Seasonal variations of arsenic at the sediment–water interface of Poyang Lake, China

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A B S T R A C T

Arsenic species including arsenite, arsenate, and organic arsenic were measured in the porewaters collected from Poyang Lake, the largest freshwater lake of China. The vertical distributions of dissolved arsenic species and some diagenetic constituents [Fe(II), Mn(II), S(−II)] were also obtained in the same porewater samples in summer and winter. In sediments the concentration profiles of total As and As species bound to Fe–Mn oxyhydroxides and to organic matter were also determined along with the concentrations of Fe, Mn and S in different extractable fractions. Results indicate that, in the summer season, the concentrations of total dissolved As varying from 3.9 to 55.8 μg/L in sediments were higher than those (5.3–15.7 μg/L) measured in the winter season, while the concentrations of total As species in the solid phase varied between 10.97 and 25.32 mg/kg and between 7.84 and 30.52 mg/kg on a dry weight basis in summer and winter, respectively. Seasonal profiles of dissolved As suggest downward and upward diffusion, and the flux of dissolved As across the sediment–water interface (SWI) in summer and winter were estimated at 3.88 mg/m^2 a and 0.79 mg/m^2 a, respectively. Based on porewater profiles and sediment phase data, the main geochemical behavior of As was controlled by adsorption/desorption, precipitation and molecular diffusion. The solubility and migration of inorganic As are controlled by Fe–Mn oxyhydroxides in summer whereas they appear to be more likely controlled by both amorphous Fe–Mn oxyhydroxides and sulfides in winter. A better knowledge of the cycle of As in Poyang Lake is essential to a better management of its hydrology and for the environmental protection of biota in the lake.

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1. Introduction

Arsenic (As) has trace abundance in the Earth's crust, yet it occurs widely in the environment, with localized high concentrations found in certain rock, soils, and groundwater and well waters, especially in the many parts of Asia (e.g. Azcue and Nriagu, 1994; Benner et al., 2008; Polya et al., 2008; Rowland et al., 2008; Song et al., 2010). Arsenic occurs naturally in lake waters as a result of hydrothermal leaching or weathering of As-containing minerals in rocks and soils or of geothermal activity (Belzile and Tessier, 1990). It can also be introduced by anthropogenic activities such as drainage from abandoned mines and mine tailings and usage as pesticides and biocides, among others (Oremland et al., 2000).

In the aquatic environment, As can occur in several oxidation states (−III, 0, III, and V) but in natural waters it is mostly found in inorganic form as oxyanions of arsenite As(III) and arsenate, As(V) (Smedley and Kinniburgh, 2002). The elemental As, and arsine(−III) occur only rarely in highly reducing environments (Cullen and Reimer, 1989; Couture et al., 2010a). Organic As associated with various inorganic and organic compounds or colloids forms may display different biological and geochemical active properties depending on the nature of the system. Inorganic arsenite and arsenate are more toxic than organic As compounds and have been recognized as human carcinogens mainly for lung and skin (Samanta et al., 2000; Couture et al., 2010b). Studies on freshwater As geochemistry in lakes are crucial to understand its geochemical behavior (solubility, mobility and transport), particularly in such a fragile and dynamic environment like Poyang Lake. In this paper we report on the speciation of dissolved As in porewaters at the sediment–water interface (SWI) and on the partitioning of solid As species in the sediments of Poyang Lake, China.
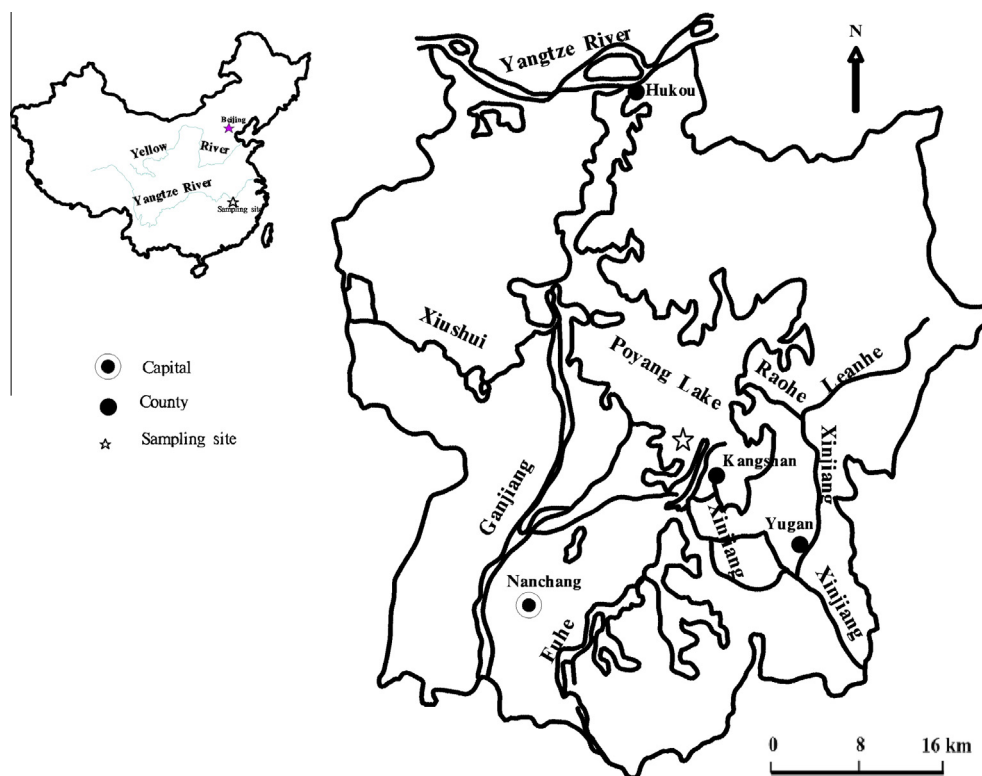
Fig. 1. Geographic map of Poyang Lake. ☆ Sampling site.

2. Experimental section

2.1. Site description

Poyang Lake, located in the north of the Jiangxi Province and to the south of mid-to-low reaches of the Yangtze River (28°24′–29°46′N, 115°49′–116°46′E) in Fig. 1, is the largest freshwater lake in China with an area of ∼3500 km² at high level. The average depth is approximately 8 m with the maximum depth being around 23 m. The lake is surrounded by eleven counties and is connected to six main rivers named Xiushui, Ganjiang, Fuhe, Xinjiang, Raohe and Leanhe. The water from these six rivers runs through the lake and discharges into the Yangtze River through a 1 km long channel at Hukou. In this study, samples were collected in Kangshan (28°49′27.6″N, 116°22′25.3″E) in August 2003 and January 2004, respectively.

2.2. Sediments and porewater sampling

Porewater samples were collected using *in situ* steady state diffusion method described in the literature (e.g. Xu et al., 1997; Liao and Deng, 2006). With this sampling approach, a volume of deoxygenated demineralized water contained within a filtration membrane (0.22 μm) is allowed to equilibrate with the overlying and interstitial waters after the Plexiglas support has been inserted in the sediments. After a 14-day *in situ* equilibration period, the peepers were retrieved from the sediment by a diver and sampled immediately on site. In each corresponding depth, a 2-mL subsample was collected for dissolved sulfide analysis (i.e. for S^{2-} analysis) and transferred to a 5-mL bottle containing the fixing agent for dissolved sulfide (the bottle was capped and brought to the laboratory where the rest of the procedure was completed (Chen et al., 2003). A 1-mL subsample was used to measure the pH close to the SWI. The remaining volume of porewater was acidified to pH < 2 with

Suprapure nitric acid, and stored at 4 °C in pre-cleaned Teflon containers. Water samples were collected at various depths to determine the dissolved oxygen (DO) using the Wrinkler method. Sediment cores were collected by a diver using a light Plexiglas corer at the same site of Kangshan where porewaters were collected. The cores were transported to the laboratory and extruded immediately in a glove-box bag filled with nitrogen gas to prevent oxidation. Sediment slices from each 1 cm depth were placed in pre-cleaned polyethylene containers and kept frozen at −80 °C until analysis.

2.3. Analysis of porewater and sediments

An atomic fluorescence spectrometer AFS-2202 equipped with a continuous flow injection hydride generation system and a boosted discharge hollow cathode lamp as the source of atomic fluorescence (FI-HG-AFS, Beijing Haiguang Geological Instrument Co.) was used for As analyses through the formation of AsH₃. For porewater and overlying water samples, the concentrations of As(III), As(V) and the fraction of total dissolved As associated to organic matter (As-org) species were measured by FI-HG-AFS according to a procedure previously developed (Liao and Deng, 2006). Briefly, for the determination of As(III), a sub-sample of pre-acidified porewater was analyzed in 0.005% (w/v) 8-hydroxyquinoline and 10% (v/v) HCl (Suprapure grade). To measure As(V), another aliquot was treated with 2.0% (w/v) KI, 0.005% (w/v) 8-hydroxyquinoline and 10% (v/v) HCl in the final solution to reduce all inorganic As to As(III) before determination by FI-HG-AFS, and As(V) was then obtained by difference. A third aliquot was digested in a Teflon bomb with concentrated nitric acid and subjected to a microwave treatment at 720 W for 5 min. After digestion and cooling at room temperature, KI, 8-hydroxyquinoline and HCl were added to obtain a final solution containing 2.0% (w/v) KI, 0.005% (w/v) 8-hydroxyquinoline and 10% (v/v) HCl before As determination. The

concentration of As-org was obtained by subtracting the sum of (As(V) + As(III)) from the total As concentration (Tot-As). The diluted solution of 8-hydroxyquinoline was used to control chemical interferences potentially caused by metals and also mask the undesired fluorescence emission signal due to As(V) during the determination of all As species (Liao and Deng, 2006). The detection limit of the instrument for As analysis was at 5 ng/L, and the precision was usually <6% on triplicate samples. Total dissolved Fe and Mn in porewaters were determined by Graphite Furnace Atomic Absorption Spectrometry (180–80 HITACHI).

Total organic carbon (TOC) in sediments refers to the total amount of organic substances present in dried sediment samples. Oven-dried samples (40 °C) of sediment (0.5 g weighed precisely) were put into clean pre-weighed crucibles. The samples were placed and heated in a muffle furnace at 750 °C for 4 h then weighed after cooling down in a desiccator. The TOC content was obtained by the difference of sample weight before and after heating. The water content of sediment samples was determined on oven dried samples at 105 °C for 4 h until constant weight. Total reducible sulfur (TRS), defined as the sum of elemental sulfur, pyritic sulfur and acid volatile sulfide (AVS) was obtained after conversion into H_2S in a hot acidic Cr^{2+} solution in the presence of ethanol. The H_2S gas is collected as a ZnS precipitate and ultimately analyzed by iodimetry (Canfield et al., 1986).

For sediment samples, a sequential extraction procedures was a modified version of the techniques developed by Tessier et al. (1979) and described in detail by Liao and Deng (2006). Wet sediment samples were submitted to sequential extraction by (i) a 0.2 M oxalic acid solution buffered to pH 2 with ammonium oxalate for 8 h to remove trace As mainly bound to amorphous Fe and Mn oxyhydroxides (identified as As-oxal); (ii) an acidic solution (pH = 2.0) of H_2O_2 30% (w/w) at 85 °C for 5 h to remove trace As bound to organic matter in the sediments after the first extraction (identified as As–H_2O_2). The As species in both extracts were chemically converted into As(III) and analyzed by FI-HGAAS (Liao and Deng, 2006). Fraction (i) containing also oxidized Fe-oxal and Mn-oxal and fraction (ii) containing reduced Fe–H_2O_2 and Mn–H_2O_2 of the sediments were obtained from the same extractions and determined by GF-AAS.

For the determination of total As, Fe and Mn, all sediment samples including certified reference standard marine sediments BCSS-1 and PACS-1 from the National & Research Council of Canada, were digested with a combination of the concentrated acids HCl, HNO_3 and HF (Suprapure grade), in a microwave oven (Elwaer and Belzile, 1995). The As recovered from this digestion procedure was analyzed after the required pre-reduction an masking step for Tot-As as described by Liao and Deng (2006). For all measured parameters, triplicate samples and standard additions were systematically run to control the quality of the analyses.

3. Results and discussion

3.1. Profiles of dissolved species in porewaters

3.1.1. pH and DO profiles of the SWI

The profiles of DO in the same littoral site were different in the two seasons (data not shown). The concentration of DO in overlying water in summer gradually decreased from 6.3 mg/L at the surface to less than 5.7 mg/L close to the SWI at 5 m deep. The water temperature in July and August was the highest of the whole year and the input of organic matter from the dying littoral vegetation increased into the lake when the water level raised, thus leading to a larger consumption of oxygen. In winter, the water level decreased by 1 m at the same site and DO was at 11.0 mg/L at 50 cm from the SWI but it was very likely much lower closer to
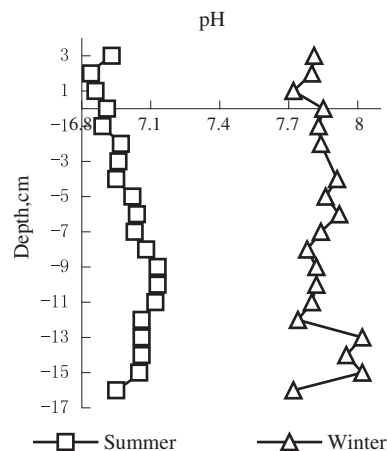

Fig. 2. Profiles of pH in porewaters in summer and winter at the Kangshan site of Poyang Lake.

the SWI as we will discuss later. Although Poyang Lake is subject to high variations of its water level, the total content of phosphorus is relatively constant in the whole lake (Lu, 1993) and DO even in summer remains still higher than 5 mg/L. Variations of pH in porewaters were also season-dependent. In summer, pH values ranged from 6.8 to 7.1 whereas pH fluctuated from 7.7 to 8.0 in winter (Fig. 2).

3.1.2. Distribution of arsenic species in porewaters

Vertical distributions of dissolved TAs, As(III), As(V), and As-org in overlying water and porewater from the SWI to a depth of 16 cm in the sediments of the Kangshan site of Poyang Lake corresponding to summer and winter are shown in Fig. 3. The concentrations of TAs, As(III), As(V), and As-org were in the ranges of 0.8–55.8, 0.7–46.2, 0.3–7.4, 3.5–16.1 µg/L in summer, and 5.5–15.7, 2.0–9.3, 0.5–5.8 and non-detectable–3.1 µg/L in winter, respectively. Average concentrations of TAs, As(III), As(V), and As-org were respectively 16.5, 10.3, 2.5 and 4.2 µg/L in summer, and 7.7, 4.2, 2.6 and 1.0 µg/L in winter. This indicates that 62% of the total dissolved As in porewaters at the sediment–water interface existed as arsenite in summer, compared to 55% in winter. Conditions of DO and As profiles suggest less oxidizing conditions at the SWI during the summer season in comparison to winter and reveal the seasonal character in the geochemical behavior of dissolved arsenic.

The concentrations of As(III), As(V) were below the detection limit (0.05 µg/L) at the SWI (Fig. 3, summer is top panel) suggesting either its oxidation into the oxic layer of the sediment or its precipitation/adsorption on Fe–Mn oxyhydroxides or organic matter present in this sediment layer. Concentrations of As(III) and As(V) increased gradually with depth corresponding to a maximum of 46.2 µg/L at 11.0 cm deep and 7.4 µg/L at 12 cm, respectively, and then both species sharply decreased between 12.0 and 16.0 cm. These maximum concentration peaks of TAs, As(III), As(V) generate a concentration gradient of total dissolved As which can favor upward or downward diffusion with possible precipitation/adsorption reactions in the upper or lower sediment layer or the diffusion of dissolved As species into the water column of the lake.

A comparison of dissolved As(III) and As(V) profiles with those of dissolved Fe(II) and Mn(II) (Fig. 4) shows similar trends and reinforces the possible association of these As species with Fe oxyhydroxides and organic matter. The simultaneous release of dissolved As(III), As(V) along with Fe(II) and Mn(II) under reducing conditions can likely occur as Fe/Mn oxyhydroxides are used as oxidants during the mineralization of organic matter (Stumm and Morgan, 1996).
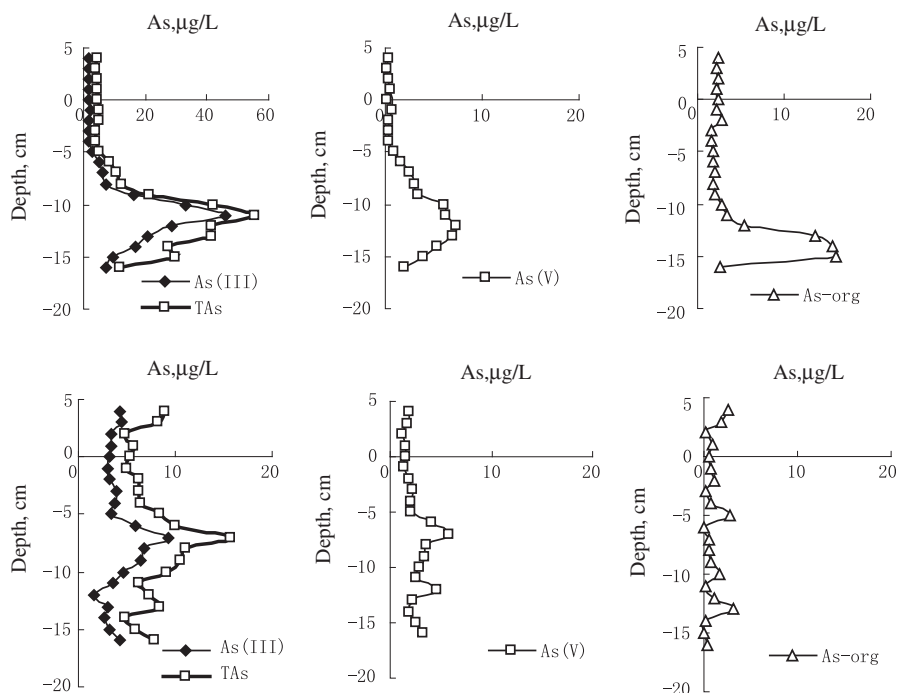
Fig. 3. Vertical profiles on arsenic species in porewaters at the Kangshan site of Poyang Lake in summer (top) and winter (bottom).
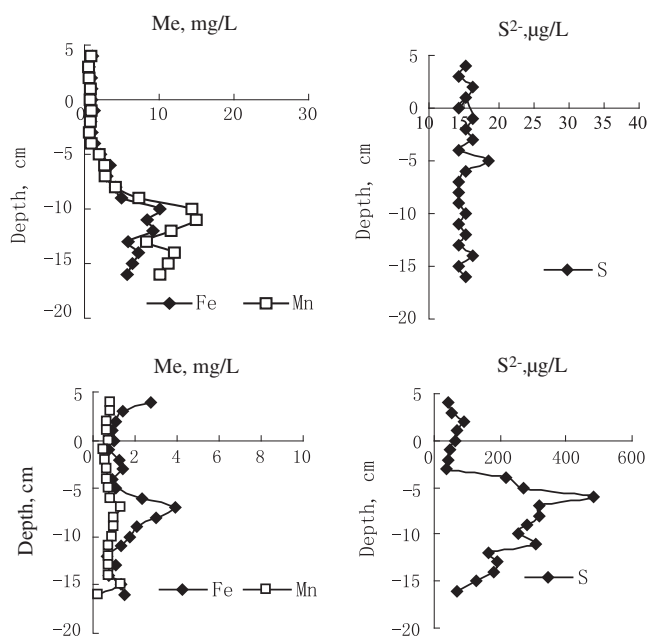

Fig. 4. Vertical profiles on dissolved iron, manganese, sulfide in porewaters at the Kangshan site of Poyang Lake in summer (top) and winter (bottom).

Amorphous iron oxyhydroxides have a pH (zpc) in the range of 7.8–8.8, therefore the surfaces of sediments are expected to be positively charged at the pH (6.8–7.1) at the Kangshan site in summer and to adsorb efficiently the negatively charged anions of arsenate and neutral arsenite. However, Fe–Mn oxyhydroxides are used as oxidizing agent in the process of organic matter mineralization following the depletion of DO in sediment. The processes of decomposition of organic matter are likely accelerated at the higher temperatures of the summer.

The average concentration of dissolved sulfide (S^{2-}) in porewaters was close to 15.0 ng/mL and the profile is shown in Fig. 3 (top panel). It was mainly associated in sediment as pyrite (as discussed later in the sediment section). Statistical correlations (r) of TAs, As(III) and As(V), respectively with Fe(II) or Mn(II) in porewaters are high as 0.91, 0.89 and 0.94 for Fe and 0.92, 0.88 and 0.89 for Mn. However, those of As-org with Fe(II), Mn(II) are only 0.41 and 0.46 respectively, and there are none between S^{2-}, and Fe(II) or Mn(II) ($r < 0.01$). It demonstrates that the absorption of organic As on Fe–Mn oxyhydroxides is weak compared with inorganic species. The importance of Fe oxyhydroxides and the critical role they play in controlling the solubility of As have been often reported (e.g. Belzile and Tessier, 1990; Couture et al., 2010c). Our result suggests that the solubility and migration of inorganic arsenic are highly controlled by Fe–Mn oxyhydroxides.

In winter, the concentrations of TAs, As(III), As(V), and As-org were at 5.3, 3.2, 1.6 and 0.5 μg/L, respectively at SWI. The percentage of As(III), As(V), and As-org in the dissolved arsenic were 60.4%, 30.1% and 9.4%, respectively. The dominance of inorganic arsenic especially As(III), suggests the existence of reducing conditions and very low levels of DO close to the SWI. Such phenomenon of a very thin layer of anoxic water at the SWI has already been observed in one of our previous studies in a Canadian lake (Chen et al., 2003). Some other significant differences with summer profiles that also suggest reducing conditions at the SWI include: concentrations of dissolved species of As (Fig. 3, bottom panel) and dissolved Fe(II) and Mn(II) (Fig. 4, bottom panel) in overlying waters that were all higher. However, the concentrations of dissolved As species and those of Mn(II) and Fe(II) measured in porewaters in winter were much lower than those measured in summer. It was likely related to the much higher concentration of S^{2-} in porewater and overlying water in winter (Fig. 4, bottom panel).

A comparison of vertical profiles of dissolved As species with those of dissolved Fe and Mn in porewaters (Fig. 3, bottom panel) shows similar trends with peaks of Fe, and Mn at 7.0 cm in depth. The statistical correlations (r) of TAs, As(III), As(V) respectively with Fe(II) or Mn(II) in porewater in winter were 0.88, 0.92 and 0.65 and 0.54, 0.53 and 0.46, respectively. A direct comparison between the profiles of dissolved Fe, Mn and inorganic As

in porewater and statistical correlations also corroborates the affinity of As from Fe–Mn oxyhydroxides. It suggests that the distribution of inorganic arsenic was controlled sequentially by Fe oxyhydroxides followed by Mn oxyhydroxides. The simultaneous increase of dissolved Fe(II), Mn(II) and As in the porewaters can again be related to the dissolution of Fe–Mn oxyhydroxides under reducing conditions with the concomitant release of the absorbed As.

Concentrations of dissolved S^{2-} were at 62.3 µg/L at SWI (Fig. 4, bottom panel), increased gradually with depth to a peak value of with 484.0 µg/L at 6.0 cm and then decreased gradually to 71.5 µg/L between 7.0 and 16.0 cm in depth. Statistical correlations (r) of TAs, As(III), As(V) with S^{2-} in porewaters in winter were 0.52, 0.53 and 0.62, respectively, demonstrating that the geochemical behavior of dissolved As in winter can also be possibly controlled by S^{2-} in porewater, which originates from the dissolution of sulfides in sediments.

The maximum concentration of As(III), As(V) peak in porewater generates a concentration gradient of dissolved As which can favor upward or downward diffusion. With upward diffusion, Fe(II) and Mn(II) would be oxidized into Fe(III) and Mn(IV) and then re-precipitated at SWI. Therefore, the concentrations of As(III) and As(V) were low for the absorption of inorganic arsenic by Fe–Mn oxyhydroxides at SWI. Downward diffusion of dissolved As in porewaters can lead to re-precipitation as red orpiment (AsS), arsenic trisulfide (As_2S_3) or its association with amorphous sulfides to form as arsenopyrite (FeAsS) and symplesite ($FeAs_2$), or only adsorption of other solid sulfides, which is discussed in the next section.

3.1.3. Flux of the dissolved arsenic

Profiles of total dissolved As in the two seasons suggest its possible diffusion along the concentration gradient generated by the maximum concentration peaks. As mentioned earlier, the downward diffusion can lead to the formation of arsenic sulfides by precipitation or to the formation of distinct mineral phases containing As. The upward diffusion can lead to the re-precipitation or adsorption of dissolved As with Fe–Mn oxyhydroxides present at the SWI or to the release of dissolved As into the overlying water. Based on the assumption that all dissolved As diffusing upwards is transferred to the overlying water, the diffusive fluxes of dissolved arsenic from sediment porewaters were estimated using the method applied by Ciceri et al. (1992):

$$J = \varphi^3 D_o (d[As]/dz)_o$$

where J is the flux of dissolved As, φ is the sediment porosity (0.49 in summer, 0.32 in winter) calculated from the water content using the following formula (Berner, 1971): $\varphi = V_{water}/V_{water} + V_{solid}$, where V is the volume obtained from mass and density of water and sediment. D_s is the bulk sediment diffusion coefficient and is obtained from $D_s = \varphi^2 D_o$ (Ullman and Aller, 1982) where D_o is the free solution diffusion coefficient for 5.3×10^{-6} cm^2/s (Li and Gregory, 1974); $(d[As]/dz)_o$ is the concentration gradient calculated from the linear portion of total dissolved As concentrations at the SWI in summer and winter respectively (Fig. 4). The estimated fluxes of dissolved As across the SWI in summer and winter using the first Fick's law were 3.88 mg/m² a and 0.79 mg/m² a, respectively. These values were comparable to those obtained in benthic fluxes reported by Millward et al. (1997).

In contrast, the average value of TAs was 16.5 µg/L (3.9–55.8) in porewaters of the Kangshan, Poyang Lake in summer; in comparison this concentration of total dissolved As in Poyang Lake is 3 times that measured in the Thames estuary and 5 times that of Plume (Millward et al., 1997) in the same season. Therefore, it is important to mention that porewaters can represent a secondary
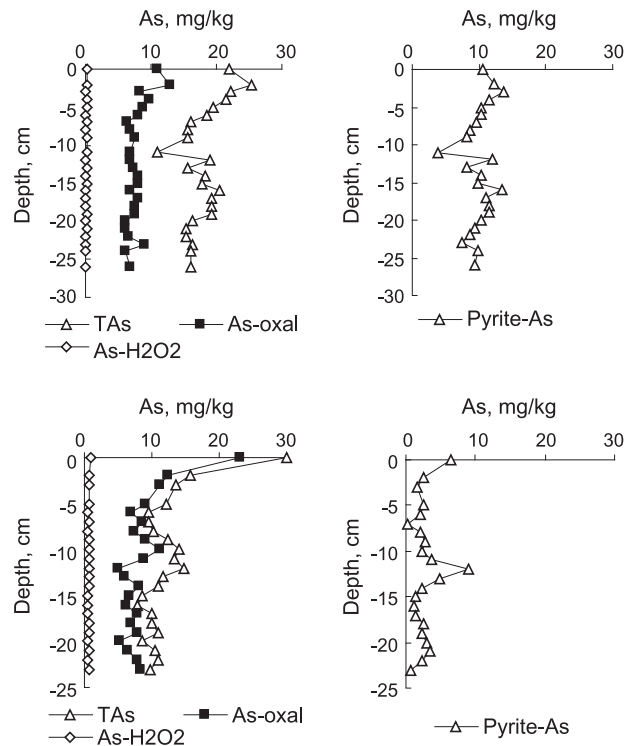

Fig. 5. Fractions of arsenic in sediments of Poyang Lake in summer (top) and winter (bottom).

pollution of As via SWI through the concentration gradient diffusion when environmental conditions change.

3.2. Profiles in sediments

The vertical profiles of total As and As fractions extracted from the sediments of Poyang Lake and the proportion of As associated to pyrite or sulfides estimated from the three sets of data by subtracting the two extracted fractions (As-oxal and As–H_2O_2) from the total are presented in Fig. 5. They clearly indicate a surficial enrichment of As associated to Fe–Mn oxyhydroxides identified as As-oxal in both seasons. A slight enrichment of As associated to pyrite or sulfides and identified as pyrite–As was also noticed in sediments of winter. Concentrations of As associated to organic matter identified as As–H_2O_2 were very low in the two seasons.

Apart from the surficial sediments, concentrations of As associated to organic matter (As–H_2O_2) represented only a minor fraction of the total with average values of 0.5–1% (in summer) and 5–8% (in winter), respectively. However, 40–45% of As in summer and 68–72% in winter was associated to the Fe–Mn oxyhydroxides fraction. The proportions of As associated to pyrite or sulfides were in average about 55% in summer and 20–25% in winter of total As in sediments. Comparison of the profiles As in sediments and dissolved As in porewaters reasonably shows that As concentrations in sediment gradually decreased (reaching the lowest level at −11.0 cm in summer and at −7.0 cm in winter) and whereas those of dissolved As in porewaters generally increased from surface to the same depths (highest at −11 cm and −7.0 respectively). This clearly shows a dynamic transport of dissolved As in porewaters from sediments to SWI accentuated by seasonal variations. Distributions of total Fe/Mn, Fe/Mn oxyhydroxides identified as Fe(oxal)/Mn(oxal), and Fe/Mn associated to organic matters as Fe(H_2O_2)/Mn(H_2O_2) in sediments (Fig. 6) are similar with the profiles of As in sediment (Fig. 5), and show that the fractions of Fe and Mn identified as oxyhydroxides, Fe-oxal/Mn-oxal were slightly higher at
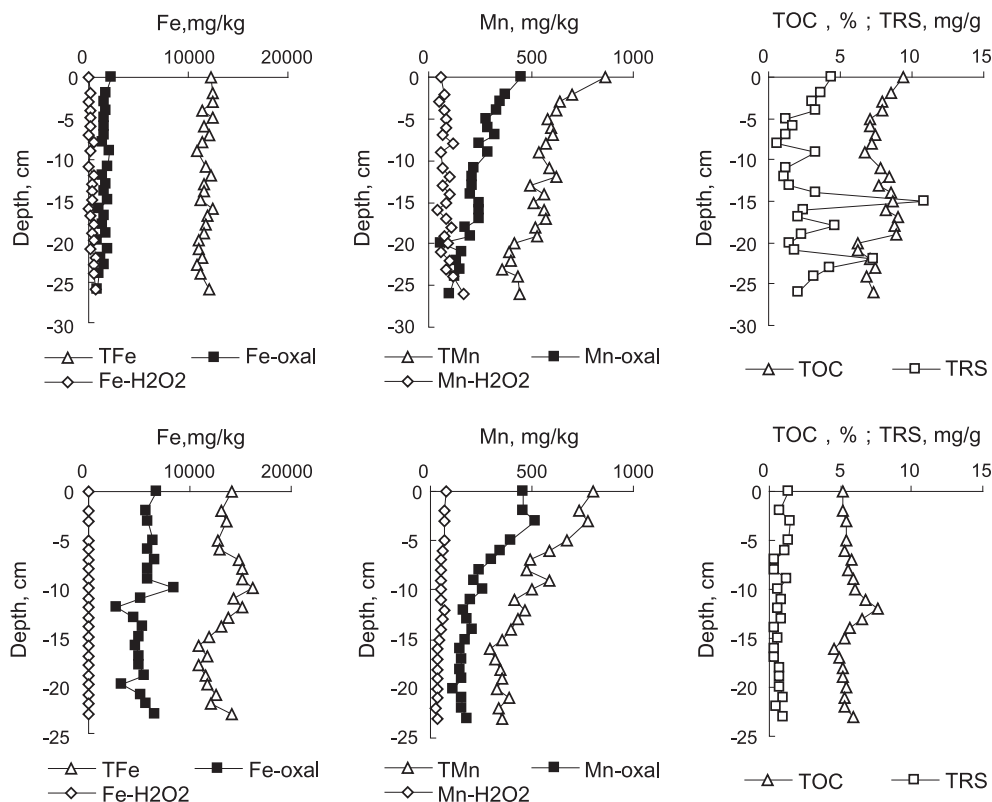
Fig. 6. Fractions of iron and manganese and profiles of TOC and TRS in sediments of Poyang Lake in summer (top) and winter (bottom).

SWI although the enrichment of Mn was more obvious. The concentrations of Fe and Mn associated to organic matter were relatively low like those of As–H_2O_2. Fig. 6 also presents the vertical distributions of TOC and TRS in the two seasons. The contents of TOC in sediments of Poyang Lake generally <5% (w/w) were lower than those reported for Canadian lakes (Chen et al., 2003). The depth-distribution trends of TRS and pyrite–As in sediment for the two seasons were particularly similar and statistical correlations between those two parameters were 0.89 and 0.95 in summer and winter, respectively. In summer however, statistical correlations between TAs or As-oxal and TMn or Mn-oxal were stronger (0.6, 0.7) than those calculated between As and Fe fractions (0.2, 0.3) while the r values between TAs or As–H_2O_2 and TOC or TRS were low (0.29, 0.14; 0.37, 0.04, respectively), suggesting that Mn oxyhydroxides could be more likely the main factor controlling the solubility of As at the SWI as discussed earlier for porewaters in summer. In winter, relatively strong statistical correlations were calculated between TAs or As-oxal between TFe, TMn, TRS (0.35, 0.67, 0.59; 0.56, 0.7, 0.55). Those results indicated that the mobility of arsenic was more likely controlled by amorphous Fe–Mn oxyhydroxides and sulfides existing in sediments in winter. The variability of all profiles between the two seasons indicates that a large dynamism is imposed on the geochemical behavior of As in Poyang Lake.

4. Conclusions

The concentrations of dissolved As species including total As, arsenite, arsenate and organic As in sediments of Poyang Lake in summer were all higher than those measured in winter. Seasonal profiles of dissolved As suggested downward and upward diffusion, and the flux of dissolved arsenic across the SWI in summer and winter were estimated to 3.88 mg/m² a and 0.79 mg/m² a, respectively. Based on porewater profiles and sediment phase data,

it has been shown that the geochemistry of As was controlled by several important processes such as adsorption/desorption, precipitation and diffusion. Chemical species such as Fe–Mn oxyhydroxides, sulfides, organic matter and TRS appear to play an important role in the geochemistry of arsenic in sediments. The solubility and migration of inorganic As are controlled by Fe–Mn oxyhydroxides in summer whereas those are more likely controlled by amorphous Fe–Mn oxyhydroxides and sulfides in winter. All profiles measured at the SWI of Poyang Lake especially those of dissolved constituents clearly indicate that the lake is a very dynamic environment with large fluctuations over the seasons. More studies are needed to better understand and manage the overall system of Poyang Lake.

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