CrossMark

RESEARCH ARTICLE

Seafood consumption among Chinese coastal residents and health risk assessment of heavy metals in seafood

Ran Zhao[1] · Shuangshuang Yan[2] · Min Liu[1] · Bi Wang[1] · Dong Hu[1] · Dongbei Guo[1] · Juan Wang[3] · Wanting Xu[1] · Chun Fan[1]

Abstract The aims of the present study were to obtain the seafood dietary patterns of coastal residents, to determine the concentrations of heavy metals, and to evaluate the possible health risks caused by seafood intake. The daily food intakes of 24 types of seafood were collected from 738 participants from Xiamen, a southern Chinese coastal city, using food frequency questionnaire (FFQ) and dietary history method. One hundred and fifty-six samples of 14 types of highest intake seafood were collected from local markets for lead (Pb), cadmium (Cd), chromium (Cr), mercury (Hg), and arsenic (As) determination. Health risks via seafood consumption were evaluated by calculating the target hazard quotient (THQ) and the total hazard index (HI) for carcinogenic and non-carcinogenic effects recommended by the US Environmental Protection Agency. The results showed that the seafood daily intake of Xiamen residents was 61.5 (2.14, 115) g/day. The concentrations of Pb, Cd, Cr, Hg, and As in seafood were ND–0.45 mg/kg, ND–0.19 mg/kg, ND–0.80 mg/kg, ND–0.70 mg/kg, and 0.32–16.9 mg/kg, respectively. Concentrations of Cd and As in some samples were higher than national limitation standards. Consumption of 14 common types of seafood would not pose non-carcinogenic risk. However, some types, such as sparuslatus, oyster, and porphyra tenera, would form a carcinogenic risk. Regardless of a carcinogenic or non-carcinogenic risk, As posed the highest risk on humans. The observed HI value for non-carcinogenic effect of all metals in all seafood reached 0.69–2.20, and the metal orders of risk can be listed as As > Hg > Cr > Cd > Pb, reiterating the risk of As is a matter of concern in seafood from Xiamen markets.

Keywords Coastal residents · Seafood · Heavy metal · Risk assessment · Target hazard quotient (THQ)

✉ Ran Zhao
  zhaoran@xmu.edu.cn

✉ Chun Fan
  fanchun65@163.com

[1] State Key Laboratory of Molecular Vaccinology and Molecular Diagnostics, School of Public Health, Xiamen University, Xiamen 361102, Fujian, People's Republic of China

[2] The First Affiliated Hospital of Henan University of Science and Technology, Luoyang 471003, Henan, People's Republic of China

[3] Department of Preventive Medicine, Xiamen Medical School, Xiamen 361008, Fujian, People's Republic of China

Introduction

Seafood is a significant source of food, especially for coastal residents, and has been widely acknowledged as an integral component of a well-balanced diet. For example, fish is known for providing a healthy source of high-quality proteins, vitamins, and omega-3 polyunsaturated fatty acids (PUFAs) (Pieniak et al. 2010). In contrast to the potential health benefits of dietary seafood intake, serious marine pollution caused by industrial usage of chemical pollutants, such as heavy metals, has arisen increasing concerns on marine food safety (Dorea 2008; Martorell et al. 2011). Typical toxic heavy metals such as lead (Pb), cadmium (Cd), chromium (Cr), mercury (Hg), and arsenic (As) exist in the marine environment

and can be bioaccumulated by marine organisms and even biomagnified through food chains (Tadiso et al. 2011), thus posing a serious threat to human health (Rainbow and Luoma 2011). With rapid urbanization and industrialization, coastal areas of China are now facing great challenges of heavy metal contamination. A large number of studies report that seafood from many coastal regions of China has been seriously polluted by heavy metals (Wan et al. 2008; Fang et al. 2009; Ding et al. 2009; Deng et al. 2010; Pan and Wang 2012). In this regard, along with economic growth, large-scale pollution monitoring and heavy metal determination have been performed in Asian developing nations (Agusa et al. 2007).

Diet comprises the main route of exposure to heavy metals in the general population (Kim and Lee 2010). Even low heavy metal concentration may threaten the health of aquatic and terrestrial organisms, including human beings (Sarmiento et al. 2011). More and more research regarding certain threats associated with the consumption of seafood contaminated with heavy metals are conducted (Wang et al. 2012; Zhang and Wang 2012; Liu et al. 2013a). Therefore, risk assessment of contaminants via dietary intake is an important issue. Contaminants can be classified as carcinogen and non-carcinogen, which lead to carcinogenic and non-carcinogenic effects on organisms. As suggested by US Environment Protection Agency (USEPA), to protect human health, the lower one of the maximum allowable consumption rate of seafood considering either carcinogenic or non-carcinogenic effect should be used (Yu et al. 2014). The USEPA has developed human health risk assessment methods for carcinogenic and non-carcinogenic contaminants (USEPA 2000). Human health risk assessments suggest that heavy metals, such as As, Hg, and Pb, in fish or mollusk from South China (Wang et al. 2013), Taihu Lake (Yu et al. 2014), Beijing markets (Fang et al. 2014), and Shanghai markets (Lei et al. 2015) all exceeded the safety limit.

In some region of China, the consumption of seafood is high, such as Xiamen city, Fujian province (Fig. 1), whose population not only consumes but also provides seafood for the rest of China and even abroad (China Statistical Yearbook, 2014, http://www.stats.gov.cn/tjsj/ndsj/2014/indexeh.htm). Thus, the living standard and dietary habit, especially seafood intake, of Xiamen's inhabitants would represent that of frequent seafood consumers. Although Xiamen is not a heavily industry polluted city, its rapid urbanization still produces various contaminants. High metal concentration in water columns and aquatic species was reported in the vicinity of Maluan Bay (a coastal lagoon in the northwest of Xiamen, considered as one of the most industrialized and urbanized coastal lagoon in China) (Wang et al. 2014), indicating that the possible health risk caused by high seafood intake in the coastal residents

should be urgently assessed. This article assessed the intakes of the widest variety of seafood species collected from 738 Xiamen residents. The specific objectives of this study were: (1) to obtain the consumer daily intakes of 24 types of seafood, (2) to quantify the concentrations of Pb, Cd, Cr, Hg, and As in common seafood, and (3) to evaluate the possible health risk of heavy metals due to seafood dietary exposure, considering both carcinogenic and non-carcinogenic effects.

## Materials and methods

Participants and design

The design and methodology of the present study have been described in detail elsewhere (Fang et al. 2014). According to the land area, study participants were enrolled (from March 2013 to May 2014) from six districts (two inside the island (Si Ming and Hu Li) and four outside the island (Xiang An, Tong An, Ji Mei, and Hai Cang)) across all regions of Xiamen city (Fig. 1). A representative sample (n 850) of the participants aged ≥18 years of either sex was randomly selected from communities and medical examination centers during their routine physical examination and asked to complete a face-to-face investigated questionnaire. Of the 850 people invited, 112 were unavailable for contact or could not provide the entire information during the survey. In total, 738 participants completed the questionnaire (87 % participation rate). Of the 738 participants, 40 were investigated again approximately 4 weeks apart for quality control. All 738 participants were included in the analyses of gender, age, weight, neighborhood, and 24 types of seafood intakes. The present study was conducted according to the guidelines laid down in the Declaration of Helsinki, and all procedures involving human subjects were approved by the Medical and Health Research Ethics Committee of School of Public Health, Xiamen University. Patients with seafood allergies or chronic diseases were excluded from the study. All participants provided written informed consent.

Dietary survey methodology

A dietary history method and a food frequency questionnaire (FFQ), the validity of which has been examined in other studies (Woo et al. 1997; Yu et al. 2011), were completed by a qualified dietitian or trained study coordinator. Dietary assessment included general information (such as age, gender, height, and weight) and information about dietary intakes for 24 types of seafood. The questionnaire consisted of 24 types in the following seven species: fish (six types), mollusk (three types), crustacean

Fig. 1 The map of the research area: Xiamen, Fujian province, southeast China
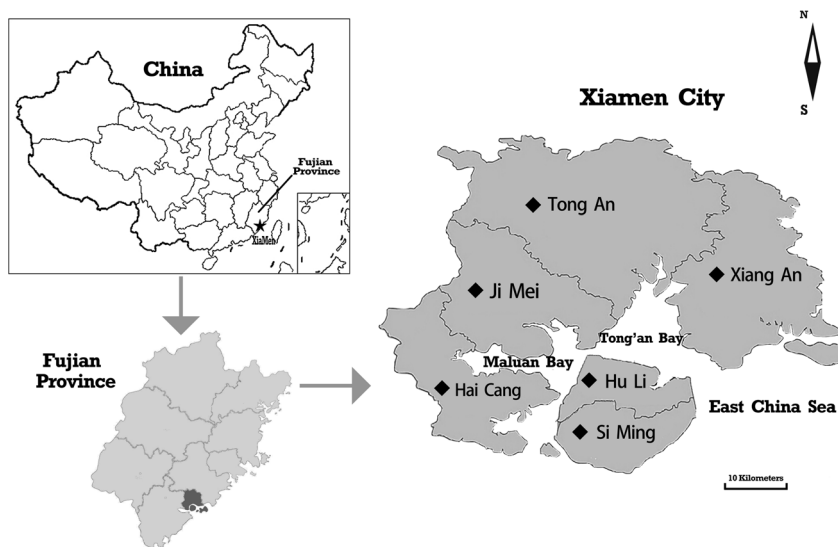
(six types), alga (two types), cephalopod (three types), and drysaltery (four types). Participants were asked to recall the seafood dietary history of the past week in a face-to-face interview, during which a survey on a week's seafood diet would be carried out. Over the interview, each participant was asked to complete the questionnaire with information about the seafood types and the size of each portion. Portion size was explained to the participants using actual samples of individual seafood portions.

Data were cross-checked by examining the general information (e.g., if weight was slipped) and dietary pattern (e.g., if alga was skipped) to determine whether it corresponded to the number of times seafood was purchased over a 1-week period. In case of discrepancies, the questionnaire was re-checked with the participant. The types of seafood consumed were specified by investigators' explaining and a booklet containing photographs of different types. Data analysis was performed by using Microsoft Excel 2013 software. Statistical analyses were conducted by using the SPSS version 16.0 statistical analysis packages (SPSS, Inc., Chicago, IL, USA). Median and quartiles were summarized as P50 (P25, P75) for description when the result of daily seafood consumption was non-normal distribution.

Sampling, preparation, and analysis

One hundred and fifty-six samples of the most four consumed seafood species (fish, mollusk, crustacean, and alga) were randomly acquired in traditional seafood markets and supermarkets from six district regions of Xiamen city. The sampling locations are illustrated in Fig. 2. Fish species include weever, *Nibea coibor*, sea perch, sparuslatus, red porgy, and white pomfret. Mollusk species include oyster, *Meretrix petechialis*, and *Ruditapes philippinarum*. Crustacean species include

Metapenaeus ensis, *Penaeus monodon*, and shrimp. Alga species include *Porphyra tenera* and *Laminaria japonica*. All samples were brought to the lab in clean plastic bags for treatment as soon as possible.

To eliminate sediment particles, samples were washed with tap water and deionized water three times and dried with tissue paper. Fish scales were scraped with stainless steel blade, and edible parts of all samples were removed, washed pureed, and placed in sealed polyethylene bags at −20 °C before analysis. Samples were treated using HNO_3–H_2O_2 or HNO_3–$HClO_4$ (Song et al. 2015) before taken into the microwave digestion system (MARS Xpress, USA CEM). Samples were analyzed by atomic absorption spectrometry (AAS) for Pb, Cd, and Cr and atomic fluorescence spectrometry (AFS) for Hg and As according to a modification of Chinese standards (GB/T 5009.12-2010, GB/T 5009.15-2014, GB/T 5009.123-2014, GB/T 5009.17-2003, and GB/T 5009.11-2003, respectively). Standard reference materials for seafood (GBW-10050), obtained from China National Center for Standard Reference Materials, were digested simultaneously for quality control. The detection recoveries of the methods were 90–105 %. Data were analyzed using the SPSS version 16.0 statistical analysis packages and evaluated by comparing with national limitation standards.

Health risk assessment

Average daily dose (ADD) was estimated based on the daily intake rates, concentrations of seafood samples, exposure frequency, exposure duration, average time of exposure, and body weight, see formula (1) (Fang et al. 2014; Song et al. 2015):

$$ADD = \frac{C \times IR \times EF \times ED}{BW \times TA}. \tag{1}$$

Fig. 2 Illustration of sampling
sites in Xiamen city, Fujian
province, southeast China

1. Qian Pu Farmer's Market

2. Rui Jing Wal-Mart Supermarket

3. No.8 Seafood Market

4. Hu Li Farmer's Market

5. Zongpu seafood market

6. Xiang An Farmer's Market

7. Tong An Farmer's Market

8. Ji Mei Farmer's Market

9. Hai Cang Farmer's Market
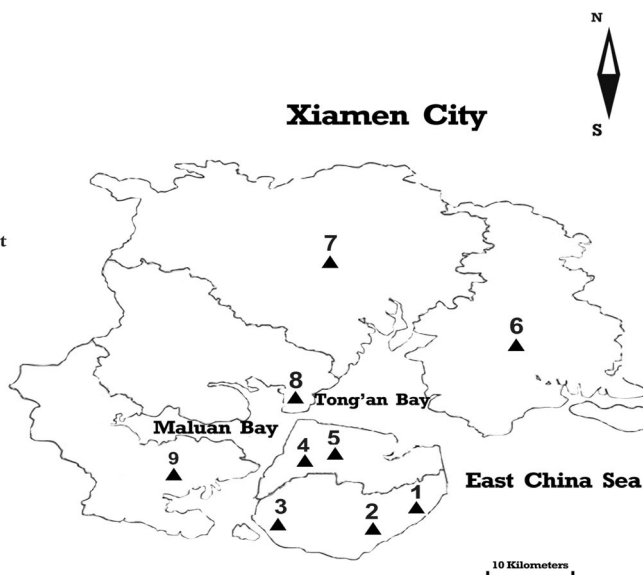
In formula (1), ADD is the average daily dose of heavy metals in seafood (mg/kg/day), C is the concentration of heavy metals in seafood samples (mg/kg wet weight (w.w.)), IR is the rate of seafood consumption (kg/day), EF is the exposure frequency (day/year), ED is the exposure duration (70 years), equivalent to the average lifetime, BW is the average body weight of the participants (kg), and TA is the average exposure time for non-carcinogens ($EF \times ED$) (Storelli 2008; Fang et al. 2014).

According to health risk assessment model (USEPA 2011), the possible health risk caused by seafood consumption of coastal residents was appraised using the method of target hazard quotient (THQ) (Storelli 2008; Yu et al. 2014; Ahmed et al. 2015). Herein, THQ is defined as the ratio of daily seafood consumption rate in relation to maximum allowable seafood consumption rate considering the carcinogenic and non-carcinogenic effects of the heavy metals, see formulas (2) and (3), respectively:

$$THQ = \frac{ADD}{ARL \big/ CSF}. \tag{2}$$

$$THQ = \frac{ADD}{RfD}. \tag{3}$$

In formulas (2) and (3), THQ is the target hazard quotient of heavy metals in seafood, ADD is the average daily dose of heavy metals in seafood (mg/kg/day), ARL is the maximum acceptable individual lifetime risk level and 10^{-5} was utilized in the present study (Yu et al. 2014), CSF is the cancer slope factor of InAs for a carcinogenic effect (1.5 $(mg/kg/day)^{-1}$) (USEPA 2000), and RfD is the oral reference dose of Pb, Cd, Cr, MeHg, and InAs for non-carcinogenic effects (4, 1, 3, 0.1, and 0.3×10^{-3} mg/kg/day, respectively) (available in the Integrated Risk Information System (IRIS) of the USEPA http://www.epa.gov/iris/). THQ value >1 means the level of

exposure is higher than the reference dose, and daily exposure at this level is likely to cause adverse effects during a person's lifetime (Yi et al. 2011; Fang et al. 2014).

The exposure is usually not to a single toxicant, but two or more. Thus, negative effects may come up and the hazard index (HI) from THQs is expressed as the sum of the hazard quotients (USEPA 2011). The calculation formula of HI was listed as follows (Storelli 2008):

$$HI = THQ_{Pb} + THQ_{Cd} + THQ_{Cr} + THQ_{Hg} + THQ_{As}. \tag{4}$$

Results and discussion

Questionnaires

General situation of the participants

After data screening, the effective investigated proportion was 87 % (738 residents), in total 850 investigated residents. Among the 738 participants, 246 (33 %) are from two inside island districts and 492 (67 %) are from four outside island districts, 380 (51 %) are male and 358 (49 %) are female. The mean (SD) body weight of the participants was 61.3 (±9.72) kg.

Seafood consumption of the participants

The results showed that among 738 participants, the population ratios of seafood consumption from high to low were alga (89.3 %), mollusk (80.4 %), fish (79.0 %), crustacean (72.8 %), drysaltery (58.3 %), and cephalopod (54.5 %). Because the result of daily seafood consumption was non-normal distribution,

median and quartile analysis were conducted and summarized in Table 1 for description. The daily intake of 24 types of seafood was 61.5 (2.14, 115) g/day, which was much higher than the data 41.62 g/day reported in China Statistical Yearbook (2014), 52.05 g/day reported in FAO Statistical Yearbook (2013) (http://www.fao.org/docrep/018/i3107e/i3107e00.htm), and the data 57.5 g/day obtained from the survey conducted by Wang et al. (Wang et al. 2005). The most four consumed seafood species by Xiamen residents are fish (16.1 (2.14, 42.9) g/day), mollusk (12.9 (3.21, 34.3) g/day), alga (9.29 (2.14, 19.3) g/day), and crustacean (4.29 (1.34, 12.0) g/day), which is lower than fish (40.8 g/day) in Shanghai (Yu et al. 2012) and fish (71.43–107.14 g/day) in Lake Phewa, Nepal (Thapa et al. 2014).

Table 1 Daily seafood consumption of Xiamen residents (g/day)

Species	Types	P_{25}	P_{50}	P_{75}
Fish		2.14	16.1	42.9
	Weever	2.14	6.43	17.1
	Nibea coibor	2.14	8.57	17.1
	Sea perch	4.29	6.43	17.1
	Sparuslatus	2.41	8.57	25.7
	Red porgy	1.79	6.43	17.1
	White pomfret	2.68	8.57	17.1
Mollusk		3.21	12.9	34.3
	Oyster	4.29	8.57	17.1
	Meretrix petechialis	3.21	8.57	17.1
	Ruditapes philippinarum	4.29	8.57	21.4
Crustacean		1.34	4.29	12.0
	Metapenaeus ensis	1.79	3.57	8.57
	Penaeus monodon	1.43	4.29	9.64
	Shrimp	2.50	3.57	8.57
	Portunus trituberculatus	2.50	2.50	10.0
	Crab	1.03	5.00	10.0
	Mud crab	5.36	2.50	5.00
Alga		2.14	9.29	19.3
	Porphyra tenera	2.14	4.29	8.57
	Laminaria japonica	2.14	4.29	12.9
Cephalopoda		2.14	4.29	10.7
	Squid	0.11	4.29	8.57
	Octopus	1.07	2.14	4.29
	Jellyfish	1.07	0.71	3.66
Drysaltery		1.07	3.21	8.57
	Dried squid	0.77	4.29	8.57
	Dried octopus	0.54	2.14	8.04
	Dried scallops	33.4	1.61	2.14
	Dried oyster	0.54	2.14	4.29
Total		2.14	61.5	115

Heavy metal concentrations

According to the intakes, 156 samples of 14 most consumed types of seafood were collected for heavy metal detection. Concentrations of Pb, Cd, Cr, Hg, and As were ND–0.45, ND–0.19, ND–0.80, ND–0.70, and 0.32–16.9 mg/kg, respectively (Table 2). Concentrations of Cd and As in some samples of sparuslatus, N. coibor, R. philippinarum, and P. tenera were higher than the safety limits of national standards (Table 3). Here, we assumed that methyl mercury (MeHg) and inorganic As (InAs) constituted 62 % of the total Hg and 10 % of the total As, respectively (Liao and Ling 2003; Wang et al. 2012; Ahmed et al. 2015; Lei et al. 2015).

The concentrations of Pb in fish, mollusk, crustacean, and alga samples were in the range of ND–0.40, ND–0.10, ND–0.04, and ND–0.45 mg/kg. The highest concentrations in fish were found in red porgy and white pomfret (0.40 mg/kg), which is a little higher than European Union limitation (0.3 mg/kg), while lower than China national standard (0.5 mg/kg, see Table 3). Xia et al. also reported lower Pb concentration (ND–0.10 mg/kg) in marine fish from coast of east China (Xia et al. 2013), which is similar to those reported in the species: cuttlefish (0.14 mg/kg), European hake (0.11 mg/kg), and common sole (0.09 mg/kg) (Storelli et al. 2012). A fairly lower Pb level was reported in canned mussels from the Canary Islands ranging from 0.006 to 0.008 mg/kg (Gutierrez et al. 2004). This study also indicated the lower concentration of Pb in crustacean (ND–0.04 mg/kg) or mollusk (ND–0.10 mg/kg) than the results reported by Figueira et al. (1.16–5.20 mg/kg) (Figueira et al. 2011) and Ahmed et al. (4.55±0.11 mg/kg) (Ahmed et al. 2015). According to the previous literatures, the Pb concentration in fish (30.7 mg/kg) from Lingdingyang, crustacean (12.0 mg/kg) from Guangxi, and mollusk (6.0 mg/kg) from Hong Kong was extremely high (Wang et al. 2013).

Cd, a highly toxic element, can be transported in the air (Ahmed et al. 2015). The range of Cd concentration in mollusk (ND–0.14 mg/kg), crustacean (ND–0.01 mg/kg), and alga (ND–0.18 mg/kg) was found to be low but a little higher in fish (ND–0.19 mg/kg). The concentration of Cd in some samples of sparuslatus (0.19 mg/kg) and N. coibor (0.15 mg/kg) was slightly higher than the safety limits of national standards (0.1 mg/kg, see Table 3) and Xia et al.'s report (ND–0.076 mg/kg) measured in fish from Chinese coast (Xia et al. 2013). Remarkable Cd level was found in cuttlefish (0.85 mg/kg) from Italy (Storelli et al. 2012) and fish (0.35 mg/kg) from Turkey (Tuzen 2009). The level of Cd contamination in fish can be largely affected by environmental pollution (Ahmed et al. 2015). The concentration of Cd in the present study suggested that the offshore of Xiamen may be slightly polluted by Cd, which may result from industrial process, some like electroplating. Shellfish can be high in Cd (WHO 1992). For example, the concentration of Cd in oyster samples from Fujian province, China, ranged between 0.44 and 4.01 mg/

Table 2 Concentrations of Pb, Cd, Cr, Hg, and As in seafood of Xiamen markets (mg/kg, w.w)

Species	Types (n)	Pb	Cd	Cr	Hg	As
Fish	Weever (22)	0.03–0.06	0.00–0.01	ND	ND	0.32–3.19
	Nibea coibor (22)	0.06–0.12	0.00–0.15	ND	ND	0.50–1.72
	Sea perch (22)	ND–0.01	ND	ND–0.07	ND	0.32–1.70
	Sparuslatus (22)	0.03–0.07	0.00–0.19	ND	ND	1.25–6.24
	Red porgy (20)	0.30–0.40	ND–0.01	ND	ND	0.78–3.24
	White pomfret (15)	0.30–0.40	ND	0.01–0.03	ND	0.41–3.15
Mollusk	Oyster (6)	0.01–0.06	0.10–0.14	ND–0.80	ND	1.10–1.60
	Meretrix petechialis (6)	0.02–0.10	0.04–0.05	ND	ND	0.50–0.80
	Ruditapes philippinarum (3)	ND	ND	ND	ND	1.80–2.00
Crustacean	*Metapenaeus ensis* (3)	0.01–0.04	ND	ND	ND	0.50–0.80
	Penaeus monodon (3)	ND	ND	ND–0.70	ND–0.70	0.70–0.90
	Shrimp (6)	ND	ND–0.01	ND–0.70	ND–0.70	0.50–0.80
Alga	*Porphyra tenera* (3)[a]	ND–0.10	ND–0.01	ND–0.50	0.01–0.02	9.84–16.9
	Laminaria japonica (3)	ND–0.45	0.06–0.18	0.40–0.66	0.01–0.10	2.10–4.45

ND not detected

[a] Means dry weight

kg (Liu et al. 2013b) which is much higher than the present findings (0.10–0.14 mg/kg).

The concentration of Cr in all fish samples ranged from ND to 0.07 mg/kg, which was extremely lower than that of the other heavy metals analyzed. Ahmed et al. observed the highest concentration of Cr in *Mastacembelus pancalus* (7.18 ± 1.38 mg/kg) among fish species (Ahmed et al. 2015). The Cr concentration in oyster was 0.80 mg/kg in the present study, which is similar to Liu et al.'s report of 0.08–0.98 mg/kg from oyster samples of Fujian province (Liu et al. 2013b), and much lower than Ahmed et al.'s detection of shellfish samples (16.05 ± 1.48 mg/kg) from Bangladesh (Ahmed et al. 2015).

Hg was not detected in the species of fish and mollusk and showed low concentrations in crustacean (ND–0.70 mg/kg) and alga (0.01–0.10 mg/kg). Similar findings have also been reported in other parts of China. For example, the concentration

Table 3 Limitation standards of Pb, Cd, Cr, Hg, and As in seafood (mg/kg)

Seafood	Pb	Cd	Cr	Hg	As	Quotation
Fish	0.5	0.1	2.0	0.5[a]	0.1[b]	GB2762-2012 (China)
	0.3	0.05	–	0.5	–	European Union
Mollusk	1.5	2.0	2.0	0.5[a]	0.5[b]	GB2762-2012 (China)
	1.5	1.0	–	0.5	–	European Union
Crustacean	0.5	0.5	2.0	0.5[a]	0.5[b]	GB2762-2012 (China)
	0.5	0.5	–	0.5	–	European Union
Alga	1.0	–	–	0.5[a]	1.5[b]	GB19643-2005 (China)
	–	3.0	–	–	–	European Union

[a] Means methyl mercury, MeHg

[b] Means inorganic As, InAs

of Hg in fish ranged from ND to 0.10 mg/kg in Guangxi province and from 0.01 to 0.04 mg/kg in Hong Kong; the samples of shellfish, shrimp, and mollusk ranged from 0.03 to 0.09, 0.02 to 0.04, and 0.04 to 0.11 mg/kg in Daya Bay, respectively (Wang et al. 2013). The concentration of Hg in shrimp detected in this study was the same as that from Fujian province in Liu et al.'s research (ND–0.70 mg/kg) (Liu et al. 2013b).

As exists in food in different chemical forms, and InAs is more toxic than organic As (Ahmed et al. 2015). Long-term or chronic exposure is thought to be linked to skin disease, hypertension, and cancer (Centeno et al. 2005). In the present study, the total As concentration was detected. The results showed a higher As concentration in alga (2.10–16.9 mg/kg) followed by in fish (0.32–6.24 mg/kg), which was lower than that in fish from Zhanjiang harbor (7.3 mg/kg) and Hainan province (0.1–16 mg/kg), China (Wang et al. 2013). While Falcó et al. (2006) and Martinez-Gómez et al. (2012) found As levels in red mullet were extremely high, ranging from 15.39 to 17.77 and from 16.57 to 49.68 mg/kg, respectively. Thus, according to the maximum As level permitted for fish samples which is 0.1 mg/kg (see Table 3), no level of As exposure is lower. Muluan Bay (Fig. 1) is a coastal lagoon in the northwest of Xiamen, which has been discharged complex effluents from Xinlin industrial complex over the past 30 years (Wang et al. 2014). Industrial processes such as smelting, tanning, and the addition of fertilizers increase the concentration of As in Maluan Bay that may lead to the high concentration of As in fish from Xiamen markets.

Risk assessment of dietary heavy metal exposure

The health risk for heavy metals in seafood is usually quantified by the target hazard quotient (THQ) (Burger and Gochfeld 2005;

Wang et al. 2005; Storelli 2008). The THQ is defined as the concentration of heavy metals divided by cancer slope factor (CSF) for carcinogen or reference dose (RfD) for non-carcinogen. If the THQ is less than one, it means safe during daily exposure (Yi et al. 2011). Conversely, the health risk should be considered. Based on the questionnaires, the data showed in Tables 1 and 2, and the CSF or RfDs of heavy metals, THQ values of heavy metals (Pb, Cd, Cr, MeHg, and InAs) from 14 types of seafood were calculated to estimate the potential carcinogenic and non-carcinogenic health risk for the P_{50} intake residents (Fig. 3).

The THQ values for the carcinogenic effects of the five metals from 14 types of seafood were 0.43–17.7 (Fig. 3a, b), which were calculated based on the CSF value of 1.5 (mg/kg/day)$^{-1}$ for InAs. However, a CSF value of 25.6 (mg/kg/day)$^{-1}$ published in a recent toxicological review of InAs was suggested for calculation (EPA/635/R-10/001, 2010) and has been used in some researches (Yu et al. 2014; Lei et al. 2015). If this

provisional CSF value was used in the calculation, the THQ values increased to 7.28–296 (Fig. 3c, d). Both current and provisional CSF values indicated that InAs was incorporated in the tested seafood. The results also indicated that the hazard for the carcinogenic effects of the five metals in multiple marine species was greater than one, which suggested that an extremely high cancer risk is associated with the consumption of multiple seafood species for local residents. Of all the seafood investigated, the consumption of *P. tenera* and sparuslatus had the highest potential carcinogenic health risk, which might be due to higher As concentration of 16.9 mg/kg (1.69 mg/kg for InAs) and 6.24 mg/kg (0.62 mg/kg for InAs), respectively, detected in these two types of seafood.

The THQ values for non-carcinogenic effects of five metals from 14 types of seafood were 0.01–0.41 (Fig. 3e, f), which were calculated based on the RfDs. All the THQ values were much lower than one, suggesting that common consumption
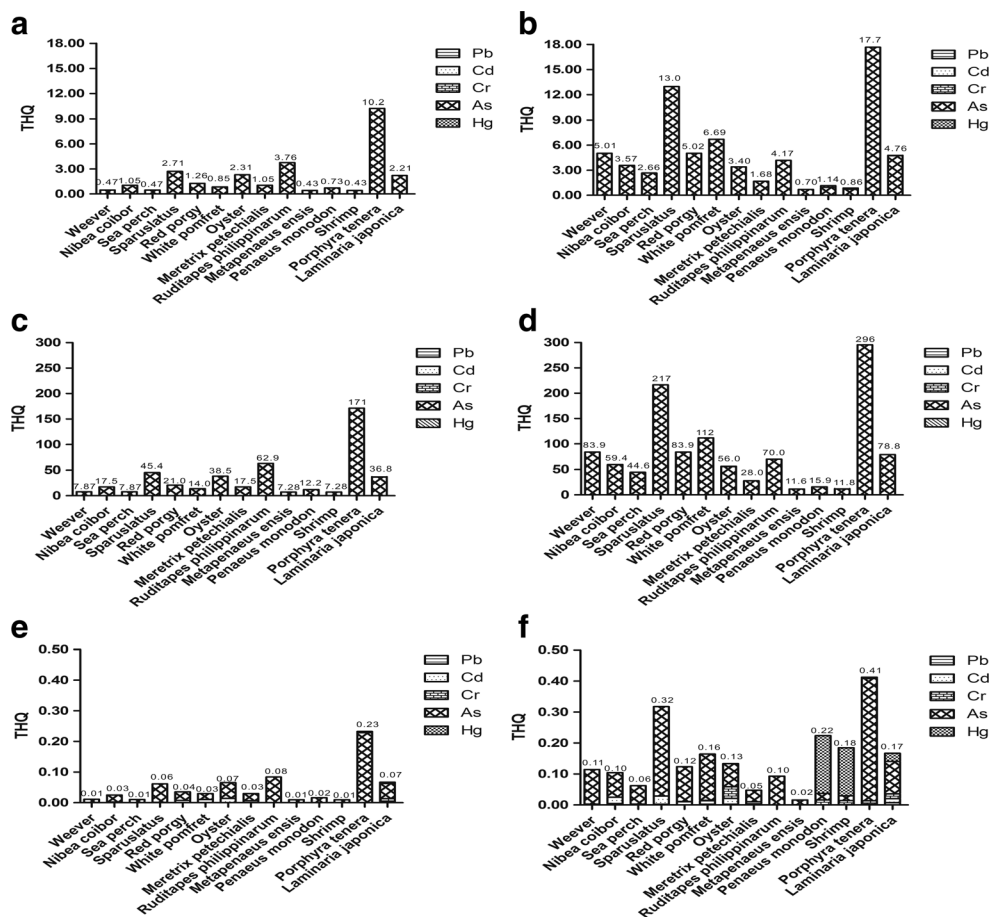

Fig. 3 The THQ values of heavy metals from 14 types of seafood for carcinogenic and non-carcinogenic effects in Xiamen residents. a THQ values for carcinogenic effect calculated from minimum concentrations of heavy metals, and the CSF value of 1.5 (mg/kg/day)$^{-1}$ for As was used. b THQ values for carcinogenic effects calculated from maximum concentrations of heavy metals, and the CSF value of 1.5 (mg/kg/day)$^{-1}$ for As was used. c THQ values for carcinogenic effect calculated from minimum concentrations of heavy metals, and the CSF value of 25.6 (mg/kg/day)$^{-1}$ for As was used. d THQ values for carcinogenic effect calculated from maximum concentrations of heavy metals, and the CSF value of 25.6 (mg/kg/day)$^{-1}$ for As was used. e THQ values for non-carcinogenic effect calculated from minimum concentrations of heavy metals. f THQ values for non-carcinogenic effect calculated from maximum concentrations of heavy metals

of the 14 types of seafood would not pose a non-carcinogenic risk. In terms of food safety, it is necessary to figure out which heavy metal in all the 14 types of seafood is the most harmful to human. Therefore, we calculated the relative contributions of the five metals to THQ. Among the metals, InAs is classified as group A—known human carcinogen, and the present results also showed the carcinogenic risk results from InAs contamination. And for the non-carcinogenic risk, the values of Pb, Cd, Cr, Hg, and As were 2.30, 4.15, 4.61, 17.05, and 71.89 %, respectively. Accounting for more than 70 % of the five heavy metals, As was the main component contributing to the potential non-carcinogenic risk. Some researchers also found that As was the contaminant causing the highest risk to humans, but they believed that Hg posed the highest non-carcinogenic risk (Yu et al. 2014; Lei et al. 2015).

The combined THQ and HI values for the carcinogenic and non-carcinogenic effects of the five metals from four species of seafood are shown in Fig. 4. The results indicated that InAs posed a very high cancer risk related to the four species of seafood intake when CSF was used in the calculation (Fig. 4a–d). For non-carcinogenic effect, the combined THQs

were 0.02–0.08 for Pb, 0.02–0.09 for Cd, 0.01–0.10 for Cr, 0.01–0.37 for MeHg, and 0.63–1.56 for InAs (Fig. 4e, f). Under these circumstances, the risk order of heavy metals can be listed as As > Hg > Cr > Cd > Pb. Significantly, the highest THQ value of samples polluted by InAs was 1.56, and the source was mainly from As bioaccumulation in fish, which was similar to Lei et al.'s research reporting that a relatively higher potential health risk might be due to higher As concentration in white pomfret collected from Shanghai (Lei et al. 2015). And a little different from Fang et al.'s research in Beijing (Fang et al. 2014) showing Cd and Pb played the most important roles of the combined THQ.

To take insight in the non-carcinogenic risk of Pb, Cd, Cr, MeHg, and InAs on health, the combined THQ and HI values for non-carcinogenic effect of five metals from four species of seafood are summarized in Table 4, which were calculated according to daily seafood consumption (IR, see Table 1), minimum, and maximum concentrations of the five metals (C_{max} and C_{min}, see Table 2). The combined THQ values for seafood were 0.17–0.88 for fish, 0.18–0.29 for mollusk, 0.04–0.42 for crustacean, and 0.30–0.61 for alga. From the results,
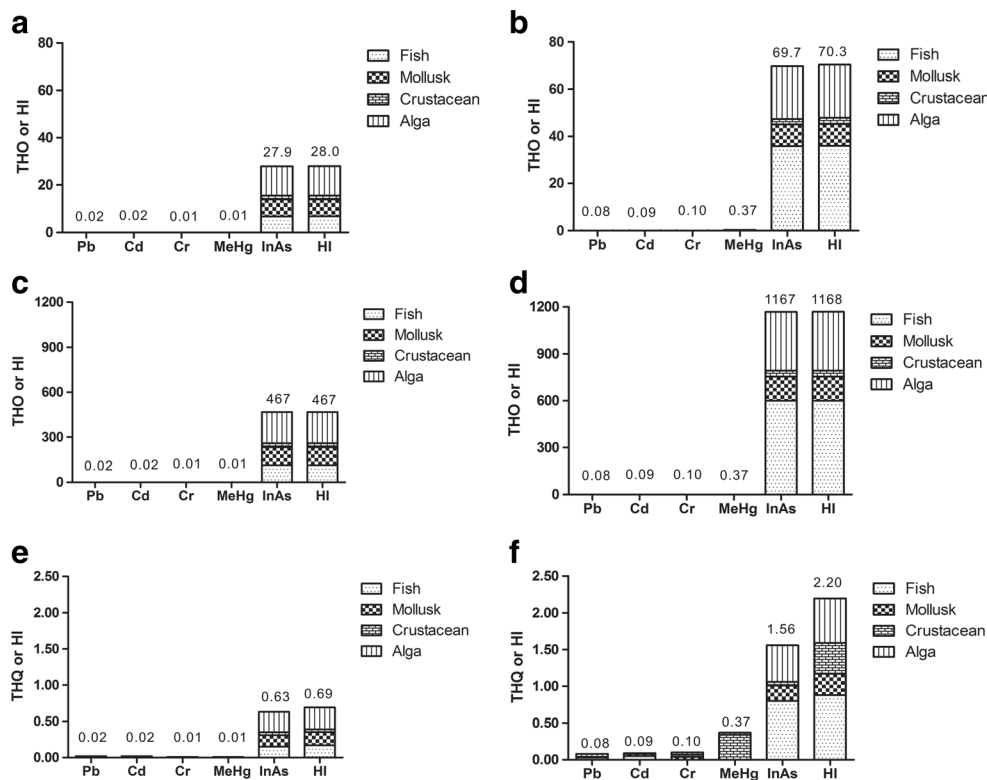

Fig. 4 The THQ and HI values of heavy metals from four species of seafood for carcinogenic and non-carcinogenic effects in Xiamen residents. **a** THQ and HI values for carcinogenic effect calculated from minimum concentrations of heavy metals, and the CSF value of 1.5 (mg/kg/day)$^{-1}$ for As was used. **b** THQ and HI values for carcinogenic effects calculated from maximum concentrations of heavy metals, and the CSF value of 1.5 (mg/kg/day)$^{-1}$ for As was used. **c** THQ and HI values for carcinogenic effect

calculated from minimum concentrations of heavy metals, and the CSF value of 25.6 (mg/kg/day)$^{-1}$ for As was used. **d** THQ and HI values for carcinogenic effect calculated from maximum concentrations of heavy metals, and the CSF value of 25.6 (mg/kg/day)$^{-1}$ for As was used. **e** THQ and HI values for non-carcinogenic effect calculated from minimum concentrations of heavy metals. **f** THQ and HI values for non-carcinogenic effect calculated from maximum concentrations of heavy metals

Table 4 THQ and HI values for non-carcinogenic effect of heavy metals from seafood in Xiamen

Metals	Assignment	RfD[a] (×10⁻³ mg/kg/day)	THQ				
			Fish	Mollusk	Crustacean	Alga	Total
Pb	IR: see Table 1	4.0	0.02–0.03	0–0.01	0	0–0.04	0.02–0.08
Cd	C_min and C_max:	1.0	0–0.05	0.02–0.03	0	0–0.01	0.02–0.09
Cr	see Table 2	3.0	0	0–0.04	0–0.03	0.01–0.03	0.01–0.10
MeHg	BW: 61.31 kg	0.1	0	0	0–0.34	0.01–0.03	0.01–0.37
In As		0.3	0.15–0.80	0.16–0.21	0.04–0.05	0.28–0.50	0.63–1.56
Combined THQ or HI			0.17–0.88	0.18–0.29	0.04–0.42	0.30–0.61	0.69–2.20

[a] RfDs of heavy metals refer to IRIS website of US EPA (http://www.epa.gov/iris/)

the single combined THQ for the four most frequently consumed seafood species was less than one, meaning that common consumption of each of these four seafood species was safe. While, the total combined THQ (HI) reached 0.69–2.20, indicating that the consumption of all seafood species together would pose a non-carcinogenic risk of the five heavy metals on ordinary residents. In particular, the THQ value of fish for InAs (0.15–0.80) was approaching one, which should be paid attention when consuming. InAs in fish (0.80) contributed to the most in HI values, followed by InAs in alga (0.50) and MeHg in crustacean (0.34). The higher THQ values of Hg were found in albacore (1.87) and rosefish (1.50) from Adriatic Sea (Storelli 2008). Compared with the data of Beijing (Fang et al. 2014), showing that the average THQ value of marine fish is 1.60, the slope of HI value of fish from Xiamen (0.17–0.88) is much lower. Thus, in the present study, the potentially non-carcinogenic risk seafood can be listed as fish > alga > crustacean > mollusk.

Conclusions

Heavy metals (Pb, Cd, Cr, Hg, and As) of seafood in markets are investigated in Xiamen city. The daily intake of the 24 types of seafood for Xiamen residents was 61.5 (2.14, 115) g/day with four most popular species: fish (16.07 (2.14, 42.9) g/day), mollusk (12.9 (3.21, 34.3) g/day), alga (9.29 (2.14, 19.3) g/day), and crustacean (4.29 (1.34, 12.0) g/day). The concentrations of the five metals were below the safety limits of Chinese national standards in most of the seafood samples. The highest As levels were found in sparuslatus (1.25–6.24 mg/kg), which exceeded the Chinese national standard limitation. The THQ was used to evaluate the health risks of the five heavy metals on humans, which suggested that the consumption of these four popular seafood species would not pose a non-carcinogenic risk. However, consumption of some types of seafood, such as sparuslatus, oyster, and *P. tenera*, would pose a carcinogenic risk, thus indicating that those

types of seafood should be limited to avoid potential harmful exposure to these heavy metals, especially As. Furthermore, HI values for non-carcinogenic effect of all metals in all seafood reached 0.69–2.20, indicating that it is necessary to pay attention to the intake of sorts of seafood and to keep a healthy and balanced diet.

Authors' contributions Ran Zhao and Chun Fan conceived and designed the experiments; Shuangshuang Yan, Wanting Xu, and Juan Wang performed heavy metals' detection; Min Liu and Bi Wang analyzed the data; Dong Hu and Dongbei Guo contributed for sampling and sample preparation; all authors participated in questionnaire survey; and Ran Zhao wrote the paper.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

Research involving human participants and/or animals Human participants were involved in the research, which was conducted according to the guidelines laid down in the Declaration of Helsinki, and all procedures involving human subjects were approved by the Medical and Health Research Ethics Committee of School of Public Health, Xiamen University.

Informed consent All participants provided written informed consent.

References

Agusa T, Kunito T, Sudaryanto A, Monirith I, Kan-Atireklap S, Iwata H, Ismail A, Sanguansin J, Muchtar M, Tana TS, Tanabe S (2007) Exposure assessment for trace elements from consumption of marine fish in Southeast Asia. Environ Pollut 145:766–777

Ahmed MK, Baki MA, Islam MS, Kundu GK, Habibullah-Al-Mamun M, Sarkar SK, Hossain MM (2015) Human health risk assessment of heavy metals in tropical fish and shellfish collected from the river Buriganga, Bangladesh. Environ Sci Pollut Res Int 22:15880–15890

Burger J, Gochfeld M (2005) Heavy metals in commercial fish in New Jersey. Environ Res 99:403–412

Centeno JA, Gray MA, Mullick FG, Tchounwou PB, Tseng CH (2005) Metal Contaminants in New Zealand: sources, treatments, and effects on ecology and human health. Resolution Press, New Zealand

Deng B, Zhang J, Zhang GR, Zhou JZ (2010) Enhanced anthropogenic heavy metal dispersal from tidal disturbance in the Jiaozhou Bay, North China. Environ Monit Assess 161:349–358

Ding ZH, Liu JL, Li LQ, Lin HN, Wu H, Hu ZZ (2009) Distribution and speciation of mercury in surficial sediments from main mangrove wetlands in China. Mar Pollut Bull 58:1319–1325

Dorea JG (2008) Persistent, bioaccumulative and toxic substances in fish: human health considerations. Sci Total Environ 400:93–114

U.S. EPA. IRIS Toxicological Review of Inorganic Arsenic (Cancer) (2010 External Review Draft). U.S. Environmental Protection Agency, Washington, DC, EPA/635/R-10/001, 2010

Falco G, Llobet JM, Bocio A, Domingo JL (2006) Daily intake of arsenic, cadmium, mercury, and lead by consumption of edible marine species. J Agric Food Chem 54:6106–6112

Fang TH, Li JY, Feng HM, Chen HY (2009) Distribution and contamination of trace metals in surface sediments of the East China Sea. Mar Environ Res 68:178–187

Fang YY, Nie ZQ, Liu F, Die QQ, He J, Huang QF (2014) Concentration and health risk evaluation of heavy metals in market-sold vegetables and fishes based on questionnaires in Beijing, China. Environ Sci Pollut Res Int 21:11401–11408

Figueira E, Lima A, Branco D, Quintino V, Rodrigues AM, Freitas R (2011) Health concerns of consuming cockles (Cerastoderma edule L.) from a low contaminated coastal system. Environ Int 37:965–972

Gutierrez A, Lozano G, Hardisson A, Rubio C, Gonzalez T (2004) Content of toxic and essential metals in canned mussels commonly consumed in Tenerife, Canary Islands, Spain. J Food Prot 67:1526–1532

Kim NS, Lee BK (2010) Blood total mercury and fish consumption in the Korean general population in KNHANES III, 2005. Sci Total Environ 408:4841–4847

Lei BL, Zhang KQ, An J, Zhang XY, Yu YX (2015) Human health risk assessment of multiple contaminants due to consumption of animal-based foods available in the markets of Shanghai, China. Environ Sci Pollut Res Int 22:4434–4446

Liao CM, Ling MP (2003) Assessment of human health risks for arsenic bioaccumulation in tilapia (Oreochromis mossambicus) and large-scale mullet (Liza macrolepis) from blackfoot disease area in Taiwan. Arch Environ Contam Toxicol 45:264–272

Liu JL, Xiang rong XU, Ding ZH, Jin MH, Wang SL, Hao Q (2013a) Heavy metals contamination in fish from coral reef ecosystem and ecology risk evaluation. Mar Environ Sci 32:262–266 (in Chinese)

Liu QM, Lin JQ, Zhang YP, Cao YL, Yao YX, Qiong WU, Mai JJ (2013b) Contents of heavy metals and distribution characteristics in different tissues and organs of oyster from Fujian coastal waters. Mar Environ Sci 32:343–1172 (in Chinese)

Martinez-Gomez C, Fernandez B, Benedicto J, Valdes J, Campillo JA, Leon VM, Vethaak AD (2012) Health status of red mullets from polluted areas of the Spanish Mediterranean coast, with special reference to Portman (SE Spain). Mar Environ Res 77:50–59

Martorell I, Perello G, Marti-Cid R, Llobet JM, Castell V, Domingo JL (2011) Human exposure to arsenic, cadmium, mercury, and lead from foods in Catalonia, Spain: temporal trend. Biol Trace Elem Res 142:309–322

Pan K, Wang WX (2012) Trace metal contamination in estuarine and coastal environments in China. Sci Total Environ 421:3–16

Pieniak Z, Verbeke W, Scholderer J (2010) Health-related beliefs and consumer knowledge as determinants of fish consumption. J Hum Nutr Diet 23:480–488

Rainbow PS, Luoma SN (2011) Metal toxicity, uptake and bioaccumulation in aquatic invertebrates—modelling zinc in crustaceans. Aquat Toxicol 105:455–465

Sarmiento AM, DelValls A, Nieto JM, Salamanca MJ, Caraballo MA (2011) Toxicity and potential risk assessment of a river polluted by acid mine drainage in the Iberian Pyrite Belt (SW Spain). Sci Total Environ 409:4763–4771

Song DP, Zhuang DF, Jiang D, Fu JY, Wang Q (2015) Integrated health risk assessment of heavy metals in Suxian County, South China. Int J Environ Res Public Health 12:7100–7117

Storelli MM (2008) Potential human health risks from metals (Hg, Cd, and Pb) and polychlorinated biphenyls (PCBs) via seafood consumption: estimation of target hazard quotients (THQs) and toxic equivalents (TEQs). Food Chem Toxicol 46:2782–2788

Storelli MM, Normanno G, Barone G, Dambrosio A, Errico L, Garofalo R, Giacominelli-Stuffler R (2012) Toxic metals (Hg, Cd, and Pb) in fishery products imported into Italy: suitability for human consumption. J Food Prot 75:189–194

Tadiso TM, Borgstrom R, Rosseland BO (2011) Mercury concentrations are low in commercial fish species of Lake Ziway, Ethiopia, but stable isotope data indicated biomagnification. Ecotoxicol Environ Saf 74:953–959

Thapa DS, Sharma CM, Kang S, Sillanpaa M (2014) The risk of mercury exposure to the people consuming fish from Lake Phewa, Nepal. Int J Environ Res Public Health 11:6771–6779

Tuzen M (2009) Toxic and essential trace elemental contents in fish species from the Black Sea, Turkey. Food Chem Toxicol 47:1785–1790

USEPA (2011) USEPA regional screening level (RSL) summary table. Available at: http://www.epa.gov/regshwmd/risk/human/Index.htm. Accessed 20 Jan 2014.

USEPA (United States Environmental Protection Agency) (2000) Guidance for assessing chemical contaminant data for use in fish advisories, EPA 823-B-00-008, vol 2, 3rd edn. Risk assessment and fish consumption limits. Office of Water, Washington, <http://water.epa.gov/citech/swguidance/fishshellfish/techguidance/risk/volume2index.cfm>

Wan L, Wang NB, Li QB, Sun B, Zhou ZC, Xue K, Ma ZQ, Tian J, Song L (2008) Distribution of dissolved metals in seawater of Jinzhou Bay, China. Environ Toxicol Chem 27:43–48

Wang XL, Sato T, Xing BS, Tao S (2005) Health risks of heavy metals to the general public in Tianjin, China via consumption of vegetables and fish. Sci Total Environ 350:28–37

Wang SF, Li B, Zhang MM, Xing DH, Jia YF, Wei CY (2012) Bioaccumulation and trophic transfer of mercury in a food web from a large, shallow, hypereutrophic lake (Lake Taihu) in China. Environ Sci Pollut Res Int 19:2820–2831

Wang SL, Xu XR, Sun YX, Liu JL, Li HB (2013) Heavy metal pollution in coastal areas of South China: a review. Mar Pollut Bull 76:7–15

Wang ZS, Dong XX, Zhou SL, Yan CZ, Yan YJ, Chi QQ (2014) Contamination assessments of surface water in coastal lagoon (Maluan Bay, China) incorporating biomarker responses and bioaccumulation in hepatopancreas of exposed shrimp (Litopenaeus vannamei)—an integrative approach. Environ Sci Pollut Res Int 21:205–219

WHO (1992) Cadmium environmental health criteria no. 134. World Health Organisation, Geneva

Woo J, Leung SSF, Ho SC, Lam TH, Janus ED (1997) A food frequency questionnaire for use in the Chinese population in Hong Kong: description and examination of validity. Nutr Res 17:1633–1641

Xia CH, Wu XG, Lam JCW, Xie ZQ, Lam PKS (2013) Methylmercury and trace elements in the marine fish from coasts of East China. J Environ Sci Health A Tox Hazard Subst Environ Eng 48:1491–1501

Yi YJ, Yang ZF, Zhang SH (2011) Ecological risk assessment of heavy metals in sediment and human health risk assessment of heavy metals in fishes in the middle and lower reaches of the Yangtze River basin. Environ Pollut 159:2575–2585

Yu R, Woo J, Chan R, Sham A, Ho S, Tso A, Cheung B, Lam TH, Lam K (2011) Relationship between dietary intake and the development of

type 2 diabetes in a Chinese population: the Hong Kong Dietary Survey. Public Health Nutr 14:1133–1141

Yu YX, Chen L, Yang D, Pang YP, Zhang SH, Zhang XY, Yu ZQ, Wu MH, Fu JM (2012) Polycyclic aromatic hydrocarbons in animal-based foods from Shanghai: bioaccessibility and dietary exposure. Food Addit Contam Part A Chem Anal Control Expo Risk Assess 29:1465–1474

Yu YX, Wang XX, Yang D, Lei BL, Zhang XL, Zhang XY (2014) Evaluation of human health risks posed by carcinogenic and non-carcinogenic multiple contaminants associated with consumption of fish from Taihu Lake, China. Food Chem Toxicol 69:86–93

Zhang W, Wang WX (2012) Large-scale spatial and interspecies differences in trace elements and stable isotopes in marine wild fish from Chinese waters. J Hazard Mater 215:65–74