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The Sedimentary Record of Environmental Contamination in Horseshoe Lake, Madison County, Illinois

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ABSTRACT

Industrial development over the last 110 years has contaminated many parts of the American Bottoms, an extensive floodplain of the Mississippi River just east of St. Louis, MO. Water resources in this region have been severely impacted by long-term mismanagement of hazardous waste disposal by local industries. Toxic refuse from metal smelting, steel making, and wood-treatment industries has been released on site to percolate into the ground or to run off into local streams (Colten 1988). A record of metal contamination exists in the sediment of Horseshoe Lake, a natural oxbow lake in the most industrialized portion of the American Bottoms. We examined two dated sediment cores from Horseshoe Lake to reconstruct the historical record of environmental contamination. We used isotopes of nitrogen to track the history of sewage contamination finding that sediment $\delta^{15}N$ increased to values > 10 $^{o}/_{oo}$ in the 1920's. Because such high values of $\delta^{15}N$ are only associated with the presence of human or animal wastes, we deduce that major contamination of the lake by sewage began at that time. Lead, cadmium, and zinc concentrations increased in the sediment after the 1940's. The increase in heavy metals is probably related either to increased input to the lake from local industrial activities or the use of lead shot by local waterfowl hunters. Our results provide a physical record of contamination that is consistent with Colten's (1988) description of hazardous waste disposal in the American Bottoms.

INTRODUCTION

The American Bottoms is a large area of the Mississippi River flood plain in Southwestern Illinois located across the river from the city of St. Louis, MO (Fig. 1). This flood plain contains a number of large, highly industrialized cities including East St. Louis and Granite City, IL. Because of its proximity to transportation, Illinois coal deposits, and the availability of water for industrial processes; the American Bottoms became a prime location for heavy industry during the 19th century (Colten 1988).

An unanticipated consequence of this early industrial development was widespread contamination of land and water by hazardous wastes. Colten (1988) emphasized that; “Factories dumped all manner of liquid waste into water courses for natural purification and dilution treatment while they heaped solids on site. This created numerous waste water sinks including most of the lakes and stream channels on the American Bottoms, where contaminated sediments were allowed to accumulate over the years." The potential for severe contamination of local water bodies from these practices is clear.

The history of heavy industry began in the American Bottoms with the establishment of the St. Louis Stamping Company in Granite City, IL in 1866 (Crayne 1999). The Granite City Steel Company, established in 1895 still occupies the northwest shore of Horseshoe Lake. Two lead smelters, National Lead (NL Industries) and Hoyt Metal were established in 1910. Midland Creosote (later Jennison-Wright) used coal tar from coking to treat railroad ties. Both National Lead and Jennison-Wright are now Superfund sites located in Granite City, IL (Fig 2).

Horseshoe Lake is a shallow (average depth about 2m deep) oxbow located in the American Bottoms adjacent to Granite City, IL (Fig. 1). Ollendorf (1993) cited unpublished radiocarbon dates indicating that the lake is approximately 3,300 years old. The main surface water source for the lake is the Cahokia Drainage Canal, a channelized natural stream. The Canal combines the flow of a series of small streams that drain the loess bluffs to the east of the lake. The Collinsville, IL, water treatment facility is located on Canteen Creek, one of these streams. Horseshoe Lake also receives urban runoff from Granite City through Nameoki ditch, agricultural runoff through Elm Slough, and treated effluent (25mgd) from Granite City Steel (Hill et al. 1981). The water level of the lake is maintained by the Metro East Sanitary District.

Horseshoe Lake is eutrophic. Between 1959 and 1981 there were 14 documented fish kills. The likely cause of these fish kills was deoxygenation under ice during the winter (Hill et al. 1981).

The sediments of lakes record the long-term history of environmental pollution (Pan and Brugam 1997). The sedimentary record of Horseshoe Lake should be a proxy for the contamination of the American Bottoms. There is no other remaining physical record of past contamination in the American Bottoms. The sedimentary record of Horseshoe Lake can be viewed as an archive of environmental contamination and compared with Colten's (1988) compilation of the written historical record.

We used the physical record of contamination stored in the lake sediment to reconstruct the impact of urbanization and industrialization on Horseshoe Lake. Our hypothesis is that the sedimentary record will show increasing levels of contaminants as industrialization increased.

METHODS

Horseshoe Lake was first surveyed to determine the best locations to take sediment cores. Each survey point was located using a Global Positioning System and the depth of the sediment was estimated by probing with steel rods. Core locations (Fig. 2) were chosen based on this survey and work by Ollendorf (1993) who also identified the locations of maximum sedimentation in the lake. Fifteen cores were taken by manually pushing a 7.5 cm wide clear plastic tube into the sediment with vanadium-steel core rods (Wright 1980). Following retrieval, the top and bottom of each tube was sealed with rubber stoppers, held in a vertical position and taken to our laboratory. The cores were extruded vertically and sliced into 1 cm sections that were placed in plastic Whirl-Pak bags. These bags were stored at 4°C until analysis. Although fifteen cores were taken, the two longest cores (H3 and H5; Fig. 2) were chosen for analysis here.

Subsamples of the sediment were processed using acetolysis to reveal pollen grains (Faegri and Iversen 1975). Pollen was counted under a microscope at 40x. Sediment from the time of the first colonization of the lakeshore by European/American farmers was identified by an increase of pollen from agricultural weeds.

The radioisotopes, ^{210}Pb and ^{14}C, were also used to date core H3. Dating of core H5 was accomplished by extrapolating dates from H3 using known changes in pollen in both cores. ^{14}C analysis was done by Geochron Laboratories, Boston, MA. ^{210}Pb dating was performed by the St. Croix Watershed Research Center in Marine-on-St. Croix, MN.

Following drying, samples for stable isotope analysis were ground in a Spex Certiprep® (6750 Freezer/Mill) cryogenic impact grinder cooled by liquid nitrogen. The entire grinding process took about three minutes for each sample. After the samples were ground in the Freezer Mill they were transferred into 2.0 ml microcentrifuge tubes with attached O-Ring Seal Caps. The microcentrifuge tubes were sealed and sent to CoBSIL (Cornell-Boyce Thompson Stable Isotope Laboratory, Ithaca, NY) for isotopic analysis. There was no pre-treatment of samples before analysis.

For phosphorus analysis, the sediment was ashed and leached overnight with hot 6N HCl. The leachate was taken to dryness and redissolved in 0.1 N HCl. Phosphate was determined using the vanadomolybdate yellow technique (American Public Health Association 1985).

Biogenic silica (diatom silica) was analyzed using the technique of DeMaster et al. (1983). The technique differentiates biogenic silica which is amorphous from crystalline forms of silica found in clays and sands.

Samples for metals analysis were prepared by grinding with an alumina mortar and pestle followed by borate fusion. Metals were analyzed using X-ray Fluorescence Analysis at the Department of Earth and Planetary Sciences, Washington University, St. Louis, MO.

Principal Components Analysis (PCA) included all core levels in which we analyzed sediment chemistry. PCA was completed using the computer program, PCord®.

RESULTS

Pollen analysis of the Horseshoe Lake cores can be used to cross correlate dates between cores (Fig 3). Changes in pollen percentages represent changes in watershed vegetation that are contemporaneous between cores. An increase in *Ambrosia spp*. pollen which represents the arrival of European/American farmers in 1808 (Anonymous 1892) occurs at 52 cm in core H3 and 56 cm in core H5. At 33 cm, pollen of Chenopodiaceae increased in both cores. This change is dated at 1920 using ^{210}Pb analysis on core H5. An increase in *Typha latifolia* pollen occurred in 1955 (dated by ^{210}Pb) in both cores.

The percent mineral matter in lake sediment is an indicator of erosion in the lake watershed and is expected to increase because of human disturbance of watershed soils from farming and urban construction. In both Horseshoe Lake cores, percent mineral matter increased from about 83% in 1800 AD to 90% in 1850 (Fig. 3). In the H3 core, percent mineral matter continued at 90% until the present. In the H5, core percent mineral matter was highly variable from 1950 to the present.

Sediment silicon, titanium, and potassium all peaked in the 1890's and declined about 1920 (Fig. 4). In contrast to these elements, calcium increased from 20 mg/gm to 120 mg/gm after 1920 (Fig. 4). Manganese was more concentrated in the sediments of core H3 than in core H5 until around 1920 (Fig. 4). After that time, both cores showed similar concentrations. Magnesium and sodium had constant concentrations throughout both cores.

The cores from Horseshoe Lake have shown little change in phosphorus concentration since 1750 AD (Fig. 5). Biogenic silica concentrations remained nearly constant from 1750 to the 1940's and then declined until the present. There was little variation in percent nitrogen in either sediment core. In contrast δ^{15}N increases from +2 $^{o}/_{oo}$ to greater than +10 $^{o}/_{oo}$ after 1920 (Fig. 5).

Both Horseshoe Lake cores showed large increases in lead, zinc and cadmium in recently deposited sediment (Fig. 5). Peak concentrations of all three metals occurred in sediment dated between 1939 and 1947. Lead and zinc declined slightly in the following year to present values.

We used PCA to estimate the similarity among samples from the two cores. Factor scores from the analysis were used to group samples with similar chemical characteristics (Fig. 6). Three groups of samples emerged. Samples from 1750 to 1889 A.D. formed one group with low concentrations of heavy metals and calcium. Samples from 1889 to 1939 had high concentrations of calcium or silicon, but low concentrations of heavy metals. Samples from 1939 to the present had high concentrations of heavy metals.

DISCUSSION

The increase in *Ambrosia* (ragweed) pollen in Horseshoe Lake (Fig. 3) is present in many sediment cores in the Midwest (Brugam and Speziale 1983; Pan and Brugam 1997). *Ambrosia* is a common agricultural weed inhabiting areas disturbed by human activities that indicates the beginning of European-style agriculture. The ragweed pollen increase at Horseshoe Lake is too old to be dated by ^{210}Pb, but it can be dated to 1808 from historical records (Anonymous 1892). The pollen increase is small because *Ambrosia* was already a significant part of the local flora before European settlement. However, in both cores the *Ambrosia* rise is followed by the appearance of small amounts of *Plantago lanceolata*, a species introduced by Europeans (Brugam 1978). We therefore used the ragweed rise to differentiate between "pre-settlement" and "post-settlement" parts of the core (Fig. 3).

Pollen of Chenopodiaceae and Amaranthaceae cannot be differentiated using normal pollen analytical methods. Many representatives of both plant families are weedy invaders of heavily disturbed areas. The reason these weedy species expanded in the 1920's is not clear, but we suspect an increase in land disturbance due to human activities.

Typha latifolia is an emergent plant of wetlands and shallow lakeshores forming nearly monospecific stands. The 1955 increase in pollen from this species in the sediment suggests a recent expansion of *Typha* swamps around the lake.

PCA is a statistical method for summarizing complex data-sets. Most of the variation in chemical variables from the Horseshoe Lake cores was summarized on the first two axes (Table 1). PCA divided the cores into three sections that generally correspond to periods in the history of industrial development around the lake. Before 1889, human impacts on the lake were minimal. This period (1889-1939) is consistent with the construction of Granite City Steel Company in 1895. The most recent change in lake regime occurred in the 1940's when heavy metals contamination became apparent in the sediment.

Recent paleolimnological investigations have included measurements of stable isotopes of nitrogen in lake sediment that indicate potential sources for nutrient contamination (Brenner et al. 1998). Human and animal waste has a $\delta^{15}N$ more than +10 $^{o}/_{oo}$ (Brenner et al. 1998; Lake et al. 2001; Roadcap et al. 2002). In most lakes, this enrichment in ^{15}N comes from contamination with nitrogen from human sewage rather than from farm animals or agricultural run-off. Thus, the increase of ^{15}N in the 1920's in Horseshoe Lake sediment indicates an increase in disposal of human waste into the lake. McClelland et al. (1997) followed high $\delta^{15}N$ values from sewage contamination of local groundwater (14.9 $^{o}/_{oo}$) into an adjacent bay and ultimately through aquatic food chains. The 1920's increase of $\delta^{15}N$ to 12 $^{o}/_{oo}$ in Horseshoe Lake sediment indicates severe sewage contamination. We do not know the date of construction of the sanitary sewage systems in the lake watershed, but these systems could account for increased inputs of high $\delta^{15}N$ nitrogen compounds into the lake.

The increase in sediment calcium which occurred at the same time as the increase in $\delta^{15}N$ is also evidence for an increase in primary production due to sewage contamination. This increased calcium likely resulted from biogenic deposition of $CaCO_3$, which commonly occurs in hardwater eutrophic lakes (Hodell et al. 1998).

Although the isotopic composition of sediment organic matter clearly indicates increased nutrient loading of Horseshoe Lake by sewage disposal, there is no increase in sediment phosphorus. Eutrophication of lakes usually occurs because of increased phosphorus loading (Schindler 1974) and sewage disposal is a major source of phosphorus for lakes in urban areas (Rechow and Chapra 1983). Some of this phosphorus should be sequestered in the lake sediment by ferric oxides (Engstrom and Wright 1974), but there is no evidence of increased sediment phosphorus resulting from our hypothesized increase in sewage contamination.

Enhanced primary production in a eutrophic lake may also increase biogenic silica in lake sediment because of increasing diatom populations (DeMaster et al. 1983). Like phosphorus, there is no increase in biogenic silica in Horseshoe Lake sediment because of the sewage loading indicated by $\delta^{15}N$ values. Perhaps diatoms that produce biogenic silica are a small part of a phytoplankton community that is dominated by bluegreen algae.

Keating's (1978) study of Linsley Pond, CT, showed that diatom abundance is low in highly eutrophic lakes.

Heavy metal concentrations increased in Horseshoe Lake sediment from the 1940's onward. Von Gunten et al (1997) showed that this kind of increase in sediment heavy metals indicates contamination of the lake by industrial processes. Lead levels in the sediment of Horseshoe Lake have reached 300 ppm - about 10 times their pre-settlement level. Similarly, zinc has increased to about 3,000 ppm. Both of these changes indicate an increase in heavy metal input into the lake. We do not know the sources of these metals. NL Industries ran a lead smelter from the early 20th century to the 1970's that is a potential source of contamination. Granite City, IL, is a large industrial city with many industries that may also have added heavy metals to Horseshoe Lake. The lake has also been managed by the Illinois Department of Natural Resources for waterfowl hunters. Lead shot may be an additional source of lead contamination of sediment, but it would not be a source of zinc and cadmium. Although the source is unknown, our results clearly show contamination of Horseshoe Lake with heavy metals since the 1940's.

CONCLUSIONS

Our study of the sediment of Horseshoe Lake shows clear effects of human disturbance. The lake appears to have become contaminated with sewage in the 1920's. An increase in calcium in the sediment suggests that eutrophication of the lake caused increased biogenic precipitation of calcium carbonate. However, sediment phosphorus and biogenic silica, two other potential indicators of eutrophication did not increase. In the 1940's, Horseshoe Lake became significantly contaminated by heavy metals. Although the section of the American Bottoms that contains Horseshoe Lake has lost much of its industrial base in recent years, Horseshoe Lake still contains significant contamination. Our work is consistent with Colten's (1988) reconstruction of industrial contamination of American Bottoms ecosystems. Thus, Horseshoe Lake contains a physical record of the industrial pollution that Colten described using historical documents.

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Table 1. Eigenvalues for Principal Components Analysis of sediment cores H3 and H5 from Horseshoe Lake, IL. Factor scores for core samples are shown on Figure 6. The analysis was completed using PCord®.

Axis	Eigenvalue	Percent of Variance Explained	Cumulative Percent of Variance Explained
1	16.023	50.071	50.071
2	6.701	20.941	71.011
3	3.684	11.513	82.524
4	1.523	4.758	87.282
5	1.012	3.162	90.444
6	0.542	1.695	92.139
7	0.470	1.470	93.609
8	0.404	1.261	94.870
9	0.326	1.019	95.889
10	0.298	0.932	96.821

Figure 1. Location map of the American Bottoms and Horseshoe Lake. Star indicates location of Horseshoe Lake. Dashed line shows boundary of the American Bottoms.

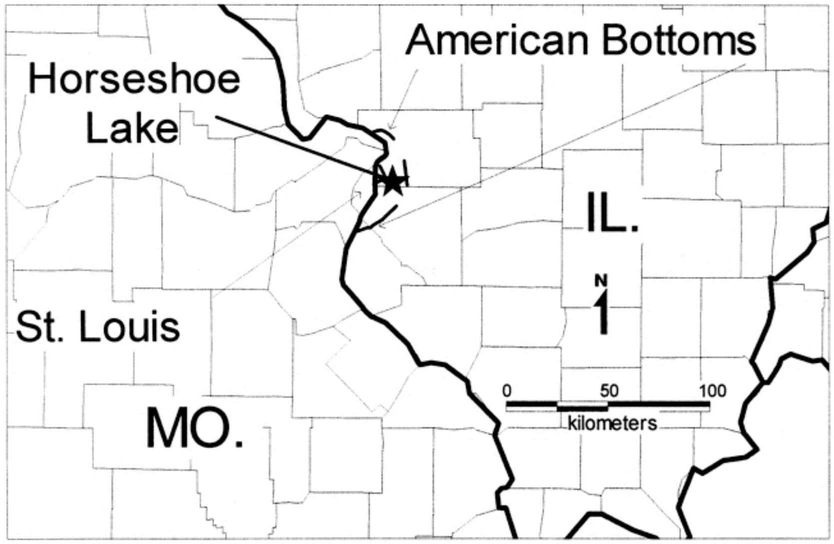

Figure 2. Map of Horseshoe Lake showing locations of NL Industries, Jennison-Wright creosote plant, Granite City Steel sites (circled dots) and sediment core sites (stars). Outline of lake shown in heavier lines.

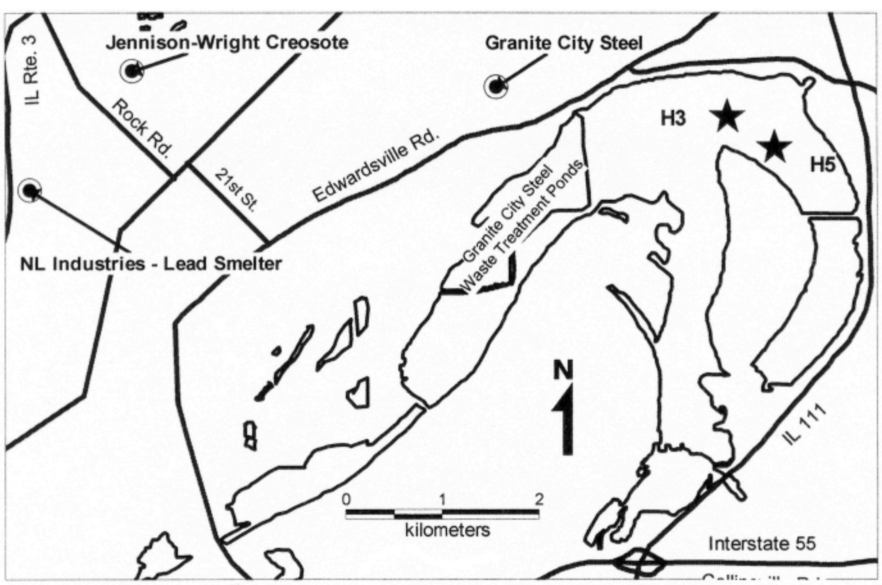

Figure 3. Pollen diagrams from Horseshoe Lake sediment cores. The y-axis of the pollen diagram is core depth in centimeters. The graphed data are percent abundances of pollen species. Solid lines represent a 5x exaggeration of pollen percentages. Dates for the cores are shown to the left of the y-axis in both cores. Dates for core H3 were determined using ^{210}Pb and ^{14}C. The dates for core H5 were extrapolated from core H3 using datable pollen changes.

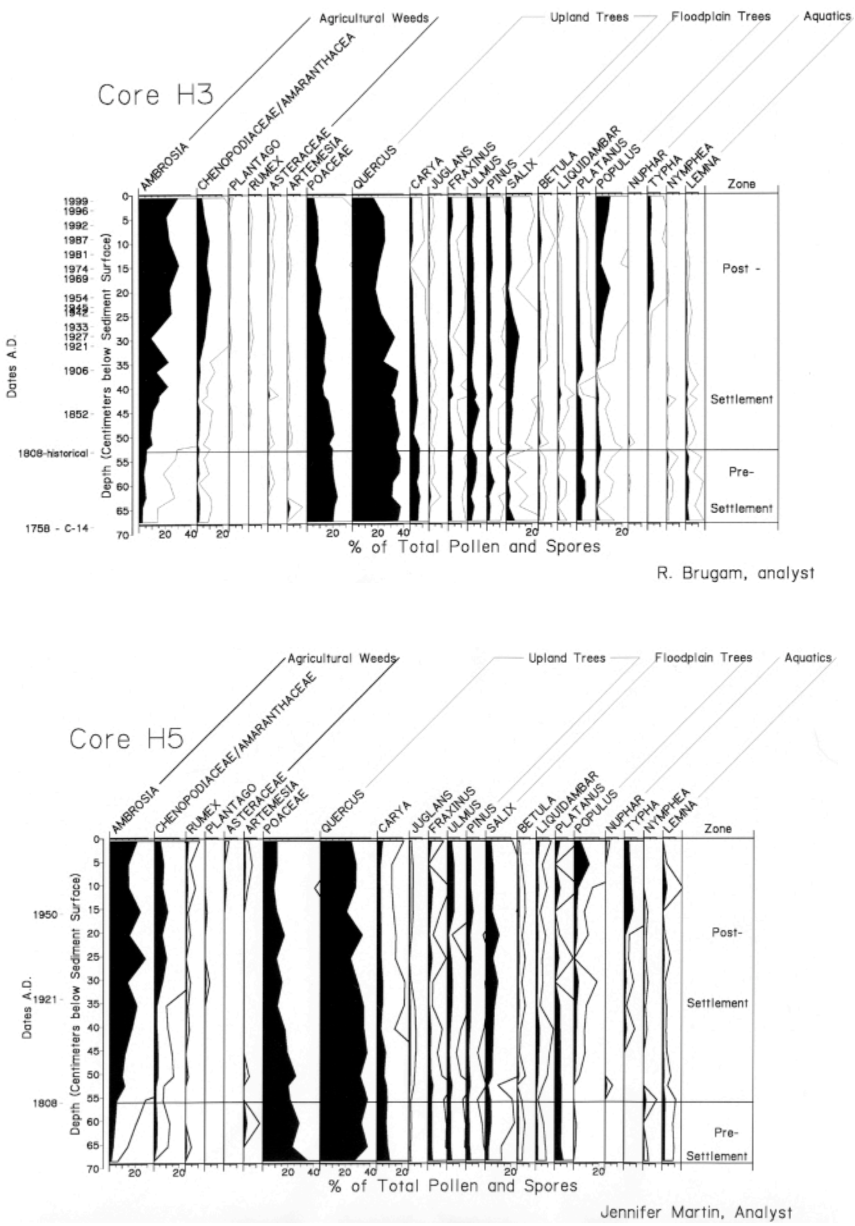

Figure 4. Major elements in Horseshoe Lake sediment cores. H3 (closed circles) and H5 (open circles).

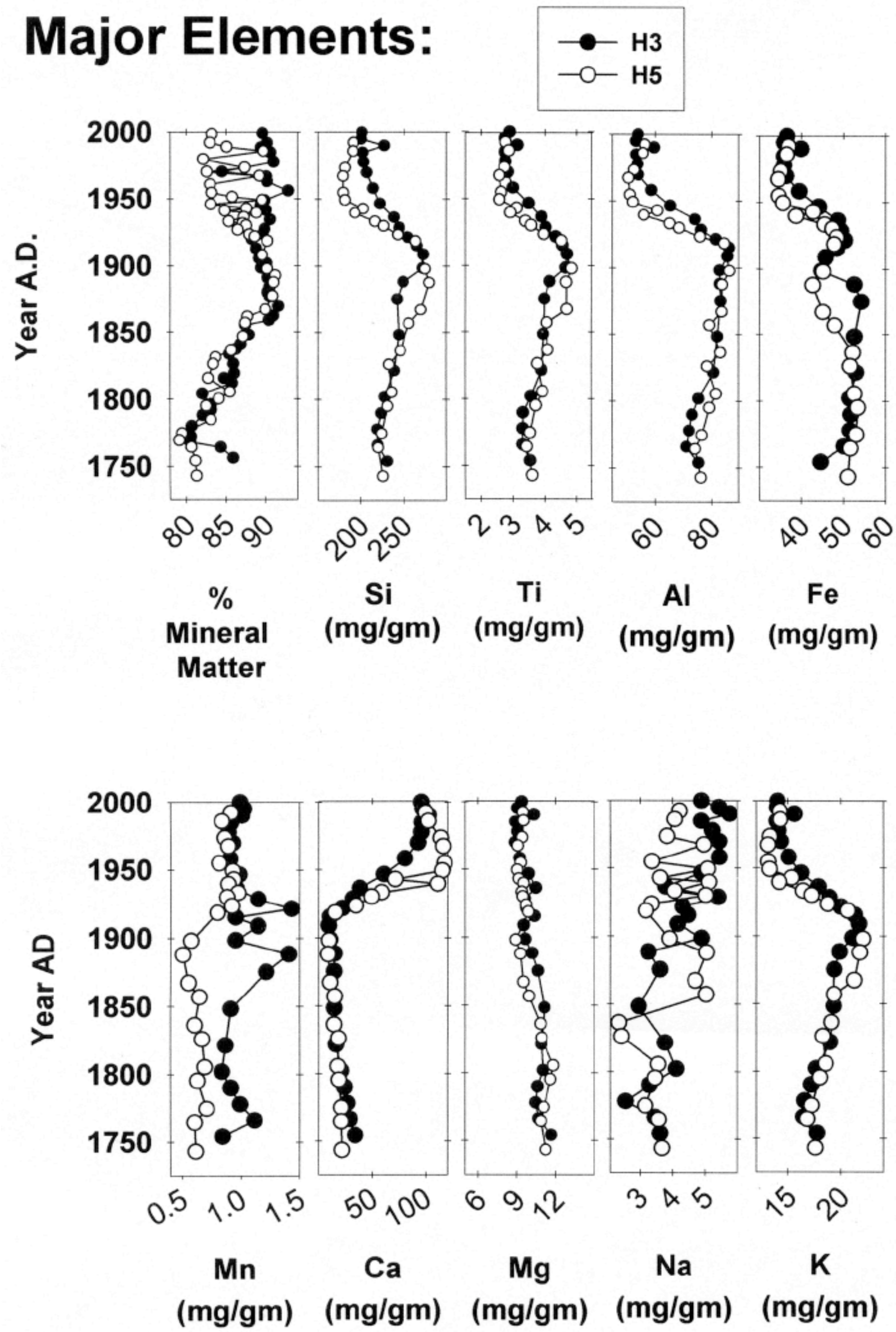

Figure 5. Trophic status indicators from Horseshoe Lake sediment cores (upper panels). Heavy metals in Horseshoe Lake cores (lower panels). H3 (closed circles) and H5 (open circles).

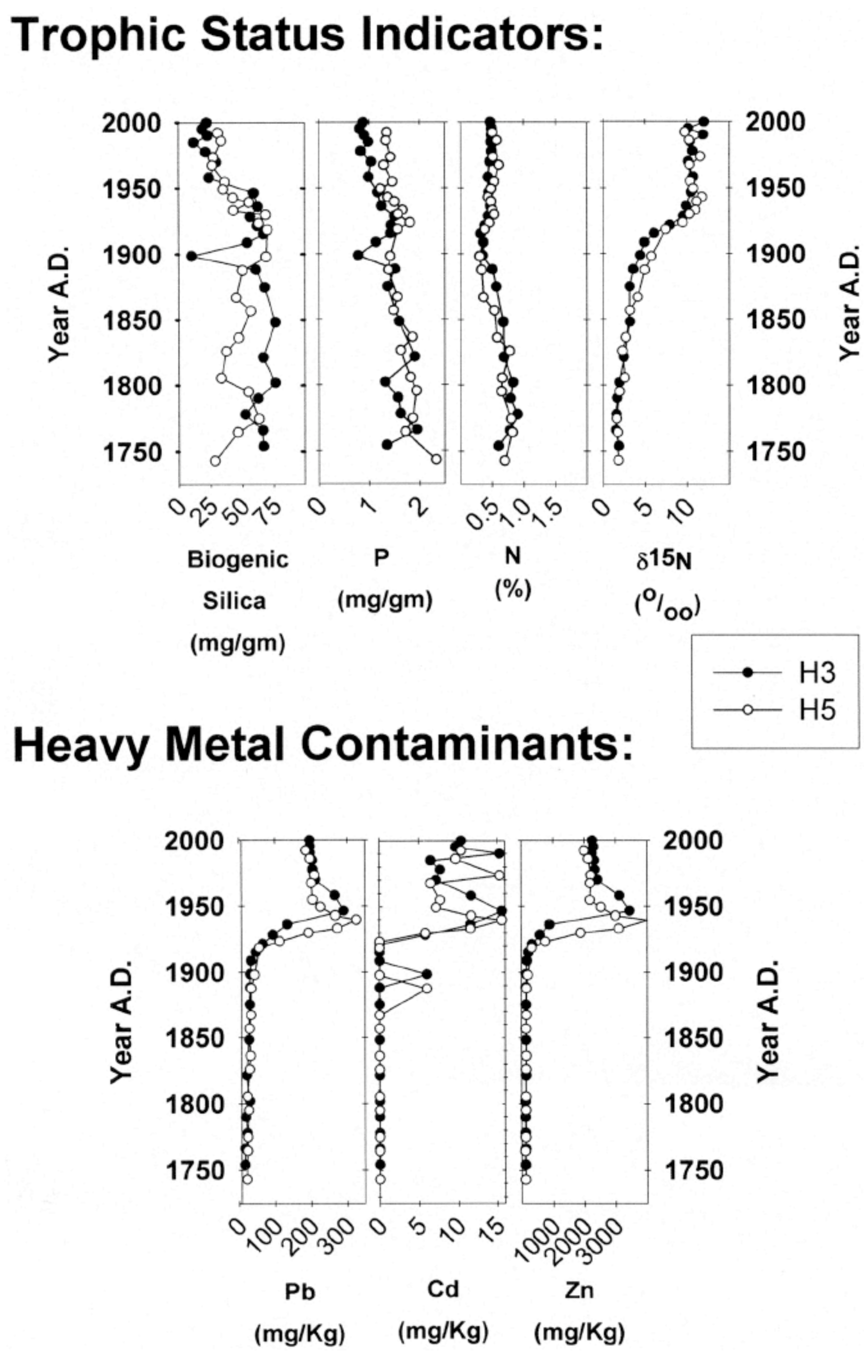

Figure 6. Principal Components Analysis of Horseshoe Lake sediment cores H3 and H5.