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Sources and Sinks of Lead and Other Trace Metals Enriched in the Surface Sediments of Remote Subalpine Lakes in Taiwan

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ABSTRACT

Sediment cores were collected from two remote subalpine lakes: the rather shallow (1.5 m) Little Ghost Lake at 2040 m elevation and the deep Great Ghost Lake (35 m) at 2150 m elevation. The distributions of metal/Al (in bulk samples) and acid-leached metal in the sediments of the Little Ghost Lake suggest that the surface enrichments of Cd and Pb (probably also Cr and Zn) are caused mainly by anthropogenic inputs. The distributions of leached Pb and Pb-206/Pb-207 ratios indicate a large anthropogenic input since about 1940, as based on Pb-210 and C-14 dating.

A natural process driven by the Fe redox cycle caused surface enrichments of acid-leached Fe, Ni, Zn and part of Pb in the Great Ghost Lake. The Pb-206/Pb-207 ratios confirm the anthropogenic Pb input in surface sediments here. According to C-14 dating, the onset of the large anthropogenic Pb increase occured in around 1945, suggesting that such large increases in these two subalpine lakes are due to widespread lead emissions from automobiles.

The Fe oxides produced during winter overturn in the Great Ghost Lake play a major role in removing the aeolian anthropogenic metals, particularly Pb, from the water column. In the Little Ghost Lake, the aeolian anthropogenic metals may be directly removed with the sinking of aeolian particulates and/or by aquatic organisms.

(Key words: Trace metals, Lake sediments, Fe redox cycle)

1. INTRODUCTION

For a number of years the authors have been studying the effect of acid rain on lakes in Taiwan (Chen *et al.*, 1988). With a strong interest in trace metal fluxes, it is necessary to understand the various pathways of trace metals among which lead is one of the easiest to study. The Earth's atmosphere has been known to transport both natural and anthropogenically

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mobilized trace elements. Atmospheric input has been shown to be dominant for common lead (Schaule and Patterson, 1981, 1983; Patterson and Settle, 1987) and Pb-210 (Nozaki *et al.*, 1976). It is known that atmospheric anthropogenic Pb has been produced from the development of cupellation technology starting about 4500 bp (Settle and Patterson, 1980), and then its production increased markedly in the late 18th century at the beginning of the Industrial Revolution. Around 1940, there was a sharp increase in atmospheric lead concentrations due to lead emissions from automobiles (Murozumi *et al.*, 1969).

The anthropogenic source of lead is relatively easy to identify as most ore leads have Pb-206/Pb-207 ratios measurably less than the natural leads of soil and soil related components (Shirahata *et al.*, 1980). Comparing the relative abundance with respect to Al or leaching with dilute acid are two methods of reducing the grain-size effects in order to evaluate the sources (natural or anthropogenic) of metals in sediments (Kemp and Thomas, 1976; Kemp *et al.*, 1976; Ng and Patterson, 1982; Forstner and Wittmann, 1983; Finney and Huh, 1989). Fe and Mn oxides, the surface of clay minerals, organic matters and carbonates are the major phases of metal association in sediments. Acid-leached metals measured here represent most of the metals in these phases but do not include those in the lattice of clay minerals and rock debris. The latter are of natural origin and are not changed by anthropogenic inputs.

In this paper, metal concentrations and Pb-206/Pb-207 ratios in the leachate as well as metal contents and metal/Al ratios in bulk sediments are reported to identify the sources of lead in two subalpine lakes in Taiwan.

2. STUDY AREA AND METHODS

The Little Ghost Lake (Hsiao-Kuei Hu), located at 22°40'N, 120°53'E, is a rather shallow, 2040-m high lake which used to be quite isolated until 1991 at which time a road was constructed to within a few km. Some wind mixing and bioturbation are expected to have occured because the maximum depth is only 1.5 m and the lake is rich in fish and other biota.

The Great Ghost (Ta-Kuei Hu) Lake, situated at 22°52'N, 120°51'E, is a 2150-m high lake and one of the best preserved natural lakes in Taiwan, with little human perturbation because of its isolation. It takes two days to reach this lake by foot from the nearest road. The sediments are well preserved as the lake is not only deep (maximum 40 m) but also anoxic in the deeper water most of the year (Wang, 1989; Chen and Wang, 1990; Wang and Chen, 1990). Little or no wind mixing or bioturbation is expected to have occured.

A home-made gravity corer with a plastic barrel was used to collect the sediment samples. The cores were sealed with wax immediately after recovery and then opened at the home laboratory by being cut with a fishing line (Chen *et al.*, 1993a).

In the leaching procedure, the dried sediment (0.3 g) was treated with 15 ml of 1.6 N nitric acid and shaken overnight (Ng and Patterson, 1982; Giblin *et al.*, 1990) in a 50-ml centrifuge tube. The leachate was separated by centrifuge and was then removed with a quartz pipette for the measurements of metals and Pb-206/Pb-207 ratios. The dry sediments (0.3 g) for the determination of the total metal content were digested with an acid mixture (40% HNO_3, 15% $HClO_4$ and 20% HF) in pressured bomb or were digested by a microwave system with an acid mixture (42% HNO_3 and 17% HF). Metals and Pb-206/Pb-207 ratios were measured by an ELAN 5000 ICP-MS with a Ryton spray chamber and cross flow nebulizer to which sample solutions were delivered with a peristaltic pump at a flow rate of 1 ml/min. Carbon-14 dating was done by both conventional and AMS methods, while Pb-210 dating was done for the core from the Little Ghost Lake (Lin, 1992; Lin and Chen, 1992; Chen *et al.*, 1993b).

The National Institute for Environmental Studies No.2 Pond Sediment was used to test the accuracy of the trace metal determinations (Table 1). Most of the accuracy deviations shown in Table 1 are less than 15%. A correction was made only when the deviation was larger than 15%. The accuracy of the Pb-206/Pb-207 measurements could not be checked, but the precision of isotope ratio measurement was roughly 1% (n=5).

Table 1. Precision and accuracy of trace metal measurements (whole sediment, n=5)

(a) Samples are digested with an acid mixture (40% HNO_3, 15% $HClO_4$ and 20% HF) in pressured bombs.

Metals	measured (ug/g)	standard (ug/g)	deviation (%)
Cd	0.81±0.07	0.82±0.06	1.2
Cr	88.3±10.8	75±5	17.7
Cu	226±12	210±12	7.6
Fe	6.29±0.47	6.53±0.53	3.7
Mn	616±41	770	20
Ni	51.2±5.7	40±3	28
Pb	117±20	105±6	11.4
V	287±26	250	14.8
Zn	294±11	343±17	14.3

(b) Samples are digested by a microwave system with an acid mixture (42% HNO_3 and 17% HF).

Metals	measured (ug/g)	standard (ug/g)	deviation (%)
Cd	0.96±0.07	0.82±0.06	17.0
Cu	185±6	210±12	11.9
Fe	5.81±0.31	6.53±0.53	11.0
Mn	711±29	770	7.7
Ni	37.4±0.8	40±3	6.5
Pb	124±13	105±6	18.1
V	238±4	250	4.8
Zn	234±9	343±17	31.8
Sr	113±13	110	2.7
Al	11.3±1.8	10.6±0.5	6.6

Standard: National Institute for Environmental Studies No. 2 Pond Sediment.

3. RESULTS AND DISCUSSION

The vertical distributions of total trace metal contents in the sediments of the Little Ghost Lake are given in Figure 1 for Al, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, V and Zn. It can be seen that the average concentrations of Al and Fe near the surface are about 30% less than those below. Al and Fe are the main elements in the earth crust and are the dominant inorganic elements in the sediment matrix (Kemp *et al.*, 1976). Al also has a strong positive relationship with clay in the sediments (Kemp *et al.*, 1976; Lin, 1992). The lower average

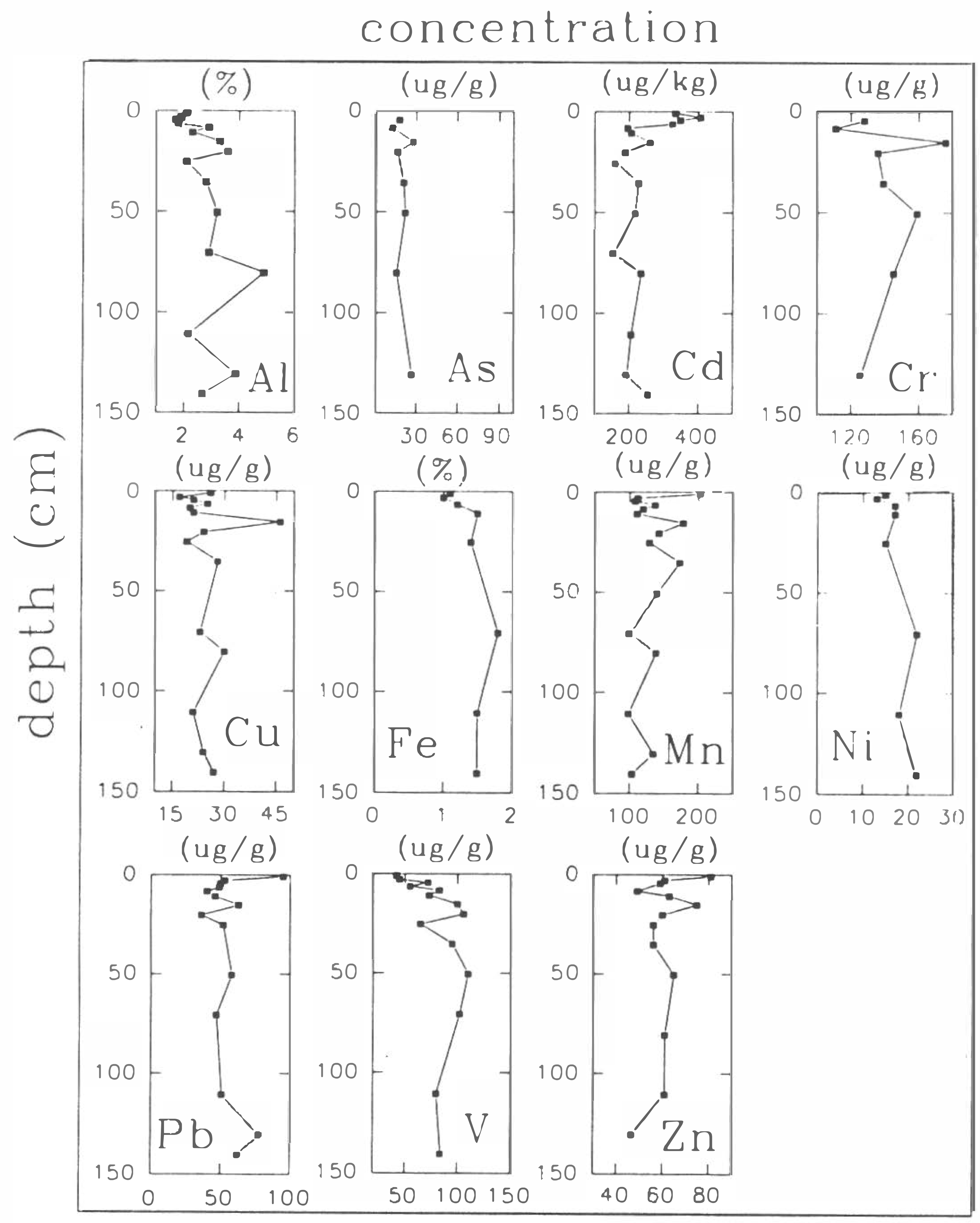

Fig. 1. Vertical distributions of trace metals in the bulk sediments of the Little Ghost Lake.

concentrations of Al and Fe near the surface are mainly caused by the dilution of organic matter (about 20%) and carbonates produced by the stonewort (Chara sp.) abundant on the surface sediment of the Little Ghost Lake. This near- surface dilution also results in a lower content of clay-size (< 4 μm) sediment (Lin, 1992), which has a relatively large enough surface area to enrich metals. These factors also probably led to the lower concentrations of Cr, Ni and V in the surface sediments.

Figure 1 shows that Cd, Mn, Pb and Zn are enriched in the bulk surface sediments of the Little Ghost Lake. The vertical distributions of metal/Al ratios in the bulk sediments of this lake are shown in Figure 2. Also seen are the surface enrichments of Cd, Mn, Pb and Zn (probably As and Cr, too), which, it seemed, may have been caused by anthropogenic inputs. In order to confirm this, the vertical distributions of acid-leached metals (Figure 3) and the Pb-206/Pb-207 ratios (Figure 4) in the sediment were measured. This confirmed that the surface enrichments of Cd and Pb were indeed mainly caused by anthropogenic inputs (acid-leached Zn and Cr were not measured). The samples for Pb-206/Pb-207 measurement were leachates leached by 1.6 N nitric acid. The use of leachate focuses on the exchangable phases of metals in the sediment, which associate with most of the anthropogenic metals (Gupta and Chen, 1975; Ng and Patterson, 1982; Giblin, 1990; Wann, 1990). The Pb-206/Pb-207 ratios

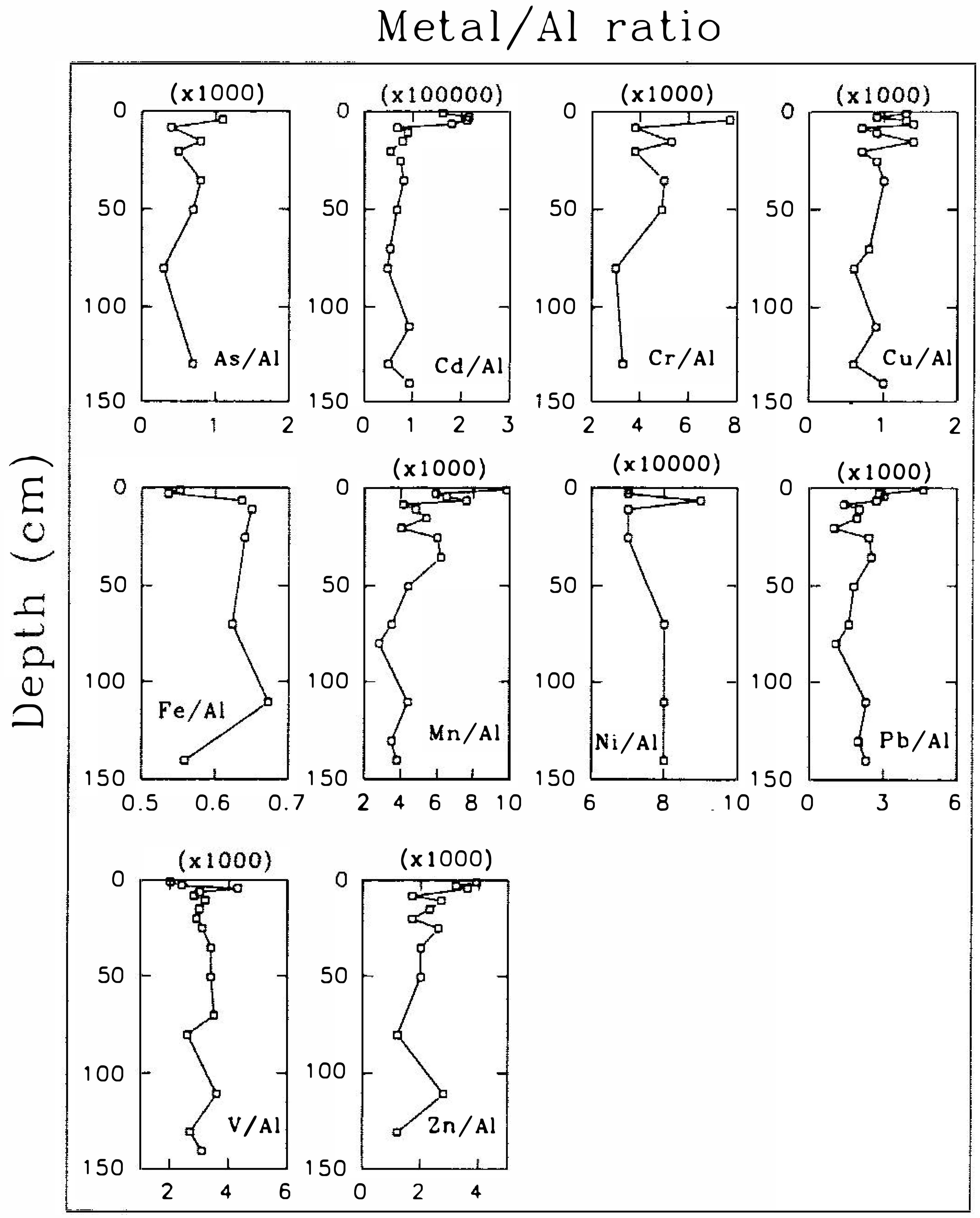

Fig. 2. Vertical distributions of the ratios of trace metal concentrations relative to Al in the sediments of the Little Ghost Lake.

increased from lower industrial-like values (roughly 1.16) near the surface to higher natural values (roughly 1.20) with depth in the Little Ghost Lake. This result is similar to those from other studies (Shirahata *et al.*, 1980; Settle and Patterson, 1982; Patterson and Settle, 1987).

The average values of acid-leached Al and Fe in the upper 8 cm are also about 30% less than the deeper values (Figure 3), as a result of dilution from organic matters and carbonates. Fe and Mn are very sensitive to redox changes in the sediments. The concentrations of acid-leached Fe and Mn are about half of their total contents in the bulk sediments. This suggests that acid-leached Fe and Mn represent most of the Fe and Mn in the Fe/Mn oxides, particularly the "fresh" ones (Forstner and Wittmann, 1983).

The vertical distribution of acid-leached Mn is more complicated but is similar to the distribution of organic carbon in the upper 13-cm (Figure 5). There is a minimum at about 8 cm (also found in the distributions of acid-leached Cu, Fe and Ni). Above the minimum, Mn increases upward and reaches its highest value at the top. Several controlling factors may account for the distribution of acid-leached Mn in the sediment: (1) a marked amount of Mn is associated with the organic matter and/or carbonates as suggested by Figure 5. (2) there is a reduced zone of Mn at about 8 cm in which Mn oxides are reduced and are dissolved in the pore water; then upward and downward diffusion and subsequent precipitation of Mn oxides and $MnCO_3$ occurs; (3) anthropogenic Mn input has increased with time and has deposited

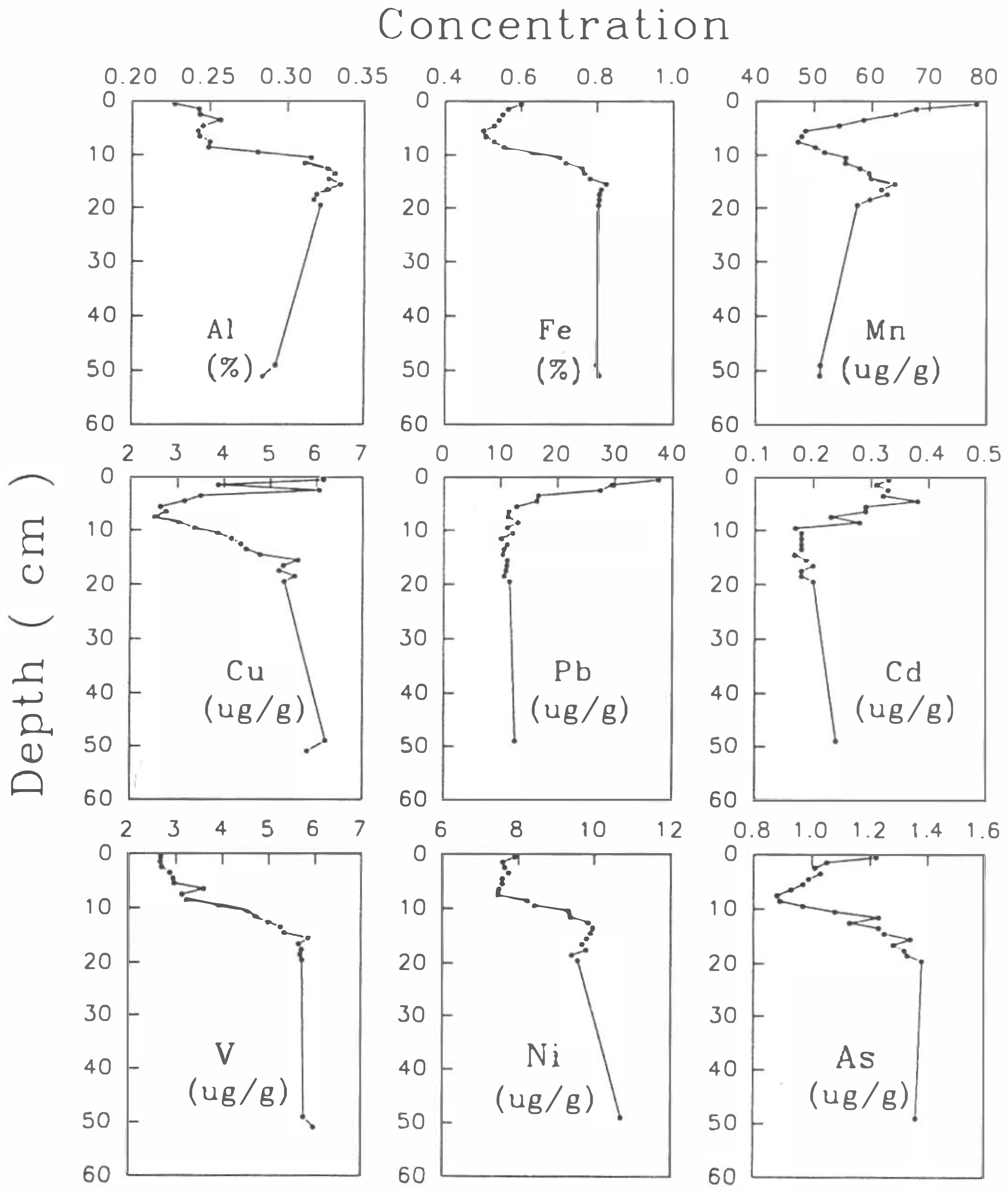

Fig. 3. Vertical distributions of acid-leached trace metals in the sediments of the Little Ghost Lake.

in the surface sediments (upper 8 cm); and (4) below the minimum, the distribution of acid-leached Mn is similar to acid-leached Al (Figure 3) and is probably controlled by the content of the clay-size sediment, which is the major phase for metal association in sediment and contains not only clay minerals but also hydrous oxides, sulfides and organic substances. These factors also more or less influence the distributions of other metals shown in Figure 3. With the exception of Pb and Cd, the distributions of acid-leached metals are similar to each other under about 8 cm. This strongly suggests that there is a similar major factor controlling the distributions of acid-leached metals below 8-cm. These authors believe that the variation of clay-size sediment content, particularly Fe and Mn oxides, could be responsible for the largest part of the distribution.

There are three distribution types in the upper 8 cm (Figure 3): a slightly upward decrease (Al and V), a slightly upward increase (Fe and Ni), and a strongly upward increase (As, Cu, Mn, Cd and Pb). This reveals that there are different processes, association phases and sources for each metal in the upper sediment column. The upward increase in Cd and Pb was probably mainly caused by anthropogenic inputs. Increases in As, Cu, and Mn were probably due to factors (1) and (2), and, to a lesser extent, to factor (3) mentioned above. The slightly upward increase trends of Fe and Ni reflect either a higher amount of Fe oxides near surface or an upward diffusion and subsequent oxidation of reduced Fe. The upward decrease trends of Al and V probably resulted from the upward decrease in clay minerals.

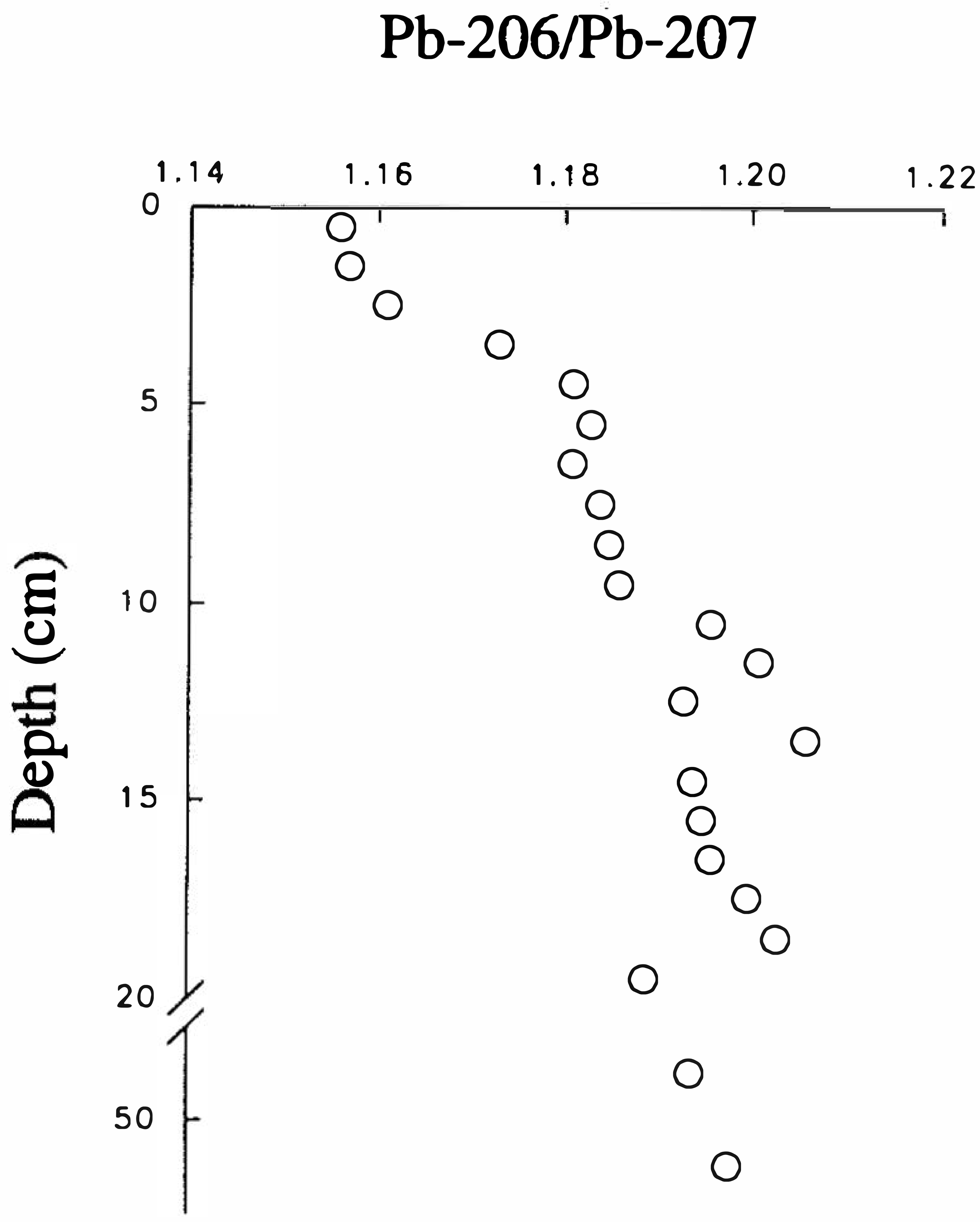

Fig. 4. Vertical distributions of Pb-206/Pb-207 ratios in the sediments of the Little Ghost Lake.

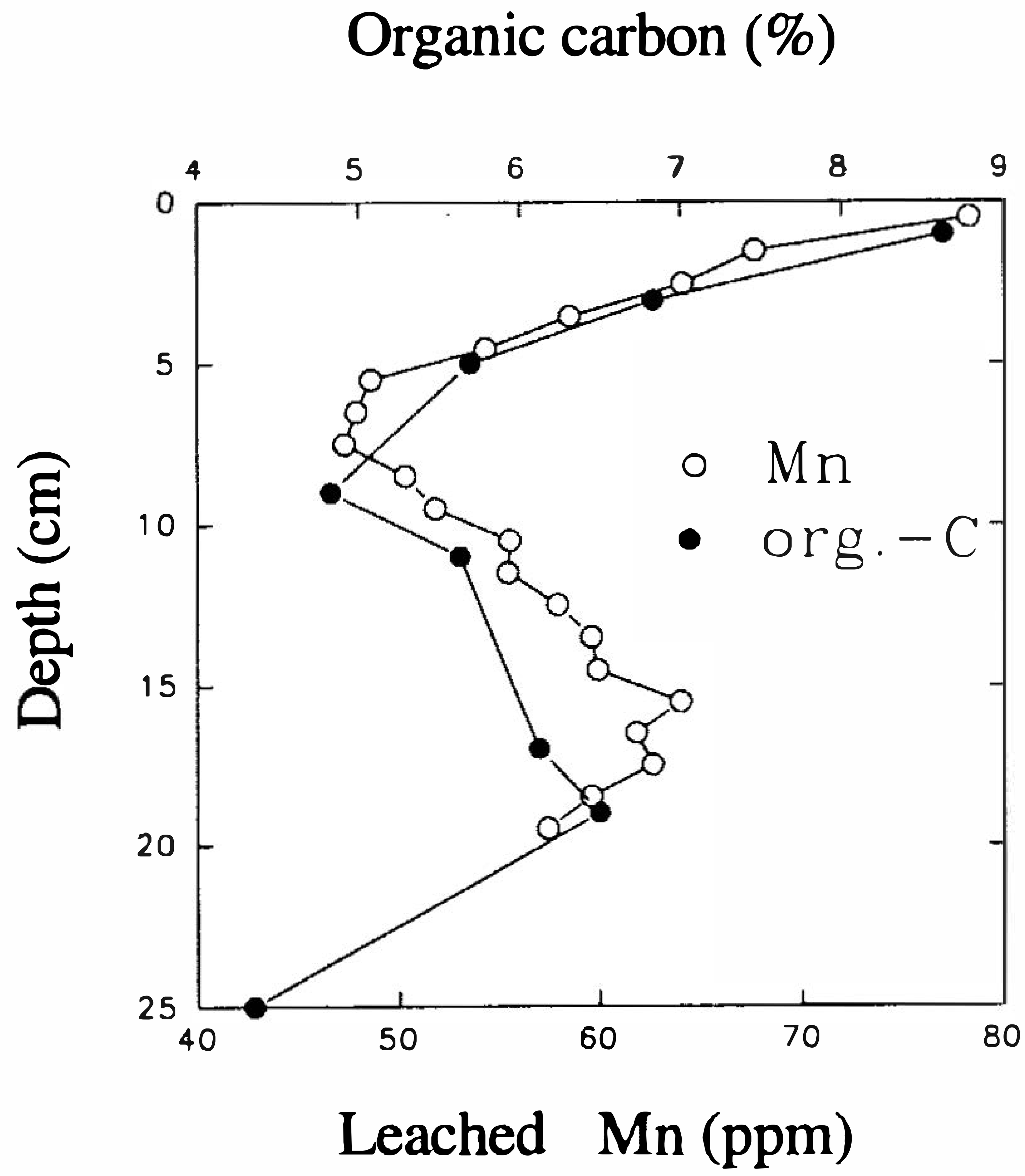

Fig. 5. Vertical distributions of acid-leached Mn and organic carbon (derived from Lin, 1992) in the sediments of the Little Ghost Lake.

The ranges of sedimentation rates in the Little Ghost Lake are 0.043-0.072 cm/yr as suggested by C-14 dating, and 0.042-0.12 cm/yr as calculated from Pb-210 dating (Figure 6) (Lin, 1992; Lin and Chen, 1992). The range obtained from C-14 dating is similar to and is covered by the range obtained from Pb-210 dating. In this study, the authors choose the medium (0.072 cm/yr) of the range obtained from Pb-210 dating to represent the average sedimentation rate of the Little Ghost Lake. Average fluxes for whole sediments thus calculated are as follows (μg/cm^2/yr): Al, 1327; As, 1; Cd, 0.01; Cr, 7; Cu, 1.2; Fe, 586; Mn, 6; Ni, 0.7;Pb, 2.3; V, 3.6; and Zn, 2.8 (Figure 7).

Figure 8 shows the vertical distributions of acid-leached trace metals in the sediments of the Great Ghost Lake. Fe, Ni, Pb and Zn show apparent near-surface enrichment. The Pb-206/Pb-207 ratios confirm the anthropogenic origin of lead near surface (Figure 9). With the exception of winter time, the hypolimnion of the Great Ghost Lake is anoxic. The redox cycles of Fe and Mn are obvious in the lake (Wang, 1989; Chen and Wang, 1990; Wang and Chen, 1990; Chen *et al.*, 1993b). Most of the recycled-dissolved Fe in the anoxic hypolimnion was oxidized and then deposited on the surface sediment during the winter overturn (Feb. to early March). This caused considerable Fe enrichment in the surface sediment. The sediment core presented here was collected in late March at which time there had not been enough time for the growth of thick anoxic hypolimnion to cause a large release of Fe from surface sediment. Therefore, the near-surface enrichment of acid-leached Fe was still observed (Figure 8). Fe oxides are good scavengers to remove trace metals in aquatic

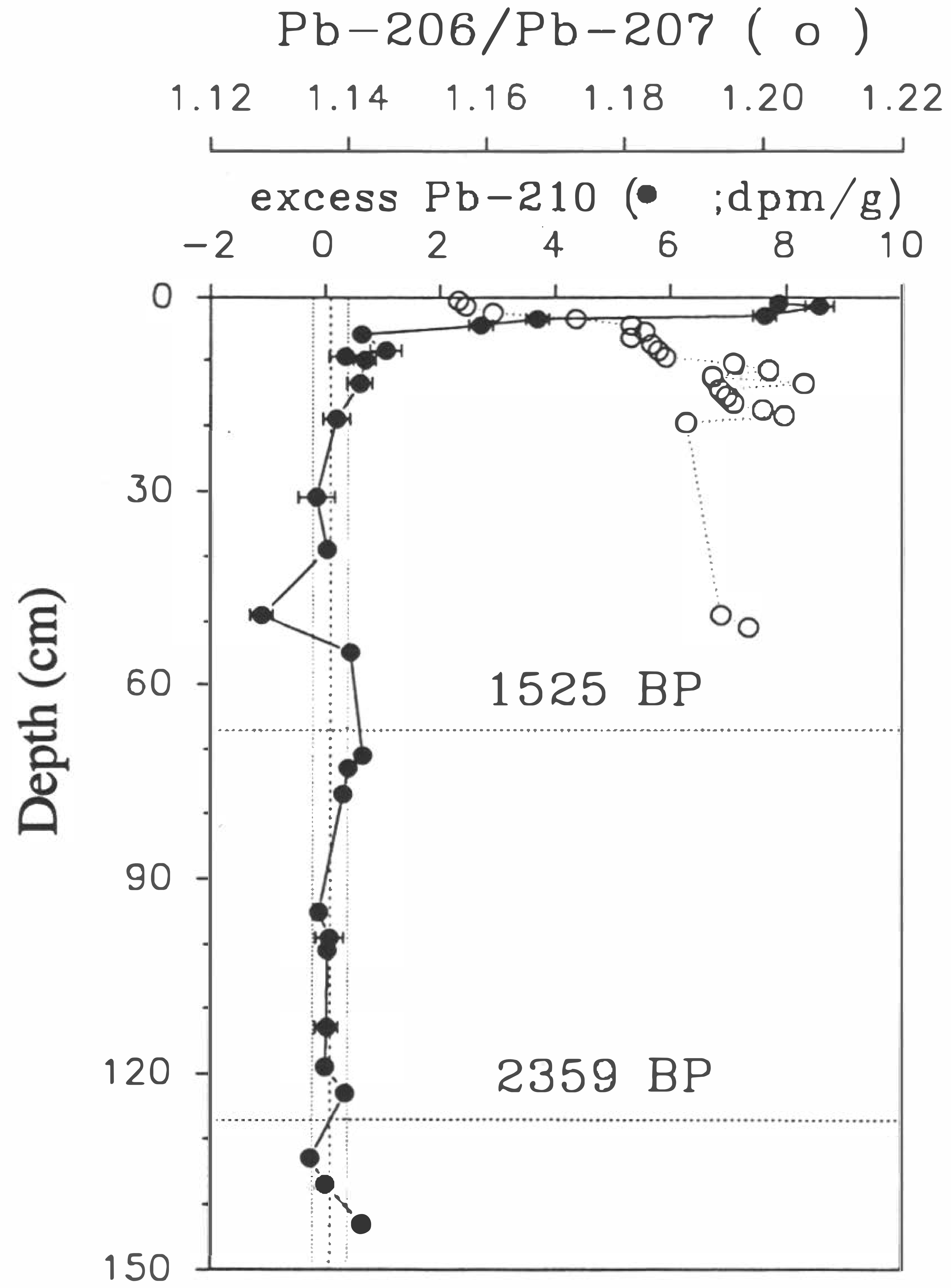

Fig. 6. Vertical distributions of excess Pb-210 activity and Pb-206/Pb-207 ratios in the sediments of the Little Ghost Lake. Two C-14 ages are marked.

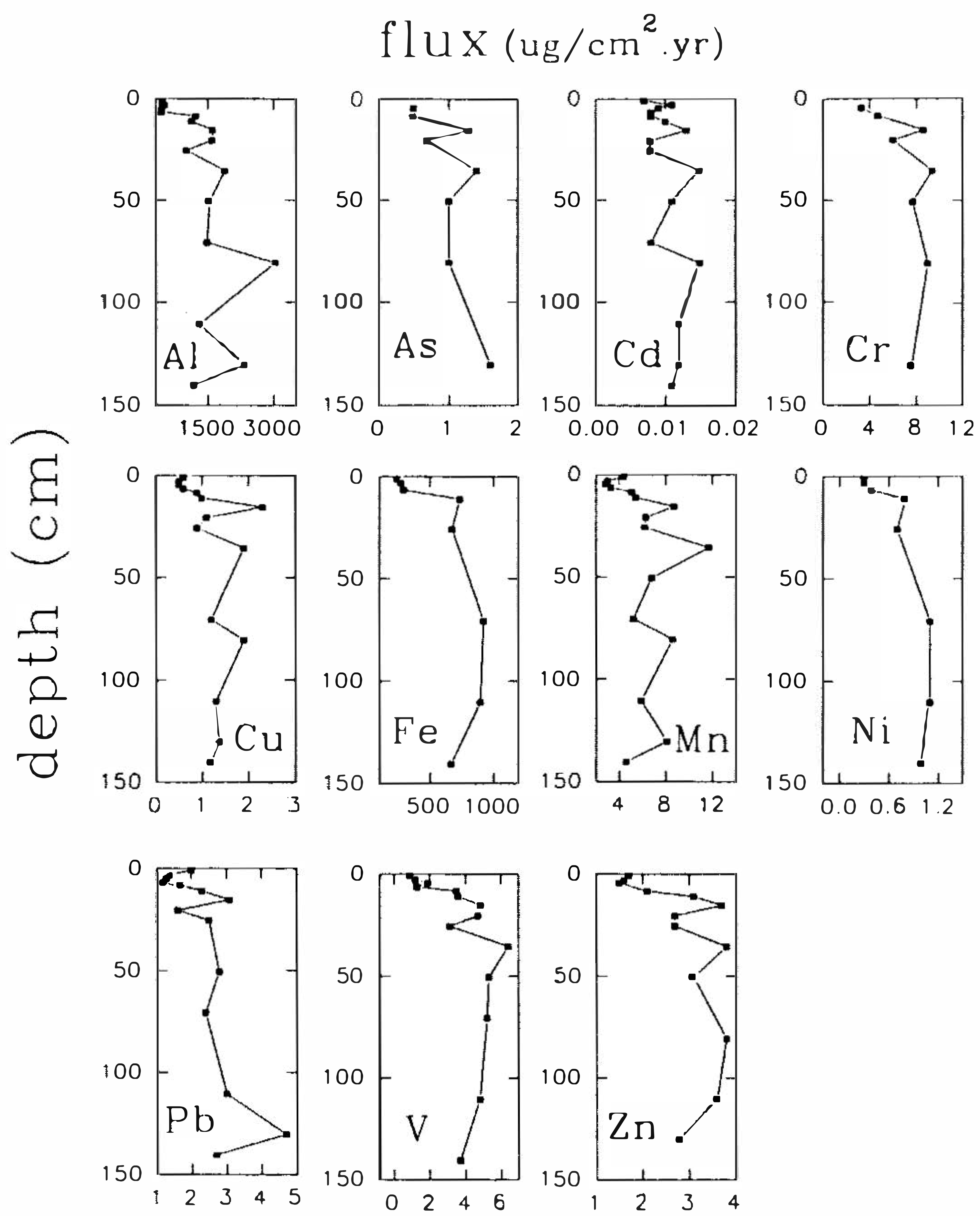

Fig. 7. Vertical distributions of trace metals fluxes in the sediments of the Little Ghost Lake based on the method of Hamilton-Taylor (1979).

systems. It is believed that the cause of the surface enrichments of Ni, Zn and part of Pb are similar to the Fe enrichment. The upward increase trend of acid-leached Fe from 23 cm to near surface of the sediment column probably resulted from the continuously upward migration and diffusion of reduced Fe.

According to C-14 dating, the sedimentation rate of the Great Ghost Lake was about 0.052 cm/yr (Figure 9). The acid-leached Pb and the Pb-206/Pb-207 ratios as a function of the dates of deposition in the Little and Great Ghost Lakes are shown in Figure 10. It is evident that the upward increase in the acid-leached Pb was consistent with the upward decrease in the Pb-206/Pb-207 ratio and was mainly caused by the upward increase in the anthropogenic Pb input. The onset of a small anthropogenic Pb increase in Little Ghost Lake was in about 1900, a little ahead of the increase (1945) in the Great Ghost Lake. Other studies show similar results (Bruland *et al.*, 1974; Skei and Paus, 1979; Shirahata *et al.*, 1980; Ng and Patterson, 1982; Finney and Huh, 1989; Giblin *et al.*, 1990). The relatively recent onset of an anthropogenic Pb increase in the Great Ghost Lake was probably caused by the underestimation of deposition dates and/or by the disturbance of the Fe cycle. The onset of the sharp anthropogenic Pb increase in the Little Ghost Lake occured in about 1940. The worldwide lead emissions from automobiles since 1940 have probably been important in these sharp increases in these two subalpine lakes.

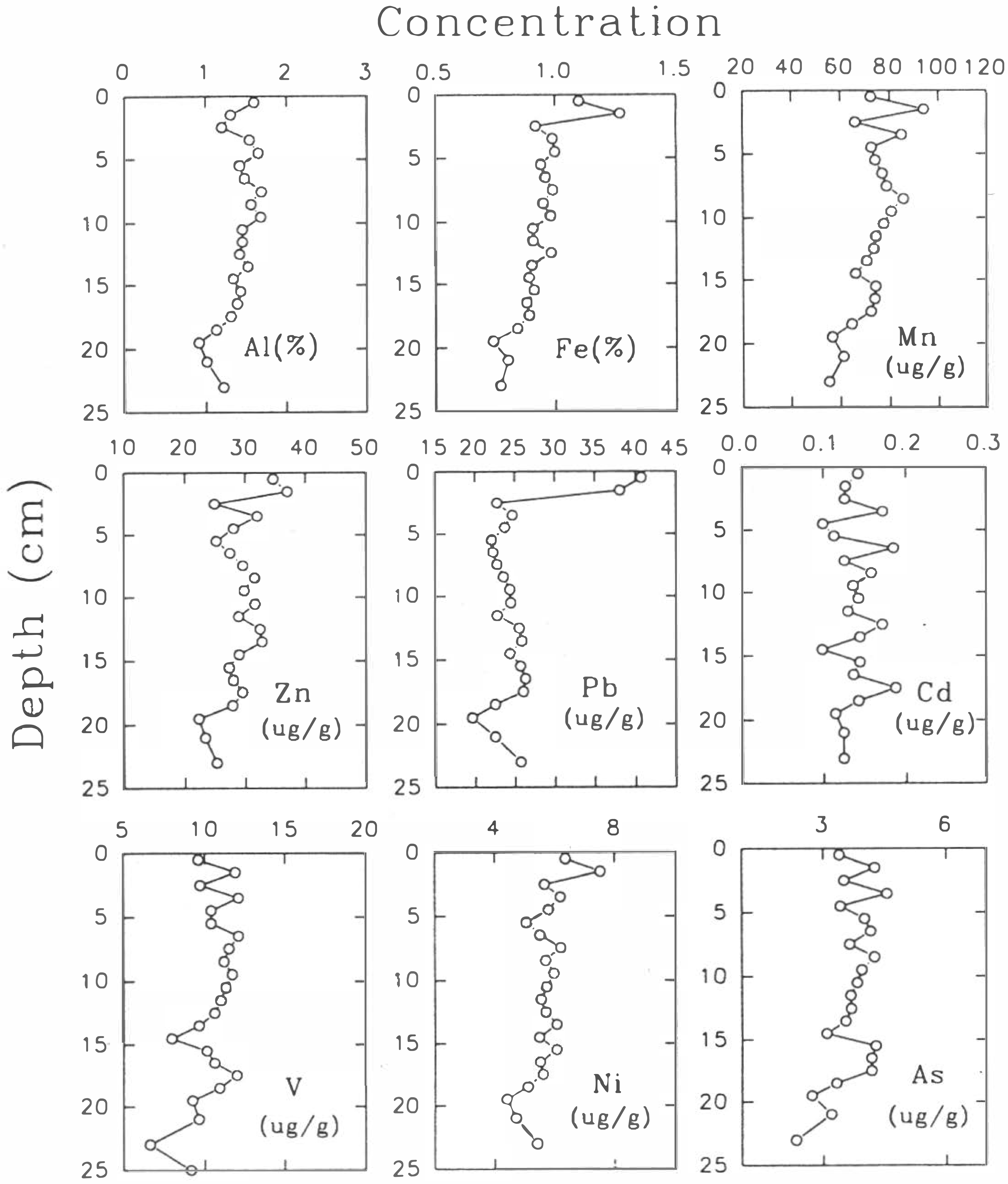

Fig. 8. Vertical distributions of acid-leached trace metals in the sediments of the Great Ghost Lake.

Since both lakes are located in remote subalpine regions, the anthropogenic metals there must have come from atmospheric fallouts. The high solubility of trace metals in atmospheric particulates was previously noted (Wallace *et al.*, 1977; Hodge *et al.*, 1978; Crecelius, 1980; Ochs and Gatz, 1980; Gatz *et al.*, 1982). It follows that most of the anthropogenic Pb in the sediment of the Great Ghost Lake was dissolved from atmospheric particulates and then removed by Fe oxides. The Fe oxides were mainly fornned by recycled Fe (reduced) during the winter overturn of the water column. In the shallow Little Ghost Lake, the atmospheric Pb may sink directly with the particulates and/or follow the way described for the Great Ghost Lake. However, based on the similar distributions of organic carbon and acid-leached Pb in the upper 8-cm of the sediment column, it seems reasonable that the scavengers of dissolved Pb in the Little Ghost Lake were probably aquatic organisms.

4. CONCLUSION

Distributions of metal/Al (whole-sediment) and acid-leached metals in the sediments of the Little Ghost Lake suggest that the surface enrichments of Cd and Pb (probably also Cr and Zn) have been caused mainly by anthropogenic inputs. The low Pb-206/Pb-207 ratios of surface sediments confirm the anthropogenic inputs of Pb. Based on Pb-210 and C-14

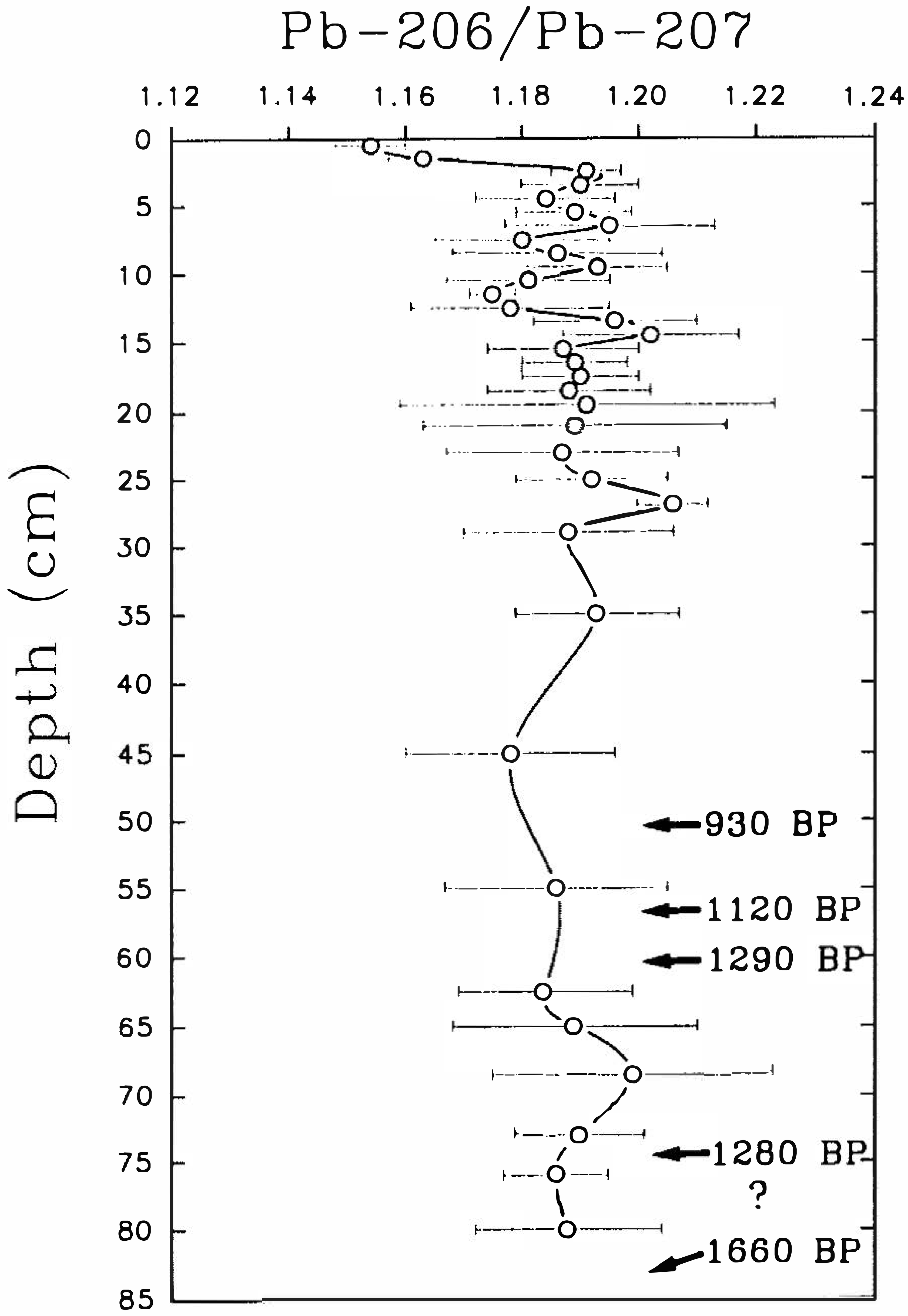

Fig. 9. Vertical distributions of Pb-206/Pb-207 in the sediments of the Great Ghost Lake. (The C-14 ages are marked)

datings, the recent onsets of small and large anthropogenic Pb increases took place in about 1900 and 1940, respectively.

The obvious surface enrichment of acid-leached Fe in the sediment and the prevailing Fe cycle in the water column suggest that the surface enrichments of acid-leached Fe, Ni, Zn and part of Pb in the Great Ghost Lake were due to a natural process. The process was driven by the Fe redox cycle and was able to remove trace metals in the water column. The low Pb-206/Pb-207 ratios of surface sediments confirm that most of the acid-leached Pb in the surface sediments of the Great Ghost Lake was anthropogenic in origin. C-14 dating indicated that the onset of the large anthropogenic Pb increase was in about 1945, probably caused by the worldwide lead emission from automobiles.

The Fe oxides produced during the winter overturn of the water column played a major role in removing the atmospheric anthropogenic metals, particularly Pb, in the Great Ghost Lake. In the shallow Little Ghost Lake, the anthropogenic metals may directly sink with atmospheric particulates and/or sink with aquatic organisms after dissolution from the atmospheric particulates.

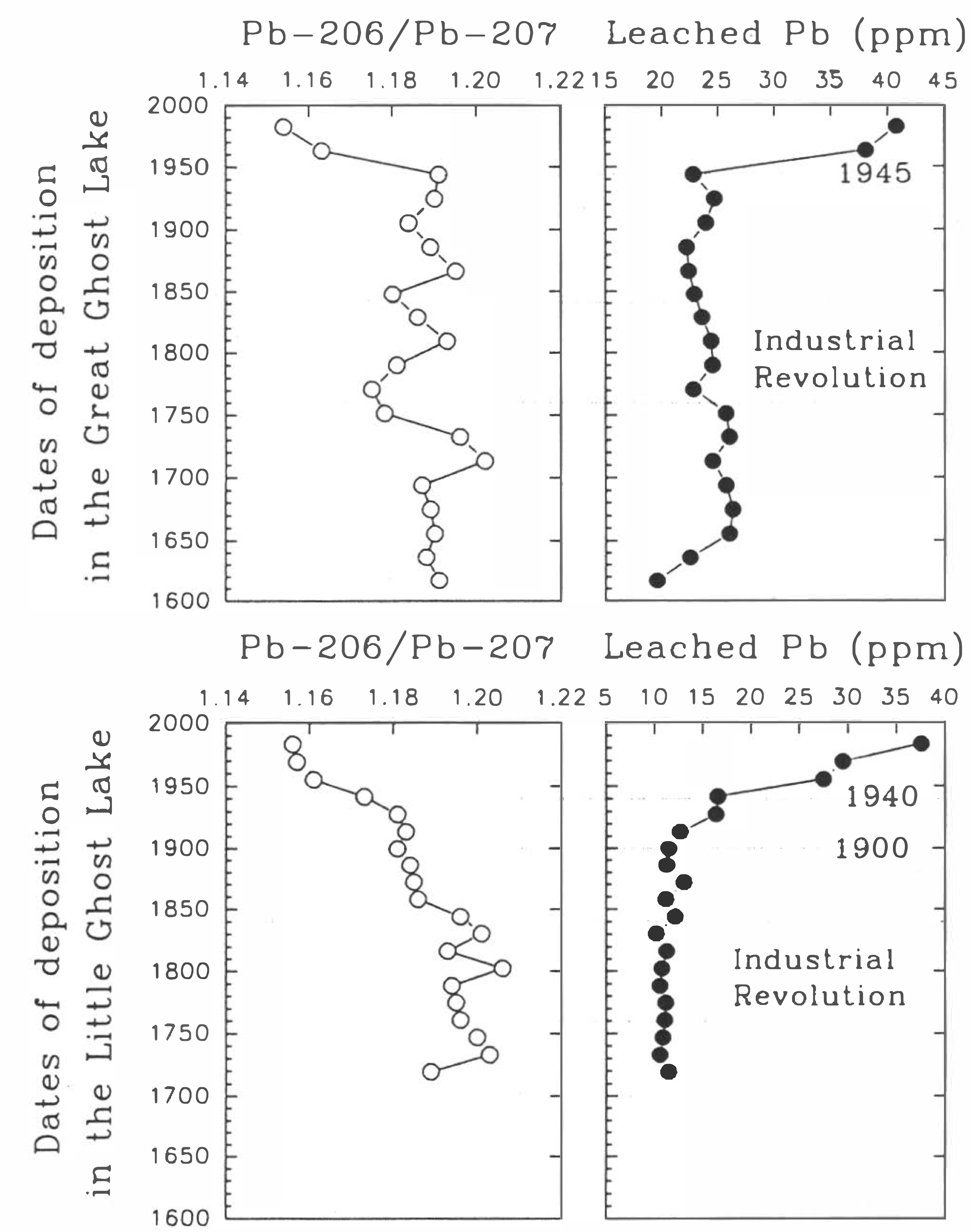

Fig. 10. The Pb-206/Pb-207 and Pb leached from the sediments of the Great Ghost Lake and the Little Ghost Lake as a function of the dates of deposition.

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