MERCURY UPTAKE IN RECENT LAKE SEDIMENTS

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ABSTRACT

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By

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The interaction of mercury (II) (as ^{203}Hg, 42 days) introduced into the overlying water with lake sediment was studied. Sediments from two Central Michigan lakes were used in the uptake experiments and the rate of uptake was used to calculate the effective mercury residence time in the overlying water. The residence in organic-rich sediment was found to be about 15 hours. A competition experiment was performed with copper and it was found that copper will replace mercury in the sediment. The total mercury adsorption capacity of lake sediment was obtained from an adsorption isotherm experiment, and it was found to be 3.1%.

Mercury (II) has a brief residence time in water which is decontaminated through complexing with the organic-rich fraction of the bottom. Copper (II) and possibly other metals are able to compete with and displace mercury back into the aqueous environment. This experimental approach provides data for modelling the behavior of metals in natural waters.

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INTRODUCTION

It was generally accepted, until five years ago that inorganic mercury released into the environment would remain biologically inert and be diluted to the point where it would pose no danger to the ecosystem. Jenson and Jernolov (1969) demonstrated that inorganic mercury is methylated by an organic sludge-type sediment into methylmercury. Wood *et al.* (1968) extracted the methylating agent in organic sediments as being a methyl-vitamin B_{12} type compound. Microorganisms initiate the conversion of mercury compounds within the sediment to the more mobile methylmercury compounds which are then readily incorporated by the biota. The actual rate of mercury loss from a sediment due to methylation has not been determined, but it has been estimated by Jernolov (1970) as being on the order of 1% per annum, assuming that during this period there was no sedimentation and no additional mercury input to the system. Although the process of methylation as a mechanism for mercury mobilization has been extensively studied very little is known about the mechanisms and rates of sediment fixation of mercury.

Previous investigations have studied the relationships between physico-chemical parameters of the sediment and the behavior of the mercury compounds. The amount of mercury

present in a sediment type is highly correlated with the organic fraction of the sediment (Thomas 1972 Anderson 1970). Within the organic fraction there is a preferential association of mercury with particles greater than 0.45 u and less than 20 u (Cranston and Buckley 1971). Heljev (1971), in his investigation of mercury interactions with humic acids, used a simplified model of the natural environment to explain uptake reactions. Huljev's simplified model of mercury uptake forms the basis by which a detailed study can be initiated.

This investigation provides a quantitative measurement of the parameters of uptake and adsorption in lake sediment from Central Michigan. The rate of uptake, which describes the effective mercury residence time in the liquid phase, and the adsorption maximum of mercury in that sediment type is found.

SAMPLE COLLECTION

The sediment samples were acquired from two Mid-Michigan lakes: Lake Lansing and Burke Lake. Samples were collected by the use of a clam-shell bottom dredge. Dredges were made in various sections of each lake and care was taken in the selection of these sites, so that a wide variety of sediment types could be obtained. The dredging operations were carried out over the side of a small boat by hand lowering the device with a rope and tripping it with a messenger. Identification of the sample sites was secured by triangulation with prominent cultural features on the lake shore. Each sample dredged contained, on the average, two liters of sediment. If, for some reason, two liters was not obtained additional dredges were made. The contents of the dredges were placed into plastic collection bags and sealed. When the samples were returned to the lab they were transferred from the plastic bags to one liter glass beakers and sealed air tight with a paraffin covering.

Utmost care was observed in the collection, handling, and storage of the sediment samples. It is very important that these samples closely represent a natural lake bottom sediment during the experiment portion of the investigation. It is difficult to remove a sample from the lake bottom and not disturb it somewhat, but the major change that would occur within a sediment would occur shortly after it was removed from the lake environment.

It is also difficult to achieve actual lake bottom conditions during a laboratory experiment, that is anything short of using a small lake as a test beaker. Lean (1972), in an investigation of the dynamics of phosphorus in lake water actually used a small lake in Ontario as a test site.

The laboratory conditions of the experiment do not exactly duplicate those conditions present in the natural lake system but it is assumed that these changes are slight and the parameters measured would closely approximate those in the natural environment.

EXPERIMENTAL TECHNIQUE

Mercury uptake is a phenomena by which mercury in the overlying water is taken up by the sediment trhough the process of diffusion into the sediment and subsequent complexing onto sediment sites. In the performance of an uptake experiment 250 ml aliquots of each sediment type were placed in a one liter beaker and distilled water was added to 900 ml. The amount of distilled water was measured carefully to calculate the concentration of mercury in the overlying water. The beakers were placed in a thermostatically controlled water bath maintained at $20^{\circ}C \pm 1^{\circ}C$ and allowed to equilibrate for one week.

The radioisotopic (^{203}Hg) tracer was prepared with sufficient activity to insure detection at the part per billion range. To achieve this detection level 0.1 gm of AR grade HgO was irradiated for two hours at a thermal neutron flux of 1×10^{12} n $cm^{-2} sec^{-1}$ in the Triga Mark I reactor in the Department of Chemical Engineering, Michigan State University. The activity resulting from the irradiation can be calculated using the equation:

$$A = N \sigma \phi (1 - e^{-\lambda t}) \qquad (1)$$

where A equals the activity in disintegrations per second (1 Curie = 3.7×10^{10} disintegrations per second), N equals the number of Hg ions, σ is the neutron capture cross section in cm^2, ϕ is the thermal neutron flux, λ is the decay constant for ^{203}Hg, and t is the time of irradiation. The value in this case is 100 uCi. The activity resulting from this irradiation is entirely due to the 46.6 day half-life of ^{203}Hg with very minor contributions from ^{197}Hg, ^{199}Hg, and ^{205}Hg. The major nuclear reaction taking place is:

$$^{202}\mathrm{Hg}\ (n,\sigma)\ ^{203}\mathrm{Hg} \qquad (2)$$

The beta (β) decay reaction of ^{203}Hg is:

$$^{203}\mathrm{Hg} = {}^{203}\mathrm{Tl} + \beta \qquad (3)$$

^{19}O with a half-life of 26.8 seconds is the only other principal activity from the irradiation.

The radioactive tracer was dissolved and prepared to a suitable concentration for addition to the sediment beakers. Without disturbing the bottom sediment in the beaker, the solution containing the ^{203}Hg was added by pipette. In order to remove any concentration gradient within the overlying water the beaker was stirred from above at about 30 rpm, a speed sufficiently slow to prevent placing the bottom in suspension.

An automatic sampling device was designed and built that will remove a 3 ml sample from the test beaker at variable time intervals from one minute to sixty minutes. The vial filler "precontaminates" itself and places the "rinse" volume into a test tube in the graction collector. This is a "flushing" procedure to remove the ^{203}Hg carry over from the previous 3 ml sample. The fraction collector is then indexed to receive the sample volume which is placed in a scintillation vial. At this point the sampling system is shut off until the preset time has elapsed. In a previous study (Spooner and Ehurlich unpublished results) a control beaker was included containing the same amount of activity as the sediment beaker in order to correct for any adsorption effects onto the glassware. It was determined that there was no adsorption onto the glassware.

A Nano-Stat syringe was used to add 2 ml of Insa-Gel, a scintillation medium, to each vial. The vials were placed in a Packard Tri-Carb scintillation counter, and a gain setting of 12% was selected to observe the 0.212 MeV beta ray given off in equation (3). The counter was set in the 10,000 count mode to achieve 1% counting statistics. In the case of the 46.6 day half-life of ^{203}Hg, the counting time for all of the samples in one experiment was short compared to the half-life. The counting results were then converted into concentration units and plotted.

An adsorption isotherm will give the amount of mercury adsorbed per gram of sediment at equilibrium with increasing concentrations of mercury in the overlying water at a constant temperature. The experimental set-up in an adsorption isotherm was similar to that used in the uptake experiment. An accurately weighed amount of sediment (about 0.33 gm) was placed in a one liter beaker and distilled water was added to make 600 ml. A control beaker was included containing 600 ml of distilled water in order to calculate the amount of mercury adsorbed onto the sediment and to correct for the decay of the radioisotope. Both beakers were sealed airtight and placed in a thermostatically controlled water bath and allowed to equilibrate for one week. The radioisotope was prepared in the same manner as in the uptake experiment and added to both beakers by pipette. The beakers were magnetically stirred, in the sample beakers the stirring rate must keep all of the sediment in constant suspension. At various times the stirring rate in the sample beaker was increased and decreased to remove the sediment ring that forms on the beaker wall.

The beakers were allowed to equilibrate for 24 hours between each addition of the radioisotope. At the end of the 24 hour period a 1 ml sample was drawn from each beaker through a 45 Å dialysis membrane with a 1 ml Centurion hand pipette. These samples were handled and counted in the same manner as the uptake samples. The concentration of the mercury in the liquid phase of the sediment beaker, subtracted from the concentration in the control beaker, gives the amount of mercury adsorbed onto the sediment. In performing a adsorption isotherm in this manner it is very important that the level of liquid remains constant in both beakers and that an equal amount of radioactive tracer be added to each beaker. These results were plotted in accordance with the accepted procedures for adsorption isotherm data (Ellis and Knezek 1972).

The methods used to determine the percent organics,

TABLE NO. 1 SEDIMENT CHARACTERISTICS

SEDIMENT TYPE	PERCENT ORGANICS	PERCENT CARBONATE	SILTS, CLAYS & OTHERS
LAKE LANSING NO. 1	37.7	6.45	QUARTZ FELDSPAR MICA (ILLITE)
BURKE LAKE NO. 1	34.7	12.05	
BURKE LAKE NO. 2	41.8	0.85	

percent calcium carbonate, and types of clays are all standard methods. The organic carbon content in the sediment was determined by the Leco Carbon Analyzer at the Department of Soils Science, Michigan State University. Calcium carbonate was determined by a titrimetric method described by Bundy and Bremner (1972). Table No. 1 shows the sediment characteristics of samples. It is important to note that even though there is a high corrolation of mercury with organics, clay plays a role in the uptake of mercury, but makes up a minor component of the sediment.

EXPLANATION OF THE UPTAKE AND ADSORPTION ISOTHERM

The rate of uptake was determined from the rate of ^{203}Hg transfer from the liquid phase to the sediment phase. This approach involves an extension of the basic kinetic relationships of a simple two compartment exchange model (Li *et al.*, 1972). For a closed system involving exchange between mercury in the sediment, the exchange reaction can be represented:

$$Hg_{sol} \underset{k_2}{\overset{k_1}{=}} Hg_{sed} \qquad (4)$$

where k_1 and k_2 are rate constants. If ^{203}Hg is added to the liquid phase, the net rate of ^{203}Hg removal from solution is given by:

$$-d\ (^{203}Hg_{total}) = k_1\ (^{203}Hg_{sol}) - k_2\ (^{203}Hg_{sed}) \qquad (5)$$

In this particular case, the phenomena is essentially an uptake process. As soon as a small amount of Hg is added there is a forward reaction (k_1) and a reverse reaction (k_2), but k_2 is small compared to k_1. Therefore the k_2 term is removed from equation (5). Integration of equation (5) gives the following:

$$^{203}Hg_{sol} = {}^{203}Hg_{total}\ e^{-k_1 t} \qquad (6)$$

For a system obeying the relationships in equation (6), a plot of log $^{203}Hg_{sol}$ against time is linear and allows for evaluation of the uptake parameters.

Evaluation of the uptake parameters for three types of sediments graphically displays that Lake Lansing No. 1 and Burke Lake No. 2 exhibit one uptake rate. Examination of the plots of Burke Lake No. 1 shows a non-linear response exhibiting rather distinct breaks, indicating more than one mode of uptake. Graphical resolution of different rates does not constitute proof that these are the only reactions involved in the uptake process. Other reactions involved may also occur but with rates not sufficiently different to allow graphical resolution. Although different rates are apparent, the graphical approach provides no indication whether these differences were controlled by adsorption or diffusion.

The half-life of the mercury in the liquid can be

Figure 1   Mercury Uptake, Lake Lansing Sediment No. 1

R value of 0.99

T test significant at the 0.001 level

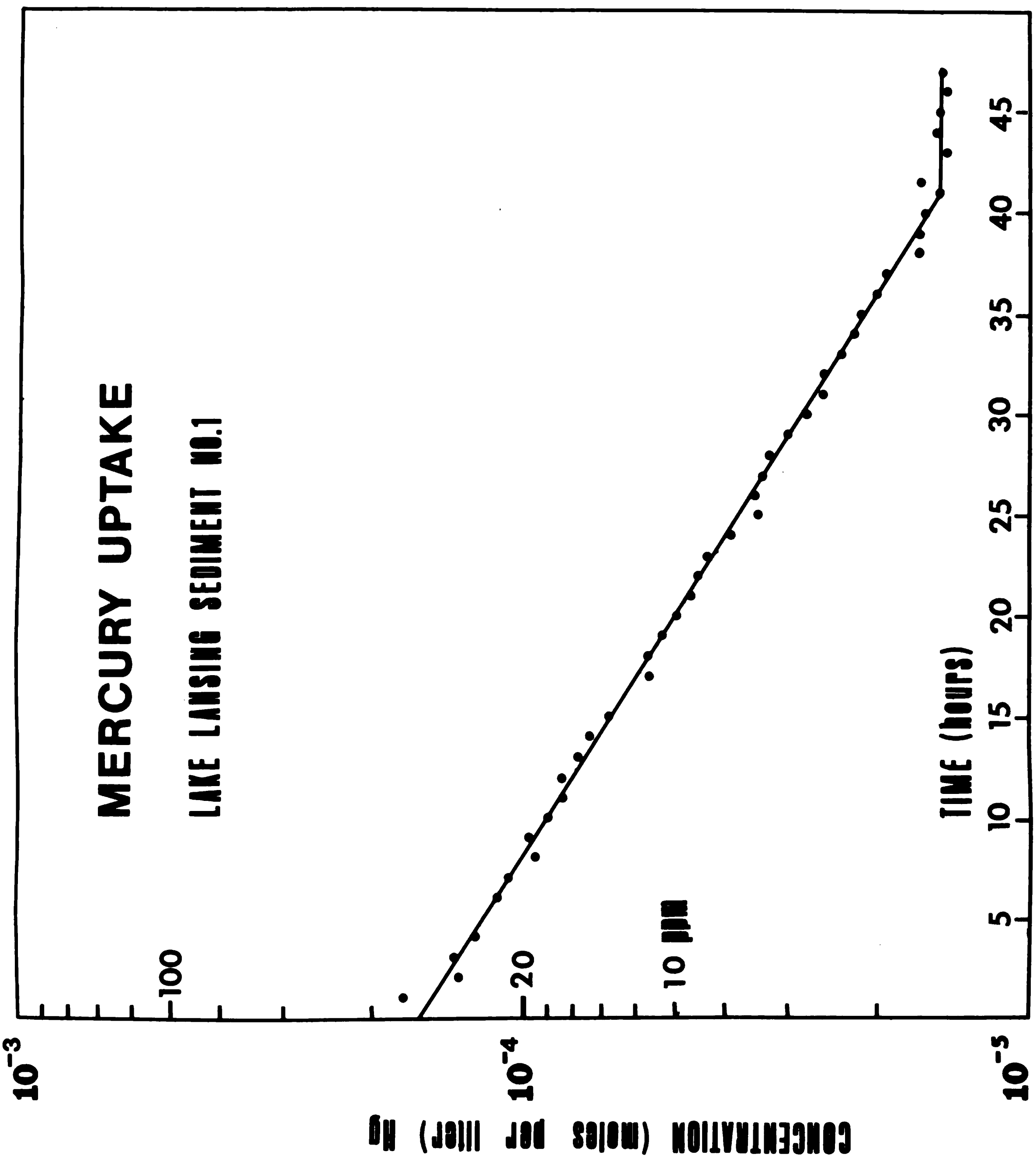

MERCURY UPTAKE
LAKE LANSING SEDIMENT NO.1
TIME (hours)
CONCENTRATION (moles per liter) Hg
10^{-3}
10^{-4}
10^{-5}
100
20
10 ppm
5
10
15
20
25
30
35
40
45

Figure 2 Mercury Uptake, Burke Lake Sediment No. 2, Showing differences between Fresh and Aged Samples

Aged Data from Spooner and Ehurlich, unpublished results

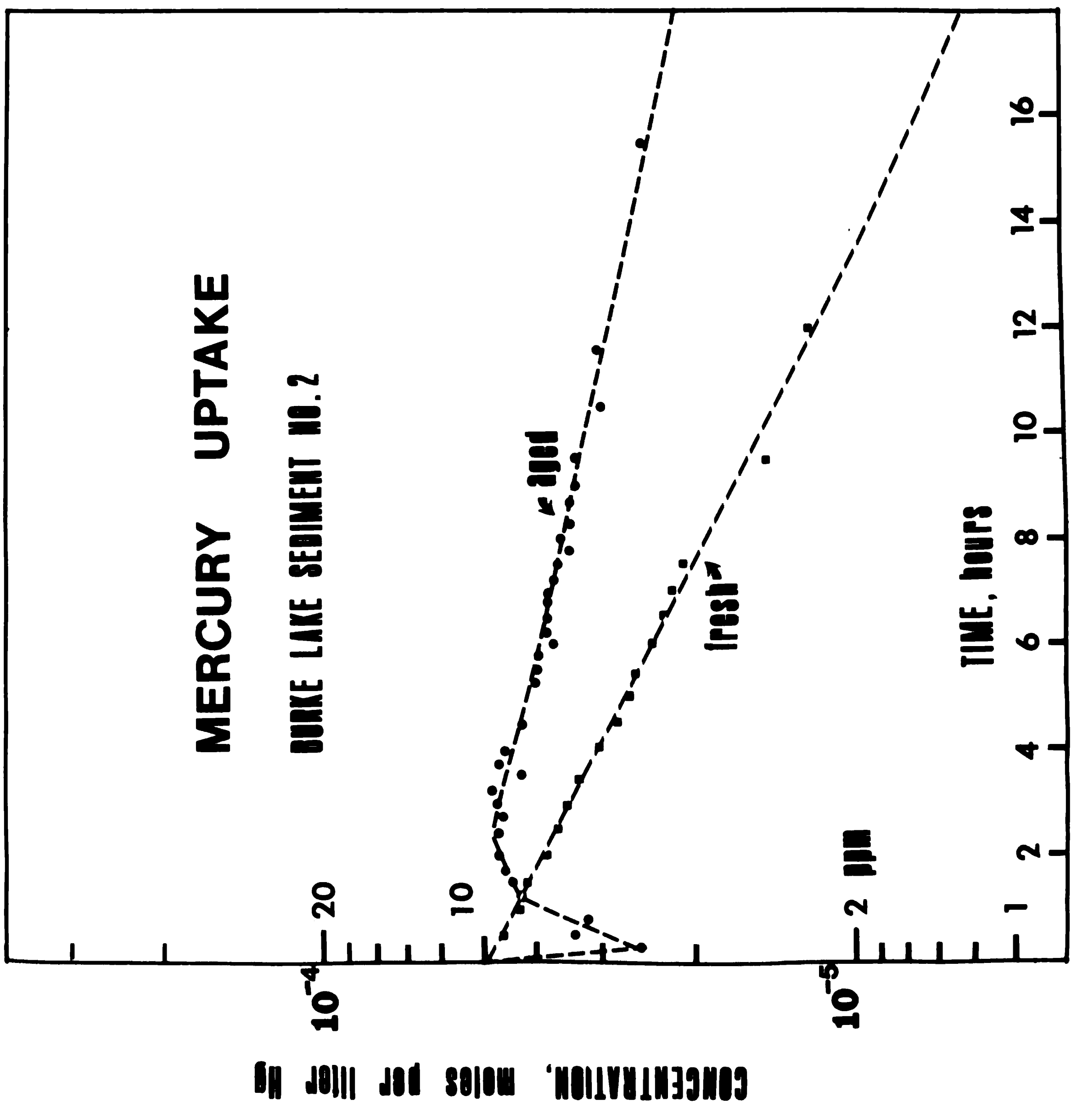
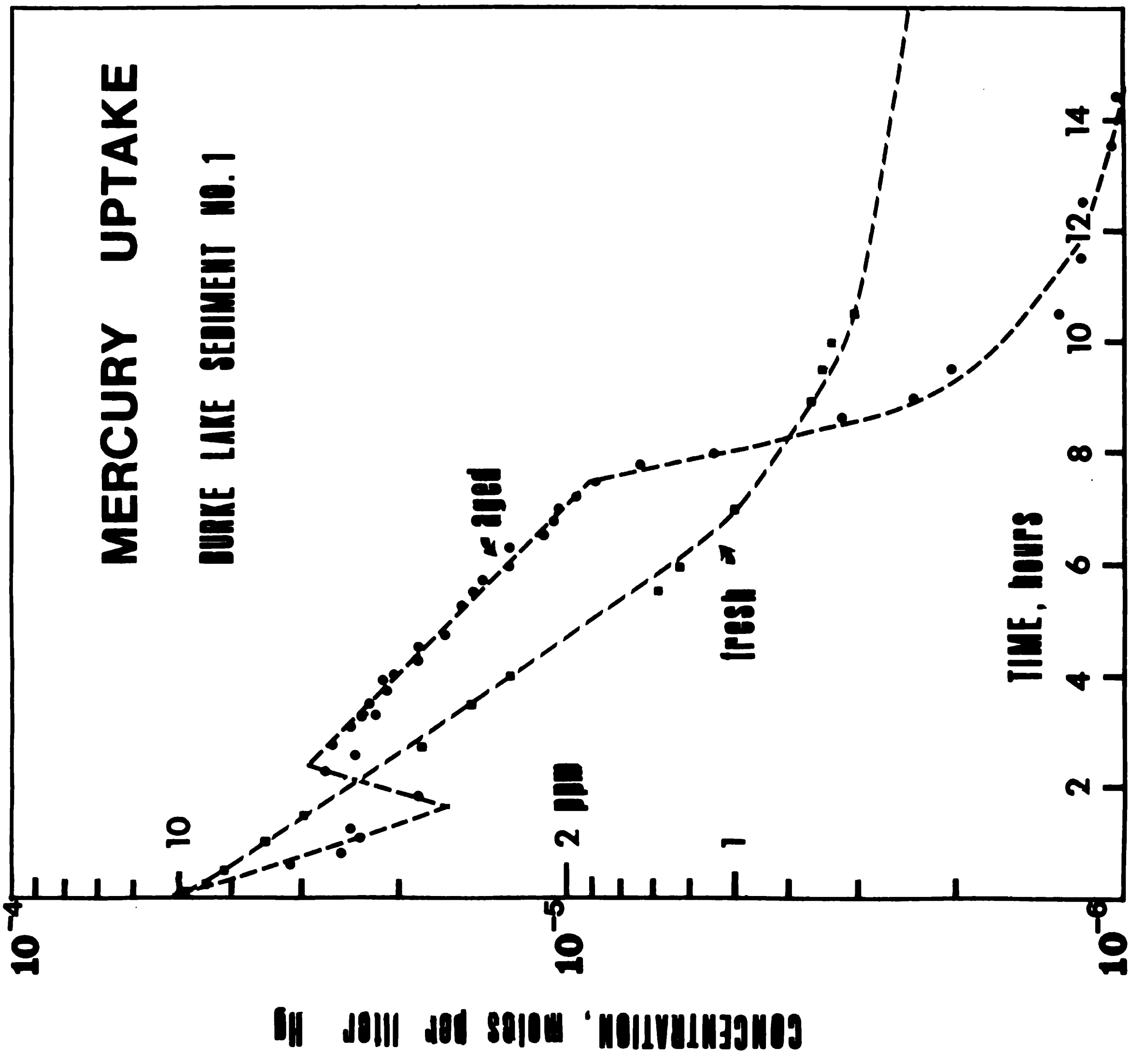

MERCURY UPTAKE
BURKE LAKE SEDIMENT NO. 2
aged
fresh
TIME, hours
1
2
4
6
8
10
12
14
16
CONCENTRATION, moles per liter Hg
10^{-4}
10^{-5}
20
10
2 ppm

Figure 3 Mercury Uptake, Burke Lake Sediment No. 1, Showing Differences between Fresh and Aged Samples


MERCURY UPTAKE
BURKE LAKE SEDIMENT NO. 1
aged
fresh
10
2 ppm
1
TIME, hours
2
4
6
8
10
12
14
10^{-4}
10^{-5}
10^{-6}
CONCENTRATION, moles per liter Hg

calculated from the rate of uptake by the equation:

$$T_{\frac{1}{2}} = \frac{.693}{\lambda} \qquad (7)$$

where λ is the rate constant and $T_{\frac{1}{2}}$ is the half-life. It is only possible to calculate a half-life from a linear equation. In plots where there are a number of distinct rates of uptake, a half-life can be calculated from each reaction (Table 2).

An important parameter defining sediment characteristics is the relative affinity of that sediment for a particular metal. This parameter can be evaluated by placing mercury in competition with other metals such as copper. This approach is graphically presented (Figure 4). Mercury was allowed to be taken up by Lake Lansing No. 1 and at some point in the experiment an equal amount of copper (0.1129 mmoles) was added to the liquid phase. It is graphically displayed that the addition of copper into the closed system containing mercury causes the mercury to be forced out of the sediment back into the liquid phase. Because there are sufficient adsorption sites in the sediment for both mercury and copper, the mercury will again be taken up at a rate similar to the rate before copper addition.

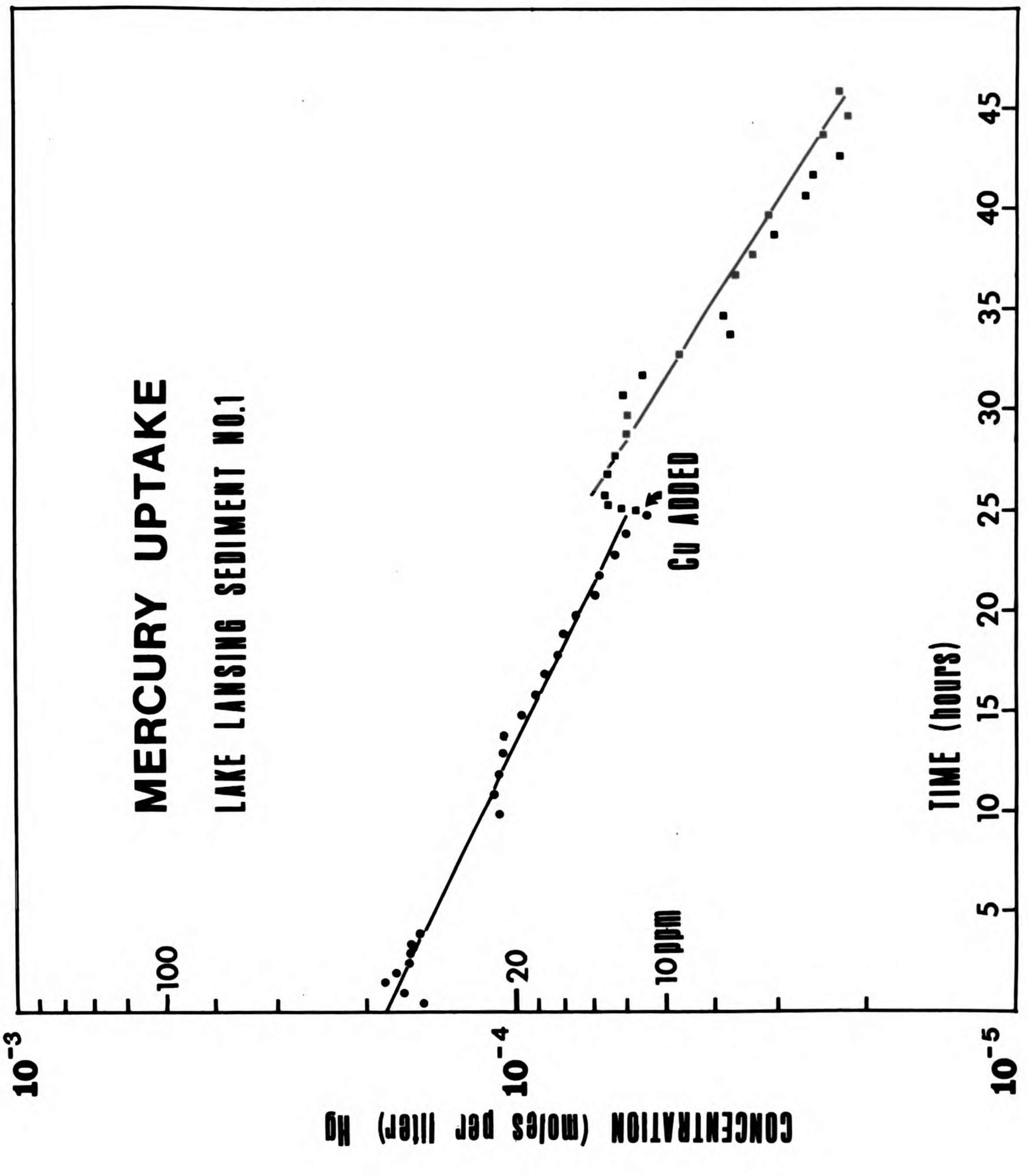
A further investigation of the sediment characteristics of Lake Lansing No. 1 was evaluated with an adsoprtion isotherm (Figure 5). The magnitude of the adsorption effect depends on the temperature, the nature of the adsorbed substance (mercury), the nature and state of the adsorbent

TABLE NO. 2 RATE OF MERCURY UPTAKE

SEDIMENT TYPE	HALF-LIFE, HOURS			
LAKE LANSING NO. 1	13.3 ± 2.3			
BURKE LAKE NO. 1	RATES	1	2	3
	FRESH	1.7	14.8	
	AGED	3.0	0.8	12
BURKE LAKE NO. 2	FRESH	5.7 ± 0.1		
	AGED	15.5		

Figure 4 Mercury Uptake, Lake Lansing Sediment No. 1, Showing Mercury Displacement with the Addition of Copper

R values of 0.99 and 0.98


MERCURY UPTAKE
LAKE LANSING SEDIMENT NO.1
Cu ADDED
100
20
10ppm
TIME (hours)
5
10
15
20
25
30
35
40
45
10^{-3}
10^{-4}
10^{-5}
CONCENTRATION (moles per liter) Hg

(sediment), and the concentration of the mercury in the liquid phase. The first theoretical attempt to describe the relationship between the adsorbed and equilibrium concentrations at a constant temperature was given by Langmuir (1918). Langmuir developed the theory for adsorption of gases on plane surfaces of glass, but the equation adequately describes the adsorption at the liquid-solid interface involving sediments (Ellis and Knezek, 1972).

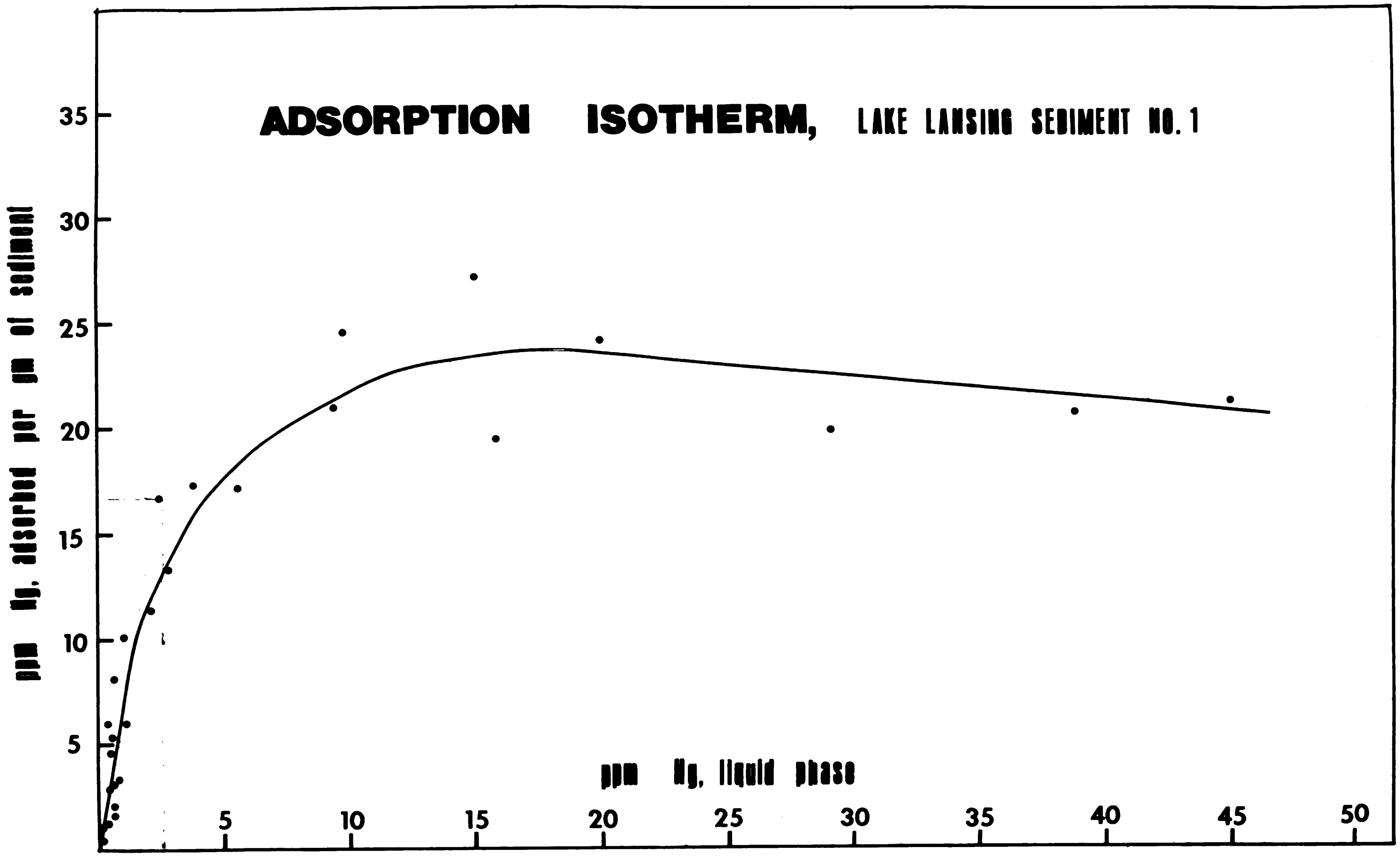
The Langmuir equation has a theoretical basis in the kinetic theory of gases. Langmuir visualized a dynamic equilibrium, such that the rate of adsorption equaled the rate of desorption with the adsorption forming a monomolecular film. The rate of adsorption will be proportional to the concentration of mercury in the liquid phase and the unoccupied sites on the sediment. Thus:

$$\text{Rate of adsorption} = k_1 (M) \left(b - \frac{x}{m} \right) \tag{8}$$

where k_1 is a constant, (M) is the equilibrium concentration of mercury in parts per million, b is the adsorption maximum of the sediment in mg of mercury per gram of sediment, and $\frac{x}{m}$ is the amount of mercury adsorbed per gram of sediment. The rate of desorption is proportional to the occupied sites.

$$\text{Rate of desorption} = k_2 \left(\frac{x}{m} \right) \tag{9}$$

Figure 5 Adsorption Isotherm, Lake Lansing Sediment No. 1



ADSORPTION ISOTHERM, LAKE LANSING SEDIMENT NO. 1

At equilibrium the opposing rates are equal.

$$k_2 \left(\frac{x}{m} \right) = k_1 (M) \left(b - \frac{x}{m} \right) \qquad (10)$$

Solving for $\frac{x}{m}$ and replacing $\frac{k_1}{k_2}$ by K gives the more usual form of the Langmuir equation.

$$\frac{x}{m} = \frac{K\,(M)\,b}{1 + K(M)} \qquad (11)$$

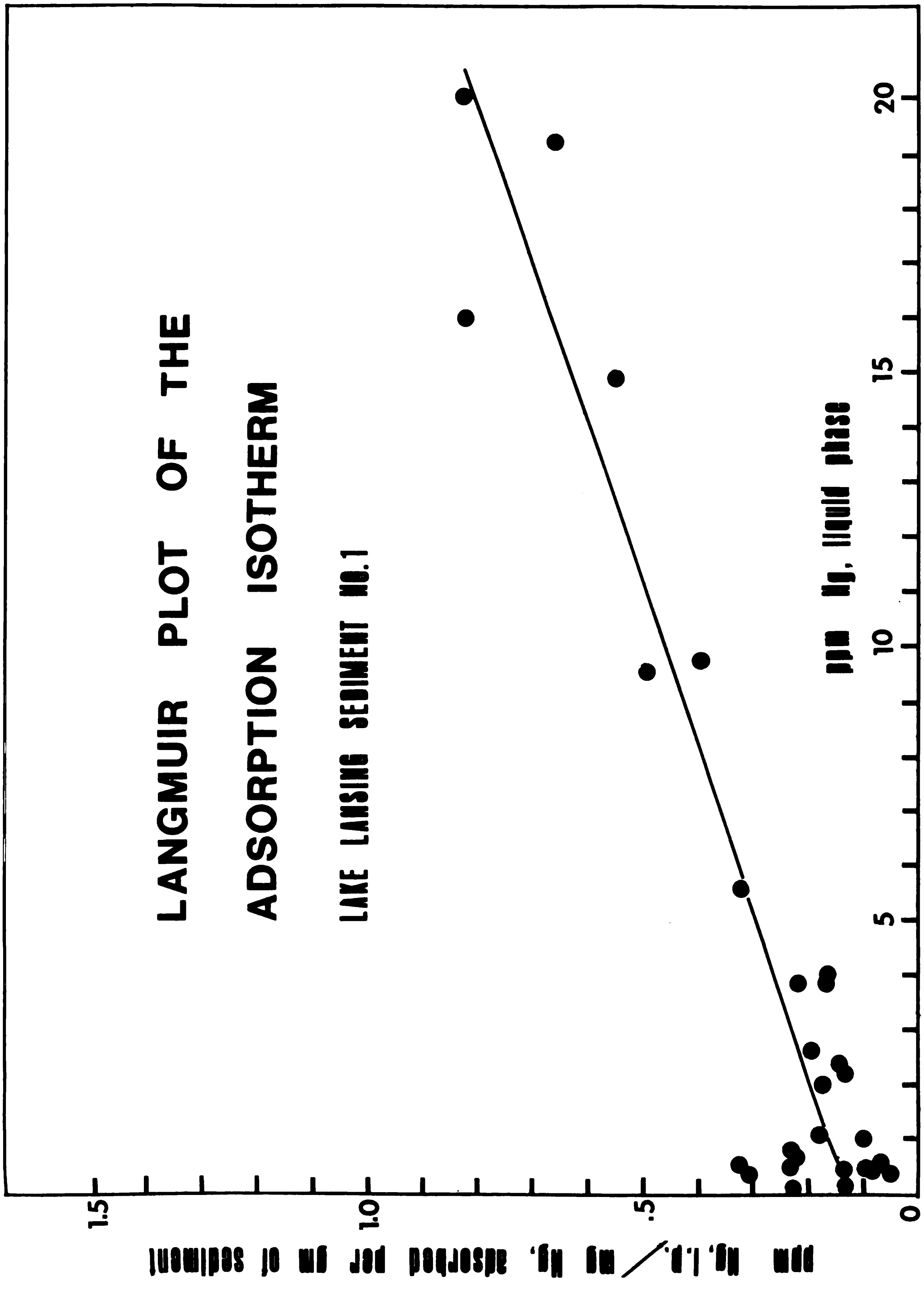
For convenience in testing by standard least square methods, equation (11) can be put in linear form.

$$\frac{(M)}{\frac{x}{m}} = \frac{1}{Kb} + \frac{(M)}{b} \qquad (12)$$

If the experimental plot of the adsorption isotherm obeys the Langmuir equation, a plot of $\frac{(M)}{\frac{x}{m}}$ against (M) should give a straight line. This type of graphical relationship is referred to as a Langmuir plot. The Langmuir plot from the Lake Lansing No. 1 adsorption isotherm yields a straight line with an R value of 0.92 (Figure 6). In experimental work of this nature a correlation coefficient greater than 0.90 constitutes a valid Langmuir relationship (Ellis, personal communication). The calculated value of the reciprocal of the slope of the regression line is the value "b" which is the adsorption maximum. This is the total amount of Hg that can be adsorbed per gram of Lake Lansing No. 1 sediment which is 31094.5 parts per million (or 3.11%). The reciprocal of the intercept of the line is "Kb" from

Figure 6 Langmuir Plot of the Adsorption Isotherm, Lake Lansing Sediment No. 1

R value of 0.92


LANGMUIR PLOT OF THE
ADSORPTION ISOTHERM
LAKE LANSING SEDIMENT NO.1
ppm Hg, liquid phase
ppm Hg, l.p./mg Hg, adsorbed per gm of sediment
0
5
10
15
20
.5
1.0
1.5

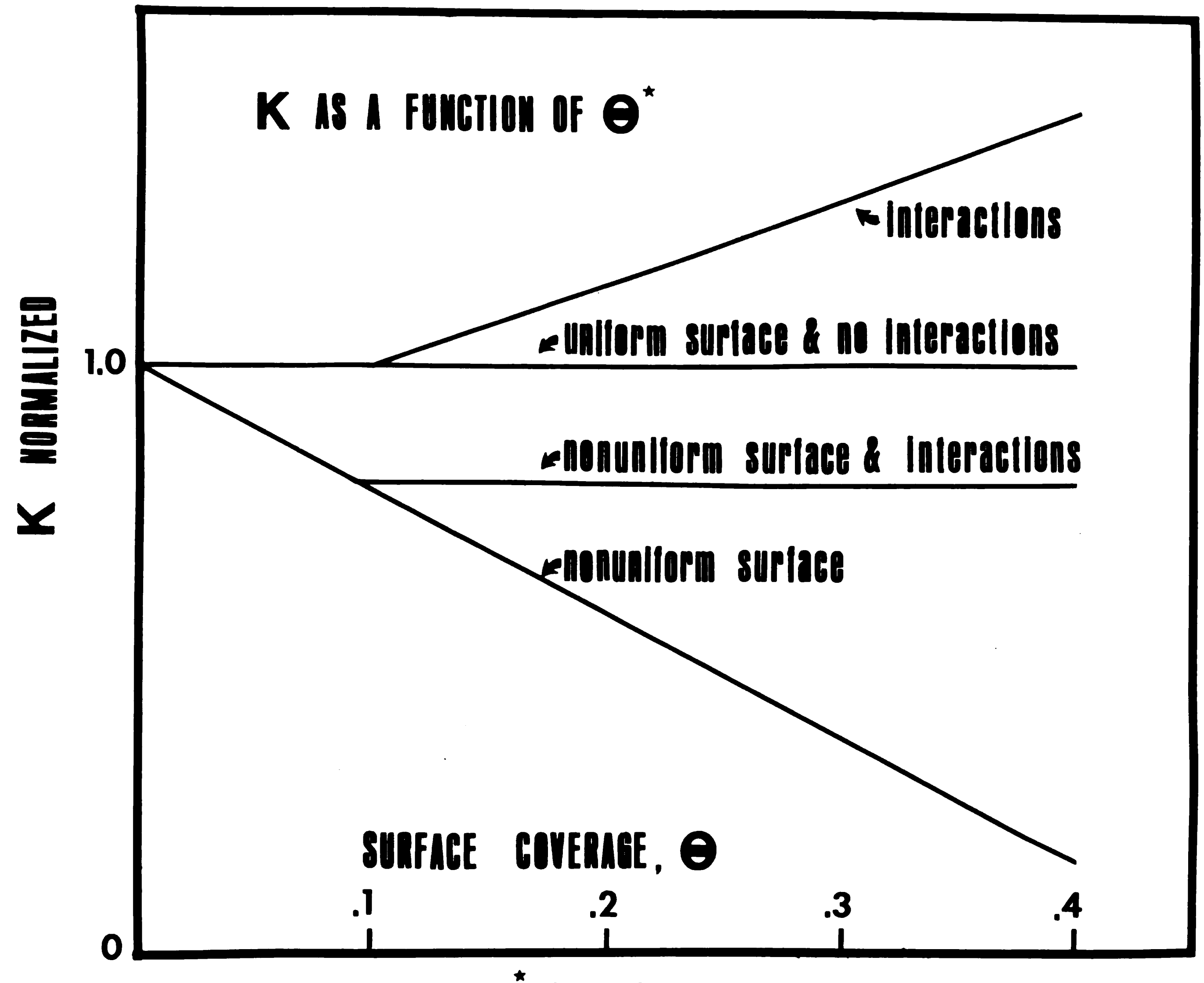
which "K" can be calculated, which is 0.661.

The Langmuir equation is obeyed only when the free energy of adsorption is constant, that is, when the sediment surfaces are uniform and there is no lateral interaction. Under these circumstances no adsorption isotherm would obey the Langmuir equation, but actually the equation is obeyed in numerous instances (Brunauer _et_ _al_., 1967). The equation for "K" is:

$$K = \frac{a\, e^{\frac{q}{kT}}}{k\,(2\ mkT)^{\frac{1}{2}}} \qquad (13)$$

The most important factor in "K" is the exponential term involving "q", the heat of adsorption. Most surfaces of sediment are energetically heterogenous, and at low coverages adsorption takes place on sites possessing the highest energies. A plot of "q" against θ, the fraction of surface covered, is ordinarily a decreasing function. Lateral interaction energies between adsorbed molecules increase the heat of adsorption. A plot of the lateral energies against "θ" is an increasing function. These two opposing effects in certain cases compensate for each other, making "q" and thus "K" approximately constant (Figure 7). If it were not for these compensating factors the Langmuir relationship would never by observed.

Figure 7 K as a Function of Surface Coverage



*FROM GRAHAM (1953)

DISCUSSION OF RESULTS

The rate of mercury uptake is a parameter capable of sensitive measurement and the effective mercury half-life in the liquid phase of an ecosystem can be calculated from that rate. Furthermore, it has been shown that the rate of uptake varies among sediment within the same lake. Initially, the rate of uptake in Burke Lake No. 1 was at least twice that of Burke Lake No. 2.

In the Burke Lake sediments a change was observed in the rates of uptake over an aging period of nine months (Figures 2 and 3). Through biologic degradation in the sediment, the components regulating the rate of mercury uptake may have been altered.

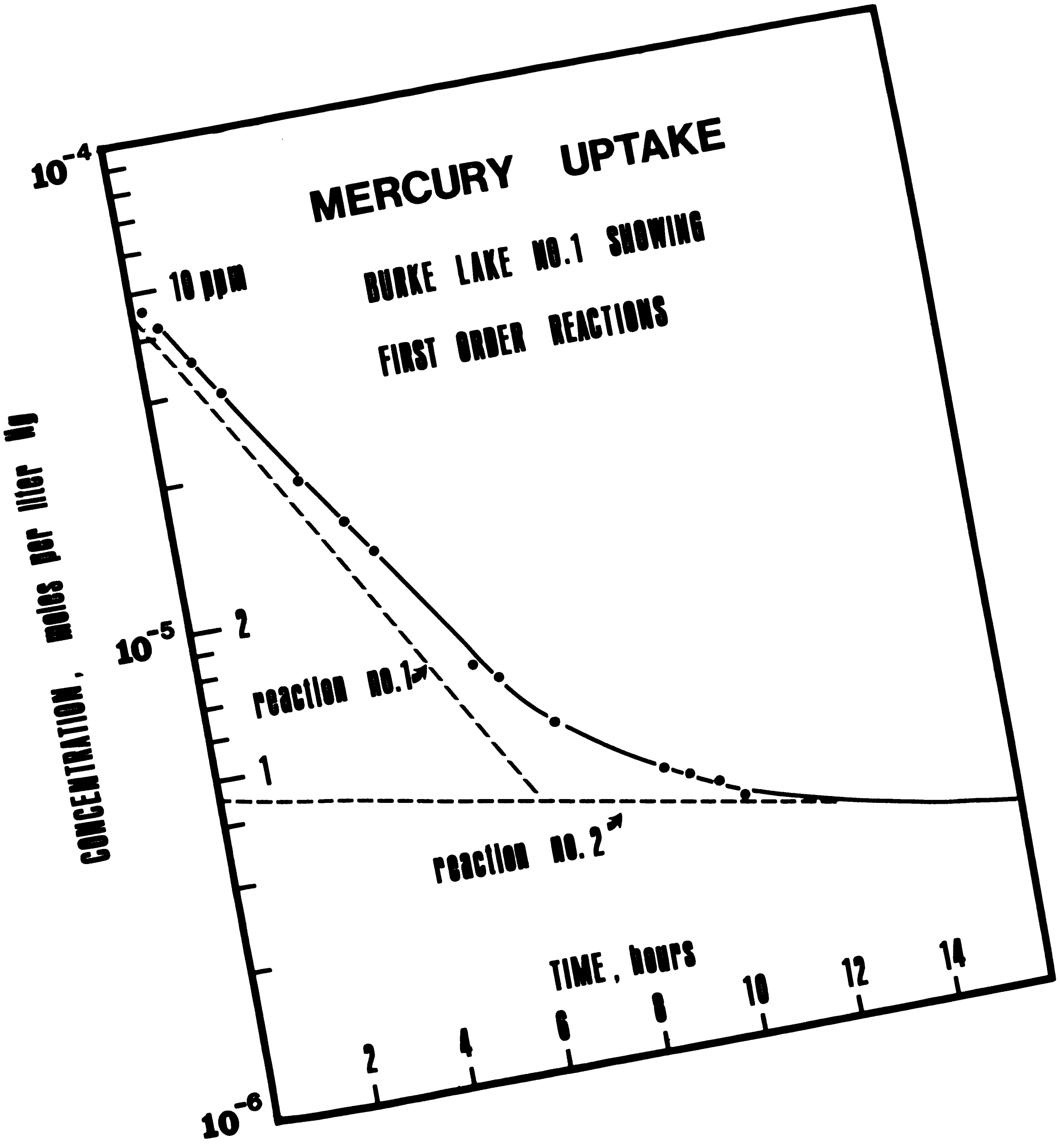
Therefore it is important that the sediment samples are tested as soon as possible after their removal from the lake bottom. In the case of Lake Lansing No. 1 and Burke Lake No. 2 the uptake phenomena follows the basic kinetic relationship of a simple two compartment exchange model. That is, it obeys a linear response with time, whereas with respect to Burke Lake No. 1 a nonlinear response was found. This would indicate several uptake reactions occuring at different rates. Separation of the different uptake rates are achieved by extrapolation of the slowest reaction and subtraction of its uptake contribution from the fastest rate (Figure 8).

Inorganic divalent mercury is the major species of mercury found in the natural environment. The conversion of other types of mercury compounds into inorganic divalent mercury ions in nature are well known (Landner, 1970). It is shown that copper(+2) will replace mercury(+2) in a lake bottom sediment thus increasing the mobility of mercury in the system. This response could become paramount in an instance where the sediment is laden with mercury and copper is introduced to the system causing a rapid increase in the mercury concentration of the body of water. This phenomena is probably due to the higher affinity of the organic fraction for complexing with copper than for mercury (Ellis, personal communication). The mercury complexing agents within the organic fraction are either amino acids or folic acids (Harlin, 1972).

The total adsorption capacity of a lake sediment was found to be extremely high: greater than 31,000 parts per million. Possibly the highest known concentrations in natural samples were 2,000 parts per million from the Detroit River (Upchurch, personal communication). For the most part, this high concentration is complexed to the organic fraction of the sediment and is tightly bonded. The "K" constant in the Langmuir equation is proportional to the bonding strength and is also a function of the shape of the adsorption iostherm curve (Ellis, personal communication). The high bonding strength is apparent in the steep initial increase in the plot of the adsorption isotherm.

The pH of the system is buffered at a pH 8 up to an

Figure 8 Mercury Uptake, Burke Lake Sediment No. 1, showing First Order Reactions


MERCURY UPTAKE
BURKE LAKE NO.1 SHOWING
FIRST ORDER REACTIONS
10 ppm
2
1
reaction no.1
reaction no. 2
CONCENTRATION, moles per liter Hg
10^{-4}
10^{-5}
10^{-6}
TIME, hours
2
4
6
8
10
12
14

equilibrium concentration of 20 parts per million by the presence of the organics (Figure 5) (Hoffman, 1969). Beyond the equilibrium concentration of 20 parts per million, the buffering capacity of the organics was overcome by the hypochloric acid used to dissolve the HgO. Mekhonina (1969) reported that the stability of mercury organic complexes is a function of pH. He stated that mercury organic complexes would dissociate at a low pH (6.3) to yield water soluble mercury. This phenomenon is apparent in the gradual decrease of the adsorption capacity of the Lake Lansing No. 1 sediment.

SUMMARY

The rate of mercury is a definite parameter of sediment type, and it is not a function of the concentration of the mercury in the overlying water (Spooner and Ehurlich, unpublished result). It is shown that mercury forms a strong organic complexing bond with the sediment (Page 30), and that the relative strength of the bond is less than that of copper (Figure 4).

The total adsorption capacity of the sediment studied was found to be extremely high (31,000 parts per million) (Page 25) and a function of the organic content of the sediment (Page 1). It is also noted that the adsorbing capacity of the organic fraction is decreased with a lower pH (6.3) (Page 30).

In studying the mercury uptake characteristics of a sediment it is important that both uptake and adsorption isotherm experiments be performed. By performing an uptake experiment the residence time of the mercury in the overlying water can be calculated. By performing an adsorption isotherm and using the concentration of mercury in the overlying water as the equilibrium concentration the amount of mercury adsorbed and its percent of the total adsorption capacity of the sediment can be determined.

Competition uptake experiments are also recommended to determine the exchange ability of mercury in the presence of other elements.

Further work in the area of elemental uptake into sediments would be very rewarding. An important parameter that can be measured is the "q" value in equation (13) which is the heat of adsorption of the mercury adsorbing sediment site. By performing adsorption isotherms at two or more different temperatures the heat of adsorption can be calculated from that data using the Van Laar equation.

Another important study would be to determine the uptake capacity of the pore waters of a sediment. Allied with the preceding study would be the investigation of the depth of diffusion of an element into the sediment, with respect to time and equilibrium concentration.

Information of this scope with respect to mercury, or any other element would be invaluable in compiling environmental impact statements.

APPENDICES

APPENDICES

DIFFUSION OF MERCURY INTO POLYETHYLENE VIALS

An important factor that must be taken into consideration when working with a highly toxic element as mercury is the possible health hazard to the experimentalist. Because of this hazard a constant and thorough monitoring of all vessels containing mercury was undertaken. Since the mercury solutions contained ^{203}Hg as a tracer the monitoring was through the measurement of radioactivity.

In the course of this monitoring procedure it was noticed that there was a loss of radioactivity, that can not be corrected for by the decay process, from the polyethylene vials used in the sample collection. This phenomena was observed when a polyethylene vial containing ^{203}Hg and Insta-Gel, a scintillation medium, is allowed to remain in the Packard liquid scintillation spectrometer for a number of days. By this method the radioactivity in the vial can be monitored over a period of several days. Several vials were monitored using this procedure (Figure 9). The Beta particles emitted through the decay of ^{203}Hg interact with the Insta-Gel to produce very small flashes of light, that are detected by a photomultiplier tube. As the ^{203}Hg ion diffuses out of the Insta-Gel mixture the probability that its Beta particle will interact with the Insta-Gel is reduced. An average diffusion rate of 12.4 percent with a standard deviation of 3.0 percent was calculated (Table 3). To combat this diffusion problem the sample vials were counted within two hours after their collection.

Figure 9 Mercury Diffusion into Polyethylene Vials

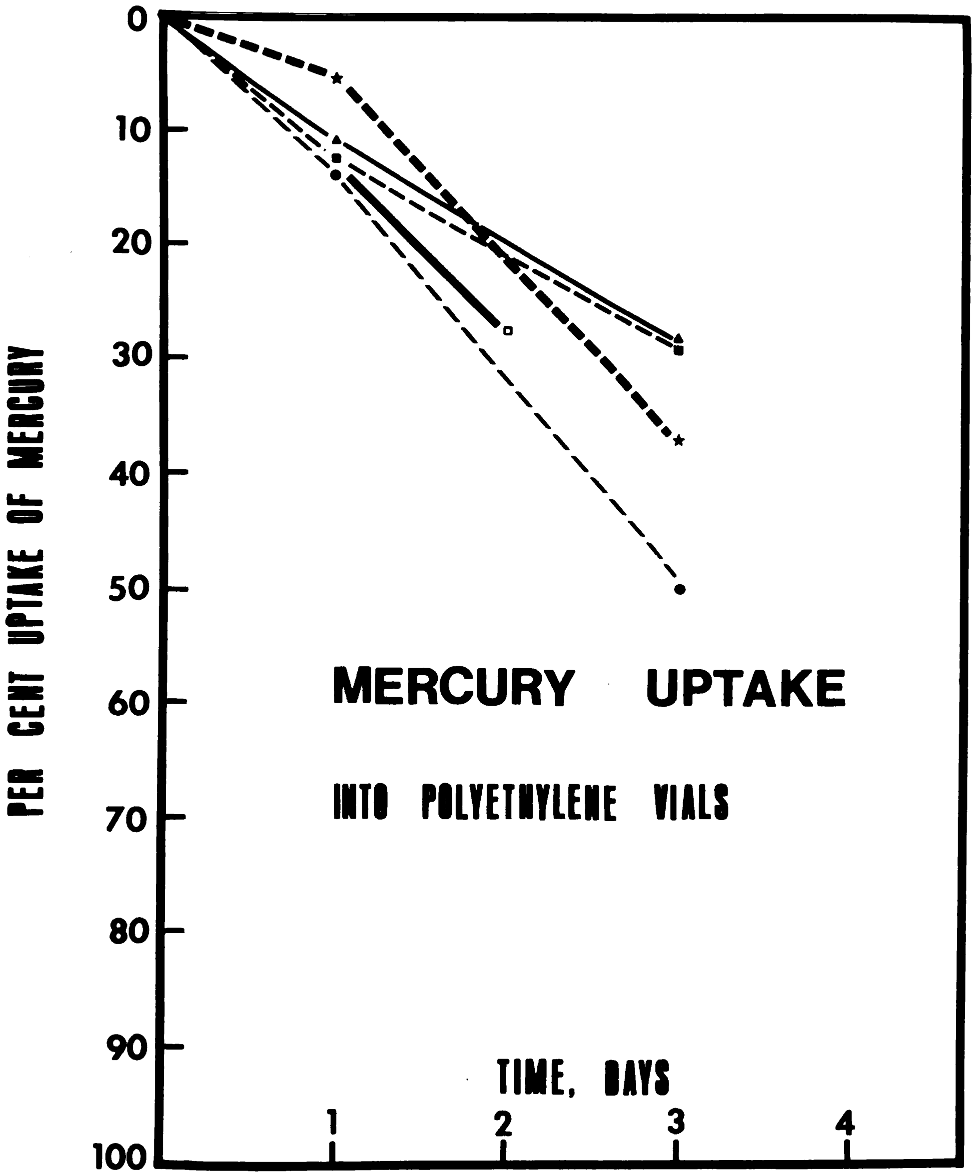
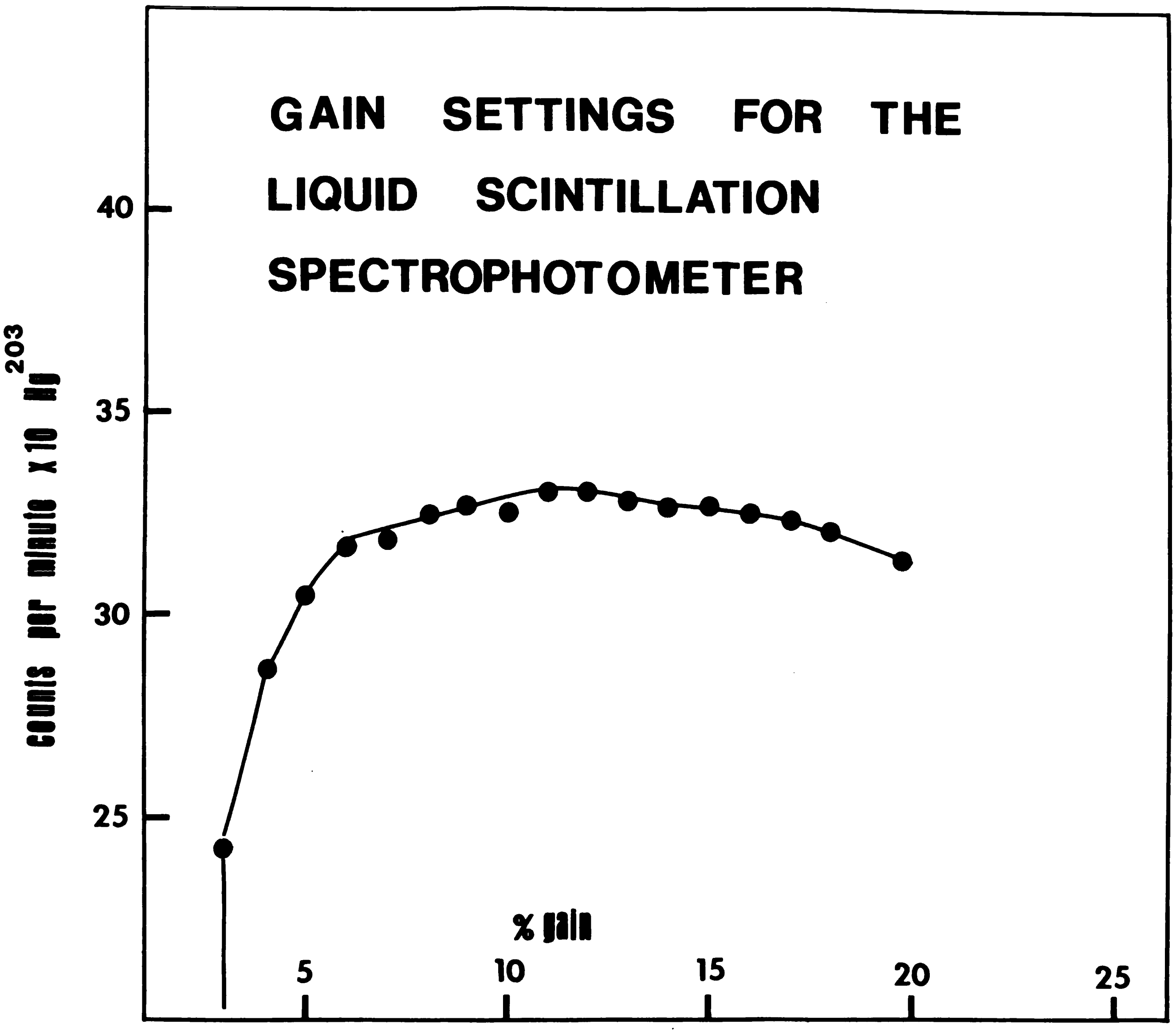

MERCURY UPTAKE
INTO POLYETHYLENE VIALS
PER CENT UPTAKE OF MERCURY
0
10
20
30
40
50
60
70
80
90
100
TIME, DAYS
1
2
3
4

TABLE NO. 3 DIFFUSION OF MERCURY INTO POLYETHYLENE VIALS

VIAL NO.	PERCENT UPTAKE PER DAY		
1	16.7		
2	9.7	MEAN	STANDARD DEVIATION
3	9.5	12.4	3.0
4	12.2		
5	13.7		

Figure 10 Gain Settings for the Packard Tri-Carb Liquid Scintillation Spectrophotometer



GAIN SETTINGS FOR THE LIQUID SCINTILLATION SPECTROPHOTOMETER

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