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## Arsenic concentrations in water and fish from Chautauqua Lake, New York

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### Synopsis

Arsenic persists in Chautauqua Lake, New York waters 13 years after cessation of herbicide (sodium arsenite) application and continues to cycle within the lake. Arsenic concentrations in lake water ranged from 22.4–114.8  $\mu\text{g l}^{-1}$ ,  $\bar{x} = 49.0 \mu\text{g l}^{-1}$ . Well water samples generally contained less than 10  $\mu\text{g l}^{-1}$  arsenic. Arsenic concentrations in lake water exceeded U.S. Public Health Service recommended maximum concentrations (10  $\mu\text{g l}^{-1}$ ) and many samples exceeded the maximum permissible limit (50  $\mu\text{g l}^{-1}$ ). Fish accumulated arsenic from water but did not magnify it. Fish to water arsenic ratios ranged from 0.4–41.6. Black crappie (*Pomoxis nigromaculatus*) contained the highest arsenic concentrations (0.14–2.04  $\mu\text{g g}^{-1}$ ),  $\bar{x} = 0.7 \mu\text{g g}^{-1}$  while perch (*Perca flavescens*), muskellunge (*Esox masquinongy*) and largemouth bass (*Micropterus salmoides*) contained the lowest concentrations (0.02–0.13  $\mu\text{g g}^{-1}$ ). Arsenic concentrations in fish do not appear to pose a health hazard for human consumers.

### Introduction

Arsenic and its compounds have been widely used in pigments, as insecticides, herbicides, as alloys in metals and as chemotherapeutic agents. Wide use and misuse of this potentially toxic element have resulted in large arsenic accumulations in soils and sediments. Bishop and Chisholm (1962) have reported values of 9.8 to 234.4  $\text{mg kg}^{-1}$  for soils from orchards in Nova Scotia and Walsh & Keeney (1975) cited examples of soil concentrations of up to 2553 ppm. Aston et al. (1975) found arsenic concentrations of over 5000 ppm in the sediments of streams in a mining area of southwest England.

Arsenic has been used as an aquatic herbicide for

decades with only minimal study as to its fate in aquatic ecosystems. An arsenic cycle may exist in lakes that have accumulated an arsenic burden from natural or human sources. Arsenic in aquatic systems can undergo oxidation-reduction, ligand exchange, precipitation, and adsorption reactions and is stable in four oxidation states (+5, +3, 0, -3). Organisms affect its distribution by accumulating, transporting and transforming it. Methylation or reduction to arsine solubilizes arsenic and diffusion or mixing by currents or burrowing organisms facilitates re-entry into the water column (Ferguson & Gavis 1972). Methyl transfer reactions require anaerobic conditions to produce dimethylarsine (Berman, 1975) with subsequent oxidation to the water-soluble form, cacodylic acid (Ridley et al. 1977). The presence of an arsenic cycle may insure the availability of methyl arsenic compounds for incorporation by higher organisms such as fish.

In model ecosystems (microcosms), biologically active forms of arsenic were concentrated from water by fish and other freshwater organisms but were not magnified in food chains (Isensee et al. 1973, Schuth & Isensee 1974, Woolson 1975). Giddings & Eddlemon (1977) reported that sediment type was an important factor affecting retention time of arsenic in the overlying water column. In microcosms with sandy bottoms, arsenic remained at much higher levels in the water than in microcosms with mud bottoms. However, these studies were conducted over short periods of time (< 3 months) and little is known about arsenic mobilization and transport in and bioaccumulation from freshwater ecosystems subjected to a chronic burden of this toxic material.

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Chautauqua Lake in western New York received arsenic from direct applications of sodium arsenite as an aquatic herbicide from 1955 to 1963 coupled with continuing runoff from farming activity. In 1971, arsenic concentrations in Chautauqua Lake water ranged from 3.5 to 43.3 ppb (Lis & Hopke 1973). Ruppert et al. (1974) found that arsenic concentrations in sediments from Chautauqua Lake ranged from 0.5 ppm to 306.0 ppm,  $\bar{x}$  = 22.1 ppm. The purpose of this study was to measure present arsenic concentrations in Chautauqua Lake waters and fish as a first step in determining the long term effects of arsenic contamination on the biota of this freshwater ecosystem.

## Methods

Water samples were collected from Chautauqua Lake, New York in June, 1976 at 30 locations. Well water samples were collected in July, 1976 from 24 locations adjacent to the lake (Figure 1). Lake water samples were collected 1 m above the bottom, using a nonmetallic water sampler, immediately acidified to 1% with double distilled  $\text{HNO}_3$  acid and filtered through a prewashed 0.45  $\mu\text{m}$  cellulose acetate filter before being frozen in polyethylene bottles. Well water samples were immediately acidified to 1% with double distilled  $\text{HNO}_3$  acid and stored frozen in polyethylene bottles. All bottles and glassware were soaked in a 2% solution of Contrad 70 (American Hospital Supply Company, McGraw Park, Illinois) for twenty-four hours and rinsed three times in de-ionized, glass distilled water before use.

Arsenic concentrations in the water samples were determined using a Perkin-Elmer model 306 atomic absorption spectrophotometer equipped with an HGA-2100 flameless atomizer and deuterium background corrector. Sensitivity was 0.5  $\mu\text{g l}^{-1}$  As. Reagent blanks which were used throughout sample collection, preparation and analysis to evaluate possible contamination contained  $< 0.5 \mu\text{g l}^{-1}$  As.

Fifteen species of fish ( $n = 129$ ) were captured in pound nets set in near-shore water between Prendergast Point and Stow in Chautauqua Lake in May, 1976 and placed immediately into ice chests. Fish were sorted by species and frozen in polyethylene bags. Samples were prepared for analysis by grinding whole bodies after removal of the head. Material was blended in a stainless steel electric blender to obtain a homogeneous mixture. For fish that were larger than 300 g, samples were taken by grinding a cross section

of the fish taken from between the operculum and the dorsal fin. Samples of *Isox masquinongy* were prepared at the New York Department of Conservation Laboratory, Rome, New York by grinding muscle and liver samples separately from each fish. The fish tissue homogenates were individually refrozen in sealed polyethylene bags and analyzed at the Nuclear Science and Technology Facility, State University of New York at Buffalo, New York. Fish tissue underwent a highly specific neutron activation analysis to determine concentration of arsenic per unit wet weight of fish tissue according to the method of Pilly et al. (1974).

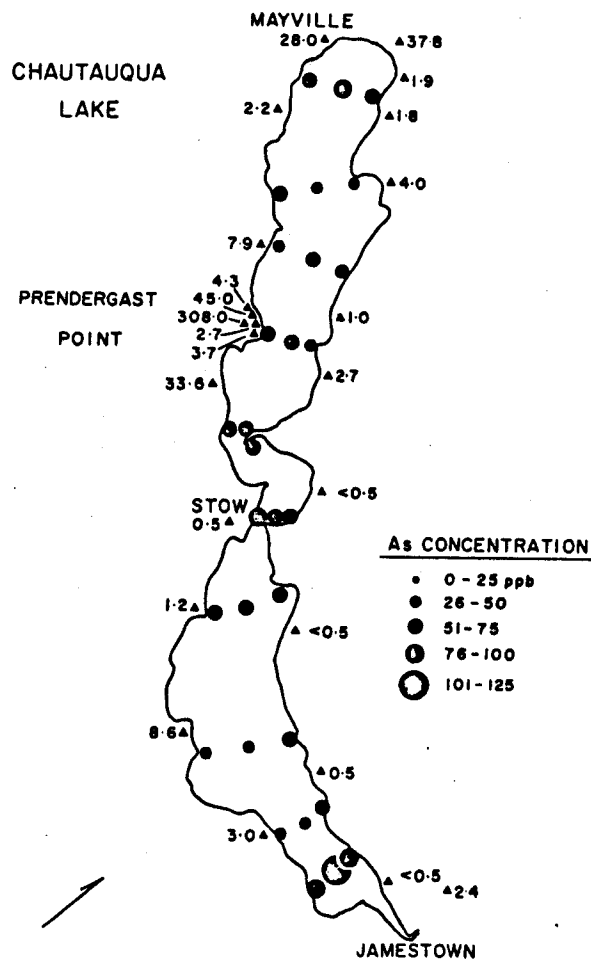


Fig. 1. Map of Chautauqua Lake.  $\blacktriangle$  mark locations of wells sampled around the lake. The value associated with each well is arsenic concentration ( $\mu\text{g l}^{-1}$ ) of the well water.  $\bullet$  indicates location of lake water collections. Size of the closed circle gives relative concentration of dissolved arsenic ( $\mu\text{g l}^{-1}$ ). The only outflow from the lake is at the Jamestown end.

## Results

Concentrations of arsenic in aquifers surrounding Chautauqua Lake were generally low. Arsenic concentrations in well water samples ranged from  $< 0.5 \mu\text{g l}^{-1}$  As to  $308 \mu\text{g l}^{-1}$  (Figure 1). Most values were below  $10 \mu\text{g l}^{-1}$  As. However, four values ranged from  $28.0$  to  $45.0 \mu\text{g l}^{-1}$  while one value was  $308 \mu\text{g l}^{-1}$ . This very high value came from a sample taken on 10 June 1976 from the main well at the state fish hatchery on Prendergast Point. A sample taken in late July 1976 gave a value of  $7.9 \mu\text{g l}^{-1}$ . Since both of these samples were retested and based on standard curves, and since reagent blanks had values  $< 0.5 \mu\text{g l}^{-1}$  As, we concluded that the values for these samples were accurate measures of the As content of that well on those given dates. This phenomenon will be addressed in the Discussion.

Dissolved arsenic levels in lake water were considerably higher than those in well water (Figure 1 and Table 1). Lake water As concentrations ranged from  $22.4$ – $114.8 \mu\text{g l}^{-1}$ ,  $\bar{x} = 49.0 \mu\text{g l}^{-1}$  with no overall distributional trend. Most values ranged from  $25$ – $60 \mu\text{g l}^{-1}$ , however, there were noticeably higher concentrations in some samples from the shallow southern end of the lake.

Fish from Chautauqua Lake contained from  $0.02$  to  $2.04 \mu\text{g g}^{-1}$  As (wet weight). Values varied among species and among individuals of a given species (Table 2). Perch (*Perca flavescens*), muskellunge (*Esox masquinongy*), largemouth bass (*Micropterus salmoides*), carp (*Cyprinus carpio*), redhorse (*Moxo-*

Table 1. Dissolved arsenic concentrations in water from Chautauqua Lake

Station	(As) $\text{g l}^{-1}$	Station	(As) $\text{g l}^{-1}$
1	50.4	16	72.8
2	61.6	17	53.2
3	57.4	18	53.2
4	42.0	19	42.0
5	33.6	20	56.0
6	33.6	21	42.0
7	25.2	22	39.2
8	44.0	23	36.4
9	33.6	24	44.8
10	50.4	25	39.2
11	56.0	26	36.4
12	22.4	27	50.4
13	42.0	28	72.8
14	47.6	29	114.8
15	56.0	30	61.6

Table 2. Accumulation of arsenic by fifteen species of fish sampled in May 1976 from Chautauqua Lake

Species	n	$\mu\text{g g}^{-1}$ As in tissue <sup>a</sup>	Bioaccumulation ratios <sup>b</sup>	Trophic level <sup>c</sup>
<i>Morone chrysops</i>	1	0.13	2.7	Piscivore
<i>Stizostedion vitreum</i>	4	0.11-0.19	2.2-3.9	Piscivore
<i>Esox masquinongy</i>				
muscle	6	0.04-0.08	0.8-1.6	Piscivore
liver	8	0.02-0.13	0.4-2.7	
<i>Micropterus salmoides</i>	14	0.04-0.10	0.8-2.0	Carnivore
<i>Pomoxis nigromaculatus</i>	12	0.14-2.04	2.9-41.6	Carnivore
<i>Pomoxis annularis</i>	13	0.12-0.17	2.5-3.5	Carnivore
<i>Ambloplites rupestris</i>	11	0.07-0.2	1.4-4.1	Carnivore
<i>Perca flavescens</i>	7	0.02-0.06	0.4-1.2	Carnivore
<i>Lepomis gibbosus</i>	14	0.10-0.55	2.0-11.2	Carnivore
<i>Moxostoma</i> sp.	1	0.11	2.2	Invertivore
<i>Catostomus catostomus</i>	2	0.06-0.21	1.2-4.3	Invertivore
<i>Cyprinus carpio</i>	2	0.14-0.15	2.9-3.1	Omnivore
<i>Ictalurus nebulosus</i>	16	0.02-0.18	0.4-3.7	Omnivore
<i>Notemigonus crysoleucus</i>	14	0.06-0.19	1.2-3.9	Omnivore
<i>Lepomis macrochirus</i>	8	0.06-0.24	1.2-4.9	Omnivore

a wet weight concentrations

b The ratio of concentration in tissue/mean concentration in water (Table 1)

c according to Scott & Crossman (1973)

*stoma* sp.), and white bass (*Morone chrysops*) had the lowest As concentrations ( $0.02\text{--}0.15\ \mu\text{g g}^{-1}$ ) while black crappie (*Pomoxis nigromaculatus*) had the highest concentrations ( $0.14\text{--}2.04\ \mu\text{g g}^{-1}$ ,  $\bar{x} = 0.70\ \mu\text{g g}^{-1}$ ) followed by pumpkinseed (*Lepomis gibbosus*) ( $0.10\text{--}0.55\ \mu\text{g g}^{-1}$ ,  $\bar{x} = 0.26\ \mu\text{g g}^{-1}$ ).

No magnification of arsenic was evident between fish trophic levels studied. Also, no trend in As concentration was evident after fish were grouped according to trophic level following Scott & Crossman (1973). Low concentrations of arsenic per unit of tissue in *Esox masquinongy* and *Micropterus salmoides* were similar to the concentrations in the most abundant forage fish of the lake *Perca flavescens* and *Notemigonus crysoleucas* (golden shiner). Predators contained lower As concentrations than many of the species available for predation in the lake including *Pomoxis* sp. and *Lepomis gibbosus*.

There was concentration of arsenic above mean environmental levels in most of the fish. Bioaccumulation ratios (BR) were computed to determine the number of times arsenic was concentrated above the mean level found in lake water (Table 2). *Pomoxis nigromaculatus* and *Lepomis gibbosus* had the highest BR, 41.6 and 11.2 respectively, while *Perca flavescens* and *Ictalurus nebulosus* had the lowest BR, 0.4. The BR for the remaining species ranged from 0.8 to 4.3 and produced no discernible trend with respect to trophic level.

Histograms of As concentration frequency distributions in tissues were plotted for six species (Figure 2). Arsenic distributions for *Notemigonus crysoleucas* and *Lepomis gibbosus* were positively and negatively

skewed respectively. Distributions for the remaining species were all positively skewed and appeared to follow log normal distributions.

## Discussion

### Well water

Although wells which were sampled in this study were of varying depth including some deep, high-flow municipal water sources, there appeared to be no possibility of contamination of surface or lake waters from underground sources of arsenic. The aquifer surrounding the lake appears to contain very little arsenic. We believe that the high As concentrations found for five of the wells are due to site specific conditions that are not representative of the aquifer in general. These wells are old and As is possibly being leached from iron well casings. The value ( $308\ \mu\text{g l}^{-1}$ ) for the state fish hatchery well is very anomalous. This well is used only during the muskellunge rearing season (May to August). The As pulse during the early portion of seasonal use may be due to As leached from the well casing or from a localized As containing shale deposit. By late July the As pulse may have been washed from the well, giving the As concentration of  $7.9\ \mu\text{g l}^{-1}$ . It is also possible that there is a seasonal cycle of As in the water from this well. However, this is unlikely and additional data would be needed to document such a cycle.

### Lake water

The persistence of elevated levels of arsenic in Chautauqua Lake 13 years after the cessation of herbicide application indicates that an arsenic cycle is present in this lake. Dissolved As concentrations in lake water in 1976 (Table 1) were almost three times greater than in 1971 ( $3.5\text{--}43.3\ \text{ppb}$ ,  $\bar{x} = 15.1\ \text{ppb}$ , Lis and Hopke 1973). In 1955 As concentrations were below the level of detectability ( $10\ \text{mg l}^{-1}$ ) (Ullmann et al. 1961). While part of the increase in As concentrations since 1971 may have been due to differences in sensitivity of analyses, there appears to have been a chronic increase in dissolved As levels in Chautauqua Lake over the five year period. This may have been due to the release of arsenic from sediments during anoxic conditions (Lis & Hopke 1973). Our results support the conclusion of Ruppert et al. (1974) that the source of As in the lake water is the sediments

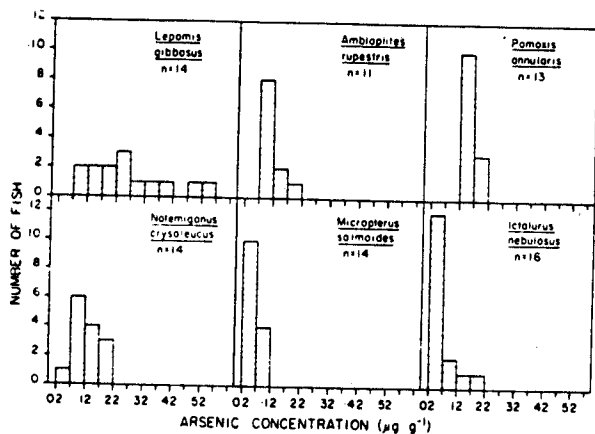


Fig. 2. Frequency distributions of the arsenic concentrations in fish from Chautauqua Lake.

which accumulated this metalloid as a result of herbicide application.

Arsenic solubility in water depends upon many factors including oxygen concentration in the water, presence of sulfur and ferric hydroxide and activity of methanogenic bacteria. Arsenic release from the sediments may occur in a reducing environment especially during anoxic conditions. Reduction of arsenate and arsenite (Ferguson & Gavis 1972) and microbial methylation (Ridley et al. 1977) can solubilize arsenic which reenters lake water by diffusion and current mixing. Low oxygen concentrations occurred in some of the deep kettleholes in Chautauqua Lake during the summer of 1975 and 1976 (Mayer et al. 1975, Storch, personal communication). These make up only a very small percent of the total lake volume. Therefore, the majority of the lake remained aerobic throughout the year and arsenic is being cycled in the water column by aerobic processes. Under anaerobic conditions we would expect even greater amounts of arsenic in the water.

### Fish

Arsenic is concentrated in fish from Chautauqua Lake but is not magnified into carnivorous fishes. Some nonessential trace elements such as mercury are concentrated across successive trophic levels, especially in storage compartments such as fat. However, concentration of Cd and Pb are generally diminished across trophic levels, due to differential exclusion by organisms. Organic As compounds might be expected to behave more like Hg, while inorganic As compounds should act more like Pb. Bioaccumulation ratios are similar to those reported for other freshwater fishes (3–30) but much less than those for saltwater fishes and crustaceans (9–4430) (Woolson 1975).

In model ecosystem studies (Isensee et al. 1973, Schuth & Isensee 1974, Woolson 1975), arsenic as cacodylic acid and dimethylarsine was rapidly accumulated by fish. Catfish had a BR value of  $< 1$  and gambusia had BR values ranging from  $< 1$  to 130. These findings and our results suggest that arsenic in fish is in equilibrium with arsenic in ambient waters. Sorenson (1976) found As accumulation by the green sunfish (*Lepomis cyanellus*) to be proportional to the arsenate water concentration. This hypothesis is supported by Penrose (1975) who reported that brown trout (*Salmo trutta*) rapidly excreted excess amounts of orally administered arsenic and that this process was mediated in part by synthesis of an organo-

arsenic compound within the gastrointestinal tract.

Arsenic is not normally distributed in the fish populations studied. Although sample sizes are small ( $< 16$  fish per species), our data support the findings of Giesy & Weiner (1977) that nonessential trace elements are not normally distributed but log normally distributed in organisms. Liebscher & Smith (1968) found As concentrations in humans to be log-normally distributed and suggested that As concentrations were not homeostatically maintained. A constant elimination rate tending toward total elimination of As, rather than elimination to a prescribed concentration would result in a positively skewed distribution. Caution must be applied in comparing mean or median concentrations between species of fish in the same or different bodies of water since these measures may not be the best estimate of a toxicant's distribution within a given fish population.

Arsenic concentrations in Chautauqua Lake fish are generally greater than, but of the same order of magnitude as, those reported for fish from Lakes Erie and Ontario (Pillay et al. 1974, Uthe & Bligh 1971) and lakes and rivers in New York (Pakkala et al. 1972). Lucas et al. (1970) report values of arsenic (dry wt) for Great Lakes fish that are much lower than those given in other studies. There has been a large increase in arsenic concentration in crappies (*Pomoxis*) in Chautauqua Lake since 1959. Ullmann et al. (1961) reported concentrations of  $< 0.10$ – $0.28 \mu\text{g g}^{-1}$ . In 1976, concentrations were  $0.12$ – $2.04 \mu\text{g g}^{-1}$ . This increase appears to be related to the rise in arsenic concentration in lake waters over the same period of time. The black crappie (*P. nigromaculatus*) concentrated the greatest amounts of arsenic of all the fishes studied. This species may be a good indicator species for monitoring arsenic concentrations in fish communities from other bodies of water as well.

### Impact on lake ecosystem

Arsenic concentrations in Chautauqua Lake are below levels toxic to adult fish. However, Spotila and Paladino (personal communication) found that  $50 \mu\text{g l}^{-1}$  As as sodium arsenite ( $\text{NaAsO}_2$ ) was toxic to newly hatched muskellunge embryos (*Esox masquinongy*). Concentrations of arsenic are as high as  $114.8 \mu\text{g l}^{-1}$  in lake water and  $308 \mu\text{g l}^{-1}$  in well water at the state fish hatchery. These arsenic levels could be toxic to sensitive early life history intervals of muskellunge.

In 7 of the last 13 years, total mortalities of muskellunge fry occurred at the state fish hatchery. In

addition, there has been a decline in the muskellunge population in the lake since the early 1960's (Bimber, personal communication). Arsenic contamination may be partly responsible for these phenomena. Further study is needed to determine arsenic chemical composition in lake and hatchery well water, its toxicity to newly hatched fish, and its impact on the reproductive success and population dynamics of resident fishes.

### Public health considerations

Arsenic concentrations in Chautauqua Lake waters are less than the maximum amounts reported for freshwater lakes and rivers in the United States (up to  $1100 \mu\text{g l}^{-1}$ ) but more than those reported for Lakes Michigan and Superior ( $0.1$ – $2.4 \mu\text{g l}^{-1}$ ) (Woolson 1975). They are above the United States Public Health Service recommended maximum concentration of  $10 \mu\text{g l}^{-1}$ . Although the average concentration of arsenic in the lake ( $49.0 \mu\text{g l}^{-1}$ ) is just below the maximum permissible limit ( $50 \mu\text{g l}^{-1}$ ) for arsenic in a public drinking water supply, many of the lake water samples exceeded this limit. Public health consequences are difficult to assess because of differential toxicity of different arsenic forms and because we do not presently know the form or forms of arsenic that are in Chautauqua Lake waters.

Arsenic concentrations in Chautauqua Lake fish do not appear to pose a health hazard for human consumers. Salt water fish contain greater concentrations of arsenic ( $0.4$ – $11.7 \mu\text{g g}^{-1}$ ) than fish in Chautauqua Lake and other freshwaters (Kennedy 1976, LeBlanc & Jackson 1973, Penrose et al. 1975). This arsenic is in an organic form that is biologically and chemically stable and probably nontoxic (Penrose et al. 1975). When humans and rats ate shrimp containing 121 ppm As more than 98% of the As was excreted after 4 days and no ill effects were noted (Coulson et al. 1935).

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