			Volume used	30 ± 0.5 1
			Poison	Nil
	2.	Sediment:	type	Core #8 0-2 cm
			amount added	1.00 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	5.90	19.30	17.0	
0h	5.80	19.90	32.6	468
1h	5.95	18.95	33.4	492
2h	5.90	20.00	28.9	357
5h	5.90	20.25	37.3	609
8h	5.95	21.45	32.3	459
12h	5.80	20.65	32.3	459
1d	5.95	20.10	27.8	324
2d	6.00	20.15	18.8	54
3d	5.90	20.45	17.5	15
4d	5.90	20.70	18.4	42
5d	5.90	20.10	18.0	30

Experiment No. 6

Initial Conditions	1. Water:	Salinity	5.2 ± 0.1^{o}/oo
		pH	7.00 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	Nil
	2. Sediment:	type	Core #8 0-2 cm
		amount added	1.05 g

Sampling time	pH	T(^{o}C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	6.90	19.55	11.5	
0h	6.95	19.75	25.1	389
1h	6.80	19.20	28.9	497
2h	6.95	18.90	26.9	440
5h	7.05	20.60	30.6	546
8h	7.00	21.50	28.6	489
12h	6.90	20.90	22.3	309
1d	7.00	20.05	28.8	494
2d	7.00	19.00	22.4	311
3d	7.05	19.85	13.6	60
4d	7.10	19.55	13.3	51
5d	7.00	19.15	13.8	66

Experiment No. 6

Initial Conditions	1.	**Water:**	**Salinity**	5.2 ± $0.1^{o}/_{oo}$
			pH	8.00 ± 0.05
			Volume used	30 ± 0.5 l
			Poison	Nil
	2.	**Sediment:**	**type**	Core #8, 0-2 cm
			amount added	1.15 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	8.05	19.30	17.5	
0h	8.00	19.15	28.3	281
1h	8.00	19.05	41.3	621
2h	7.95	19.30	41.3	621
5h	8.10	21.20	32.5	391
8h	8.05	22.00	34.5	444
12h	8.10	20.15	24.5	183
24h	8.00	19.85	25.3	203
2d	8.05	18.95	28.1	277
3d	8.00	19.75	26.0	222
4d	8.00	20.15	25.7	214
5d	8.00	19.60	24.9	193

Experiment No. 6

Initial Conditions	1.	Water:	Salinity	5.2 ± 1°/oo
			pH	8.95 ± 0.05
			Volume used	30 ± 0.5 1
			Poison	Nil
	2.	Sediment:	type	Core #8 0-2 cm
			amount added	1.19 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	8.95	19.5	15.0	
0h	8.80	19.0	20.5	139
1h	9.00	19.0	17.9	73
2h	8.85	19.5	16.7	43
5h	8.90	19.9	16.5	38
8h	8.90	20.1	13.0	-50
12h	8.90	20.9	113.0*	
1d	8.90	20.5	20.7	144
2d	8.90	20.0	11.1	-98
3d	8.90	19.4	9.9	-129
4d	8.90	20.0	10.6	-111
5d	8.90	20.0	11.0	-101

*sample contaminated

Experiment No. 6

Initial Conditions	1. Water:	Salinity	5.2 ± 0.1°/oo
		pH	7.70 ± 0.05
		Volume used	30 ± 0.5 1
		Poison	Nil
	2. Sediment:	type	
		amount added	Nil

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	7.65	18.90	17.0	
0h	7.70	19.45	21.7	
1h	7.70	19.60	19.1	
2h	7.70	19.55	22.6	
5h	7.65	20.95	16.1	
8h	7.75	21.10	16.1	
12h	7.75	19.90	19.0	
1d	7.70	19.90	13.7	
2d	7.65	19.65	15.8	
3d	7.65	19.50	15.3	
4d	7.70	19.95	16.0	
5d	7.70	20.05	16.9	

Experiment No. 7

Initial Conditions	1. Water:	Salinity	16.2 ± 0.1°/oo
		pH	3.90 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	Nil
	2. Sediment:	type	Core #8, 0-2 cm
		amount added	1.03 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	3.95	6.40	25.8	
0h	3.90	6.35	54.2	827
1h	4.00	6.25	95.5	2030
2h	4.00	6.00	100.0	2161
4h	3.85	5.90	80.0	1579
5h	3.90	5.85	67.6	1217
7h	3.90	6.15	73.7	1395
12h	3.95	6.50	72.3	1354
18h	4.00	6.45	55.4	862
1d	4.00	6.45	40.7	434
2d	4.00	6.45	58.8	961
3d	4.00	6.45	38.9	382
4d	4.05	6.30	41.8	466
5d	3.95	6.05	42.5	486

Experiment No. 7

Initial Conditions	1. Water:	Salinity	16.2 ± 0.1 °/oo
		pH	4.10 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	Nil
	2. Sediment:	type	Core #8, 0-2 cm
		amount added	1.09 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	4.05	6.55	27.1	
0h	4.10	6.50	57.1	826
1h	4.10	6.75	75.7	1338
2h	4.05	6.40	55.2	773
4h	4.05	6.30	50.4	641
5h	4.10	6.15	44.0	465
7h	4.00	6.00	50.2	636
12h	4.00	5.70	44.2	471
18h	4.00	5.95	40.7	374
1d	4.00	6.40	39.3	336
2d	4.00	6.35	43.3	446
3d	4.00	6.25	42.5	424
4d	4.00	6.10	38.9	325
5d	4.00	6.40	44.5	479

Experiment No. 7

Initial Conditions	1. Water:	Salinity	16.0 ± 0.1°/oo
		pH	8.90 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	Nil
	2. Sediment:	type	Core #8, 0-2 cm
		amount added	1.02 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	8.95	6.40	25.2	
0h	8.90	6.35	35.9	315
1h	9.00	6.10	48.1	674
2h	9.10	5.95	58.3	974
4h	8.85	5.95	64.9	1168
5h	8.90	5.80	60.0	1024
7h	8.90	6.40	48.1	674
12h	9.00	6.50	39.2	412
18h	9.00	6.85	64.6	1159
1d	9.10	5.90	30.8	165
2d*	8.95	6.15	43.3	532
3d	8.95	6.30	41.3	474
4d	8.90	6.40	34.2	265
5d	9.10	6.35	22.3	-85

*stir rod and drill replaced

Experiment No. 7

Initial Conditions	1. Water:	Salinity	25.3 ± 0.1°/oo
		pH	3.90 ± 0.05
		Volume used	30 ± 0.5 1
		Poison	Nil
	2. Sediment:	type	Core #8, 0-2 cm
		amount added	1.18 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	3.85	6.45	38.2	
0h	3.95	6.75	62.5	618
1h	3.80	6.80	129.9	2331
2h	3.90	6.40	Lost	
4h	4.00	6.30	140.8	2608
5h	4.00	6.15	134.6	2450
7h	3.90	5.90	Lost	
12h	3.85	5.80	164.9	3221
18h	3.85	6.05	135.8	2481
1d	3.95	6.35	63.6	646
2d	3.90	6.00	82.3	1121
3d	4.00	6.20	57.1	481
4d	4.00	6.10	55.7	445
5d	3.95	6.15	55.7	445

Experiment No. 7

Initial Conditions	1.	Water:	Salinity	25.3 ± 0.1°/oo
			pH	6.00 ± 0.05
			Volume used	30 ± 0.5 1
			Poison	Nil
	2.	Sediment:	type	Core #8, 0-2 cm
			amount added	1.00 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	6.00	6.70	39.6	
0h	6.00	6.80	118.2	2358
1h	6.00	6.80	Lost	
2h	6.00	6.70	103.3	1911
4h	6.05	6.50	86.9	1419
5h	6.00	6.20	82.8	1296
7h	6.00	6.10	69.6	900
12h	5.95	6.30	72.8	996
18h	5.90	6.75	76.9	1119
1d	6.00	5.95	48.7	273
2d	6.00	6.10	58.8	576
3d	6.00	6.20	47.7	243
4d	6.00	6.45	44.2	138

Experiment No. 7

Initial Conditions			
1.	Water:	Salinity	25.6 ± 0.1°/oo
		pH	6.90 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	Nil
2.	Sediment:	type	Core #8, 0-2 cm
		amount added	1.45 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	7.00	6.50	44.4	
0h	6.90	6.60	79.8	732
1h	6.95	6.75	68.5	499
2h	7.00	6.20	109.9	1355
4h	7.00	6.10	99.5	1140
5h	7.00	6.25	77.7	689
7h	7.00	6.40	85.4	848
12h	6.95	6.80	78.2	699
18h	7.00	6.35	66.7	461
1d	7.10	6.45	48.5	85
2d	7.00	6.30	53.4	186
3d	7.05	6.20	44.2	-4.
4d	7.00	6.35	44.6	4
5d	6.90	6.00	44.1	-6

Experiment No. 7

Initial Conditions	1. Water:	Salinity	25.1 ± 0.1 °/oo
		pH	8.05 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	Nil
	2. Sediment:	type	Core #8, 0-2 cm
		amount added	1.14 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	8.10	6.55	45.5	
0h	8.00	6.40	51.3	153
1h	8.00	6.30	39.2	-166
2h	8.05	6.15	87.6	1108
4h	8.00	5.90	76.5	816
5h	7.90	6.40	66.0	539
7h	7.95	6.70	54.4	234
12h	7.95	6.90	66.7	558
18h	7.95	6.75	41.2	-113
1d	7.95	6.85	37.4	-213
2d	8.15	6.30	33.3	-321
3d	8.10	6.45	36.8	-229
4d	8.10	6.10	33.0	-329
5d	8.00	6.25	32.6	-339.

Experiment No. 7

Initial Conditions	1. Water:	Salinity	25.2 ± 0.1°/oo
		pH	9.00 ± 0.05
		Volume used	30 ± 0.5 1
		Poison	Nil
	2. Sediment:	type	Core #8, 0-2 cm
		amount added	1.00 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	8.85	6.50	40.5	
0h	8.80	6.30	51.6	333
1h	8.95	6.15	Lost	
2h	9.05	5.95	53.0	375
4h	9.00	6.10	52.3	354
5h	9.05	5.80	68.4	837
7h	9.10	6.15	73.8	999
12h	9.10	6.20	51.2	321
18h	9.10	6.30	58.1	528
1d	8.95	6.35	27.3	-396
2d	8.90	6.40	28.7	-354
3d	8.95	6.55	24.7	-474
4d	9.00	6.60	23.5	-510
5d	9.10	6.35	24.8	-471

Experiment No. 8

Initial Conditions	1. Water:	Salinity	4.5 ± 0.1°/oo
		pH	4.00 ± 0.05
		Volume used	30 ± 0.5 1
		Poison	Nil
	2. Sediment:	type	#2 0-2 cm
		amount added	7.97 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Amount Hg released/g sed.
Background	4.00	6.40	30.8	
0h	4.00	6.30	93.0	234.1
1h	4.00	6.25	193.4	612.0
3h	4.00	6.35	79.7	184.1
7h	4.00	6.70	74.9	166.0
12h	4.00	6.60	64.0	125.0
1d	4.00	6.75	47.7	63.6
2d	4.00	6.80	45.1	53.8
3d	4.00	6.40	42.2	42.9
4d	4.00	6.50	44.6	51.9
5d	4.00	6.30	48.3	65.9

Experiment No. 8

Initial Conditions	1. Water:	Salinity	4.5 ± 0.1°/oo
		pH	6.00 ± 0.05
		Volume used	30 ± 0.5 1
		Poison	Nil
	2. Sediment:	type	#2 0-2 cm
		amount added	8.77 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Amount Hg released/g sed.
Background	6.05	6.90	30.7	
0h	6.00	6.85	43.9	45.2
1h	6.15	6.40	97.2	227.5
3h	6.00	6.20	137.5	365.3
7h	5.95	6.60	160.9	445.4
12h	5.90	6.70	104.6	252.8
1d	5.95	6.30	62.3	108.1
2d	6.00	6.80	26.9	-13.0
3d	6.00	6.70	26.7	-13.7
4d	6.00	6.50	26.6	-14.0
5d	6.00	6.40	27.1	-12.3

Experiment No. 8

Initial Conditions	1. Water:	Salinity	4.5 ± 0.1°/oo
		pH	6.80 ± 0.05
		Volume used	30 ± 0.5 l
		Poison	Nil
	2. Sediment:	type	#2 0-2 cm
		amount added	8.47 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Amount Hg released/g sed.
Background	6.90	6.85	28.7	
0h	6.85	6.15	88.2	210.7
1h	6.95	5.95	98.3	246.5
3h	6.90	6.80	65.2	129.3
7h	6.90	6.70	46.4	62.7
12h	6.95	6.50	48.1	68.7
1d	6.90	6.75	23.2	-19.5
2d	6.95	6.70	32.4	13.1
3d	7.10	6.40	31.5	9.9
4d	6.95	6.60	29.0	1.1
5d	6.85	6.30	30.0	4.6

Experiment No. 8

Initial Conditions	1. Water:	Salinity	4.5 ± 0.1°/oo
		pH	7.95 ± 0.05
		Volume used	31 ± 0.5 1
		Poison	Nil
	2. Sediment:	type	#2 0-2 cm
		amount added	7.75 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Amount Hg released/g sed.
Background	7.85	6.85	28.2	
0h	7.95	6.80	33.3	20.4
1h	8.00	6.40	58.8	122.4
3h	8.05	6.90	76.2	192.0
7h	7.95	6.85	50.9	90.8
12h	7.90	6.80	50.9	90.8
1d	7.95	6.70	27.5	-2.8
2d	7.90	6.90	32.4	16.8
3d	7.90	6.95	33.4	20.8
4d	7.95	6.40	27.7	-2.0
5d	7.90	6.60	79.0*	

* Contaminated sample

Experiment No. 8

Initial Conditions	1.	Water:	Salinity	4.5 ± 0.1°/oo
			pH	9.00 ± 0.05
			Volume used	30 ± 0.5 l
			Poison	Nil
	2.	Sediment:	type	#2 0-2 cm
			amount added	7.89 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Amount Hg released/g sed.
Background	9.10	6.90	29.9	
0h	9.00	6.70	81.3	195.4
1h	8.80	6.70	85.1	210.0
3h	8.95	6.75	50.2	77.2
7h	9.10	6.70	75.9	174.9
12h	9.15	6.80	50.9	79.8
1d	9.00	6.40	47.0	79.8
2d	9.15	6.30	40.8	65.0
3d	9.00	6.50	33.2	12.5
4d	9.05	6.80	30.6	2.7
5d	9.10	6.20	30.9	3.8

Experiment No. 8

Initial Conditions	1. Water:	Salinity	23.3 ± 0.1°/oo
		pH	3.95 ± 0.05
		Volume used	31 ± 0.5 1
		Poison	Nil
	2. Sediment:	type	#2 0-2 cm
		amount added	7.54 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Amount Hg released/g sed.
Background	4.05	6.90	32.4	
0h	4.00	6.85	66.3	139.4
1h	4.00	6.50	323.5	1196.8
3h	4.15	6.55	123.1	372.9
7h	3.90	6.50	96.5	263.5
12h	3.95	6.80	86.7	223.2
1d	3.95	7.00	51.3	77.7
2d	3.95	6.40	57.7	104.0
3d	4.00	6.90	48.3	65.4
4d	4.00	6.80	63.4	127.5
5d	3.90	6.85	56.9	97.5

Experiment No. 8

Initial Conditions	1. Water:	Salinity	23.3 ± 0.1°/oo
		pH	6.00 ± 0.05
		Volume used	30 ± 0.5 1
		Poison	Nil
	2. Sediment:	type	#2 0-2 cm
		amount added	8.64 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Amount Hg released/g sed.
Background	6.00	6.05	37.8	
0h	6.05	6.15	99.0	212.5
1h	6.00	6.20	157.1	414.2
3h	6.00	6.30	78.5	141.3
7h	6.00	6.00	79.4	144.4
12h	6.05	6.90	65.5	96.2
1d	5.95	6.85	50.2	43.1
2d	5.90	6.90	45.3	26.0
3d	6.05	6.80	37.8	0
4d	6.00	6.40	41.4	12.5
5d	6.00	6.45	49.1	39.2

Experiment No. 8

Initial Conditions	1.	Water:	Salinity	23.3 ± 0.1°/oo
			pH	7.00 ± 0.05
			Volume used	30 ± 0.5 l
			Poison	Nil
	2.	Sediment:	type	#2 0-2 cm
			amount added	7.70 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Amount Hg released/g sed.
Background	7.00	6.95	28.5	
0h	7.00	6.85	66.4	147.7
1h	7.00	6.40	162.9	523.6
3h	7.00	6.30	115.4	338.6
7h	7.00	6.80	41.6	51.0
12h	6.90	6.80	54.3	100.5
1d	6.90	6.20	27.0	-5.8
2d	7.00	6.10	27.1	-5.5
3d	7.00	6.50	28.0	-1.9
4d	7.00	6.90	34.1	21.8
5d	6.95	6.60	31.1	10.1

Experiment No. 8

Initial Conditions	1.	Water:	Salinity	23.3 ± 0.1°/oo
			pH	8.00 ± 0.05
			Volume used	30 ± 0.5 l
			Poison	Nil
	2.	Sediment:	type	#2 0-2 cm
			amount added	8.06 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Amount Hg released/g sed.
Background	8.00	6.20	31.6	
0h	8.00	6.30	73.6	156.3
1h	8.10	6.40	138.5	397.9
3h	8.05	6.45	72.9	153.7
7h	8.00	6.40	86.2	203.2
12h	8.00	6.10	31.6	0
1d	8.00	6.00	25.6	-22.3
2d	8.00	6.05	27.4	-15.6
3d	7.95	6.35	24.8	-25.3
4d	7.90	6.90	22.6	-33.5
5d	8.05	6.55	26.5	-15.3

Experiment No. 8

Initial Conditions	1.	Water:	Salinity	23.3 ± 0.1°/oo
			pH	9.00 ± 0.05
			Volume used	30 ± 0.5 l
			Poison	Nil
	2.	Sediment:	type	#2 0-2 cm
			amount added	8.53 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Amount Hg released/g sed.
Background	9.10	6.40	34.6	
0h	9.00	6.40	42.2	26.7
1h	8.85	6.30	86.4	182.2
3h	8.95	6.30	16.2	-64.7
7h	8.95	6.35	27.3	-25.7
12h	8.95	5.90	22.6	-42.2
1d	8.95	5.85	23.9	-37.6
2d	8.95	6.80	20.9	-48.2
3d	8.90	7.00	21.4	-46.4
4d	8.95	7.10	23.5	-39.0
5d	8.95	6.75	25.7	-31.3

Experiment No. 8

Initial Conditions	1. Water:	Salinity	23.3 ± 0.1°/oo
		pH	6.90 ± 0.05
		Volume used	30 ± 0.5 1
		Poison	Nil
	2. Sediment:	type	
		amount added	Nil

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Amount Hg released/g sed.
Background	6.95	6.40	38.0	
0h	7.00	6.30	124.7*	
1h	7.05	6.30	203.0*	
3h	7.00	6.80	39.6	
7h	7.00	6.10	52.5	
12h	7.15	6.40	37.1	
1d	6.90	5.95	37.8	
2d	6.95	6.80	38.7	
3d	7.05	6.70	35.2	
4d	7.00	6.80	27.3	
5d	6.95	6.40	39.2	

*Contaminated sample

Experiment No. 9

Initial Conditions	1. Water:	Salinity	7.6 ± 0.1 °/oo
		pH	7.65 ± 0.05
		Volume used	40 ± 0.5
		Poison	Nil
	2. Sediment:	type	Core #1, 2-4 cm
		amount added	11.67 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	7.60	8.95	19.2	
0h	7.60	8.95	17.6	- 5.5
1h	7.65	8.90	22.7	12.0
2h	7.65	8.90	23.4	14.4
4h	7.65	8.85	23.5	14.7
8h	7.60	8.70	26.8	26.0
12h	7.65	8.30	17.8	- 4.8
18h	7.70	7.90	27.4	28.1
24h	7.70	8.95	33.3	48.3
1.5d	7.65	8.90	30.4	38.4
2d	7.65	8.80	29.8	36.3
2.5d	7.60	8.70	34.2	51.4
3d	7.65	8.90	35.2	54.8
3.5d	7.60	8.40	22.9	12.7
4d	7.60	8.10	54.3	120.3
4.5d	7.65	8.95	54.2	120.0
5d	7.70	8.70	36.3	58.6
5.5d	7.70	8.90	31.4	41.8
6d	7.65	8.30	29.4	35.0
8d	7.70	8.45	24.5	18.2
10d	7.65	8.95	27.5	28.4
11d	7.65	8.90	1415.1**	
12d	7.60	8.55	392.9**	

Experiment No. 9 Cont'd

Initial Conditions	1. **Water:**	**Salinity**	7.6 ± 0.1 ^{o}/oo
		pH	7.65 ± 0.05
		Volume used	40 ± 0.5
		Poison	Nil
	2. **Sediment:**	**type**	Core #1, 2-4 cm
		amount added	11.67 g

Sampling time	**pH**	**T(^{o}C)**	**Corrected Hg conc (ng/l)**	**Released Hg (ng) per g sediment**
13d	7.55	8.65	92.3**	
14d	7.60	8.60	69.2**	
15d	7.65	8.20	28.2	30.8

**Possible air contamination from broken Hg thermometer

Experiment No. 9

Initial Conditions	1. Water:	Salinity	7.6 ± 0.1 ^{o}/oo
		pH	7.75 ± 0.05
		Volume used	40 ± 0.5 ℓ
		Poison	2 g $CuSO_4$
	2. Sediment:	type	core #1, 2-4 cm
		amount added	10.88 g

Sampling time	pH	T(^{o}C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	7.75	8.90	20.9	-
0h	7.70	8.85	27.5	24.3
1h	7.70	8.85	33.6	46.7
2h	7.70	8.85	34.0	48.2
4h	7.80	8.85	50.9	110.3
8h	7.80	8.60	34.2	48.9
12h	7.75	8.10	27.8	25.4
18h	7.75	8.00	28.5	27.9
24h	7.70	8.80	35.4	53.3
1.5d	7.80	8.80	33.3	45.6
2d	7.85	8.70	30.0	33.5
2.5d	7.85	8.70	27.8	25.4
3d	7.80	8.85	26.7	21.3
3.5d	7.75	8.85	29.8	32.7
4d	7.80	8.75	27.5	24.3
4.5d	7.80	8.80	28.2	26.8
5d	7.75	8.90	36.2	56.3
5.5d	7.75	8.90	37.2	60.0
6d	7.80	8.90	32.8	43.8
8d	7.70	8.50	38.1	63.2
10d	7.70	8.80	37.1	59.6
11d	7.70	8.80	1464.7**	-
12d	7.70	8.65	249.5**	-
13d	7.70	8.75	66.1**	-
14d	7.75	8.55	36.4	57.0
15d	7.75	8.25	40.6	72.4

* hole in nuclepore filter

** possible air contamination from broken Hg thermometer

Experiment No. 9

Initial Conditions 1. Water: Salinity 7.6 ± 0.1 ^{o}/oo

pH 7.65 ± 0.05

Volume used 45 ± 0.5 ℓ

Poison 2 g $CuSO_4$

2. Sediment: type –

amount added Nil

Sampling time	pH	T(^{o}C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	7.65	8.75	22.7	
0h	7.60	8.70	112.4*	
1h	7.60	8.70	27.9	
2h	7.65	8.95	18.5	
4h	7.65	8.85	18.2	
8h	7.65	8.85	18.6	
12h	7.75	8.40	18.5	
18h	7.65	8.35	20.0	
24h	7.65	8.05	17.2	
1.5d	7.70	8.80	17.7	
2d	7.65	8.85	18.9	
2.5d	7.65	8.75	19.8	
3d	7.70	8.75	19.4	
3.5d	7.70	8.90	23.8	
4d	7.75	8.55	17.5	
4.5d	7.80	8.75	65.2*	
5d	7.65	8.15	22.2	
5.5d	7.65	8.80	23.8	
6d	7.65	8.80	19.9	
8d	7.65	8.95	18.3	
10d	7.65	8.40	20.7	
11d	7.65	8.55	756.7**	
12d	7.70	8.80	153.3**	
13d	7.65	8.70	112.6**	
14d	7.65	8.60	55.8**	
15d	7.65	8.20	28.7	

* hole in nuclepore filter

** possible air contamination from broken Hg thermometer

Experiment No. 9

Initial Conditions	1. Water:	Salinity	19.6 ± 0.1 °/oo
		pH	7.50 ± 0.05
		Volume used	40 ± 0.5 ℓ
		Poison	Nil
	2. Sediment:	type	core #1, 2-4 cm
		amount added	10.04 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	7.50	8.90	36.4	-
0h	7.50	8.80	51.7	61.0
1h	7.55	8.70	50.6	56.6
2h	7.55	8.80	61.8	101.2
4h	7.60	8.80	61.9	101.6
8h	7.55	8.40	43.0	26.3
12h	7.50	7.90	44.7	33.1
18h	7.50	7.90	47.5	44.2
24h	7.50	8.75	787.8*	-
1.5d	7.50	8.80	54.9	73.7
2d	7.50	8.80	50.1	54.6
2.5d	7.50	8.85	35.1	-5.2
3d	7.50	8.60	42.8	25.5
3.5d	7.50	8.55	44.4	31.9
4d	7.65	8.40	42.2	23.1
4.5d	7.55	8.85	46.5	40.2
5d	7.55	8.85	38.6	8.8
5.5d	7.50	8.45	47.4	43.8
6d	7.55	8.35	44.1	30.7
8d	7.55	8.85	46.9	41.8
10d	7.55	8.85	44.7	33.1
11d	7.55	8.85	129.0**	-
12d	7.55	8.90	90.7**	-
13d	7.60	8.85	535.6**	-
14d	7.60	8.50	452.0**	-
15d	7.55	8.65	67.0**	-

* hole in nuclepore filter

** possible air contamination from broken Hg thermometer

Experiment No. 9

Initial Conditions	1. Water:	Salinity	19.6 ± 0.1 ^{o}/oo
		pH	7.45 ± 0.05
		Volume used	40 ± 0.5
		Poison	2 g $CuSO_4$
	2. Sediment:	type	core #1, 2-4 cm
		amount added	9.26 g

Sampling time	pH	T(^{o}C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	7.55	8.75	31.7	-
0h	7.45	8.65	40.0	35.9
1h	7.45	8.70	60.2	123.1
2h	7.50	8.70	50.3	80.3
4h	7.50	8.65	55.3	101.9
8h	7.40	8.40	43.2	49.7
12h	7.50	8.00	42.8	47.9
18h	7.55	7.90	39.2	32.4
24h	7.55	8.15	72.9	178.0
1.5d	7.55	8.80	44.7	56.2
2d	7.55	8.80	37.5	25.1
2.5d	7.55	8.75	47.0	66.1
3d	7.55	8.75	43.3	50.1
3.5d	7.50	8.80	47.7	69.1
4d	7.55	8.75	65.0	143.8
4.5d	7.55	8.85	93.5	267.0
5d	7.55	8.75	55.6	103.2
5.5d	7.55	8.75	43.4	50.5
6d	7.55	8.70	64.6	142.1
8d	7.55	8.60	42.9	48.4
10d	7.55	8.65	44.2	54.0
11d	7.55	8.55	1401.9**	-
12d	7.55	8.80	233.5**	-
13d	7.55	8.80	249.7**	-
14d	7.55	8.70	127.8**	-
15d	7.55	8.70	79.4**	-

* hole in nuclepore filter

** possible air contamination from broken Hg thermometer

Experiment No. 9

Initial Conditions				
	1.	Water:	Salinity	19.6 ± .1 $^{o}/oo$
			pH	7.45 ± 0.05
			Volume used	40 ± 0.5 ℓ
			Poison	Nil
	2.	Sediment:	type	-
			amount added	Nil

Sampling time	pH	T(^{o}C)	Corrected Hg conc (ng/l)	Released Hg (ng) per g sediment
Background	7.50	8.85	19.0	
0h	7.45	8.80	19.9	
1h	7.50	8.70	15.6	
2h	7.55	8.90	90.6*	
4h	7.45	8.80	23.5	
8h	7.50	8.50	20.5	
12h	7.50	8.45	21.2	
18h	7.50	8.40	17.4	
24h	7.45	8.35	30.0	
1.5d	7.50	8.90	19.6	
2d	7.50	8.85	18.3	
2.5d	7.45	8.70	17.6	
3d	7.55	8.85	19.0	
3.5d	7.45	8.80	17.2	
4d	7.50	8.75	25.2	
4.5d	7.45	8.80	159.2*	
5d	7.50	8.85	21.0	
5.5d	7.50	8.85	25.4	
6d	7.50	8.90	19.4	
8d	7.50	8.75	20.9	
10d	7.45	8.80	18.5	
11d	7.50	8.80	580.9**	
12d	7.50	8.80	216.9**	
13d	7.50	8.40	75.6**	
14d	7.45	8.20	43.5**	
15d	7.50	8.45	40.4	

* hole in nuclepore filter
** possible air contamination from broken Hg thermometer

Experiment No. 9

Initial Conditions	1.	Water:	Salinity	19.6 ± 0.1 $^{o}/oo$
			pH	7.55 ± 0.05
			Volume used	40 ± 0.5 ℓ
			Poison	2 g $CuSO_4$
	2.	Sediment:	type	-
			amount added	Nil

Sampling time	pH	T(^{o}C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	7.50	8.80	20.8	
0h	7.55	8.85	18.1	
1h	7.60	8.85	23.0	
2h	7.60	8.85	97.0*	
4h	7.60	8.80	21.4	
8h	7.60	8.40	25.1	
12h	7.50	8.30	19.7	
18h	7.60	8.80	24.6	
24h	7.55	8.90	21.8	
1.5d	7.60	8.95	21.2	
2d	7.60	8.40	21.8	
2.5d	7.55	8.80	22.9	
3d	7.55	8.55	23.0	
3.5d	7.55	8.80	22.0	
4d	7.55	8.30	22.2	
4.5d	7.55	8.55	22.9	
5d	7.55	8.80	43.1	
5.5d	7.60	8.85	28.0	
6d	7.55	8.90	25.7	
8d	7.55	8.90	25.3	
10d	7.40	8.95	23.9	
11d	7.55	8.75	991.2**	
12d	7.55	8.85	288.8**	
13d	7.60	8.55	105.7**	
14d	7.60	8.55	39.3	
15d	7.55	8.55	19.0	

* hole in nuclepore filter

** possible air contamination from broken Hg thermometer

Experiment No. 9

Initial Conditions	1. Water:	Salinity	28.8 ± 0.1 o/oo
		pH	7.80 ± 0.05
		Volume used	40 ± 0.5 ℓ
		Poison	Nil
	2. Sediment:	type	core #1, 2-4 cm
		amount added	9.68 g

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	7.75	8.75	19.2	-
0h	7.85	8.95	24.9	23.6
1h	7.80	8.95	56.5	154.1
2h	7.70	8.90	66.3	194.6
4h	7.70	8.85	63.2	181.8
8h	7.70	8.80	46.0	110.7
12h	7.75	8.35	50.0	127.3
18h	7.80	8.05	53.0	139.7
24h	7.90	8.80	40.0	86.0
1.5d	7.85	8.75	33.4	58.7
2d	7.85	8.80	32.0	52.9
2.5d	7.80	8.85	33.2	57.9
3d	7.80	8.85	17.8	-5.8
3.5d	7.80	8.20	33.9	60.7
4d	7.80	8.05	34.5	63.2
4.5d	7.80	8.80	37.0	73.6
5d	7.75	8.70	31.3	50.0
5.5d	7.80	8.85	29.7	43.4
6d	7.75	8.10	31.6	51.2
8d	7.75	8.15	28.7	39.3
10d	7.80	8.20	29.3	41.7
11d	7.80	8.95	702.1**	-
12d	7.85	8.80	88.6**	-
13d	7.80	8.75	65.9**	
14d	7.80	8.70	30.4	46.3
15d	7.80	8.45	37.3	74.8

* hole in nuclepore filter

** possible air contamination from broken Hg thermometer

Experiment No. 9

Initial Conditions			
1.	Water:	Salinity	28.8 ± 0.1 $^{o}/oo$
		pH	7.75 ± 0.05
		Volume used	40 ± 0.5 ℓ
		Poison	2 g $CuSO_4$
2.	Sediment:	type	core #1, 2-4 cm
		amount added	11.53 g

Sampling time	pH	T(^{o}C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	7.80	8.60	25.0	-
0h	7.80	8.60	29.6	16.0
1h	7.80	8.65	23.8	-4.2
2h	7.70	8.60	27.9	10.1
4h	7.75	8.60	23.3	-5.9
8h	7.75	8.50	30.2	18.0
12h	7.75	8.00	52.0	93.7
18h	7.80	8.60	99.3	257.8
24h	7.80	8.65	60.7	123.9
1.5d	7.85	8.65	53.0	97.1
2d	7.80	8.65	49.0	83.3
2.5d	7.80	8.70	58.4	115.9
3d	7.80	8.65	91.2	230.0
3.5d	7.80	8.65	97.1	250.1
4d	7.85	8.35	99.0	256.7
4.5d	7.75	8.30	119.7	328.5
5d	7.85	8.70	61.5	126.6
5.5d	7.75	8.70	44.3	67.0
6d	7.80	8.65	44.9	69.0
8d	7.80	8.60	39.6	50.7
10d	7.75	8.40	42.8	61.8
11d	7.75	8.45	534.9**	-
12d	7.85	8.70	121.6**	-
13d	7.75	8.75	83.9**	-
14d	7.80	8.60	45.2	70.1
15d	7.75	8.40	44.3	67.0

* hole in nuclepore filter
** possible air contamination from broken Hg thermometer

Experiment No. 9

Initial Conditions	1.	Water:	Salinity	28.8 ± 0.1 °/oo
			pH	7.80 ± 0.05
			Volume used	40 ± 0.5 ℓ
			Poison	Nil
	2.	Sediment:	type	-
			amount added	Nil

Sampling time	pH	T(°C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	7.80	8.80	28.1	
0h	7.80	8.80	23.7	
1h	7.80	8.75	38.7	
2h	7.80	8.75	23.1	
4h	7.75	8.75	22.9	
8h	7.75	8.60	21.4	
12h	7.80	8.25	22.2	
18h	7.80	7.80	20.8	
24h	7.85	8.65	19.0	
1.5d	7.85	8.70	24.7	
2d	7.80	8.70	21.8	
2.5d	7.80	8.65	24.9	
3d	7.80	8.80	20.1	
3.5d	7.80	8.20	23.0	
4d	7.80	8.00	22.8	
4.5d	7.80	8.80	28.7	
5d	7.85	8.70	20.0	
5.5d	7.85	8.75	26.9	
6d	7.75	8.45	24.4	
8d	7.80	8.55	22.0	
10d	7.80	8.75	25.1	
11d	7.80	8.85	703.1**	
12d	7.80	8.35	172.6**	
13d	7.80	8.35	68.1**	
14d	7.85	8.80	23.4	
15d	7.80	8.30	89.8**	

* hole in nuclepore filter

** possible air contamination from broken Hg thermometer

Experiment No. 9

Initial Conditions	1. Water:	Salinity	28.8 ± 0.1 $^{o}/oo$
		pH	7.80 ± 0.05
		Volume used	42 ± 0.05 ℓ
		Poison	2 g $CuSO_4$
	2. Sediment:	type	Blank
		amount added	Nil

Sampling time	pH	T(^{o}C)	Corrected Hg conc (ng/1)	Released Hg (ng) per g sediment
Background	7.80	8.60	29.4	
0h	7.80	8.80	28.8	
1h	7.85	8.85	223.4*	
2h	7.90	8.80	23.5	
4h	7.80	8.80	27.0	
8h	7.80	8.40	31.4	
12h	7.85	8.20	34.7	
18h	7.75	7.90	34.4	
24h	7.80	8.45	32.4	
1.5d	7.80	8.80	30.7	
2d	7.80	8.85	25.2	
2.5d	7.85	8.80	25.3	
3d	7.80	8.80	26.4	
3.5d	7.80	8.85	27.0	
4d	7.85	8.90	31.4	
4.5d	7.85	8.85	66.2	
5d	7.80	8.80	27.9	
5.5d	7.80	8.90	28.7	
6d	7.90	8.90	28.7	
8d	7.80	8.65	35.1	
10d	7.75	8.70	24.9	
11d	7.75	8.80	228.1**	
12d	7.75	8.80	201.5**	
13d	7.75	8.65	132.4**	
14d	7.75	8.40	33.7	
15d	7.75	8.40	72.5**	

* hole in nuclepore filter

** possible air contamination from broken Hg thermometer

COULTER COUNTER CODING FORM

EFFICIENCY OF STIRRING TEST			Date Coded:			Date Keypunched:			Vol. - Volume of sample Type 01 FAST STIR 02 SLOW STIR						Page of			
SAMPLE Nº	TYPE	VOL	Counts Per Channel															
			1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
1	01	2	0	0	3958	2003	1200	655	428	194	118	99	63	35	24	5	2	0
2	01	2	0	0	3529	1530	1086	655	419	246	141	95	44	38	19	6	1	0
3	01	2	0	0	4126	1780	1171	674	439	262	211	101	55	28	13	1	0	0
4	01	2	0	0	4249	1765	1118	681	421	193	121	40	13	9	1	1	0	0
5	01	2	0	0	4231	1661	1004	563	379	220	133	103	105	71	32	9	1	0
6	01	2	0	0	4041	1552	1209	643	378	246	174	103	103	68	25	0	2	0
7	01	2	0	0	4253	1966	1234	648	385	268	150	102	74	48	36	24	4	0
8	01	2	0	0	3660	1500	1173	681	454	324	220	183	109	69	38	12	9	0
9	01	2	0	0	3693	1514	1045	640	415	279	177	172	146	55	31	5	2	0
10	01	2	0	0	4134	1752	1131	705	474	225	191	181	125	40	23	5	1	0
1	02	2	0	0	3226	368	163	102	74	56	35	36	15	12	6	0	1	0
2	02	2	0	0	3250	335	144	91	62	63	59	50	46	25	8	3	0	0
3	02	2	0	0	3951	500	246	202	176	134	112	85	29	10	4	0	0	0
4	02	2	0	0	3488	555	311	214	175	171	162	151	88	54	33	8	1	0
5	02	2	0	0	1808	575	561	338	216	178	123	128	81	25	4	1	0	0
6	02	2	0	0	2815	544	246	123	56	53	21	13	12	12	9	2	0	0
7	02	2	0	0	2318	750	462	261	182	154	88	58	114	79	31	5	0	0
8	02	2	0	0	2391	703	442	259	175	194	91	99	133	102	31	8	1	0
9	02	2	0	0	2641	616	262	162	99	64	29	21	8	1	1	0	0	0
10	02	2	0	0	2724	499	276	124	94	68	60	35	0	3	1	0	0	0

Appendix A.4

Data for Static Release Experiments

Aerobic Static Release Experiment
Daily Averaged Mercury Flux

Time (days)	Mercury Concentration in Solution (ng/l)	Total Δ Hg (ng)	Hg Flux (ng/cm^2/day)
0	398		
1	697	3348	8.4
2	782	1507	3.8
3	2204	14962	37.4
4	3200	12124	30.3
5	3860	9660	24.2
6	4308	8390	21.0
7	5328	14468	36.2
8	8098	32988	82.5
9	6030	-12622	- 31.6
10	7720	22890	57.2
11	8050	10980	27.5
12	10170	29210	73.0
13	10926	17690	44.2
14	11860	20226	50.6
15	8550	-21280	- 53.2
16	9520	18210	45.5
17	17726	91540	228.9
18	23798	78406	196.0
19	40250	188278	470.7
20	23400	128290	320.7
21	9855	-122090	-280.2
22	7330	- 15434	- 38.6
23	7078	4770	11.9
24	2173	- 42012	-105.0
25	6665	47053	117.6
26	6288	2855	7.1
27	6320	6568	16.4
28	6028	3360	8.4
29	12358	69228	-173.2
30	3388	- 28519	71.3

Concentration of Mercury in Sediment (ppb) at Completion of Aerobic Static Release Experiment, t = 30 d

Depth Along Core (cm)	1	2	3	4	5	6	7	8	9
0-1	471 (2.0)*	109 (2.0)	508 (1.8)	566 (1.9)	423 (1.8)	421 (2.0)	385 (1.6)	324 (2.1)	602 (2.0)
4-5	374 (1.1)	339 (1.0)	428 (1.6)	441 (1.7)	360 (1.5)	199 (1.0)	217 (1.2)	576 (1.1)	559 (1.1)
9-10	543 (0.8)	91 (1.1)	511 (1.4)	440 (0.5)	361 (0.9)	428 (1.1)	332 (1.0)	466 (0.6)	453 (0.3)
14-15	347 (0.3)	460 (0.2)	888 (1.3)	894 (1.5)	402 (0.6)	233 (0.4)	501 (1.0)	570 (0.8)	328 (0.1)
19-20	125 (.1)	952 (.1)	440 (.1)		368 (.2)	210 (.3)	278 (.1)	944 (.1)	281 (.2)

* Bracketted numbers refer to % organic carbon content of sediment

Key				
	7	8	9	
	4	5	6	Plan view of box core in container Sediment injected into 5 through 2 at a depth of 10 cm.
	1	2	3	

Appendix B.1

Data for In Situ Study

Dissolved Mercury in Water and Interstitial Waters
Squamish River Estuary and Mud Flats

FMC Outfall 1.0 µg/l

Station	Sampling Period	Dissolved Mercury (ng/l)	Interstitial Mercury (µg/l)
1	Background	240	
	1	320	2.0
	2		
	3		
	4	390	1.5
	5	460	
2	Background	235	
	1	260	14.4
	2		12.1
	3		
	4	510	2.0
	5	310	
3	Background	290	
	1	300	14.1
	2		16.0
	3	310	5.5
	4	380	1.4
	5	310	
4	Background	42	
	1	140	16.7
	2		14.6
	3	170	4.9
	4	270	1.5
	5	170	
5	Background	33	
	1	92	1.2
	2		2.1
	3	69	1.9
	4	91	1.8
	5	60	

Particulate Mercury in Waters of Squamish River Estuary

Station	Sampling Period	Suspended Solids	Particulate Hg (ng/l)
1	1	30.2	280
	2		
	3		
	4	680.5	860
	5	860.3	78000
2	1	41.6	520, 480
	2		
	3		
	4	785.4	3200, 3400
	5	880.2	1400
3	1	95.1	3800
	2		
	3	65.3	3200
	4	159.8	2800
	5	585.5	1440
4	1	17.8	780
	2		
	3	173.4	320
	4	496.7	300
	5	182.8	220
5	1	1.0	40
	2		
	3	1.8	44
	4	560.3	360
	5	77.7	1340

COULTER COUNTER CODING FORM

Coded by: IN SITU STUDY

Date Coded:

Date Keypunched:

Vol. = Volume of sample

Page of

	Sample Time	Sample Site	Vol.	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
				Counts Per Channel															
	B	1	2	0	0	11140	6120	1518	3337	2514	1185	1065	49	128	8	6	0	0	0
	B	2		0	0	12129	6499	1493	3141	245	1154	1119	95	52	25	24	19	0	0
	B	3		0	0	15911	11125	1800	525	369	945	1000	33	5	1	0	0	0	0
	B	4		0	0	18335	6182	366	295	183	1604	30	5	1	3	0	0	0	0
	B	5		0	0	15550	11135	1775	694	415	1235	115	20	5	1	0	0	0	0
	1	1		0	0	14360	1515	3112	200	122	136	13	2	0	0	0	0	0	0
	1	2		0	0	14336	528	1246	222	1105	56	10	13	0	0	1	0	0	0
	1	3		0	0	14300	9045	1415	345	1192	110	275	49	10	1	4	1	0	0
	1	4		0	0	3360	11455	1591	533	350	244	188	34	45	35	12	0	0	0
	1	5		0	0	15583	1530	385	331	1225	1120	53	13	5	12	0	0	0	0
	3	3		0	0	42133	11132	1563	680	535	406	295	152	~~73~~ 37	~~460~~ 46	~~185~~ 73	66	0	0
	3	4		0	0	5802	11520	894	682	5914	449	289	142	49	12	1	0	0	0
	3	5		0	0	41162	855	1630	4644	1440	248	121	443	09	5	1	1	0	0
	4	1		0	0	39420	11060	8640	6660	14840	3280	11440	710	180	10	10	0	0	0
	4	2		0	0	44000	11520	4520	7530	16580	5900	13340	3450	1650	510	260	90	0	0
	4	3		0	0	53360	24830	8710	6540	15520	4270	21500	850	220	10	10	0	0	0
	4	4		0	0	33130	8450	5530	4450	13500	2590	10400	390	50	10	0	0	0	0
	4	5		0	0	25440	16620	4760	6650	15000	3720	23600	1300	520	220	50	50	0	0
	5	2		0	0	25900	11250	18310	16220	14050	23500	17500	450	100	50	10	0	0	0
	5	1		0	0	10740	21560	11600	[illegible]	14490	37400	23400	1370	550	350	100	50	0	0
	5	7		0	0	30020	15440	30430	17380	15400	3311	16550	600	100	40	0	0	0	0
	5	4		0	0	13740	16720	14600	38600	15260	2710	12200	1130	320	290	230	20	0	0
	5	5		0	0	9480	6610	15110	3590	13520	1950	16000	440	120	30	20	0	0	0
	6																		