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Mercury levels and estimated total daily intakes for children and adults from an electronic waste recycling area in Taizhou, China: Key role of rice and fish consumption

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ABSTRACT

In order to assess the potential health risks of Hg pollution, total mercury (T-Hg) and methyl mercury (MeHg) concentrations were determined in air, dust, surface soil, crops, poultry, fish and human hair samples from an electronic waste (e-waste) recycling area in Taizhou, China. High concentrations of T-Hg and MeHg were found in these multiple matrices, and the mean concentration was 30.7 ng/m³ of T-Hg for atmosphere samples, 3.1 µg/g of T-Hg for soil, 37.6 µg/g of T-Hg for dust, 20.3 ng/g of MeHg for rice and 178.1 ng/g of MeHg for fish, suggesting that the e-waste recycling facility was a significant source of Hg. The inorganic Hg (I-Hg) levels (0.84 µg/g) in hair samples of e-waste workers were much higher than that in the reference samples. Pearson's correlation coefficients showed that strong positive correlations ($p < 0.01$) between hair I-Hg and time staying in industrial area ($r = 0.81$) and between MeHg and fish consumption frequency ($r = 0.91$), imply that workers were mainly exposed to Hg vapor through long-time inhalation of contaminated air and dust, while other population mainly exposed to MeHg through high-frequency fish consumption. The estimated daily intakes of Hg showed that dietary intake was the major Hg exposure source, and Hg intakes from rice and fish were significantly higher than from any other foods. The estimated total daily intakes (TDIs) of MeHg for both children (696.8 ng/(kg·day)) and adults (381.3 ng/(kg·day)) greatly exceeded the dietary reference dose (RfD) of 230 ng/(kg·day), implying greater health risk for humans from Hg exposures around e-waste recycling facilities.

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Introduction

China is rich in mercury (Hg) and exploration of Hg in China has led to serious environmental pollution and local ecosystem deterioration (Horvat et al., 2003; Sakamoto et al., 2007; Feng et al., 2008; Cheng and Hu, 2012). Numerous studies on Hg pollution, including our former studies, have focused on major industries, such as Hg mining, coal combustion,

chlorin-alkali production and gold mining (Cheng et al., 2009; Zhang et al., 2012). However, e-waste recycling industry has received little scientific attention. According to Electronic Industries Alliance, Hg can be found in at least 26 categories of electronic devices, including electrical lighting and switching devices, thermostats, devices with fluorescence lamps in LCDs (e.g., laptop computers, mobile phones and flat-panel TVs), and semiconductors (Jang and Townsend, 2003; Wong et al.,

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2006). A report prepared by Silicon Valley Toxics Coalition (SVTC) suggested that 22% of the world's annual consumption of Hg was used in electronics. It was estimated that, between 1997 and 2004, approximately 315 million computers were disposed of in the US alone, from which 180,000 kg of Hg could have been released to the environment (Wong et al., 2006). Therefore, e-waste recycling industry is one of the major sources of environmental Hg emissions.

In fact, Hg pollution in e-waste recycling areas in China has become increasingly severe due to its huge increase in the amount of e-waste generated annually and poor disposal methods. Approximately, 20 to 50 million metric tons of e-wastes are produced worldwide every year, 70% of which are exported to China for recycling (Wang, 2007; Ma et al., 2008). Taizhou, situated in Zhejiang province of eastern China, is a popular e-waste destination, and has nearly 30 years of e-waste unregulated disposal history. E-waste recycling in Taizhou employs around 40,000 people. Approximately 2.2 million metric tons of e-wastes are dismantled in Taizhou annually (Chan et al., 2007). These primitive recycling centers use simple operation methods such as sorting, incineration, acidic and alkaline baths, manual assembly, and strong acid leaching, which are usually carried out with no or very little personal protection equipment or pollution controlling measures. The incineration of e-wastes generates a large amount of particulates laden with heavy metals and other toxic materials, resulting in the contamination of food, soil, and surface water, which leads to increased human exposure (Leung et al., 2007; Li et al., 2007). In recent years, emission of toxic heavy metals and organic pollutants from e-waste recycling industry of China has received considerable attention (Huo et al., 2007; Wong et al., 2007; Ma et al., 2009, 2011); and Zhang et al. (2014) reported the concentrations of 16 PCBs and 7 heavy metals (Pb, Cd, Cr, Zn, As, Hg, and Cu) in soils near an abandoned e-waste recycling plant in Taizhou. Their results showed that both PCBs and heavy metal residue pose high risk to the ecosystem. However, there is still a paucity of specific information on Hg and MeHg pollution and its potential noxious effects related to the exposure to contaminants generated in the e-waste recycling industry. In this study, Hg and MeHg levels in workshop-floor dust, surface soil, atmosphere, crops, poultry, fish and hair samples were analyzed, and human exposures to Hg and MeHg via soil and dust ingestion, dermal exposure absorption, daily air inhalation, drinking water intake and daily dietary intake from e-waste sites were estimated.

1. Materials and methods

1.1. Sampling sites

Samples were collected from three types of sampling sites (Fig. 1), Fengjiang e-waste recycling industrial area (Site 1–5), Baifengao village (Sites 6–10) and reference site (Site 11). Fengjiang is a small town of Taizhou City, while it is one of the most important and intensive e-waste recycling bases of Taizhou. Fengjiang e-waste recycling industrial area, about 3 km south of the center of Fengjiang town, covers an area of 1.1 km² and comprises more than 40 corporations engaged in e-waste recycling industries.



Fig. 1 – Sampling sites (1–10) in Taizhou City, Zhejiang Province, China.

Baifengao village, about 3 km north of the center of Fengjiang town, had about 100 small-scale e-waste dismantling factories before 2007. Wenling, a small city located 25 km south of Fengjiang town, was selected as a reference location.

1.2. Sampling and sample preparation

1.2.1. Air, dust, soil, and crop samples

CD-1 ambient vapor analyzer (Beijing Detector Instruments Ltd.) was set at Sites 1 to 11 for collecting air samples. Every 10 min, 5 L of air sample was collected in a cartridge packed with gold-coated particles and subsequently analyzed by cold atomic fluorescence spectrometry. Sampling inlets were placed at 1.1 m above the ground level, where they were close to children stature. Workshop-floor dust (fine particles on the cement floor) and surface soil were collected from five large-scale e-waste recycling plants (Fig. 1, Sites 1–5) of the industrial area. Soil samples were taken from 2 paddy fields (Sites 6, 7) and three vegetable fields (Sites 8–10) of Baifengao village; rice (Sites 6, 7), corn (Site 8), soybean (Site 9) and cole leaf (Site 10) samples were collected to show the Hg exposure. Soil samples were collected from five quadrates from the top 5–10 cm of the surface layer at each site. All the samples (but cole) were air-dried, ground and sieved to pass a 100 mesh, stored in sealed glass containers, and frozen in refrigerator until analysis. Cole samples were cut and homogenized, then stored at 4°C until analysis.

1.2.2. Foodstuff samples

Hens and pigs are the major poultry for the local residents. Hens and eggs were purchased from the peasants of Baifengao village. These hens were 12–18 months old, herded under natural conditions, and lived on rice and sometimes food from the fields. Hens were then sacrificed by cervical dislocation, and chicken, brain and liver tissues were collected. Pork (the major meat consumed by the local residents), brain, liver and kidney samples of male pigs with ages of 10 to 12 months were

collected from the local villagers, which were fed with local feedstuff. Four types of fish representative of what is commonly consumed in Taizhou, were purchased from the local fishers of Baifengao village. The edible parts of the samples were dissected on site to collect a small muscle sample (10–20 g) from the dorsolateral part of the body, and then the samples were cut and homogenized and stored at -40°C until analysis. Drinking water samples were collected from the local inhabitants. The milk, bean oil and salt were purchased from the local shops of Baifengao village.

1.2.3. Hair samples

Hair samples were collected from 67 individuals (healthy volunteers) from different sampling locations. For each volunteer, the mean stay time every day in the industrial area and fish consumption frequency every month were inquired and recorded. According to the occupations (consecutive length of service for more than 12 months until sampling), the samples were divided into four groups: workers of industrial area, farmers of Baifengao village, civilians of Fengjiang town and civil servants of reference. Hair samples were cut from the base of the scalp, located behind the ear, and then washed with detergent, rinsed two times with acetone to dry. The dried hair was cut into small pieces shorter than 2 mm with scissors and kept at 4°C until analysis.

1.3. Hg analysis

T-Hg and MeHg concentrations in the samples were analyzed using cold vapor atomic adsorption spectrometry and gas chromatography with electron capture detector, respectively, according to the method of the Japanese National Institute for Minamata Disease (NIMD) (NIMD, 2001; Cheng et al., 2006, 2008). In addition, blank samples were analyzed as the same way as tissue samples. The precision and accuracy of the T-Hg and MeHg measurements were checked using certified reference materials from the Community Bureau of Reference BCR.

1.4. Exposure assessment

The children and adult exposure routes in this area were included: soil and dust ingestion (SDI), dermal exposure absorption (DEI), daily air inhalation (DAI), drinking water intake (DWI) and daily dietary intake (DDI). In this study, the disposition model for estimated daily intakes via soil/dust ingestion and dermal exposure was used (Eqs. (1) and (2)). This model was original from Nouwen et al. (2001) and simplified by Minh et al. (2003). The different kinds of daily intake of Hg were assessed according to the following Eqs. (1)–(5):

SDI (ng/(kg-day)):

$$\text{SDI} = \text{SDI}_{\text{soil}} + \text{SDI}_{\text{dust}} = (\text{AID} \times \text{IF}_{\text{soil}} \times C_{\text{soil}}) / \text{Wt} + (\text{AID} \times \text{IF}_{\text{dust}} \times C_{\text{dust}}) / \text{Wt} \quad (1)$$

wherein: SDI_{soil} and SDI_{dust} : ingestion of Hg for soil and dust, respectively (ng/(kg-day)); AID: ingestion of soil/dust particles amounts (6.3×10^{-2} g/day for children and 2.6×10^{-2} for adults); IF_{soil} and IF_{dust} : ingestion factor for soil (0.302 for children and 0.237 for adults) and dust (0.49 for children and 0.55 for adults) respectively; C_{soil} and C_{dust} : T-Hg and MeHg

concentration in soil and dust (ng/g dry wt); and Wt: body weight (15 kg for children and 60 for adults). In this study the mean levels of soil or dust concentrations in the industrial area were used to estimate SDI.

DEI (ng/(kg-day)):

$$\text{DEI} = \text{DEI}_{\text{soil}} + \text{DEI}_{\text{dust}} = (\text{DAE}_{\text{out}} \times \text{EF}_{\text{out}} \times C_{\text{soil}}) / \text{Wt} + (\text{DAE}_{\text{in}} \times \text{EF}_{\text{in}} \times C_{\text{dust}}) / \text{Wt} \quad (2)$$

wherein: DEI_{soil} and DEI_{dust} : dermal absorption for soil and dust, respectively (ng/(kg-day)); DAE_{out} : skin coverage with dust outside ($5.1 \text{ g}/(\text{m}^2\text{-day})$ for children and 37.5 for adults); DAE_{in} : skin coverage with dust inside ($0.56 \text{ g}/(\text{m}^2\text{-day})$ for both children and adults); EF_{out} and EF_{in} : exposure factors for appropriate absorptions of outside (0.0017 m^2 for children and 0.0011 for adults) and inside (0.0005 m^2 for children and 0.0131 for adults) skin, respectively.

DAI (ng/(kg-day)):

$$\text{DAI} = V_{\text{air}} \times C_{\text{air}} / \text{Wt} \quad (3)$$

wherein: V_{air} : Inhaled volume of air ($10 \text{ m}^3/\text{day}$ for Children and 20 for adults (Horvat et al., 2003)); and C_{air} : T-Hg or MeHg concentration in atmosphere (ng/ m^3). In this study the mean levels of atmosphere concentrations in the industrial area were used to estimate DAI.

DWI (ng/(kg-day)):

$$\text{DWI} = V_{\text{water}} \times C_{\text{water}} / \text{Wt} \quad (4)$$

wherein: V_{water} : the volume of drinking water (800 mL/day for Children and 1200 for adults, according to Guide to Chinese Diet (Chinese Ministry of Health, 2007); and C_{water} : T-Hg or MeHg concentration in drinking water (ng/mL).

DDI (ng/(kg-day)):

According to the Guide to Chinese Diet (Chinese Ministry of Health, 2007), the suggested daily meal quantities for Chinese children were about 150–200 g of grain, 300–400 g of vegetables and fruits, 20–30 g of fish, 20–30 g of meat, 50–70 g of egg, 200–300 mL of milk, 20–30 g of soybean, 10–20 g of food oil and 1–3 g of salt, for adults were 250–400, 500–900, 50–100, 50–75, 25–50, 200–300, 30–50, 25–35, and 6 g, respectively.

$$\text{DDI} = (\text{DM}_{\text{rice}} \times C_{\text{rice}} + \text{DM}_{\text{vegetable}} \times C_{\text{vegetable}} + \text{DM}_{\text{fish}} \times C_{\text{fish}} + \text{DM}_{\text{meat}} \times C_{\text{meat}} + \text{DM}_{\text{egg}} \times C_{\text{egg}} + \text{DM}_{\text{milk}} \times C_{\text{milk}} + \text{DM}_{\text{soybean}} \times C_{\text{soybean}} + \text{DM}_{\text{oil}} \times C_{\text{oil}} + \text{DM}_{\text{salt}} \times C_{\text{salt}}) / \text{Wt} \quad (5)$$

wherein: DM: daily meal quantity of every kind of food (g/day), in this study we selected the medium levels. C: the mean levels of T-Hg and MeHg in every of kind food (ng/g), according to the custom and the Guide to Chinese Diet, the concentrations of rice, soybean and salt were calculated using dry weight, others using wet weight. C_{meat} was the mean levels of pork, chicken, liver and kidney.

For calculation, absorption efficiencies of Hg species in diet by human body were considered as approximately 8% for I-Hg and 95% for MeHg, respectively (WHO, 1990, WHO, 1991). It was assumed absorption efficiencies through SDI and DEI were the same to intake through diet. For inhalation of Hg

vapor, approximately 80% was retained in the body (WHO, 1991). The total daily intakes (TDIs) of T-Hg and MeHg were assessed according to the following Eq. (6), and the resultant estimates are shown in Table 5:

$$\text{TDIs} = \text{SDI} + \text{DEI} + \text{DAI} + \text{DWI} + \text{DDI}. \quad (6)$$

1.5. Statistical analysis

All results were expressed as means \pm S.D. Statistical analysis was performed using SPSS. Correlation coefficients were studied using Pearson correlation analysis. The differences were regarded as statistically significant at $p < 0.05$.

2. Results and discussion

2.1. Hg in air, dust, soil, and crop samples

The mean T-Hg concentration of atmosphere samples in industrial area was approximately twice the value of Baifengao village and 4.3 times the value of Wenling (Table 1). The mean concentration of T-Hg in air in Fengjiang town (including the industrial area and Baifengao village) was 23.1 ng/m^3 , which was higher than those reported for Guiyang ($5.2\text{--}8.5 \text{ ng/m}^3$, Feng et al., 2003) and Beijing ($6.2\text{--}10.7 \text{ ng/m}^3$, Liu et al., 2002), and the data was several to even tens of times higher than the background Hg value of atmosphere in China (0.6 to 1.7 ng/m^3). For soil samples, the critical value for T-Hg recommended by the Chinese National Standard Agency in arable soil is $0.15 \text{ }\mu\text{g/g}$ (CNSA, 1995). Evidently, concentrations for T-Hg in arable soil exceeded this value at all sampling sites (Table 1). T-Hg concentration in soil from reference location ($0.1 \text{ }\mu\text{g/g}$), which represented the regional baseline concentration, was much lower. T-Hg concentrations in worldwide uncontaminated soils

were in the range of 0.01 to $0.50 \text{ }\mu\text{g/g}$ (Qiu et al., 2006). The critical value for MeHg has not been issued by CNSA, so comparison with critical data was impossible. However, the proportion of soil MeHg in T-Hg was in accordance with previous studies, which reported that MeHg in soil usually accounts for less than 1% of T-Hg (Mailman and Bodaly, 2005). For dust samples, T-Hg and MeHg concentrations in workshop-floor dust samples were approximately 12 and 9 times greater than those of soil samples from the industrial area, 29 and 21 times higher than those of Baifengao village, and 376 and 168 times higher than those of reference locations ($p < 0.01$, independent t-test, Table 1), respectively. Results for T-Hg and MeHg in atmosphere, soil and dust samples clearly indicated that the concentrations rapidly decreased with the distance from the industrial area. This gradient indicated regional contamination due to atmospheric transport and deposition of Hg from the e-waste recycling facilities. Pearson's correlation coefficients between dust and soil samples in industrial area were very high, with a significant correlation ($r = 0.39$, $p < 0.01$). To our knowledge, there were no chemical factories located near our sampling locations in Fengjiang town. Thus, the elevated concentrations of Hg in soil, atmosphere, and crop in our study implicated e-waste recycling as the major source of Hg emission.

The average T-Hg concentrations of each crop exceeded the maximum value of $0.020 \text{ }\mu\text{g/g}$ recommended by CNSA (CNSA, 1994) to the tolerance limit of Hg in human foods, indicating that almost all of the agricultural crops growing in the study area were adversely affected by Hg contamination. Elevated concentrations of MeHg were obtained with the peak value of 41.6 ng/g in rice sample from Site 6. Because MeHg is readily bioavailable, the high MeHg concentration in rice could pose a threat to the health of local residents (Cheng et al., 2005; Feng et al., 2008). MeHg concentrations in crop samples were higher than those in the arable soil samples. And different kinds of crops showed different accumulation

Table 1 – Concentrations of T-Hg and MeHg in atmosphere, soil, dust and crop samples from industrial area, Baifengao village and reference locations ($n = 15$).

Location	Site	Atmosphere	Soil			Species	Dust or crop		
		T-Hg (ng/m^3)	T-Hg ($\mu\text{g/g}$)	MeHg (ng/g)	% MeHg		T-Hg ($\mu\text{g/g}$)	MeHg (ng/g)	% MeHg
Industrial area	1	27.6	2.8 ± 0.7	1.8 ± 0.5	0.06	Dust	65.4 ± 21.1	9.5 ± 1.5	0.01
	2	36.2	7.1 ± 1.2	0.8 ± 0.2	0.01		81.5 ± 23.7	19.1 ± 3.3	0.02
	3	43.4	1.8 ± 0.4	0.7 ± 0.3	0.04		19.0 ± 5.3	4.7 ± 0.9	0.03
	4	29.8	2.0 ± 0.7	0.8 ± 0.3	0.04		12.2 ± 2.1	5.4 ± 1.1	0.04
	5	16.7	1.9 ± 0.5	0.6 ± 0.4	0.03		10.1 ± 3.0	3.0 ± 1.0	0.03
Baifengao village	Mean	30.7 ± 9.9	3.1 ± 2.4	0.9 ± 0.5	0.03	Mean	37.6 ± 31.3	8.4 ± 6.9	0.02
	6	19.6	2.8 ± 0.3	0.6 ± 0.2	0.02	Rice	0.081 ± 0.015	41.6 ± 10.3	51.3
	7	21.1	1.3 ± 0.4	0.4 ± 0.1	0.03	Rice	0.020 ± 0.006	10.7 ± 1.6	53.5
	8	15.5	1.2 ± 0.3	0.3 ± 0.2	0.03	Corn	0.068 ± 0.010	8.5 ± 1.9	12.5
	9	9.3	0.8 ± 0.4	0.5 ± 0.2	0.06	Soybean	0.043 ± 0.003	7.8 ± 1.8	18.1
Wenling	10	11.5	0.4 ± 0.2	0.2 ± 0.1	0.05	Cole ^a	0.025 ± 0.005	1.1 ± 0.3	4.4
	Mean	15.4 ± 5.1	1.3 ± 1.0	0.4 ± 0.2	0.03	Grain ^b	0.056 ± 0.032	20.3 ± 18.5	36.2
	11	7.2	0.1 ± 0.1	0.05 ± 0.05	0.05	Rice	0.012 ± 0.008	4.1	34.2

Pearson's correlation coefficient of T-Hg between soil and dust in industries area, $r = 0.39$, $p < 0.01$.

Pearson's correlation coefficient of T-Hg between soil and crop in Baifengao village, $r = 0.09$, $p > 0.05$.

Pearson's correlation coefficient of T-Hg between atmosphere and crop in Baifengao village, $r = 0.20$, $p > 0.05$.

Pearson's correlation coefficient between T-Hg and MeHg, $r = 0.15$, $p > 0.05$.

^a Wet weight.

^b Mean levels of rice and corn.

abilities for MeHg, indicating different sources of MeHg. Most probably Hg in crop in this area reflected combined uptake of Hg from the soil and from the atmosphere through absorption by plant leaves (Horvat et al., 2005). It was difficult to interpret the concentrations in crop vs. Hg in soil or atmosphere. There was no correlation between T-Hg in soil ($r = 0.01$, $p > 0.05$) or atmosphere ($r = 0.20$, $p > 0.05$) and in crop. This clearly indicated that the uptake and retention of Hg in crop were influenced by a number of factors, of which the concentration of T-Hg in soil and atmosphere may only be external environmental factors, and not the most important, of the variables. However, the intramural mechanism of these crop uptaken Hg was still unclear.

The results showed that the percentage of MeHg in soil sample was rather low, and the percentage of MeHg in soils rarely exceeded 1%, which was in accordant with literature data (Gnamus et al., 2000; Horvat et al., 2005). The proportions of MeHg to T-Hg in rice samples were found up to 51.3% and 53.5% in Sites 6 and 7, which was corresponding to the results obtained in similar kinds of samples from Qinzhenh chemical plant and Wanshan Hg mine areas (Cheng et al., 2005; Cheng et al., 2006). There was no correlation between the concentrations of T-Hg and MeHg ($r = 0.15$, $p > 0.05$), which reflected the well-known fact that the T-Hg concentration was not the only factor influencing transformation mechanisms of Hg in different environmental conditions (Gray et al., 2000; Horvat et al., 2005).

2.2. Hg in foodstuff samples

Mean concentrations of T-Hg and MeHg in hen, pig, fish, milk, water, oil and salt samples analyzed in this study are presented in Table 2. For hen samples, the highest T-Hg and MeHg concentrations were found in liver, followed by chicken, egg, and brain. For pig, the highest T-Hg and MeHg concentrations were found in kidney, followed by liver, meat and brain. The percentage of MeHg in tissue sample was rather high, and ranged from 36% to 82.1%. T-Hg content in different tissues (except brain tissues) of hens and pigs both exceeded the maximum concentration of 50 ng/g recommended by CNSA (1994) to the tolerance limit of Hg in human foods,

indicating that poultry growing in the area was adversely affected by Hg contamination. However, the Hg levels in milk, drinking water, bean oil and common salt were all below critical data, because these commodities were usually made in other areas. Domestic hens and pigs from Baifengao village lived on rice and local feedstuff; they closely shared the same environment as their owners, and were therefore exposed, at least in part, to the same pollutants. Former studies have suggested that domestic animals such as dogs could serve as surrogate indicators of human metal exposure (López-Alonso et al., 2007). So, more effort is necessary to protect the local ecosystem and human health in the e-waste recycling areas.

In this study, the average T-Hg and MeHg concentrations for all species of fish were 229.8 and 178.1 ng/g, respectively. This result is more severe than the study by Li et al. (2012), which found that MeHg concentrations ranged from 12.0 to 38.5 ng/g in fish tissue from 7 provinces, south China. Approximately 93% of the samples showed T-Hg levels below the limit established by CNSA (0.3 ng/g). Fish are usually considered to be one of the most important sources of MeHg exposure for human beings. Bioaccumulation of MeHg in fish depends on the trophic level and it is also influenced by fish age, length or weight (Zhang and Wong, 2007), so different kinds of fish have different MeHg levels. The proportion of MeHg relative to T-Hg in the fish ranged from 70% to 93%, the mean value was 79%, which was consistent with mercury accumulation in fish as documented in other studies (Fréry et al., 2001).

2.3. Hg in hair samples

Hair is mostly used as an indicator for human exposure levels because it is easy and relatively nonintrusive to sample and it can give information on exposure integrated over time. Harada et al. (1999) reported that the upper limit of normal hair T-Hg level was 10 µg/g. The Joint FAO/WHO Expert Committee on Food Additives (2003) value for provisional tolerable weekly intake is 1.6 µg/(kg·week), corresponding to 2.2 µg/g in the hair. Hair T-Hg mean concentrations in this study were shown in Table 3, which indicates that different

Table 2 – Concentration of T-Hg and MeHg in hen, pig, fish, milk, drinking water, bean oil and common salt samples from Baifengao village ($n = 15$, wet weight).

Tissue samples	Hen and pig			Tissue samples	Fish, water, oil, milk and salt		
	T-Hg (ng/g)	MeHg (ng/g)	% MeHg		T-Hg (ng/g)	MeHg (ng/g)	% MeHg
Hen				Pomfret	312.7 ± 120.4	220.3 ± 101.1	70.5
Chicken	53.7 ± 10.3	19.5 ± 7.6	36.0	Ribbon fish	243.2 ± 89.3	178.9 ± 64.1	73.6
Brain	14.3 ± 3.6	10.8 ± 4.3	75.5	Small yellow croaker	171.1 ± 69.9	134.7 ± 81.7	78.7
Liver	59.0 ± 22.5	40.5 ± 10.1	68.6	Horsehead	192.1 ± 78.6	178.3 ± 71.2	92.8
Egg	52.3 ± 19.8	26.9 ± 13.2	51.4		229.8 ± 63.1	178.1 ± 35.2	78.9
Pork	50.8 ± 12.3	27.7 ± 9.3	54.5	Fish mean			
Pig ^a				Milk	2.9 ± 0.2	–	–
Brian	11.2 ± 3.9	9.2 ± 3.1	82.1	Drinking water (ng/L)	13.8 ± 2.6	0.10 ± 0.04	0.7
Liver	72.5 ± 20.1	41.2 ± 18.2	56.8	Food oil	5.4 ± 1.8	–	–
Kidney	82.1 ± 28.9	39.5 ± 14.5	48.1	Common salt	4.5 ± 1.7	–	–
Meat mean ^b	60.1 ± 10.8	32.1 ± 9.9	53.4				

^a $n = 5$.

^b Mean levels of chicken, hen liver, pork, pig liver and pig kidney; – under detection limit.

Table 3 – Hg levels in hair samples and investigated factors (n = 67).

	I-Hg ($\mu\text{g/g}$)	MeHg ($\mu\text{g/g}$)	T-Hg ($\mu\text{g/g}$)	% MeHg	Fish meals per month	Time stay at industry area (hr/day)
<i>Worker</i>						
Administrators of industrial area	0.45 ± 0.08	0.39 ± 0.21	0.84 ± 0.18	44.0	3.0 ± 1.3	4.9 ± 1.6
E-waste dismantling workers	0.77 ± 0.18	0.30 ± 0.10	1.07 ± 0.21	27.9	1.9 ± 1.0	11.1 ± 1.3
Recover metal residues workers	1.27 ± 0.30	0.26 ± 0.07	1.53 ± 0.33	17.3	1.8 ± 0.7	9.8 ± 1.6
Acid baths workers	1.41 ± 0.37	0.23 ± 0.07	1.64 ± 0.38	14.4	1.7 ± 0.6	10.2 ± 1.6
Mean	0.84 ± 0.48	0.35 ± 0.21	1.19 ± 0.41	33.5	2.5 ± 1.4	8.1 ± 3.9
<i>Farmer</i>						
Farmers of Baifengao village	0.22 ± 0.06	0.66 ± 0.29	0.88 ± 0.30	73.4	4.8 ± 0.9	1.3 ± 0.9
<i>Civilian</i>						
Civilian of Fengjiang town	0.28 ± 0.10	1.25 ± 0.58	1.52 ± 0.62	80.6	10.0 ± 3.1	1.4 ± 0.8
<i>Reference</i>						
Civil servants of Wenling	0.13 ± 0.06	1.21 ± 0.61	1.40 ± 0.59	89.0	10.9 ± 2.3	–
Pearson's correlation coefficient between I-Hg and MeHg levels in the hair, $r = -0.65$, $p < 0.01$.						

sampling group population hair T-Hg concentrations were lower than $2.2 \mu\text{g/g}$.

Hair inorganic Hg (I-Hg) concentrations were the difference between T-Hg and MeHg concentrations. The workers had significantly higher I-Hg levels ($0.84 \mu\text{g/g}$) than the other three groups ($p < 0.01$, independent t-test, Table 3). The mean concentrations of I-Hg in the worker samples were almost 7 times higher than those in the references. There were significant correlations between T-Hg and I-Hg concentrations in hair samples from workers ($r = 0.51$, $p < 0.05$), and I-Hg concentrations in hair samples accounted for 66.5%, on average for workers. This indicated a certain level of I-Hg exposure for the workers in the industrial area. The highest mean concentrations of I-Hg ($1.41 \mu\text{g/g}$) and T-Hg ($1.64 \mu\text{g/g}$) were both detected in the acid bath workers, followed by burning to recover metal residue workers, electronic waste dismantling workers and administrators in the industrial area. It could be mainly due to inhalation of contaminated air, dust and smoke when they dismantled electronic wastes by employing very primitive working methods and very little or no personal protection measures.

However, samples from civilians in Fengjiang town and civil servants in Wenling showed highest hair MeHg levels with mean values of 1.25 and $1.21 \mu\text{g/g}$, respectively. Significant correlations between T-Hg and MeHg concentrations were found in hair samples from civilians ($r = 0.96$; $p < 0.01$) and civil servants ($r = 0.99$, $p < 0.01$). And the average percentages of MeHg accounted for 81% of civilians and 89% of civil servants. Most studies in the former studies reported that MeHg accounted for above 70%–80% of the total hair Hg and the major route of MeHg exposure was through fish consumption (Fréry et al., 2001; Tsuji et al., 2002). In this study, the investigated factors showed that those civilians and references had both high fish consumption frequency (Table 3).

Further Pearson's correlation coefficient studies showed that a strong positive correlation ($r = 0.81$; $p < 0.01$) was observed between hair I-Hg and stay time in the industrial area, but a strong negative correlation ($r = -0.82$; $p < 0.01$) was found between hair MeHg and stay time (Table 4). And the Pearson's correlation coefficients between hair I-Hg and atmosphere T-Hg

showed a strong positive correlation ($r = 0.68$; $p < 0.01$). However, the correlation coefficients between fish consumption frequency and hair Hg levels showed contrary to response, a strong positive correlation ($r = 0.91$; $p < 0.01$) was observed between MeHg and frequency, but a strong negative correlation ($r = -0.75$; $p < 0.01$) was found between I-Hg and frequency. I-Hg and the MeHg levels in all hair samples showed significant negative correlations ($r = -0.65$; $p < 0.01$). These results confirmed that I-Hg and MeHg exposure came from different sources. Workers in this area were poor and the fish price was much higher than other kinds of food. Therefore, high fish consumption frequency was not expected for them (Table 3). They were mainly exposed to Hg vapor through long-time inhalation of contaminated air and dust and larger amount of rice consumption, however other populations in this area and in references were mainly exposed to MeHg through high-frequency fish consumption.

2.4. Exposure assessment

Tables 5 and 6 present estimates of intakes of T-Hg, MeHg and I-Hg subdivided by different routes and food types for Chinese population. In all routes, DDI was the major source (>82%) of T-Hg, MeHg and I-Hg exposures, while non-dietary sources contributed only a small proportion (<18%) to TDIs. In all foods, T-Hg and MeHg intakes from rice and fish were significantly higher than from any other food, and the proportion range from both foods was 66%–83% of TDIs. Our results are in line with previous studies conducted in southern China (Li et al., 2012; Shao et al., 2013), which showed that both rice and fish consumption were the two main routes of Hg exposure. While

Table 4 – Pearson's correlation coefficients between mercury levels and investigated factors.

	I-Hg	MeHg	T-Hg
Stay time every day in the industrial area	0.81^*	-0.82^*	0.17
Fish consumption frequency every month	-0.75^*	0.91^*	0.21
Atmosphere T-Hg levels	0.68^*	-0.23	0.61^*
* Significant correlation at $p < 0.01$.			

Table 5 – Estimated daily intakes of T-Hg, MeHg and I-Hg for children.

		T-Hg		MeHg		I-Hg ^a	
		T-Hg (ng/(kg·day))	%TDIs	MeHg (ng/(kg·day))	%TDIs	T-Hg (ng/(kg·day))	%TDIs
SDI		6.5	0.8	0.02	0.003	6.48	5.0
DEI		0.2	0.02	0.0006	0.00009	0.1994	0.2
DAI		16.3	2.0	–	–	16.3	12.7
DWI		0.06	0.007	0.005	0.001	0.055	0.04
DDI	Rice	258.3	31.3	225.0	32.3	33.3	25.9
	Vegetable	69.0	8.4	24.4	3.5	44.6	34.7
	Fish	288.9	35.0	282.0	40.5	6.9	5.4
	Meat	54.5	6.6	50.8	7.3	3.7	2.9
	Egg	110.3	13.4	102.2	14.7	8.1	6.3
	Milk	3.8	0.5	–	–	3.8	3.0
	Soybean	17.0	2.1	12.4	1.8	4.6	3.6
	Oil	0.4	0.05	–	–	0.4	0.3
	Salt	0.05	0.006	–	–	0.05	0.04
	Total DDIs	802.3	97.2	696.8	99.99	105.45	82.1
TDIs		825.3	100	696.8	100	128.48	100

SDI: soil and dust ingestion; DEI: dermal exposure absorption; DAI: daily air inhalation; DWI: drinking water intake; DDI: daily dietary intake; TDI: total daily intakes.

^a I-Hg=T-Hg – MeHg.

vegetables and rice contribute quantity of I-Hg were much higher than any other foods. DDI and DAI were the dominant pathways of exposure to I-Hg. The results also reflected the origin of hair Hg of this area, in which high I-Hg concentrations in hair from workers were attributed to occupational exposure of Hg vapor and large amount of rice consumption. However, high MeHg levels in hair from other population were mainly from high-frequency fish consumption. The estimated intakes of T-Hg, MeHg and I-Hg from SDI, DAI, DWI and DDI were greater in children than in adults, and DEI contributed to greater T-Hg and MeHg intakes in adults than in children. The rates of SDI, DEI, DAI, DWI and DDI could vary between children and adults, due to the differences in the ingestion rate, body surface, and body weight. The estimated TDIs of I-Hg intakes for both children (128.5 ng/(kg·day)) and adults (58.6) did not exceed

570 ng/(kg·day) recommended by WHO (2010), T-Hg intakes for children (825.3 ng/(kg·day)) greatly exceeded 710 ng/(kg·day) according to WHO (1990), and TDIs of T-Hg for adults (439.9 ng/(kg·day)) were below the WHO recommended values. However, MeHg intakes of both children (696.8 ng/(kg·day)) and adults (381.3) greatly exceeded the dietary reference dose of 230 ng/(kg·day) as recommended by WHO (2003). Therefore, the residents, especially young children and pregnant women might be at high risk of Hg exposure.

3. Conclusions

Our results suggested that elevated Hg levels in ambient air, soil and crops were seriously impacted by artisanal e-waste

Table 6 – Estimated daily intakes of T-Hg, MeHg and I-Hg for adults.

		T-Hg		MeHg		I-Hg ^a	
		T-Hg (ng/(kg·day))	%TDIs	MeHg (ng/(kg·day))	%TDIs	I-Hg (ng/(kg·day))	%TDIs
SDI		0.7	0.2	0.001	0.0003	0.699	1.2
DEI		0.5	0.1	0.001	0.0003	0.499	0.9
DAI		8.2	1.9	–	–	8.2	14.0
DWI		0.02	0.005	0.002	0.0005	0.018	0.03
DDI	Rice	119.9	27.3	104.9	27.5	15	25.6
	Vegetable	34.5	7.8	12.2	3.2	22.3	38.1
	Fish	216.6	49.2	211.5	55.5	5.1	8.7
	Meat	34.1	7.8	31.8	8.3	2.3	3.9
	Egg	17.2	3.9	16.0	4.2	1.2	2.0
	Milk	1.0	0.8	–	–	1	1.7
	Soybran	6.9	1.6	4.9	1.3	2	3.4
	Oil	0.2	0.05	–	–	0.2	0.34
	Salt	0.04	0.009	–	–	0.04	0.07
	Total DDIs	430.4	97.9	381.3	99.99	49.1	83.9
TDIs		439.9	100	381.3	100	58.6	100

^a I-Hg=T-Hg – MeHg.

recycling. Workers of e-waste recycling plant and residents in Fengjiang Town showed high hair I-Hg or MeHg levels, indicating that they were at high risk of Hg vapor exposure via inhalation and rice or fish consumption. The TDI estimation presented that population in this area was at potential risk of Hg exposures through fish and rice consumption.

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