



# Human exposure to mercury in a compact fluorescent lamp manufacturing area: By food (rice and fish) consumption and occupational exposure



Peng Liang<sup>a, b</sup>, Xinbin Feng<sup>b, \*\*</sup>, Chan Zhang<sup>c</sup>, Jin Zhang<sup>a</sup>, Yucheng Cao<sup>a</sup>, Qiongzhi You<sup>a</sup>, Anna Oi Wah Leung<sup>d</sup>, Ming-Hung Wong<sup>a</sup>, Sheng-Chun Wu<sup>a, \*</sup>

<sup>a</sup> School of Environmental and Resource Sciences, Zhejiang Agricultural and Forest University, Lin'an, Zhejiang Province 311300, PR China

<sup>b</sup> State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, PR China

<sup>c</sup> College of Law and Political Science, Zhejiang Agricultural and Forestry University, Lin'an, Zhejiang Province 311300, PR China

<sup>d</sup> Department of Biology, Hong Kong Baptist University, Hong Kong, PR China

## ARTICLE INFO

### Article history:

Received 18 August 2014

Received in revised form

17 December 2014

Accepted 22 December 2014

Available online 13 January 2015

### Keywords:

Mercury

Compact fluorescent lamps

Methylmercury exposure

Rice

## ABSTRACT

To investigate human Hg exposure by food consumption and occupation exposure in a compact fluorescent lamp (CFL) manufacturing area, human hair and rice samples were collected from Gaohong town, Zhejiang Province, China. The mean values of total mercury (THg) and methylmercury (MeHg) concentrations in local cultivated rice samples were significantly higher than in commercial rice samples which indicated that CFL manufacturing activities resulted in Hg accumulation in local rice samples. For all of the study participants, significantly higher THg concentrations in human hair were observed in CFL workers compared with other residents. In comparison, MeHg concentrations in human hair of residents whose diet consisted of local cultivated rice were significantly higher than those who consumed commercial rice. These results demonstrated that CFL manufacturing activities resulted in THg accumulation in the hair of CFL workers. However, MeHg in hair were mainly affected by the sources of rice of the residents.

© 2014 Elsevier Ltd. All rights reserved.

## 1. Introduction

Human exposure to Hg compounds has been of great public concern around the world due to their high toxicity, and has been associated with several diseases such as neurologically related problems, myocardial infarction and autism (Li et al., 2008b).

The primary environmental source of human Hg exposure is through the digestion system, such as consumption of freshwater fish and seafood which have accumulated a considerable amount of Hg via biomagnification through the food chain/web (USEPA, 1997; WHO/IPCS, 1990). Recently, some studies showed that rice, rather than fish, may be one of the major Hg exposure pathways in inland China, where local residents consume few seafood products (Li et al., 2011b; Zhang et al., 2010). The probable daily intake (PDI)

of methylmercury (MeHg) from rice consumption indicated potential human health risk in Hg mining areas (Feng et al., 2008; Li et al., 2011b; Zhang et al., 2010). Neurobiological disruptions and increase of *c-fos* gene expression have been observed in experiments involving rats fed with rice contaminated with MeHg (Cheng et al., 2006; Ji et al., 2006), although the bioavailability of Hg in humans is not well known. Current international safety guidelines established by the Joint Food and Agriculture Organization (FAO) and World Health Organization (WHO) Expert Committee on Food Additives (JECFA) recommend a maximum Provisional Tolerable Weekly Intake level (PTWI) for Hg of 1.6 µg/kg (body weight) for women of childbearing age and 3.3 µg/kg for the general population (JECFA, 2003). In 1997, the United States Environmental Protection Agency (USEPA) set the limit of 0.1 µg kg<sup>-1</sup> d<sup>-1</sup> as a reference dose (RfD) for MeHg (USEPA, 1997).

Besides the ingestion pathway, occupational exposures such as via inhalation of Hg vapor or dermal contact with elemental Hg or its compounds also pose a potential threat to human health, especially to people engaged in Hg mining, chloralkali plant operation and compact fluorescent lamp (CFL) industries. It has been

\* Corresponding author.

\*\* Corresponding author.

E-mail addresses: [fengxinbin@vip.skleg.cn](mailto:fengxinbin@vip.skleg.cn) (X. Feng), [shengchunwu@126.com](mailto:shengchunwu@126.com) (S.-C. Wu).

well documented that occupational exposure may cause damage to the central nervous system (e.g., tremor and mental changes) and the kidneys (e.g., proteinuria) (WHO/IPCS, 1991).

CFL is a type of fluorescent lamp which relies on Hg as a source of ultraviolet radiation for the production of visible light. Good practice in the manufacturing of CFLs contained as low as 1.4–2.7 mg elemental Hg per lamp (Dunmire et al., 2003). There were 8.0 billion lamps produced in China in 2010 (Li et al., 2013). This implies that the annual elemental Hg consumption for CFL production in China reached up to 11.2–21.6 Mg.

Most of the previous studies reported potential human Hg exposure from broken CFL in the indoor environment (Li and Li, 2011; Sarigiannis et al., 2012). Measures to mitigate health risks to Hg exposure after the breakage of a CFL have been proposed, such as all windows and doors should be immediately opened and all occupants should vacate the room and wait 15–30 min after breakage before cleaning up. The room should also be ventilated for several hours (Sarigiannis et al., 2012). Our previous study revealed that CFL manufacturing activities resulted in Hg contamination to the local soil and sediment environment in a CFL production base in China due to the primitive technology used and the poorly-equipped facilities (Shao et al., 2012). Xu et al. (2007) reported that extremely high Hg concentrations in air were detected in a CFL manufacturing department and reached as high as  $16,000 \text{ ng m}^{-3}$ . However, data on human Hg exposure in CFL manufacturing areas are still insufficient and the sources of the Hg exposure have not been fully investigated. Some recent studies have confirmed that population in a Hg mining area who were expected to be mainly exposed to inorganic Hg (IHg), were also facing a serious threat of MeHg exposure via consumption of the local Hg-contaminated rice (Feng et al., 2008; Zhang et al., 2010). Hence, we hypothesize that the population in the CFL manufacturing area may be at the risk of co-exposure to both inorganic Hg (IHg) through inhalation and MeHg via food consumption. The objectives of this study were to: (1) investigate Hg contamination in rice samples from the CFL manufacturing area; and (2) to identify sources of possible human co-exposure through either inhalation of IHg or ingestion of MeHg via consumption of contaminated food using speciation analyses of human hair samples collected from the CFL production and control areas.

## 2. Materials and methods

### 2.1. 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 CFLs” since there are more than 180 CFL manufacturing factories in Gaohong producing 25% of the world production of CFLs in 2010 (SBZP, 2011).

Residents from four villages namely Shimen (SM), Nima (NM), Maling (ML) and Hongqiao (HQ) were chosen for this study (Fig. 1). Most of the CFL factories are located in NM and HQ villages, while a few is found in ML village. There are no factories located in SM villages, which was selected as the control site.

### 2.2. Sample collection

Hair samples were collected from 128 participants, in which 84 donors also provided samples of rice samples that their family consumed on a daily basis. In addition, 46 participants only provided rice samples, but refused to donate hair samples. A written consent form for the sample donation was obtained from each participant, together with a short questionnaire recording details concerning gender, age, occupation, source of the rice, weekly consumption amount of rice and fish. Among all of the hair samples, 63 were collected from CFL workers and 65 were from residents. Of the rice samples, 77 were locally cultivated and 53 were non-locally cultivated.

The sources of rice included the locally cultivated and commercial rice. Hair samples were cut with stainless steels scissors from the occipital region of the scalp, bundled together with srip, placed and sealed in polyethylene bags. It is acknowledged that a complete sample set of rice and hair could not be obtained from all the participants since some of the participants were reluctant to supply either their hair or rice samples.

Five species of fish were collected from the local market in 2011 which included grass carp (*Ctenopharyngodon idellus*) ( $n = 6$ ), northern snakehead (*Channa argus*) ( $n = 4$ ), oriental weatherfish (*Misgurnus anguillicaudatus*) ( $n = 20$ ), mud skipper (*Periophthalmus argentilineatus*) ( $n = 25$ ) and yellow-headed catfish (*Pelteobagrus*

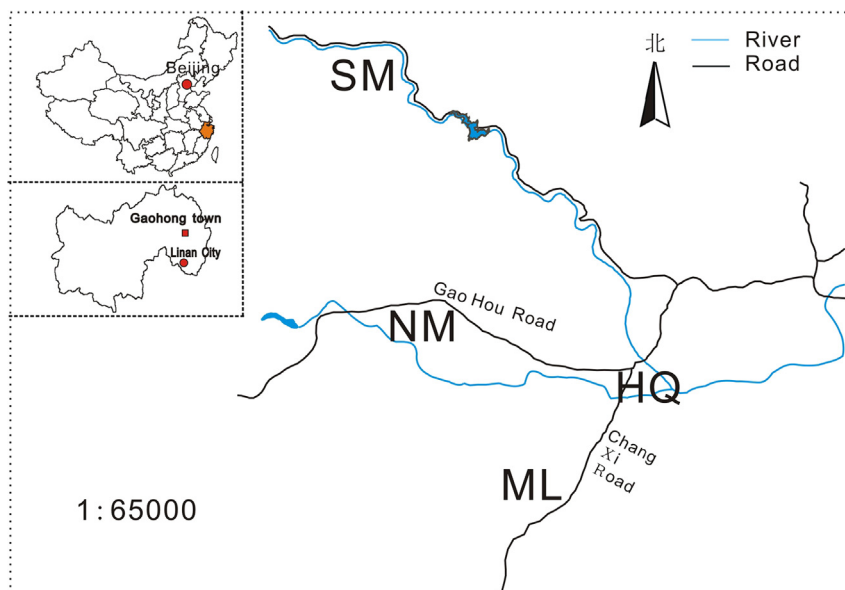


Fig. 1. Sampling site for this study. SM: Shimen NM: Nima; ML: Maling; HQ: Hongqiao.

*fulvidraco*) ( $n = 5$ ). The MeHg concentrations in these fish were reported in our previous study (Shao et al., 2012).

### 2.3. Sample analyses

THg concentrations in rice and human hair samples was detected by using cold vapor atomic fluorescence spectrometry (CVAFS) after a set of pretreatment including BrCl oxidation, SnCl<sub>2</sub> reduction, purge and gold trap according to USEPA Method 1631 (USEPA, 2002).

MeHg concentration in rice and human hair were detected by GC-CVAFS (Brooks Rand MERX) after extraction using KOH–methanol/solvent (Liang et al., 1994, 1996) in accordance with USEPA method 1630 (USEPA, 2001).

The IHg concentration in human hair was calculated by subtracting the MeHg concentration from the THg concentration.

### 2.4. Calculation of PDI

To estimate MeHg intake from rice and fish consumption, the PDI values of the participants were calculated according to the following Formula (1):

$$PDI = (C \times IR \times 10^{-3}) / bw \quad (1)$$

where PDI is given in micrograms per kilogram of body weight per day ( $\mu\text{g kg}^{-1} \text{d}^{-1}$ ); C is MeHg concentration in rice or fish ( $\text{ng g}^{-1}$ ); IR is daily intake rate ( $\text{g d}^{-1}$ ); bw represents the bodyweight. The average bodyweight of a 60 kg for adult population was adopted according to the second National Physique Monitoring Bulletin (GASC, 2005). Herewith, it is assumed that there was no MeHg loss during the rice cooking process (Li et al., 2010).

### 2.5. QA/QC

Two method blanks as well as four certified reference materials were accompanied with each sample batch (up to 40 samples). The measured THg concentration in the rice certified reference material (GBW10010, National Research Center for Certified Reference Materials, China) was  $5.1 \pm 0.4 \mu\text{g kg}^{-1}$  ( $n = 16$ ) which was comparable with the certified concentration of  $5.3 \pm 0.5 \mu\text{g kg}^{-1}$ ; the measured THg and MeHg concentrations in human hair certified reference material (NIES-13, National Institute for Environmental Studies, Japan) were  $4.24 \pm 0.21$  and  $3.72 \pm 0.11 \mu\text{g g}^{-1}$  ( $n = 12$ ), respectively, which were also comparable with the certified values of  $4.42 \pm 0.40$  and  $3.80 \pm 0.40 \mu\text{g g}^{-1}$ , respectively. The standard deviation of the THg and MeHg concentrations in the replicates of all samples ranged from  $-27\%$  to  $+32\%$ .

### 2.6. Data analyses

All of the statistical results were analyzed by SPSS 16.0 for Windows. Differences in Hg concentrations in rice and hair among all sampling sites were performed by multiple-way ANOVA (S–N–K test). Prior to multiple-way ANOVA, the data was log-transformed to obtain equal variance. The correlations among different parameters were conducted using Spearman's rho correlation coefficient (two tailed test). The significant level was set at 0.05.

## 3. Results and discussion

### 3.1. Hg contamination in rice samples

The mean THg concentration of all the rice samples was  $10.6 \pm 6.9 \mu\text{g kg}^{-1}$  with a range from 1.30 to  $41.2 \mu\text{g kg}^{-1}$ , while the average MeHg concentration of all the rice samples was  $5.29 \pm 3.62 \mu\text{g kg}^{-1}$ , ranging from 0.09 to  $21.5 \mu\text{g kg}^{-1}$ . MeHg constituted a large proportion of the THg in the rice grain, with an average of 40.1%. A significant correlation between THg and MeHg in rice samples ( $r = 0.695$ ,  $p < 0.01$ ) were observed.

Fig. 2 shows the THg and MeHg concentrations in rice samples collected from different villages. THg and MeHg concentrations in the locally cultivated rice samples (THg:  $12.9 \pm 7.14 \mu\text{g kg}^{-1}$   $n = 77$ ; MeHg:  $6.01 \pm 3.65 \mu\text{g kg}^{-1}$   $n = 52$ ) were significantly higher ( $p < 0.05$ ) than the non-local rice samples (THg:  $6.79 \pm 4.81 \mu\text{g kg}^{-1}$   $n = 72$ ; MeHg:  $3.39 \pm 2.97 \mu\text{g kg}^{-1}$   $n = 29$ ). It was noted that the THg concentrations in 11 rice samples exceeded the national tolerance limit of Hg (GB2762-2012) in human foods, which is  $20 \mu\text{g kg}^{-1}$ , and 10 samples were locally cultivated. It was evident that CFL manufacturing activities indeed pose a threat to the safety of the local rice production.

For individual villages, a higher concentration of THg and MeHg in the local rice sample was noted in NM and HQ villages where CFL factories are the most densely distributed. There are two rivers in Gaohong town, namely the You River and Qiu River, which received the most Hg exhausts from the CFL factories. Unfortunately, these

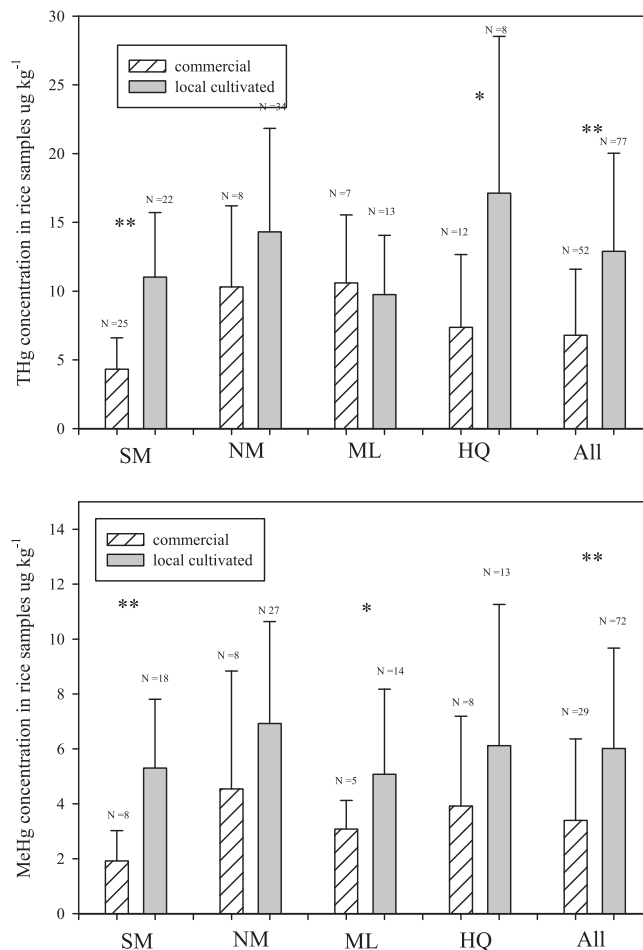


Fig. 2. THg and MeHg concentrations in rice samples from different sampling villages.

two rivers provide water for the irrigation of the local paddy rice fields. Hg species in the irrigating water could be transformed to MeHg as the flooding soil provides ideal anaerobic conditions for Hg methylation (Meng et al., 2011). Moreover, compared to other forms of Hg, MeHg would be more easily accumulated in rice plants due to its extended residence time in the anaerobic paddy soil (Meng et al., 2011). In comparison, the atmosphere is the principal source of inorganic Hg to the aboveground parts of the rice plant, and the accumulated IHg in aboveground parts of rice plants cannot be transported to seeds (Meng et al., 2012). Therefore, most of the paddy soil system in Gaohong town might have been affected by CFL manufacturing activities via irrigation with river water contaminated with Hg, especially in the village where CFL factories are densely distributed.

Table 1 shows the Hg concentrations in rice from literature. So far, most of studies that report Hg concentration in rice were conducted in China, especially in Hg mining areas. Both THg and MeHg concentrations in rice cultivated in the CFL manufacturing area were much lower than those found in the Hg mining areas (Feng et al., 2008; Horvat et al., 2003; Li et al., 2011b; Qiu et al., 2012, 2013). THg concentrations in locally cultivated rice from the high density area of the CFL factories located in Gaohong town (mean value:  $15.2 \mu\text{g kg}^{-1}$ , range:  $2.20\text{--}41.2 \mu\text{g kg}^{-1}$ ) were higher than the average value reported from seven provinces in southern China (mean value:  $10.1 \mu\text{g kg}^{-1}$ , range:  $0.86\text{--}47.2 \mu\text{g kg}^{-1}$ ) (Li et al., 2012). Moreover, MeHg concentrations in the locally cultivated rice samples collected from the high density area of the CFL factories (mean value:  $6.66 \mu\text{g kg}^{-1}$ , range:  $0.41\text{--}21.50 \mu\text{g kg}^{-1}$ ) were two times higher than those reported from seven provinces in southern China (mean value:  $2.47 \mu\text{g kg}^{-1}$ , range:  $0.13\text{--}18.2 \mu\text{g kg}^{-1}$ ) (Li et al., 2012) and several villages in Guizhou province, e.g., Changshun,  $2.5 \mu\text{g kg}^{-1}$  (Feng et al., 2008), Huaxi,  $2.9 \mu\text{g kg}^{-1}$  (Meng et al., 2010) and Leigong,  $2.1 \mu\text{g kg}^{-1}$  (Zhang et al., 2010), respectively.

### 3.2. Human MeHg exposure through food consumption

The PDI of MeHg through for people whose diet consisted of

**Table 1**  
THg and MeHg concentrations in rice samples from China.

| Location                           | Rice THg concentration ( $\mu\text{g kg}^{-1}$ ) | Rice MeHg concentration ( $\mu\text{g kg}^{-1}$ ) | Reference                     |
|------------------------------------|--|---|-------------------------------|
| Wanshan Hg mining area, Guizhou    | 36.2 (4.9–214.7)                                 | 8.5 (1.9–27.6)                                    | Feng et al., 2008             |
| Wanshan Hg mining area, Guizhou    | 11.1–569   | 8.03–144  | Horvat et al., 2003           |
| Gouxu Hg mining area, Guizhou      | $134 \pm 79.7$ (40.9–277)                        | $15.4 \pm 7.39$ (6.81–33.5)                       | Li et al., 2011               |
| Laowuchang Hg mining area, Guizhou | $138 \pm 64.1$ (29.5–258)                        | $14.4 \pm 9.75$ (6.37–34.1)                       | Li et al., 2011               |
| Yanwuping Hg mining area, Guizhou  | $22 \pm 9.5$ (10–45)                             | $11 \pm 7.6$ (3.2–39)                             | Qiu et al., 2013              |
| Xunyang Hg mining area, Shaanxi    | 50–200   | 8.2–80  | Qiu et al., 2012              |
| Seven provinces in southern China  | 10.1 (0.86–47.2)                                 | 2.47 (0.13–18.2)                                  | Li et al., 2012               |
| Changshun village, Guizhou         | $7.0 \pm 2.8$ (3.2–15.1)                         | $2.5 \pm 1.2$ (0.80–4.3)                          | Feng et al., 2008             |
| Leigong village, Guizhou           | 3.2  | 2.1   | Zhang et al., 2010            |
| CFL manufacturing area             | $12.9 \pm 7.14$ (1.30–41.2)                      | $6.01 \pm 3.65$ (0.09–21.5)                       | This study (local cultivated) |

locally cultivated rice is  $0.028 \pm 0.021 \mu\text{g kg}^{-1} \text{d}^{-1}$ , which was significantly higher ( $F = 6.30$ ,  $p < 0.05$ ) than that for people who consumed commercial rice ( $0.017 \pm 0.017 \mu\text{g kg}^{-1} \text{d}^{-1}$ ).

Rice consumption has been considered as the major source of MeHg exposure for inland residents in mainland China because they consume few fish meals and rice is their staple food (Zhang et al., 2010). Rice is the staple food for the residents in Gaohong town. Our questionnaire survey showed that  $2.04 \text{ kg}$  rice was consumed per week per person.

Although MeHg concentrations in fish were not determined in this study, our previous study reported MeHg concentrations in different kinds of fish samples collected from the local market in Gaohong town. The mean value of MeHg in local fish samples was  $40.3 \mu\text{g kg}^{-1}$  (Shao et al., 2012) (Table 2). According to our questionnaire survey, the daily intake rate of fish for people in Gaohong town was only  $12.4 \pm 17.7 \text{ g}$  per day. Therefore, the average MeHg PDI through fish consumption for the local residents was only  $0.008 \mu\text{g kg}^{-1} \text{d}^{-1}$ , which accounted for 28.8% and 47% of the MeHg intake through rice consumption for people who consume local and commercial rice, respectively. Fig. 3 shows the PDI of MeHg through rice and fish consumption for the residents of Gaohong town in this study. Most of the PDIs of MeHg through rice consumption were much higher than that through fish consumption. Thus, rice consumption is the major food source for MeHg exposure to local residents in this study.

### 3.3. Accumulation of Hg in human hair

In order to evaluate the contribution of occupational exposure and food exposure on Hg accumulation in human hair, all participants were divided into four groups according to their occupation and source of rice, i.e., workers who consumed locally cultivated rice (WFL), other residents who consumed locally cultivated rice (RFL); workers consumed commercial rice (WFI) and other residents who consumed commercial rice (RFI).

Table 3 summarizes the distribution of different Hg species in human hair from different groups. The THg concentration in human hair ranged from  $143$  to  $22800 \mu\text{g kg}^{-1}$  with a mean value of  $983 \mu\text{g kg}^{-1}$ . THg concentrations in human hair for WFL were significantly higher ( $p < 0.05$ ) than the other three groups. Human hair can be used as a good indicator of Hg exposure as either exogenous Hg contamination via consumption of Hg-contaminated food or Hg deposited endogenously could be accumulated in human hair and it is difficult to be washed out (Li et al., 2008a, 2011a). Thus, the results of this study demonstrated that both occupational exposure and food consumption contributed to Hg accumulation in human hair.

MeHg concentrations in human hair samples ranged from  $4.0$  to  $1190 \mu\text{g kg}^{-1}$  with a mean value of  $148 \mu\text{g kg}^{-1}$ . MeHg constituted a proportion of THg in human hair, with an average of 15.5% (0.11–94.4%). A significantly positive correlation ( $r = 0.617$ ,  $p < 0.01$ ) was observed between THg and MeHg concentrations in hair of the other residents (Fig. 4a) and for the CFL workers ( $r = 0.430$ ,  $p < 0.01$ ) (Fig. 4b). An abnormally low ratio of MeHg to

**Table 2**  
THg and MeHg concentration in local fish samples.<sup>a</sup>

| Fish species         | THg ( $\mu\text{g kg}^{-1}$ ) | MeHg ( $\mu\text{g kg}^{-1}$ ) | n  |
|----------------------|-------------------------------|--------------------------------|----|
| Mud skipper          | $170 \pm 45$                  | $143 \pm 37$                   | 25 |
| Yellowhead catfish   | $41.2 \pm 16.3$               | $32.6 \pm 11.8$                | 5  |
| Northern snakehead   | $31.2 \pm 6.4$                | $25.3 \pm 5.83$                | 4  |
| Grass carp           | $23.6 \pm 10.9$               | $18.6 \pm 7.2$                 | 6  |
| Oriental weatherfish | $14.1 \pm 4.3$                | $10.5 \pm 3.5$                 | 20 |

<sup>a</sup> Published in Shao et al. (2012).

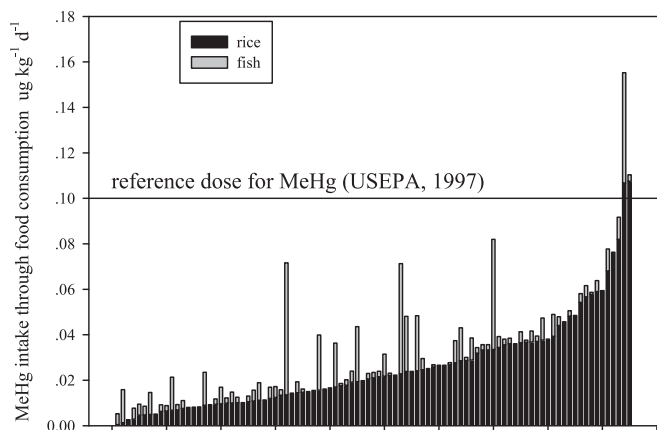


Fig. 3. PDI of MeHg through rice and fish consumption for resident in Gaohong town.

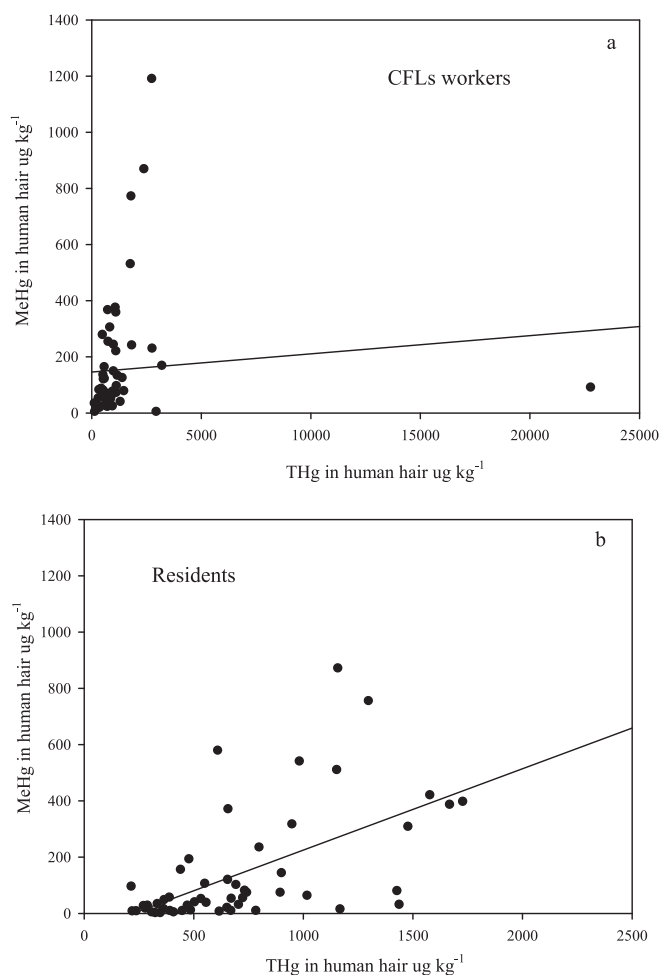


Fig. 4. Correlation between THg and MeHg in human hair for CFL workers (a) and other residents (b).

THg was noted in several hair samples collected from the workers of the CFL factories. A possible explanation might be that Hg vapor released from the CFL manufacturing factories could adhere to the hair surface and may not be easily washed away during pretreatment. In addition, it has been reported that IHg can also be accumulated in hair through the consumption of contaminated rice (Meng et al., 2012), although the mechanism has not been well

addressed.

To evaluate the influence of occupation and rice exposure on MeHg accumulation in human hair, two way ANOVA (occupation [fixed] \* rice source [fixed]) was employed for testing the difference of MeHg concentration in human hair. There was no significant interaction between the two factors ( $F = 0.569$ ,  $p > 0.05$ ). Occupational exposure (CFLs workers and other residents) did not have a significant effect on MeHg concentration in human hair ( $F = 0.172$ ,  $p > 0.05$ ), but rice source (local cultivated and commercial) did have a significant effect on MeHg concentration in human hair ( $F = 21.4$ ,  $p < 0.001$ ). This indicated that MeHg concentration in human hair of the residents who consumed locally cultivated rice (WFL and RFL) were significantly higher than residents who consumed commercial rice (WFI and RFI).

MeHg concentration in human hair can be used as a good indicator of human MeHg exposure since, generally, there is almost no exogenous contamination of MeHg in human hair (Li et al., 2008a). Thus, occupational exposure of THg from CFL manufacturing activities did not result in MeHg accumulation in human hair. In contrast, MeHg intake through local rice consumption promoted MeHg accumulation in human hair.

IHg concentration in human hair ranged from 30 to 22710  $\mu\text{g kg}^{-1}$  with a mean value of 913  $\mu\text{g kg}^{-1}$ . Two-way ANOVA was employed for testing the difference in IHg concentrations in human hair. No significant interacting effect between the two factors existed ( $F = 0.669$ ,  $p > 0.05$ ). Both occupational exposure ( $F = 1.643$ ,  $p > 0.05$ ) and rice source ( $F = 1.066$ ,  $p > 0.05$ ) did not have a significant effect on IHg concentration in human hair.

Li et al. (2011a) found that IHg in human hair can be a good biomarker of occupational exposure to Hg vapor. The higher concentration of IHg in human hair for WFL and WFI than that for RFL and RFI indicated that CFL manufacturing activities may lead to IHg accumulation in workers, although the differences were not significant. Moreover, IHg in hair for WFL and RFL were higher than WFI and RFI, respectively since IHg can also be accumulated in hair through rice consumption (Meng et al., 2012).

Table 4 shows the comparison of hair Hg concentrations between this study and previous studies. Hg concentrations from our study were lower than those of residents from the Hg mining area in Guizhou Province, China, where rice consumption is also the major MeHg exposure source for the local residents. Moreover, hair Hg levels in CFL manufacturing area were similar to the fish eating population in the Pearl River Delta region in Southern China (Shao et al., 2013). Finally, the hair Hg levels in Gaohong town were much lower than those from the Amazon (Vieira et al., 2013), Faroe Islands (Choi et al., 2009), and Japan (Endo and Haraguchi, 2010), where fish served as the major Hg exposure for the local residents.

Fig. 5 shows the correlation between MeHg in human hair and PDI of MeHg. Positive correlations were noted for local residents and negative correlations were found for CFL workers, however, both of these correlations were not significant, which indicated that other MeHg exposure pathways may exist.

The results of PDI of MeHg for this study were higher than the previous report on MeHg exposure from fish consumption in 12 provinces in China, which was 0.006  $\mu\text{g kg}^{-1} \text{d}^{-1}$  (Shang et al., 2010) and the residents from seven southern province China by rice consumption, 0.004–0.008  $\mu\text{g kg}^{-1} \text{d}^{-1}$  (Li et al., 2012). Moreover, the average PDI of MeHg obtained in our study was comparable with the average value for US adult women (0.02  $\mu\text{g kg}^{-1} \text{d}^{-1}$ , Mahaffey et al., 2004), Korean population (0.03  $\mu\text{g kg}^{-1} \text{d}^{-1}$ , Lee et al., 2006) and Hong Kong residents (0.05  $\mu\text{g kg}^{-1} \text{d}^{-1}$ , Liang et al., 2011), but was lower than that reported in Japanese populations with high fish consumption (0.14  $\mu\text{g kg}^{-1} \text{d}^{-1}$ , Zhang et al., 2009). Overall, 2 PDI of MeHg values would exceed the MeHg RfD established by the USEPA (Fig. 3). Thus,

**Table 3**THg and MeHg concentrations in human hair ( $\mu\text{g kg}^{-1}$ ).

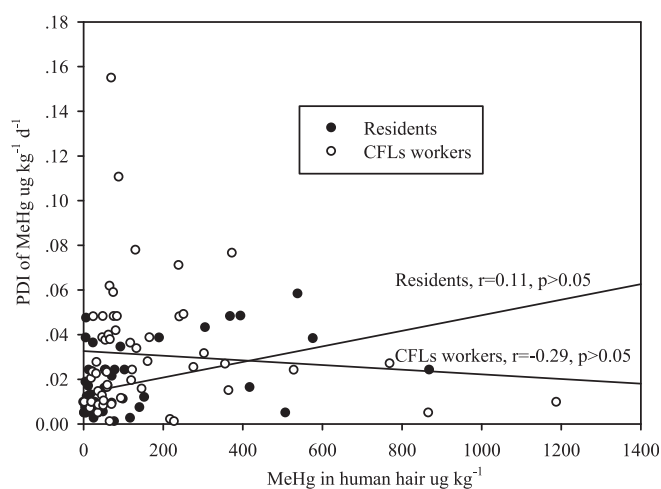
|      | WFL                         | RFL                       | WFI                           | RFI                        | All                        |
|------|-----------------------------|---------------------------|-------------------------------|----------------------------|----------------------------|
| THg  | 1631 $\pm$ 3551 (143–22800) | 874 $\pm$ 462 (217–1730)  | 744 $\pm$ 561 (165–2970)      | 580 $\pm$ 247 (240–1430)   | 983 $\pm$ 2060 (143–22800) |
| MeHg | 209 $\pm$ 257 (21.1–1189)   | 252 $\pm$ 258 (0.44–869)  | 65.0 $\pm$ 59.0 (3.43–252.81) | 52.4 $\pm$ 57.7 (2.83–233) | 133 $\pm$ 195 (0.44–1189)  |
| IHg  | 1484 $\pm$ 3636 (110–22710) | 621 $\pm$ 411 (34.3–1410) | 724 $\pm$ 568 (162–2967)      | 533 $\pm$ 256 (233–1351)   | 913 $\pm$ 2185 (110–22710) |

WFL: workers who consume local cultivated rice; RFL: other residents who consume local cultivated rice; WFI: workers who consume commercial rice; RFI: other residents who consume commercial rice.

**Table 4**

Comparison of hair Hg concentrations from different studies.

| Location                      | Population          | Hair THg ( $\mu\text{g g}^{-1}$ ) | Hair MeHg ( $\mu\text{g g}^{-1}$ ) | Exposure sources | References               |
|-------------------------------|---------------------|-----------------------------------|------------------------------------|------------------|--------------------------|
| Gouxi Hg mining area, China   | Workers             | 25.5 $\pm$ 30.7 (4.8–108)         | 7.55 $\pm$ 2.63 (3.72–12.1)        | Rice             | Li et al., 2011          |
|                               | Local residents     | 5.05 $\pm$ 2.81 (2.35–10.6)       | 3.53 $\pm$ 1.19 (1.87–5.65)        |                  |                          |
| Wuchuan Hg mining area, China | Smelting workers    | 33.9 $\pm$ 24.1 (7.63–93.1)       | 0.95 $\pm$ 0.37 (0.52–1.73)        | Rice             | Li et al., 2008          |
|                               | Other inhabitants   | 2.71 $\pm$ 0.99 (1.13–4.27)       | 1.84 $\pm$ 0.77 (0.92–3.12)        |                  |                          |
| Wanshan Hg mining area, China | Inhabitants         | 7.3 $\pm$ 11.6 (2.1–58.5)         | 2.8 $\pm$ 1.4 (0.8–5.6)            | Rice             | Feng et al., 2008        |
|                               | Local residents     | 1.08 $\pm$ 0.94 (0.14–7.15)       | 0.58 $\pm$ 0.59 (0.03–4.64)        | Fish             | Shao et al., 2013        |
| Amazon                        | Riparian population | 8.24 (0.72–20.08)                 | 2.14 (0.07–5.70)                   | Fish             | Vieira et al., 2013      |
| Faroe Islands                 | Whaling men         | 7.31 (0.92–46.0)                  |                                    | Pilot whale      | Choi et al., 2009        |
| Japan                         | Local residents     | 17.7 $\pm$ 11.5                   |                                    | Fish             | Endo and Haraguchi, 2010 |
| This study                    | CFL workers         | 1.36 $\pm$ 2.90 (0.143–22.8)      | 0.154 $\pm$ 0.215 (0.003–1.19)     | Rice             | This study               |
|                               | Other residents     | 0.716 $\pm$ 0.399 (0.217–1.73)    | 0.141 $\pm$ 0.202 (0.004–0.869)    |                  |                          |

**Fig. 5.** Correlation between MeHg in human hair and PDI of MeHg.

CFL manufacturing activities have already led to a PDI of MeHg that has exceeded the guidelines for public health for the local residents.

#### 4. Conclusions

THg and MeHg concentrations in the locally cultivated rice samples from CFL manufacturing area were significantly higher than those in commercial rice samples. This means that CFL manufacturing activities indeed resulted in Hg contamination of the local rice samples. Residents who consumed the locally cultivated rice samples have significantly higher MeHg concentration in their hair compared to people who consumed the commercial rice samples, which indicated potential health risk of MeHg exposure for the local residents through rice consumption. MeHg intake from fish consumption were generally lower than that from rice consumption. Generally, the PDI of MeHg through food consumption for the local residents was below the guidelines for public health concerns.

#### Acknowledgments

Financial support from the Natural Science Foundation of Zhejiang Province (No. LQ12D03001), the National Natural Science Foundation of China (No. 21307114) and Postdoctoral Science Foundation of China (No. 2013M540719) are gratefully acknowledged.

#### References

- Cheng, J., Wang, W., Jia, J., Zheng, M., Shi, W., Lin, X., 2006. Expression of c-fos in rat brain as a prelude marker of central nervous system injury in response to methylmercury-stimulation. *Biomed. Environ. Sci.* 19, 67–72.
- Choi, A., Weihe, P., Budtz-Jørgensen, E., Jørgensen, P., Salonen, J., Tuomainen, T., Murata, K., Nielsen, H., Petersen, M., Askham, J., 2009. Methylmercury exposure and adverse cardiovascular effects in Faroese whaling men. *Environ. Health Perspect.* 117, 367.
- Dunmire, C., et al., 2003. Mercury in Fluorescent Lamps: Environmental Consequences and Policy Implications for NRDC. Ecos Consulting, prepared for the Natural Resources Defense Council (NRDC).
- Endo, T., Haraguchi, K., 2010. High mercury levels in hair samples from residents of Taiji, a Japanese whaling town. *Mar. Pollut. Bull.* 60, 743–747.
- Feng, X., Li, P., Qiu, G., Wang, S., Li, G., Shang, L., Meng, B., Jiang, H., Bai, W., Li, Z., 2008. Human exposure to methylmercury through rice intake in mercury mining areas, Guizhou Province, China. *Environ. Sci. Technol.* 42, 326–332.
- GASC, 2005. The Second National Physique Monitoring Bulletin.
- Horvat, M., Nolde, N., Fajon, V., Jereb, V., Logar, M., Lojen, S., Jacimovic, R., Falnoga, I., Liya, Q., Faganelli, J., 2003. Total mercury, methylmercury and selenium in mercury polluted areas in the province Guizhou, China. *Sci. Total Environ.* 304, 231–256.
- JECFA, 2003. In: Summary and Conclusions of the Sixty-first Meeting of the Joint FAO/WHO Expert Committee on Food Additives, Rome, pp. 1–22.
- Ji, X., Wang, W., Cheng, J., Yuan, T., Zhao, X., Zhuang, H., Qu, L., 2006. Free radicals and antioxidant status in rat liver after dietary exposure of environmental mercury. *Environ. Toxicol. Pharmacol.* 22, 309–314.
- Li, L., Wang, Y., Zhang, X., Yan, F., Jian, X., Zhang, H., Li, S., Huang, Y., 2013. Analysis on reduction trend of mercury releases from chinese light source production industry for execution the global legally binding instrument on mercury. *Zhaoming Gongcheng Xuebao* 24, 23–30 (in Chinese).
- Lee, H.-S., Cho, Y.-H., Park, S.-O., Kye, S.-H., Kim, B.-H., Hahm, T.-S., Kim, M., Ok Lee, J., Kim, C.-i., 2006. Dietary exposure of the Korean population to arsenic, cadmium, lead and mercury. *J. Food Compos. Anal.* 19, S31–S37.
- Li, P., Feng, X., Qiu, G., 2010. Methylmercury exposure and health effects from rice and fish consumption: a review. *Int. J. Environ. Res. Public Health* 7, 2666–2691.
- Li, P., Feng, X., Qiu, G., Li, Z., Fu, X., Sakamoto, M., Liu, X., Wang, D., 2008a. Mercury exposures and symptoms in smelting workers of artisanal mercury mines in Wuchuan, Guizhou, China. *Environ. Res.* 107, 108–114.
- Li, P., Feng, X., Qiu, G., Wan, Q., 2011a. Hair can be a good biomarker of occupational exposure to mercury vapor: simulated experiments and field data analysis. *Sci.*

- Total Environ. 409, 4484–4488.
- Li, P., Feng, X., Shang, L., Qiu, G., Meng, B., Zhang, H., Guo, Y., Liang, P., 2011b. Human co-exposure to mercury vapor and methylmercury in artisanal mercury mining areas, Guizhou, China. *Ecotoxicol. Environ. Saf.* 74, 473–479.
- Li, P., Feng, X., Yuan, X., Chan, H.M., Qiu, G., Sun, G.-X., Zhu, Y.-G., 2012. Rice consumption contributes to low level methylmercury exposure in southern China. *Environ. Int.* 49, 18–23.
- Li, Y.D., Li, J., 2011. Environmental release of mercury from broken compact fluorescent lamps. *Environ. Eng. Sci.* 28, 687–691.
- Li, Y.F., Chen, C.Y., Li, B., Li, W., Qu, L., Dong, Z.Q., Nomura, M., Gao, Y.X., Zhao, J.X., Hu, W., Zhao, Y.L., Chai, Z.F., 2008b. Mercury in human hair and blood samples from people living in Wanshan mercury mine area, Guizhou, China: an XAS study. *J. Inorg. Biochem.* 102, 500–506.
- Liang, L., Horvat, M., Bloom, N., 1994. An improved speciation method for mercury by GC/CVAFS after aqueous phase ethylation and room temperature pre-collection. *Talanta* 41, 371–379.
- Liang, L., Horvat, M., Cernichiari, E., Gelein, B., Balogh, S., 1996. Simple solvent extraction technique for elimination of matrix interferences in the determination of methylmercury in environmental and biological samples by ethylation-gas chromatography-cold vapor atomic fluorescence spectrometry. *Talanta* 43, 1883–1888.
- Liang, P., Shao, D.-D., Wu, S.-C., Shi, J.-B., Sun, X.-L., Wu, F.-Y., Lo, S., Wang, W.-X., Wong, M.H., 2011. The influence of mariculture on mercury distribution in sediments and fish around Hong Kong and adjacent mainland China waters. *Chemosphere* 82, 1038–1043.
- Mahaffey, K.R., Clickner, R.P., Bodurow, C.C., 2004. Blood organic mercury and dietary mercury intake: national Health and Nutrition Examination Survey, 1999 and 2000. *Environ. Health Perspect.* 112, 562.
- Meng, B., Feng, X., Qiu, G., Cai, Y., Wang, D., Li, P., Shang, L., Sommar, J., 2010. Distribution patterns of inorganic mercury and methylmercury in tissues of rice (*Oryza sativa* L.) plants and possible bioaccumulation pathways. *J. Agric. Food Chem.* 58, 4951–4958.
- 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.
- Meng, B., Feng, X., Qiu, G., Wang, D., Liang, P., Li, P., Shang, L., 2012. Inorganic mercury accumulation in rice (*Oryza sativa* L.). *Environ. Toxicol. Chem.* 31, 2093–2098.
- Qiu, G., Feng, X., Meng, B., Sommar, J., Gu, C., 2012. Environmental geochemistry of an active Hg mine in Xunyang, Shaanxi Province, China. *Appl. Geochem.* 27, 2280–2288.
- Qiu, G., Feng, X., Meng, B., Zhang, C., Gu, C., Du, B., Lin, Y., 2013. Environmental geochemistry of an abandoned mercury mine in Yanwuping, Guizhou Province, China. *Environ. Res.* 125, 124–130.
- Sarigiannis, D.A., Karakitsios, S.P., Antonakopoulou, M.P., Gotti, A., 2012. Exposure analysis of accidental release of mercury from compact fluorescent lamps (CFLs). *Sci. Total Environ.* 435, 306–315.
- SBZP, 2011. Statistics Bureau of Zhejiang Province.
- Shang, X., Li, X., Zhang, L., Zhao, Y., Wu, Y., 2010. Estimation of methylmercury intake from the 2007 Chinese Total Diet Study. *Food Addit. Contam.* 3, 236–245.
- Shao, D., Kang, Y., Cheng, Z., Wang, H., Huang, M., Wu, S., Chen, K., Wong, M.H., 2013. Hair mercury levels and food consumption in residents from the Pearl River Delta: South China. *Food Chem.* 136, 682–688.
- 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.
- USEPA, 1997. Mercury Study Report to Congress. In: Volume V: Health Effects of Mercury and Mercury Compounds. Office of Air Quality Planning and Standards and Office of Research and Development, United States Environmental Protection Agency.
- 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.
- Vieira, S.M., de Almeida, R., Holanda, I.B.B., Mussu, M.H., Galvão, R.C.F., Crispim, P.T.B., Dórea, J.G., Bastos, W.R., 2013. Total and methyl-mercury in hair and milk of mothers living in the city of Porto Velho and in villages along the Rio Madeira, Amazon, Brazil. *Int. J. Hyg. Environ. Health* 216, 682–689.
- WHO/IPCS, 1990. Methylmercury. *Environmental Health Criteria*, no.101. World Health Organization, Geneva.
- WHO/IPCS, 1991. Inorganic mercury. *Environmental Health Criteria* No 118. In: International Programme on Chemical Safety Geneva, Switzerland. World Health Organisation.
- Xu, J., Zhang, M., Fan, L.Z., 2007. Alleviation of occupational mercury hazards after technical modification in an energy-saving lamps plant. *Occup. Health Emerg. Res.* 25, 127–128 (in Chinese).
- Zhang, H., Feng, X., Larssen, T., Qiu, G., Vogt, R.D., 2010. In inland China, rice, rather than fish, is the major pathway for methylmercury exposure. *Environ. Health Perspect.* 118, 1183.
- Zhang, Y., Nakai, S., Masunaga, S., 2009. An exposure assessment of methyl mercury via fish consumption for the Japanese population. *Risk Anal.* 29, 1281–1291.