

# Mercury speciation, distribution, and bioaccumulation in a river catchment impacted by compact fluorescent lamp manufactures

Peng Liang<sup>1,2,3</sup> · Xinbin Feng<sup>2</sup> · Qiongzi You<sup>1</sup> · Jin Zhang<sup>1,3</sup> · Yucheng Cao<sup>1,3</sup> · Anna Oi Wah Leung<sup>4</sup> · Shengchun Wu<sup>1,3</sup>

Received: 17 July 2015 / Accepted: 1 February 2016 / Published online: 22 February 2016  
© Springer-Verlag Berlin Heidelberg 2016

**Abstract** The influence from the manufacturing of compact fluorescent lamps (CFL) on mercury (Hg) speciation and distribution in river catchments nearby a typical CFL manufacturing area in China was investigated. Water, sediment, river snail (*Procambarus clarkii*), and macrophyte (*Paspalum distichum* L.) samples were collected. Total Hg (THg) and methylmercury (MeHg) concentrations in water ranged from 1.06 to 268 ng · L<sup>-1</sup> and N.D. –2.14 ng · L<sup>-1</sup>, respectively. MeHg was significantly positively correlated with THg in water. THg and MeHg in sediment ranged from 15.0 to 2480 and 0.06 to 1.85 ng · g<sup>-1</sup>, respectively. River snail samples exhibited high concentrations of THg (206–1437 ng ·

g<sup>-1</sup>) and MeHg (31.4–404 ng · g<sup>-1</sup>). THg and MeHg concentrations in root of *P. distichum* L. were significantly higher than those in shoot, indicating that THg and MeHg in the plant were mainly attributed to root assimilation. A very high bioaccumulation factor (20.9 ± 22.1) for MeHg in *P. distichum* L. was noted, suggesting that *P. distichum* L. might have a potential role in phytoremediating MeHg contaminated soil due to its abnormal uptake capacity to MeHg.

**Keywords** Mercury contamination · Methylmercury · *Procambarus clarkii* · *Paspalum distichum* L. · Aquatic ecosystem

Responsible editor: Philippe Garrigues

**Capsule abstract** Compact fluorescent lamp manufacturing activities result in Hg pollution in local river catchment. *Paspalum distichum* L. might be potentially useful for the accumulation of MeHg.

✉ Xinbin Feng  
fengxinbin@vip.skleg.cn

✉ Shengchun Wu  
shengchunwu@126.com

<sup>1</sup> School of Environmental and Resource Sciences, Zhejiang Agricultural and Forest University, Lin'an, Zhejiang Province 311300, People's Republic of China

<sup>2</sup> State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, People's Republic of China

<sup>3</sup> Key Laboratory of Soil Contamination Bioremediation of Zhejiang Province, Hangzhou 311300, China

<sup>4</sup> Department of Biology and Croucher Institute for Environmental Sciences, Hong Kong Baptist University, Hong Kong, People's Republic of China

## Introduction

Mercury (Hg) is one of the most toxic pollutants in the world. Both natural and anthropogenic activities contribute to the Hg burden in the environment, such as volcanic explosion and Hg industrial activities. So far, Hg pollution has been drawing increasing concern worldwide; Hg can stay in the atmosphere for about 0.5–2 years (Fu et al. 2010) and can be transported to remote areas without Hg emission sources (Fitzgerald et al. 1998). In addition, inorganic Hg can be converted to methylmercury (MeHg), a more toxic and bioaccumulative Hg compound, especially in anoxic aquatic environment. Some bacteria (e.g., sulfate/iron-reducing bacteria) are regarded as the principal agents responsible for MeHg production in the sediments of water bodies (King et al. 2000). It is believed that accumulation of MeHg in aquatic systems may continue to pose a threat to fish and other biota, including humans. However, there are few promising technologies available to deal with Hg pollution in aquatic system.

Compact fluorescent lamps (CFL) rely on Hg as a source of ultraviolet radiation for the production of visible light. Extremely high Hg concentrations in air, up to  $16,000 \text{ ng} \cdot \text{m}^{-3}$ , have been reported in CFL manufacturing zone, resulting in a severe exposure of CFL workers to Hg vapor through inhalation (Xu et al. 2007). Our previous study has also demonstrated that CFL manufacturing activities may lead to Hg contamination of the local soil and agricultural products (Shao et al. 2012). Moreover, elevated concentrations of THg and MeHg in local cultivated rice samples were observed when compared with the commercial rice samples from the markets. It was doubted that the two streams, serving as the major source of irrigation water for the local paddy farmland, may have been contaminated by Hg when passing through the CFL manufacture zone (Liang et al. 2015). However, detailed information is still lacking in terms of the concentrations and speciation of Hg in the different compartments of local aquatic environment, including water, sediment, aquatic animals and macrophytes. Based on our previous study, we hypothesized that (i) Hg emitted from CFL manufacture may result in a severe Hg pollution in the ambient waterbody via wastewater discharge and wet/dry deposition, and (ii) part of inorganic Hg entering into the aquatic environment may be converted to MeHg in the sediment and then accumulated in organisms such as macrophyte and river snail. Therefore, THg and MeHg concentrations in surface water, sediment, river snails, and aquatic plants from local watershed were examined in this study. Moreover, concentrations of particulate Hg (PHg), dissolved Hg (DHg), particulate methylmercury (PMeHg), and dissolved MeHg (DMeHg) in surface water were also determined. The

objectives of this study were (i) to figure out the distribution of Hg species in surface water and sediment in river catchments, (ii) to determine the contamination levels in terms of THg and MeHg in river snails and macrophytes near the CFL manufacturing zone, and (iii) to screen potential aquatic plants for phytoremediating Hg-contaminated sites.

## Materials and methods

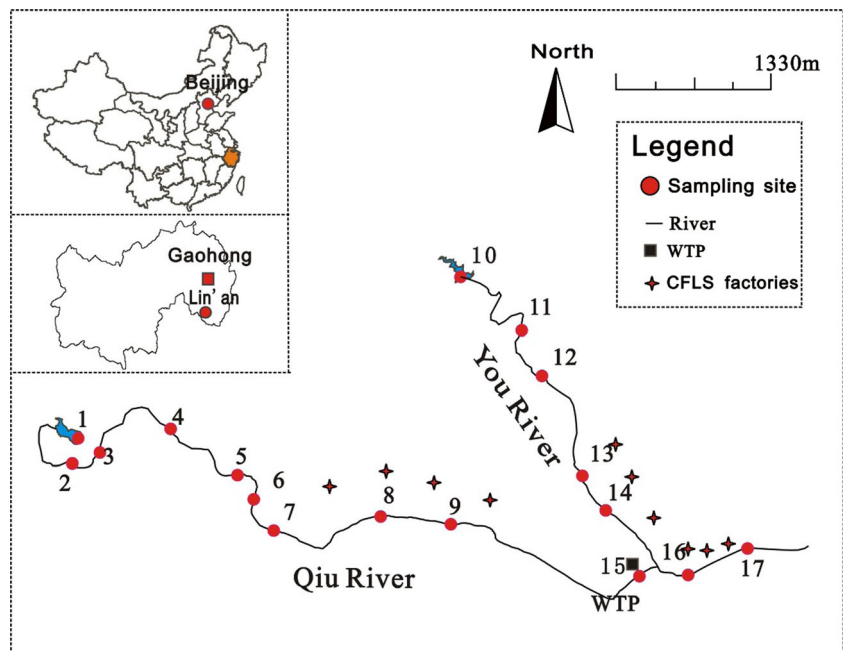
### Sampling area

Gaohong Town ( $30^{\circ}19'N$ ,  $119^{\circ}40'E$ ) is located in northern Zhejiang Province, eastern China. It is commonly known as the “land of CFL” because there are more than 180 CFL manufacturing factories in this small town, accounting for 20 % of the global production in 2010 (SBZP 2011). There are two streams namely Qiu stream and You stream passing through Gaohong Town. A wastewater treatment plant (WTP) is located at the confluence of the two streams. Most of the CFL factories are situated along Qiu stream and downstream of the wastewater treatment plant. Fewer factories are distributed along You stream (Fig. 1).

### Sample collection

A total of 17 sampling sites were selected along the two streams according to the spatial distribution of the CFL factories (as shown in Fig. 1). Sampling sites 1 to 6 and 10 to 12 were located at the upstream of Qiu stream and You stream, where fewer CFL manufacturing factories were located.

**Fig. 1** The sampling sites used in this study



Sampling sites 7 to 9 and 13 to 14 were closed to the zone where the CFL were densely concentrated. Sampling sites 15 to 17 were located at the confluence of two streams and the downstream.

River water samples were collected three times in September 2013, July 2014, and August 2014. All the water samples were acidified with ultra-pure HCl to a final acid concentration of 0.4 % (v/v) within 24 h after collection.

Sediment, river snail (*P. clarkia*), and macrophyte (*P. distichum* L.) samples were collected from the same sampling sites as the water samples. However, river snail samples were only available at nine sites. All samples were stored in double-sealed polyethylene bags to avoid cross contamination. After returning to laboratory, macrophyte samples were washed by tap water and then rinsed by miliQ water three times. The roots and shoots were separated for further analyses. The river snail samples were washed by tap water and then rinsed by miliQ water for three times before the shells were removed. All of the solid samples were freeze-dried, ground, and homogenized for further analyses.

**Sample analyses**

THg concentrations in water samples were determined by BrCl oxidation, SnCl<sub>2</sub> reduction, purging, gold trapping, and cold vapor atomic fluorescence spectrometry (CVAFS) detection according to USEPA Method 1631 (Feng et al. 2009, USEPA 2002). THg in sediment, macrophyte, and river snail samples were determined with a direct Hg analyzer (Lumix, Russia) based on thermal decomposition, amalgamation, and atomic absorption spectrophotometry, following the US EPA Method 7473 (Li et al. 2011, USEPA 2007).

MeHg in water was measured following USEPA Method 1630 (USEPA 2001). An aliquot of 50-mL sample was placed in a 60-mL fluoropolymer distillation vessel. The distillation was carried out at 125 °C under Hg-free N<sub>2</sub> flow until approximately 45 mL of water was collected in the receiving vessel. MeHg in soil and sediment was extracted with HNO<sub>3</sub> and CuSO<sub>4</sub> and then leached with dichloromethane and back-leached with water (Feng et al. 2009, Liang et al. 2004).

MeHg concentrations in hydrophyte samples and river snail were analyzed after a digestion using KOH–methanol/solvent extraction. MeHg was determined using aqueous ethylation, purging, and gas chromatography–CVAFS detection (Books Rand), following USEPA Method 1630 (Meng et al. 2011, USEPA 2001).

pH and Eh values of water samples were determined by the portable pH and Eh meter (SX751, Luheng, China) in field.

**QA/QC**

Quality assurance and quality control of THg and MeHg determinations were carried out using duplicates, method blanks, matrix spikes, and certified reference materials. The detailed information is shown in Table 1.

**Data analyze**

Differences of Hg speciation and concentrations in environmental samples among all the sites were performed by one-way ANOVA (data could be transformed to obtain equal variance) by SPSS 16.0 for windows. All of figures were performed by Sigma plot 10.0.

**Results and discussion**

**Hg speciation in river water**

Table 2 shows the concentration of THg, MeHg, DHg, and PHg in the water samples. THg concentration in water samples exhibited a wide range of 1.06 to 268 ng · L<sup>-1</sup> with a mean value of 15.8 ± 44.6 ng · L<sup>-1</sup>. THg concentrations in Qiu stream (4.82 ± 3.73 ng · L<sup>-1</sup>) were slightly higher than those in You stream (3.43 ± 1.69 ng · L<sup>-1</sup>). Peak concentrations of THg were noted at the sampling sites near the wastewater treatment plant and sites downstream of the plant (sites 15, 16, and 17) and were significantly higher (*p* < 0.01, SNK test) than the THg concentrations at the upstream sites of two streams.

**Table 1** Detailed information for QA/QC results

Analysis items	THg				MeHg			
	Surface water	Sediment	Aquatic plant	River snails	Surface Water	Sediment	Aquatic plant	River snails
LOD	0.01 ng · L <sup>-1</sup>	0.5 ng · g <sup>-1</sup>	0.5 ng · g <sup>-1</sup>	0.5 ng · g <sup>-1</sup>	0.01 ng · L <sup>-1</sup>	0.008 ng · g <sup>-1</sup>	0.07 ng · g <sup>-1</sup>	0.07 ng · g <sup>-1</sup>
LOQ	0.05 ng · L <sup>-1</sup>	1.5 ng · g <sup>-1</sup>	1.5 ng · g <sup>-1</sup>	1.5 ng · g <sup>-1</sup>	0.05 ng · L <sup>-1</sup>	0.040 ng · g <sup>-1</sup>	0.35 ng · g <sup>-1</sup>	0.35 ng · g <sup>-1</sup>
CRMs		GBW-0730-5a (GSD-5a)	GBW-10020	NRCC-Tort-2		ERM-CC580	NRCC-Tort-2	NRCC-Tort-2
Certified values		290 ± 30	150 ± 20	270 ± 60		77.5 ± 3.7	152 ± 30	152 ± 30
Determined values ( <i>n</i> = 5)		300 ± 8.16	147.33 ± 3.44	272.67 ± 13.19		83.09 ± 0.14	149 ± 9.66	149 ± 9.66

**Table 2** Concentrations of THg, DHg, PHg, and MeHg (ng/L) in surface water of Qiu (QS) and You (YS) streams

Site	Location	Sampling date													
		Sep. 2013						Jul. 2014		Aug. 2014					
		THg	DHg	PHg	TMeHg	DMeHg	PMeHg	THg	MeHg	THg	DHg	PHg	TMeHg	DMeHg	PMeHg
1	QS upstream	1.06	0.53	0.52	0.78	0.76	0.02	4.17	0.07	5.18	3.39	1.80	0.05	0.02	0.03
2	QS upstream	2.64	0.94	1.69	0.06	0.05	0.01	5.53	0.11	3.75	3.38	0.37	0.12	0.09	0.03
3	QS upstream	2.13	0.92	1.21	0.26	0.2	0.06	4.37	0.09	4.82	2.84	1.99	0.11	0.07	0.04
4	QS upstream	1.53	0.91	0.62	0.07	0.03	0.04	2.83	0.06	4.17	2.03	2.14	0.05	0.02	0.03
5	QS upstream	2.14	1.02	1.12	0.13	0.09	0.04	9.79	0.10	3.47	2.13	1.33	–	–	–
6	QS upstream	2.20	0.81	1.39	0.16	0.07	0.09	6.08	0.12	2.67	2.40	0.28	0.06	0.03	0.03
7	QS CFLs factories	2.50	1.32	1.18	0.14	0.12	0.02	4.96	0.11	3.55	2.84	0.70	0.07	0.03	0.04
8	QS CFLs factories	5.51	2.33	3.18	0.23	0.23	N.A.	18.78	0.32	3.24	2.37	0.88	0.07	0.04	0.03
9	QS CFLs factories	2.96	0.32	2.64	0.19	0.03	0.16	12.08	0.31	8.19	3.23	4.95	0.19	0.06	0.13
10	YS upstream	4.36	1.58	2.78	0.08	0.02	0.06	3.60	0.16	1.78	1.30	0.48	0.04	0.02	0.02
11	YS upstream	1.96	1.88	0.08	0.08	0.04	0.04	2.68	0.03	3.09	0.97	2.11	0.03	0.02	0.01
12	YS upstream	1.98	1.89	0.09	0.08	0.02	0.06	2.58	0.02	7.09	1.53	5.56	0.04	0.02	0.02
13	YS CFLs factories	2.39	1.36	1.03	0.09	0.03	0.06	3.87	0.03	2.89	1.58	1.31	0.04	0.02	0.02
14	YS CFLs factories	4.31	2.26	2.05	0.14	0.02	0.12	7.00	0.03	1.91	1.15	0.75	–	–	–
15	WTP	11.20	7.16	4.04	0.73	0.50	0.23	98.47	0.68	9.77	4.46	5.31	0.13	0.12	0.01
16	Downstream of WTP	38.29	14.76	23.53	1.24	1.17	0.07	164.88	1.68	12.94	9.99	2.95	0.29	0.13	0.16
17	Downstream of WTP	13.90	4.8	9.1	1.17	0.37	0.80	268.40	2.14	7.25	4.52	2.73	0.85	0.28	0.57

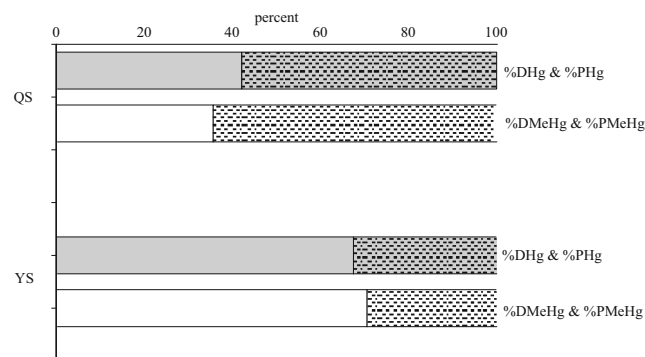
WTP wastewater treatment plant

Although most of the CFL factories are located along Qiu stream, there is no significant difference in THg concentration between You and Qiu streams. This is because the wastewater containing Hg exhausts from CFL factories was usually discharged into the wastewater treatment plant through municipal pipelines before discharged to the stream at the confluence. Thus, it led to significantly higher THg concentrations in the water samples collected near the treatment plant and the downstream sites (sites 16 and 17). Fortunately, the THg concentrations in all the water samples from the watershed of Gaohong town were still lower than  $1000 \text{ ng} \cdot \text{L}^{-1}$ , the upper limit of type V water quality in accordance with the Chinese national standard. It means the water could be used for agricultural purposes.

MeHg concentrations in water samples ranged from N.D.  $-2.14 \text{ ng} \cdot \text{L}^{-1}$ . The highest MeHg concentrations were observed at sites 16 and 17 as well. In general, a significantly positive correlation ( $r=0.825$ ,  $p<0.001$ ,  $n=49$ ,  $T$  test) was observed between MeHg and THg concentrations in all the surface water samples. This result is in line with Ullrich et al. (2001) who reported that a higher MeHg concentration may appear in a waterbody with lower pH and Eh value as well as a higher THg concentration. In the present study, no significant differences ( $p>0.05$ , SNK test) were observed among all the water samples with respect to Eh (182–205 mv) or pH value (7.8–8.4). However, it is not surprising that THg

concentrations at sites 16 and 17 were significantly higher ( $p<0.01$ , SNK test) than those at the other sites, resulting in high MeHg concentrations observed there as well.

The ratios of DHg and PHg to THg (i.e., %DHg and %PHg) and the ratios of DMeHg and PMeHg to TMeHg (i.e., %DMeHg and %PMeHg) were different in the two streams. Higher %DHg and %DMeHg were observed in You stream than in Qiu stream. In contrast, higher ratios of %PHg and %PMeHg in Qiu stream were noted compared to You stream (Fig. 2). These differences may be attributed to the differences in the riverbed of the two streams. Most of the riverbed of You stream



**Fig. 2** Ratios of DHg and PHg to THg (%DHg and %PHg) and the ratios of DMeHg and PMeHg to TMeHg (%DMeHg and %PMeHg) in the two streams

was lined with cement whereas the riverbed of Qiu stream was unaltered (mainly sediment). Therefore, the amount of suspended particles in Qiu stream was higher than that in You stream due to the presence of sediment. Higher concentrations of suspended particles resulted in higher ratios of %PHg and %PMeHg, as the partition coefficient between solid and liquid phases of Hg and MeHg reached  $10^3$ – $10^5$  and  $10^{1.5}$ – $10^{3.5}$ , respectively (Zhong and Wang. 2011).

It was clearly observed that the THg concentrations in water samples that were collected in July 2014 were significantly higher ( $p < 0.01$ , SNK test) than in the samples collected on the other two occasions, especially in the case of water samples near the wastewater treatment plant and downstream. Moderate to heavy rain was encountered during the sampling day in July 2014. Elevated Hg concentrations in water samples might be attributed to surface runoff from both sides of the river or illegal effluent discharge. However, there was no significant increase in MeHg concentration in water samples among all the three sampling times. It is therefore likely that effluent discharge might be responsible for the elevated Hg concentrations on days with heavy rain.

**Hg speciation in sediment**

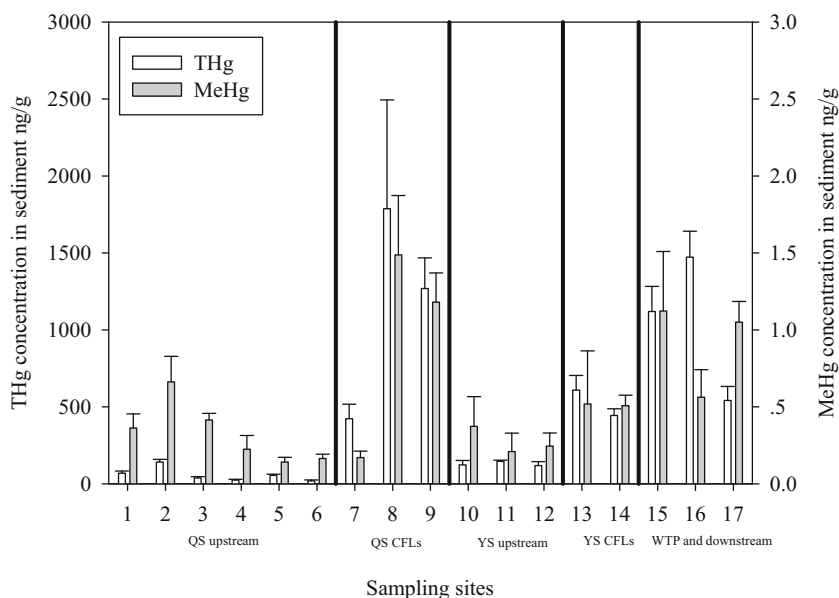
Figure 3 shows the distribution of THg and MeHg in surface sediment. The mean concentrations of THg in sediment for all of the sampling sites varied from 15.0 to 2480  $\text{ng} \cdot \text{g}^{-1}$ . THg concentrations in sediment samples collected at the upstream of the two rivers ( $81.4 \pm 50.4 \text{ ng} \cdot \text{g}^{-1}$ ) were significantly lower ( $p < 0.01$ , SNK test) than those collected at CFL factories (906

$\pm 626 \text{ ng} \cdot \text{g}^{-1}$ ) and the downstream sites of treatment plant ( $1044 \pm 421 \text{ ng} \cdot \text{g}^{-1}$ ). Elevated THg concentrations in sediment were observed at sites 8, 9, 15, and 16, which were located near CFL factories and the wastewater treatment plant. MacDonald et al. (2000) have evaluated the threshold effect concentration (TEC) and probable effect concentration (PEC) for establishing freshwater sediment quality guidelines. TEC values refer to those below which adverse effects are not expected to occur, while PEC values refer to those above which adverse effects are expected to occur more often. TEC and PEC values for Hg in freshwater sediment are 180 and 1060  $\text{ng} \cdot \text{g}^{-1}$ , respectively. In the present study, THg concentrations at all of the CFL factory sites were higher than TEC, and even higher than PEC at sites 8, 9, 15, and 16. Therefore, this indicates that CFL manufacturing sites could potentially cause adverse effects to ambient aquatic environment.

Compared with other recent studies conducted in different industrial areas, the THg concentrations in river sediment near CFL plants were higher than those in sediments adjacent to a coal-fired power plants (55 to 201  $\text{ng} \cdot \text{g}^{-1}$ ) (Liang et al. 2010), but were lower than those near chlor-alkali plants (110 to 617,000  $\text{ng} \cdot \text{g}^{-1}$ ) (Ullrich et al. 2007), organic chemical plants (260 to 38,900  $\text{ng} \cdot \text{g}^{-1}$ ) (Yan et al. 2008), and Zn smelters (150 to 15,400  $\text{ng} \cdot \text{g}^{-1}$ ) (Zheng et al. 2011).

MeHg concentration in sediment ranged from 0.06 to 1.85  $\text{ng} \cdot \text{g}^{-1}$  and is of top concern in local society because of its high bioavailability and toxicity. Similar to the distribution pattern of THg, the highest MeHg concentrations were noted at sites 8, 9, 15, and 17, which were also close to the CFL factories and the wastewater plant. A significant correlation between MeHg and THg

**Fig. 3** THg and MeHg concentrations in sediment



**Table 3** THg, MeHg, %MeHg, and BAF of the river snail samples

Site	Location	THg (ng/g)	MeHg (ng/g)	%MeHg	BAF(THg)	BAF(MeHg)
5	QS upstream	244.75	63.41	26	4.49	450.51
6	QS upstream	329.00	52.42	16	17.09	318.73
7	QS CFLs factories	424.00	159.26	38	1.00	934.67
8	QS CFLs factories	1436.67	335.98	23	0.80	284.55
9	QS CFLs factories	1172.25	170.79	15	0.92	114.81
13	YS CFLs factories	205.67	31.38	15	0.35	60.54
14	YS CFLs factories	378.00	84.23	22	0.85	165.86
15	WTP	1286.67	403.72	31	1.15	359.64
17	Downstream of WTP	1313.00	389.72	30	2.42	371.09

concentrations in sediments was observed ( $r=0.714$ ,  $p<0.01$ ,  $T$  test), indicating the impact of THg concentration on MeHg production in sediment.

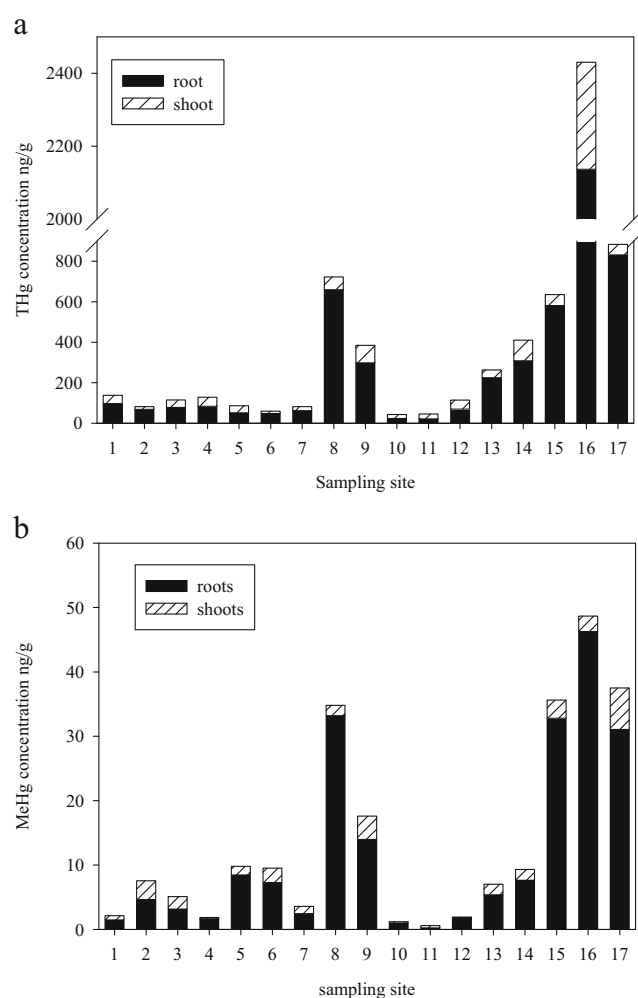
### Accumulation of Hg in the river snails

THg concentrations in the river snails ranged from 206 to 1437  $\text{ng}\cdot\text{g}^{-1}$ . In comparison, MeHg concentrations ranged from 31.4 to 404  $\text{ng}\cdot\text{g}^{-1}$  (Table 3). It is well known that MeHg can accumulate in aquatic organisms through biomagnification. About 75 to 90 % of THg was MeHg in some carnivorous fish species (Liang et al. 2011). However, river snails do not belong to the top level of the food web and are not carnivorous. The percentage of MeHg to THg in the river snails accounted for only 14.6–37.6%. Shao et al. (2012) reported that THg and MeHg concentrations in fish from the local market in Gaohong, and ranged from only 14 to 170 and from 11 to 143  $\text{ng}\cdot\text{g}^{-1}$ , respectively; which were significantly lower than those in the river snails that lived in sediment. Furthermore, THg and MeHg concentrations in fish samples from the abandoned Hg mines in China ranged from 61 to 680 and from 24 to 98  $\text{ng}\cdot\text{g}^{-1}$ , respectively (Qiu et al. 2009). These values were also lower than those observed in the present study.

We found that THg concentrations in the river snails from CFL manufacturing areas were also significantly higher than those in Lengshui River, Hunan province, China and in a reservoir close to an antimony ore (Fu et al. 2011). The river snails were thus highly contaminated with Hg at the CFL manufacturing areas.

Bioaccumulation is a process in which a chemical substance is absorbed in an organism thorough any routes of exposure, i.e., dietary and ambient environmental sources. This process is measured using the bioaccumulation factor (BAF) which is defined as the ratio of the concentration of a chemical accumulated inside an organism (from food and direct exposure) to the concentration in the surrounding environment (Ivanciuc et al. 2006). In this study, BAF of the river snail for MeHg ( $340\pm 257$ ) was 105 times higher than that for

THg ( $3.23\pm 5.34$ ). Moreover, significantly positive correlations for THg ( $r=0.705$ ,  $p<0.05$ ,  $n=9$ ,  $T$  test) and MeHg ( $r=0.907$ ,  $p<0.01$ ,  $n=9$ ,  $T$  test) were noted between the river snails and sediment. These results suggested that sediment was the major source of THg and



**Fig. 4** THg (a) and MeHg (b) concentrations in the roots and shoots of *Paspalum distichum* L.

**Table 4** Pearson correlation coefficients among THg in the sediment, MeHg in the sediment, THg in the roots, MeHg in the roots, THg in the shoots, and MeHg in the shoots ( $n = 17$ )

	THg in sediment	MeHg in sediment	THg in the roots	MeHg in the roots	THg in the shoots
MeHg in the sediment	0.806 <sup>b</sup>				
THg in the roots	0.699 <sup>b</sup>	0.403			
MeHg in the roots	0.824 <sup>b</sup>	0.683 <sup>b</sup>	0.890 <sup>b</sup>		
THg in the shoots	0.599 <sup>a</sup>	0.207	0.914 <sup>b</sup>	0.693 <sup>b</sup>	
MeHg in the shoots	0.806 <sup>b</sup>	0.603 <sup>b</sup>	0.410	0.572 <sup>a</sup>	0.180

<sup>a</sup> Correlation significant at 0.05 level

<sup>b</sup> Correlation significant at 0.01 level

MeHg in the river snails, and MeHg was more readily bioaccumulated in the river snails compared to THg.

**Accumulation of Hg in *P. distichum* L.**

THg and MeHg in the roots and shoots of *P. distichum* L. are shown in Fig. 4. Elevated root THg and MeHg concentrations were observed at the sampling sites close to the CFL factories and the wastewater treatment plant, e.g., sites 8, 9, 15, 16, and 17. Table 4 shows the correlation between Hg concentration in sediments and plant tissues. THg concentrations in sediment were significantly positively correlated ( $p < 0.01$ ,  $T$  test) with THg contents in roots, indicating that Hg in *P. distichum* L. roots was derived mainly from the sediment. However, the correlations between THg in the roots and shoots were not significant. It had been demonstrated that most of the Hg in plant roots is unlikely to be transported to the shoots because Hg in the roots is bound to different proteins and phytochelatins (Carrasco-Gil et al. 2013, Greger et al. 2005, Krupp et al. 2009). Moreover, THg concentration in the shoots of plant samples was related to THg in air as plant leaves can accumulate Hg from the air (Rutter et al. 2011, Vannini et al. 2014). Hyper-accumulator plants for heavy metals (e.g., nickel, copper, cadmium, and arsenic) have been reported to exist (Liu et al., 2007); however, so far, no plants have been identified as hyper-accumulator of Hg (Wang et al. 2012). In this study, BAF of THg for *P. distichum* L. was  $1.00 \pm 0.93$  (0.15–3.42). More importantly, the THg concentration in the shoot of *P. distichum* L. was lower than that in the root; therefore, *P. distichum* L. cannot be regarded as a hyper-accumulator of Hg.

Although numerous studies have reported THg accumulation in different plant samples, few have reported on the MeHg concentrations in different plant tissues and the accumulation of MeHg in plants. Meng et al. (2011) suggested that rice (*Oryza sativa*. L) can accumulate MeHg from soil through root accumulation and the BAF of MeHg were found to range from 2.5 to 6 in different treatments. In the present study, MeHg concentrations in sediment were significantly positively correlated with MeHg in the roots. Moreover, MeHg contents in the roots were also significantly positively

correlated with MeHg in the shoots. These results indicate that MeHg in the roots of *P. distichum* L. was derived mainly from the sediment and MeHg could be further translocated to the shoots. More importantly, the BAF of MeHg in *P. distichum* L. roots reached  $20.9 \pm 22.1$  (1.08–82.1), which was significantly higher than that for the rice samples (Meng et al., 2011). These results indicate that *P. distichum* L. may be used as a potential hyper-accumulator, but the process of MeHg accumulation in the plant requires further studies.

**Conclusions**

CFL manufacturing activities indeed resulted in Hg pollution of the local environment. High Hg contamination in river water and sediment was observed at the sites downstream of the wastewater treatment plant where the three CFL factories were also located. High THg and MeHg concentrations were observed in the river snails, reaching  $754 \pm 527$  and  $187 \pm 149$  ng g<sup>-1</sup>, respectively. THg contamination in *P. distichum* L. was also noted in CFL manufacture areas around the treatment plant. An elevated BAF of MeHg in *P. distichum* L. was noted, indicating that *P. distichum* L. has a potential role to be used in phytoremediating the sites contaminated by MeHg.

**Acknowledgments** Financial support from the Innovative Team Foundation of Zhejiang Province (2013TD12), the National Natural Science Foundation of China (No. 21307114, 21577130), and Zhejiang Provincial Public Techniques Research and Social Development Project (2015C33050) are gratefully acknowledged.

**References**

Carrasco-Gil S, Siebner H, LeDuc DL, Webb SM, Millán R, Andrews JC, Hernández LE (2013) Mercury localization and speciation in plants grown hydroponically or in a natural environment. *Environ Sci Technol* 47:3082–3090

Feng X, Jiang H, Qiu G, Yan H, Li G, Li Z (2009) Mercury mass balance study in Wujiangdu and Dongfeng Reservoirs, Guizhou, China. *Environ Pollut* 157:2594–2603

- Fitzgerald WF, Engstrom DR, Mason RP, Nater EA (1998) The case for atmospheric mercury contamination in remote areas. *Environ Sci Technol* 32:1–7
- Fu XW, Feng XB, Zhang G, Xu WH, Li XD, Yao H, Liang P, Li J, Sommar J, Yin RS, Liu N (2010) Mercury in the marine boundary layer and seawater of the South China Sea: concentrations, sea/air flux, and implication for land outflow. *J Geophys Res-Atmos* 115
- Fu Z, Wu F, Mo C, Liu B, Zhu J, Deng Q, Liao H, Zhang Y (2011) Bioaccumulation of antimony, arsenic, and mercury in the vicinities of a large antimony mine, China. *Microchem J* 97:12–19
- Greger M, Wang Y, Neuschütz C (2005) Absence of Hg transpiration by shoot after Hg uptake by roots of six terrestrial plant species. *Environ Pollut* 134:201–208
- Ivanciuc T, Ivanciuc O, Klein DJ (2006) Modeling the bioconcentration factors and bioaccumulation factors of polychlorinated biphenyls with poset quantitative structure-activity relationships (QSSAR). *Mol Divers* 10:133–145
- King JK, Kostka JE, Frischer ME, Saunders FM (2000) Sulfate-reducing bacteria methylate mercury at variable rates in pure culture and in marine sediments. *Appl Environ Microbiol* 66:2430–2437
- Krupp EM, Mestrot A, Wielgus J, Meharg AA, Feldmann J (2009) The molecular form of mercury in biota: identification of novel mercury peptide complexes in plants. *Chem Commun* 28:4257–4259
- Li P, Feng X, Qiu G, Wan Q (2011) Hair can be a good biomarker of occupational exposure to mercury vapor: simulated experiments and field data analysis. *Sci Total Environ* 409:4484–4488
- Liang L, Horvat M, Feng XB, Shang LH, Lil H, Pang P (2004) Re-evaluation of distillation and comparison with HNO<sub>3</sub> leaching/solvent extraction for isolation of methylmercury compounds from sediment/soil samples. *Appl Organomet Chem* 18:264–270
- Liang Y, Liu X, Yuan D, Gong Z, Zhang Z (2010) Mercury species in seawater and sediment of Xiamen western sea area adjacent to a coal-fired power plant. *Water Environ Res* 82:335–341
- Liang P, Shao D-D, Wu S-C, Shi J-B, Sun X-l, Wu F-Y, Lo SCL, Wang W-X, Wong MH (2011) The influence of mariculture on mercury distribution in sediments and fish around Hong Kong and adjacent mainland China waters. *Chemosphere* 82:1038–1043
- Liang P, Feng X, Zhang C, Zhang J, Cao Y, You Q, Leung AOW, Wong M-H, Wu S-C (2015) Human exposure to mercury in a compact fluorescent lamp manufacturing area: by food (rice and fish) consumption and occupational exposure. *Environ Pollut* 198:126–132
- Liu P, Qiu GL, Shang LH (2007) Phytoremediation of mercury contaminated soil: a review. *Chinese J Ecol* 26:933–937 (in Chinese)
- MacDonald DD, Ingersoll CG, Berger T (2000) Development and evaluation of consensus-based sediment quality guidelines for freshwater ecosystems. *Arch Environ Contam Toxicol* 39:20–31
- Meng B, Feng X, Qiu G, Liang P, Li P, Chen C, Shang L (2011) The process of methylmercury accumulation in rice (*Oryza sativa* L.). *Environ Sci Technol* 45:2711–2717
- Qiu G, Feng X, Wang S, Fu X, Shang L (2009) Mercury distribution and speciation in water and fish from abandoned Hg mines in Wanshan, Guizhou province, China. *Sci Total Environ* 407:5162–5168
- Rutter AP, Schauer JJ, Shafer MM, Creswell J, Olson MR, Clary A, Robinson M, Parman AM, Katzman TL (2011) Climate sensitivity of gaseous elemental mercury dry deposition to plants: impacts of temperature, light intensity, and plant species. *Environ Sci Technol* 45:569–575
- SBZP (2011) Statistics Bureau of Zhejiang Province, <http://www.zj.stats.gov.cn/tjsj/tjnj/DesktopModules/Reports/07.%E6%B5%99%E6%B1%9F%E7%BB%9F%E8%AE%A1%E5%B9%B4%E9%89%B42010/indexch.htm>, Hangzhou.
- Shao D, Wu S, Liang P, Kang Y, Fu W, Zhao K, Cao Z, Wong M (2012) A human health risk assessment of mercury species in soil and food around compact fluorescent lamp factories in Zhejiang Province, PR China. *J Hazard Mater* 221:28–34
- Ullrich SM, Tanton TW, Abdrashitova SA (2001) Mercury in the aquatic environment: a review of factors affecting methylation. *Critical Rev Environ Sci Technol* 31:241–293
- Ullrich SM, Ilyushchenko MA, Kamberov IM, Tanton TW (2007) Mercury contamination in the vicinity of a derelict chlor-alkali plant. Part I: sediment and water contamination of Lake Balkyldak and the River Irtysh. *Sci Total Environ* 381:1–16
- USEPA (2001): Method 1630: Methylmercury in water by distillation, aqueous ethylation, purge and trap, and CVAFS U.S. Environmental Protection Agency, Office of Water, Office of Science and Technology Engineering and Analysis Division (4303), 1200 Pennsylvania Avenue NW, Washington, D.C, pp. 1–41
- USEPA (2002): Method 1631: mercury in water by oxidation, purge and trap, and cold vapor atomic fluorescence spectrometry. Revision E. United States Environmental Protection Agency. EPA 821-R-95-027
- USEPA (2007) Method 7473: mercury in solids and solutions by thermal decomposition amalgamation, and atomic absorption spectrophotometry., United States Environmental Protection Agency
- Vannini A, Nicolardi V, Bargagli R, Loppi S (2014) Estimating atmospheric mercury concentrations with lichens. *Environ Sci Technol* 48:8754–8759
- Wang J, Feng X, Anderson CWN, Xing Y, Shang L (2012) Remediation of mercury contaminated sites—a review. *J Hazard Mater* 221–222:1–18
- Xu J, Zhang M, Fan LZ (2007) Alleviation of occupational mercury hazards after technical modification in an energy-saving lamps plant. *Occup Health Emergency Resc* 25:127–128 (in Chinese)
- Yan H, Feng X, Shang L, Qiu G, Dai Q, Wang S, Hou Y (2008) The variations of mercury in sediment profiles from a historically mercury-contaminated reservoir, Guizhou province, China. *Sci Total Environ* 407:497–506
- Zheng N, Liu J, Wang Q, Liang Z (2011) Mercury contamination due to zinc smelting and chlor-alkali production in NE China. *Appl Geochem* 26:188–193
- Zhong H, Wang WX (2011) Bioavailability of mercury in sediment: a review. *Environ Chemistry* 30:165–178 (in Chinese)