Current Research (1993) Newfoundland Department of Mines and Energy, Geological Survey Branch, Report 93-1, pages 419-433

THE EFFECT OF URBAN AND INDUSTRIAL DEVELOPMENT ON THE GEOCHEMISTRY OF THE WATERSHEDS IN THE ST. JOHN'S AREA: PRELIMINARY RESULTS

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ABSTRACT

A geochemical survey of lake sediments and waters conducted during 1990 revealed that urban development and industrialization in St. John's have affected the geochemistry of the drainage network. To define these changes more accurately, lake-sediment cores were collected during the winter of 1992 from four ponds within the city limits and a 'background pond' in a relatively undisturbed but geologically similar area. ^{14}C *dates from basal samples of these cores range from 2260 to 3960 years BP, well before settlement.*

Stratigraphic variations of major and trace elements, pollen, diatoms, soot, charcoal, oil droplets and stable $^{206/207}Pb$ *and* $^{208/207}Pb$ *isotopic ratios are being used to document the chemical, physical, and biological history and, as far as possible, to identify the sources of the contaminants that have been added to the watersheds.*

Both geochemistry and pollen show all cores in the study have been affected to some degree. The background pond shows the smallest changes in both magnitude and thickness of sediment affected, and Quidi Vidi Lake the largest, where Pb values, for example, have increased from approximately 15 to 20 ppm at the base (the natural background level for this lake) to over 600 ppm near the top, an increase of about 30 fold.

Pollen profiles have been established for Quidi Vidi Lake, Long Pond and the background pond and these reflect the farming activities, which commenced in 1770 around Quidi Vidi Lake and in 1820 near Long Pond, by changes in the relative abundances of spruce (Picea) *and grass* (Gramineae) *pollen. Pollen assemblages in sediment from the background pond show little evidence of terrestrial vegetation change.*

Preliminary results suggest that three general periods of widespread anthropogenic change in the St. John's area are recorded in the lake-sediment cores: an initial period of forest clearance and farming, a period of urban development with the common use of coal as fuel for heating and industry, and a later period of urban and suburban expansion and road construction that continues to the present. The use of leaded gasoline has left a clear signal of elevated Pb levels in the uppermost sediment layers in all lakes sampled.

INTRODUCTION

Human activities within a watershed, which can vary from forest clearance to heavy urban development and industrialization, lead to chemical, physical and biological changes. Such activities cause the erosion and transportation of material, some local such as topsoil, and some foreign such as oil and heavy metals, to sites of sedimentation such as lakes. Lake sediments provide a record of man-made changes in their catchments. In urban environments, the anthropogenic effects in lake sediments may extend to depths as a great as metre or more, depending on the sedimentation rate and the period of settlement, necessitating the collection of long lake-sediment cores to obtain the natural background signature.

The lakes and ponds of a city like St. John's with a 400-year history of human influence can be expected to show clear evidence of change. To examine the impact of

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This project is jointly funded by the Newfoundland Department of Mines and Energy and the Canada–Newfoundland Cooperation Agreement on Mineral Development (1990-1994); project carried by Geological Survey Branch, Department of Mines and Energy, Government of Newfoundland and Labrador.

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urbanization and industrialization upon the lakes and ponds in and around St. John's, a local lake-sediment survey was conducted during the fall of 1990 (Christopher, 1991). This study revealed that enrichment in many elements, including heavy metals, has occurred with respect to the surrounding areas. St. John's lies on a lead-enriched shale of the St. John's Group resulting in elevated values of Pb. The Pb values encountered in the city, however, were 10 to 15 times the values in the surrounding areas overlying this Group. Peak Pb values within the city were above the 99th percentile for the whole of Newfoundland. Other elements such as Sb were also locally enriched but, in relation to the whole of Newfoundland, levels were not as high as are found naturally in areas of mineralization (Davenport *et al.*, 1992). The 1991 study did not, however, precisely define a background signature nor did it document the chemical, physical, and biological changes through time.

Macpherson and MacKinnon (1988) collected a core from Kenny's Pond to examine the chemical and pollen profiles. Some of the changes in Kenny's Pond were correlated by Macpherson and MacKinnon (*op. cit.*) with events such as forest clearance, eutrophication, foundries and tinsmithing, and road and hotel construction. The present study takes a broader, multi-disciplinary approach to document the history and, as far as possible, to identify sources of the contaminants within the St. John's area by examining the stratigraphic variation of 49 major and trace elements, pollen and diatom profiles, stable $^{206/207}$Pb and $^{208/207}$Pb isotopic ratios, charcoal, soot, and oil droplets.

The charcoal profiles can be used to identify specific major fires whereas soot counts will provide information about the history of coal burning and automobile use. The pollen profiles provide an effective way to trace vegetation changes due to human activities within both the watershed and the lake itself. For example, by examining changes in relative abundance of *Picea* (spruce) and *Gramineae* (grass) pollen, one can determine approximately when the forest was cleared for farming purposes. The diatom assemblage will help to understand the effect of anthropogenic activities upon the lake water pH through time, which may have important consequences for the mobility of various elements. The stable isotopic ratios of Pb can determine whether increased Pb levels in recent sediments are due to enhanced leaching from soil and bedrock or are from exotic sources. From oil-droplet frequency measurements, the proportion of oil within the sediments can be estimated and their relation to the chemical and physical stratigraphy determined. The detailed chemical profiles for the cores and the various other aspects provide a database for change through time, which can be correlated with the historical records to complete a comprehensive history of anthropogenic effects within the St. John's area. Radiometric dating by ^{137}Cs, ^{210}Pb and ^{14}C will provide corroborative evidence to link the sedimentary and historical records.

METHODS

Sampling

Lake-sediment cores were collected during the winter of 1992 from Quidi Vidi Lake, Mundy Pond, Georges Pond and Long Pond, and a pond in a relatively isolated area along the Witless Bay Line, Long Pond (Figure 1). For the sake of clarity, the Long Pond near the Witless Bay Line will be referred to as 'background pond'. In total, 14 cores were collected, with duplicate cores from six sites, using a modified percussion coring system (Reasoner, 1986). This coring system had an internal diameter of 10 cm, which allowed subsamples to be taken at intervals as small as 2 cm in thickness while still providing enough sediment for the various analyses.

After retrieval, the cores were transported to the cold-storage room in the Centre for Earth Resources Research, Memorial University, where they were extruded from the core barrels and sampled over sections varying in thicknesses from 2 to 5 cm. In most cases, the sampling was carried out at 2 cm intervals for the upper 30 to 50 cm and then 5 cm intervals to the base of the core. At this point, subsamples for geochemistry, pollen, diatoms, water content and loss on ignition (LOI) were taken.

Chemical Analysis

Before analysis, all subsamples were dried and pulverized using an alumina swing-mill. Blind duplicates and standards were inserted to check for analytical precision and accuracy. In total, 49 elements were determined on the dried sediments. Total-element content was measured by instrumental neutron-activation analysis (INAA) for As, Au, Ba, Br, Ce, Co, Cs, Eu, Fe, Hf, La, Mo, Na, Ni, Rb, Sb, Sc, Se, Sm, Ta, Tb, Th, U, W and Yb, and by inductively coupled emission spectrometer (ICP-ES) following an HF-$HClO_4$-HCl digestion for Al, Ba, Be, Bi, Ca, Cd, Ce, Co, Cr, Cu, Dy, Fe, Ga, K, La, Li, Mg, Mn, Na, Nb, Ni, P, Pb, Rb, Sc, Sr, Th, Ti, V, Y, Zn and Zr. Acid-extractable concentrations were determined by atomic absorption spectrophotometry (AAS) of Cd, Co, Cu, Fe, Mn, Ni, Pb, and Zn following a 4M HNO_3−1M HCl digestion, of Ag following digestion in concentrated HNO_3, and of Mo following digestion in aqua regia (Wagenbauer *et al.*, 1983).

Pb Isotopes

Stable $^{206/207}$Pb and $^{208/207}$Pb isotope ratios were determined by inductively-coupled-mass-spectrometer (ICP-MS) following the digestion of ashed sediment in 8N HNO_3, HF, and H_3BO_3 (5000 ppm B) (Longerich *et al.*, 1987).

Pollen, Diatoms, Charcoal, Soot, and Oil droplets

Samples for pollen analysis from Quidi Vidi Lake, Long Pond and 'background pond' and for diatoms from Quidi Vidi Lake were processed and mounted on slides, which were

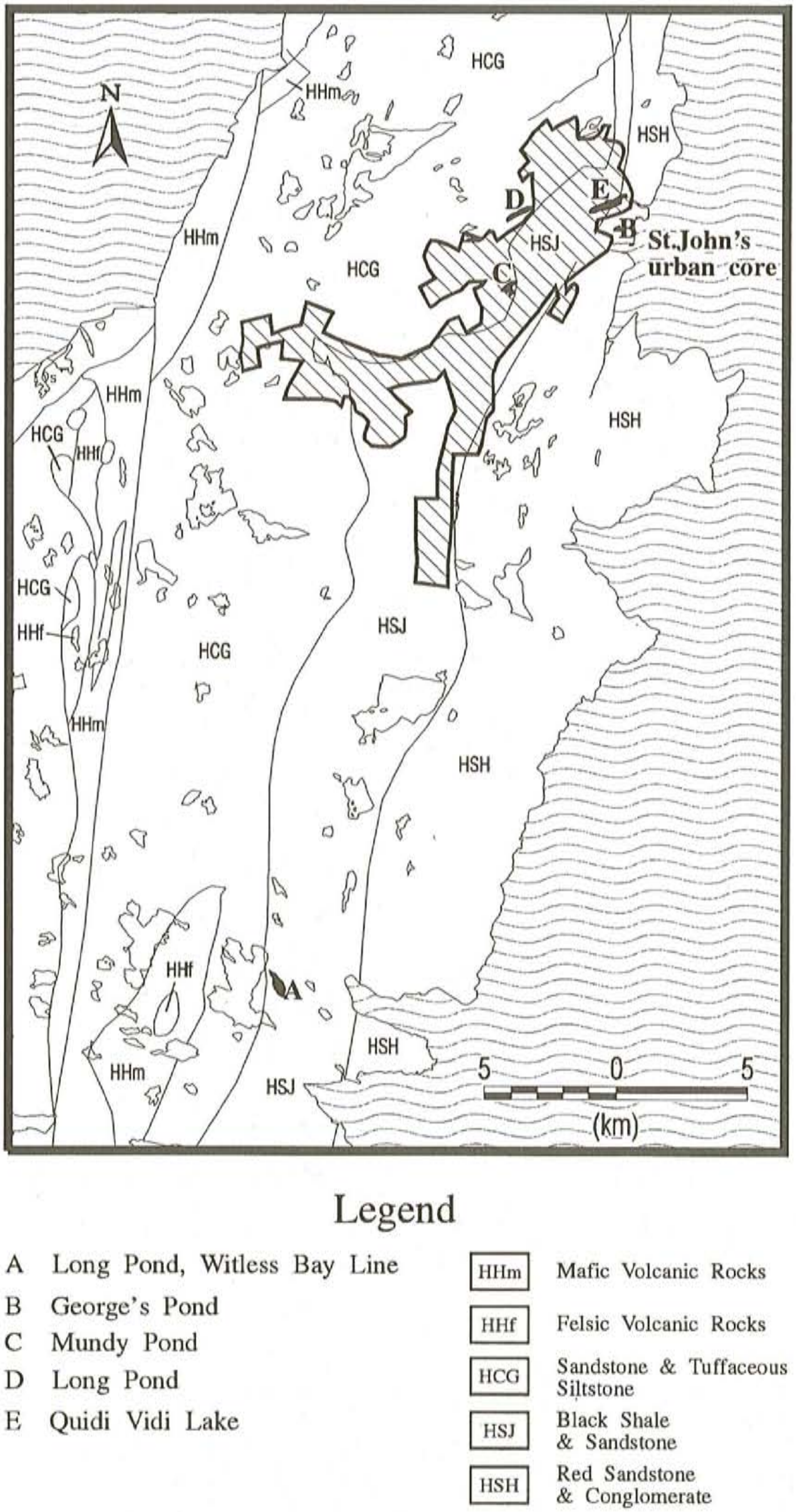

Figure 1. *Location map showing the lakes and ponds cored in relation to regional geology (from King, 1988).*

counted under a microscope at magnifications varying from x400 to x1000 using standard techniques. The pollen slides also allowed examination and counts of charcoal, soot, and oil droplets. Charcoal and soot counts were completed with the pollen counts. The oil droplet count was carried out afterwards at a magnification of x1000. The oil droplets varied in size and thus were counted into size categories of 5 μm increments (i.e., 5-10, 10-15, 15-20 μm, etc.).

RESULTS

Long Pond (Background Pond)

Forested on all sides, Long Pond near Bay Bulls on Witless Bay Line (Figure 1) was chosen as the background pond because of its relatively undisturbed and geologically similar setting. A small paved road to the north is separated from the pond by approximately 100 m of forest. A few small cabins on cleared lots along the edge of the pond constitute the most obvious human disturbance. Water depth at the sample site was 4.58 m, and three stratigraphic units were recognized in the core based on the colour of the wet sediment: Unit 1, from the bottom of the core at 135 cm to 89 cm, is a light-brown gyttja; Unit 2, from 89 to 85 cm, is a black gyttja, and Unit 3, from 85 cm to the top of the core, is a brown gyttja. Layers of plant fragments occur at 46, 62, 78 and 83 cm.

Sediment geochemistry (Figure 2) shows rather minor variations throughout the core, consistent with minimal anthropogenic activity. The only chemical disturbance occurred within the upper 12 cm for the elements K, Mg, Na, Pb and Sb, with Pb (total) showing the largest jump increasing from 17 ppm at 12 cm to 34 ppm at the top (Figure 2). One notable irregularity is recorded at 30 cm depth, where LOI rises abruptly to 46 percent (compared with a range of 27 percent to 34 percent for the whole core) and the concentrations for Al, Ba, Be, Ca, Ga, K, La, Mg, Na, Pb, P, Sr, Ti, V and Zn show corresponding decreases. These chemical changes coincide with the sharp increase in *Isoetes* pollen (an aquatic plant pollen) and the counts of pollen and spores per cc (Figure 3). Counts for the terrestrial pollen species remain constant throughout the

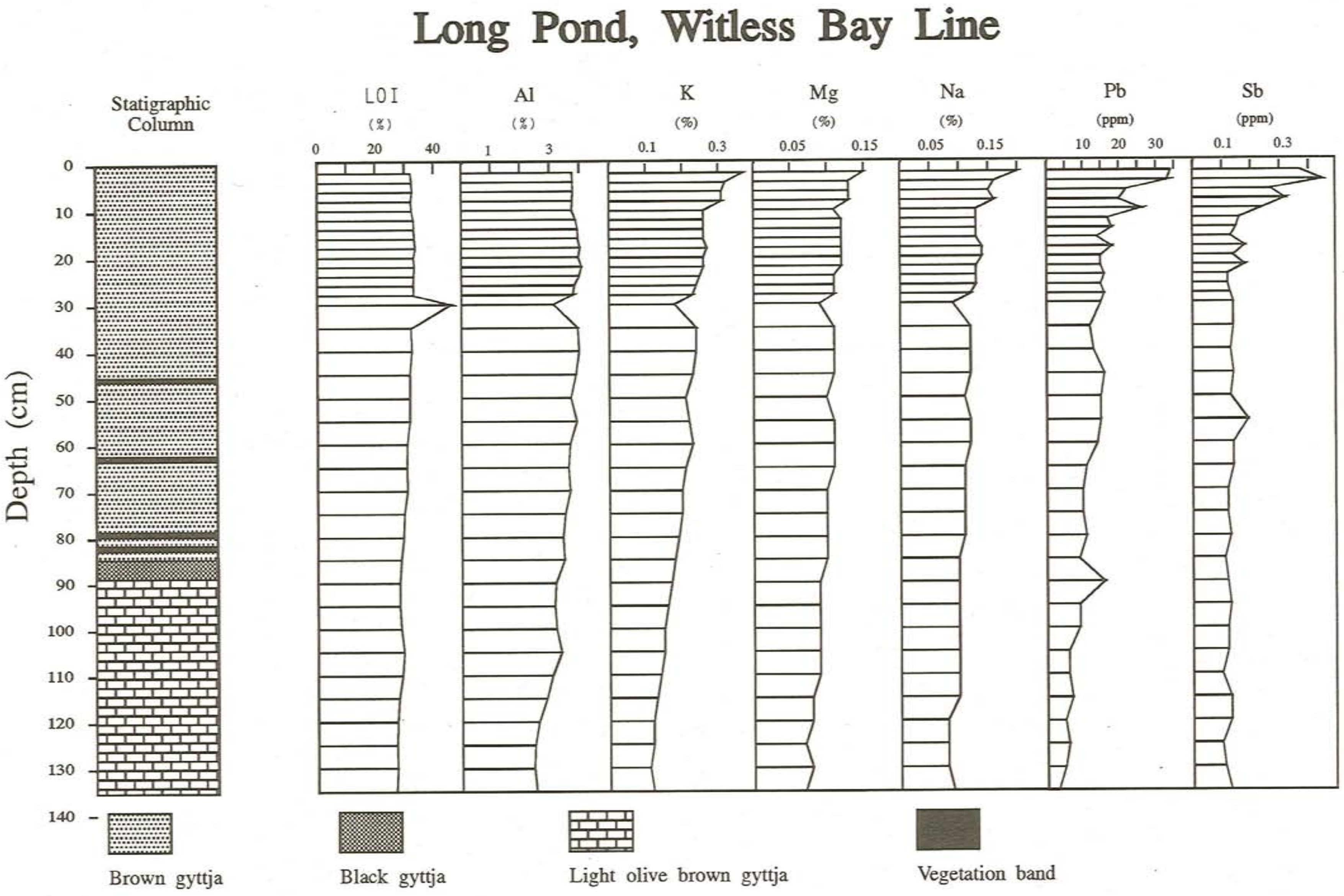

Figure 2. *Stratigraphy and geochemical profiles for LOI, Al, K, Mg, Na, Pb and Sb from Long Pond, Witless Bay Line.*

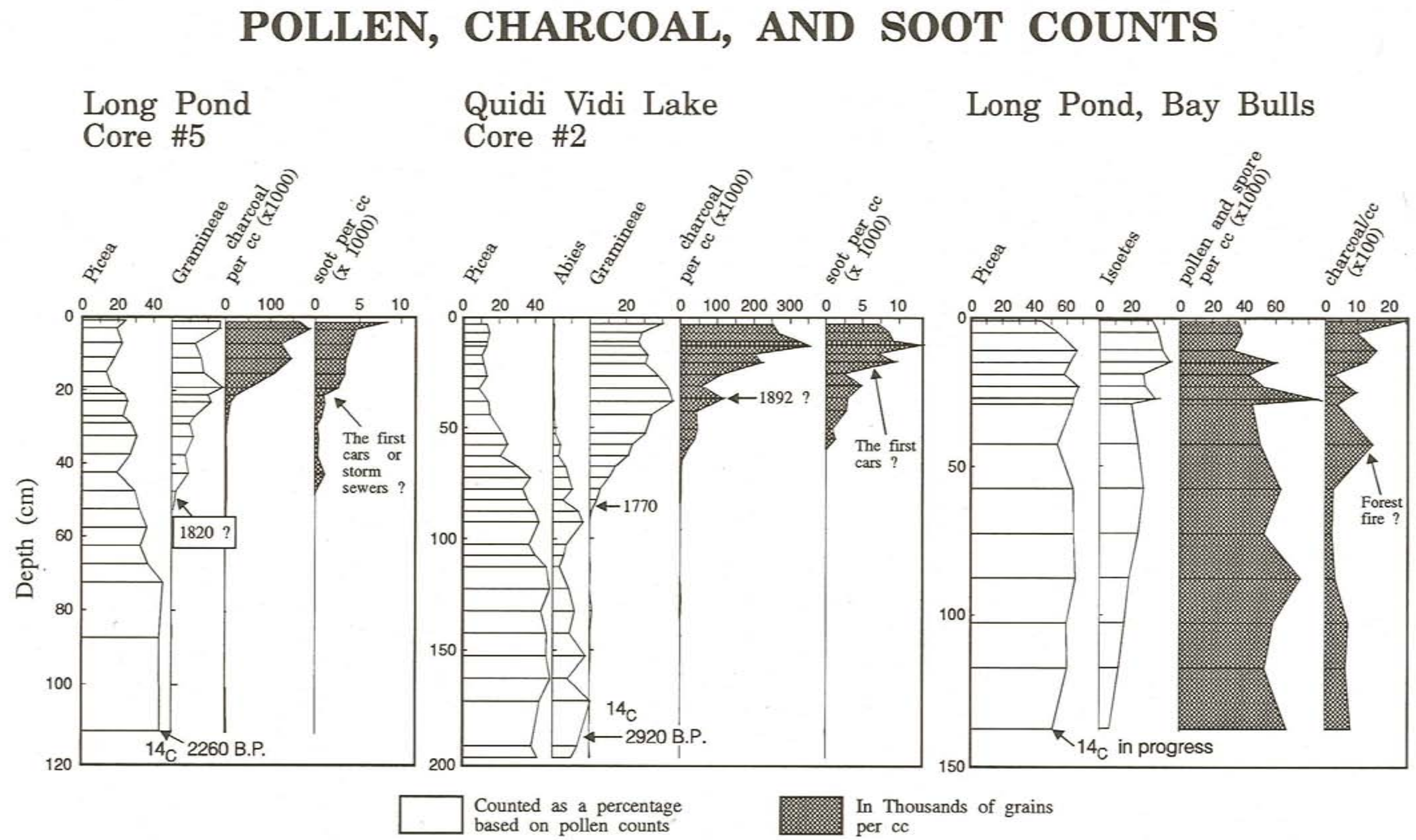

Figure 3. *Pollen, charcoal, and soot profiles from Long Pond core #5, Quidi Vidi Lake #2, and Long Pond, Witless Bay. Pollen for each species shown as a percentage of total pollen count, soot and charcoal as number of grains per cc. Note different vertical scales for each core.*

whole core, reflecting the minimal vegetation disturbance by land clearance, so the only change occurs within the pond itself. The changes in chemistry and aquatic pollen counts coincide with a charcoal peak that may further reflect the first human disturbance at 30 cm depth, perhaps occurring with the appearance of cabins along the pond. The charcoal profile also exhibits evidence of a forest fire in the large peak at 45 cm depth. Only one soot spheroid was counted in this core, and it was in the top 2 cm. The absence of soot is thought to be a result of the tree barrier separating the road and the pond, and low traffic volume on the road itself.

Georges Pond

The core from Georges Pond on Signal Hill was collected in 7.6 m of water and the sediment type is gyttja throughout its 130 cm length. Four units were defined, based on colour changes in the wet sediment: Unit 1, from the bottom of the core at 130 cm to 55 cm is black; Unit 2, from 55 to 24 cm is olive brown; Unit 3, from 24 to 18 cm is light green-brown; and Unit 4, from 18 cm to the top is dark green-brown. A ^{14}C date on a bulk-sediment sample from near the base of the core at 120 cm, gives an age of 3960 ± 70 years BP (GSC 5485).

Only minor chemical change was expected as anthropogenic disturbance appears to be limited to a paved road and a few nearby buildings. The chemical profiles for LOI and many of the elements (e.g., Fe and Co) indicate the first signs of human activity are recorded at a depth of 55 cm (Figure 4). Records show that this pond was the first water supply for St. John's with water being pumped for use by 1852 (Malcolm Pirnie, 1951).

For Pb (total), background values in the core below 55 cm range from 2 to 8 ppm whereas the peak values reach as high as 320 ppm in the upper units, a 40 fold increase. This was unexpected since the pond is somewhat isolated from the city, and lies within the Signal Hill Historic Park, a protected area. Total Zn levels also increase up the core at this depth from 147 ppm at 18 cm to 188 ppm at 16 cm. These increases may be related to up-grading and paving of the road up to Signal Hill, which would imply very much greater sedimentation rates in Units 2, 3 and 4 than Unit 1. The Zn values are attributed to galvanized railings and culverts used in road construction. Other possible effects of road construction can be seen in LOI where the lowest values are between 18 and 14 cm depth.

The physical stratigraphy appears to correlate well with the chemical stratigraphy. The first chemical increase and peak in elements such as Au, Fe, Co and Pb, possibly due to fallout from coal burning, occurs within Unit 2, whereas increases for elements such as Zn, Al, and Na, which may be related to road construction, occur within Unit 4.

Mundy Pond

The sediments of this shallow pond, located in one of the older parts of the city (Figure 1), show chemical changes to a depth of 65 to 70 cm (Figure 5). The core, which was collected in 1 m of water, contains four stratigraphic units. Unit 1, from the base of core at 100 cm to 48 cm, is light-brown gyttja; Unit 2, between 48 and 25 cm, is an olive-grey-brown to light-brown gyttja; Unit 3, from 25 to 15 cm, is an olive-black gyttja, and Unit 4 from 15 cm to the top is an olive-grey-brown clay-gyttja. Three horizons of plant debris occur between 40 and 60 cm.

The geochemistry displays several features that may correlate with the history of settlement in the area. At 25 cm, the boundary between Units 2 and 3, an abrupt increase occurs in several elements such as As, Au, Cd, Fe and Zn, (Figure 5). This overall increase corresponds with a decrease in organic content (LOI) and Br, suggesting that it's due to a much greater rate of clastic sediment influx and deposition. Superimposed on this overall trend, there are variations in particular elements or groups of elements, such as the peaks at 18 cm in Cd, Cu, Fe and Zn, which indicate an additional source of heavy metals and, associated with the Mn peak at 20 cm, coincident spikes in Ag, As, Bi, Co, Cr, and P. Chromium (not shown in the Figure 5) increases dramatically from 40 ppm at 28 cm to a peak of over 1600 ppm at 22 cm and drops off to 100 ppm at 16 cm depth. This suggests a major but relatively short-lived change in water chemistry leading to co-precipitation of some of the pollutants with Mn oxides. The earliest sign of chemical distubance in the lake-sediment record are the increases in rare-earth elements such as La, Ce, Sm, Eu, Tb, Dy and Yb in Unit 2, which starting at 60 cm depth, increase to a peak between 28 and 24 cm, then drop off until around 16 cm, after which they remain more or less constant through the top stratigraphic unit.

This chemical and sedimentary stratigraphy records a history of human activity, which perhaps starts with a farming era detectable at a depth of 60 cm. Subsequent activities including coal burning with consequent airborne pollution, initial suburban development when sewage effluent was dumped into the pond, and more recent urbanization with the construction of roads, storm sewers and houses is also recorded in the sedimentary record. The 'sewage era' possibly started when the pond area was first settled and became intensified during the period corresponding to the interval between 25 and 15 cm depth. A report on sewage systems in the city (Sunderland *et al.*, 1970) indicated that the sewage for the Mundy Pond area was first diverted to the harbour in 1967, although the area west and north of the pond remained without a sewer system for some time after this. The report makes no reference to where the sewage was dumped before 1967, but most likely, it was into the pond as suggested by Bland (1946). Thus, the change in the stratigraphy at 15 cm depth could be associated with this change in sewage discharges or be related to the first installation of storm sewers (or both). The LOI profile, which drops from 29 percent at 20 cm to about 17 percent at 16 cm, likely indicates when construction (including the construction of houses, roads, storm sewers and sanitary sewers) was at a peak.

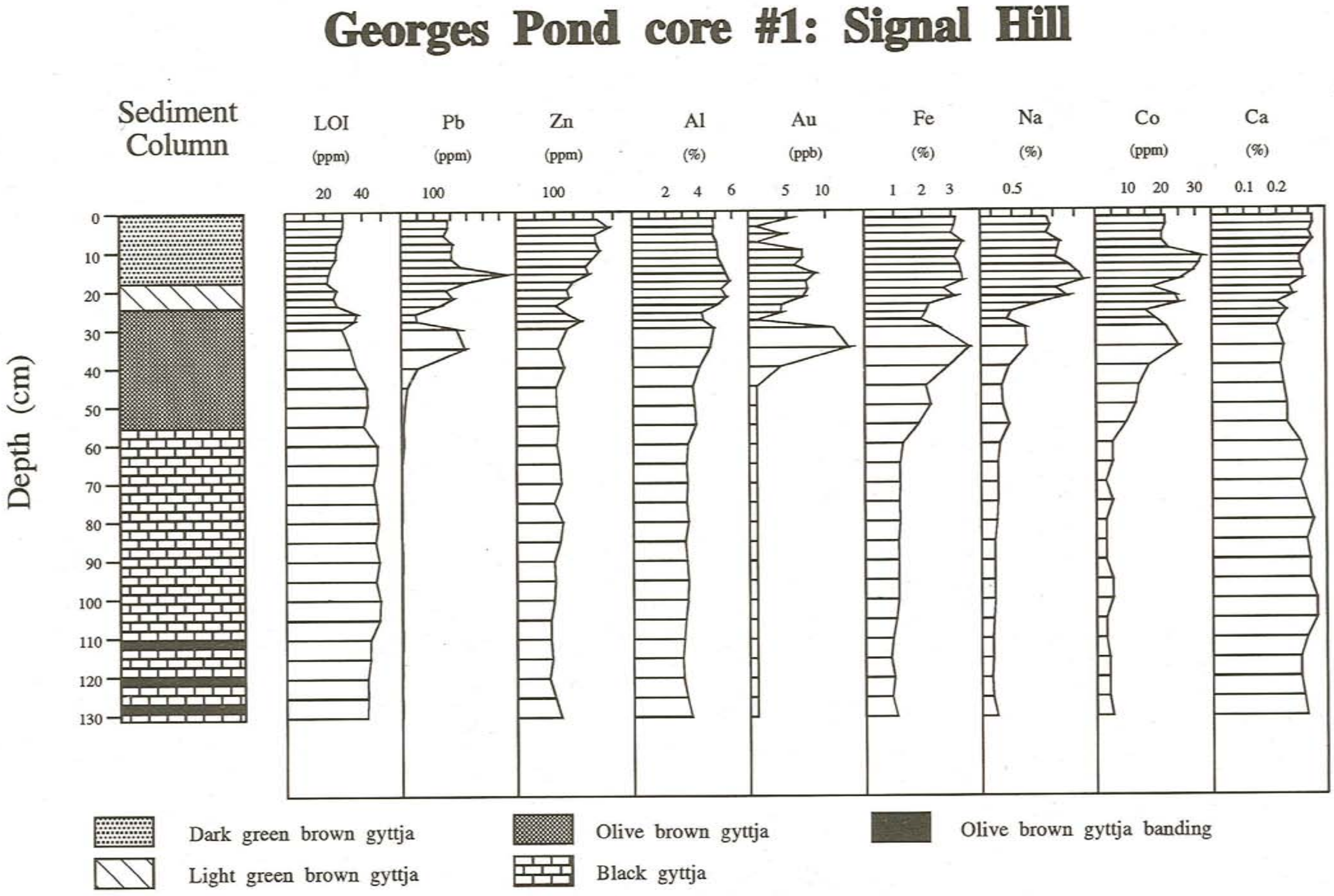

Figure 4. *Stratigraphy and geochemical profiles for LOI, Pb, Zn, Al, Au, Fe, Na, Co and Ca in lake-sediment core #1 from Georges Pond.*

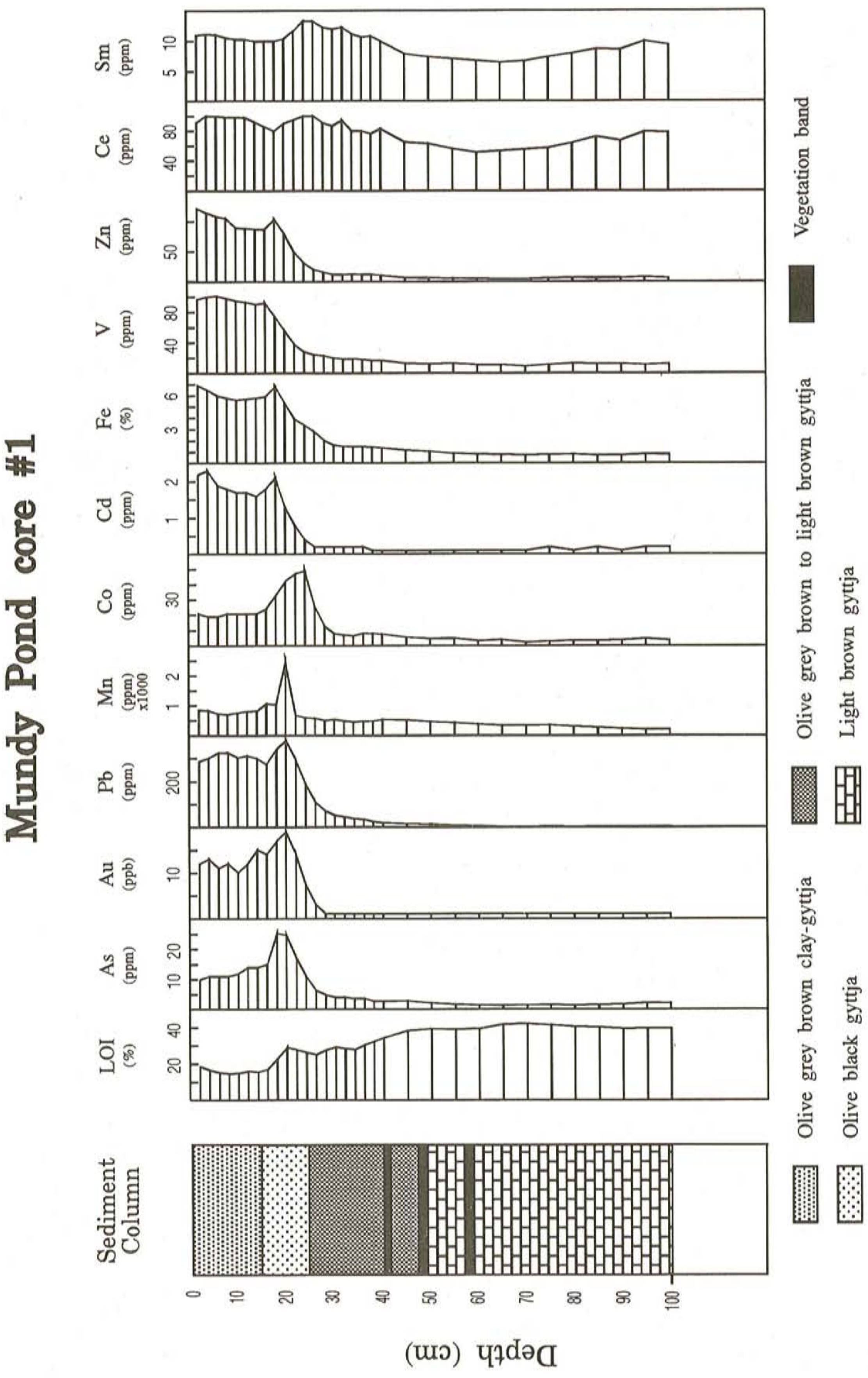

Figure 5. *Stratigraphy and geochemical profiles for LOI, As, Au, Pb, Mn, Co, Cd, Fe, V, Zn, Ce and Sm in lake-sediment core #1 from Mundy Pond.*

Long Pond

Water depth at the coring site in Long Pond was 8.82 m. The stratigraphy, which is divided into three units (Figure 6), is characterized by a change in sediment type at 20 cm depth. The bottom section from 115 to 48 cm (Unit 1) is composed of brown gyttja; Unit 2, between 48 and 20 cm, is composed of light-brown gyttja, while the upper 20 cm (Unit 3) is composed of olive-grey, clay-rich gyttja. A ^{14}C date on a bulk sediment sample from near the base of the core at 115 cm gave an age of 2260 ± 70 years BP (GSC 5480).

The pollen profile (Figure 3) from this pond first records farming activity by the first appearance of Gramineae at a depth 55 cm. Historical records indicate farming started in this area around 1820 (MacKinnon, 1981). Chemical evidence suggests the watershed was affected earlier at 60 cm depth (Figure 6), as concentrations of elements such as Al and Fe, which indicate increased soil erosion and the deposition of an increased proportion of silt, increase slightly at this point from background values.

The chemical stratigraphy in this core, as with most other cores, correlates well with the sediment stratigraphy. Some element concentrations peak in Unit 2 and others in Unit 3 (Figure 6). At the boundary between Units 2 and 3 at 20 cm, LOI drops sharply (from 32 percent at 24 cm to only 14 percent at 16 cm). Several elements, including Al, Cs, Fe, Ga, Hf, K, Ba, Ca, Cd, Li, Mg, Na, Ni, Pb, Rb, Sr, Th, U, V, W, Zn and Zr show a corresponding increase. Oil droplets and soot from oil burning, as described by Goldberg *et al.* (1981) first appear at this depth. In the Michigan Lake area, Goldberg *et al.* (1981) related this first appearance of soot from oil combustion to the first installation of oil-burning furnaces there between 1948 and 1953. Presumably this feature has a similar explanation in St. John's where the first oil furnaces likely did not arrive until after World War II. The coincidence of occurrence of the first appearance of oil droplets and the sudden drop in LOI at this depth, however, suggest the possibility that these changes are related to the construction of roads and storm sewers in the Long Pond catchment. Storm sewers, which drain roads and parking lots receive much debris including oil, and as several discharge into Long Pond via Leary's Brook, they are probably a source for some of the oil.

At 20 cm depth there is a dramatic increase in Pb levels correlating well with the influx of mineral matter, an increase in soot (Figure 3) and the first occurrence of the oil droplets (Figure 6). The increases in Pb and Na are about three-fold at this point, whereas other elements indicative of the increased clastic component in the sediment such as LOI and Al change more modestly. This suggests that there were additional inputs at this time for Na (possibly road salt) and Pb (possibly leaded gasoline).

Gold shows an interesting trend in this pond (Figure 6), increasing from below the detection (<2 ppb) below 26 cm to 6 ppb at 24 cm and rising to 10 ppb at the top. Several other elements show similar trends including As, Fe, Na and, to some extent, Pb. The peak in As between 22 and 24 cm is associated with Sb, Au and Fe, an element suite that is thought to reflect fall-out from coal burning in the city.

Quidi Vidi Lake

Quidi Vidi Lake is the largest lake in this study area and is located in one of the oldest parts of the city. Several cores were collected from this lake, but most work has been carried out on core #2 which was collected in 12.5 m of water. Core #1 was collected 3 m away from core #2 in 11.9 m of water.

Core #2

Four stratigraphic units are recognized; Unit 1, from the bottom of the core at 200 cm to 83 cm, is a dusky-brown gyttja; Unit 2, from 83 to 55 cm, is a dusky yellowish-brown gyttja; Unit 3, from 55 cm to 44 cm, is a brownish-black clayey gyttja; and Unit 4, from 44 cm to the top, is an olive-black clayey gyttja (Figure 7). Thin horizons of plant debris occur at 32, 122 and 188 cm. A ^{14}C date on a bulk sediment sample from near the base of the core at 190 cm gave an age of 2920 ± 60 years BP (GSC 5469).

Pollen profiles first record change based on the *Picea* counts as deep as 110 cm. The presence of *Gramineae* pollen is indicative of farming, which according to historical records began in this area around 1770 (Head, 1976), and this species first appears at 80 cm (Figure 3).

Chemically, this lake shows evidence of human activity as deep as 85 cm. As in the other ponds, there is a relationship between chemical patterns and the sediment stratigraphy (Figure 7). Several elements commonly associated with rural development (Engstrom and Wright, 1984) such as Al, Ca, Fe, Mg and Rb increase in concentration up through the core from 82 cm. Some of these elements such as Ca continue to increase up through the core after the first evidence of human activities, whereas other elements such as K increase to a certain horizon (approximately 33 cm) and then remain constant to the top.

Background values for Pb (total) of 15 to 23 ppm occur in the lower 125 cm of the core. At about 75 cm, Pb shows a progressive and rapid increase, reaching peak values of 480 ppm to 610 ppm in the top 18 cm. Thus, there has been as much as a 30 fold increase in Pb values in the recent sediments. In detail, there are two peaks for Pb and elements such as Au, Fe, Ni, As, Co, Ag, Sb and Zn, with the second between 55 and 33 cm. This lower peak is the maximum for Ag, As, Au, Co and Sb. Preliminary data indicate $^{208/207}Pb$ and $^{206/207}Pb$ ratios form three clusters: one group in the lower metre of the core where Pb values are background, a second at 47 cm within the lower Pb peak, and a third within the upper Pb peak above 22 cm.

The charcoal profile shows a peak at 39 cm depth (Figure 3), but the maximum occurs toward the top of the core. The

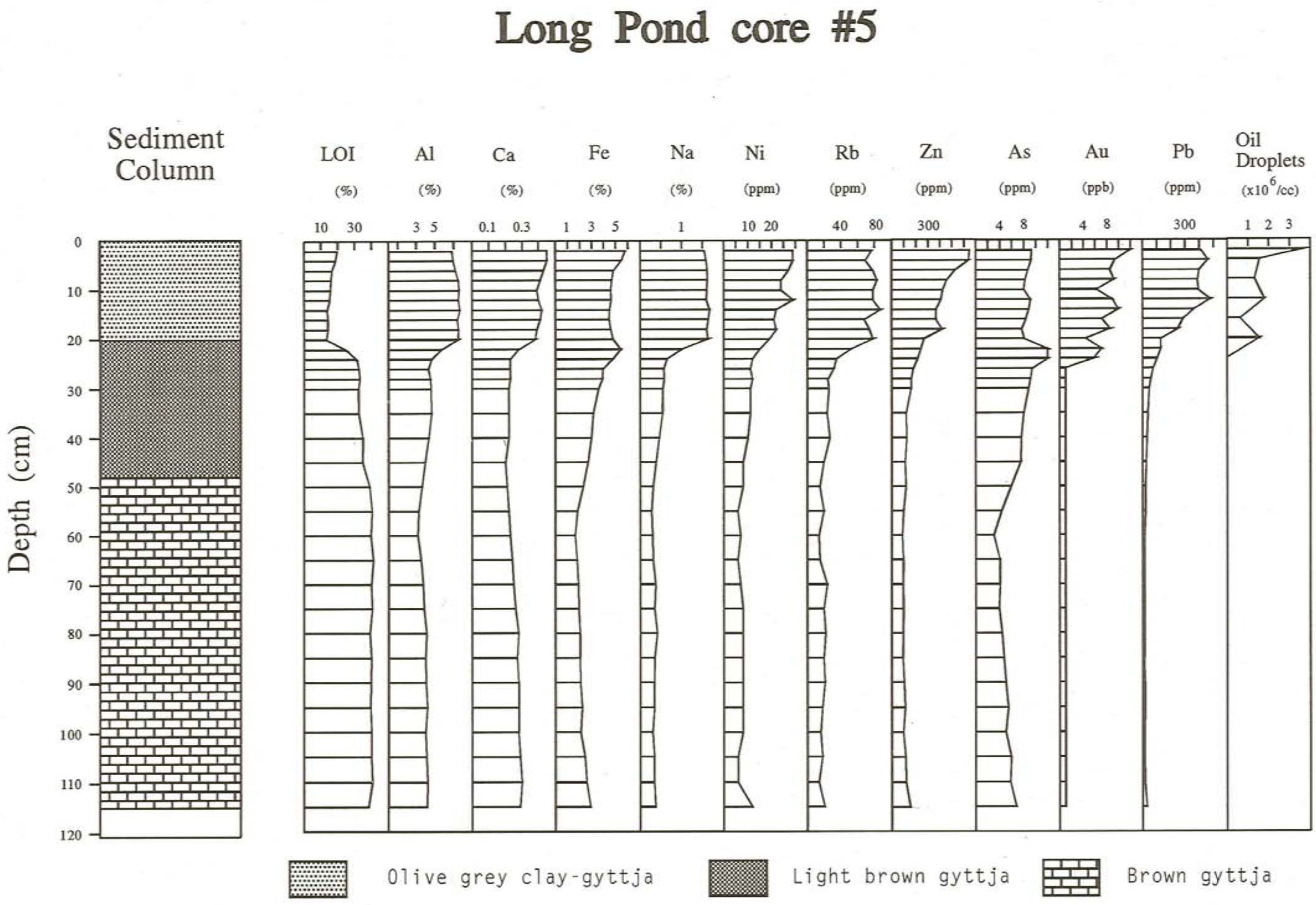

Figure 6. *Stratigraphy, geochemical profiles for LOI, Al, Ca, Fe, Na, Ni, Rb, Zn, As, Au and Pb, and distribution of oil droplets in lake-sediment core #5 from Long Pond.*

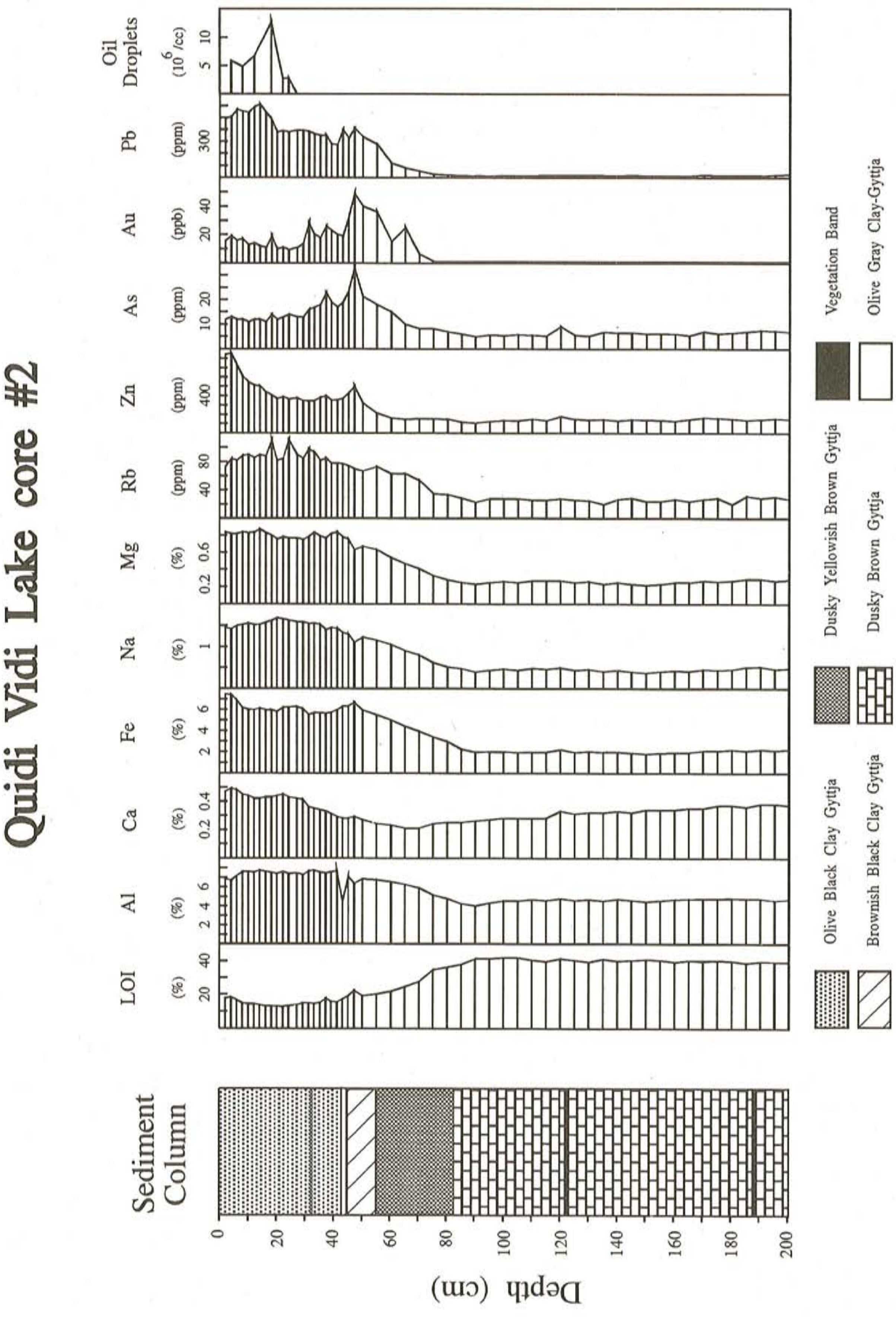

Figure 7. *Stratigraphy, geochemical profiles for LOI, Al, Ca, Fe, Na, Mg, Rb, Zn, As, Au and Pb, and distribution of oil droplets in lake-sediment core #2 from Quidi Vidi Lake.*

soot profile shows a gradual increase from 55 to 27 cm, followed by a sharp rise to 12 cm. Oil droplets were observed only in the upper 20 cm (Figure 7), coinciding with the peaks in charcoal, soot and Pb.

Core #1

This core was collected next to core #2 to evaluate the variability of physical and chemical stratigraphy at nearby sites in a lake, and to compare the magnitude of these site variations with those in cores from opposite ends of the same lake, and in cores from different lakes. The physical stratigraphy is very similar in the two cores (Figure 8), although the thicknesses of the units in core #1 are consistently less than in core #2. For example, in core #1 the first sediment change occurs at 72 cm depth whereas in core #2 it occurs at 83 cm. The water depth at the site of core #1 (11.9 m) was 0.6 m less than at site #2 which, together with the slightly condensed sediment column, suggests that core #1 was collected at a site where there is a relative high on the surface beneath the recent lake sediments, and this relative elevation difference in the lake bottom has persisted to the present.

The chemical stratigraphy closely mirrors the physical stratigraphy in being slightly condensed in core #1 compared with core #2. Both the trends and amplitudes of the chemical parameters in the two cores are very similar. In particular, the similarity in the Pb peak in the upper 20 cm confirms that the attenuation of Unit 1 in core #1 relative to core #2 is real and not due to loss of the uppermost, rather fluid sediment during core recovery.

There are some small-scale differences in core #1 that are not apparent in core #2, such as changes in LOI at 145 and 75 cm. The latter change in LOI occurs at the boundary between Units 3 and 4 and the decrease in LOI is accompanied by corresponding increases in several elements such as Fe. These local features may be in part due to normal random errors in sampling and analysis or to local diagenetic effects. They do not obscure the main features of the chemical stratigraphy, however, but clearly small, single-sample variations in chemistry must be interpreted with caution unless supported by corroborating data from physical or biological parameters.

DISCUSSION

The record of anthropogenic change in lakes and their catchments, which is contained in the variations of the physical, chemical and biological parameters measured in the sediment cores, is a complex temporal and spatial web of information. Some parameters reflect mainly local events that affect an individual lake at one or more periods in time. For example, the installation of a storm-sewer system will only be recorded in lakes in the drainage network receiving the newly channelled run-off, and this type of development may occur at different times in different catchments. Features associated with this type of change will be similar from lake to lake, but the first appearance of these features will not provide a common time datum. Airborne pollution transported over long distances, such as ^{137}Cs deposition from atmospheric nuclear explosions, will affect wide areas, and within the study area its first appearance and peak in the sediment record from all lakes will provide temporal benchmarks (1945 and 1963 in this case). Many of the parameters measured in this study reflect anthropogenic processes that affected areas over scales intermediate between these two extremes.

The lack of trends in the abundances of pollen from terrestrial species in the background pond over the time interval represented by the 120 cm core confirm the limited degree of disturbance of the vegetation observed in its catchment, although the variation in the pollen of the aquatic species Isoetes indicates that the pond is not pristine. The distributions of pollen from Picea and Gramineae in the two urban lakes studied, Long Pond and Quidi Vidi, show similar trends that can be related to the clearing of the indigenous forest and the establishment of pastureland (Macpherson and MacKinnon, 1988). These events took place around 1770 near Quidi Vidi Lake and around 1820 near Long Pond. The changes are more intense and prolonged in Quidi Vidi Lake, reflecting the longer history and greater intensity of human disturbance in this catchment (Figure 3). The onset of these changes is diachronous and is dated at each lake from historical records.

If the assumption is correct that the first appearance of Gramineae at 80 cm occurred at or shortly after the start of farming in the area in 1770, then this would imply that the sediment influx for this pond has been enormous over the last 200 years considering the size of the lake. In Kenny's Pond, a much smaller pond near Long Pond, Macpherson and MacKinnon (1988) established that since the start of farming in that area in 1820, 75 cm of sediment have accumulated, implying an average sedimentation rate of 22 times pre-settlement rate. For Quidi Vidi Lake, if the changes in chemistry and pollen profiles at 75 to 80 cm date from around 1770 when farming commenced in the area, then the average sedimentation rate since European settlement (0.35 cm per year) is almost 10 times the pre-settlement rate of 0.04 cm per year. Generally, similar changes are suspected for Georges Pond and Long Pond, and probably Mundy Pond too. Whereas a greater degree of compaction in the deeper sediments would partly offset these apparent differences in sedimentation rates, the differences are too large to be completely explained in this way. Moreover, the preliminary ^{137}Cs results suggest sedimentation rates over the past 30 years in Quidi Vidi Lake may have exceeded 0.6 cm per year.

Chemical parameters in the sediment cores reflect several anthropogenic processes that vary in their region of influence from local (i.e., an individual lake) to the entire area. The most widespread feature is the increase in levels of Pb (both total and acid-extractable) at the top of all cores, including the 'background pond'. This result, together with evidence of higher Pb levels in recent lake sediments from south-central Newfoundland (Davenport *et al.*, 1992), indicates that the regional background Pb levels in sediment have increased by 10 to 20 ppm, presumably due to the long-range transport

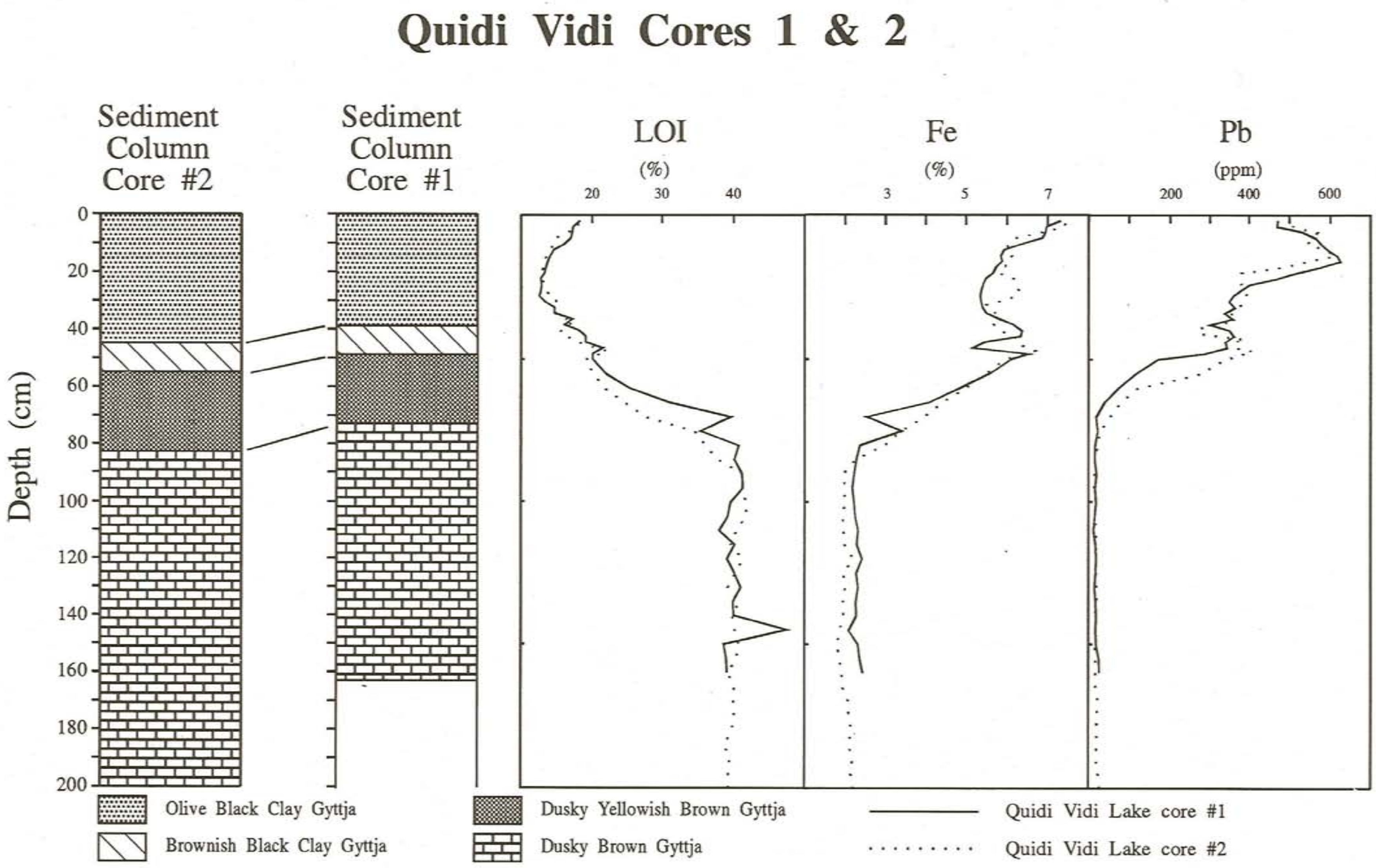

Figure 8. *Stratigraphy, LOI, Fe and Pb profiles from Quidi Vidi Lake cores 1 and 2.*

and deposition of Pb. In the urban lakes, the increase has been far greater: an additional 250 ppm has been added to the sediments of Mundy Pond, 300 ppm in Long Pond and almost 600 ppm to Quidi Vidi Lake sediments. Even the recent sediments in Georges Pond within the Signal Hill National Historic Park have Pb levels more than 100 ppm above the natural background. The widespread distribution of this Pb pollution and its spatial variation suggest leaded gasoline to be the most probable source. In these urban lakes, the contribution from local traffic dominates over the Pb from distant sources. Associated with this Pb peak are increases in Sb in the sediment of both the 'background pond' and city lakes.

This rise in Pb and Sb within the most recent sediment is the most striking feature in the 'background pond', but in the urban lakes it forms only part of a more complex pattern. In Quidi Vidi Lake, Georges Pond and Mundy Pond there is an earlier prominent rise in Pb, together with As, Au, Co, Fe, Sb and in some cores other heavy metals such as Cd and Zn. The presence of this feature in the lakes closest to the 'oldtown', including Georges Pond, but its absence or very weak expression in Long Pond on the edge of a large parkland, suggests that it may be the result of local air pollution from the widespread use of coal for domestic and industrial fuel. Coal was commonly burnt in St. John's from the beginning of the 19th century until the 1950's (Newfoundland Statistics Agency, 1981). Given its contined use until quite recently, it is surprising that the pollution effects attributed to coal-burning peak in relatively old sediments. Their diminished signature in sediments thought to date from the mid-20th century may be due to a switch in the dominant source of coal. Historical records indicate the first coal was imported from Britain as early as 1810 (Prowse, 1895). By the 1880's most of the coal came from Sydney, N.S. (Johnson, 1959), and the Sydney coal appears to be lower in heavy elements than the early sources resulting in a drop in the concentrations of these elements.

The $^{208/207}$Pb and $^{206/207}$Pb isotopic ratios on seven sediment samples from Quidi Vidi Lake core #2 cannot resolve the question of coal sources. They do establish, however, that the lower Pb peak at 47 cm in this core is distinct in its $^{206/207}$Pb ratio from the samples at 60, 100 and 170 cm that form a cluster of similar values that are taken to be the local, natural background ratios, thus indicating that a significant component of the Pb at 47 cm is exotic. Similarly, the ratios at 22 and 14 cm are distinctly different from both the sample at 47 cm and the background ratios at 60 cm and below, indicating a second exotic source for the upper Pb peak.

These peaks in heavy metals and related elements are superimposed on a general increase in their levels and those of the major elements contained in silicate rocks (Al, Fe, Ca, Mg, Na and K, as well as minor elements such as Ti, Rb and Sr). These general increases that are observed in all city cores, and which are mirrored by a decrease in organic content (LOI), are the result of increasing rates of erosion leading to a larger clastic component in the sediments. The initial cause of this was forest clearance and the establishment of farms, both of which increased the rate of soil erosion. These initial changes in sediment composition are quite subtle, and the start of these activities is recorded more clearly in the pollen record. Major changes occur higher in the cores of city lakes during suburban and urban construction in their catchments. These large chemical changes are reflected in the physical appearance of the sediment itself, which becomes noticeably richer in silt and clay in the upper sections of Quidi Vidi Lake, Long Pond and Mundy Pond. These changes are less apparent in Georges Pond, where the disturbance from construction is much less. The dates, at which the effects of urban development are recorded, are likely to vary from lake to lake as the city expanded from the old town by the harbour into the catchments of Quidi Vidi Lake, Mundy Pond and Long Pond.

The distributions of soot and charcoal in the cores (Figure 3) provide information on the distribution in space and time of airborne particulate pollution. Charcoal particles, from the burning of wood, are persistent but not abundant (generally < 20 per cc) in the sediment of the 'background pond', and occur with about the same frequency in the lower, undisturbed sections of the Long Pond and Quidi Vidi cores. Values in the upper sections of the cores from these two city lakes are up to 100 times higher than even the peak in the 'background pond' core that is tentatively attributed to a forest fire. These very high values in the urban sediments are presumably due to the almost year-round burning of firewood for domestic heating. The soot particles are from three main sources; coal burning, oil burning and automobile exhaust. Under some conditions the three may be distinguished on their morphology and surface texture (Goldberg *et al.*, 1981), but such distinctions have not been made in the present study. Soot is absent from the sediment in the background pond, but in Quidi Vidi Lake it occurs in all samples above 60 cm and in Long Pond above 45 cm. The Pb and soot profiles for Quidi Vidi Lake (Figures 8 and 8 respectively) correlate well, suggesting a possible relationship between the two and lending support for a link between the initial Pb rise and coal consumption.

Oil droplets were observed in the slides from the cores from both urban lakes that were prepared for pollen analysis. They are restricted to the top 27 cm in Quidi Vidi Lake and the top 20 cm in Long Pond, and are therefore a fairly recent feature. Their distribution is somewhat similar to that of Pb, but there are differences in detail. The most likely sources of the oil are furnace oil (incomplete combustion, spills and leaks) and automobile lubricating oil (spills and waste). Oil furnaces did not become common until the 1960's, whereas automobiles and service stations were common in the 1920's and 1930's (Adams, 1991).

CONCLUSION

To fully decipher the history of environmental change recorded in the lake-sediment cores of St. John's, an absolute time scale is necessary. The pollen records provide mainly catchment-scale change, which can be diachronous. The

^{137}Cs, ^{210}Pb and ^{14}C dating currently in progress should provide this framework and refine the interpretations presented here by constraining many of the tentative links between sedimentary features and the historical record.

ACKNOWLEDGMENTS

We would like to thank the following who helped with the coring during the winter of 1992: Skip Pile, Rick Bourgeois, and Martin Blake, without whose help sampling would have been impossible; Drs. Joyce Macpherson, Ali Aksu, and John Kingston for help at various stages logistically and academically; Paul Edwards, Department of Earth Sciences, Memorial University, and C-Core for their logistical help; Dr. Roger McNeely of the Terrain Sciences Division, Geological Survey of Canada, for the ^{14}C age-dating of sediment samples. The Faculty of Graduate Studies and the Department of Mines and Energy for their financial support making this project possible. The helpful comments of Dr. David Liverman on a draft of this paper are gratefully acknowledged.

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