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A history of acid mine contamination, recovery, and eutrophication in Sandy Lake, Pennsylvania

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Abstract Sandy Lake, PA, was subject to acid mine contamination in the 1860s, which led to complete extirpation of fish. The mine effluent was soon diverted, but the watershed experienced continued deforestation, agricultural development, and population growth to the beginning of the twentieth century. Paleolimnological analyses of metals and diatoms clearly show the onset of acid mine drainage with rapid increases in concentrations of iron, manganese and sulfur, and decreases in diatom-inferred pH in the sedimentary profile, and a very rapid recovery following remediation. Diatom-inferred phosphorus suggests a period of oligotrophication following diversion of the acid mine drainage and reduction in lake catchment area. However, analyses of nutrients and algal pigments suggest continued eutrophication to the present in spite of increasing forest cover and a stable population over the last century. Pigments indicate a tenfold increase in phytoplankton, with cyanobacteria becoming a more significant portion of the biomass. Accumulation of sedimentary phosphorus has increased by a factor of 4 and sedimentary organic carbon by a factor of 5 since the mid-1800s.

Keywords Acid mine drainage · Metals · Oligotrophication · Eutrophication · Diatoms · Sandy Lake

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Introduction

Sandy Lake, Mercer County, Pennsylvania, is the southern-most natural lake in the glaciated region of western Pennsylvania lying within 20 km of the terminal moraine of the Wisconsin glaciation. It is a lake of remarkably high water quality considering its location (within 100 km of major population centers: Pittsburgh, Cleveland, Erie, with few other natural lakes nearby), its geological setting (Pennsylvanian-era sandstone, limestone, and shale overlain with calcareous glacial outwash), and current land use in its watershed (39.7% pasture/hay, 3.65% row crop). Springtime total phosphorus (TP_{spr}) is about 12–14 μg l^{-1}. The water is moderately hard (alkalinity 60–70 mg l^{-1} as $CaCO_3$), and Secchi disk transparency averages 3.5 m during the summer. The current condition of the lake is probably due to a number of factors perhaps the most important are that the shoreline is in private ownership with no public access, 60% of the shoreline is low-lying wetlands that have prevented extensive lakeshore development, and the few seasonal homes that are on the lakeshore have sewage holding tanks rather than on-lot septic systems. The lake has a diverse recreational fishery, and a diverse aquatic macrophyte community, although the dominant species is the invasive Eurasian watermilfoil (*Myriophyllum spicatum* L.), and since 2000, the lake has supported an established population of zebra mussels (*Dreissena polymorpha* Pallas).

However, the lake water quality had been severely affected by mine drainage in the mid- to late-nineteenth century. A report of the State Commission of Fisheries (Busch 1897) indicates that the operators of coal mines within the watershed turned "sulphur and coal water" into the lake leading to the complete extirpation of fish. Public protest "put a stop to the evil" and the lake returned to "better form" and restocking of pickerel, pike, perch, and bass was underway. The report gives neither the dates of contamination nor the date and method of remediation. Neither the Pennsylvania Fish and Boat Commission nor municipal records of Mercer County or the Borough of Stoneboro contain any mention of this aspect of lake history, and it is probable that the remediation was conducted by the now-defunct Mercer Iron and Coal Company that operated the mines. Nevertheless, an atlas of Mercer County (Anonymous 1873) contains two maps that provide a clue (Fig. 1). The first is a map of Lake Township, showing Sawmill Run as a main tributary of the lake draining the southern portion of the catchment area that contained a number of coal mines. On this map Sawmill Run discharges into the lake at approximately the midpoint along the south shoreline. The second map is a plat map of the Borough of Stoneboro that shows Sawmill Run clearly channelized as it approaches the lake and diverted to discharge into the lake outlet stream approximately 1.1 km east. These maps limit the diversion to a 9 year period between the opening of the first mine in 1864 (Anonymous 1888) and publication of the atlas in 1873. The effect of this diversion was to allow the mine drainage captured by Sawmill Run to bypass the lake. It also reduced the lake's catchment area from 12.9 to 7.0 km^2.

When sulfide-rich minerals in mine spoils are exposed and oxidized, the acid generated results in runoff with low pH and elevated concentrations of heavy metals (Brugam et al. 1988; Friese et al. 1998; Salonen et al. 2006). Coal mine effluent is particularly rich in S, Fe, Mn, and Al (Rose and Cravotta 1998). In a reanalysis of the USEPA's National Stream Survey data, Herlihy et al. (1990) determined that small (median catchment area 8.1 km^2, median length 2.8 km) streams affected by acid mine drainage had an average sulfate concentration of 157 mg l^{-1}, a mean pH of 4.3, dissolved Fe of 6.7 mg l^{-1}, Mn of 2.4 mg l^{-1} and Al of 1.9 mg l^{-1}. A survey of 71 acid mine drainage-affected streams in the Appalachian coal producing region of Pennsylvania showed an average concentration of 214 mg l^{-1} sulfate, 2.65 mg l^{-1} aluminum, 1.54 mg l^{-1} manganese and 2.68 mg l^{-1} iron (Biesecker and George 1966) and another set of 57 perennial first order acid mine drainage-affected streams in Pennsylvania had average concentration of 397 mg l^{-1} sulfate, 28.5 mg l^{-1} manganese, and 23.8 mg l^{-1} iron. These contrast with 29 nearby unaffected streams having mean concentrations of 23.3 mg l^{-1} sulfate, 0.34 mg l^{-1} manganese, and 0.18 mg l^{-1} iron (Dyer 1982). Consequently, we hypothesize that Sawmill Run was similarly affected once the mining operations of the Mercer Iron and Coal Company started within its catchment area in 1864. Since Sawmill run drained almost half of the historical watershed of the lake, there must have been a significant impact on lake water quality. The effects of low pH and elevated concentrations of dissolved metals on both fish and invertebrates are well known (Driscoll et al. 1980; Burton and Allan 1986; Wong et al. 1999; Borgmann et al. 2004) and the reported extirpation of fish in the lake probably occurred soon after the mine began operation.

Increased sulfate concentrations in the lake water would have resulted in both an increase in the S content of seston (assimilatory sulfate reduction) and an increase in bacterial (dissimilatory) sulfate reduction, particularly if the sediments contained a reservoir of degradable organic carbon (Mitchell et al. 1985; Herlihy et al. 1988; Friese et al. 1998). Both processes would have led to the accumulation of reduced sulfur compounds in the sediments. The diversion of Sawmill Run between 1864 and 1873 would have abruptly ended acid and metal loading, and the lake would have begun to recover. Lake recovery from acid mine drainage is dependent on the extent of water quality and biotic degradation and even the most severely degraded lakes show improvement with time once contamination is reduced or eliminated (Fritz and Carlson 1982; Hutchinson and Havas 1986; Keller et al. 1999; Couillard et al. 2004). The reduction in the lake's catchment area would also have led to reduced nutrient loading, and empirical watershed phosphorus loading models (Dillon and Rigler 1974) predict a decrease in lake phosphorus concentration largely as a result of the effect of reduced hydraulic discharge on the P retention coefficient (Ostrofsky 1978).

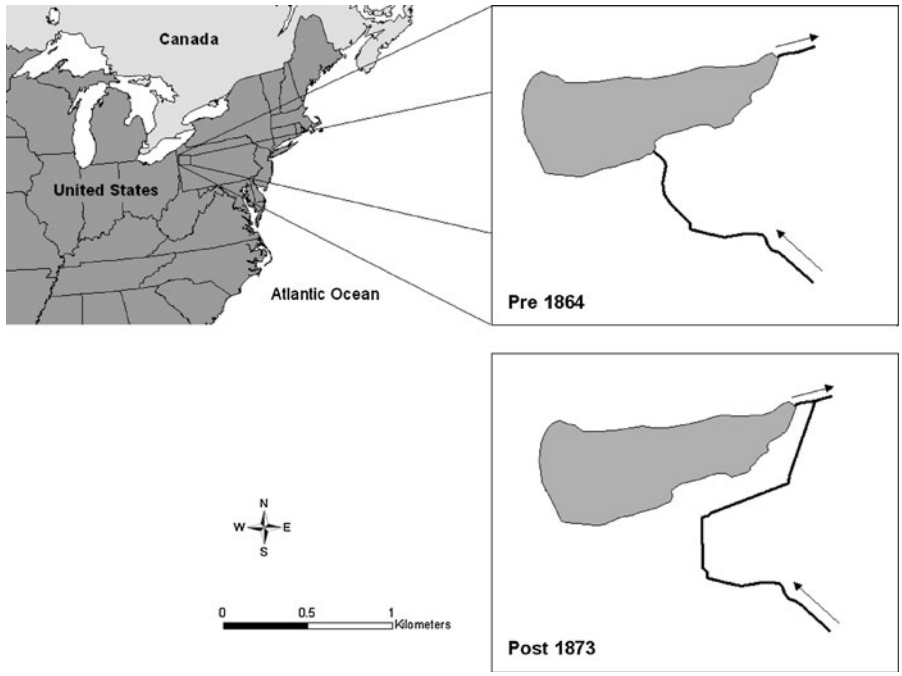

Fig. 1 Map of Sandy Lake showing Sawmill Run entering midway along the south shoreline prior to 1864 (*top right panel*), and with Sawmill Run channelized and diverted to the outlet sometime between 1864 and 1873 (*lower right panel*). Redrawn from Anonymous (1873)

Aside from noting fish mortality in Sandy Lake there is no record of the nature or extent of the water quality or ecological impairment caused by mine runoff, nor is there any record of the trajectory of recovery following its diversion. In addition to the acute impact of mine drainage on the lake and the anticipated oligotrophication from the reduction in catchment area, the watershed has experienced deforestation, the development of agriculture, and limited urbanization. In the absence of water quality records, the deterioration and recovery of the lake ecosystem from mine drainage, the effect of reducing the lake's catchment by almost 50%, and the more gradual process of enrichment resulting from anthropomorphic land use changes might only be inferred from evidence in the sedimentary record. Here we present the results of a paleolimnological analysis Sandy Lake to test our hypothesized history of acid mine contamination and recovery, oligotrophication, and cultural enrichment.

Study area

Sandy Lake (41°20.71′N, 80°06.43′W; elevation 353 m) is located in Mercer County, northwestern Pennsylvania. The area has a humid continental climate, with average annual precipitation of 109 cm, mean annual runoff of approximately 53.4 cm, and average January minimum and July maximum temperatures of −8.6° and 27.5°C, respectively, measured at Mercer, PA, 17 km southwest of the lake. The lake outlet stream is a tributary of Sandy Creek which in turn enters the Allegheny River south of Franklin, PA., and ultimately the Ohio River at Pittsburgh. Forests are dominated by sugar maple, beech, red oak, and cherry, with hemlock and red maple in wetter areas. The lake is a small (area = 60 ha, z_{max} = 14 m, z_{mean} = 6.6 m) kettle with a relatively small watershed (7 km^2 current, 12.9 km^2 historical). Its current hydraulic retention time is 1.12 year. It supports a diverse community of aquatic vascular plants, including state endangered *Myriophyllum verticillatum* L. and *Potamogeton gramineus* L., and state threatened *Schoenoplectus subterminalis* (Torr.) Soják, as well as a diverse population of fish. Sandy Lake is used primarily for fishing, swimming, and waterskiing, with a small park and beach on the south shore, and fewer than 30 seasonal cottages. The village of Stoneboro (population 1104, 2000 census) lies immediately south of the lake although only a small fraction of the village lies within the current watershed boundary. The lake stratifies from May through mid-October, with the thermocline at approximately 5 m. Hypolimnetic water below 9 m becomes anoxic by early July. *Dreissena polymorpha* were first observed in the lake in 2000 (J. Widel, Lakeside Park Co., personal communication) and have been a conspicuous feature on hard surfaces ever since (Bodamer and Ostrofsky 2010).

Historical records indicate that the first permanent settlement in the Sandy Lake watershed was established around 1825, and consisted of a small colony of black freedmen, escaped slaves seeking refuge in a free state, and Native Americans (White 1909). An 1840 census listed a population of only 2 dozen individuals. The colony dwindled to just a few individuals by the 1850s. Early geological surveys indicated the presence of coal in the area coincident with the increasing scarcity of wood to power expanding railroads. In 1862, the Mercer Iron and Coal Company acquired land and mineral rights totaling about 1500 ha. within the watershed and opened the first mine in 1864. This mine was in the Sawmill Run watershed, the major tributary stream of Sandy Lake, and over the next 10 years, over 650,000 tons of coal were mined. The village of Stoneboro was incorporated in 1866 to house an expanding population of mine and railroad workers. By 1870 the borough population had reached 471 residents, and 1186 residents by 1880 (Anonymous 1888). A second mine was opened in 1870, a short distance to the west, and the first mine was phased out by 1880. This second mine was closed in 1906, to be replaced by a third mine nearby, but outside the watershed. During this time, other developed industries included the Sandy Lake Ice Company, Stoneboro Milling Company, and a glass factory. In 1924, the operations of the Mercer Iron and Coal Company were abruptly terminated due to financial and labor problems, and the company was liquidated shortly thereafter. The only business in the watershed to survive the end of the coal era was an amusement park under a series of owners, in a series of locations around the lake. Agriculture within the watershed probably peaked in the late nineteenth or very early twentieth century. The 1913 Stoneboro USGS quadrangle shows that only about 26% of the watershed was forested. The 1970 Sandy Lake USGS quadrangle shows that forests had increased to 38% of the watershed. Current estimates based on USGS National Land Cover Data set (2000) extracted using ArcGIS 9 software (Environmental Systems Research Institute 2004) show forest cover at 53%.

Methods

We collected a 67-cm core in the approximate center of Sandy Lake in February, 2008 using a gravity corer. The water depth was 12 m. The core was extruded in the lab within 24 h. Each 1-cm depth increment was partitioned and a 2-g portion of the fresh sediment was immediately placed into 90% aqueous acetone for the extraction of sedimentary pigments. A second 1-g portion of fresh sediments was used for diatom analysis, and the remaining fresh sediment was weighed, dried at 60°C in an oven, reweighed to determine water content, and ground with a mortar and pestle. These dried samples were used for ^{210}Pb dating (CRS model, D. Engstrom, University of Minnesota), and for the remaining analyses.

Analysis of sedimentary algal pigments followed Swain (1985). Fresh sediments were extracted four times in 10 ml 90% aqueous acetone. The extracts were pooled and made up to 100 ml. A portion of this sample was used to determine chlorophyll derivatives (CD) directly. The remaining portion of the sample was washed with petroleum ether, and the aqueous acetone phase evaporated and redissolved in ethanol for the analysis of oscilloxanthin and myxoxanthophyll. Chlorophyll derivatives are recorded as optical density units per organic C mass (OD_{665} g C^{-1}) whereas oscilloxanthin and myxoxanthophyll are recorded in µg pigment g C^{-1}.

Wet sediment samples for diatom analysis were cleared using HNO_3, HCl, and H_2O_2. Samples were rinsed and decanted several times with distilled water and mounted on permanent slides with Naphrax (Brunel Microscopes Ltd). Rather than examine diatoms in all sections of the core, 13 samples were chosen based on their depth and proximity to major environmental changes as indicated by the C:N ratios and ^{210}Pb dating. Approximately 350 diatoms were identified from each of these 1-cm sections using Patrick and Reimer (1966), Dodd (1987), and Camburn et al. (1986) as taxonomic references. Weighted averaging was used to calculate diatom-inferred pH (DI-pH) and diatom-inferred phosphorus concentrations (DI-P) based on published pH and P optima for diatom species in northeastern U.S. lakes (Dixit et al. 1999).

Sediment carbon, sulfur, and nitrogen were measured on oven-dried sediments using a LECO elemental analyzer, model CNS-2000. Biogenic silica (BSi) was measured using the method outlined by DeMaster (1981). Dry sediment samples (approximately 40 mg) were extracted for 2, 3, 4, and 5 h in hot (85°C) 1% sodium carbonate. Silica was

measured on these extracts colorimetrically using the procedure outlined by Strickland and Parsons (1968). Silica concentrations were graphed as a function of extraction time and the extrapolated Y-intercept was taken as a measure of biogenic silica.

Metals (Fe, Mn, Al, Zn, Pb) were analyzed by atomic absorption (Perkin Elmer AAnalyst 700) on HNO_3–H_2O_2–HCl digested samples (EPA undated). Mercury was measured with a Milestone DMA-80 direct mercury analyzer.

Sedimentary phosphorus species were determined using a sequential extraction with NH_4Cl, NaOH and HCl (Hieltjes and Lijklema 1980). The technique isolates interstitial and loosely bound-P (NH_4Cl-extractible), metal bound-P (NaOH-extractible) and calcium-bound (apatite)-P (HCl-extractible). NH_4Cl- and NaOH-extractible-P fractions are both potentially available for biological uptake in the sediments, and can be recycled to the water column to support planktonic production so they are combined here as "available-P". Total sedimentary phosphorus was determined on oven-dried sediments following combustion at 550°C (Anderson 1976). All phosphorus analyses were completed using the heteropoly blue colorimetric technique (Strickland and Parsons 1968). The difference between total sediment P and the sum of the inorganic fractions was assumed to be organically bound P.

Concentrations (mass × mass sediment^{-1}) of pigments, metals and nutrients were converted to flux rates (mass cm^{-2} sediment surface year^{-1}) using sediment accumulation rates derived from ^{210}Pb dates.

Results

Total ^{210}Pb activity declines from the surface of the core to a background (supported ^{210}Pb) below 29 cm. The break between sediments containing unsupported ^{210}Pb and the older sediments is very clear (Fig. 2). Dates calculated according to the constant rate of supply (CRS) model have an uncertainty of less than ±5 year for the last century and ±15 year for the oldest modeled date of 1852. This analysis indicated sedimentation rates of about 0.014 g cm^{-2} year^{-1} in the middle of the

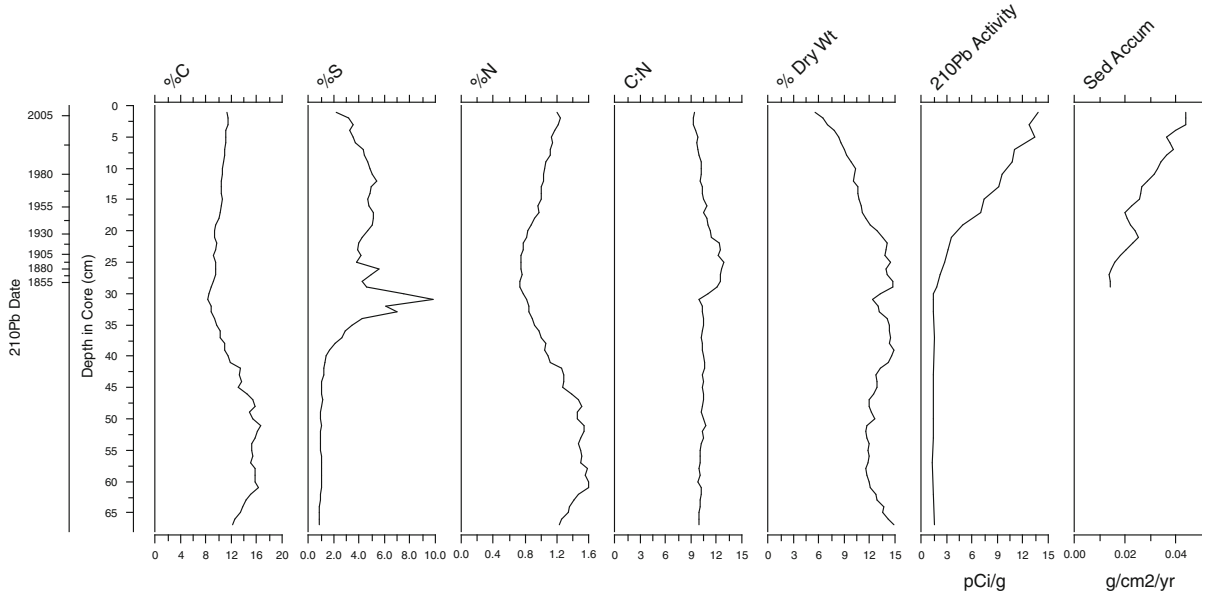

Fig. 2 Bulk (%) organic carbon, sulfur, and nitrogen, C:N weight ratio, and dry mass for the Sandy Lake core. Sediment accumulation rates (g cm^{-2} year^{-1}) were determined from ^{210}Pb activity in the core. The oldest date determined was 1852 (±15.53 year) at the 28/29 cm interval

nineteenth century increasing to approximately 0.045 g cm^{-2} $year^{-1}$ at present. The increase in sedimentation rate was nearly monotonic except for a brief decrease between 16 and 20 cm (1930–1950) that might be a result of a decline in agricultural activity in the mid-twentieth century. Sediment accumulation rates increase again after 1950 possibly due to increasing recreation, increasingly mechanized agriculture, and the use of synthetic fertilizers, all of which contribute to eutrophication and increased suspended sediment loads in streams.

The carbon and nitrogen contents of the sediment tracked each other fairly closely ($r = 0.97$, Fig. 2). Both increased over the interval 66/67–60/61 cm (from 12 and 1.2%, respectively), remained relatively constant at about 15 and 1.5%, respectively, until both began to decrease from about 45 cm to lows at about 30 cm. From about 30 cm both C and N gradually increased toward the surface from 8 to 0.8%, respectively, to about 11 and 1.2%. These fluctuations in %C were closely matched with changes in % dry mass–increasing organic matter correlated with decreased % dry matter and vice versa. The notable exception was at the 30-cm depth where both %C and % dry mass reached (local) minima.

In spite of the pre-settlement and pre-mine variations in %C and %N, the C:N ratio (Fig. 2) from core depths between 66/67 and 30/31 cm was remarkably constant (mean 10.3, SE 0.04). From a core depth of 30/31 cm the C:N increased to a maximum of 12.9 at 24/25 cm, then declined almost monotonically to 9.4 at the surface.

The sulfur content of the sediment was constant and low (approximately 1%) from 66/67 cm up to about 39/40 cm where it rapidly increased to a maximum of almost 10% at 30/31 cm (Fig. 2). Concentrations rapidly fell back to about 4–5% around 27/28 cm and remained at these concentrations until 11/12 cm when they gradually decreased further to 2–3% at the surface.

All metal (Fe, Mn, Al, Zn, Hg, Pb) concentrations were historically low (deeper than 35–40 cm) and relatively stable (Fig. 3). Means (±standard errors) between 40 and 67 cm were 30.4 (±0.6) mg g^{-1} sediment dry weight for iron, 0.56 (±0.01) mg g^{-1} for manganese, 21.8 (±0.4) mg g^{-1} for aluminum, 0.11 (±0.002) mg g^{-1} for zinc, 0.049 (±0.005) mg g^{-1} for lead, and 119(±1.5) ηg g^{-1} for mercury. Iron, manganese and aluminum concentrations increased reaching maxima between 25 and 30 cm

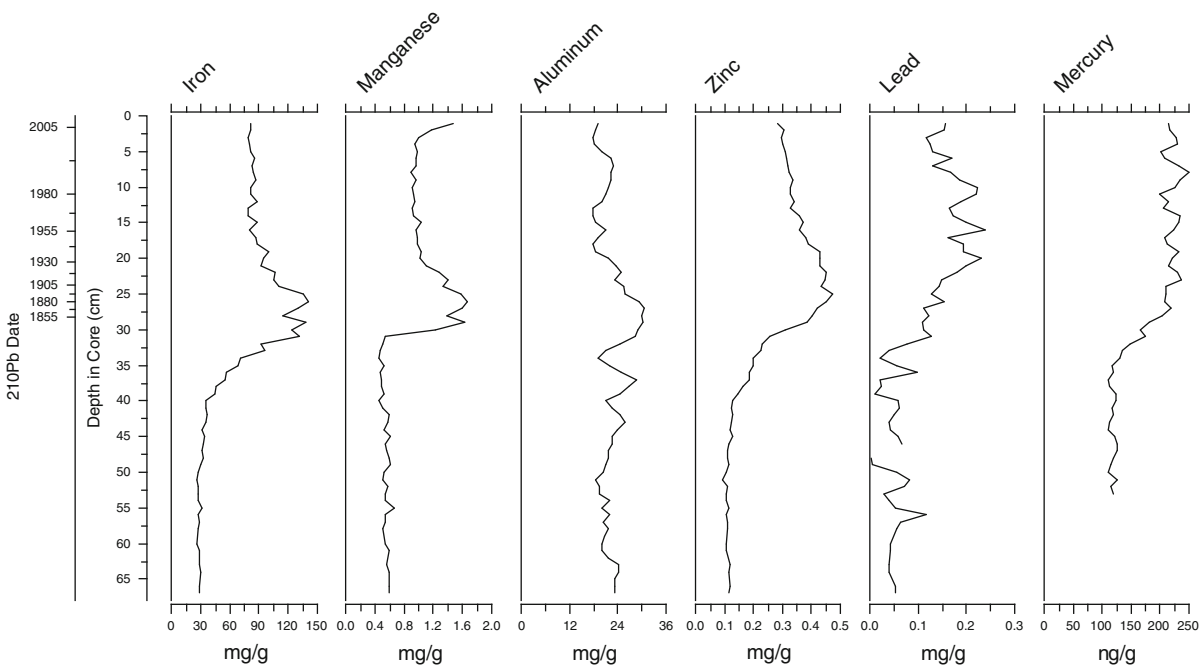

Fig. 3 Distribution of iron, manganese aluminum, zinc, and lead (mg g^{-1}) and mercury (ηg g^{-1}) in the Sandy Lake core

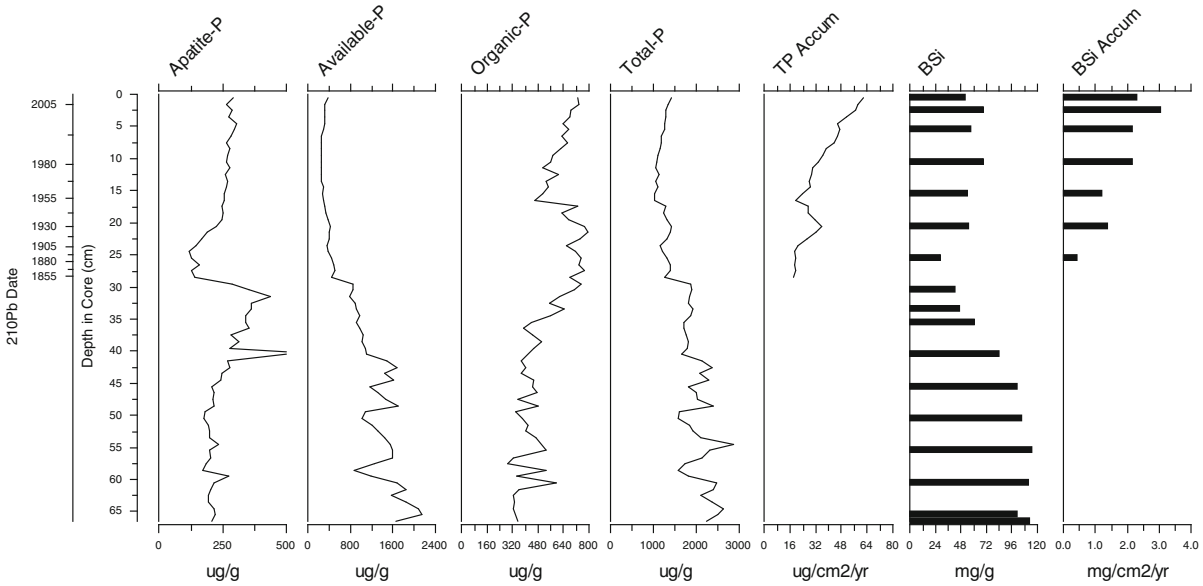

Fig. 4 Distribution of phosphorus species and biogenic silica in the Sandy Lake core. Apatite- and available-P are the HCl-extractable and the NH_4OH + NaOH extractable-P fractions, respectively. Organic-P is the difference between total-P and the sum of apatite- and available-P. Accumulation rates were calculated as the product of P or Si concentration (mg g^{-1}) and sediment accumulation rate (g cm^{-2} $year^{-1}$) calculated from ^{210}Pb dates

of 140 mg g^{-1} for iron, 1.7 mg g^{-1} for manganese and 30.5 mg g^{-1} for aluminum. Iron, manganese and aluminum are metals commonly associated with coal mining (Rose and Cravotta 1998), and following these large increases, concentrations decreased between 25 and 20 cm, although Fe and Mn concentrations remained relatively constant at 2–4 times historical concentrations through to the present. In contrast, mercury and lead, which can be released from mining operations but also have a significant atmospheric component and can enter lakes and their watersheds from distant sources, did not show the same recent (post-25 cm) decline, and concentrations remained elevated at about 2.5 times historic concentrations. Mercury, for example, increased from its historic low concentration and has remained at 200–250 ηg g^{-1} through to the present. Zinc was intermediate in its response, and showed an increase between 25 and 30 cm to a maximum concentration of almost 0.5 mg Zn g^{-1} from its mean historic concentration of 0.1 mg g^{-1}. Zinc concentrations declined gradually from its peak to a current concentration of about 0.3 mg g^{-1}.

In the interval between 40 and 67 cm in the core total-P fluctuated erratically (Fig. 4); however, these fluctuations were closely mirrored in the profile of available-P which was measured independently. Consequently, we interpret the period bounded by these sediment depths to be one of marked changes in phosphorus loading, phosphorus sedimentation and/or phosphorus retention and burial. During this same interval organic-P showed much less variability. Between 29 and 40 cm in the core the inorganic fractions and total-P stabilize as organic-P increases from approximately 400 to almost 800 μg g^{-1} sediment dry weight. At 29 cm there was an abrupt decrease in the inorganic fractions and total-P, but no change in organic-P and between 16 and 20 cm there was a decrease in organic- and total-P. Available- and apatite-P showed little further change to the present, while increasing organic-P was responsible for a gradual increase in total-P toward the surface of the core. Total-P accumulation rates generally increased over the dated section of the core with the exception of an interval between 16 and 20 cm. Overall, accumulation rates have increased from <20 μg cm^{-2} $year^{-1}$ at 28/29 cm to >60 μg cm^{-2} $year^{-1}$ at present.

Biogenic silica concentrations were highest deep in the core (Fig. 4) and decreased to a minimum between 25 and 30 cm. Concentrations were stable at approximately 60 mg g^{-1} sediment dry weight from

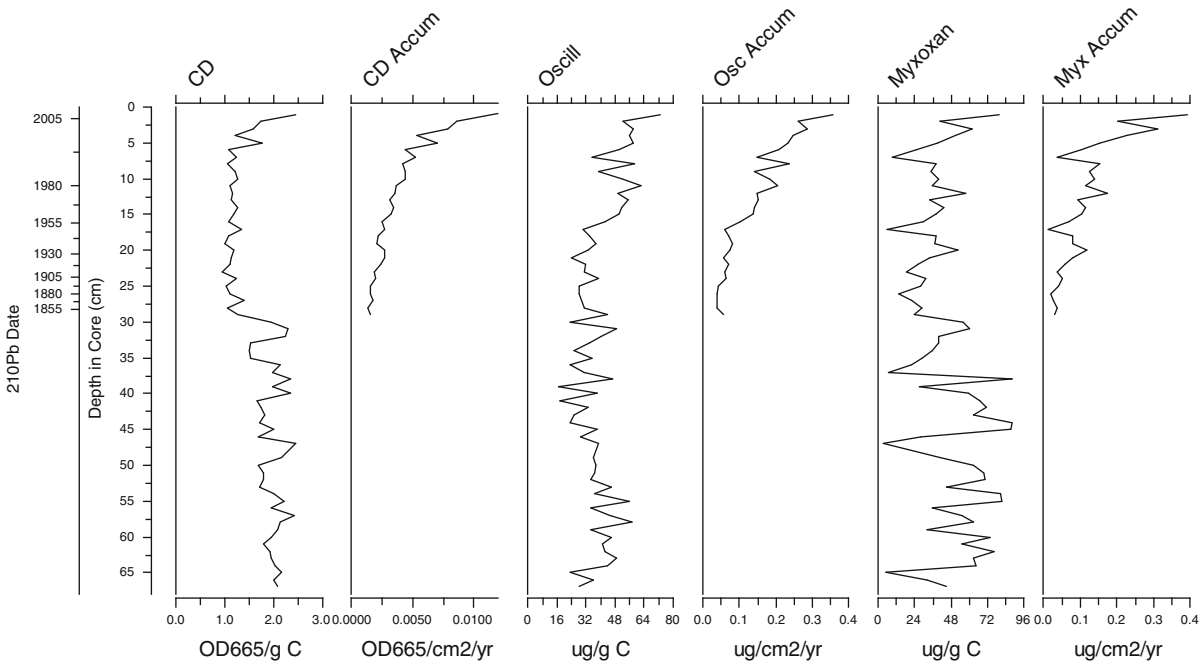

Fig. 5 Distribution and accumulation rates of chlorophyll derivatives, oscilloxanthin, and myxoxanthophyll in the Sandy Lake core

a depth of 20 cm to the surface. Biogenic silica accumulation rates showed a continuous increase over the dated section of the core from a low of <0.5 mg cm^{-2} $year^{-1}$ at 25/26 cm to approximately 2.5 mg cm^{-2} $year^{-1}$ at the surface.

The method used to determine chlorophyll derivatives (CD) gives results in OD_{665} g C^{-1}, a rather unconventional unit. However, OD_{665} is assumed proportional to chlorophyll mass in a number of trichromatic equations used to quantify the concentration of chlorophyll in lake water samples (e.g., Strickland and Parsons 1968) and we used OD_{665} as an analog measure of chlorophyll mass to present both concentrations and accumulation rates of CD. Chlorophyll derivatives (CD) varied between 1.6 and 2.4 OD_{665} g C^{-1} in the older portion of the core deeper than 30 cm with no apparent trend (Fig. 5). At 30 cm CD abruptly decreased to 1.0–1.2 OD_{665} g C^{-1}, and only began to increase again above 4 cm. CD accumulation rates (OD_{665} cm^{-2} $year^{-1}$) indicate a gradual increase over the dated portion of the core from 0.015 OD_{665} cm^{-2} $year^{-1}$ at 28/29 cm to 0.040 OD_{665} cm^{-2} $year^{-1}$ at 5/6 cm. There is a rapid increase in the most recent sediment to 0.15 OD_{665} cm^{-2} $year^{-1}$ at the surface. Oscilloxanthin, in contrast is lowest in the deeper sections of the core varying between 20 and 50 μg g C^{-1} below 18 cm. Concentrations increase between 18 cm and the surface from about 35–75 μg g C^{-1}. Accumulation rates of oscilloxanthin are relatively constant at about 0.06 μg cm^{-2} $year^{-1}$ between 16 and 29 cm, then increase from 16 cm to 0.36 μg cm^{-2} $year^{-1}$ at the surface. Myxoxanthophyll concentrations were highly variable showing no evident trend, although calculated accumulation rates show a constant increase from 29 cm (<0.05 μg cm^{-2} $year^{-1}$) to 8 cm (0.15 μg cm^{-2} $year^{-1}$). Accumulation rates increase from 8 cm to the surface of the core (0.39 μg cm^{-2} $year^{-1}$).

We identified 53 diatom taxa from the 13 core samples examined. The most commonly occurring diatoms included species of *Aulacoseira* (*ambigua* (Grunow) Simonsen, *granulata* (Ehrenberg) Simonsen), *Cyclotella* (*comta* (Ehr.) Kütz., *stelligera* Cleve & Grunow (=*Discotella stelligera* (Cleve & Grunow) Houk & Klee)), *Tabellaria* (*fenestrata* (Lyngb.), *flocculosa*(Roth) Kütz., *quadrisepta* Knuds.), and *Fragilaria construens* (Ehr.) Grun. (=*Staurosira construens* Ehrenberg)). The *Aulacoseira* species and *T. fenestrata* were most dominant in the deeper samples whereas the *Cyclotella* species, *T. flocculosa* and *F. construens* increased in importance in the more recent cores sections. Several other species

were common only a few depths, notably *Eunotia exigua* (Bréb. *ex* Kütz.) and *Frustulia rhomboides* (Ehr.) which are indicative of more acidic waters in both Europe and North America (Camburn and Charles 2000; Kwandrans 1993), with reported pH optima of 5.9 and 6.2, respectively (Dixit et al. 1999). Diatom-inferred pH indicated that the lake water was historically circumneutral up until 30 cm. In the two samples dominated by the acidophiles between 28 and 30 cm the DI-pH dropped by almost a full pH unit to 6.4. Above 28 cm the pH quickly returned to historical levels to the present. Mean diatom-inferred P from core samples deeper than 30 cm was 20 µg P l^{-1}. Post-Sawmill Run diversion DI-P was relatively stable; DI-P of the 7 post-diversion diatom assemblages gave a mean of 15 µg P l^{-1}. The stratigraphy of the common diatom species and diatom-inferred pH and P are shown in Fig. 6.

Discussion

In describing the history of Sandy Lake from sediment proxies we considered samples from between 35 and 67 cm to represent the pre-settlement period. It was not possible to assign dates to this interval; however, assuming that sediment accumulation rates calculated from the oldest ^{210}Pb dated section of the core represented a maximum pre-settlement rate then the interval is bounded by approximately 1400 and 1770 C.E. Sediments examined from this interval are characterized by relatively higher carbon and nitrogen content, higher dry weight, relatively higher concentrations of total P largely because of high concentrations of apatite-P and available-P, relatively low C:N ratio, and low concentrations of sulfur and all metals. Organic P concentrations were relatively low. Indicators of phytoplankton biomass (BSi, CD) were relatively high although specific cyanobacterial pigments were low. Actual concentrations are misleading, however, and without reliable dates for this period accumulation rates cannot be calculated. The high concentrations of BSi and CD are most likely a result of low accumulation rates that are undiluted by allochthonous material from an undisturbed watershed. Dominant diatoms from this interval included *Aulacoseira ambigua*, *A. granulata*, *Tabellaria fenestrata* and *T. quadrisepta*. All of these species are indicative of oligo-mesotrophic waters (Reavie et al. 1995; Dixit et al. 1999; Reavie and Smol 2001) although the *Aulacoseira* species have also indicated mesotrophy (Ramstack et al. 2003). We interpret these data to suggest that pre-settlement Sandy Lake was oligo-mesotrophic with an undisturbed watershed that contributed little allochthonous sediment. The sediment organic matter is dominated by planktogenic material based on its C:N ratio of 10, indicative of algal rather than terrigenous origin (Meyers 1994). The relatively low concentrations of organic-P suggest a well oxygenated hypolimnion that would support rapid breakdown of settling and recently settled seston. The ratio of BSi to the cyanobacterial pigments oscilloxanthin and myxoxanthophyll suggest phytoplankton assemblages dominated by diatoms, mostly species with relatively low P optima.

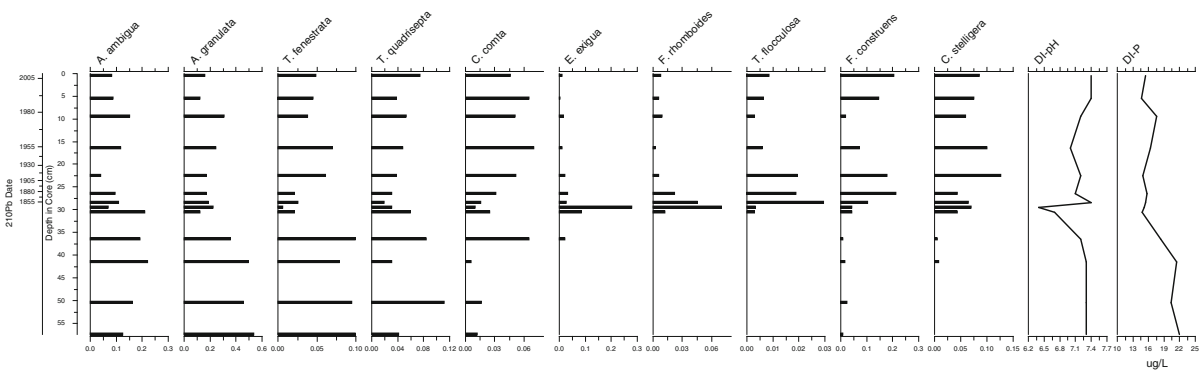

Fig. 6 Relative abundance of the most common diatoms (*Aulacoseira ambigua, A. granulata, Tabellaria fenestrata, T. quadrisepta, Cyclotella compta, Eunotia exigua, Frustulia rhombiodes, T. flocculosa, Fragilaria construens,* and *C. stelligera*) identified in the Sandy Lake core, and diatom-inferred pH and P

The largest changes in the sedimentary profile are centered around 30 cm which appears to slightly predate the opening of the coal mining era although there is some uncertainty in the early ^{210}Pb dating (28/29 cm = 1852 ± 15.5 year.) and bioturbation, resuspension, and diffusion can blur the onset of changes in the sedimentary record. Coincident at 30 cm are minima, or local minima, of C, N, %dry weight, apatite-P, TP, BSi, CD, and diatom-inferred pH, as well as maxima, or increases, in S, C:N, Fe, Mn, Zn, Pb, Hg, and organic P. Increases in S, Fe and Mn in particular are consistent with the lake receiving acid mine drainage (Rose and Cravotta 1998). Other metals examined (Pb, Hg, and Zn) showing increases in the sediment profile approximately coincident with the spikes in Fe, Mn, and S may be associated with mining activity, but they have also been widely used to mark the onset of the Industrial Revolution due to the widespread dispersal and ultimate precipitation of volatile chemical species (Galloway and Likens 1979). The observation that Fe, Mn, and S concentrations all decline simultaneously consistent with the time of the Sawmill Run diversion but that Hg does not decline at all up to the present, that Pb declines only above 9.5 cm (early 1980s) consistent with the removal of lead from automotive fuel, and that Zn concentrations decline only gradually, all suggest that that the source of these metals is primarily atmospheric rather than local mine effluent.

The atmospheric flux of Pb and Zn to lake sediments is proportional to lake area (Dillon and Evans 1982, Blais and Kalff 1993) and mean Sandy Lake pre-settlement lead concentration of 47 (±1.9) μg g^{-1} is similar to historic concentrations reported from Lake Michigan (Edgington and Robbins 1976), from eight southern Ontario lakes (Dillon and Evans 1982), and from seven lakes in Connecticut (Siver and Wozniak 2001). Sandy Lake pre-settlement mean zinc concentration of 110 (±1.7) μg g^{-1} is similar to the historic Zn concentrations from ten southern Ontario lakes (Evans, et al. 1983). Recent sediment lead and zinc concentrations among lakes are more variable due to differences in local industrial and municipal activities (Callender and Rice 2000). In contrast to lead and zinc, mercury loading to lakes has been shown to be proportional to watershed area (Swain et al. 1992). Mercury concentrations in Sandy Lake sediments are similar to concentrations measured elsewhere in the northeastern US (Kamman et al. 2005) and although some lake sediment profiles have shown a recent decrease in Hg flux rates (Engstrom and Swain 1997), there has been no apparent decrease in Sandy Lake.

During this interval of elevated metal concentration in the sediments, diatom-inferred pH showed a decrease of one pH unit, due principally to the abundance of the acidophiles *Eunotia exigua* and *Frustulia rhomboides*, which approached 30 and 7% of the individuals identified, respectively. This interval also marked slight declines in the abundance of *A. ambigua* and *A. granulata*, and increases in *C. stelligera, T. flocculosa*, and *F. construens* which had previously been unimportant or absent in the sedimentary diatom assemblage. These latter species have slightly lower P optima than the earlier assemblage (Reavie et al. 1995; Dixit et al. 1999; Reavie and Smol 2001) and result in a lower DI-P for the more recent (post-diversion) samples.

The relatively modest decrease in pH may be the result of a number of factors, the two most important are in situ alkalinity generation through dissimilatory sulfate reduction in the sediments, and the loading of alkalinity from the unaffected portion of the watershed. Bacterial reduction of 1 eq l^{-1} sulfate generates 1 eq l^{-1} alkalinity, and the factors controlling the rate of S reduction include ready supplies of sulfate, iron, and degradable organic matter, and an anaerobic environment—all of which were present in Sandy Lake. For this alkalinity to be permanent, the sulfide must be held in the reduced form and removed either by the evolution and outgassing of H_2S, or by burial in the sediments as metal sulfides, elemental sulfur, or organic-S (Herlihy et al. 1988). The loss of H_2S was probably unlikely given a 5 m thick oxygenated epilimnion. However, the sediments deposited in the interval between the opening of the mines and the Sawmill Run diversion contain up to 10% S suggesting considerable permanent loss of reduced S. In experimentally acidified Lake 223 in the Experimental Lakes Area between 66 and 81% of the added acid was neutralized by alkalinity generated in this manner (Cook et al. 1986). Using the estimated sediment accumulation rate and the measured sediment sulfur concentration, and assuming that all S entered the lake as sulfate, we estimated a net storage of approximately 1.4 mg cm^{-2} $year^{-1}$ at the coring site. This net storage is equivalent to approximately 3.5 × 10^5 mol H^+ $year^{-1}$ neutralized

across the area of the hypolimnetic sediments. This generated alkalinity does not include any from S reduction that may have occurred in the epilimnetic sediments which might occur at a slower rate, but due to the larger area involved, may make a significant contribution (Kelly and Rudd 1984).

The relative significance of in situ alkalinity generation in Lake 223 is due in part to that lake's setting in poorly buffered geological strata (Cook et al. 1986). Approximately half of the Sandy Lake watershed was unaffected by acid mine drainage, and a survey of 9 unaffected first-order streams within 5 km of the lake gave a mean Ca of 34.6 ± 4.8 mg l^{-1} and a mean Mg of 7.4 ± 1.1 mg l^{-1} (Navarre and Ostrofsky, unpubl. data). Direct groundwater inputs to the lake make an unknown additional contribution. Estimating the watershed contribution to acid neutralization using estimated surface flow alkalinity from the Ca + Mg data, and mean annual runoff gives 4.2 × 10^6 mol H^+ $year^{-1}$ neutralized.

Although the diatom-inferred acidification of Sandy Lake was modest, significant disruption of biotic communities can be expected at pH values around 6.0 (Neary et al. 1990), and the elevated concentrations of metals on the sediment surface would negatively affect zoobenthos even if metal concentrations in the water column remained low due to rapid precipitation. The report of the State Commission of Fisheries (Busch 1897) makes no comment on the status of benthic invertebrates, only documenting the extirpation of fish. Fish populations may be affected directly by low pH and high metal concentrations through direct water-borne or diet-borne exposure, or indirectly through the elimination of pH- or metal-sensitive prey (Schindler et al. 1985; Rasmussen et al. 2008). Our paleolimnological analysis sheds no light on the relative importance of either of these pathways in Sandy Lake.

Chlorophyll derivatives (CD) show a clear decrease at the same time the metals increase, from a pre-settlement mean of about 2.0 OD_{665} g C^{-1} to a mean of 1.1 OD_{665} g C^{-1} above 30 cm. CD concentrations do not recover as metals decrease and as pH increases. Either conditions change decreasing pigment preservation, or more likely, the diversion of almost 50% of the lakes catchment area reduced the lake's nutrient loading supporting a reduced phytoplankton biomass. This latter conclusion is supported by the DI-P data showing a 25% decrease in lake water P concentration. Consistent with this interpretation is the observation of carbon and nitrogen minima and an increase in C:N ratio. Taken together these data suggest an overall decrease in organic matter loading to the lake with a relative increase in allochthonous sources, either from a decrease in settling phytoplankton, or an increase in erosion from the watershed, or both. Detrital apatite-P, largely derived from eroded soils, reached a peak at this time, then abruptly decreased consistent with suddenly decreasing watershed size from 12.9 to 7 km^2 with the Sawmill Run diversion.

At about 28 cm, the concentration of sulfur in the sediment drops from its high of almost 10% to about 4.5%. Iron and manganese concentrations decrease as well, but not as quickly possibly due to migration of reduced ferrous and manganous compounds within the anoxic sediment column. Metal sulfides are considerably less mobile. Concurrent with the decrease in sulfur and metals is the rise in diatom-inferred pH back to pre-settlement levels. These rapid changes are consistent with acid mine drainage being diverted away from the lake, and occur in the sedimentary record at the inferred time of the Sawmill Run diversion. The data suggest a very rapid recovery, most likely a result of the moderately hard water received from the unaffected portion of the watershed, in situ alkalinity generation associated with sulfate reduction and storage of reduced sulfur species, and the short hydraulic retention time of the lake that allowed rapid flushing of the contaminated water mass.

However, occurring almost simultaneously with the acid mine drainage degradation and recovery are the land use changes in the watershed associated with deforestation, developing agriculture, and increasing human populations. At this point ^{210}Pb dates were available and sediment and proxy accumulation rates could be calculated. All indicators other than DI-P suggest gradual eutrophication following the recovery from acid mine drainage. From 28 cm to the present the accumulation rate of carbon increases from about 0.1–0.5 g cm^{-2} $year^{-1}$. The continued decline in C:N ratio suggests that a growing fraction of this sedimented organic matter is of planktonic origin. CD accumulation rate increases by a factor of 10. BSi accumulation, indicative of diatom productivity, increases by a factor of 6, whereas the cyanobacterial pigments increase by a factor of 10. There has been a significant change in the

sedimentary diatom assemblage, from a pre-settlement community dominated by *Aulacoseira ambigua, A. granulata*, and *Tabellaria fenestrata* to a modern community where these former species are still important, but share dominance with *Fragilaria construens* and *Cyclotella stelligera*. Historic and modern pigment ratios suggest that cyanobacteria are increasing relative to diatoms in the lake's phytoplankton community.

The rapidity with which the lake has recovered from acid mine drainage with levels of coal mine-specific proxies (Fe, Mn, S, pH, acidophilic diatoms) all responding rapidly to the Sawmill Run diversion is consistent with other studies where acid and metal loadings have been reduced or eliminated (Fritz and Carlson 1982; Hutchinson and Havas 1986; Keller et al. 1999; Cattaneo et al. 2004). The lake's response to the reduction in catchment area and anthropogenic land use changes is not as clear cut. The DI-P suggests oligotrophication, with a 25% decline in predicted water column P concentration, and modern DI-P is in good agreement with contemporary P concentrations measured chemically. All other proxies (BSi, pigments, C:N, sediment accumulation rate), however, suggest progressive enrichment in spite of increasing forest cover in the watershed over the last century, a nearly static human population around the lake since the late nineteenth century, and a halving of drainage basin area and associated near-complete removal of urban drainage. Although Sandy Lake has not experienced the problems associated with eutrophication evident in many other nearby lakes (low Secchi transparency, high springtime TP, high suspended chlorophyll-α) it is troubling that sediment analyses suggest an approximate 3-times increase in sediment accumulation rate, 10-times increase in chlorophyll derivatives and cyanobacterial-specific pigments, and 6-times increase in BSi since the mid-nineteenth century. Why trophic indicators should all point to increasing eutrophication with the continued increase in watershed forest cover over the last century is not clear. One possibility is that the development of an anoxic hypolimnion allows the more efficient utilization of nutrients through internal loading of previously-sedimented P (Nürnberg and Peters 1984). Another is the observation that while the area of land in agriculture has decreased, agricultural practices have intensified with the extensive use of imported synthetic fertilizers. In any case, Sandy Lake seems to be very resilient with respect to the stressors associated with acid mine drainage, but very much less so with respect to eutrophication.

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