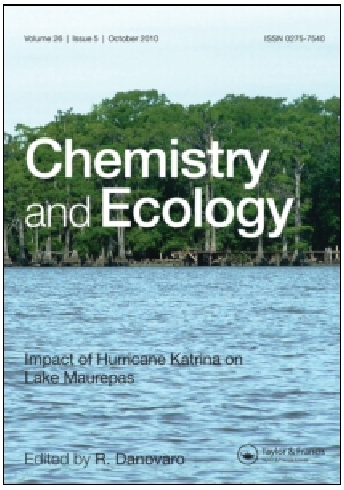

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Distribution of Trace Metals in the Surface Sediments of Two Pristine Subalpine Lakes in Taiwan

Jen-Kang Wann[a]; Chen-Tung Arthur Chen[a]
[a] Institute of Marine Geology, National Sun Yat-Sen University, Kaohsiung, Taiwan, R.O.C.

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DISTRIBUTION OF TRACE METALS IN THE SURFACE SEDIMENTS OF TWO PRISTINE SUBALPINE LAKES IN TAIWAN

JEN-KANG WANN and CHEN-TUNG ARTHUR CHEN

Institute of Marine Geology, National Sun Yat-Sen University,
Kaohsiung, Taiwan, R.O.C.

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Sediment cores were collected from two remote subalpine lakes: the rather shallow (1.5 m) Little Ghost Lake at 2040 m elevation and the deeper, Great Ghost Lake (40 m) at 2150 m elevation. Different early diagenesis of metals were observed. The seasonally anoxic hypolimnion drives the annual iron redox cycle and causes the remobilization of metals in the mobile fractions of sediments in the Great Ghost Lake. These result in the redistribution of metals in the mobile fractions of sediment and poor correlation between metal concentrations. In the Little Ghost Lake, the shallow water column is always oxic with less change of metals in the mobile fractions of sediments. As a result, most metals have strong positive correlations with each other.

The distributions of metal/Al ratios, total metal contents and acid-leached metal concentrations in the near-surface sediments of these two lakes suggest that the anthropogenic inputs from the atmospheric fallouts in recent decades are one of the major factors affecting the distributions of metals such as cadmium, lead and, to a lessor extent, zinc. The Pb-206/Pb-207 ratios confirm the anthropogenic lead input in these surface sediments.

KEY WORDS: Limnology, trace metals, sediments, diagenesis, Taiwan.

1. INTRODUCTION

For a number of years the authors have been studying the effect of acid rain on lakes in Taiwan. With a strong interest in the remobilization of trace metals from the sediment, it is necessary to understand the factors affecting their distributions in the lake sediments (Chen *et al.*, 1988, 1993a, 1994; Wann *et al.*, 1995; Wann and Chen, 1995).

The Earth's atmosphere is known to transport both natural and anthropogenically mobilized trace elements. Anthropogenic activities, such as the burning of fossil fuels, wastes, and leaded gasoline, have increased the flux of certain elements to the atmosphere by up to several orders of magnitude (Lantzy and Mackenzie, 1979; Li, 1981; Salomons and Forstner, 1984). The input of atmospheric anthropogenic matters plays a major role in controlling the distribution of certain trace metals, especially lead, in the aquatic sediments (Schaule and Patterson, 1981, 1983; Patterson and Settle, 1987; Salomons and Forstner, 1984).

Water chemistry, early diagenesis, variable input mechanisms and the preservation of sediments are also major factors regulating the distributions of trace metals in the aquatic sediments. The most intensive early diagenesis reactions are usually found where variations of redox condition take place (Salomons and Forstner, 1984). The subject of our study, the Great Ghost Lake, is seasonally stratified and anoxic (Chen and Wang, 1990; Wang and Chen, 1990). The hypolimnion is anoxic through most of the year and may cause marked anaerobic mineralization processes in the hypolimnion (Chen and Wang 1990; Wang and Chen, 1990) and sediment, and subsequently affect the distribution of elements. Distinctive variations of well preserved organic carbon are also found in the sediment of the Great Ghost Lake (Wann and Chen, 1995). In contrast, the shallow water column of the Little Ghost Lake is always oxic (Lin and Chen, 1992) with the relatively short retention time of dissolved and particulate metals. The differences between water chemistries, sediment redox conditions and preservations of organic matters in these two lakes give us a chance to compare different factors in regulating distribution of trace metals in lake sediments.

Comparing the relative abundance of elements with respect to aluminium, or leaching the sediments with dilute acid are two methods of reducing the grain-size effects, in order to differentiate the natural or anthropogenic sources (Kemp and Thomas, 1976; Kemp *et al.*, 1976; Ng and Patterson, 1982; Forstner and Wittmann, 1983; Finney and Huh, 1989). Iron and manganese oxides/hydroxides, the surface of clay minerals, organic matters and carbonates are the major mobile fractions and the major phases of metal association in the sediments. Acid-leached metals measured here represent most of the metals in these phases but do not include those within in the lattice of clay minerals and in rock debris. The latter are of natural origin and are not changed by the anthropogenic inputs or the early diagenesis.

In this paper, metal concentrations and Pb-206/Pb-207 ratios in the leachate, total metal contents, metal/Al and acid-leached metal/total metal ratios, and correlations between metals in the sediments are reported in order to understand the factors affecting the distributions of trace metals in these two subalpine lakes in Taiwan. This is a follow up of our preliminary investigation (Wann *et al.*, 1995) which was based on fewer elements at much coarser vertical spacing.

2. MATERIALS 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 occurred because the maximum depth is only 1.5 m and the lake is rich in fish and other biota.

The Great Ghost Lake (Ta-Kuei Hu), 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 through most of the year (Chen and Wang, 1990; Wang and Chen, 1990). Little or no wind mixing or bioturbation is expected to have occurred.

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.*, 1993b). The near-surface sediments (< 25 cm) in the cores are studied here including one core (140 cm) from the Little Ghost Lake and two cores (82 cm and 93 cm) from the Great Ghost Lake. The cores collected from the Great Ghost Lake show signals of climatic changes (Chen *et al.*, 1993b; Lou *et al.*, 1995).

In the leaching procedure, the dried sediment (0.3 g) was treated with 15 ml of 1.6N nitric acid and was 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 dried sediment (0.3 g) for the determination of the total metal content was digested with an acid mixture (40% HNO_3, 15%$HClO_4$ and 20% HF) in a pressure bomb or was 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 $\min^{-1}$. Carbon-14 dating was done by both conventional and AMS methods, Pb-210 dating was done using the method of Chung and Craig (1983) (Lin and Chen, 1992; Chen *et al.*, 1993b). The total carbon was measured by the LECO CS-244 element analysis system (Chen *et al.*, 1993b).

The National Institute for Environmental Studies No.2 Pond Sediment was used to test the accuracy and precision of the trace metal determinations. Most of the accuracies (the relative error) are better then 10%. In every set of sample treatment and metal determination, the NIES sediment was included. A correction was made only when the accuracy (the relative error) of the NIES sediment determination was worse than 10% (Wann *et al.*, 1995). The precision (the relative standard deviation) for the measurements of Al, As, Cd, Ce, Cr, Cs, Cu, Fe, Mg, Mn, Ni, Pb, Rb, Sr, V and Zn are roughly ± 15.9, 10.7, 7.3, 9.5, 8.4, 7.1, 3.2, 5.3, 4.5, 4.1, 2.1, 10.5, 4.1, 11.5, 1.7 and 3.8%, respectively. The accuracy of the Pb-206/Pb-207 determination was calibrated by the NIST SRM981 but we did not have a reference sediment to check for the bias of the sample preparation. The precision of the isotope ratio measurement was roughly $\pm 1\% (n = 5)$.

3. RESULTS AND DISCUSSION

The vertical distributions of metal concentrations in the near-surface sediments of the Little Ghost Lake are given in Figure 1 for Al, As, Cd, Ce, Cr, Cs, Cu, Fe, Mg, Mn, Ni, Pb, Rb, V and Zn. It can be seen that the average concentrations of aluminium and iron near the surface are about 30–40% less than those below. Aluminium and iron are the main elements in the earth crust and are the dominant inorganic elements in the sediment matrix (Kemp *et al.*, 1976). Aluminium also has a strong positive relationship with clay in the sediments (Kemp *et al.*, 1976; Lin and Chen, 1992). The lower concentrations of aluminium and iron near the surface are probably caused by the dilution of organic matter (about 10–20%), carbonates (about 10%) and quartz, which usually contain low metal concentrations. This near-surface dilution also results in a lower content of clay-size (< 4 μm) sediment (Wann *et al.*, 1995), which has a relatively large surface area to enrich metals. These factors also led to the lower concentrations of Ce, Cr, Cs, Cu, Mg, Ni, Rb and V in the surface sediments.

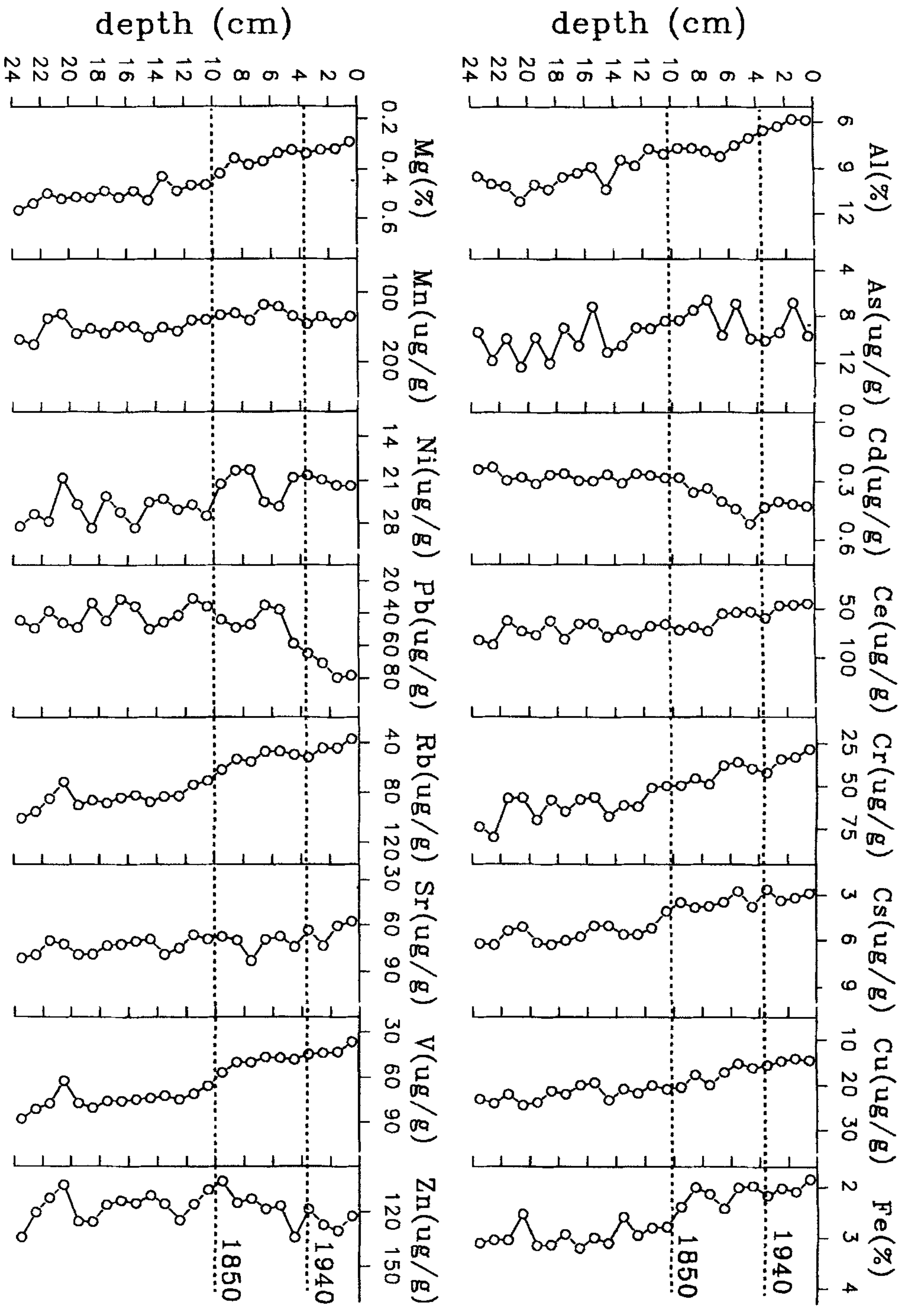

Figure 1 The distributions of metal concentrations in the surface sediments of the Little Ghost Lake.

The regression coefficients (*R*) between these metals in the surface sediments are shown in Table I. Ce, Cr, Cs, Cu, Mg, Rb and V have strong positive correlations with aluminium and iron. This suggests that, like aluminium, these metals mainly come from the lithogenic sources, and their distributions are almost unaltered in the early diagenesis. Arsenic, manganese, nickel and strontium show weak positive correlations ($R < 0.6$) with aluminium, probably because of the other source inputs and/or redistributions of these metals in the mobile fractions of the sediments in the early diagenesis. The distributions of acid-leached arsenic and manganese are very similar to that of organic carbon in the surface sediments (above 20 cm depth). This suggests that large amounts of arsenic and manganese are associated with organic matters (Wann *et al.*, 1995).

Figure 1 shows that cadmium and lead (probably also zinc) are enriched in the surface sediments of the Little Ghost Lake. They also have negative correlations with aluminium. The vertical distributions of metal/Al ratios in the sediments of this lake are shown in Figure 2. Also seen are the marked surface enrichments of cadmium, lead and zinc, which may have been caused by the anthropogenic inputs. These metals have significantly enriched in the atmospheric particles because of the increases in industrial activities and the use of lead alkyls in the gasoline (Kemp *et al.*, 1976; Lantzy and Mackenzie, 1979; Forstner and Wittmann, 1983; Salomons and Forstner, 1984). In order to confirm this, the vertical distributions of acid-leached lead and the Pb-206/Pb-207 ratios in the sediments (Fig. 3) were measured (Chen *et al.*, 1993b; Wann *et al.*, 1995). The anthropogenic source of lead is relatively easy to identify as most lead in the ores have Pb-206/Pb-207 ratios measurably less than the natural lead of soil related components (Shirahata *et al.*, 1980). The higher acid-leached lead concentrations but lower Pb-206/Pb-207 ratios near surface (Fig. 3) confirm that the surface enrichment of lead was mainly caused by anthropogenic inputs, especially the widespread lead emissions from automobiles since about 1940. The marked surface enrichments of acid-leached cadmium and lead (acid-leached zinc was not measured) are also found (Wann *et al.*, 1995).

The vertical distributions of ratios of acid-leached metal/total metal for Al, As, Cd, Cu, Fe, Mn, Ni, Pb and V are shown in Figure 4. These ratios are relatively constant throughout the core, with the exceptions of higher ratios for copper, iron, manganese and lead near the surface. These reveal that the association phases of metals and the sediment composition have not changed very much. Instead, the anthropogenic input and the early diagenesis are involved to increase the metal content in the mobile fractions and/or to change the association phases of copper, iron, manganese and lead in the upper sediments. Iron and manganese are redox-sensitive elements. The upward migrations of reduced iron and manganese in the pore water are usually observed in the anoxic sediments and may have caused the high ratios of acid-leached metal/total Pb mainly result from increase of the anthropogenic lead, which is usually associated with the mobile fractions of sediment (Forstner and Wittmann, 1983; Salomons and Forstner, 1984). The surface enrichments of total cadmium, Cd/Al and acid-leached cadmium are probably also caused by the anthropogenic input. But, since a large amount of cadmium ($>60\%$) is associated with the mobile fractions of sediment (Fig. 4), this will diminish the signals of anthropogenic input observed in the distribution of ratios of acid-leached Cd/total Cd shown in Figure 4.

Table I The regression coefficients (*R*) between metal concentrations in the surface sediments of the Little Ghost Lake.

	Al	*As*	*Cd*	*Ce*	*Cr*	*Cs*	*Cu*	*Fe*	*Mg*	*Mn*	*Ni*	*Pb*	*Rb*	*Sr*	*V*	*Zn*
Al	1	0.562	−0.749	0.724	0.831	0.825	0.906	0.817	0.903	0.475	0.535	−0.618	0.835	0.564	0.820	−0.278
As		1	−0.301	0.259	0.437	0.499	0.474	0.423	0.468	0.343	0.195	−0.085	0.429	0.242	0.399	−0.004
Cd			1	−0.817	−0.840	−0.802	−0.876	−0.828	−0.898	−0.620	−0.554	−0.568	−0.856	−0.442	−0.856	0.358
Ce				1	0.916	0.731	0.888	0.670	0.794	0.673	0.281	−0.477	0.808	0.610	0.756	−0.271
Cr					1	0.901	0.921	0.857	0.918	0.814	0.539	−0.474	0.959	0.657	0.922	−0.089
Cs						1	0.846	0.890	0.904	0.728	0.633	−0.514	0.946	0.651	0.949	0.056
Cu							1	0.816	0.929	0.601	0.456	−0.559	0.885	0.604	0.858	−0.313
Fe								1	0.945	0.689	0.790	−0.623	0.941	0.448	0.952	−0.105
Mg									1	0.671	0.679	−0.620	0.955	0.533	0.953	−0.192
Mn										1	0.439	−0.025	0.784	0.458	0.732	0.265
Ni											1	−0.600	0.697	0.249	0.759	0.087
Pb												1	−0.554	−0.400	−0.617	0.431
Rb													1	0.588	0.990	−0.066
Sr														1	0.579	0.166
V															1	−0.048
Zn																1

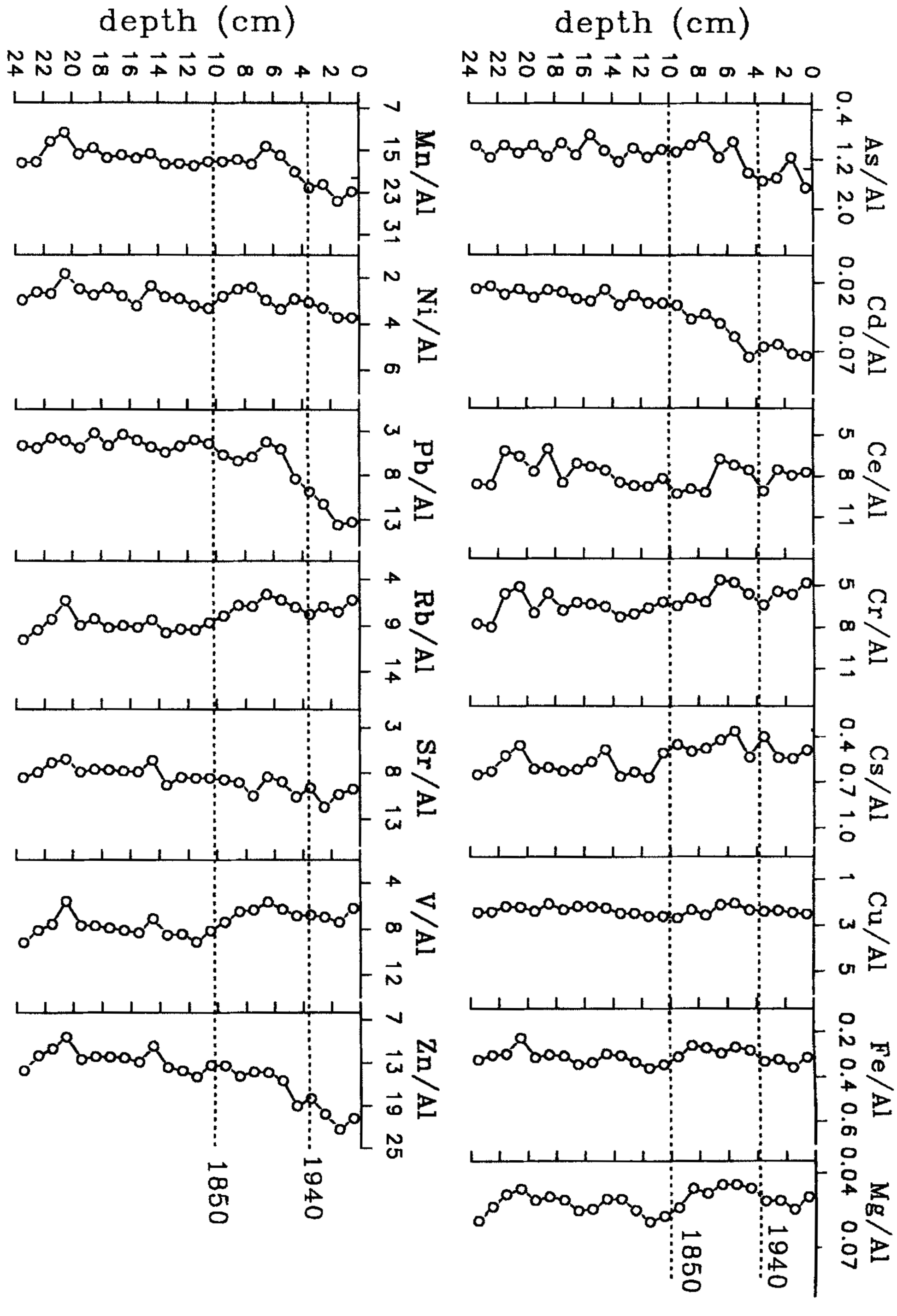

Figure 2 The distributions of metal/Al ratios (x 10000) in the surface sediments of the Little Ghost Lake.

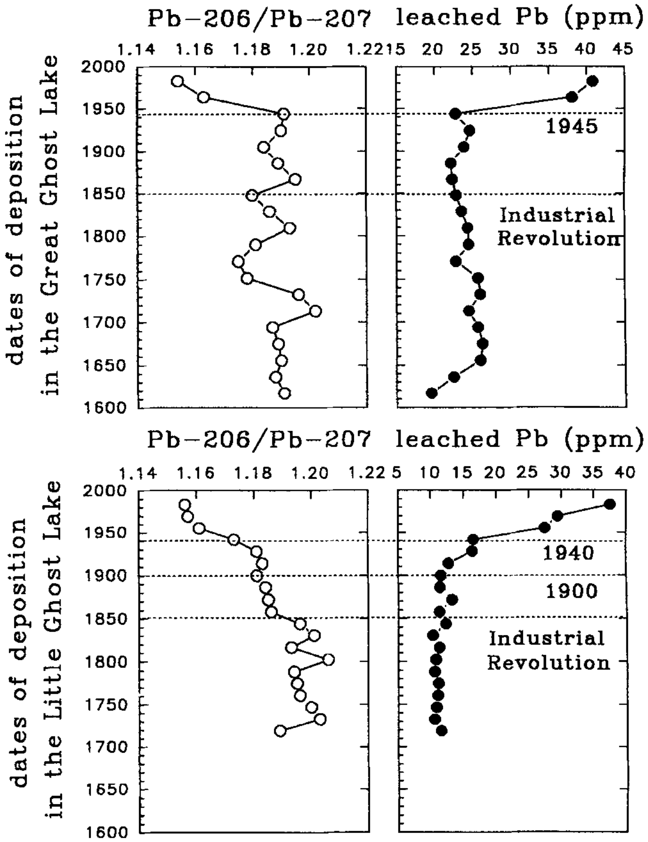

Figure 3 The Pb-2066/Pb-207 and lead leached from the sediments of the Great Ghost Lake and the Little Ghost Lake as a function of the dates of deposition (taken from Wann *et al.*, 1995).

Averages of the ratios of acid-leached metal/total metal for Al, As, Cd, Cu, Fe, Mn, Ni, Pb, and V are 0.036, 0.126, 0.690, 0.231, 0.263, 0.410, 0.371, 0.305 and 0.069, respectively. These ratios suggest that less aluminium and vanadium exist in the mobile fractions of the sediments, with little in the acid-leached phase. In contrast, more cadmium, manganese, nickel and lead are associated with the mobile fractions of the sediments, with more in the acid-leached phase. As a result, the mobility of metals in the sediments is in this order: Cd > Mn > Ni > Pb > Fe > Cu > As > V > Al. The probability of remobilization of metals from the sediments caused by leaching of lake water should the lake become acidified is also in this order.

The distribution of Al, As, Cd, Ce, Cr, Cs, Cu, Fe, Mg, Mn, Ni, Pb, Rb, V and Zn in the surface sediments in two cores (82 cm and 93 cm) collected from the Great Ghost Lake are shown in Figures 5 and 6. Only lead enriches significantly in the surface sediments of the 82 cm core (Fig. 5). In Figure 6, cadmium, manganese and lead (probably zinc, too) show slight surface enrichment, whereas the concentrations of other metals are nearly uniform vertically. Similar distributions are also found in the

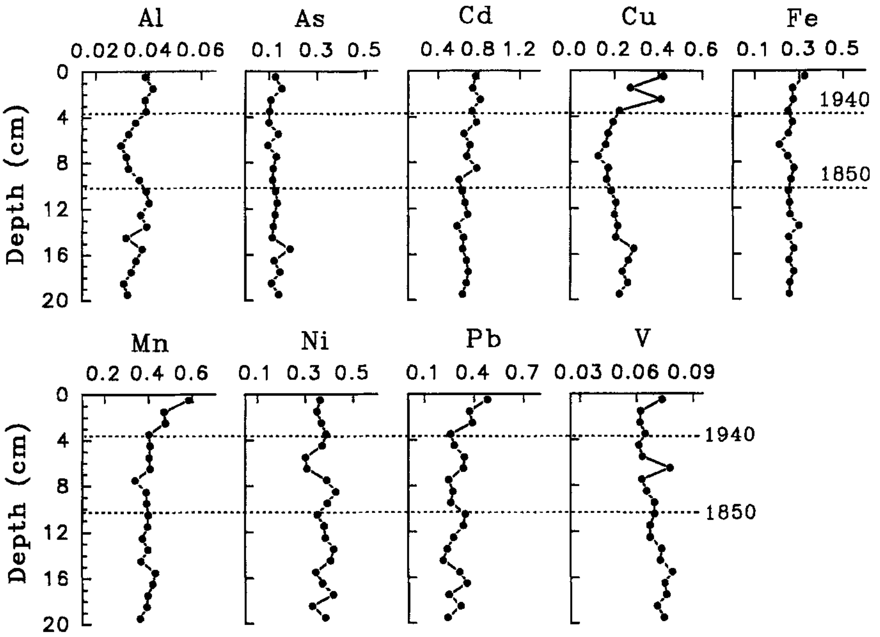

Figure 4 The distributions of ratios of acid-leached metal/total metal in the surface sediments of the Little Ghost Lake.

ratios of metal/Al in the same cores (Figs. 7 and 8). The Pb/Al ratios, which eliminate the grain size effect, again show an increase near the surface. The surface enrichment of lead caused by the anthropogenic input is also confirmed by the distributions of acid-leached lead and the Pb-206/Pb-207 ratio (Fig. 3), but the surface enrichments of cadmium and manganese shown in Figure 8 are not found in Figure 7.

The correlations between the metal concentrations in the 82 cm and 93 cm cores are shown in Tables II and III (organic TC is included), respectively. There are negative correlations ($R < -0.4$) between TC and caesium, magnesium, rubidium and vanadium (Tab. III), but positive correlations among caesium, magnesium, rubidium and vanadium ($R > 0.4$) (Tab. II and III). These metals also have low ratios of acid-leached metal/total metal (< 0.2; Fig. 9). These suggest that caesium, magnesium, rubidium and vanadium mainly came from the same lithogenic sources and have not changed much in the mobile fractions of the sediments during the early diagenesis. Although aluminium and strontium also have low ratios of acid-leached metal/total metal (< 0.2) in the sediments, their correlations with TC, magnesium and rubidium (Mg and Rb have strong negative correlations with TC throughout the 93 cm core; Wann and Chen, 1995) are not clear. These suggest that the mobile fractions of aluminium and strontium in the sediments have markedly changed in the early diagenesis and/or these metals came from different lithogenic sources.

The interactions between organic matters and the clay surface may also cause weak correlations between aluminium and TC and other lithogenic metals. There are no obvious correlations between other metals listed in both Tables II and III. This is

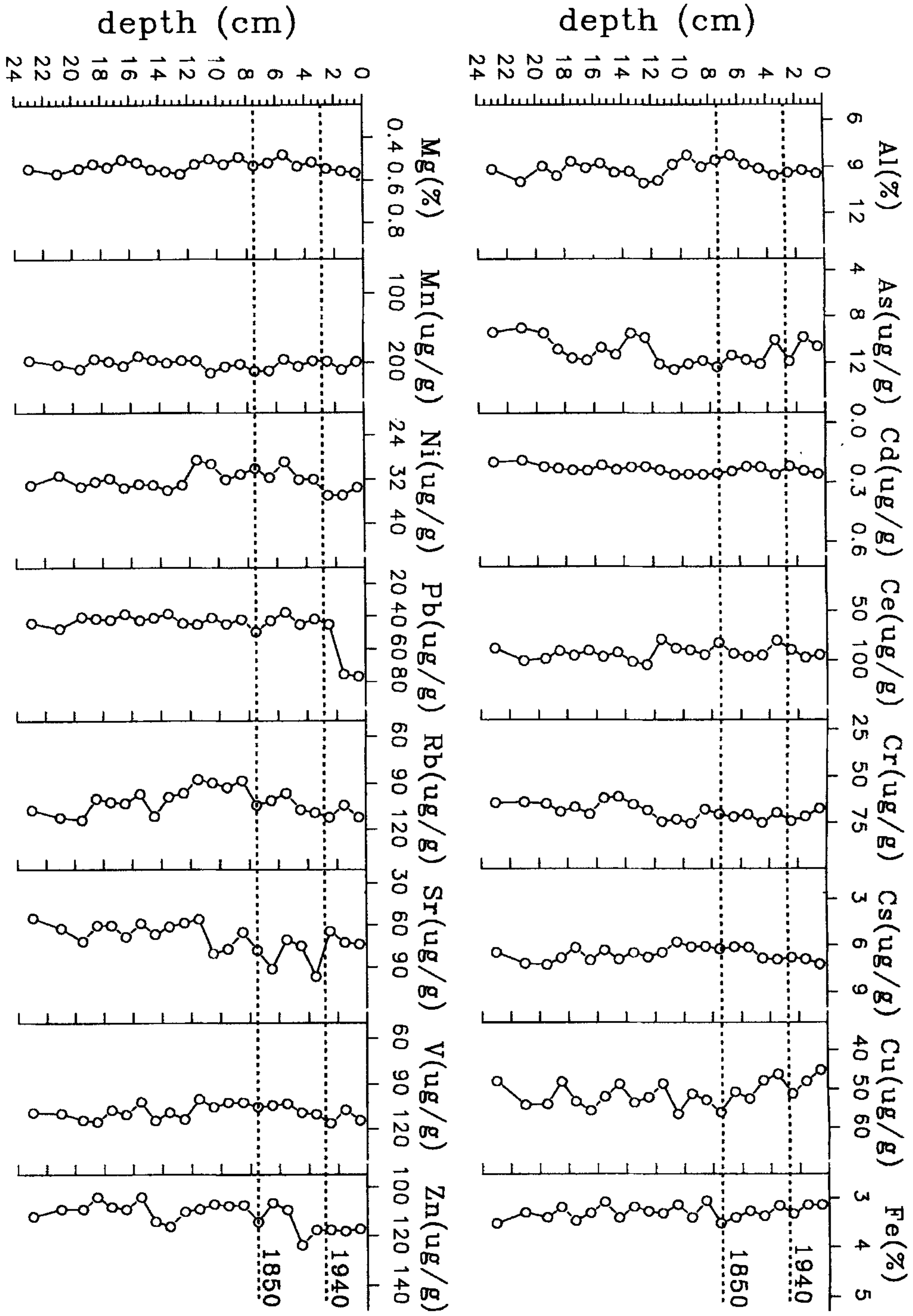

Figure 5 The distributions of metal concentrations in the surface sediments of the Great Ghost Lake (82 cm core).

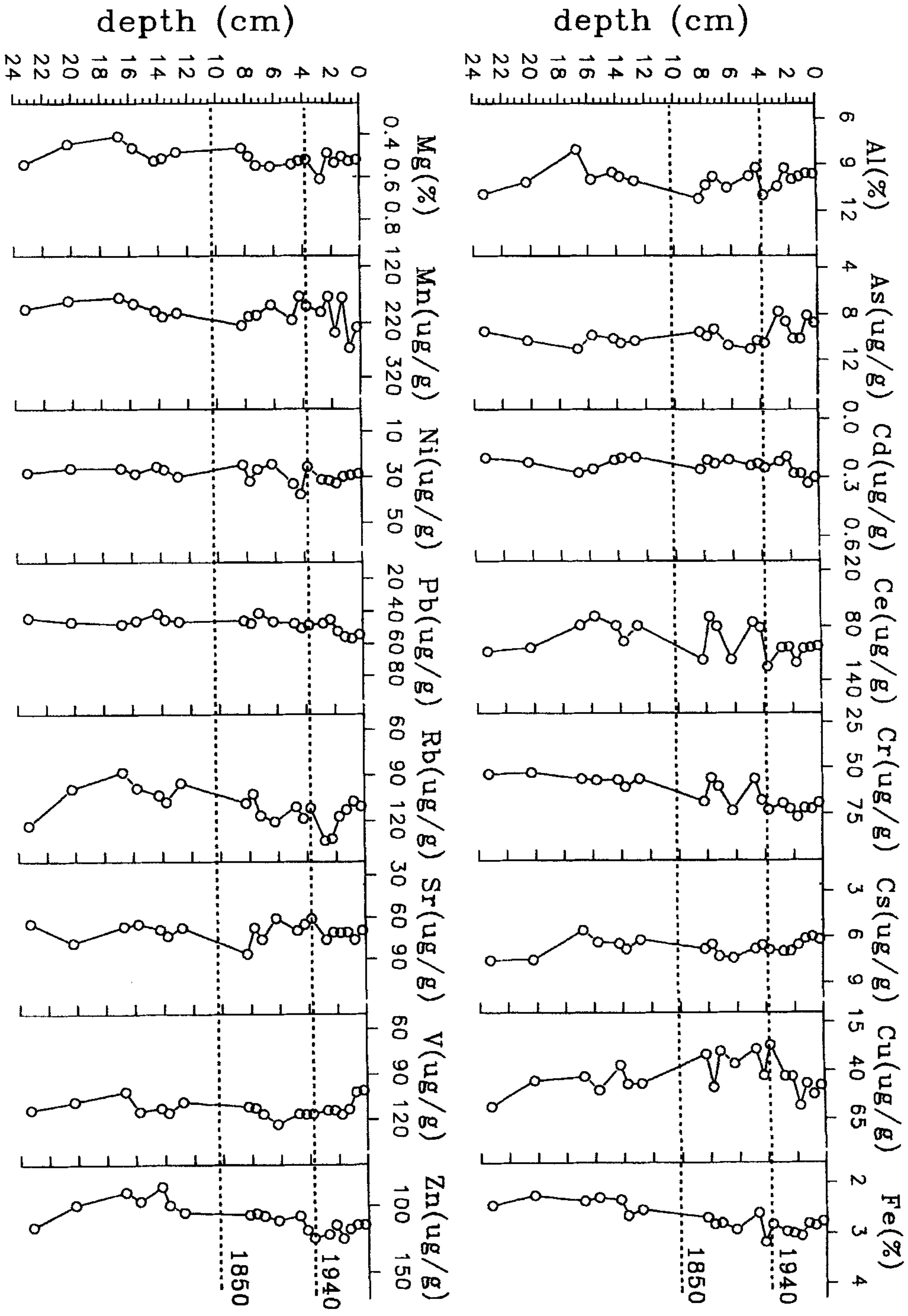

Figure 6 The distributions of metal concentrations in the surface sediments of the Great Ghost Lake (93 cm core).

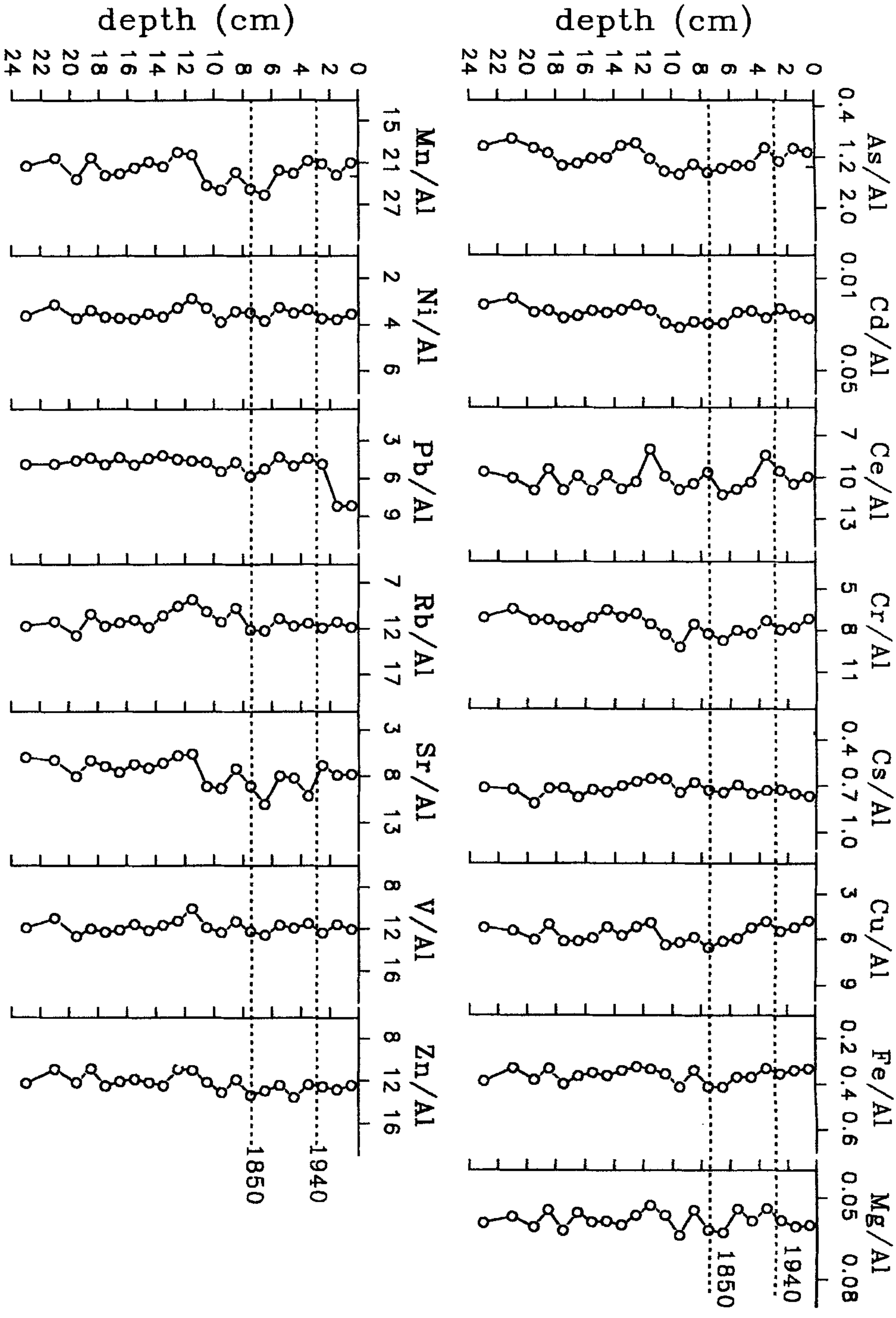

Figure 7 The distributions of metal/Al ratios (x 10000) in the surface sediments of the Great Ghost Lake (82 cm core).

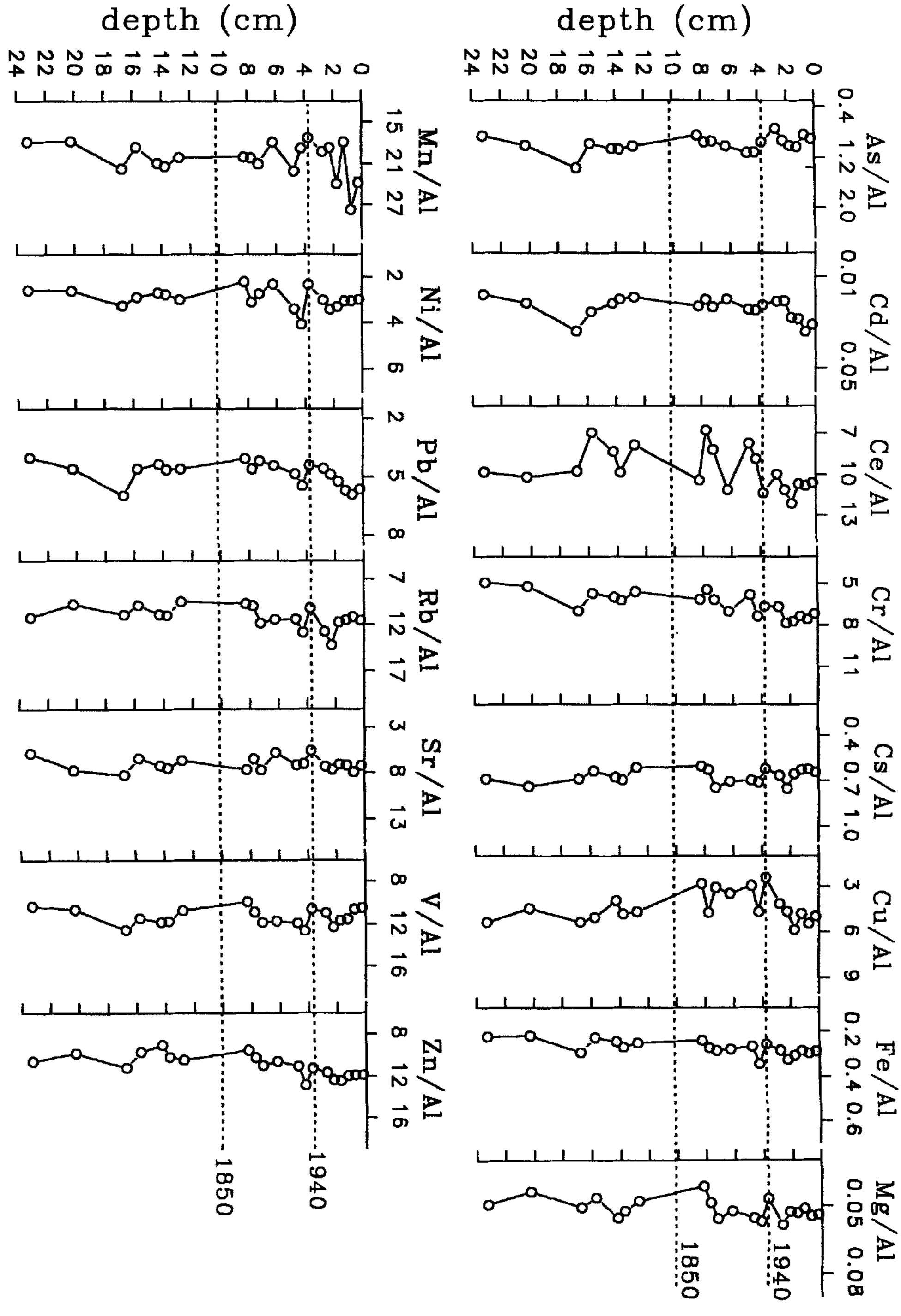

Figure 8 The distributions of metal/Al ratios (x 10000) in the surface sediments of the Great Ghost Lake (93 cm core).

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Table II The regression coefficients (R) between metal concentrations in the surface sediments (82 cm core) of the Great Ghost Lake.

	Al	*As*	*Cd*	*Ce*	*Cr*	*Cs*	*Cu*	*Fe*	*Mg*	*Mn*	*Ni*	*Pb*	*Rb*	*Sr*	*V*	*Zn*
Al	1	−0.452	−0.365	0.099	−0.188	0.608	−0.329	−0.273	0.471	−0.443	0.135	−0.148	0.182	−0.411	0.481	0.231
As		1	0.551	−0.482	0.618	−0.565	0.214	0.113	−0.606	0.221	−0.506	−0.190	−0.454	0.216	−0.408	−0.082
Cd			1	−0.450	0.437	−0.314	−0.054	−0.222	−0.368	0.399	−0.222	0.218	−0.327	0.607	−0.252	0.046
Ce				1	−0.401	0.202	0.197	−0.198	0.402	−0.078	0.403	0.084	0.119	−0.305	0.231	−0.050
Cr					1	−0.285	−0.048	0.041	−0.310	0.427	−0.280	0.101	−0.345	0.391	−0.272	0.209
Cs						1	−0.395	−0.033	0.592	−0.161	0.571	0.355	0.752	−0.074	0.743	0.423
Cu							1	0.118	−0.258	0.402	−0.268	−0.456	−0.281	−0.088	−0.252	−0.431
Fe								1	0.195	0.175	−0.070	−0.226	0.323	−0.040	0.095	0.072
Mg									1	−0.057	0.589	0.482	0.546	−0.285	0.568	0.409
Mn										1	−0.117	0.183	−0.001	0.569	−0.169	0.121
Ni											1	0.320	0.551	−0.139	0.632	0.360
Pb												1	0.272	−0.106	0.131	0.419
Rb													1	0.156	0.730	0.519
Sr														1	−0.090	0.248
V															1	0.357
Zn																1

Table III The regression coefficients (R) between total carbon (TC) and total metals in the surface sediments (93 cm core) of the Great Ghost Lake.

	TC	*Al*	*As*	*Cd*	*Ce*	*Cr*	*Cs*	*Cu*	*Fe*	*Mg*	*Mn*	*Ni*	*Pb*	*Rb*	*Sr*	*V*	*Zn*
TC	1	−0.075	0.381	0.583	0.073	0.010	−0.422	−0.001	−0.187	−0.709	0.138	−0.156	0.421	−0.659	0.133	−0.430	−0.111
Al		1	−0.119	−0.221	0.501	0.108	0.602	−0.116	0.055	0.346	0.189	−0.270	−0.168	0.291	0.127	0.412	0.383
As			1	−0.201	−0.197	−0.339	−0.042	−0.248	−0.324	−0.400	−0.381	−0.127	−0.190	−0.507	−0.422	0.285	−0.367
Cd				1	0.205	0.417	−0.581	0.146	0.102	−0.150	0.512	0.004	0.772	−0.231	0.191	−0.527	0.202
Ce					1	0.691	0.350	0.025	0.365	0.227	0.193	−0.297	0.303	0.471	0.169	0.155	0.612
Cr						1	−0.074	−0.036	0.794	0.355	0.237	0.127	0.560	0.553	0.041	0.197	0.718
Cs							1	−0.213	0.058	0.394	−0.179	−0.190	−0.525	0.548	0.097	0.613	0.223
Cu								1	−0.039	−0.071	0.190	0.312	0.365	−0.032	−0.084	−0.302	0.103
Fe									1	0.544	0.167	0.513	0.399	0.682	−0.021	0.357	0.780
Mg										1	0.286	0.257	0.019	0.737	−0.077	0.474	0.560
Mn											1	0.004	0.359	−0.035	0.410	−0.307	0.189
Ni												1	0.349	0.300	−0.132	0.107	0.425
Pb													1	−0.009	0.010	−0.392	0.501
Rb														1	0.029	0.521	0.706
Sr															1	−0.274	−0.064
V																1	0.276
Zn																	1

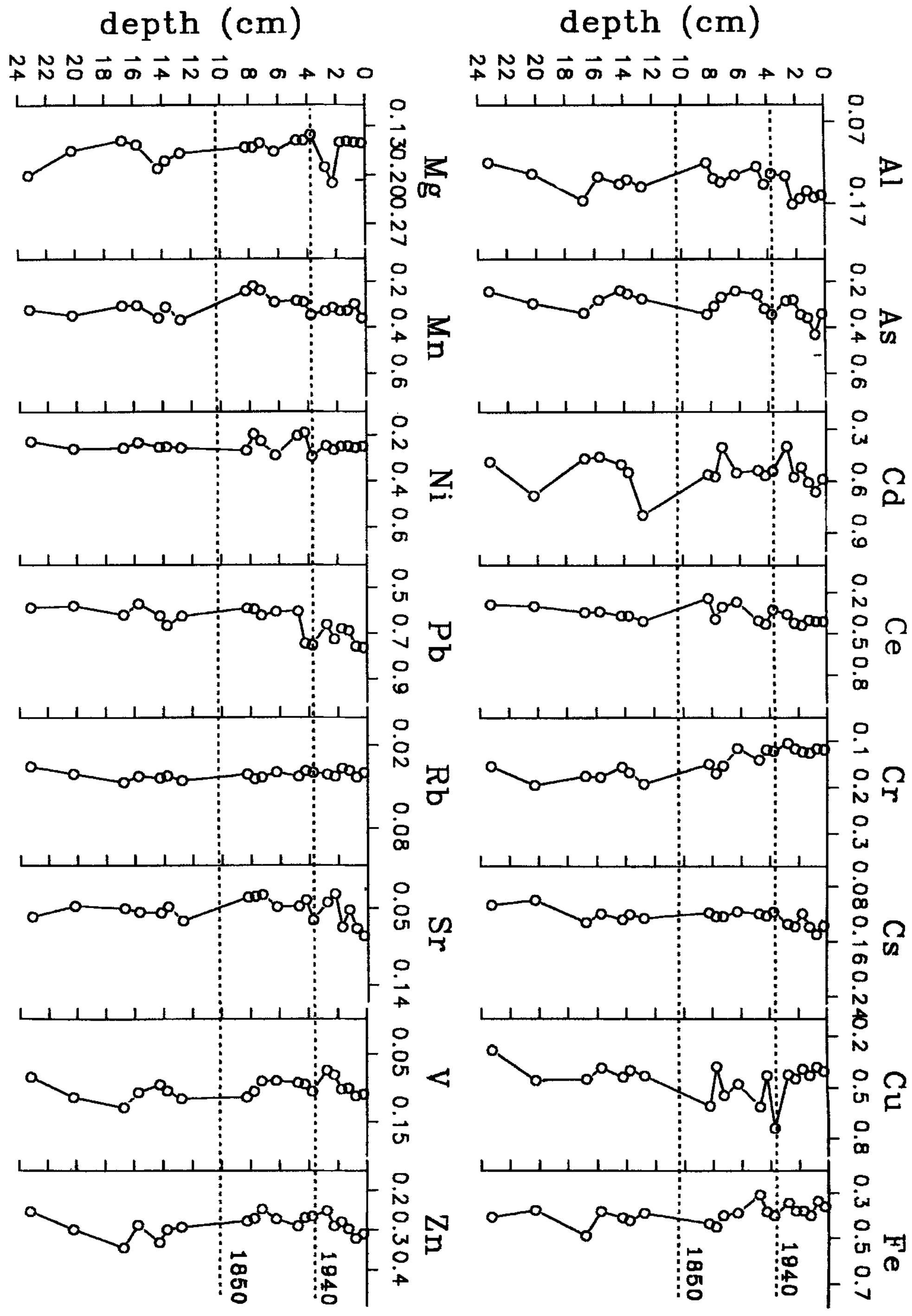

Figure 9 The distributions of ratios of acid-leached metal/total metal in the surface sediments of the Great Ghost Lake (93 cm core).

mainly due to the variation of source input (such as anthropogenic inputs of lead and cadmium), high ratios of acid-leached metal/total metal (> 0.2) of these metals, and redistribution of mobile fractions of these metals in the sediments during early diagenesis. The redistribution of these metals is mainly driven by the annual redox cycle of iron in this lake, and/or is caused by the remobilization of these metals and subsequent association with other phases (especially organic phases), and/or reprecipitation. Strontium and manganese tend to form carbonates in the reduced sediments, this is probably one of the factors resulting in the positive correlation ($R > 0.4$) between manganese and strontium. We believe that the remobilization of these metals is mainly caused by the dissolution of metal oxides in the reducing condition as iron and manganese oxides/hydroxides are good scavengers. Complexing with dissolved organic substances can markedly increase the solubilities of metals.

The distributions of acid-leached metals in the surface sediments of the Great Ghost Lake (93 cm core) are shown in Figure 10. Cadmium, caesium, lead, strontium and zinc are enriched in the surface sediments, in part, due to the anthropogenic input. The high ratios of Cd/Al, Pb/Al and, to a lesser extent, Zn/Al are also found in the surface sediments (Fig. 8). In addition to the anthropogenic input, the variation of organic matter contents also plays a major role in regulating the distribution of metals (Wann and Chen, 1995).

The concentrations of arsenic and cadmium have good positive correlations with organic carbon throughout the 93 cm core ($R = 0.77$ for arsenic and $R = 0.65$ for cadmium; Wann and Chen, 1995). Good correlations between organic carbon and acid-leached arsenic ($R = 0.87$) and acid-leached cadmium ($R = 0.71$) are also found (Wann and Chen, 1995). These suggest that arsenic and cadmium are mainly associated with organic matter. In contrast, aluminium, cerium, magnesium, rubidium and vanadium have negative correlations with organic carbon in the sediments. The R values between organic carbon and aluminium, cerium, manganese, rubidium and vanadium throughout the 93 cm core are -0.74, -0.81, -0.86, -0.83 and -0.69, respectively (Wann and Chen, 1995). These metals mainly came from the lithogenic sources.

The vertical distributions of ratios of acid-leached metal/total metal in the surface sediments of the 93 cm core and the 82 cm cores are shown in Figures 9 and 11. Average ratios of Al, As, Cd, Fe, Mn, Ni, Pb, V and Zn in the 82 cm core are 0.154, 0.337, 0.597, 0.284, 0.363, 0.178, 0.558, 0.097 and 0.261, respectively. In the 93 cm core, average ratios of Al, As, Cd, Ce, Cr, Cs, Cu, Fe, Mg, Mn, Ni, Pb, Rb, Sr, V and Zn are 0.144, 0.302, 0.551, 0.364, 0.144, 0.124, 0.455, 0.394, 0.165, 0.309, 0.245, 0.654, 0.041, 0.053, 0.101 and 0.288, respectively. The probability of remobilization of metals from the sediments if the lake becomes acidified is thus Cd > Pb > Mn > As > Fe > Zn > Ni > Al > V in the 82 cm core and Pb > Cd > Cu > Fe > Ce > Mn > As > Zn > Ni > Mg > Al = Cr > Cs > V > Sr > Rb in the 93 core. Both sequences are very similar and are also similar to the sequence found for the Little Ghost Lake. These also suggest that aluminium and vanadium mainly occur in the immobile fractions of the sediments whereas more cadmium, manganese and lead are associated with the mobile fractions of the sediments in both lakes.

However, the ratios of acid-leached metal/total metal for lead (0.558 and 0.654) and arsenic (0.337 and 0.302) in the Great Ghost Lake are much higher than those (0.305 for lead, 0.126 for arsenic) in the Little Ghost Lake. The relatively higher ratios of lead and

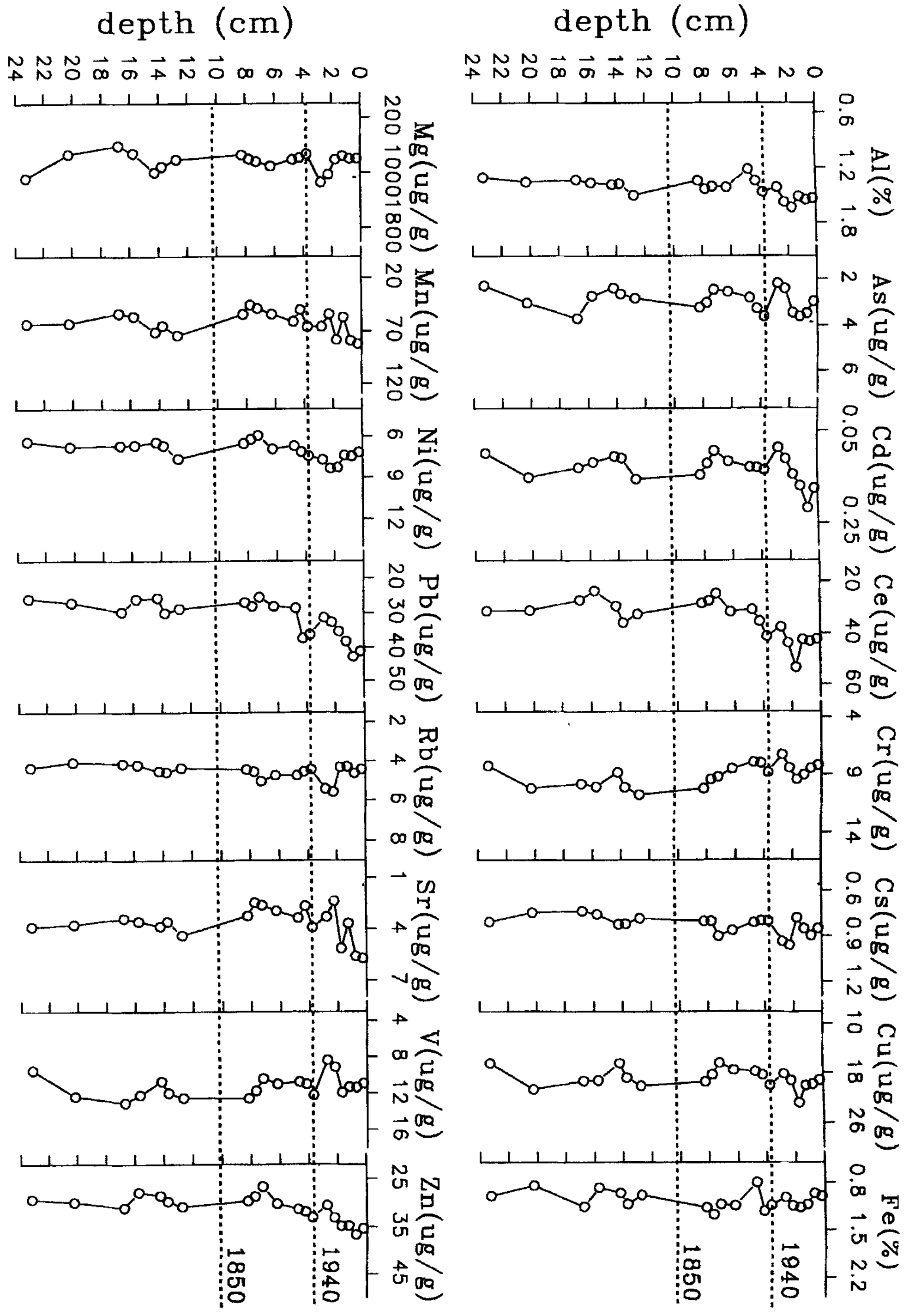

Figure 10 The distributions of acid-leached metals in the surface sediments of the Great Ghost Lake (93 cm core).

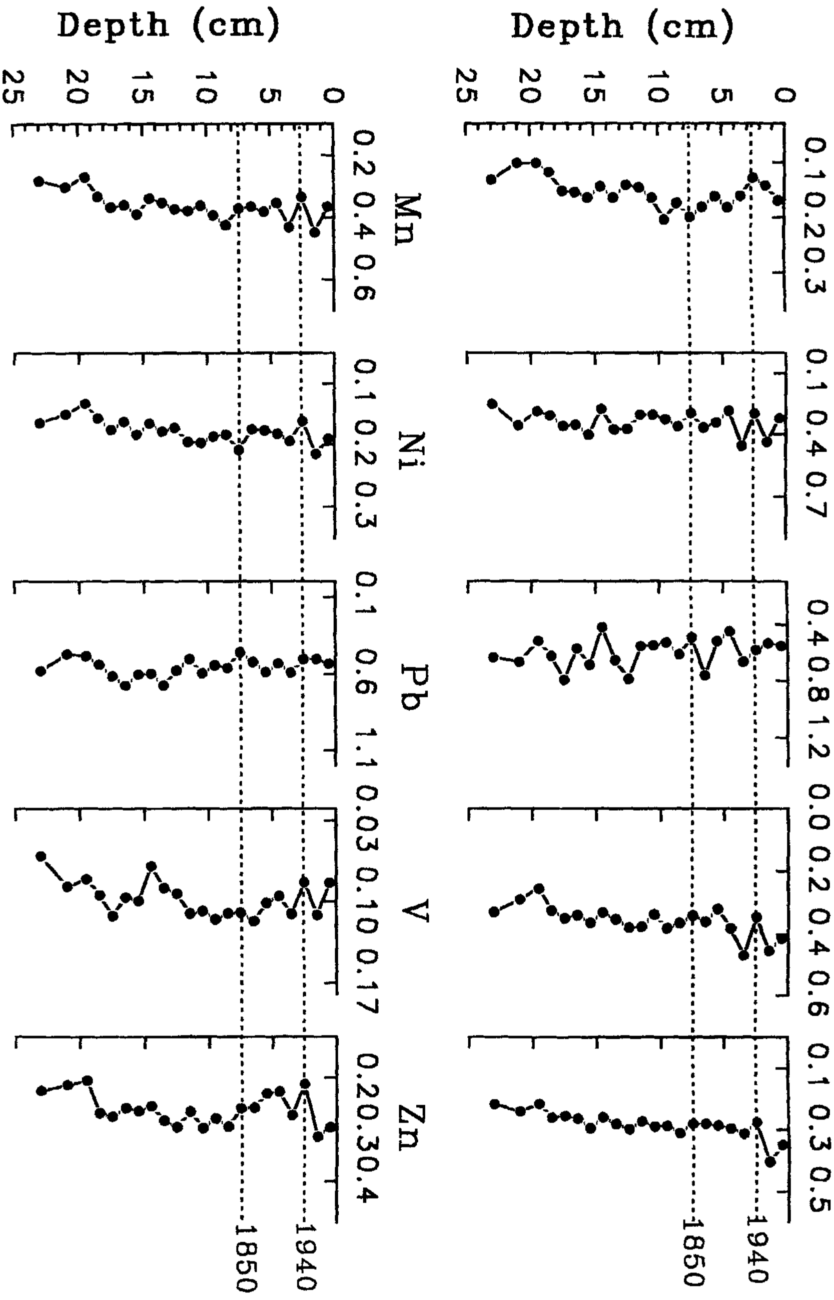

Figure 11 The distributions of ratios of acid-leached metal/total metal in the surface sediments of the Great Ghost Lake (82 cm core).

arsenic in the Great Ghost Lake are mainly caused by the higher concentrations of acid-leached lead (averaging 25.3 μg^{-1}) and arsenic (averaging 3.68 μg^{-1} comparing to those of the Little Ghost Lake (the averages of lead and arsenic are 14.6 and 1.16 μg^{-1}, respectively). The higher acid-leached arsenic in the Great Ghost Lake is probably caused by the high organic matter content which has good positive correlations with acid-leached arsenic ($R = 0.84$) and total arsenic ($R = 0.77$) through the 93 cm core (Wann and Chen, 1995). The acid-leached aluminium contents (averaging 1.40%) in the Great Ghost Lake is much higher than those (averaging 0.29%) of the Little Ghost Lake. This may result from the contents of the amorphous gibbsite and the clay-size minerals (about 10%) in the surface sediments of the Great Ghost Lake are higher than that of the Little Ghost Lake (the clay-size minerals are about 6%). A large amount of acid-leached lead is associated with the amorphous gibbsite and the surface of clay minerals.

Although both lakes are located in the mountainous area of southern Taiwan and have similar elevation and geological background, the water chemistry is different. The anoxic hypolimnion in the deep Great Ghost Lake results in good preservation of organic matters in the sediments, and drives the iron redox cycle in the lake. Subsequently metals in the mobile fractions of sediments, especially the iron and manganese oxides/hydroxides, are redistributed during the early diagenesis. All these cause the weak correlations between metals in the Great Ghost Lake (Tab. II and III). In contrast, the shallow Little Ghost Lake is always oxic and the retention time of dissolved and particulate metals in the water column is relatively short. As a result, metals in the mobile fractions of the sediment does not change much in the early diagenesis, resulting in strong positive correlations between most metals, especially those of lithogenic origin (Tab. I).

The correlations between the acid-leached metal and total metal in these two lakes are also very different (Tab. IV). Most of the metals in the Little Ghost Lake show strong positive correlations between the acid-leached metal and total metal. In the Great Ghost Lake, most of the correlations between the acid-leached metal and total metal are not significant. Some differences are even found between the two cores collected from the same lake because of different carbon content (Tab. IV).

Aluminium and iron are the most abundant metals in the earth's crust. Aluminium is also a major lithogenic metal occurring in the aluminium silicates and clay minerals. In the hydrocycle, a large amount of iron forms the iron oxides/hydroxides, which are very sensitive to the redox conditions, occur in the oxic sediments. In the Little Ghost Lake, strong positive correlations between the acid-leached aluminium and total aluminium ($R = 0.799$) and the acid-leached iron and total iron ($R = 0.918$) are found. In contrast, weaker correlations are found in the Great Ghost Lake (Tab. IV). All of these suggest that the metals in the mobile fractions of sediments in the Little Ghost Lake are not much changed during the early diagenesis. In contrast, the redistributions of metals in the mobile fractions of the sediments are obvious in the Great Ghost Lake during the early diagenesis.

Table IV The regression coefficients (*R*) between the acid-leached and total metal concentrations in the surface sediments of the Little Ghost Lake (LGL) and the Great Ghost Lake (GGL).

	Al	*As*	*Cd*	*Ce*	*Cr*	*Cs*	*Cu*	*Fe*	*Mg*	*Mn*	*Ni*	*Pb*	*Rb*	*Sr*	*V*	*Zn*
LGL	0.799	0.494	0.968	–	–	–	0.199	0.918	–	0.388	0.682	0.873	–	–	0.977	–
GGL (82cm)	–0.340	0.361	0.329	–	–	–	–0.092	–0.361	–	0.301	0.277	0.886	–	–	–0.435	0.295
GGL	–0.064	0.315	0.747	0.625	–0.375	0.214	0.034	0.600	0.694	0.603	0.395	0.893	0.747	0.066	–0.284	0.565

4. CONCLUSIONS

Marked difference of water chemistry causes different early diagenesis of metals in these two lakes. The seasonally anoxic hypolimnion in the Great Ghost Lake drives the annual iron redox cycle and causes the remobilization of metals in the mobile fractions of the sediments. These result in the redistribution of metals in the mobile fractions of sediments and cause weak correlations between metals. In the Little Ghost Lake, the shallow water column is always oxic with less change of metals in the mobile fractions of sediment. As a result, most metals have strong positive correlations with each other. Strong positive correlations also exist between most acid-leached metal and total metal in the Little Ghost Lake but the correlations are relatively weak in the Great Ghost Lake.

The Little and Great Ghost Lakes located in the isolated mountain regions of Taiwan have been affected by metal pollution mainly coming from the atmospheric fallouts which have played a major role in regulating the distributions of metals in recent lake sediments. The worldwide lead emissions from automobiles since 1940 have probably been important in the sharp increase of lead in the sediments of these two subalpine lakes.

The ratios of acid-leached metal/total metal in the surface sediments of these two lakes indicate that aluminium and vanadium are mainly from the lithogenic sources and are associated with the immobile fractions of the sediments. On the other hand, cadmium, manganese and lead are relatively mobile with higher ratios of acid-leached metal/total metal, as large amounts of these metals mainly occur in the mobile fractions of the sediments.

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