Ecotoxicology and Environmental Safety 73 (2010) 1812–1819

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

journal homepage: www.elsevier.com/locate/ecoenv

Dietary intake and potential health risk of DDTs and PBDEs via seafood consumption in South China

Jianyang Guo [a,b], Fengchang Wu [b], Rulang Shen [a], Eddy Y. Zeng [a,*]

[a] State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China
[b] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China

ARTICLE INFO

Article history:
Received 13 November 2009
Received in revised form
7 August 2010
Accepted 9 August 2010

Keywords:
Dietary intake
Risk assessment
Polybrominated diphenyl ethers
Dichlorodiphenyltrichloroethane
Seafood
South China

ABSTRACT

A total of 602 seafood samples, including fish, shrimps, crabs and molluscs, were analyzed for a suite of persistent halogenated compounds. The residual levels of DDTs (sum of *o,p′*- and *p,p′*-DDT, DDD, and DDE) and polybrominated diphenyl ethers (PBDEs) varied significantly among different species, and ranged from non-detectable (nd) to 699 ng/g and nd to 5.93 ng/g, respectively. Comparison of the levels of DDTs and PBDEs in mussel samples worldwide suggested that South China is probably one of the most DDT-polluted areas, but is moderate at most in terms of PBDE contamination. Combined with a recent dietary survey at the same sampling locations, dietary intakes of DDTs and PBDEs by local residents via seafood consumption for all age groups were estimated to be 147–564 and 4.7–18.5 ng/day, or 8.5 – 12.9 and 0.27 – 0.46 ng/kg bw/day, respectively. Among the different seafood types, fish contributed the largest portion of the dietary intakes of DDTs (57%), followed by molluscs (38%). Similarly, the dietary intakes of PBDEs were also dominated by fish (45%) and molluscs (45%). Assessment based on several available guidelines suggested that though no significant human health risk associated with the dietary intake of PBDEs, a lifetime cancer risk from dietary exposure to DDTs remains a probability. Because dietary intake of DDTs was dominated by fish and molluscs, added concern should be paid to fish and molluscs.

1. Introduction

The notorious persistent halogenated compounds (PHCs), such as organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs), are widely distributed in various environmental compartments. The lipophilic and persistent properties are probably the main causes for these compounds to be bioaccumulated through the food chain, and the toxicity of PCBs and OCPs has been documented (Smith and Gangolli, 2002). Although no toxic effects of PBDEs to humans have been convincingly confirmed, particularly at the levels often found in the environment, certain low-brominated congeners at high levels are likely to induce toxicity, including endocrine disruption (Hallgren and Darnerud, 2002) and neurological deficiency (Branchi et al., 2003).

Generally, PHCs are transferred to humans mainly via inhalation, dermal absorption, and food consumption (Bayarri et al., 2001; Sjodin et al., 2003). In most cases, dietary intake is the main route of PHCs. For example, more than 90% of the average human intake of dioxins and PCBs originates from foods, especially those of animal origin (Liem et al., 2000). In the case of PBDEs, however, inhalation may be another important route for human exposure in addition to dietary intake, especially for occupational exposure (Sjödin et al., 2001; Harrad et al., 2004; Meng et al., 2007b).

The Pearl River Delta (PRD), located in a subtropical zone of South China, is one of the economically most prosperous regions in China. Because of the mild climatic conditions all year around and rich aquatic resources, aquaculture has become an increasingly popular economic activity in the PRD. On the other hand, dichlorodiphenyltrichloroethane (DDT) has been extensively used in the past (Guo et al., 2009), and PBDEs, as an extensive used brominated flame retardants, have become ubiquitous environmental contaminants due to intensifying manufacturing and e-waste recycling activities in the PRD (Guan et al., 2007; Ni and Zeng, 2009). As a result, seafood produced in this region has possibly been subject to heavy PHCs contamination, and consumption of these products could be an important route for human exposure to PHCs. Because a large quantity of seafood products are produced in and exported from the PRD region, dietary intake of PHCs via seafood consumption should be a concern not only in South China, but also throughout the globe.

The objectives of the present study were to examine the dietary intake of DDTs (sum of *o,p′*- and *p,p′*-DDT, DDD, and DDE) and PBDEs and to assess health risk related to these PHCs via

* Corresponding author. Fax: +86 20 85291421.
E-mail address: eddyzeng@gig.ac.cn (E.Y. Zeng).

0147-6513/$ - see front matter © 2010 Elsevier Inc. All rights reserved.
doi:10.1016/j.ecoenv.2010.08.009

seafood consumption, which could provide critical information for the local governments to establish seafood consumption guidelines.

2. Materials and methods

2.1. Sample collection

Seafood samples were collected based on a market basket-based survey. A total of 602 samples, representing thirteen species of fish, six species of shrimps, two species of crabs, and fourteen species of molluscs that are commonly consumed by local residents, were randomly collected from local fishery markets in 11 coastal cities of Guangdong Province, South China, between November 2004 and October 2005 (Fig. 1). Upon collection, samples were stored in polyethylene bags, kept in ice, and brought back to the laboratory immediately. They were stored at −20 °C until analyzed. Because all seafood samples were purchased dead, no ethical issues were involved in the present study.

2.2. Analytical methods and instrumentation

The methodological details, including extraction, cleanup, fractionation, and the instrumental analysis have been described in our previous publications (Guo et al., 2007a, 2007b; Meng et al., 2007a). Briefly, approximately 20 g (wet weight) of muscle or soft tissue were taken, homogenized, freeze-dried, and ground into fine powders. After spiked with the surrogate standards of 2,4,5,6-tetrachloro-*m*-xylene (TMX), PCB 67, PCB 191, PCB 209 and ^{13}C-PCB 141, each sample was Soxhlet extracted with an acetone and hexane mixture (1:1 in volume) for 48 h. The extract was first subject to a gel permeation chromatograph for lipid removal, then subject to silica/alumina columns for purification. For DDTs, an aliquot (2 mL) of the extract was purified on a silica/alumina column packed, from the bottom to top, with neutral alumina (6 cm, 3% deactivated) and neutral silica gel (12 cm, 3% deactivated). The first fraction was eluted with 5 mL of hexane and discarded. The second fraction was eluted with 70 mL of dichloromethane and hexane mixture (3:7 in volume) and collected. For PBDEs, the remaining 2 mL of the extract was cleaned on a silica/alumina column packed, from bottom to top, with neutral alumina (6 cm, 3% deactivated), neutral silica gel (2 cm, 3% deactivated), 25% sodium hydroxide silica (5 cm), neutral silica gel (2 cm, 3% deactivated) and 50% sulfuric acid silica (6 cm). The PBDE fraction was eluted with 70 mL of dichloromethane and hexane mixture (1:1 in volume). Both extracts were further concentrated to 100 μL under a gentle stream of N_2. Finally, the internal standards (PCB 82 for DDTs and ^{13}C-PCB 208 for PBDEs) were added before instrumental analysis.

Analyses of DDTs were carried out on a Hewlett-Packard 5890 gas chromatograph (GC)/5972 mass spectrometer (MS) in the selective ion monitoring (SIM) mode. A fused-silica capillary column (DB-5, 30 m × 0.25 mm × 0.25 μm) was used for separation. High-purity helium was used as carrier gas at a constant pressure of 10.0 psi. Samples (1.0 μL each) were injected in the splitless injection mode. The temperatures of the injector and ion source were 280 and 250 °C, respectively. The oven temperature was programmed from 80 °C (held for 1.0 min), raised to 200 °C at 12 °C/min, increased to 220 °C at 1 °C/min, and finally ramped to 290 °C at 15 °C/min (held at 290 °C for 5 min).

For PBDEs, analyses were performed with a Shimadzu Model 2010 GC coupled with a Model QP 2010 MS (Shimadzu, Japan) using the negative chemical ionization and the selective ion monitoring mode. GC columns used for separation were a DB-XLB (30 m × 0.25 mm × 0.25 μm) capillary column for low-brominated congeners (BDE 28 to BDE 183) and a CP-Sil 13 CB (12.5 m × 0.25 mm × 0.2 μm) capillary column for BDE 209. Manual injection was conducted in the splitless mode with a split time of 1.0 min. The temperatures of the ion source and interface were 200 and 280 °C, respectively. Ion fragments used for monitoring were m/z 79 and 81 ($[Br]^-$) for tri- to hepta-BDEs, m/z 79, 81, 486.7, and 488.7 for the highly brominated BDE, m/z 372, 374, and 376 for ^{13}C-PCB 141, m/z 496, 498, and 500 for PCB 209, and m/z 474, 476, and 478 for the internal standard (^{13}C-PCB 208).

2.3. Dietary survey

A questionnaire-based dietary survey was conducted in the same area from where seafood samples were collected. Dietary data were collected through face-to-face interviews with 1626 local residents, and 12 food categories (fish, shrimp, crab, molluscs, meat, egg, vegetable, bean, milk, fruit, rice, and wheat products) were included. In addition, information on the age and gender of each interviewee was also collected. Statistical analyses were conducted using Sybase SQL Server 1.0 (Dublin, CA, USA). In the present study, only the food groups of fish, shrimps, crabs, and molluscs were used for further discussions (Table 1). The selection of these food groups was based on previous studies demonstrating that the major proportion of total dietary intake of PHCs was originated from consumption of fish (Kiviranta et al., 2004; Darnerud et al., 2006).

2.4. Dietary exposure assessment

Dietary exposure was estimated based on the results from our dietary survey conducted in April 2006 and the data analyses from the present study. All concentrations were not surrogate recovery corrected and normalized to wet sample weights except where specified. The reporting limits (RLs) (defined as the lowest concentrations of the calibration curves divided by the actual sample weights) were 0.02–0.10 ng/g for DDTs, 0.002–0.005 ng/g for all tri- to hepta-BDEs, and 0.1 ng/g for BDE 209. For samples with an analyte concentration below the RL, half of the RL value was used for the estimation. The estimated daily intake (EDI) was calculated as: EDI (ng/g bw/day) = seafood consumption (g/kg bw/day) × contaminant concentration (ng/g). In the present study, the average body weights of different age groups were derived from recent surveys (Yang et al., 2005; Zhang et al., 2008).

Fig. 1. Map of the sampling sites.

Table 1
Dietary survey in coastal cities of Guangdong Province, Southern China (g/day)[a].

	Age					
	2 – 5		6 – 18		> 18	
Gender	Male	Female	Male	Female	Male	Female
Fish	**16.0**	**15.3**	**37.9**	**36.3**	**63.6**	**55.0**
Shrimps	**3.1**	**1.9**	**10.0**	**6.2**	**6.2**	**5.9**
Crabs	**1.3**	**0.8**	**4.1**	**2.5**	**2.5**	**2.3**
Molluscs	**12.3**	**7.4**	**39.6**	**24.5**	**24.5**	**23.2**
Meat	50.4	81.7	135	94.1	145	117
Egg	37.5	62.8	58.2	50.5	38.0	37.8
Vegetable	163	110	132	176	235	239
Beans	91.9	60.7	63.7	56.0	53.2	52.3
Milk	119	150	128	102	70.6	84.9
Fruit	93.8	75.7	127	109	94.4	116
Rice	87.5	110	242	194	256	198
Wheat products	2.63	39.4	87.6	61.4	69.9	47.1

[a] Only food groups in bold letters were used in the present study.

3. Results and discussion

3.1. Selection of target analytes

Persistent halogenated compounds include a suite of compounds such as OCPs, PCBs, and PBDEs, as well as more toxic compounds dioxins, etc. In the present study, edible parts of the 602 seafood samples were analyzed for OCPs, PCBs, and PBDEs, but not for dioxins. The results indicated that PHCs in seafood products from South China were dominated by DDTs (sum of *o,p′*- and *p,p′*-DDT, DDD and DDE) and to a less extent by PBDEs (sum of BDE 28, 47, 66, 99, 100, 138, 153, 154, 183, and 209). The concentrations of hexachlorocyclohexanes (HCHs) were far below those of DDTs. The results were in accordance with the well-documented findings that the residual levels of HCHs in foodstuff in China declined remarkably from 1978 to 2001 (Nakata et al., 2002) and that low concentrations of HCHs were found in aquatic samples from the coastal region of North and East China (Jiang et al., 2005; Yang et al., 2006). Other OCP compounds, such as heptachlor, heptachlor epoxide, aldrin, dieldrin, endosulfan (I and II), endosulfan sulfate, endrin, endrin aldehyde, endrin ketone, and methoxychlor, were rarely detected and/or their concentrations were rather low when detectable. Historically, the production and usage of PCBs were relatively insignificant in China compared to those of DDTs and HCHs (Mai et al., 2005), which is well reflected in the previous studies conducted in the coastal region of North and East China (Jiang et al., 2005; Yang et al., 2006). With all these considerations, only the DDT and PBDE data are further utilized in the remainder of the paper.

3.2. DDTs and PBDEs in seafood from South China

Levels of DDTs and PBDEs in seafood from South China are summarized in Table 2. The residual levels of DDTs were highly variable among different species, and ranged from non-detectable (nd) to 699, nd to 52.5, nd to 56.3, and nd to 620 ng/g in fish, shrimps, crabs, and molluscs, respectively, suggesting that bioaccumulation of DDTs in seafood products was highly species-specific, probably due to their different ecological characteristics such as feeding habits and habitats. Like DDTs, PBDEs in seafood also varied in abundance with different species (Table 2), with concentration ranges of nd–5.93, nd–1.11, nd–1.55, and nd–2.20 ng/g in fish, shrimps, crabs, and molluscs, respectively. It has been a challenge to conduct a comparison of contamination levels among different countries because bioaccumulation in different organisms may vary significantly. The inclusion of mussel, which is an excellent bioindicator of trace toxic contaminants, in the present study allowed us to compare the results from the present study to those from other investigations around the world. The residual levels of DDTs in mussel from South China (3.95–507 ng/g with a mean of 65.7 ng/g) were higher than those from most other locations worldwide (Table 3), except for Egypt where DDT has still been used in agriculture and vector control (Khaled et al., 2004), indicating that South China is probably one of the most DDT-polluted areas in the world.

It should be noted that comparison of PBDE levels in mussel is somewhat compromised, because the number of BDE congeners varied greatly among different studies, probably reflective of the variability in local PBDE sources. Therefore, only BDE 47 and BDE 99, the most frequently analyzed congeners, were used for comparison (Table 4). Contrary to those of DDTs, the levels of BDE 47 and BDE 99 in mussel from the present study were lower than those obtained by most previous studies. Some published data were normalized to dry or lipid sample weight. If converted into wet sample weight assuming a moisture content of 80% and lipid content of 2%, these concentrations were also much higher

Table 2
Concentrations (ng/g) of DDTs and PBDEs in seafood products from South China.

	DDTs[a]			PBDEs[b]		
	Range	Average	Median	Range	Average	Median
Fish[c]	nd–699	32.2	5.99	nd–5.93	0.24	0.15
Shrimps[d]	nd–52.2	5.77	0.91	nd–1.11	0.24	0.11
Crabs[e]	nd–56.3	12.8	5.51	nd–1.55	0.44	0.32
Molluscs[f]	nd–619.7	42.3	6.66	nd–2.20	0.41	0.26

[a] Sum of BDE 28, 47, 66, 99, 100, 138, 153, 154, 183, and 209.
[b] Sum of *o,p′*- and *p,p′*-DDT, DDD, and DDE.
[c] Including *Tilapia*, *Ctenopharyngodon idellus*, *Aristichthys nobilis*, *Megalobrama amblycephala*, *Micropterus salmoides*, *Siniperca chuatsi*, *Ophicephalus argus*, *Trichiurus lepturu*, *Nemipterus virgatus*, *Mugil cephalus*, *Sciaenops ocellatus*, *Trachinotus blochii*, and *Lutjanus erythopterus*.
[d] Including *Penaeus monodon*, *P. japonicus*, *Metapenaeus ensis*, *Macrobrachium rosenbergii*, *Proambarus clarkia*, and *Squilla oraloria*.
[e] Including *Scylla serrata* and *Ovalipes punctatus*.
[f] Including *Tegillarca granosa*, *Scapharca subcrenata*, *Argopectens irradias*, *Patinopecten yessoensis*, *Haliotis diversicolor*, *Terebra maculate*, *Meretrix meretrix*, *Cyclina sinensis*, *Venerupis variegate*, *Sinonovacula constricta*, *Solen grandis*, *Perna uiridis*, and *Crassostrea gigas*.

Table 3
Comparison of DDT concentrations (ng/g) in mussels obtained from the present study with those reported from different countries and regions.

Location	Date	Concentration	Reference
Japan	1994	3.5 (0.80–12)[a]	Monirith et al. (2003)
Greenland	1994–1995	0.39 [a]	Cleemann et al. (2000)
Thailand	1994–1995	1.2–38 [a]	Tanabe et al. (2000)
Demark	–	2.4–67 [a]	Granby and Spliid (1995)
Brazil	1996	1.1–10 [a]	de Brito and Bruning (2002)
Vietnam	1997	40 (2.4–310) [a]	Monirith et al. (2003)
Italy	1997	14–64 [b]	Binelli et al. (2001)
England	1998	0.20–17 [a]	Connor et al. (2001)
India	1998	4.2 (0.6–15) [a]	Monirith et al. (2003)
Indonesia	1998	1.0 (0.10–3.1) [a]	Monirith et al. (2003)
Hong Kong	1998–1999	120 (7.0–1000) [a]	Monirith et al. (2003)
Egypt	2000	125–772 [a]	Khaled et al. (2004)
Bohai Sea	2002	4.46–129 [c]	Yang et al. (2004)
South China	2005	65.7 (3.95–507) [b]	Present study

[a] Including *p,p′*-DDT, DDE and DDD.
[b] Including *o,p′*- and *p,p′*-DDT, DDE, DDD.
[c] Including *p,p′*-DDT, DDE, DDD and *o,p′*-DDT.

Table 4
Comparison of PBDE concentrations (ng/g) in mussels obtained from the present study with those reported from different countries and regions.

Location	Sampling time	BDE 47	BDE 99	Reference
U.K. [a]	1996	3.5	3.9	Allchin et al. (1999)
Denmark [a]	2000	0.05–0.49	0.02–0.25	Christensen and Platz (2001)
France [a]	2001–2002	0.11–1.49	0.02–0.62	Johansson et al. (2006)
U.S.A. [a]	2002	nd–3.7	nd–2.0	Oros et al. (2005)
Norway [a]	2003	0.03–1.2	0.01–0.07	Bethune et al. (2004)
Korea [a]	2005	0.02–3.3	0.01–4.4	Ramu et al. (2007)
Netherlands [b]	1999	0.9–4.3	0.3–1.6	de Boer et al. (2003)
Singapore [b]	2002	0.72–11	0.62–18	Bayen et al. (2003)
Hong Kong [b]	2004	0.62–9.12	1.34–25.9	Liu et al. (2005)
Italy [c]	2005	23.1–309.5	5.0–69.5	Binelli et al. (2008)
South China [a]	2005	nd–0.063	nd–0.059	Present study

[a] On a wet weight basis.
[b] On a dry weight basis.
[c] On a lipid weight basis.

Table 5
Estimated daily intake (ng/day) and body weight normalized estimated daily intake (in parentheses, ng/kg bw/day) of PBDEs and DDTs via seafood consumption.

	Age					
	2–5		6–18		> 18	
Gender	Male	Female	Male	Female	Male	Female
PBDEs[a]						
Fish[b]	2.5 (0.14)	2.4 (0.15)	5.8 (0.14)	5.6 (0.14)	9.8 (0.15)	8.5 (0.15)
Shrimps[c]	0.3 (0.02)	0.2 (0.01)	1.1 (0.03)	0.7 (0.02)	0.7 (0.01)	0.6 (0.01)
Crabs[d]	0.4 (0.02)	0.3 (0.02)	1.3 (0.03)	0.8 (0.02)	0.8 (0.01)	0.7 (0.01)
Molluscs[e]	3.2 (0.18)	1.9 (0.12)	10.3 (0.25)	6.4 (0.16)	6.4 (0.10)	6.0 (0.11)
Total	**6.4 (0.36)**	**4.7 (0.30)**	**18.5 (0.46)**	**13.4 (0.33)**	**17.6 (0.27)**	**15.9 (0.28)**
DDTs[f]						
Fish[b]	95.8 (5.38)	91.6 (5.73)	227 (5.61)	217 (5.37)	381 (5.74)	330 (5.84)
Shrimps[c]	2.8 (0.16)	1.7 (0.11)	9.1 (0.22)	5.6 (0.14)	5.6 (0.08)	5.4 (0.10)
Crabs[d]	7.2 (0.40)	4.4 (0.28)	22.6 (0.56)	13.8 (0.34)	13.8 (0.21)	12.7 (0.22)
Molluscs[e]	81.9 (4.60)	49.3 (3.08)	264 (6.51)	163 (4.03)	163 (2.46)	155 (2.74)
Total	**188 (10.5)**	**147 (9.2)**	**522 (12.9)**	**400 (9.9)**	**564 (8.5)**	**502 (8.9)**

[a] Sum of BDE 28, 47, 66, 99, 100, 138, 153, 154, 183 and 209.
[b] Including *Tilapia*, *C. idellus*, *A. nobilis*, *M. amblycephala*, *M. salmoides*, *S. chuatsi*, *O. argus*, *T. lepturu*, *N. virgatus*, *M. cephalus*, *S. ocellatus*, *T. blochii*, and *L. erythopterus*.
[c] Including *P. monodon*, *P. japonicus*, *M. ensis*, *M. rosenbergii*, *P. clarkia*, and *S. oraloria*.
[d] Including *S. serrata* and *O. punctatus*.
[e] Including *T. granosa*, *S. subcrenata*, *A. irradias*, *P. yessoensis*, *H. diversicolor*, *T. maculate*, *M. meretrix*, *C. sinensis*, *V. variegate*, *S. constricta*, *S. grandis*, *P. uiridis*, and *C. gigas*.
[f] Sum of *o,p′*- and *p,p′*-DDT, DDD, and DDE.

than those from the present study. This indicates that the magnitude of PBDE contamination in South China is moderate globally.

3.3. Estimated daily intake

Because the residual levels of DDTs and PBDEs were highly variable among individual samples, the median concentrations instead of mean concentrations in each seafood group (i.e. fish, shrimps, crabs, and molluscs) were chosen for assessment. The EDIs of DDTs and PBDEs (both non-body and body weight normalized) via seafood consumption for different age groups were 147 – 564 and 4.7 – 18.5 ng/day, or 8.5 – 12.9 and 0.27 – 0.46 ng/kg bw/day, respectively (Table 5). Because seafood products accounted for less than 10% of total food consumption from our dietary survey (Table 1), higher dietary intake of DDTs or PBDEs can be expected via all food sources. In addition, for both DDTs and PBDEs, the EDIs of male were slightly higher than those of female independent of different age groups, while the EDIs of the 2–5 age group were much less than those of the 6–18 and > 18 age groups, probably due to the different seafood consumptions for different age groups. If they are normalized to the body weight, the dietary intakes of both PBDEs and DDTs decreased in the order of youth (6–18) > children (2–5) > adults (> 18).

With respect to DDTs, the EDIs (5.74 – 5.84 ng/kg bw/day) via fish consumption for adults in South China were not only higher than those (3.62–5.80 ng/kg bw/day) in Beijing, North China (Li et al., 2008), but also higher than that (4.73 ng/kg bw/day) calculated from a previous study conducted in Zhoushan, East China (Jiang et al., 2005). Besides, the total EDIs (502–564 ng/day) of DDTs for adults residing in South China were comparable to the dietary intake (523 ng/day) from a market basket survey in Sweden (Darnerud et al., 2006).

Dietary intake of PBDEs via consumption of various food types has been examined worldwide. Based on market surveys, the dietary intakes of PBDEs via fish consumption were 23 ng/day in Finland (Kiviranta et al., 2004) and 23.1 ng/day in Sweden (Darnerud et al., 2006), respectively. For most food items of animal origin, the total dietary intake of PBDEs was estimated to be 44 ng/day in Canada (Ryan and Patry, 2001) and 40.8 ng/day in Sweden (Lind et al., 2002). Recently, dietary intake of PBDEs was estimated at 20.8 ng/day for 14 edible marine species (fish and molluscs) in Catalonia of Spain (Domingo et al., 2006), and the median PBDE intake was 14 ng/day via seafood consumption in Belgium (Voorspoels et al., 2007). The dietary intake of PBDEs for adult (15.9–17.6 ng/day) obtained from the present study was comparable to or less than all the previously obtained results.

A summary of the dietary intake values of PBDEs (mostly via fish consumption) obtained by different studies is presented in Table 6. It should be noted that such comparison, as well as the comparison of the concentration data earlier, is qualitative at best, because sampling strategy (e.g., the type of foodstuff included), available data format (mean, median, or upper or lower bound values), the number of BDE congeners analyzed and sampling time were quite variable among the different studies. As an example to illustrate the consequence, a recent survey conducted in Hong Kong (Cheung et al., 2008) included 22 BDE congeners and obtained EDIs of PBDEs at 331–1677 ng/day, much higher than those from other previous studies employing fewer BDE congeners.

The relative contribution of each seafood group to the total EDIs of DDTs and PBDEs is displayed in Fig. 2. Fish contributed the most to the EDI of DDTs (an average of 57%), followed by molluscs (an average of 38%). The significantly higher contribution from fish is mainly resulted from its relatively high consumption rate compared to other seafood groups and the opposite is true for shrimps and crabs. Overall, the patterns of relative contributions from the seafood types to the total EDIs of PBDEs were similar to those of DDTs (Fig. 2), i.e., the contributions were dominated by fish (an average of 45%) and molluscs (an average of 45%). As a consequence of the high levels of PBDEs and DDTs in fish and molluscs, the EDIs of DDTs and PBDEs via consumption of fish and molluscs accounted for a major portion of the total dietary intake for all seafood species under investigation.

Table 6
Estimated dietary intake of PBDEs (ng/day) in different countries.

Country	Sample	Sampling	EDI	BDE congeners	References
Finland	Fish	1997-1999	23	47, 99, 100, 153, 154	Kiviranta et al. (2004)
Sweden	Fish	1999	23.1(ND=1/2 LOD)	47, 99, 100,153, 154	Darnerud et al. (2006)
Canada	Food of animal origin	1998	44	47, 99, 100,153, 154	Ryan and Patry (2001)
Sweden	Food of animal origin	1999	40.8 (ND=0)	47, 99, 100,153, 154	Lind et al. (2002)
Spain	Marine species	2005	20.8	47, 99, 100, 153, 154, 183	Domingo et al. (2006)
Belgium	Seafood	2005	14 (median bound)	47, 99, 100, 153, 154, 183	Voorspoels et al. (2007)
Hong Kong	Fish	2004	311–1677	22 BDE congeners	Cheung et al. (2008)
USA	Fish	–	8.94–15.7	Thirteen BDE congeners	Schecter et al. (2006)
Dutch	Fish and molluscs	–	8.3 (median bound)	28, 47, 99, 100, 153, 154, 183, and 209	van Leeuwen and de Boer (2008)
South China	Seafood	2005	15.9–17.6	28, 47, 66, 99, 100, 153, 154, 138, 183, and 209	Present study

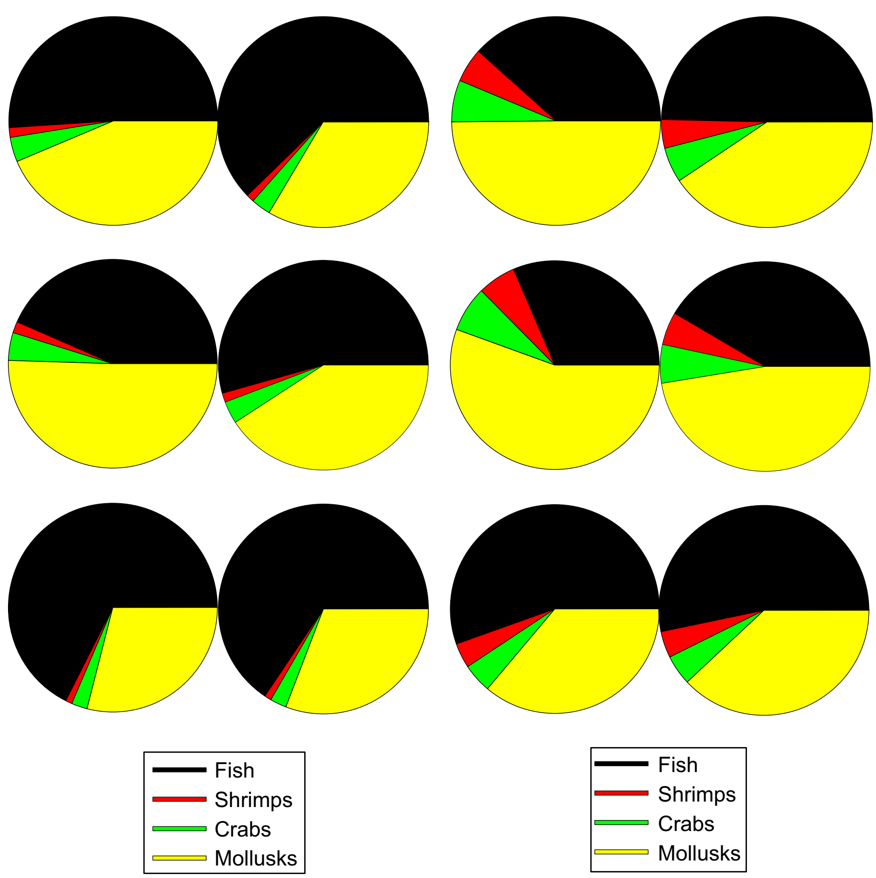

Fig. 2. Relative contribution of each sample group to the total dietary intake. The left two columns are for DDTs and the right two columns are for PBDEs.

3.4. Risk assessment

Several guidelines established by different international organizations can be used for assessment of health risk of PHCs to humans. These guidelines include the acceptable daily intake (ADI) recommended by the World Health Organization (WHO) (Food and Agriculture Organization/World Health Organization, 2001), the minimal risk level (MRL) formulated by the Agency for Toxic Substances and Disease Registry (ATSDR) (Schafer and Kegley, 2002), and the potential risk assessment method based on the benchmark concentration (Jiang et al., 2005). The EDI of DDTs for adults (8.5–8.9 ng/kg bw/day) via seafood consumption from the present study is far below the ADI (20 μg/kg bw/day) proposed by the WHO (Food and Agriculture Organization/World Health Organization, 2001) and the MRL (500 ng/kg day for *p,p′*-DDT) recommended by ATSDR (Schafer and Kegley, 2002). This, however, does not necessarily suggest no potential health risk associated with seafood consumption in South China. The present results indicate that DDTs were detected in all the seafood samples and the levels of DDTs in nearly one-third of the samples (31%) were higher than the maximum admissible concentrations suggested by the USEPA (14.4 ng/g wet wt) (EPA, 2000).

To screen the potential health significance of the estimated dietary exposure to DDTs, a method based on the benchmark concentration was also used for assessment. The detailed method has been described elsewhere (Jiang et al., 2005). Briefly, two hazard ratios (HRs), i.e., from the 50th and the 95th percentile measured concentrations (50th and 95th MEC), were used for assessing the potential health risk to humans, as calculated by

$$\text{Hazard ratio (HR)} = \frac{\text{Estimated daily intake}}{\text{Benchmark concentration}} \tag{1}$$

$$\text{Benchmark concentration} = \frac{\text{Risk} \times \text{body weight}}{\text{Fish consumption} \times \text{Slope factor}} \tag{2}$$

where the benchmark concentration for carcinogenic effect was derived from the USEPA cancer slope factor (obtained from the USEPA's Integrated Risk Information System (http://www.epa.gov/iris/)) by setting cancer risk to one in one million due to lifetime exposure. The benchmark concentration for non-cancer carcinogenic effect was the USEPA reference dose. Because the dietary intake of DDTs was mainly originated from fish consumption, human health risk associated with fish consumption could well represent that with seafood consumption.

For different age groups, the 50th MEC in fish was used for evaluating the non-cancer and cancer risks. The non-cancer HRs for different age groups are all substantially lower than one (Fig. 3). This is well in accordance with the results from a similar risk evaluation conducted in Zhoushan, East China (Jiang et al., 2005). On the other hand, the 50th percentile cancer HRs for all age groups are considerably higher than one, and there was no significant difference among the 50th percentile cancer HRs of different age groups. This suggests that dietary intake of DDTs via fish consumption may be of serious concern and a lifetime cancer risk remains a possibility. As for different seafood groups, two cancer HRs, i.e. the 50th and 95th MEC HRs, in different seafood groups were used for assessment, and the results (for adults only) are presented in Fig. 4. Both 50th MEC and 95th MEC HRs of shrimps and crabs are less than one, indicating the health risk related to consumption of shrimps and crabs is neglected. On the contrary, both HRs of fish and the 95th MEC HR of molluscs are higher than one. This suggests that fish and molluscs may be of human health concern. A refined risk assessment should be considered to further ascertain the real risk via molluscs consumption, and an initiation of appropriate management strategies is deemed urgent for fish.

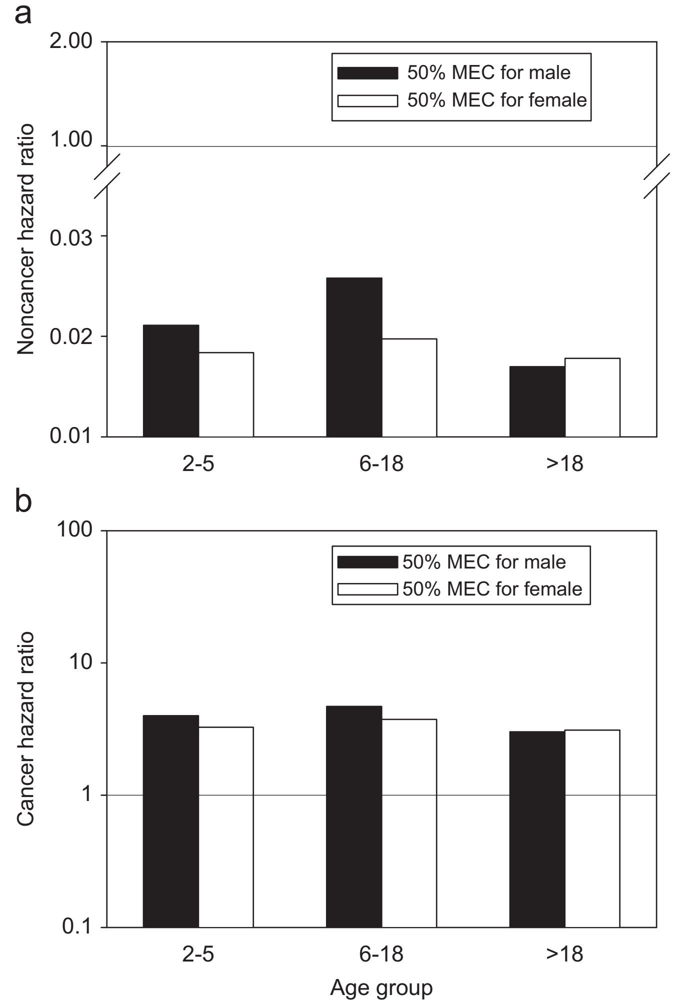

Fig. 3. Non-cancer hazard and cancer hazard ratios of DDTs for different age groups derived from fish consumption in South China. 50% MEC denote the 50th percentile measured concentrations.

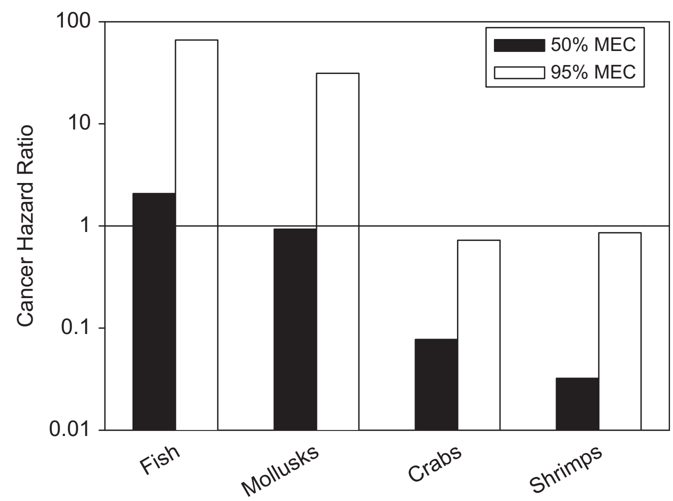

Fig. 4. Cancer hazard ratios of DDTs derived from consumption of different seafood types (only for adults). 50% MEC and 95% MEC denote the 50th percentile and 95th percentile measured concentrations, respectively.

It is worthwhile to note that shrimps are extensively farmed in South China to meet the growing domestic and international demand. In 2005, the amount of shrimps exported from China accounted for 15% of the global need (http://www.99sj.com.cn/

News/62711.htm), 60% of which came from Zhanjiang District, one of the sampling locations in the present study. The assessment above suggests that the health risk related to consumption of shrimps is minimal, while added concern should be paid to fish and molluscs. In conclusion, a lifetime cancer risk from dietary intake of DDTs has remained a possibility in South China, especially for coastal residents who tend to consume more fish and molluscs than inland residents. Possible health risk caused by DDTs via seafood consumption should not be neglected.

It is still a challenge to assess the potential health risk of PBDEs, because toxicological and epidemiological data associated with PBDEs are extremely limited. Currently, only a lowest observed adverse effect of level (LOAEL) of 1 mg/kg/day, on the basis of the most sensitive endpoints of toxic effects of PBDEs (Darnerud et al., 2001; Bocio et al., 2003), and MRLs, which are 0.007 and 10 mg kg/day for lower brominated congeners and Deca-BDE, respectively, derived by the ATSDR (http://www.atsdr.cdc.gov/mrls.html), have been suggested. The total dietary intake of PBDEs for adults (0.27–0.46 ng/kg bw/day) from the present study is several orders of magnitude lower than the suggested LOAEL and the oral MRLs. Therefore, potential health risk of PBDEs via seafood consumption in South China is deemed minimal based on the current toxicological knowledge.

In the present work, the estimation of EDIs and the related health risk assessment were based on the residual levels of PHCs in seafood samples and the consumption data of different food items. Both of them could introduce considerable amounts of uncertainty. For example, the number of children (2–5 years old) interviewed in our dietary survey was only 9, likely to result in a heavy bias for the estimated food consumption. If these and other potential types of uncertainty (including those with the occurrence of PHCs in seafood samples and sample representation) are taken into account, the results of EDIs and health risk assessments are obviously qualitative only and should be used with caution. A more refined assessment of the dietary intakes and subsequently the potential health risk assessment are deemed necessary in the future when more information becomes available.

4. Conclusion

A total of 602 seafood samples from South China were analyzed for DDTs and PBDEs. The results indicated that levels of DDTs and PBDEs varied significantly among different species and ranged from nd to 699 and nd to 5.93 ng/g, respectively. Comparison of the levels of DDTs and PBDEs in mussel worldwide suggested that South China is probably one of the most DDT-polluted areas, but the magnitude of PBDE contamination in South China is moderate globally. Combined with a recent dietary survey conducted at the same sampling locations, dietary intakes of DDTs and PBDEs by local residents via seafood consumption for different age groups were estimated at 147–564 and 4.7–18.5 ng/day or 8.5–12.9 and 0.27–0.46 ng/kg bw/day, respectively. On average, fish contributed the largest proportion of the dietary intake of DDTs (57%), followed by molluscs (38%). Similarly, the dietary intakes of PBDEs were also predominated by fish (45%) and molluscs (45%). Assessments based on several available guidelines suggested insignificant human health risk for dietary intake of PBDEs associated with consumption of the seafood under investigation. However, lifetime cancer risk from dietary exposure to DDTs has remained a possibility, especially for coastal residents who tend to consume more fish and molluscs than inland residents.

Acknowledgments

This research was financially supported by the National Natural Science Foundation of China (40821003, U0633005 and 40532013). The present study did not involve humans or experimental animals; the dietary survey was conducted based on a question–answer procedure. We also thank T.S. Xiang for assistance in GC/MS analysis. This is contribution No. IS-1235 from GIGACS.

References

Allchin, C.R., Law, R.J., Morris, S., 1999. Polybrominated diphenylethers in sediments and biota downstream of potential sources in the UK. Environ. Pollut. 105, 197–207.

Bayarri, S., Baldassarri, L.T., Iacovella, N., Ferrara, F., di Domenico, A., 2001. PCDDs, PCDFs, PCBs and DDE in edible marine species from the Adriatic Sea. Chemosphere 43, 601–610.

Bayen, S., Thomas, G.O., Lee, H.K., Obbard, J.P., 2003. Occurrence of polychlorinated biphenyls and polybrominated diphenyl ethers in green mussels (*Perna viridis*) from Singapore, Southeast Asia. Environ. Toxicol. Chem. 22, 2432–2437.

Bethune, C., Nielsen, J., Julshamn, K., 2004. Current levels of primary polybrominated diphenyl ethers (PBDEs) in Norwegian seafood. Organohalogen Compd. 66, 3814–3819.

Binelli, A., Bacchetta, R., Vailati, G., Galassi, S., Provini, A., 2001. DDT contamination in Lake Maggiore (N. Italy) and effects on zebra mussel spawning. Chemosphere 45, 409–415.

Binelli, A., Guzzella, L., Roscioli, C., 2008. Levels and congener profiles of polybrominated diphenyl ethers (PBDEs) in Zebra mussels (*D. polymorpha*) from Lake Maggiore (Italy). Environ. Pollut. 153, 610–617.

Bocio, A., Llobet, J.M., Domingo, J.L., Corbella, J., Teixido, A., Casas, C., 2003. Polybrominated diphenyl ethers (PBDEs) in foodstuffs: human exposure through diet. J. Agric. Food Chem. 51, 3191–3195.

Branchi, I., Capone, F., Alleva, E., Costa, L.G., 2003. Polybrominated diphenyl ethers: neurobehavioral effects following developmental exposure. Neurotoxicology 24, 449–462.

Cheung, K.C., Zheng, J.S., Leung, H.M., Wong, M.H., 2008. Exposure to polybrominated diphenyl ethers associated with consumption of marine and freshwater fish in Hong Kong. Chemosphere 70, 1707–1720.

Christensen, J.H., Platz, J., 2001. Screening of polybrominated diphenyl ethers in blue mussels, marine and freshwater sediments in Denmark. J. Environ. Monit. 3, 543–547.

Cleemann, M., Riget, F., Paulsen, G.B., Klungsør, J., Dietz, R., 2000. Organochlorines in Greenland marine fish, mussels and sediments. Sci. Total Environ. 245, 87–102.

Connor, L., Johnson, M.S., Copplestone, D., Leah, R.T., 2001. Recent trends in organochlorine residues in mueesl (*Mytilus edulis*) from the Mersey Estuary. Mar. Environ. Res. 52, 397–411.

Darnerud, P.O., Atuma, S., Aune, M., Bjerselius, R., Glynn, A., K, P.G., Becker, W., 2006. Dietary intake estimations of organohalogen contaminants (dioxins, PCB, PBDE and chlorinated pesticides, e.g. DDT) based on Swedish market basket data. Food Chem. Toxicol. 44, 1597–1606.

Darnerud, P.O., Eriksen, G.S., Jóhannesson, T., Larsen, P.B., Viluksela, M., 2001. Polybrominated diphenyl ethers: occurrence, dietary exposure, and toxicology. Environ. Health Perspect. 109 (Suppl. 1), 49–68.

de Boer, J., Wester, P.G., van der Horst, A., Leonards, P.E.G., 2003. Polybrominated diphenyl ethers in influents, suspended particulate matter, sediments, sewage treatment plant and effluents and biota from the Netherlands. Environ. Pollut. 122, 63–74.

De Brito, A.P.X., Bruning, I.M.R.D.A., 2002. Chlorinated pesticides in mussels from Guanabara Bay, Rio de Janeiro, Brazil. Mar. Pollut. Bull. 44, 71–81.

Domingo, J.L., bocio, G.F., Llobet, J.M., 2006. Exposure to PBDEs and PCDEs associated with the consumption of edible marine species. Environ. Sci. Technol. 40, 4394–4399.

EPA, 2000. Guidance for assessing chemical contaminant, data for use in fish advisories. Fish Sampling and Analysis, vol. 1, third ed. EPA 823-R-95-007. Office of Water, Washington, DC.

Food and Agriculture Organization/World Health Organization, 2001. Food Standards Program, Codex Committee on Food Additives and Contaminants, The Netherlands.

Granby, K., Spliid, N., 1995. Hydrocarbons and organochlorines in common mussels from the Kattegat and the belts and their relation to condition indices. Mar. Pollut. Bull. 30, 74–82.

Guan, Y.F., Wang, J.Z., Ni, H.G., Luo, X.J., Mai, B.X., Zeng, E.Y., 2007. Riverine inputs of polybrominated diphenyl ethers from the Pearl River Delta (China) to the coastal ocean. Environ. Sci. Technol. 41, 6007–6013.

Guo, J.-Y., Wu, F.-C., Mai, B.-X., Luo, X.-J., Zeng, E.Y., 2007a. Polybrominated diphenyl ethers in seafood products of South China. J. Agric. Food Chem. 55, 9152–9158.

Guo, J.-Y., Zeng, E.Y., Wu, F.-C., Meng, X.-Z., Mai, B.-X., Luo, X.-J., 2007b. Organochlorine pesticides in seafood products from southern China and health risk assessment. Environ. Toxicol. Chem. 26, 1109–1115.

Guo, Y., Yu, H.-Y., Zeng, E.Y., 2009. Occurrence, source diagnosis, and biological effect assessment of DDT and its metabolites in various environmental compartments of the Pearl River Delta, South China: a review. Environ. Pollut. 157, 1753–1763.

Hallgren, S., Darnerud, P.O., 2002. Polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyls (PCBs) and chlorinated paraffins (CPs) in rats—testing interactions and mechanisms for thyroid hormone effects. Toxicology 177, 227–243.

Harrad, S., Wijesekera, R., Hunter, S., Halliwell, C., Baker, R., 2004. Preliminary assessment of UK human dietary and inhalation exposure to polybrominated diphenyl ethers. Environ. Sci. Technol. 38, 2345–2350.

Jiang, Q.T., Lee, T.K., Chen, K., Wong, H.L., Zheng, J.S., Giesy, J.P., Lo, K.K., Yamashita, N., Lam, P.K., 2005. Human health risk assessment of organochlorines associated with fish consumption in a coastal city in China. Environ. Pollut. 136, 155–165.

Johansson, I., Héas-Moisan, K., Guiot, N., Munschy, C., Tronczyński, J., 2006. Polybrominated diphenyl ethers (PBDEs) in mussels from selected French coastal sites: 1981–2003. Chemosphere 64, 296–305.

Khaled, A., Nemr, A.E., Said, T.O., El-Sikaily, A., Abd-Alla, A.M.A., 2004. Polychlorinated biphenyls and chlorinated pesticides in mussels from the Egyptian Red Sea coast. Chemosphere 54, 1407–1412.

Kiviranta, H., Ovaskainen, M.-L., Vartiainen, T., 2004. Market basket study on dietary intake of PCDD/Fs, PCBs, and PBDEs in Finland. Environ. Int. 30, 923–932.

Li, X., Gan, Y., Yang, X., Zhou, J., Dai, J., Xu, M., 2008. Human health risk of organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) in edible fish from Huairou Reservoir and Gaobeidian Lake in Beijing. China: Food Chem. 109, 348–354.

Liem, A.K.D., Furst, P., Rappe, C., 2000. Exposure of populations to dioxins and related compounds. Food Addit. Contam. 17, 241–259.

Lind, Y., Aune, M., Atuma, S., Becker, W., Bjerselius, R., Glynn, A., Darnerud, P.O., 2002. Food intake of the brominated flame retardants PBDEs and HBCD in Sweden. Organohalogen Compd. 58, 181–184.

Liu, Y., Zheng, G.J., Yu, H., Martin, M., Richardson, B.J., Lam, M.H.W., Lam, P.K.S., 2005. Polybrominated diphenyl ethers (PBDEs) in sediments and mussel tissues from Hong Kong marine waters. Mar. Pollut. Bull. 50, 1173–1184.

Mai, B.X., Zeng, E.Y., Luo, X.J., Yang, Q.S., Zhang, G., Li, X.D., Sheng, G.Y., Fu, J.M., 2005. Abundances, depositional fluxes, and homologue patterns of polychlorinated biphenyls in dated sediment cores from the Pearl River Delta, China. Environ. Sci. Technol. 39, 49–56.

Meng, X.-Z., Zeng, E.Y., Yu, L.-P., Mai, B.-X., Luo, X.-J., Ran, Y., 2007a. Persistent halogenated hydrocarbons in consumer fish of China: regional and global implications for human exposure. Environ. Sci. Technol. 41, 1821–1827.

Meng, X.Z., Zeng, E.Y., Yu, L.P., Guo, Y., Mai, B.X., 2007b. Assessment of human exposure to polybrominated diphenyl ethers in China via fish consumption and inhalation. Environ. Sci. Technol. 41, 4882–4887.

Monirith, I., Ueno, D., Takahashi, S., Nakata, H., Sudaryanto, A., Subramanian, A., Karuppiah, S., Ismail, A., Muchtar, M., Zheng, J., Richardson, B.J., Prudente, M., Hue, N.D., Tana, T.S., Tkalin, A.V., Tanabe, S., 2003. Asia-Pacific mussel watch: monitoring contamination of persistent organochlorine compounds in coastal waters of Asian countries. Mar. Pollut. Bull. 46, 281–300.

Nakata, H., Kawazoe, M., Arizono, K., Abe, S., Kitano, T., Shimada, M., Li, W., Ding, X., 2002. Organochlorine pesticides and polychlorinated biphenyl residues in foodstuffs and human tissues from China: status of contamination, historical trend and human dietary exposure. Arch. Environ. Contam. Toxicol. 43, 473–480.

Ni, H.G., Zeng, E.Y., 2009. Law enforcement and global collaboration are the keys to containing e-waste tsunami in China. Environ. Sci. Technol. 43, 3991–3994.

Oros, D.R., Hoover, D., Rodigari, F., Crane, D., Sericano, J., 2005. Levels and distribution of polybrominated biphenyl ethers in water, surface sediments, and bivalves from the San Francisco Estuary. Environ. Sci. Technol. 39, 33–41.

Ramu, K., Kajiwara, N., Isobe, T., Takahashi, S., Kim, E.-Y., Min, B.-Y., We, S.-U., Tanabe, S., 2007. Spatial distribution and accumulation of brominated flame retardants, polychlorinated biphenyls and organochlorine pesticides in blue mussels (*Mytilus edulis*) from coastal waters of Korea. Environ. Pollut. 148, 562–569.

Ryan, J.J., Patry, R., 2001. Body burdens and food exposure in Canada for polybrominated diphenyl ethers (BDEs). Organohalogen Compd. 51, 226–229.

Schafer, K.S., Kegley, S.E., 2002. Persistent toxic chemicals in the US food supply. J. Epidemiol. Community Health 56, 813–817.

Schecter, A., Papke, O., Harris, T.R., Tung, K.C., Musumba, A., Olson, J., Birnbaum, L., 2006. Polybrominated diphenyl ether (PBDE) levels in an expanded market basket survey of US food and estimated PBDE dietary intake by age and sex. Environ. Health. Perspect. 114, 1515–1520.

Sjödin, A., Carlsson, H., Thuresson, K., Sjölin, S., Bergman, Å., Östman, C., 2001. Flame retardants in indoor air at an electronic recycling plant and at other work environments. Environ. Sci. Technol. 35, 448–454.

Sjodin, A., Patterson Jr., D.G., Bergman, Å., 2003. A review on human exposure to brominated flame retardants—particularly polybrominated diphenyl ehters. Environ. Int. 29, 829–839.

Smith, A.G., Gangolli, S.D., 2002. Organochlorine chemicals in seafood: occurrence and health concerna. Food Chem. Toxicol. 40, 767–779.

Tanabe, S., Prudente, M.S., Kan-atireklap, S., Subramanian, A., 2000. Mussel watch: marine pollution monitoring of butyltins and organochlorines in coastal waters of Thailand, Philippines and India. Ocean Coastal Manage. 43, 819–839.

van Leeuwen, S.P.J., de Boer, J., 2008. Brominated flame retardants in fish and shellfish—levels and contribution of fish consumption to dietary exposure of Dutch citizens to HBCD. Mol. Nutr. Food Res. 52, 194–203.

Voorspoels, S., Covaci, A., Neels, H., Schepens, P., 2007. Dietary PBDE intake: a market-basket study in Belgium. Environ. Int. 33, 93–97.

Yang, N., Matsuda, M., Kawano, M., Wakimoto, T., 2006. PCBs and organochlorine pesticides (OCPs) in edible fish and shellfish from China. Chemosphere 63, 1342–1352.

Yang, R.Q., Yao, Z.W., Jiang, G.B., Zhou, Q.F., Liu, J.Y., 2004. HCH and DDT residues in molluscs from Chinese Bohai coastal sites. Mar. Pollut. Bull. 48, 795–799.

Yang, X.-g., Li, Y.-p., Ma, G.-s., Hu, X.-q., Wang, J.-z., Cui, Z.-h., Wang, Z.-h., Wang, J.-z., Yu, W.-t., Yang, Z.-x., Zhai, F-y., 2005. Study on weight and height of the Chinese people and the differences between 1992 and 2002. Chin. J. Epidemiol. 26, 489–493 in Chinese.

Zhang, S.-y., Han, Y.-s., Shen, X.-z., Ma, Z.-g., Liu, G., Liu, l.-j, 2008. Growth charts of height, weight and body mass index for Han Chinese children and adolescent from large and middling cities of China. Chin. J. Child Health Care 16, 257–259 in Chinese.