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# Sedimentary evidence for changes in the pollution status of Taihu in the Jiangsu region of eastern China

N.L. Rose[1,*], J.F. Boyle[2], Y. Du[3], C. Yi[3], X. Dai[4], P.G. Appleby[5], H. Bennion[1], S. Cai[3] and L. Yu[4]

*[1]Environmental Change Research Centre, University College London, 26 Bedford Way, London WC1H 0AP, UK; [2]Department of Geography, University of Liverpool, P.O. Box 147, Liverpool L69 3BX, UK; [3]Institute of Geodesy and Geophysics, Chinese Academy of Sciences, 54 Xu Dong Road, Wuchang 430077, Hubei, People's Republic of China; [4]Department of Geography, East China Normal University, Shanghai 200062, People's Republic of China; [5]Environmental Radioactivity Research Centre, Department of Mathematical Sciences, University of Liverpool, P.O. Box 147, Liverpool L69 3BX, UK; *Author for correspondence (e-mail: nrose@geog.ucl.ac.uk)*

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## Abstract

As part of a study using lake sediments to determine the extent and causes of human impacts to lakes along an east–west transect following the Yangtse River, sediment cores were taken from Taihu in eastern China. Previous studies have focussed on the impacts of direct inputs of pollutants from municipal and industrial wastewater but little work has been undertaken on trends in atmospheric deposition from the many industrial sources surrounding the lake. Analysis of the Taihu sediment cores for atmospheric pollutant indicators such as trace metals, magnetic parameters and spheroidal carbonaceous particles (SCPs) show the lake has become increasingly contaminated over the last 40–50 years. Sediment levels of atmospherically deposited pollutants are currently similar to some of the more contaminated lakes in Europe. Further, sediment nitrogen, phosphorus and geochemical analyses confirm the dramatic increase in eutrophication at the site and periods of recent soil erosion in the catchment.

## Introduction

Over the last few decades China has undergone rapid and extensive industrial growth particularly in the more densely populated eastern areas (e.g., Nanjing Statistical Bureau; Figure 1). This has led, inevitably, to dramatic increases in emissions of pollutants to the atmosphere and, as a consequence, to increases in the deposition of these pollutants over wide areas. These have the potential to severely impact water quality in lakes of the region (Boyle et al. 1999; Qu et al. 2002), but currently, there is little information on temporal trends in pollutant deposition to freshwater systems.

The importance of water resources in China cannot be overestimated and therefore determining the extent, rate and direction of change in water quality is a national priority. In the absence of long-term monitoring programmes, lake sediments remain one of the few ways whereby this information can be determined retrospectively (Smol 2002). Furthermore, such records can potentially ascertain whether any detriment in quality is as a result of natural changes, or due to agricultural or industrial impacts.

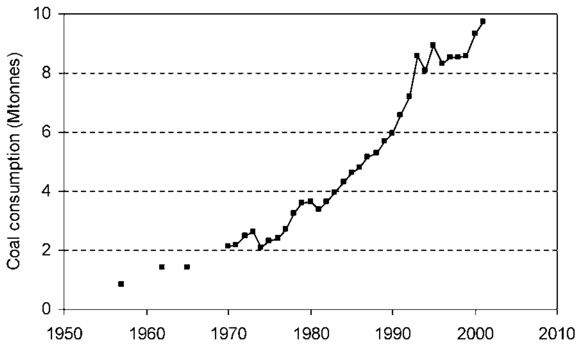


*Figure 1.* Coal consumption for Nanjing as an example of evidence for rapid industrial development in eastern China. (Data source: Nanjing Statistical Bureau).

In 1998 a collaborative project, jointly funded by the UK's Royal Society and the Chinese Academy of Sciences, was initiated to use lake sediments from three regions of China to determine the extent to which impacts to freshwaters have changed through time and to identify the causes of these changes. The three areas were selected on an east–west transect along the Yangtse River thus complimenting previous studies at other sites along the river (Boyle et al. 1999). This paper discusses the results from Taihu in the first of these areas, the lowland Jiangsu region in eastern China. Previous studies on Taihu have discussed the impacts of direct inputs of pollutants from municipal and industrial wastewater and domestic sewage (e.g., Zou et al. 1996; Shen et al. 2000; Yu and Yang 2001; Dickman et al. 2001; Qu et al. 2001). Here, we focus on the identification of changes in input of pollutants to the lake from atmospheric deposition.

## Site

Taihu, the third largest lake in China, is located in the delta of the Yangtse River in eastern China (30°05′–32°08′N; 119°08′–121°55′E). It is a shallow lake with a surface area of 2338 km$^2$ (Qin 1999), a catchment area of 36,895 km$^2$ and a mean depth of 1.9 m. Seven large and medium size cities and 31 county towns are to be found around the lake (Shen et al. 2000) (Figure 2) whilst the catchment includes 36 million people or 3% of the nation's population. The catchment also contains 2.66 million hectares of farmland to which 2–3 million tons of chemical fertiliser and 70–80,000 tons of pesticides are applied annually. This results in ca. 1.5 million tons of chemical pesticides and fertilisers being washed into the lake each year (Yu and Yang 2001). However, despite banning the use of organochlorine pesticides in China in 1983, DDT and HCH residues can still be detected in river water, sediment and fishes in the region (Feng et al. 2003). High levels of nutrients, polycyclic aromatic hydrocarbons and heavy metals have been ascribed to the input of partially and untreated domestic and industrial sewage (Qu et al. 2001, 2002) from the cities of Wuxi, Changzhou and Wujin, amongst others, mainly via the Zhihugang River joining the lake from the north (Qu et al. 2002).

However, apart from receiving wastewater, the lake is also the main source of drinking water for the city of Wuxi (population 4.3 million) (Figure 2) as well as being used for flood control, shipping, waste disposal (Qu et al. 2001) and a major source of fish for human consumption and macrophytes for fish food. Intensive fish-farming further increases nutrient inputs to the lake. It is therefore no surprise that water quality has decreased over recent decades (Qin 1999; Qu et al. 2000) and the lake is now hypertrophic (Pu et al. 1998; Dickman et al. 2001) having been oligotrophic in the 1950s (Qu et al. 2000). Blue-green algae dominate the west basin of Taihu whilst the east basin is generally covered by vascular plants (Qu et al. 1998; Dickman et al. 2001). Current diatom assemblages of the lake are indicative of highly productive alkaline conditions, being dominated by planktonic taxa, principally *Stephanodiscus* spp., *Aulacoseira granulata*, *Cyclotella meneghinaina* and *Nitzschia* spp. and biodiversity is thought to have decreased since 1970 (Li 1999). However, our investigation found that the diatom dissolution index was exceptionally high in the sediments of Taihu and prevented any interpretation of historical floristic change.

Since 1997 major investment by Shanghai Municipality, Jiangsu and Zhejiang Provinces has created 30 new water treatment plants and 200 'eco-agricultural' zones whilst livestock farms in the Taihu Class-1 Protection Zone now control their discharges and, as a consequence, phosphate from rivers feeding Taihu have been reduced by 16%. These measures have led to some recent water quality improvement although Qu et al. (2001) attribute a major reduction in phosphorus in the

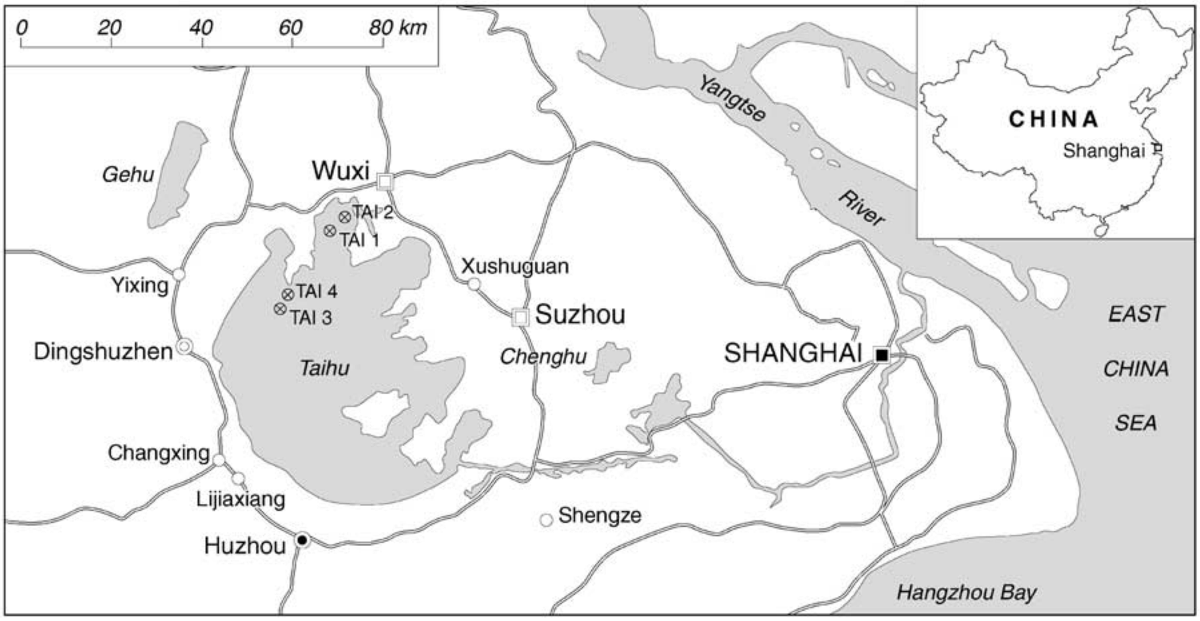


*Figure 2.* Map of Taihu showing coring locations and proximity to major cities and the Yangtse (Changjiang) River.

sediments of East Taihu to macrophyte removal. However, it has been suggested that it is reduced light, as a result of resuspended solids, rather than phosphorus that limits productivity in the lake (Dickman et al. 2001).

## Methods

### *Coring and lithostratigraphy*

The Taihu lake basin is flat and the major sediment accumulation occurs to the west (Qu et al. 2000) along a wide palaeo-channel. Sediment cores were taken from four locations within this area of Taihu (Figure 2) in October 1998. At each location short gravity cores (TAI-1S to TAI-4S) and longer piston cores (TAI-1L to TAI-4L) were taken and extruded in 1 cm intervals. Analysis of sediment dry weight and organic content as loss-on-ignition were undertaken using standard techniques (Stevenson et al. 1987).

### *Dating*

Cores from each location were screened for the quality of their records of fallout radionuclides and subsequently TAI-1S and TAI-4S were selected for more detailed study. Radiometric dates were obtained for the sediment cores by analysing for $^{210}Pb$, $^{226}Ra$, and $^{137}Cs$ by direct gamma assay using a well-type coaxial low background intrinsic germanium detector using the methods described in Appleby et al. (1986) and Appleby (2001). Lithostratigraphic, geochemical and spheroidal carbonaceous particle (SCP) data were used to cross-correlate the chronology to the TAI-1L core.

### *Magnetic susceptibility*

Magnetic parameters were measured on core TAI-1L. Low field AC magnetic susceptibility was measured using a dual frequency (470 Hz = $\chi_{LF}$, 4700 Hz = $\chi_{HF}$) Bartington Instruments MS2 sensor. Anhysteretic remnant magnetisation (ARM) was induced in a steady field of 0.1 mT with a parallel peak alternating field of 100 mT using a DTECH AF demagnetiser and measured on a Molspin spinner magnetometer. Acquisition of isothermal remanent magnetisation (IRM) in fields of 1 T (SIRM), −20 mT ($IRM_{-20\ mT}$), and −300 mT ($IRM_{-300\ mT}$) was carried out using a Molspin pulse magnetiser and measured on a Molspin spinner magnetometer. SOFT is calculated as (SIRM-$IRM_{-20\ mT}$)/2 and HIRM as (SIRM-$IRM_{-300\ mT}$)/2, both on a mass specific

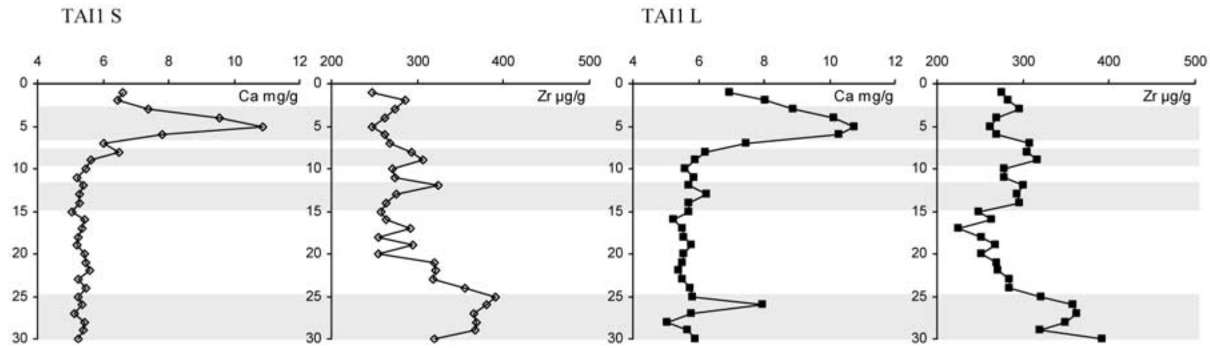


*Figure 3.* Sediment concentrations of Ca and Zr for the cores TAI-1S and TAI-1L showing the good depth correlation between the cores.

basis. HARD% is 100 × (HIRM/SIRM). FD% is $100 \times (\chi_{LF} - \chi_{HF})/\chi_{LF}$.

*Sediment geochemistry*

Si, K, Ca, S, Fe and Zr were measured on both TAI-1S and TAI-1L using energy dispersive isotope-source X-Ray fluorescence (XRF) following the techniques described in Boyle (2000). Cd, Cu, Pb and Zn were measured on nitric acid extracts by flame atomic absorption spectrometry. A STATS tube was used for Cd and Pb to enhance sensitivity.

*Spheroidal carbonaceous particles*

SCPs are produced from the high temperature combustion of fossil-fuels and have no natural sources. Therefore, in lake sediments they provide an unambiguous record of industrially-derived, atmospherically deposited pollution. The procedure for extraction and enumeration of SCPs from the TAI-1S and TAI-1L sediment cores followed Rose (1994). Sediment concentrations were calculated as 'number of SCPs per gram dry mass of sediment' or $gDM^{-1}$.

*Nitrogen and phosphorus*

Nitrogen and phosphorus analysis was undertaken on the TAI-1S core. Sediment nitrogen analysis followed an adapted Kjeldahl digestion followed by a distillation procedure (Allen 1989) whilst sediment phosphorus analysis employed a perchloric–sulphuric acid digest followed by a colorimetric measurement (Allen 1989).

**Results**

Following initial screening of the Taihu cores it was concluded that the TAI-1L and TAI-1S cores contained the best resolved and most reliable records. These were therefore the subjects of subsequent detailed analyses at institutes in both the United Kingdom and China. The remainder of this paper deals with the results from these cores. A comparison of element geochemistry (Figure 3) and mineral magnetics showed an excellent correlation between these two cores, and no difference in the timing of stratigraphic changes. We therefore have confidence in the direct comparison of the sediment records of these two cores.

*Dating*

$^{210}Pb$ activity in TAI-1S (Figure 4) declines irregularly with depth with little net decline in the top 20 cm and a maximum at 14.5 cm. Unsupported concentrations decline abruptly below 25 cm and equilibrium with the supporting $^{226}Ra$ is reached at 30 cm. The $^{137}Cs$ profile has a relatively well-resolved peak between 15 and 25 cm that most likely records the 1963 fallout maximum from the atmospheric testing of nuclear weapons. The presence of this peak together with the sharp decline in $^{137}Cs$ concentration above 18 cm suggests that the irregular profile is not due to mixing. $^{210}Pb$ dates calculated using the $^{137}Cs$ date as a reference point suggest a mean accumulation rate of 0.30 g $cm^{-2}$ $year^{-1}$ (0.41 cm $year^{-1}$) between 1952 and 1986, increasing during the past decade

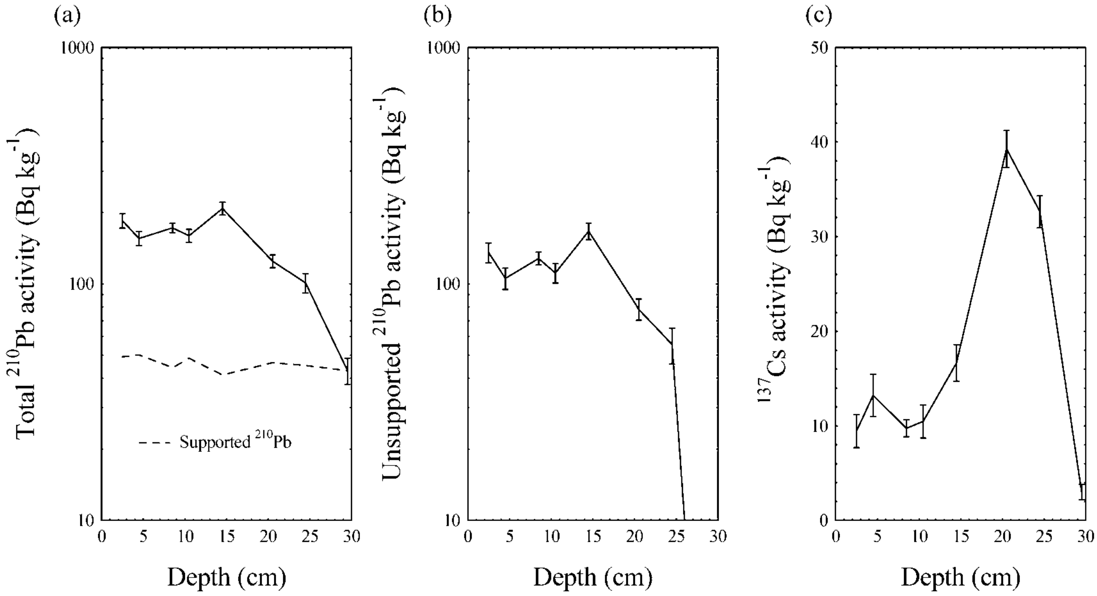


*Figure 4.* Fallout radionuclide concentrations in Taihu core TAI-1S showing (a) total and supported $^{210}$Pb; (b) unsupported $^{210}$Pb; (c) $^{137}$Cs.

to 0.52 g cm$^{-2}$ year$^{-1}$ (0.88 cm year$^{-1}$). The mean post-1963 $^{210}$Pb flux is more than twice the atmospheric flux and almost certainly reflects intensive sediment focussing at the coring site. Further details are given in Yi et al. (in press). Dates from the chronology constructed from $^{210}$Pb and $^{137}$Cs data are shown on the sediment profiles in Figures 5, 6 and 9.

*Magnetic susceptibility*

In the post-1940s sediment of TAI-1L there is great similarity between the $\chi$LF, SIRM, and SOFT profiles, and a degree of similarity with the HIRM profile (Figure 5) with values generally increasing to the sediment surface and a number of coincident peaks between the profiles. By contrast, the HARD% profile, uniquely amongst the magnetic parameters, shows a decline over the period. $\chi$ARM and $\chi$FD% show contrasting trends, with $\chi$ARM showing a decline over the early part of the record (pre-1950) followed by a general increase to the present.

*Sediment geochemistry*

Geochemical and trace element data for TAI-1L are presented in Figure 6. Below 25 cm (i.e., prior to the mid-1960s) the sediment is rich in Si and Zr and relatively depleted in the other major and minor elements. The correlation of Zr with Si ($R = 0.66$) is typical where a silt component mixes with other sediment components (Boyle 2001). The fall in both Si and Zr above 25 cm, and complementary increase in Ti and K is compatible with an increase in the clay concentration, a conclusion which is supported by particle size analysis in an adjacent core which shows an increased clay content in the top 25–30 cm. This situation, clearly illustrated by the Ti/Zr ratio profile, shows a partial return to initial values above ca. 15 cm (early 1980s).

A marked increase in trace metal concentration is observed above 25 cm. Cd, Cu, Pb and Zn all show the same pattern, increasing in concentration sharply at first, and then flattening off towards the sediment surface. Cd and Pb show little variation in concentration above ~20 cm (since ~1970) while Cu and Zn continue to rise up to a depth of ~12 cm (mid- to late-1980s). However, these near surface changes are subtle, and the concentrations are fairly constant in the last 20 years.

*Spheroidal carbonaceous particles*

The SCP profile for TAI-1L (Figure 6) shows low concentrations up to 30 cm (ca. 1930s) followed by

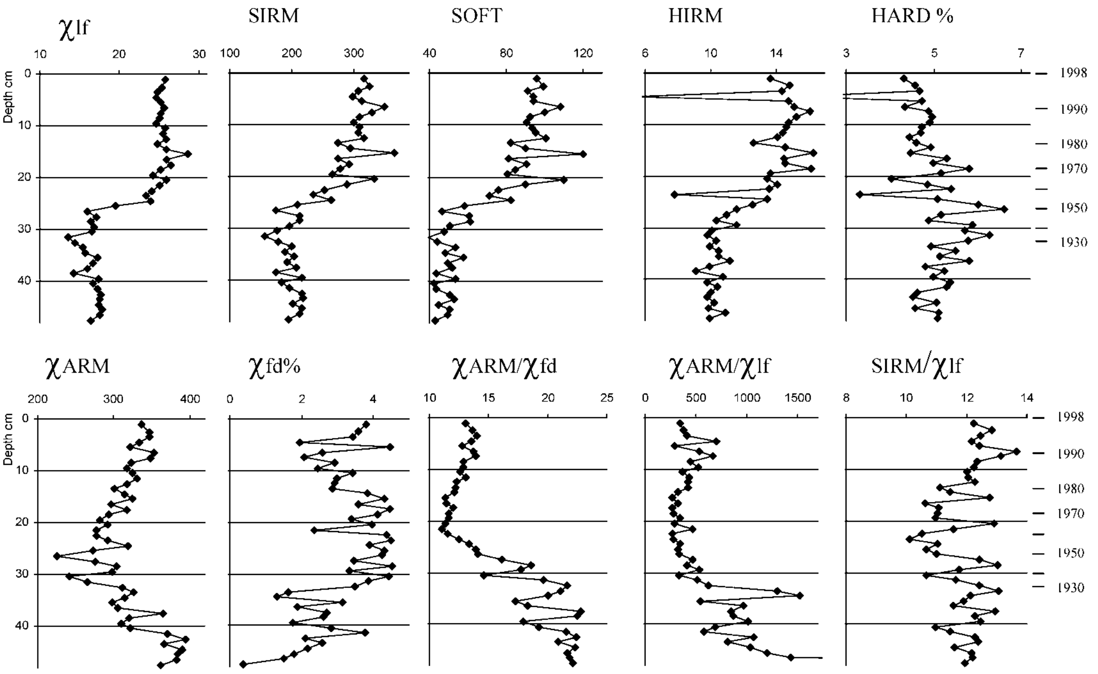


*Figure 5.* Magnetic parameters for core TAI-1L. See text for definitions.

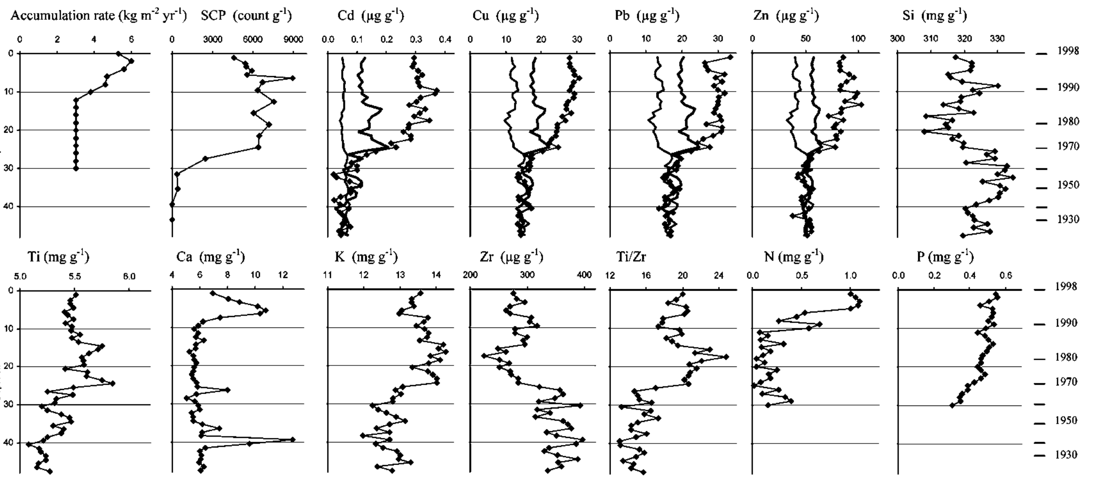


*Figure 6.* Sediment accumulation rate, SCP, trace metal, geochemical and nutrient element concentration profiles for the core TAI-1L. Filled symbols (◆) show sediment concentrations; thick and thin lines on the trace metal profiles show baseline estimates using Zr and Ti, respectively. Data are shown on a sediment depth axis whilst the corresponding $^{210}$Pb chronology is given on the right.

a massive increase in concentration at 25–30 cm (1940s and 1950s). Above this the concentration profile flattens with a peak of 9000 $gDM^{-1}$ at 6–7 cm (ca. 1990) followed by a decline to the sediment surface.

## Discussion

The similarity between the profiles of the magnetic parameters $\chi$LF, SIRM, SOFT and HIRM suggests that the magnetic signature is dominated by ferrimagnetic minerals. Beyond this, there are a number of differences between the profiles of the various magnetic parameters, showing that four independent magnetic components contribute to the total magnetic record.

First, the HARD% profile, unlike the other magnetic parameters, shows a decline over the top 30 cm (the last 60 years). This suggests that there is an antiferromagnetic component that varies independently of the main ferrimagnetic fraction. This is most easily explained if it is caused by independent variation in goethite or haematite. A crude correlation of HARD% with the Si and Zr concentration profiles, which vary independently of the other mineral elements (Figure 6), points to an association with coarser grain sizes, probably silt.

Second, the $\chi$ARM profile resembles no other magnetic parameter and, as this is a specific indicator of stable single domain ferrimagnetic grains, this second component might reflect a pedogenic or bacterial magnetosome source. The contrasting behaviour of $\chi$ARM and $\chi$FD% points away from a pedogenic source, while the $\chi$ARM/$\chi$FD and $\chi$ARM/$\chi$LF ratios indicate a magnetosome contribution (Oldfield 1999). The similarity of the SIRM/$\chi$LF ratio to the $\chi$ARM profile supports this owing to the inverse grain size dependence of SIRM (Walden 1999).

These first two fractions are distinctive but do not greatly influence the main susceptibility and remanence parameters. However, $\chi$LF, SIRM and SOFT all show remarkably similar profiles and this third group of parameters point to a ferrimagnetic component that varies independently of both the antiferromagnetic fraction, and the stable, single domain ferrimagnetic component. The low magnitude, and contrasting profile of the $\chi$FD%, show that there is no superparamagnetic contribution and this is compatible with a coarse grained multi-domain ferrimagnetic fraction typical of atmospheric pollution. This interpretation is reinforced by strong correlations of $\chi$LF, for example, with Cd, Cu, Pb and Zn ($R^2$ respectively 0.92, 0.90, 0.90, 0.87).

The $\chi$FD% profile differs strikingly from the third component. First, the step increase occurs at ~32 cm, significantly deeper down the sediment profile. Second, the value declines slightly (but significantly) over the top 15 cm (the last 20 years or so). This suggests that there is a fourth component, showing a subtle but distinct difference from the pollution signal. Of the other magnetic parameters only HARD% shows any sign of response at 32 cm, exhibiting a slight increase. Therefore, this fourth fraction might reflect a pedogenic source, possibly suggesting enhanced soil erosion. Previous magnetic susceptibility studies in Taihu (Qu et al. 2000) support evidence for enhanced human activity and periods of soil erosion in recent sediments. However, such an interpretation receives no support from the geochemical data, though it may be that local soils have a similar major element composition to the sediment in the lake, which would make the geochemistry a poor indicator of erosion.

If the trace metal increases are to be interpreted in terms of enhanced pollution, then the chemical properties of the pre-pollution sediment must be characterised. A passive tracer element can be used to evaluate this (Norton and Kahl 1987; Boyle 2001) whilst the method employed by Hilton et al. (1985) can be used to decide upon an appropriate model. The essence of the approach is to identify a depth in the sediment above which contamination by pollutants disrupts the previous relationship between a heavy metal and a passive tracer element. In this instance, two plausible tracers emerge, Ti and Zr, which lead to significantly different results. The correlations of trace metals with Zr are weak but statistically significant for samples deeper than 26 cm (Figure 7). For the same depths correlations with Ti are not significant. However, if the depth intervals 24 and 25 cm are included, good correlations are observed. Baseline concentrations estimated using Ti or Zr are both shown in Figure 6 whilst the corrected trace metal enrichment profiles are shown in Figure 8. If the Ti model is correct, then the recent trace metal enrichment starts at a depth of 22 cm (early 1960s). Conversely, if the Zr model is correct, then the enrichment started earlier, at a depth of 25 cm (mid-1950s). While there is no solid evidence to distinguish the two cases, there are two reasons to favour the Zr model. First, the concentration profiles for all four pollutant metals are then essentially identical, whereas they differ widely if a Ti model is assumed. Second, the pollution profiles based on Zr baseline correction are essentially identical to the SCP profile, while differing greatly if a Ti model is assumed. By application of Occam's razor, the simpler of these scenarios is

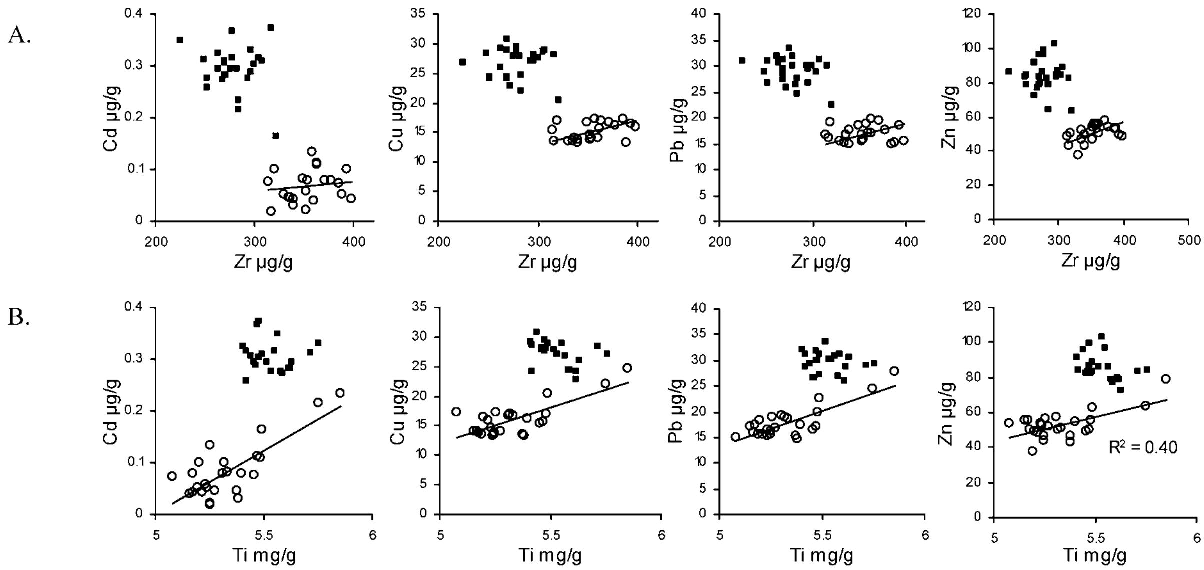


*Figure 7.* Baseline estimates based on the procedure of Hilton et al. (1985). The filled squares represent sample that have a metal contribution from pollution. The open circles are those samples that are assumed to have no contamination, and thus can be modelled using a passive tracer such as Zr or Ti.

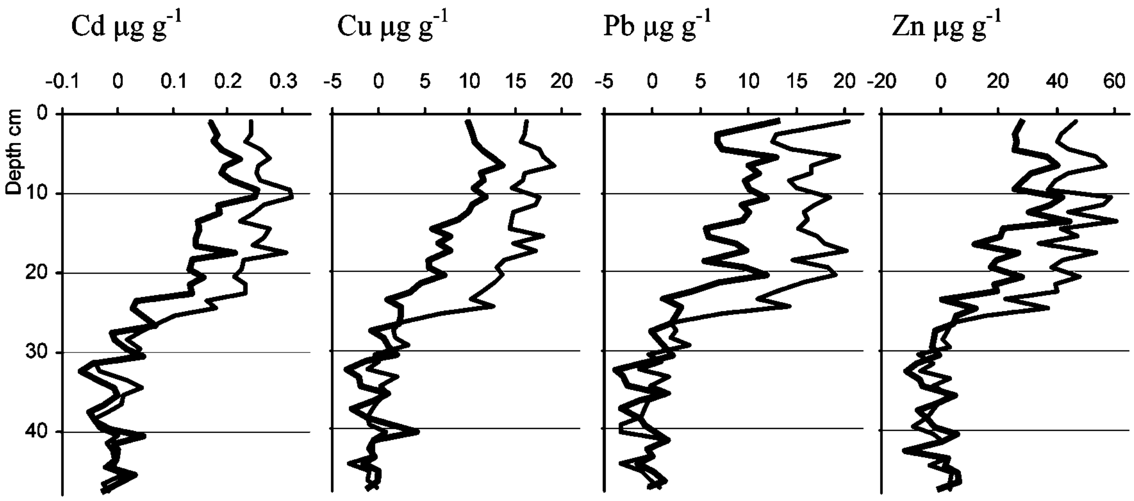


*Figure 8.* Estimates of the trace metal contamination record. Concentrations have been corrected for the natural contribution using Ti (thick line) and Zr (thin line).

favoured pointing to similar atmospheric pollution signals for SCP and heavy metals.

The SCP concentration profile (Figure 6) and the Zr-corrected profiles for Cd, Cu, Pb and Zn (Figure 8) show relatively little change over the top 25 cm (1960 to present day) of the sediment record. The peak concentrations are also relatively low compared with comparable European sites. However, if we take sediment mass accumulation rates into account a different picture emerges. Figure 9 shows SCP and heavy metal fluxes (calculated as the product of sediment mass

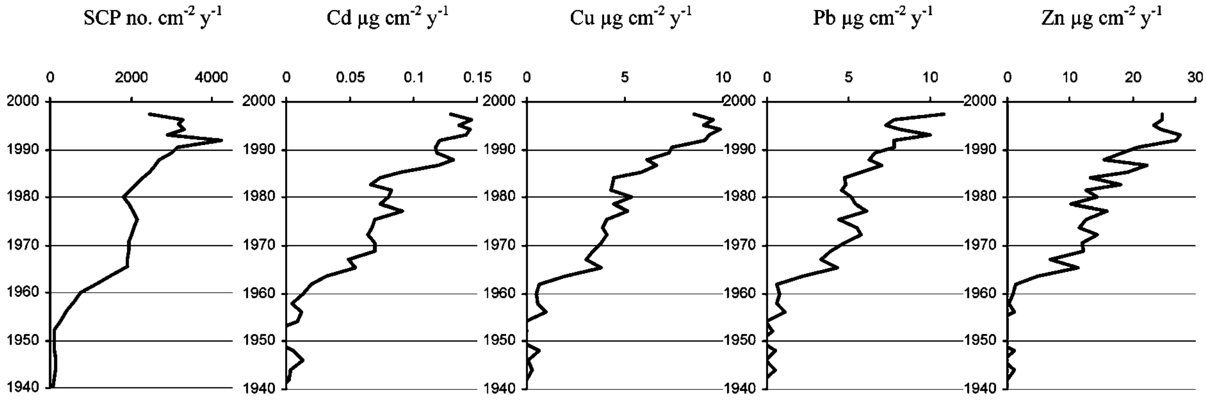


*Figure 9.* Accumulation rate profiles for SCPs, Cd, Cu, Pb and Zn for the core TAI-1L.

accumulation rate and pollutant concentration) plotted though time. This reveals a more or less continuous increase in atmospheric pollutant flux from 1960 to the present day, though with a slight recent decrease in the SCP flux. Further, there appears to be good temporal agreement with regional industrial development (e.g., regional coal consumption; Figure 1). The maximum fluxes, in the most recent sediment, are considerably higher than those obtained for lakes on the Jianghan Plain (Boyle 1999) apart from Donghu, located within the city of Wuhan and known to receive significant atmospheric deposition. This site shows peak SCP accumulation rates of a similar order to Taihu. These values are equivalent to levels observed in some of the more contaminated lakes in Europe.

The sediment N and P profiles (Figure 6) show agreement with the known eutrophication and decreasing water quality of Taihu over a period of decades. P concentrations increase steadily from the base of the core (ca. 1930s) to the present and this may be interpreted as a record of aqueous P (see Boyle 2001 for a review). Nitrogen levels increase dramatically from the mid-1980s and this is in agreement with N concentrations in lake water which have been reported to have increased from 1.84 mg $L^{-1}$ in 1987 to 2.5 mg $L^{-1}$ in 1994 (Sun and Huang 1993; Fan et al. 1997; both cited from Qin 1999). High N/C ratios are generally associated with algae and hence the dramatic increase in sediment N may reflect the increased severity of algal blooms in the lake in recent years. Such an interpretation receives support from the Ca profile; the pulse in Ca at 3–8 cm (Figure 6) can plausibly be explained by algal blooms severe enough to cause precipitation of calcite (e.g., Dean 1999). However, the low N concentration deeper in the core may simply be due to loss by microbial breakdown and therefore further work is required to fully interpret the nutrient record.

## Conclusions

The sediment record of Taihu reveals a complex history of changes in recent decades. Magnetic susceptibility data indicate changes in sediment supply, such as enhanced soil contributions, whilst nitrogen, phosphorus and geochemical sediment data show confirmation of eutrophication since at least the 1960s, with a possible acceleration in the last 15–20 years as a result of domestic and agricultural inputs via direct discharge and run-off. Superimposed on these records is evidence for increases in atmospheric deposition of pollutants.

At ca. 25 cm (1950s) there is a sharp increase in the concentration of heavy metals, SCPs and magnetic indicators of pollution indicating significant increases in atmospheric emissions as a result of developing industry in the cities of Shanghai, Suzhou, Wuxi, Changzhou, Jiaxing, Huzhou and Hangzhou around the lake.

Above 10–15 cm (1987–1976) there is a reduction or levelling in the pollutant concentrations. This coincides with an increase in the sediment accumulation rate, and an increase in the concentration of Si and Zr. Conversion of concentration data to pollutant accumulation rates indicates a continued increase over the period in agreement with trends in regional industrial development.

Any decrease in pollutant sediment concentrations at this time is therefore due to dilution rather than reduced pollutant loading. Furthermore, although concentrations of contaminants in the sediments of Taihu are only moderate, accumulation rate values of, for example, SCPs are found to be at an equivalent level to those of some of the most contaminated sites in Europe. This represents evidence for high levels of atmospheric deposition which added to the other stresses, could lead to further ecological damage at this or other lakes in the region.

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