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ARSENIC PROFILES IN
SEDIMENTS AND SEDIMENTATION PROCESSES
ALONG THE SLOPE OF A LAKE BASIN

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ARSENIC PROFILES IN SEDIMENTS AND SEDIMENTATION PROCESSES ALONG THE SLOPE OF A LAKE BASIN

By

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ABSTRACT

ARSENIC PROFILES IN SEDIMENTS AND SEDIMENTATION PROCESSES ALONG THE SLOPE OF A LAKE BASIN

By

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Lake Lansing, Michigan was treated with sodium arsenite for macrophyte control in 1957. Seven 1.5 m sediment cores taken on a line through the littoral zone to the deepest portion of the basin were analyzed for arsenic in 5 cm increments. The objectives were to determine rates that sediment surfaces at different depths were returning to pre-treatment concentrations and to evaluate sedimentation processes affecting those rates.

Arsenic concentrations going downward from the surface in each core increased to some maximum. Below the maximum, there was a recession to background concentrations. Depth of peak concentrations followed two patterns; three littoral cores showed peak arsenic at 0.13 m from the sediment surface; four cores from progressively deeper regions of the lake showed a regular decrease in peak depth from 0.32 m to 0.17 m.

Magnitude of peak arsenic in each core increased with depth of water from which the core was taken. This suggested that 1957 treatment arsenic quantitatively precipitated to

the sediments as a function of depth of overlying water.

Sediment accumulation rates were calculated. They were low in the littoral, highest at 3.75 m, and decreased going into deeper water. Particle-size sorting of sediments along the basin's slope was measured. This work suggested that sediments originated from wetland vegetation at the edge of the lake. Turbulent movement of water in the shallows caused suspension and down-slope movement of small particles. Fewer particles of wetland origin were available for sedimentation beyond the region of highest fallout (3.75 m), thus accounting for progressively lower sedimentation rates in deeper portions of the basin.

In each sediment profile, there was a decline in arsenic from peak concentration to the 1980 sediment surface. Exponential curves were fit to these data. From them, the littoral sediment surface was predicted to reach pre-treatment concentration >100 years after treatment. Using this model, the pelagial sediment surface would return to background in 28 to 43 years. The latter rates are unrealistic; the rate of approach of deep sediments to background will be limited by the rate of approach of shallow sediments to pre-treatment arsenic concentrations. For Lake Lansing, that prediction is >100 years.

DEDICATION

To my wife, Lili

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INTRODUCTION

Inorganic and organic compounds of arsenic exist as natural components of terrestrial systems and are detectable in nearly all soils (Peoples, 1975; Walsh and Keeney, 1972). Arsenic is naturally distributed in high levels in rocks and minerals that contain iron, sulfur, and manganese where the element can be concentrated up to 2,000 ppm (Fleischer, 1973). The arsenic content of soils is generally much lower, averaging 6 ppm and ranging from 1 to 40 ppm (Bowen, 1966; Vallee et al., 1960). Where higher soil arsenic levels are found the source can usually be traced to anthropogenic activities. Mining, disposal of industrial wastes and widespread use of arsenical pesticides can elevate arsenic to concentrations several fold natural levels (Bishop and Chisholm, 1962; Vallee et al., 1960). Natural concentrations of arsenic in marine waters are usually low, ranging from 0.15 to 6.0 μg As l^{-1} and averaging near 2 μg As l^{-1} (Woolson, 1975). The highest concentrations in inland waters have been found in hot springs, like those in Nevada and Wyoming, where two examples showed arsenic levels of 2,300 and 500 μg As l^{-1} (Hem, 1959). Ritchie (1961) reported 8,500 μg As l^{-1} in a New Zealand hot spring. A survey of other fresh water systems in the U.S. revealed concentrations ranging from 10 μg As l^{-1} to 140 μg As l^{-1}, with 76% of the 726 water samples analyzed falling below 10 μg As l^{-1} (Durum et al., 1974). A survey of arsenic content of U.S. lakes placed 94% of the 1577 lakes sampled between 10 and 340 μg As l^{-1}, with an average value of 60 μg As l^{-1} (Kopp and Kroner, 1967). As with soil, high arsenic levels in lake water can often be attributed to human impact. Release of industrial and domestic waste, burning of fossil fuels, and application of arsenical pesticides are principal causes of artificially elevated arsenic in lakes and streams (Shapiro, 1971; Lis and Hopke, 1973; Aston et al., 1975; Domogalla, 1926). Of particular interest here is the large scale use over the last several decades of sodium arsenite as an herbicide to control aquatic vegetation (Kobayashi and Lee, 1978; Ferguson and Gavis, 1972; Ruppert et al., 1974; Mackenthun, 1950). This activity, coupled with the ensuing potential of acute, or more likely, chronic toxicity of arsenic to humans and non-target aquatic organisms, has generated interest in the fate of applied arsenic in lake systems (Bails and Ball, 1966; Cowell, 1965; Gilderhus, 1966; Crosby, 1966).

Studies of the movement of arsenic in lakes or ponds treated with the herbicide sodium arsenite (Na_2AsO_2) show that aqueous levels decrease within a period of weeks after application. The mechanism of this decline has generally been attributed to coprecipitation of arsenic from the water column with iron oxides, followed by sorption of the iron-arsenic complex by sediment particles (Crecelius, 1975; Seydel, 1972; Sohacki, 1968; Mackenthun, 1964). This mechanism is supported by observed increases of iron-associated arsenic in surficial sediments (Crecelius, 1975; Kanamori, 1965; Kobayashi and Lee, 1978). Because of the complex chemistry of arsenic and unknown patterns of lake sedimentation, the fate of sediment arsenic has not been fully described. Most of the work toward this end has involved analysis of arsenic content with depth in single sediment cores, or in several cores taken from widely separated locations in a lake (Crecelius, 1975; Kobayashi and Lee, 1978). Arsenic profiles in single cores have been used to reflect the timing of arsenic loading to the sediments (e.g., Crecelius, 1975), and to suggest the change in potential of sediment arsenic to be recycled into overlying water. However, interpretation of results from a single or widely separated cores is limited in that it cannot be extended to develop a model for sediment arsenic distribution in the basin as a whole.

In this study, a series of cores was taken along a line running from a wetland fringe, on Lake Lansing, Michigan, across the littoral zone of the lake, and down the pelagial slope to the deep plain of the lake. Batterson (1980) has shown that the vertical profile of arsenic in cores from the deep basin have an arsenic peak related to a single sodium arsenite treatment applied in 1957 for macrophyte control. The Lake and Stream Improvement Section of the Michigan Department of Conservation treated areas with a total of 3800 liters of sodium arsenite in June of that year (Roelofs, 1958). This treatment resulted in an input of 2920 kilograms of arsenic. Historical records indicate that this has been the sole arsenic treatment of the lake. Batterson (Ibid.) showed arsenic loading to the lake from atmospheric fallout and overland flow was negligible. He also demonstrated that the arsenic content of the upper 5 cm of sediments was 2 to 6 times pre-treatment concentrations found in deep portions of cores. For example, the arsenic content of pre-treatment sediments in the south basin was in the range of 17 to 20 μg g^{-1} dry weight; 46 μg As g^{-1} were found in surficial sediments near shore and 125 μg As g^{-1} just beyond 7 m contour (cf. Figure 1). The significance of high sediment arsenic levels stems from the potential of sediment to contribute soluble arsenic to overlying water. This was suggested when in 1978 the arsenic of the lower pelagial water of the south basin increased from 14 to 115 μg As l^{-1} during a period of summer stratification.

In his cores from the deep plain of the lake, Batterson (Ibid.) observed a recession in the arsenic concentration from 1957 peak with the addition of recent sediments to the lake bottom, and calculated the rate at which sediments of the pelagial plain were returning to the background concentration. Since sedimentation rates and sediment mixing processes are likely different at different depths along the slope of the lake basin, he could not predict from his data the time necessary for sedimentation to ameliorate the effects of the 1957 treatment in the basin as a whole. The purposes of this study were: (1) determine the nature of the arsenic profiles in littoral sediments and sediments of the pelagial slope, (2) to use these profiles to determine sedimentation rates and the degrees of mixing of new sediments with base sediments at different depths in the lake, and (3) to use the profiles to predict the rates at which sediments at different depths would approach background, thus bringing sediment surfaces to pre-treatment arsenic concentrations.

MATERIALS AND METHODS

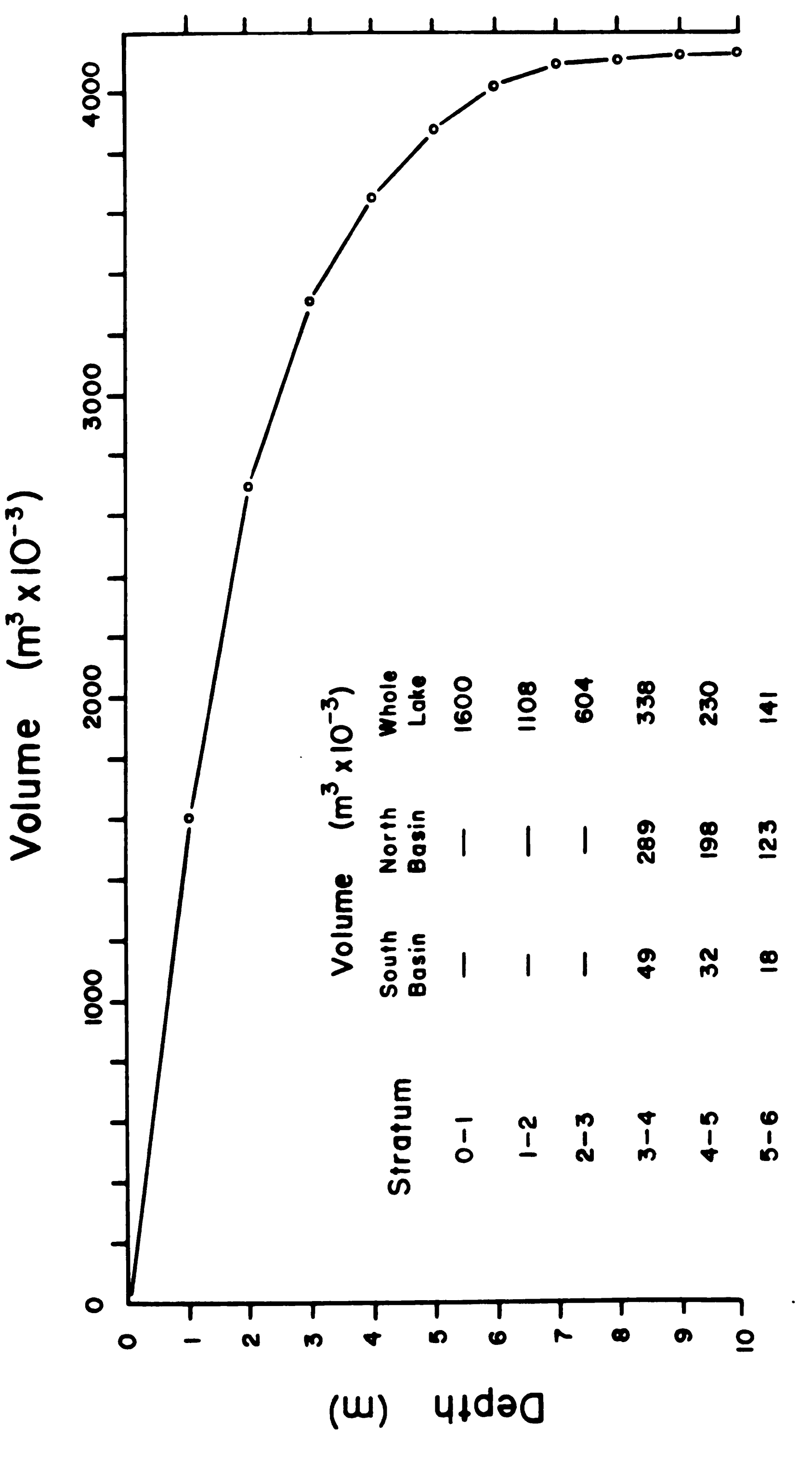
Lake Lansing is located approximately 5.6 kilometers northeast of the city of East Lansing, Michigan. The lake has a surface area of 1816 x 10^3 m^2, mean depth of 2.3 m, and a maximum depth of 10 m. The littoral zone of the lake extends to the 3 m contour; 77% of the lake surface area lies over the littoral zone (Figure 1). The volume of the lake is approximately 4,124 x 10^3 m^3 (Figure 2). The bathymetry of Lake Lansing shows it is divided by a shallow bar into a north and a south basin. Each basin has a tendency to thermally stratify in the summer (Figure 3) and develop anoxic conditions in the lower pelagial regions. The tendency for oxygen loss is particularly evident in the south basin (Figure 4).

The slope of the lake basin along transect line AB shown in Figure 1 was determined by gauging the depth of water at measured distances from the shore. This was accomplished by lowering a plumb through augered holes in the ice cover. The plumb weight consisted of a 25 cm diameter disc to minimize error caused by sinking into the soft sediments.

Seven cores were obtained along line AB during July to August, 1980. Since the sediments along the transect were loose and unconsolidated, they were sampled by freezing the sediment onto the exterior surface of tubing which extended

Figure 1. The Lake Lansing basin showing areas treated with sodium arsenite in 1957 (stippled), μg As g^{-1} of dry surficial sediments (from Batterson, 1980), and the position of the sampling transect (AB) used in this study.

Figure 2. Depth-volume curve for Lake Lansing, with tabled volumes for strata of the two deep holes and the lake as a whole.



Stratum	Volume (m^3 x 10^{-3}) South Basin	North Basin	Whole Lake
0–1	—	—	1600
1–2	—	—	1108
2–3	—	—	604
3–4	49	289	338
4–5	32	198	230
5–6	18	123	141
6–7	8	61	69
7–8	1	24	25
8–9	—	8	8
9–10	—	1	1

Figure 3. Temperatures (C^{o}) in the south basin of Lake Lansing during 1978.

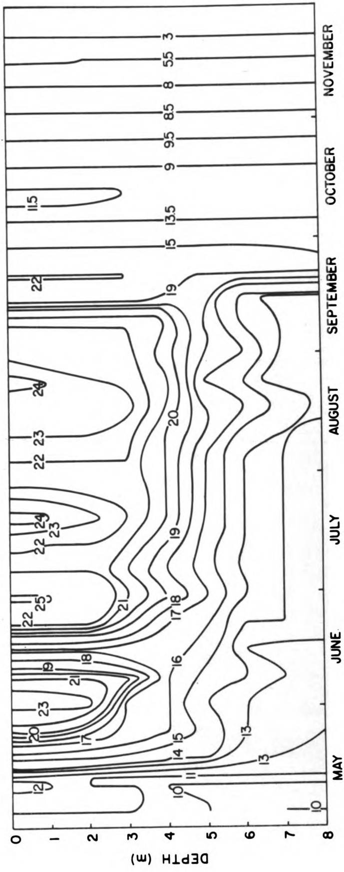
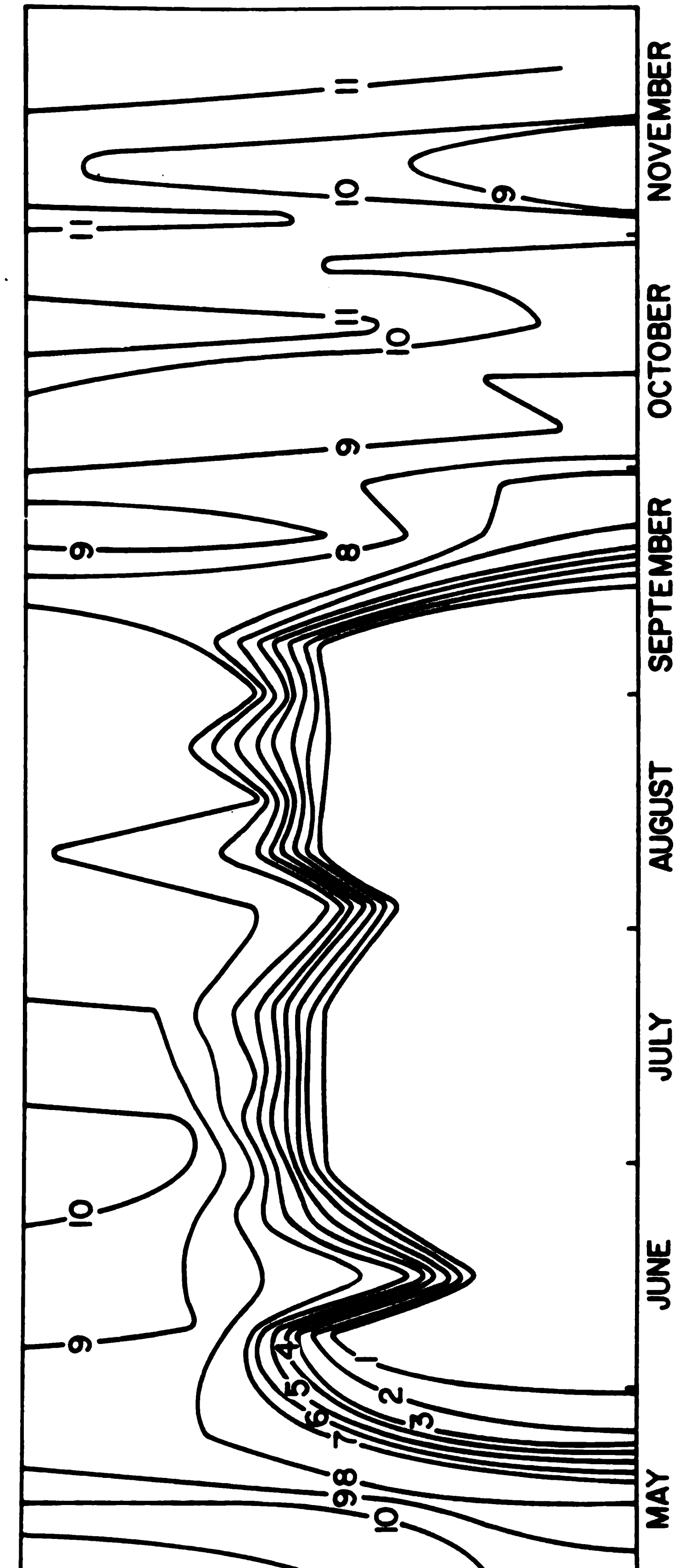

DEPTH (m)
MAY
JUNE
JULY
AUGUST
SEPTEMBER
OCTOBER
NOVEMBER

Figure 4. Dissolved oxygen concentrations (mg l^{-1}) in the south basin of Lake Lansing during 1978.


DEPTH (m)
0
1
2
3
4
5
6
7
8
MAY
JUNE
JULY
AUGUST
SEPTEMBER
OCTOBER
NOVEMBER

from the water surface and penetrated the sediments a known distance. Lengths of 5.08 cm o.d. thin-walled aluminum electrical conduit were used which were threaded and joined by couplings. Added to the water depth at each coring site was the length of the sediment core desired. Sections of tubing were then selected which would exceed that length by several feet to provide excess tubing above the water. The joints were not water-tight so silicone sealant was applied to the threads to accomplish this. The bottom of the sampling tube was stoppered and lowered into the water. Additional lengths were added until the stoppered end was just above the sediment surface. When the last section was attached, the tube was carefully pushed into the sediments to the appropriate depth. After insertion into the sediments, pelletized dry ice was added to the end of the tube extending above the water surface. The amount added was enough to freeze the sediments as well as a small portion of water above the sediment-water interface. Replenishment of dry ice was maintained at a rate to offset sublimation. Thirty minutes after the initial addition of dry ice, the samples were retrieved. As the tube was pulled out of the water the sections were uncoupled down to the frozen sample. After the sediment sample was removed from the lake, the unfrozen exterior layer was stripped away. The sample was then placed in plastic and the tube repacked with dry ice for transportation to the laboratory. In the laboratory, the dry ice was removed from the tube and replaced with tap water. This melted the sediment in contact with the tubing and allowed for the tube to be pulled free. The frozen sample was then cut into 5 cm sections using an electric band saw. The exterior of each doughnut-shaped piece was rinsed with ion-free water and placed in a labeled plastic bag. There were two reasons for the rinsing: to wash away any contamination that might have resulted from the sectioning process or sediment contact with the aluminum tube, and to remove dislocated particles from the core surfaces.

The frozen samples were then dried in a Napco model 630 forced air drying oven at 75^{o}C for 72 hours and dry samples were ground with mortar and pestle. From each of the well-mixed ground samples approximately one gram of sediment was removed and dried at 105^{o}C for 24 hours. The sample was then introduced into an acid-washed and pre-weighed two dram polyvial and weighed. After weighing, the polyvials were heat-sealed and taken to Michigan State University's nuclear reactor facility for neutron activation analyses. For each group of samples that was irradiated there were three standards for quantifying the analyses. Two of the standards were obtained from the Natural Bureau of Standards and prepared for introduction to the polyvials according to the procedure recommended by the Bureau. These were Standard Reference Material 1645 (River sediment) and 1571 (Orchard leaves). The other standard was a 2 ml solution containing 150 μg As ml^{-1}.

A Triga Mark I nuclear reactor was used for irradiation.

Thirty-seven sediment samples and three standards were introduced into a 40-position specimen rack that was rotated during irradiation to establish uniform flux for all sample positions. A flux rate of 10^{12} neutrons cm^{-2} sec^{-1} was used. Sixteen to twenty hours following irradiation (allowing for the partial decay of ^{24}Na activity), the samples were counted for 1000 seconds live-time with a 76.2 cm^3 active volume Ge (Li) detector having a relative efficiency of 15% and an energy resolution of 1.8 KeV FWHM at the 1.333 MeV photopeak of ^{60}Co. The source-to-detector geometry was kept constant for all counts and the detector resolution was sufficient to completely resolve the ^{76}As peak (559 KeV) and the adjacent peak of ^{82}Br (554 KeV). The gamma-ray spectrum from each sample and standard was analyzed by a Canberra Series 80 multi-channel analyzer. This analyzer computed the peak net area which is the number of counts in a peak that are above an average background level. Standards and samples were corrected for decay during counting by the following equation:

$$A_c = Ae^{\frac{0.693t}{\lambda^{1/2}}}$$

where A_c = Area corrected for decay between counting time of the sample and standards (net count)

A = Area of ^{76}As (net count)

e = Base of the natural logarithms

$\lambda^{1/2}$ = Half-life of ^{76}As = 1584 minutes

t = Finishing time in minutes

The mass of arsenic in the sample was derived using the time corrected counts of the standards.

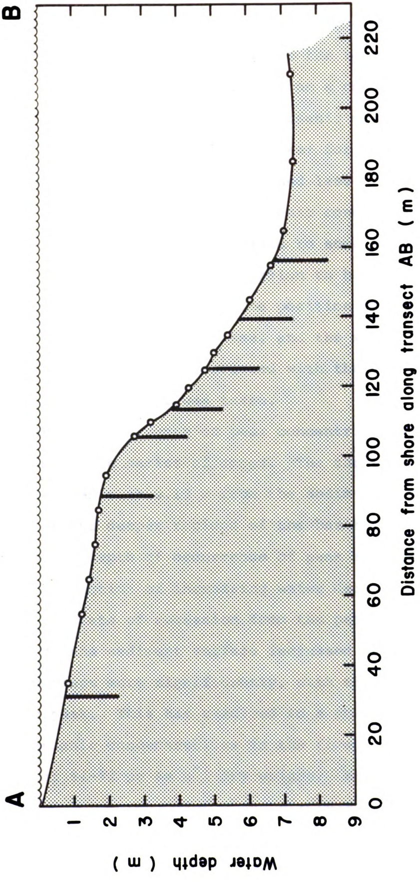
Estimates were made of the mass of dry sediments in each 5 cm section of core. The volume of cores was obtained by determining the cross-sectional area of frozen sediments plus the sampling pipe. The cross-sectional area of the pipe was subtracted from the total area and the remainder was multiplied by the length of the core section (5 cm). The density of sediments in core sections was measured by a water displacement method. A known volume of ion-free water was added to a graduated cylinder. Longitudinal sections of frozen core material were placed in the cylinder. When the frozen piece thawed, volume in the cylinder was recorded. The volume of the piece of the core was calculated by subtracting the initial volume in the cylinder from the final volume. Contents of the cylinder were rinsed with ion-free water into a pre-dried and weighed aluminum tray and dried in an oven at $95^{0}C$ to a constant weight. The weight of the dried material was divided by the calculated volume of the core fragment to obtain an estimate of the density of the sediments in the fragment; g DW cm^{-3} in the fragment multiplied by the volume of 5 cm core sections yielded the total dry weight in core sections. Additional analyses of the physical features of cores were made during this study. The results of this work are included here as Appendix Table 1.

In February of 1981, surficial sediment samples were collected along line AB for particle size fractionation. Samples were collected with an Ekman dredge at points corresponding to those from which cores had been taken. Each sample was mixed thoroughly and three 200 ml volumes were withdrawn. These replicates were dried to a constant weight at 95°C. In addition, three 200 ml subsamples from each dredge were rinsed through a stack of U.S. Standard Sieves; numbers 10, 20, and 50 (pore sizes 2, 0.833, and 0.227 mm, respectively) were used. A measured volume of tap water was used for the rinse. Particles retained by each sieve were emptied into pre-weighed aluminum trays and dried at 95°C to constant weight. The weight of particles passing through the smallest sieve (0.227 mm) was calculated by subtracting the sum of the dry weights of the larger size classes from the dry weight of whole samples. The data obtained were used to calculate the percent dry weight contribution of each particle size fraction in the surficial sediments along the slope of the lake basin.

RESULTS

The profile of the south basin along transect AB showed two zones with distinct gradients (Figure 5). Within the first zone extending from shore to approximately 105 m lakeward, the basin gradually declined to 2 m below the lake surface. The slope of the basin in this region was approximately 1:50. Within the second zone, which extended from the edge of this shallow shelf to a point about 50 m lakeward, the basin dropped 5 m; the slope increased nearly fivefold. The basin profile suggests that there was an extensive shallow region where the sediment surface was subject to wind-generated water movement that could resuspend previously sedimented materials. Since aquatic plant cover tends to stabilize the sediments, resuspension processes may be most intense during periods when submersed plant biomass is low, as during spring overturn. However, potential for contact between these sediments and moving water is higher than in the deep region of the basin. The vertical bars of Figure 5 indicate the position and depths of cores taken in this study. Core sampling was concentrated in the portion of the basin where slope was greatest. One would expect sedimentation and mixing processes to change most rapidly in that region of the lake where influence of water movement changes rapidly (Hutchinson, 1957).

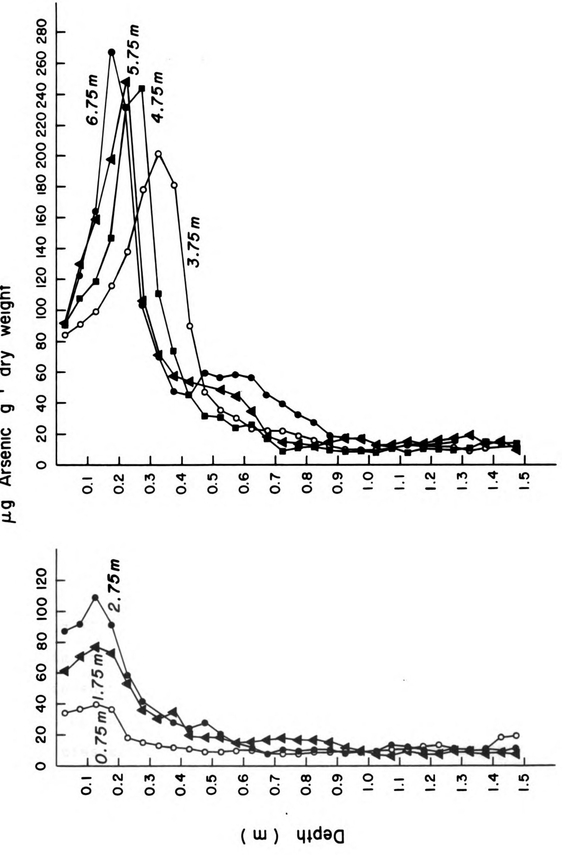
Figure 5. Shape of the south basin of Lake Lansing along line AB based on measurements of depth of water at metered distances from the edge of the lake. Bars indicate location of sediment core sampling.


A
B
Water depth (m)
1
2
3
4
5
6
7
8
9
0
20
40
60
80
100
120
140
160
180
200
220
Distance from shore along transect AB (m)

The concentration of total arsenic with depth in each sediment core taken along the slope of the south basin is present in Figure 6. It is obvious from a comparison of the curves that the total amount of arsenic was different in cores from different depths in the lake; for example, cores from the shallows of the lake contained less arsenic than cores from the pelagial region. Arsenic concentrations going downward in each core increased to some maximum point. The maximum was followed by a recession to background levels in deep portions of the cores. The position of the arsenic trace with respect to the abscissa, and the magnitude of peak arsenic in each core, increased with the depth of the water from which the core was taken.

The depth of occurrence of peak concentrations followed two patterns in the series of cores. The three shallow cores showed peak arsenic at 0.13 m from the sediment surface. The four cores of the deeper regions of the basin showed a regular decrease in depth of occurrence of peak arsenic concentration as a function of increasing water depth. Figure 6 shows that the rate of recession from the peak arsenic concentration to the sediment surface increased with depth of water, or perhaps more significantly, with the magnitude of the arsenic peak. This has resulted in a convergence of the surficial arsenic concentrations in the five deepest cores to a range of 84-92 μg As g^{-1} dry weight. Arsenic profiles were obtained for cores taken along transect 1 in Figure 1, as well as along line AB. However, stations on transect 1

Figure 6. Concentrations of total-arsenic with depth in sediment cores taken along line AB of the south basin of Lake Lansing.

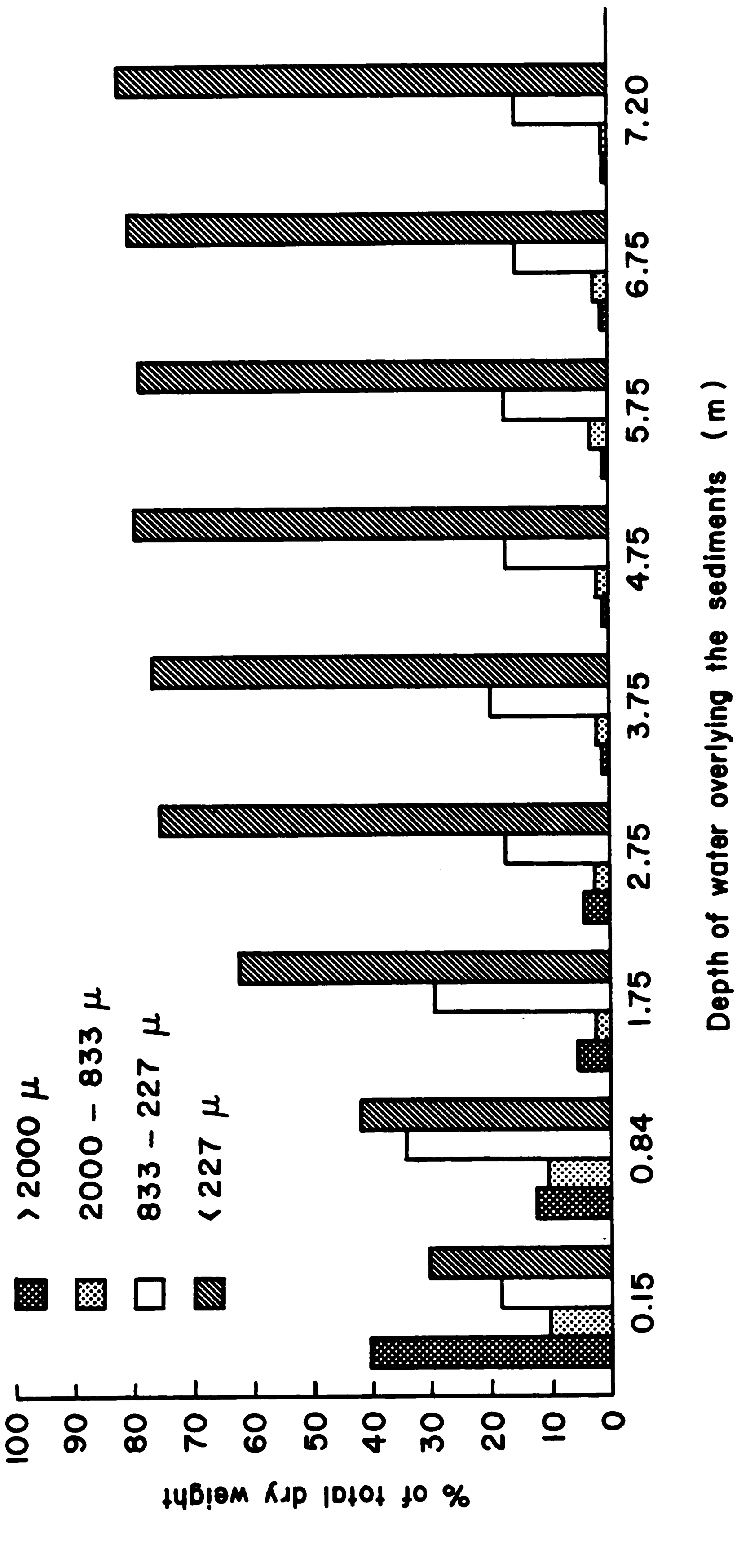

µg Arsenic g⁻¹ dry weight
Depth (m)
6.75 m
5.75 m
4.75 m
3.75 m
2.75 m
1.75 m
0.75 m
0 20 40 60 80 100 120 140 160 180 200 220 240 260 280
0 20 40 60 80 100 120
0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0 1.1 1.2 1.3 1.4 1.5

were used during 1978 and 1979 in intensive sampling programs for water chemistry and zooplankton and benthic populations. The data from transect 1 is included here in Appendix Tables 3 and 4. In general, the depth profiles for arsenic along transect 1 showed patterns similar to those described for line AB. Portions of individual profiles appear to have been badly disturbed by sampling.

Decreases in arsenic from peak concentrations toward the surface in each core suggests that newly deposited sediments of relatively low arsenic concentration were burying the arsenic introduced in 1957. The low relief topography of the land surrounding the lake, the relatively small size of the drainage basin, the absence of appreciable stream flow to the lake, and the lack of eroding beaches (Batterson, 1980) argue for the position that inorganic soil materials do not contribute substantially to the buildup of sediments. They appear to accumulate from the breakdown of vegetation from residential shorelines and wetlands around the lake (Knoecklein, 1981) and from submersed plant remains. Wetlands dominate the shoreline of the south basin of Lake Lansing (Figure 1). The shoreward origin of the transect used in this study was located at the edge of a wetland. That in-shore sediments originated in the wetland was suggested by the common occurrence of macroscopic fragments of plant tissues in the shallows. Currents generated by wind action on the lake were expected to sort out particles on the sediment surface in relation to their size and the velocity of the currents in a manner that is well known for streams (Wetzel, 1975).

The percent dry weight contribution of each of four particle sizes constituting the surficial sediments along line AB are presented in Figure 7. Regular changes occurred in the largest and the smallest size categories. The largest particles (> 2000 μ) were made up of fibrous fragments of the wetland vegetation. The largest size class made up 40.6% of the sediments by weight at 0.15 m water depth. This size category dropped to 1.3% at 3.75 m. At depths greater than 3.75 m the change in this size was less than 1%. In contrast, the smallest particle size (< 227 μ) made up 30.4% of the sediments at 0.15 m water depth, and increased to 75% at 2.75 m. This size class did not change appreciably beyond 2.75 m. The data of Figure 7 show that particles of the sizes measured were sorted by currents primarily in depths of 2.75 m and less; size distribution was essentially the same for depths of 3.75 m and more. There may have been significant differences in particle size distribution within the smallest size class (< 227 μ) between the four deepest stations; if so they would not be evident with the techniques used here.

Figure 7. Mean of percent dry weight contribution of each of four particles sizes constituting the surficial sediment of the south basin along line AB.



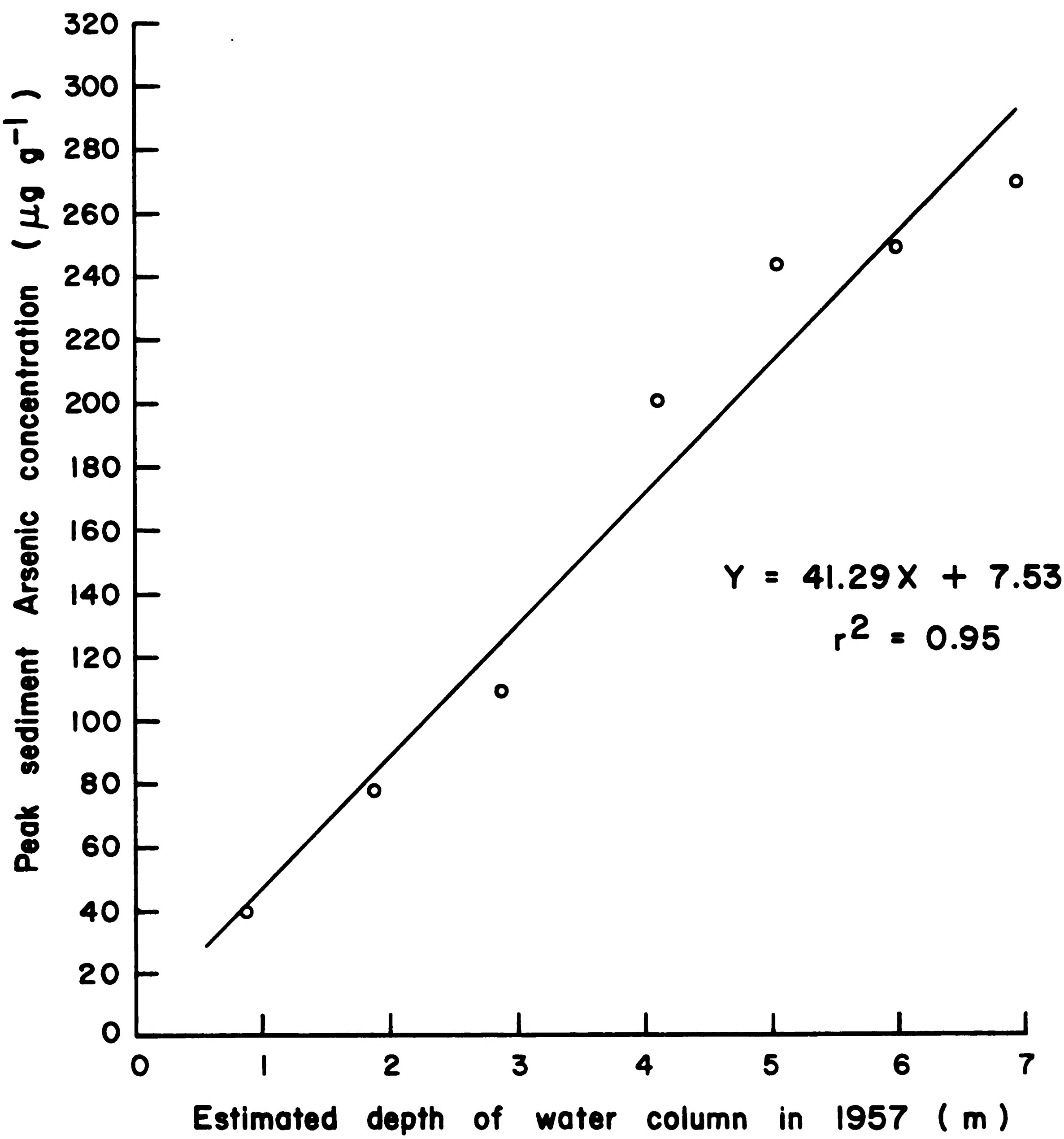
> 2000 μ
2000 – 833 μ
833 – 227 μ
< 227 μ
% of total dry weight
0
10
20
30
40
50
60
70
80
90
100
0.15
0.84
1.75
2.75
3.75
4.75
5.75
6.75
7.20
Depth of water overlying the sediments (m)

DISCUSSION

The profiles of arsenic concentration in the cores taken along line AB in the south basin of Lake Lansing (Figure 6), show that the peak concentrations increase as the depth of water overlying the sediments increased. This suggests that 1957 treatment arsenic quantitatively precipitated to the sediment surface as a function of depth of overlying water. That relationship is presented in Figure 8. The horizontal scale in the figure was obtained using estimated sedimentation rates at sampling points along the slope of the basin; these are discussed later in this section. Part of the scatter in this relationship may result from decreases in the magnitude of arsenic peaks since 1957 due, for example, to the transport of precipitated arsenic from the sediment surface downward in the sediment profile.

Three conditions must be met for the relationship in Figure 8 to be accepted as valid. Arsenic sprayed over weed-beds at the time of lake treatment must have become well mixed in the volume of water in the lake before precipitation. Some mechanisms for the removal of arsenic from the water column were required. Once on the sediment surface, arsenic must have remained relatively immobile, thus allowing the maximum concentration in each core profile to represent the depth in the sediments of the 1957 sediment surface.

Figure 8. Regression of maximum arsenic concentrations found in cores from Lake Lansing on depth of the water column at coring stations corrected for sediment accumulation since 1957.



Peak sediment Arsenic concentration (μg g⁻¹)
320
300
280
260
240
220
200
180
160
140
120
100
80
60
40
20
0
0
1
2
3
4
5
6
7
Estimated depth of water column in 1957 (m)
Y = 41.29X + 7.53
r² = 0.95

Evidence for extensive mixing of arsenic in the lake prior to fallout on the sediments comes from the work of Batterson (1980). His core taken from the north deep basin of Lake Lansing had an arsenic profile and peak arsenic concentrations remarkably similar to his core from the deep portion of the south basin. This was observed even though the distances from 1957 treatment areas to his coring areas were substantially different. Batterson's surficial sediment data is given in Figure 1 of this paper. They show that arsenic concentrations in surface sediments along six transects in the lake decreased from the shallows to deep water in the same manner observed along line AB. The prediction from the relationship presented here between peak arsenic concentration and depth of overlying water in 1957 (Figure 8) is that peak arsenic concentrations marking the time of treatment in 1957 occur beneath the surface sediments sampled by Batterson over most of the lake bottom. That Lake Lansing was likely well mixed after the arsenic treatment in July of 1957 is further suggested by the weak thermal stratification that exists in the lake in summer. An example of this is shown here in Figure 3. Vertical temperature differences occur only in the small volume of water over the deep holes in the lake. High south and southwest winds in summer tend to prevent the development of a stable metalimnion.

The principal mechanisms involved in the loss of inorganic arsenic from the water column of lakes has received considerable attention in the literature. Ferguson and Gavis

(1972) suggest that arsenite, As (III), tends to be oxidized to arsenate, As (V), in aerobic water. Arsenite is most likely to exist as the anion $HAsO_4^{2-}$. Chemically similar to phosphate, it can be absorbed, occluded or precipitated with hydrous ferric oxides. Kobayashi and Lee (1978) studied accumulation of arsenic in sediment of five Wisconsin lakes treated with sodium arsenite. They found a strong coefficient of correlation between arsenic and iron in the sediments of Lake Mendota. They concluded that iron controls arsenic levels in the water column through sorption of arsenate by ferric hydroxides, followed by precipitation to the sediment. Crecelius (1975), studying the geochemical cycle of arsenic in Lake Washington, found a strong correlation between sediment iron and arsenic ($r^2 = 0.94$). He suggested that arsenic is associated with the iron phase which causes a major portion of arsenic to be removed from Lake Washington water and accumulated in the sediments. Seydel (1972) studied the distribution and circulation of arsenic through water, organisms, and sediments of Lake Michigan. She suggested that accumulation of the arsenic in the sediments up to 28.8 ppm was due to the coprecipitation of arsenic with iron.

In snaerobic water of a hypolimnion or in anaerobic sediments, arsenate tends to be reduced to arsenite (Ferguson and Gavis, 1972). Ferguson and Anderson (1974) reported that at low Eh in the presence of sulfide (S^{2-}), arsenite should be effectively removed from the water column as insoluble sulfides. The experiments of Batterson (1980) lead to the conclusion that iron controlled the solubility of inorganic arsenic in aerated freshwater systems, while sulfide controlled the solubility in anoxic systems. Because of these mechanisms, significant quantities of soluble arsenic are expected only where the redox status permits oxidized sulfur and reduced iron to exist simultaneously. Batterson (1980) showed that these conditions can occur in the hypolimnion of Lake Lansing; for example, arsenic increased from 14 to 115 $\mu g\ l^{-1}$ in deep water of the south basin in a two-week period in the summer of 1978. However, he showed the conditions were short-lived and were not typical. Similar elevations in hypolimnetic arsenic were not observed in the winter, spring or summer of 1979. This discussion argues for the position that arsenic, well mixed in the volume of Lake Lansing, would fall out on the sediment surface and tend to stay there as insoluble compounds of iron or sulfur.

A question can arise as to the immobility of the arsenic peak deposited on the sediments as a result of the 1957 treatment. Carighan and Flett (1981) showed that phosphorus in lake sediments could migrate upward and accumulate near the mud-water interface. In spite of the similarity between arsenic and phosphorus chemistry, an important difference is that phosphorus does not combine with sulfide as arsenic does. Crecelius et al. (1975) found that the concentration of total arsenic was high in the surface sediment of Puget Sound in Washington and dropped to background levels of 10 ppm with the depth in the core. They suggested that high arsenic at surface sediment was the result of a recent additional input of arsenic from a large copper smelter. In the same study, sediment accumulation rates were determined by the lead-210 technique. They showed that the arsenic level started to increase in the cores at the time when the copper smelter started to operate. Crecelius (1975) also found that the position of peak concentrations of arsenic for five different locations in Lake Washington varied with sedimentation rate. In areas with lower sedimentation rates, peak concentration occurred at a shallower depth in sediment cores.

Kobayashi and Lee (1978) studied accumulation of arsenic in sediments of lakes treated with sodium arsenite. Arsenic profiles were developed for cores from five lakes. They used sedimentation rate for eutrophic lakes in the study area from Bartleson (1970) and showed that the depth of peak concentration corresponded to treatment time. In two lakes (Big Cedar and Pewaukee) with the same sedimentation rates, the difference in depth of peak concentration was due to time of treatment. From these considerations, it is concluded that arsenic deposited on Lake Lansing sediments following treatment in 1957 has been relatively immobile.

Sediment accumulation rates along line AB can be calculated using the depth of peak arsenic concentration in each core to represent the 1957 sediment surface. The density of core sections (g DW cm^{-3}) was used with depth of the peaks to express sedimentation rate in units of g DW m^{-2} yr^{-1}; these data are presented in Figure 9. Net sedimentation rates were low in shallow portions of the lake. The rate was highest at 3.75 m, and diminished from this maximum going into deeper water. Particle-size sorting of sediments along line AB has been demonstrated in this study (Figure 7). It is postulated that sediments originate largely from fragmenting vegetation of the wetland at the edge of the lake. Wind-induced turbulent movement of water in the shallows causes suspension and down-slope movement, particularly of small particles. The region of highest sedimentation rate corresponds to the point along the slope of the basin where the mean turbulent energy of water is diminished rapidly with sudden increase in depth (Wetzel, 1975). Fewer numbers of particles of marsh origin are available for sedimentation beyond the region of highest fallout, thus accounting for progressively lower sedimentation rates in portions of the south basin deeper than 3.75 m.

In each sediment profile presented in this study, there was a decline in arsenic from the peak concentration to the 1980 sediment surface. It is proposed that the rates of these declines are a function primarily of sedimentation rates, concentration of arsenic in sedimenting materials, concentration of arsenic in base sediments, and the degree to which sedimenting materials are mixed with base sediments. The relationship between these factors is expressed in Figure 10. An underlying assumption of this figure is that diffusion processes are not important in establishing observed arsenic

Figure 9. Sedimentation rates at the points of sampling along the slope of the Lake Lansing basin.

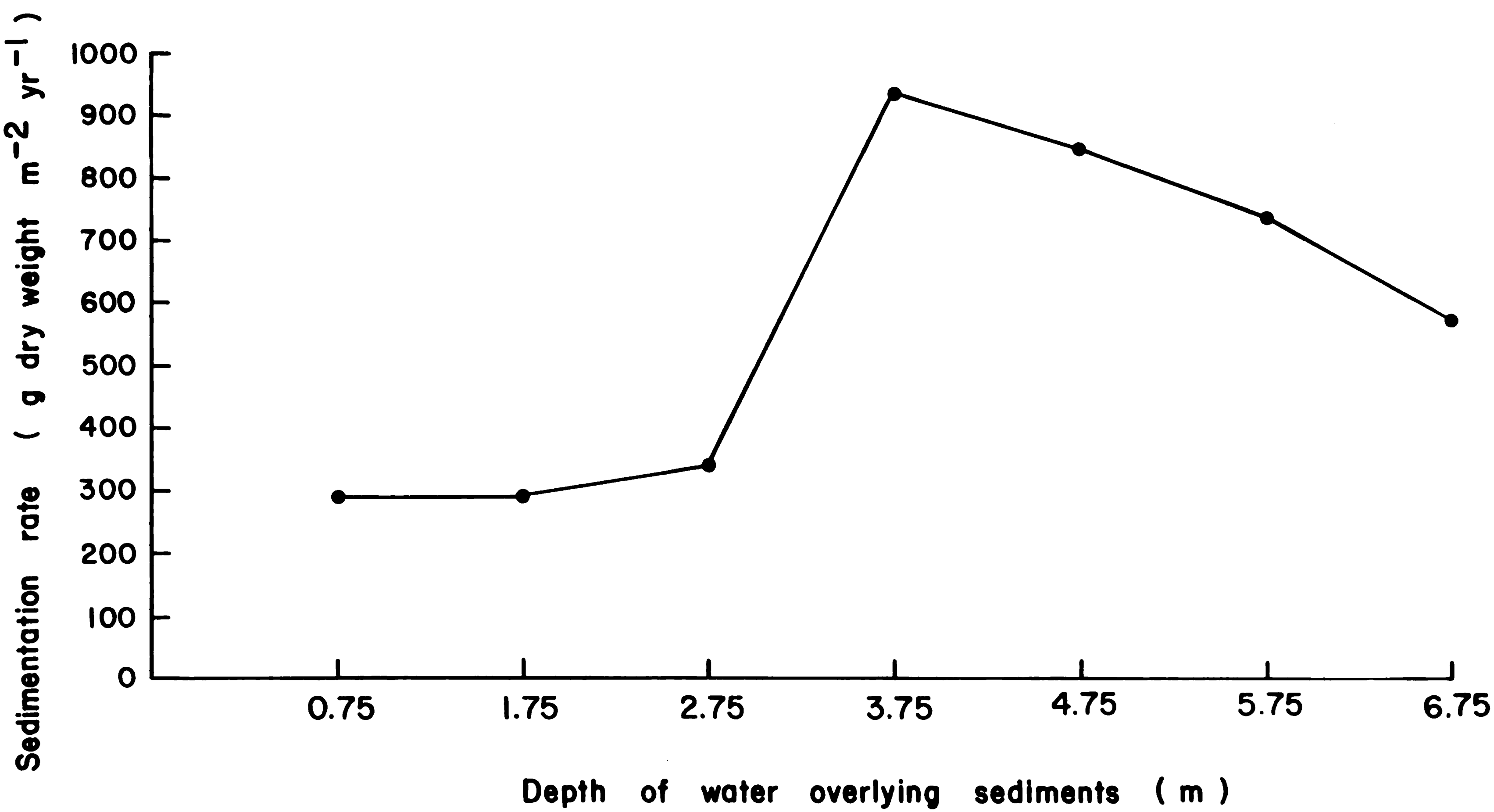
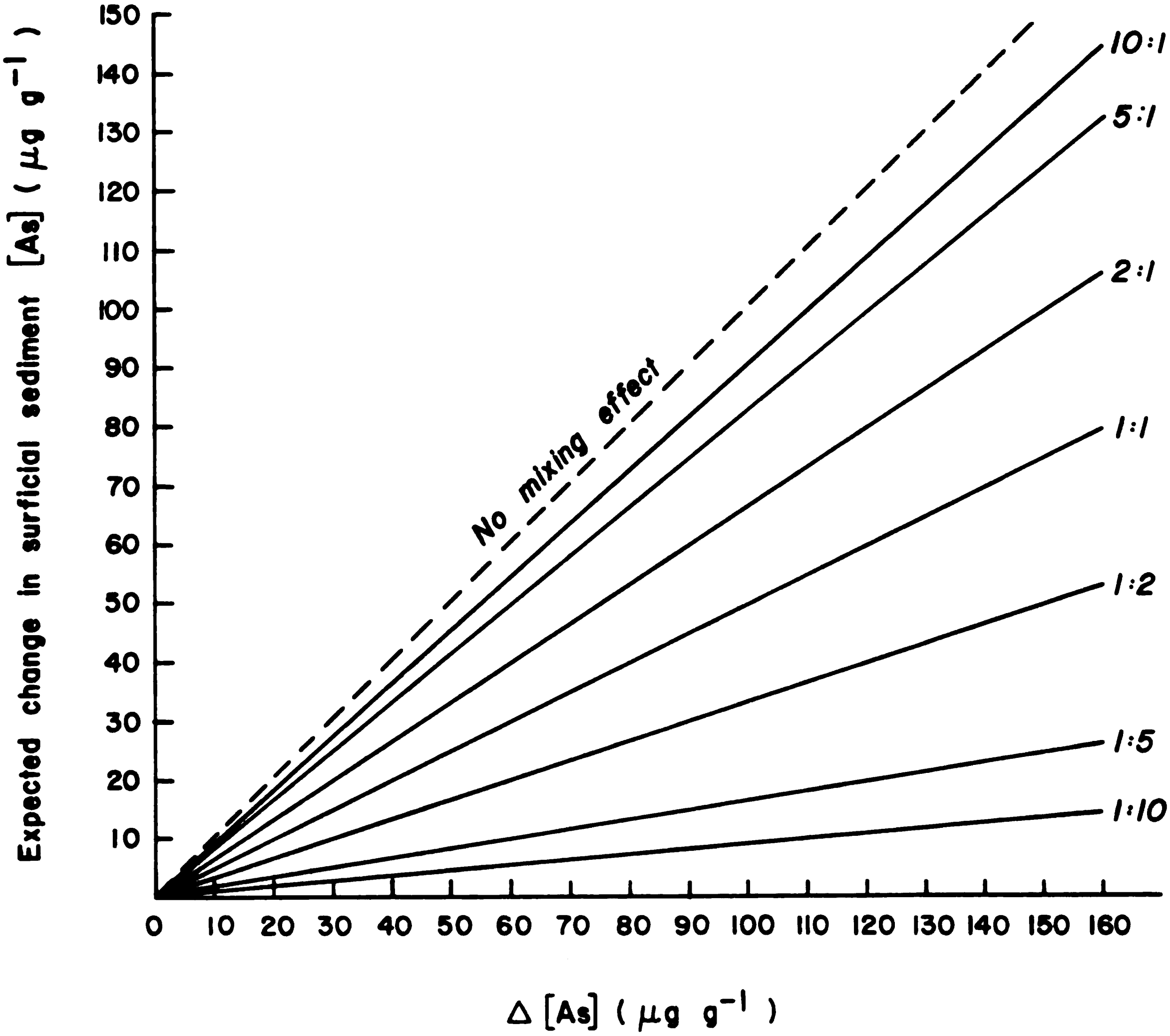

Sedimentation rate (g dry weight m^-2 yr^-1)
1000
900
800
700
600
500
400
300
200
100
0
0.75
1.75
2.75
3.75
4.75
5.75
6.75
Depth of water overlying sediments (m)

Figure 10. Expected change in arsenic concentration of surficial sediments as a function of the difference in arsenic concentration between sedimenting and base materials. Curves reflect extent of influence of mixing newly sedimented material with base sediments. Dashed line represents maximum influence of newly sedimented material on surficial sediment. Ratios are for new sediment: base sediment mixing.


Expected change in surficial sediment [As] (μg g⁻¹)
Δ [As] (μg g⁻¹)
No mixing effect
10:1
5:1
2:1
1:1
1:2
1:5
1:10
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160

profiles.

The horizontal axis of Figure 10 represents the difference in arsenic concentration between sedimenting and base materials. This scale can be used independently of the arsenic concentrations of these materials. For example, if base sediment has 100 μg As g^{-1} and sedimenting material 50, or base 1100 and sedimenting material 1050, or base 50 and sedimenting material 0, then all of these conditions are represented by the same point on the abscissa (Δ [As] 50 μg g^{-1}). To facilitate use of the figure, it is best to be consistent by subtracting sedimenting arsenic concentration from that of base material. Note that when sedimenting arsenic concentration is lower than that in base material, the expected change would be a negative value.

The vertical axis marks the expected change in surficial arsenic concentration following sedimentation and mixing of the materials under consideration. This value is negative when sedimenting arsenic concentration is lower than base concentration, and positive when it is higher. This scale can also be used independent of the arsenic concentrations in the sediment materials.

The curves of the figure represent the degree of mixing at the sediment surface under consideration. Mixing is viewed as a ratio; for example, when the degree of mixing of new sediments with base sediments is low, the ratio is high. In general, this situation is likely in lakes where turbulent flow is diminishing and suspended materials fall out on a sediment surface that is not exposed to appreciable turbulence. As shown in Figure 10, the surface arsenic concentration is expected in this case to be heavily influenced by the concentration of arsenic in sedimenting materials. The family of curves given in Figure 10 illustrates a range of cases.

The shapes of recession curves in cores of this study from peak arsenic concentrations to the arsenic concentrations of 1980 sediment surfaces suggest that the relationships of Figure 10 were operative along line AB since 1957. These declines can be described as exponential decreases in arsenic concentration with distance from the depth of the peak. Using the core data, exponential coefficients were calculated. Employing the described function, which takes the general form $y = ae^{bx}$ where $y = [As]$ and x = depth, the estimated times for sediment surfaces to reach background were calculated. The time element was obtained by using sedimentation rates estimated from the depths of peak concentration due to 1957 treatment. These data are presented in Table 1.

The results in the last column of the table show that shallow sediments are expected to take a long period of time (> 100 yrs.) to reach pre-treatment background. The surface of these sediments, covered primarily by materials of fringing wetland and submersed macrophyte origin, could experience relatively high mixing due to the action of waves and wind-generated currents. Mixing arsenic-bearing surface sediments with newly sedimented materials of lower concentration slows

Table 1. Calculated time for arsenic to reach background concentrations in surface sediments along the slope of the south basin of Lake Lansing.

Depth of Water 1980 m	Depth of Water 1957 m	Surface Arsenic $\mu g\ g^{-1}$	Background[1] Arsenic $\mu g\ g^{-1}$	Sediment Accumulation Rate $cm\ yr^{-1}$	Regression Coefficients for Exponential Curves a, b, r^2	Time From Peak to Background yrs
0.75	0.87	34	9	0.543	32.68, 0.02, 0.999	>100*
1.75	1.87	62	14	0.543	59.27, 0.02, 0.988	>100*
2.75	2.87	87	11	0.543	80.69, 0.02, 0.920	>100*
3.75	4.07	84	12	1.413	72.11, 0.03, 0.97	43
4.75	5.02	91	10	1.195	77.24, 0.04, 0.95	40
5.75	5.97	92	15	0.978	85.99, 0.05, 0.99	37
6.75	6.92	91	16	0.760	73.56, 0.07, 0.98	28

[1] Means of concentrations in deep portions of cores.

*Only three data points were available to obtain coefficients for these exponential curves; the accuracy of this best estimate is reduced by that.

the process of burying 1957 arsenic. The data of Table 1 further suggest that sediments in the deep portion of the lake would return to background much faster than shallow sediments. As an explanation for this, it is proposed that sediments from shallow water with relatively low arsenic concentrations have been a dominant source of new material for deep sediments since the time of treatment. If these were mixed poorly in deep water with the heavily contaminated base sediments there, the arsenic concentration of the deep sediment surfaces would recede rapidly toward the level of the incoming materials. Poor mixing of surface sediments by turbulence is expected in deep portions of the lake. It must be noted that by this model, the rate of approach of deep sediments to background arsenic concentration will be limited by the rate of approach of shallow sediments to background. Because of this, it is proposed that the years to reach background calculated for deep sediments and given in Table 1 are unrealistic. If shallow sediments are predicted to reach background in > 100 years, then on the assumptions of this dissertation, a similar length of time can be predicted for deep sediments as well.

The data of this study, and the assumptions used to examine them, provide the framework for an experimental approach to answer a question of considerable ecological importance. The processes involved in burying contaminants in lake sediments, and the time required to accomplish this are not generally known.

APPENDIX

During February of 1981, cores were taken from transect AB from the same depth as those used for total arsenic analysis. The top 5 cm section was removed from each frozen core and cut longitudinally. Density of the surficial sediments as well as bulk density of constituent particles was measured by a water displacement method. The results are given in Appendix Table 1.

A known volume of distilled water was added to a graduated cylinder; weight of cylinder and water was measured and volume of water was recorded. Longitudinal sections were placed in the cylinder. When the frozen piece thawed, weight and volume of the cylinder and contents were measured and water temperature recorded. The weight of core section, consisting of particulate and dissolved solids plus core water, was obtained from the increase in weight. The volume of core section was calculated by subtracting the initial volume in the cylinder from the final volume. Contents of the cylinder were rinsed into a pre-dried and weighed aluminum tray and dried in an oven at 95°C to a constant weight. The weight of dry material was divided by the calculated volume of core section to obtain the density of particulate and dissolved solids in the core section. The weight of the core water was calculated by subtracting the weight of dry solids from the weight of solids plus core water. The volume of core water was calculated from the weight of the water corrected for density at the temperature at which the weight was measured. To obtain the volume of solids, the

the volume of core water was subtracted from the volume of solids plus core water. The bulk density of solids was calculated by dividing solids weight by solids volume.

Core total solids density and bulk total solids density were corrected for dissolved solids. To measure the dissolved solids component of core water, a frozen core fragment was thawed on a glass fiber filter and the water was drawn through its 0.5 pores. A measured volume of filtrate was placed in a pre-washed aluminum tray and dried in an oven at 95°C to a constant weight. The resultant dry weight of dissolved solids was divided by the filtrate volume to obtain the concentration of dissolved solids in the core water. The mass of dissolved solid was subtracted from estimates of the core total solids density and from the bulk total solids density.

The percent-ash was determined in dissolved solids fractions of cores. After weighing, the contents of aluminum trays containing dried core filtrate were combusted at 550°C for one hour (APHA, 1976). The weight of the residue was used to calculate ash-weight; weight loss on ignition was taken to represent organic material.

Table A-1. Organic and inorganic content of particulate and dissolved components, and density of <u>surficial</u> sediments along line AB of this study.

		PARTICULATE FRACTION						DISSOLVED FRACTION			
Water Depth (m)	Total Dry Weight ($g\ cm^{-3}$)	Dry Weight ($g\ cm^{-3}$)	Organic Matter[1] (%)	Organic Matter ($g\ cm^{-3}$)	Inorganic Matter ($g\ cm^{-3}$)	Density[2] ($g\ cm^{-3}$)	Volume[3] (%)	Dry Weight ($mg\ cm^{-3}$)	Organic Matter (%)	Organic Matter ($mg\ cm^{-3}$)	Inorganic Matter ($mg\ cm^{-3}$)
0.15	0.0515	0.0510	63.9	0.0326	0.0184	1.1743	4.3	0.45	35.1	0.16	0.29
0.75	0.535	0.0530	32.0	0.0170	0.0360	1.4274	3.7	0.52	41.4	0.22	0.31
1.75	0.0593	0.0588	32.8	0.0193	0.0395	1.5692	3.8	0.53	----	----	----
2.75	0.0611	0.0606	34.6	0.0210	0.0396	1.5850	3.8	0.55	54.9	0.30	0.25
3.75	0.0661	0.0653	32.5	0.0219	0.0441	2.0181	3.2	0.80	40.0	0.32	0.49
4.75	0.0707	0.0699	34.0	0.0238	0.0461	2.4344	2.9	0.81	42.6	0.35	0.47
5.75	0.0738	0.0664	29.8	0.0189	0.0444	2.7025	2.5	7.59	24.5	1.86	5.73
6.75	0.0752	0.0670	29.7	0.0199	0.0471	2.7576	2.4	8.41	26.4	2.22	6.19

[1] Values taken from Appendix Table 2.

[2] Particle dry weight per cm^3 of original particle volume.

[3] Percent sediment volume (water volume and particle volume) occupied by particulate material.

Table A-2. Mean and one standard error of three replicates of percent organic matter in sediment cores from Lake Lansing along line AB.[1]

Depth from sediment surface (cm)	Depth of overlying water (m) 0.00	0.75	1.75	2.75	3.75	4.75	5.75	6.75
0 - 5	63.9±0.33	32.0±0.32	32.8±0.86	34.6±0.24	32.5±0.04	34.0±0.19	29.8±0.54	29.7±0.35
5 - 10	63.9±0.40	31.9±0.15	31.4±0.57	35.5±0.11	33.5±0.11	34.9±0.07	33.4±0.19	33.5±0.30
10 - 15	57.4±0.20	32.5±0.43	33.3±0.21	34.1±0.58	34.1±0.34	34.9±0.02	33.0±0.20	32.6±0.34
15 - 20	42.4±0.35	34.5±0.55	32.2±0.89	37.0±0.38	34.5±0.23	34.4±0.12	33.0±0.22	32.3±0.27
20 - 25	35.9±0.27	33.0±0.21	30.0±0.32	38.8±0.49	35.4±0.23	34.1±0.22	35.0±0.17	33.0±0.24
25 - 30	34.5±0.18	35.0±0.69	33.2±0.69	39.0±0.13	35.7±0.09	34.4±0.04	35.7±0.59	33.1±0.30
30 - 35	34.6±0.45	36.8±0.28	35.7±0.49	39.8±0.18	35.6±0.33	34.8±0.16	36.7±0.16	33.4±0.17
35 - 40	35.9±0.12	38.3±0.29	36.0±0.44	40.1±0.19	37.2±0.17	32.5±0.39	39.6±0.20	32.7±0.01
40 - 45	38.4±1.07	38.5±0.08	36.6±0.45	40.2±0.30	37.1±0.20	32.8±0.25	43.3±0.14	31.9±0.04
45 - 50	40.9±0.79	38.1±0.14	41.2±0.44	45.7±0.45	37.5±0.11	38.6±0.57	---------	36.3±0.42

[1]Three sub-samples were taken from each section of core. Dry weight was obtained at 105°C after 24 hours. Samples were combusted by exposure to 550°C for 1 hour; loss of weight by combustion was taken as an estimate of organic weight.

Table A-3. Concentrations of total As (μg As g^{-1} dry weight) with depth in sediment cores taken along line AB of the south basin of Lake Lansing.

Depth below sediment surface (cm)	Depth of water overlying sediment (m) 0.75	1.75	2.75	3.75	4.75	5.75	6.75
0 - 5	34	62	87	84	91	92	91
5 - 10	37	71	92	91	108	131	123
10 - 15	40	77	109	99	119	160	165
15 - 20	37	73	91	116	147	199	268
20 - 25	18	53	59	138	232	249	232
25 - 30	15	36	42	178	244	107	104
30 - 35	13	31	32	201	111	72	70
35 - 40	12	35	28	181	74	58	48
40 - 45	11	20	24	90	45	54	46
45 - 50	9	19	28	47	32	--	60
50 - 55	9	19	21	35	31	49	57
55 - 60	10	15	15	30	24	45	59
60 - 65	10	16	12	23	26	35	57
65 - 70	8	17	8	22	17	18	46
70 - 75	8	18	11	22	9	15	40
75 - 80	8	17	10	19	11	14	33
80 - 85	9	17	11	16	12	12	28
85 - 90	9	15	10	12	10	14	20
90 - 95	10	12	9	10	9	17	18
95 - 100	9	10	9	10	9	17	18
100 - 105	10	8	10	10	8	13	14
105 - 110	11	7	14	11	11	13	12
110 - 115	11	9	13	13	8	15	14
115 - 120	13	8	11	12	10	14	14
120 - 125	14	8	9	--	10	16	15
125 - 130	12	10	12	11	10	17	16
130 - 135	10	10	11	9	11	19	--
135 - 140	11	8	12	11	15	13	--
140 - 145	19	9	10	--	14	15	--
145 - 150	20	8	12	12	14	10	--

Table A-4. Concentrations of total As (µg As g^{-1} dry weight) with depth in sediment cores taken along transect 1 of the south basin of Lake Lansing.

Depth below sediment surface (cm)	Depth of water overlying sediment (m)							
	0.5	1.5	2.5	3.5	4.5	5.5	6.5	7.5
0 - 5	51	54	80	142	105	120	117	114
5 - 10	53	62	86	160	135	138	162	198
10 - 15	72	70	92	148	153	166	227	210
15 - 20	73	81	94	133	160	170	209	299
20 - 25	53	34	96	109	170	175	271	169
25 - 30	20	18	162	73	---	215	93	101
30 - 35	15	20	145	52	171	123	87	84
35 - 40	9	20	123	42	78	89	74	68
40 - 45	7	19	87	42	26	73	73	55
45 - 50	7	19	54	20	15	53	65	52
50 - 55	11	24	37	23	11	29	51	50
55 - 60	9	15	36	13	13	18	56	51
60 - 65	10	17	26	12	11	19	45	--
65 - 70	9	16	15	17	9	15	49	59
70 - 75	10	12	13	17	9	15	48	58
75 - 80	11	16	8	11	14	26	57	58
80 - 85	9	17	9	10	15	28	57	25
85 - 90	11	16	9	13	13	10	19	24
90 - 95	10	15	8	--	11	11	24	25
95 - 100	10	15	10	13	12	13	22	23
100 - 105	11	12	10	13	11	12	20	17
105 - 110	11	9	12	14	10	11	19	14
110 - 115	11	9	12	15	13	16	21	16
115 - 120	9	6	11	12	12	16	20	17
120 - 125	10	8	9	--	14	16	24	18
125 - 130	10	7	10	--	14	15	21	20
130 - 135	11	10	11	--	13	16	18	19
135 - 140	10	8	7	--	13	16	22	22
140 - 145	9	9	9	--	9	14	25	16
145 - 150	--	10	10	--	13	17	19	22

Table A-5. Concentrations of arsenic in macrophytes collected along line AB; July 1, 1980.[1]

Location	Water Depth (m)	Species	Plant Parts	μg As g^{-1} DW ± 1 SE
10-15 m within wetland	0	*Lythrum salicaria* L.	stems and leaves	0
			roots	19.24 ± 2.39
lake edge	0	*Lythrum salicaria* L.	stems and leaves	0
			roots	44.91 ± 5.58
		Typha angustifolia L.	stems and leaves	0
			rhizomes and roots	28.72 ± 12.58
0-20 m from shore	0-0.5	*Nuphar advena* Ait.	petioles and leaves	0
			rhizomes and roots	17.49 ± 14.02
20-40 m from shore	0.5-1.0	*Elodea canadensis* (Michx.) Planchon	whole plants	26.60 ± 6.84
70-105 m from shore	1.5-2.0	*Potamogeton spp.*	whole plants	28.18 ± 2.74
105-110 m from shore	2.0-2.5	*Ceratophyllum demersum* L.	whole plants	33.79 ± 2.06

[1]Three sub-samples of each plant collection were dried, ground, and analyzed for total-As with procedures similar to those for sediment samples (neutron activation).

Table A-6. Concentrations of arsenic in macrophytes collected along transect 1; July 1, 1980.[1]

Location	Water Depth (m)	Species	Plant Parts	μg As g^{-1} DW ± 1 SE
10-15 m within wetland	0	*Lythrum salicaria* L.	stems and leaves	0
			roots	17.22 ± 9.60
		Typha angustifolia L.	stems and leaves	0
			rhizomes and roots	15.20 ± 4.50
lake edge	0	*Lythrum salicaria* L.	stems and leaves	0
			roots	60.56 ± 13.79
0-20 m from shore	0-0.5	*Nuphar advena* Ait.	petioles and leaves	0
			rhizomes and roots	15.27 ± 7.80
		Najas flexilis (Willd.) Rostk and Schmidt	whole plants	46.95 ± 4.04
20-100 m from shore	0.5-1.0	*Chara globularis* (Thuill)	whole plants	22.41 ± 4.07
100-190 m from shore	1.0-2.0	*Elodea canadensis* (Michx.) Planchon	whole plants	28.80 ± 5.77
190-200 m from shore	2.0-2.5	*Ceratophyllum demersum* L.	whole plants	34.05 ± 4.58

[1]Three sub-samples of each plant collection were dried, ground, and analyzed for total-As with procedures similar to those for sediment samples (neutron activation).

Table A-7. The slope of the south basin of Lake Lansing.

TRANSECT 1		LINE AB	
Distance from shore	Depth cm	Distance from shore	Depth cm
0	0	0	0
80	84	35	84
100	93	55	123
130	109	65	140
160	138	75	158
190	192	85	191
200	260	95	191
210	344	105	271
220	391	110	320
230	421	115	392
240	455	120	429
250	497	125	477
260	549	130	504
270	606	135	540
280	676	145	604
290	711	155	665
300	730	165	702
310	750	185	730
		210	723

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