

# History of Toxic Metal Discharge to Surface Waters of the Aberjona Watershed

HENRY M. SPLIETHOFF AND  
HAROLD F. HEMOND\*

*Ralph M. Parsons Laboratory, Department of Civil and Environmental Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139*

A history of waterborne export of toxic metals from the industrial/residential Aberjona Watershed, north of Boston, MA, has been reconstructed from an analysis of sediment cores from the Upper Mystic Lake, together with historical records of industrial activity on the watershed. The lake sediments exhibit complex profiles of arsenic, cadmium, chromium, copper, lead, and zinc. These profiles are interpretable in terms of historical industrial activities on the watershed and transport via the Aberjona River to the Upper Mystic Lake. High concentrations (in excess of 1000 mg/kg arsenic and chromium) occur in two zones, at depths between 25 and 35 cm and between 50 and 70 cm in the sediments of this lake. The deeper zone corresponds to the early 1900s, a period of intense industrial activity during which chemical and leather industries are known to have made direct discharges of metals to surface waters. Sharp decreases in metals concentrations subsequently occurred as industry declined, and concurrently, much of the watershed was sewered in the mid-1930's. The more recent period of metals transport to the lake, which occurred ca. 1960, evidently was the result of remobilization of wastes deposited on the watershed decades earlier.

## Introduction

The history of anthropogenic disturbance of a watershed can often be studied through chemical and geochronological analysis of lake sediment cores (1-7). In the present study, such analysis has been carried out on cores from the Upper Mystic Lake, which receives drainage from the Aberjona Watershed in eastern Massachusetts, in an effort to assess the timing and magnitude of historical toxic metal releases in the watershed.

The Aberjona Watershed covers 65 km<sup>2</sup> of densely populated land in the northern suburbs of Boston, MA (Figure 1). Historically, the dominant industries in this area were leather tanning (8) and chemical manufacture (9). Currently, numerous hazardous waste sites are being

investigated by the Massachusetts Department of Environmental Protection (DEP), and two sites, Industri-Plex and Wells G & H (10-14), have been designated as Superfund sites.

Aquatic sediments throughout the watershed contain significant levels of metal contamination (8, 9, 15), suggesting that transport by the Aberjona River has had a dominant role in determining the distribution of these contaminants. There is thus concern about the possibility of present or past human exposure to toxic metals outside of, as well as within, waste site boundaries. The present study was undertaken to elucidate the historical transport of metals by the river and, thus, the possibilities for human exposure to toxic metals via the surface waters of the watershed. Cadmium, chromium, copper, zinc, lead, and the metalloid arsenic (hereafter all referred to as "metals") were examined.

## Methods and Materials

**Study Site.** The Upper Mystic Lake (Figure 1), formed in a 58-ha kettlehole, receives the entire flow of the Aberjona River. The lake has two other minor inlets that account for less than an estimated 5% (0.037 m<sup>3</sup>/s) (16) of the average inflow from the Aberjona River (0.78 m<sup>3</sup>/s), and they are not believed to be a significant source of metallic contaminants. Two shallow embayments at the north end have a combined area of 23 ha. The main basin has an area of 35 ha, a mean depth of 13 m, a maximum depth of 25 m, and a volume of  $5.4 \times 10^6$  m<sup>3</sup>. Volume of the entire lake is  $5.7 \times 10^6$  m<sup>3</sup>. Average hydraulic residence time is 100 days (16), shoreline length is 5300 m, and shoreline development is 1.8. The lake is dimictic. The Upper Mystic Lake was tidally influenced (but presumably nonsaline) until 1864, when a dam was constructed, separating it from the Lower Mystic Lake and the Mystic River and raising its water level by 2 m. In addition to its use as a drinking water supply between 1864 and 1898, the Upper Mystic Lake has been heavily used for recreation.

**Sediment Coring and Core Subsectioning.** Sediment cores were collected from the deep center region of the Upper Mystic Lake, at depths equal to or greater than 22.5 m (Figure 1). Cores were obtained by freeze coring, which allows sediments to be frozen in place, preventing disruption by bubbling of trapped gases and maintaining the integrity of sediment layering. Two freeze corers (Figure 2) were developed as modifications of previously published designs (17-20). Freeze corer no. 1 was constructed of 7.6 cm diameter aluminum tubing with a flat cut-away section for the freezing of sediment, fins for stability during descent, a Teflon separator to facilitate core removal, and internal PVC insulation to minimize heat transfer through the surfaces where frozen sediment is not desired. When data from corer no. 1 revealed the need for longer cores, a second device (corer no. 2) was constructed from a  $7.6 \times 7.6 \times 125$  cm section of Extren (fiberglass reinforced resin-based) square tubing, one face of which was machined away and fitted with an aluminum plate. Extren is relatively non-conductive to heat (conductivity of 7 versus 2000 cal·cm·cm<sup>-2</sup>·hr<sup>-1</sup>·K<sup>-1</sup> for aluminum), which results in most of the recovered sediment adhering to the aluminum plate, and conservation of the coolant (a slurry of denatured ethanol and crushed dry ice).

\* To whom correspondence should be addressed; telephone: 617-253-1637; fax: 617-253-7475; e-mail address: HFHemond@mit.edu.

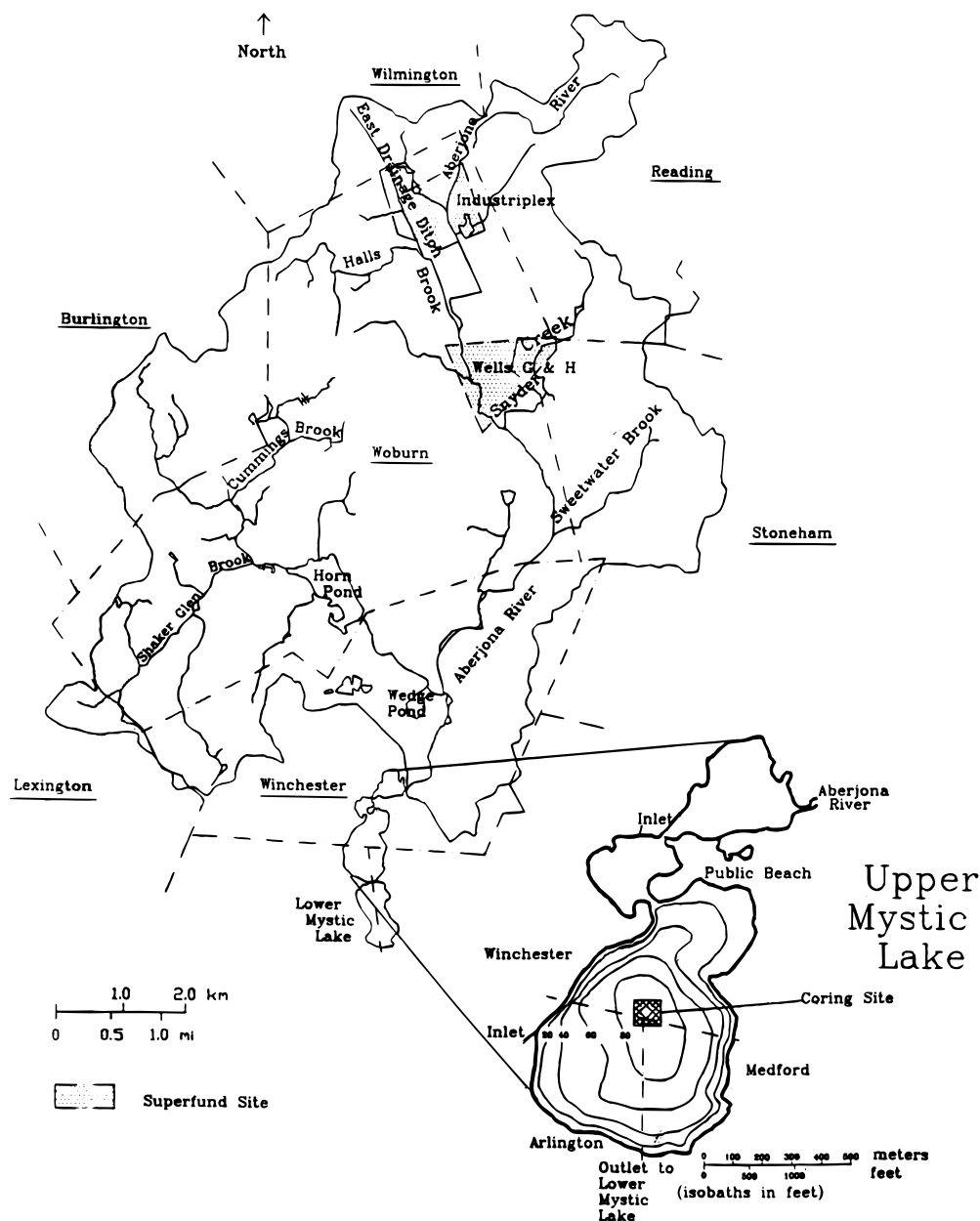


FIGURE 1. Aberjona Watershed map showing location of EPA Superfund sites, town boundaries, and sediment core sampling locations.

A 12-ft-long square-ended aluminum “duck” boat was modified for sampling purposes. A 25 × 25 cm hole was cut into the center of the bottom of the boat, and an open-ended aluminum box was welded in place to provide a coring/sampling well through which large forces could be exerted on the corer without compromising the stability of the vessel. A wooden frame with a free-swinging block was built over this well, and a hand-powered winch was anchored to a thwart to facilitate withdrawal of the corers.

Control of the penetration of corer no. 1 was accomplished by varying the number of small removable floats attached to the tether. Penetration test drops were made by filling the corer with water to approximate the submerged mass of the coolant and monitoring the depth of penetration with vertical smears of vacuum grease along the outside of the corer. Corer no. 2 was lowered gradually into the sediment; penetration was adjusted with lead weights placed internally at its tip.

After penetration conditions had been established, coolant was added, and the corer placed in the sediment.

When CO<sub>2</sub> bubbles reaching the lake surface had begun to subside (approximately 15 min), the corer was swiftly retrieved and cleaned of unfrozen sediment. Frozen slabs of sediment were gently pried off after filling the corer with lake water to warm it. Sediment slabs were sealed in plastic wrap and stored on dry ice for transport to the lab. Slabs of frozen sediment 30–40 cm long with a thickness of 0.5–1.5 cm were obtained from corer no. 1, while corer no. 2 produced slabs about 80–100 cm in length, with a thickness of 1.0–2.0 cm.

Cores were subsectioned with an electrically heated tungsten wire stretched across a hacksaw frame. Prior to cutting, cores were tempered at 0 °C over an ice bath to improve cutting rate and to prevent refreezing of the cut before its completion. Cores were divided into 2.0 cm-long increments, weighed, dried at 95 °C for 24 h, and reweighed.

**Analytical Techniques.** Dried sediment increments of core no. 4 (UML932904#2) were homogenized by mortar and pestle and combined, when necessary, to form 2-g

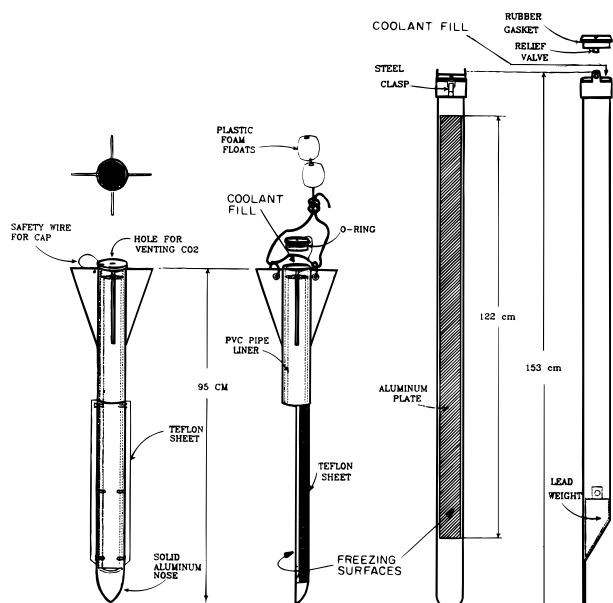


FIGURE 2. Freeze corer designs no. 1 (left) and no. 2 (right).

subsamples, which were analyzed for Cs-137 by counting  $\gamma$  emission at 661 KeV with a germanium detector (Canberra, Model GL2020) and multichannel analyzer (Canberra, Series 40).

One portion of each increment of core no. 1 (UML311090) and core no. 2 (UML310791#2) was combusted at 550 °C for 5 h to determine the fraction of organic matter (FOM) (21). Another portion was extracted in hot concentrated reagent-grade nitric acid and 30% hydrogen peroxide and then refluxed in concentrated reagent-grade hydrochloric acid (22). This procedure closely approximates EPA Method 3050 for sediments (23). Reagent blanks were digested along with every group of samples.

Total metal analysis was performed on digestates using a sequential inductively coupled argon plasma (ICAP) emission spectrometer (Atomscan 25, Thermo Jarrell Ash Corp., Franklin, MA). Approximately 10% of the samples were analyzed in duplicate to determine instrumental precision (~5%). Generally, standards and blank solutions were analyzed after every four samples to check for instrumental drift (<10%). As a further quality control measure, surface sediments collected in the proximity of the coring sites were digested with analyte spikes; recovery was generally high (>80%).

Half of the digestate from each increment of core no. 1 and three-quarters of the digestate of each increment of core no. 2 were reserved for Pb-210 dating. Actual counts were of Po-210, a granddaughter nuclide assumed to be in secular equilibrium with Pb-210 in the sediment (and hence to have the same activity). Each subsample was spiked with a known amount of Po-209, which served as an internal standard, accounting for both detector efficiency and plating efficiency. Both nuclides were simultaneously plated onto silver planchets by the method described by Benoit and Hemond (24).  $\alpha$ -Emission was counted using an  $\alpha$ -spectrometer with a surface barrier detector (Ortec, Model 576) and a multichannel analyzer (Canberra, Series 40).

## Results and Discussion

All cores were taken from the deep central basin of the lake. Core no. 1 was retrieved from a water depth (surface at lake to top of sediment) of 22.5 m (Figure 1); it was 43

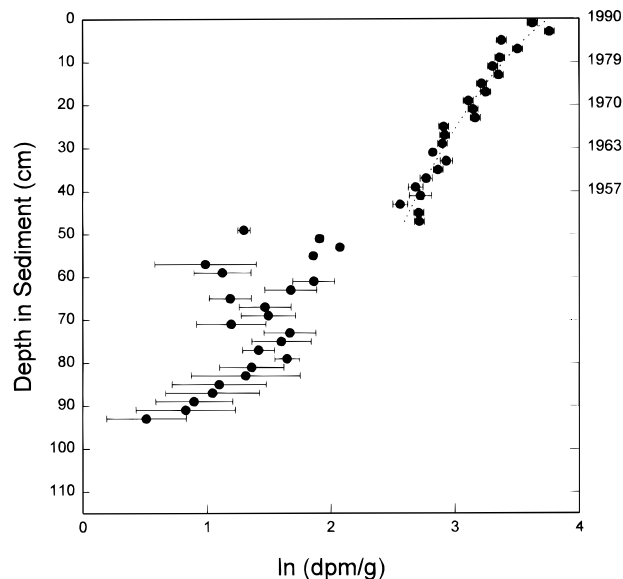


FIGURE 3. Unsupported Po-210 activity in the Upper Mystic Lake (0–30 cm data from core no. 1; 30–93 cm data from core no. 2).

cm in length and included a well-demarcated sediment–water interface. Cores 2–4 were retrieved from a water depth of 24 m and were 96, 92, and 84 cm, respectively, in overall length. At several locations throughout the cores (5–9, 13–20, and 57–71 cm), groups of regularly spaced laminations could be seen. The preservation of these laminations through the sampling process provides evidence that the freeze-coring procedure caused little mixing or smearing of the sediments. The upper 20 cm of sediment was not recovered in core no. 2 because of coolant depletion; however, the vertical pattern of laminations as well as the metals profiles indicated that cores no. 1 and no. 2 overlapped between the depths of 20–45 cm, making it possible to accurately construct a composite profile from the two cores. Because of the sharp and distinctive features of the metals data, uncertainty of matching within the region of overlap appears to be no greater than the resolution of individual increments (2 cm). This was confirmed independently with core no. 3, which replicated and gave excellent agreement with the composite of cores no. 1 and no. 2, as illustrated by the arsenic and cadmium profiles (see below). Core no. 4 matched very well with cores 1–3 with respect to the two parameters that were measured, fraction solids and fraction organic matter.

**Chronological Inferences. Pb-210.** The unsupported Pb-210 profile in the Upper Mystic Lake sediment is presented in Figure 3. Unsupported activity is total activity minus supported activity; the latter was taken as average activity at the base of the profile (1.9 dpm, between 93 and 113 cm). Activity is plotted against depth rather than mass accumulation, because the freeze-coring process may have altered the fraction solids content of the core, rendering mass accumulation rates uncertain. Statistical counting error ( $1\sigma$ ) is indicated by horizontal error bars or is less than the width of the symbols.

The Pb-210 profile shows no evidence of a surface mixed layer. However, there is considerable scatter between 47 and 71 cm. Anthropogenic disturbances may have resulted in a dilution of Pb-210 activity in the bottom sediments during the time period represented by this lower region. Of particular note is that this region of high scatter corresponds to the period of heaviest contamination of the watershed;

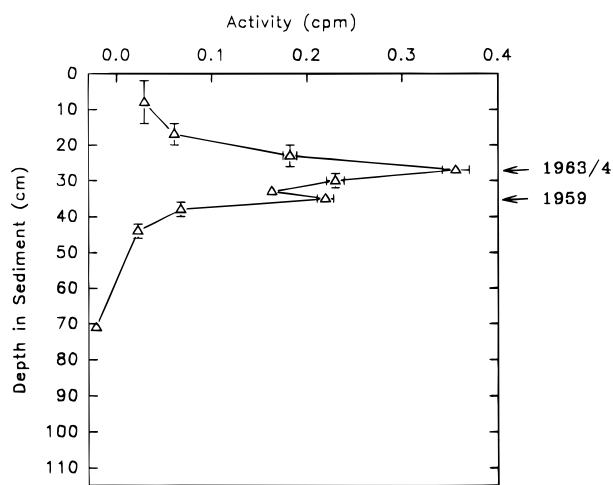


FIGURE 4. Profile of Cs-137 activity in Upper Mystic Lake (core no. 4).

metal contamination may have altered the scavenging efficiency of the lakes by biological (e.g., toxicity to plankton) or chemical means. Accordingly, dates were inferred from the Pb-210 profile only in the smoothly varying region of 0–47 cm, using a polynomial curve fit of the data, and the constant input concentration (CIC) model (25).

**Cs-137 Horizons.** The atmospheric flux of Cs-137 from fallout in the Northern Hemisphere during the years of atmospheric nuclear weapons testing reached a maximum in 1959 and, after a short period of decline, reached a second and greater maximum in 1963–1964 (26). The Cs-137 profile (Figure 4) exhibits a bimodal distribution of activity, with a primary peak in the 26–28-cm increment and a secondary peak in the 34–36-cm increment (core no. 4, from which the Cs-137 data were obtained, was collected 2.4 years later than core no. 1 and 1.7 years later than core no. 2; therefore, the depth axis shown in Figure 4 has been offset 2 cm downward relative to the core so that zero depth corresponds to mid-1991, the time at which core no. 2 was taken). The Cs-137 peak structure in core no. 4 is highly coincident with the temporal pattern of atmospheric deposition, and provides a basis for assigning the dates 1959 and 1963–1964 to the 34–36- and 26–28-cm increments, respectively.

**Industrial Horizons.** In the 19th century, the Aberjona Watershed, like many other areas in the northeastern United States, entered a period of rapid industrial development. Development resulted in the release of pollutants, many of which were transported by the Aberjona River to the Upper Mystic Lake. Lead, associated with the increased combustion of fossil fuels ca. 1850, is likely to have been the earliest (27). In 1970, lead began to be phased out as a gasoline additive in the United States, resulting in a decrease in local and regional atmospheric deposition of this metal (28). Anthropogenic arsenic release to the Aberjona River began with the use of arsenic-containing pyrites in sulfuric acid manufacture in 1888 and the manufacture of arsenical pesticides in 1899. Production of sulfuric acid reached a peak in 1916 (9). The tanning industry grew throughout the 19th century and began releasing chromium to the river after the introduction of chromium salts to the tanning process ca. 1900. Cadmium, copper, and zinc were also released in association with both the tanning and chemical industries. In 1925, the tanning industry reached its peak production year (8). Shortly thereafter, economic decline

coupled with the development of local sewer systems contributed to a general decrease in metal loading to the Aberjona River.

This relatively well-documented industrial history correlates with metal horizons in the deeper portion of the cores. The rises of lead, arsenic, and chromium concentrations above background in the sediment column were assigned dates of 1850, 1888, and 1900, respectively. The peak years of arsenic release (1916) and chromium release (1925) were assigned to depths at which the maxima of these metals were found (the chromium and arsenic maxima between 30 and 40 cm are more recent, as discussed below). The more shallow lead maximum was assigned the date of 1970. The dates so assigned are consistent among themselves and consistent with both Pb-210 and Cs-137 dates.

**Interpretation of Core Chronology.** Because the Cs-137 peaks occur within the top 47 cm of the sediment column, the profile of Cs-137 is an independent test of dates derived from Pb-210 in this interval. Dates inferred from the Pb-210 profile for the two maxima of Cs-137 activity (1960 and 1965, respectively) are in excellent agreement with the dates inferred from Cs-137 (1959 and 1964). Further, the approximate 1-cm spacing of visible sediment laminations between 5 and 9 cm in core no. 1 and between 13 and 20 cm in core no. 2 compares well with the average accumulation rate of 1 cm/year in the upper portion of the sediment column that is implied by both Pb-210 and Cs-137 dating. Moreover, the date of maximum atmospheric lead deposition (1970) is consistent with the date of the lead maximum in the sediment as determined from Pb-210 (1973).

Because of the aforementioned scatter in Pb-210 between 47 and 71 cm and the existence of sharp, highly resolved metal profiles in this depth interval, we considered the documented industrial history of the watershed to be the more reliable tool to assign dates to the lower portion of the sediment column (although the results are not inconsistent with Pb-210 in this interval). A composite geochronological chart (including all dates or sediment accumulation rates derived from Pb-210, Cs-137, laminations, and industrial history) is presented in Figure 5. Dates inferred from the early industrial history of the watershed indicate that the present-day thickness of an annual increment of sediment deposition is approximately 0.5 cm, in good agreement with the approximate 0.5-cm spacing of laminations visible between 57 and 71 cm in core no. 3. The greater thickness of the annual sediment increment in the more recent sediments may reflect an historical increase in deposition rate due to watershed development as well as less compaction.

**Temporal Patterns for Individual Metals. Chromium.** A major source of chromium on the watershed was the leather industry, which over its lifetime included approximately 100 tanning and other leather processing companies. Prior to 1900, vegetable tanning processes were dominant. The industry reached a peak in 1925 (Figure 6), after which a precipitous drop in leather tanning occurred (8). The lower Cr deposition episode in the sediment profile (53–63 cm) is thus constrained above and below by these dates (see above).

A second peak in sedimentary chromium concentration begins at 35 cm, corresponding to a calculated date of 1959, with concentration remaining high (>1000 mg/kg) until the mid 1960's (approximately 25 cm). Records of com-

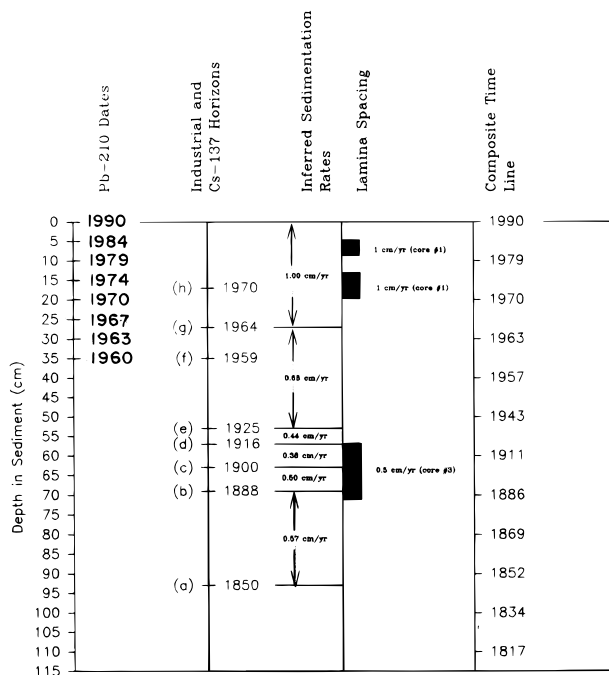


FIGURE 5. Composite of date horizons as established from Pb-210, Cs-137, industrial history, and visible sediment laminations. (a) Appearance of Pb: increase in fossil fuel burning (33). (b) Appearance of As: introduction of sulfuric acid manufacture (9). (c) Appearance of Cr: introduction of chrome tanning (8). (d) Peak of As: peak year of sulfuric acid manufacture (9). (e) Peak of Cr: peak year of leather industry (8). (f) Peak of Cs-137: peak of Cs-137 fallout (26). (g) Peak of Cs-137: maximum Cs-137 fallout (26). (h) Peak of Pb; beginning of leaded gas phaseout (35).

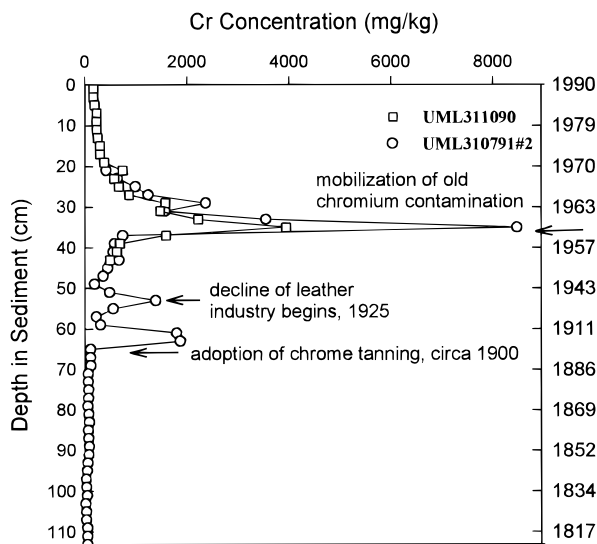


FIGURE 6. Chromium profile in the Upper Mystic Lake.

merce do not reveal any manufacturing activity during this time period that is likely to be responsible for this chromium deposition episode. However, between 1934 and 1969, a major industrial site north of present-day route 128 was occupied by rendering operations, which utilized chrome-tanned hides in the process of making glue (12). Rendering wastes were disposed of in several locations throughout the site. Comparison of photographs from 1952 and 1967 (29) show that earth- and/or waste-moving on this site near the Aberjona River may have been ongoing during the time period when high levels of chromium were being deposited in the lake. According to Darcy (30), earth-moving activities

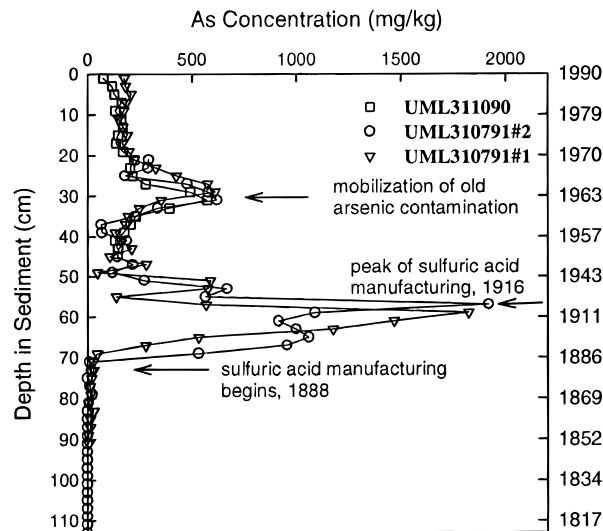


FIGURE 7. Arsenic profile in the Upper Mystic Lake. The extent of agreement of data from three cores is illustrated.

were most intense in the early 1960's, although they continued on a sporadic basis into the 1970's. Thus, it is possible that wastes from the rendering of chrome-tanned hides were physically mobilized and became a major source of chromium to the Upper Mystic Lake. In addition, the organic-rich and reducing conditions established in rendering waste piles could have enhanced the mobility of chromium (via sorption to organic colloids) (31) and other metal contaminants present in adjacent soils. A similar explanation has been given for groundwater plumes of chromium and arsenic beneath and downgradient from the Industri-Plex Site (10, 31). Another possible release mechanism is the failure of settling lagoons for rendering waste, which were reportedly poorly maintained and were frequently observed during the late 1950s to be overflowing (12).

**Arsenic.** Arsenic, like chromium, exhibits two major maxima in the sediment profile, with the lower region extending from 69 cm (slightly deeper than the lowermost portion of the chromium peak) to 51 cm (Figure 7). The position and magnitude of this deeper arsenic deposition episode are consistent with known industrial history, and provide constraints, as discussed above, to the dates at which this deposition occurred.

Arsenical pesticide manufacture in the northern part of the watershed began in 1899 (9). The burning of arsenic-containing iron pyrites in sulfuric acid manufacture began in 1888, thus predating the widespread application of chrome tanning, a conclusion that is consistent with the sediment profiles. The peak of sulfuric acid manufacture, the largest source of arsenic release to the watershed, occurred in 1916. Arsenical pesticide and sulfuric acid production on the watershed declined sharply with the acquisition of Merrimack Chemical by Monsanto Corporation and the relocation of its chemical manufacturing facilities to Everett, MA (outside of the watershed), in 1931 (9).

The more recent arsenic maximum, at 29 cm in depth, corresponds to a date of 1964; the width of this peak implies that the arsenic release continued throughout the 1960s. It is possible that this upper arsenic peak is associated with mobilization from the Industri-Plex site, triggered by the same kind of earth-moving activities that may have led to

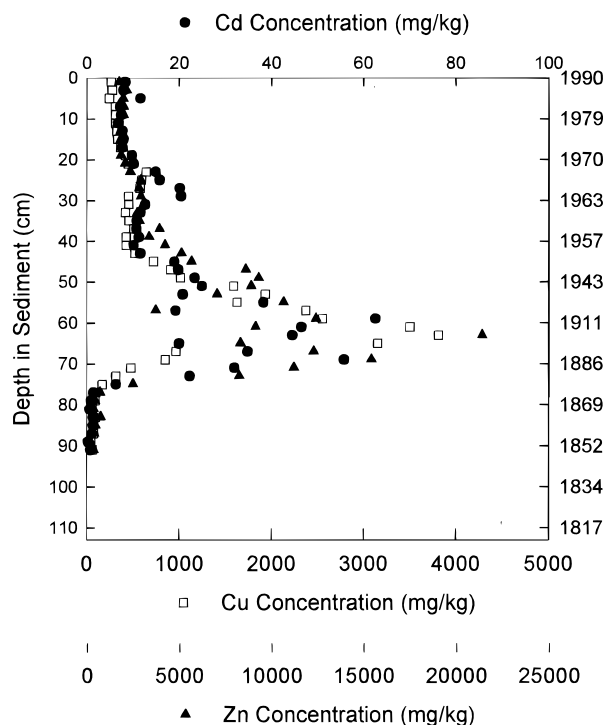


FIGURE 8. Cadmium, copper, and zinc concentration profiles in the Upper Mystic Lake.

chromium release. However, the As peak is slightly (2–4 cm) higher in the profile than the upper chromium peak. This small, but definite, difference in depth between the uppermost chromium and arsenic peaks may reflect a progression of disturbances across a heterogeneously contaminated parcel of land.

**Cadmium, Copper, and Zinc.** The most significant source of copper, which was used in dyes and pigments and as a bactericide, was probably the tanneries. Indeed, the copper profile between 50 and 70 cm matches well with the historical release pattern of copper by the tanning industry between about 1910 and 1930 (8). Copper exhibits its highest concentrations at very nearly the same locations as the lowermost chromium maxima (Figure 8), further suggesting a leather industry source. However, elevated levels of copper in the bottom sediments of the lake are found several centimeters below the beginning of elevated chromium levels, as might be expected given that chromium use was introduced into leather industry that would have already been using copper. There is only a slight elevation in the concentration of copper near 25 cm however. Thus, copper was not co-released with chromium or arsenic ca. 1960. Only a minimal association between copper and chromium contamination has been reported in the Industri-Plex Site (13).

The profile of zinc (Figure 8) is similar to the copper profile, in that it exhibits its highest concentrations between 50 and 80 cm in depth. Zinc was used in large amounts in leather tanning (8), although it was also present in iron pyrites at significant concentrations and was used in the production of various salts including zinc sulfate and zinc chloride on the Industri-Plex Site in the early 1900s (12). Its correspondence with copper suggests that the tanneries may have been the largest source.

The cadmium profile (Figure 8) is similar to the arsenic profile, although the elevated concentrations of cadmium begin at a somewhat lower depth than for arsenic.

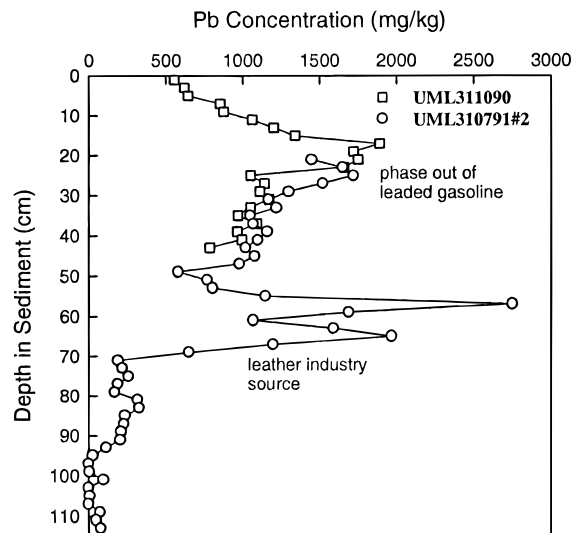


FIGURE 9. Lead concentration profile in the Upper Mystic Lake.

Cadmium may have a mixed industrial source in which both tanning and chemical manufacturing were prominent. Cadmium was found in trace quantities in pyrites utilized during the period when sulfuric acid was being manufactured at the Industri-Plex Site (12). The strong correlation between cadmium and zinc concentrations in the sediment profile as well as in surface sediments throughout the watershed (15) is consistent with the known natural association of these elements (32).

**Lead.** Lead has a major peak between 50 and 70 cm in depth (Figure 9), coincident with a period of peak industrial activity within the watershed. Several industrial operations were probable early sources of lead: leather tanning (8), the manufacture of lead arsenate (9), the production of lead-containing dyes (13), the disposal of lead-containing pyrites, and fossil fuel combustion.

In contrast to other metals, the lead profile exhibits (a) significant elevation above background as deep as 93 cm and (b) a steadily increasing concentration from approximately 50 cm in depth to approximately 20 cm, above which the concentration drops off almost monotonically to the surface of the sediment. The early increase of lead above background levels at 93 cm implies a source that predates that of the other metals and is consistent with the findings of studies that correlate the first appearance of lead in sediments with an increase in coal combustion in the middle of the 19th century (27). The shape of the upper lead peak suggests that, unlike arsenic and chromium, the more recent deposition of lead (ca. 1950 to present) is not strongly associated with an earth-moving disturbance. Instead, the lead profile tracks the use of tetraethyl lead, introduced as a gasoline additive in 1923 (33); the decline above 17 cm, a depth for which Pb-210 data indicate a date of 1973, is consistent with decreasing levels of environmental lead as a consequence of the phaseout of leaded gasoline beginning in 1970 in the United States (28). The absolute levels of lead in these sediments are higher than those levels (<500 mg/kg) typically attributed to auto emissions by other researchers (7, 34). However, this may not be inconsistent given the heavily congested, extensively paved nature of the Aberjona Watershed. Not only is the traffic density high, but in addition, atmospherically deposited lead may be transmitted to aquatic sediments more efficiently from paved surfaces than from vegetated

surfaces, the latter exhibiting high infiltration capacity, little or no surface runoff, and high sorptivity for lead. The presence of similar lead profiles from sediments of water bodies in the less industrial and otherwise less contaminated western part of the Aberjona Watershed (15) also implicates a primarily atmospheric source for lead pollutant in recent decades.

**Iron, Manganese, and Fraction Organic Matter.** To rule out the possibility that strata with elevated metal concentrations could have resulted from the existence of layers of relatively higher metal sorbing capability, the sediments were examined for strata of elevated iron, manganese, and organic matter. The sediments generally have a high organic content (between 20 and 40%), with the exception of the depths 93–107 cm, where a sandy layer was found. The sandier sediment may have been deposited when the dam between the Upper and Lower Mystic Lakes raised water levels in the upper lake and created the upper forebay in 1864. The absence of large fluctuations in organic content above 93 cm appears to preclude a major influence of organic matter on the toxic metal profiles.

The iron profile shows variations in concentration that are also of small magnitude when compared with those of the toxic metals. The manganese profile has no significant peaks corresponding to the positions of the toxic metals maxima and, therefore, may also be presumed to not be instrumental in trapping and concentrating metals. Very narrow peaks of manganese were found at 59 and 73 cm in both cores 2 and 3 and are not readily explained. It is possible that they represent an unidentified industrial source of brief duration.

## Conclusions

The Aberjona Watershed has been heavily affected by industrial activity throughout the past century. Major industrial activities as well as land and waterway alterations can be identified in the historical record. Sediment deposition in the Upper Mystic Lake is rapid, consistent with a eutrophic lake in a developing urban watershed. Sediments below approximately 80 cm (93 cm in the case of lead) have constant, low metal concentrations with respect to depth and appear to represent natural background levels for these elements. The change in land use from agriculture to manufacturing industry, dominated by the leather and chemical industries, and then to residential and light industrial/commercial usage is reflected in the patterns of metals concentrations in the sediments above this depth.

Sediment metal concentrations decreased considerably after the 1930s because of the depression of the leather tanning industry, the relocation of the chemical industry, and the sewerage of much of the watershed. Subsequently, metal concentrations in the sediments remained constant until the late 1950s to early 1960s (with the exception of lead, whose concentration became controlled by atmospheric deposition) when a disturbance, possibly earth-moving, resulted in the mobilization, transport, and redeposition of metals originally deposited during former industrial activity. Following this disturbance, the metals concentrations in the sediments of Upper Mystic Lake decreased to present, relatively steady levels that are still significantly above natural background, especially in the case of arsenic. The present input of metals to the lake

most likely represents ongoing mobilization of historically deposited material (35), although the possibility of continuing industrial sources cannot be ruled out entirely.

## Acknowledgments

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