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Human health risk assessment of organochlorines associated with fish consumption in a coastal city in China

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Health risk assessment of organochlorines associated with fish consumption reveals potential cancer risks for some contaminants in a coastal population in China.

Abstract

Food consumption is an important route of human exposure to organochlorines (OCs). In order to assess the potential health risks associated with these contaminants due to fish consumption, five species of fish were collected from a local market in Zhoushan City, an island in the East China Sea. Dioxin-like compounds, such as polychlorinated dibenzo-*p*-dioxins/ dibenzofurans, in the fish samples were screened by H4IIE-luc cell bioassay, and the concentrations of specific organochlorines were measured by gas chromatograph-electron capture detector (GC-ECD). The bioassay results indicated that concentrations of dioxin-like compounds in the fish samples were below detection limit (0.64 pg/mL). The concentrations of OC pesticides and PCBs ranged from 0.67 to 13 and 0.24 to 1.4 ng/g wet wt., respectively. Significantly, concentrations of *p,p′*-DDE in fish meat were comparatively high (average 3.9 ng/g wet wt.) compared with the other OC pesticides. The daily fish consumption, based on a dietary survey conducted among 160 local healthy residents, was determined to be 105 g/person. The relevant cancer benchmark concentrations of HCB, dieldrin, chlordane, DDTs and PCBs were 0.36, 0.04, 1.6, 1.7, and 0.29 ng/kg per day, respectively, based on the local diet. The hazard ratios (HRs), based on non-cancer endpoints were all less than 1.0, while the HRs based on cancer were greater than 1.0 for certain contaminants based on the 95th centile concentration in fish tissue.

Keywords: Health risk assessment; Organochlorines; Dioxin-like compounds; Diet consumption; Coastal city

1. Introduction

In China, the use of organochlorine (OC) pesticides, such as DDTs (dichlorodiphenyltrichloroethanes) and HCHs (hexachlorocyclohexanes), started in the 1950s, and reached a maximum in the 1970s and early 1980s. Although the agricultural use of DDTs and HCHs has been banned in China for nearly two decades, relatively great concentrations of these OC pesticides can still be found in river sediments (Hong et al., 1995, 1999; Wu et al., 1999). Compared with concentrations in the environment, very great concentrations of HCHs (ca. 1400 ng/g wet wt.) were reported in foodstuffs, such as fish and eggs, collected in 1978 (Chen and Gao, 1993). One study showed that Ya-er Lake in Wuhan, Hubei Province, was very contaminated by dioxins (Wu et al.,

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2002). Polychlorinated biphenyls (PCBs) were banned in China in the 1980s. Recent surveys have found that concentrations of PCBs ranged from 15.1 to 57.9 ng/g (mean: 34.5 ng/g) in the sediments of Minjiang River in southern China (Zhang et al., 2003), and the concentrations ranged from 1.7 to 14.3 ng/g in some Chinese estuaries (Yuan et al., 2001). Many OCs are still present in the environment due to their high environmental persistence. However, the concentrations of PCBs recorded in China were generally less than those found in developed industrial countries such as the U.S., probably due to the lesser usage historically. For this reason, PCBs are not generally considered a contaminant of major concern in China (Yuan et al., 2001).

Organochlorines (OCs), such as chlorinated pesticides and PCBs, dioxin-like compounds, have been monitored routinely in the environment and foodstuff in various countries to evaluate their potential health risk to humans (NRC, 1993; MacIntosh et al., 1996). Consumption of contaminated food is an important route of human exposure to OCs. Following the banning of these chemicals in the 1980s, the concentrations of OCs in general foodstuff had decreased significantly (Zhang et al., 1997; Li, 1999). There is, however, a lack of information on OC concentrations in seafood in China. In coastal cities, consumption of contaminated seafood is the main pathway of human exposure. A large percentage (75%) of DDT intake by Chinese was attributed to the consumption of marine food (Nakata et al., 2002). Exposure to other OCs via fish consumption contributed large percentages (67% of chlordane and 60% of PCBs) in the total exposure from all foods (Dougherty et al., 2000). Concentrations of OC contaminants in fish had led to health concerns, particularly for high-risk groups, such as pregnant women and children (EPA, 1998). High concentrations of DDTs (7 600 ng/g lipid wt.) had been found in human tissues from China (Nakata et al., 2002). Another study reported elevated concentrations of DDT, chlordanes and PCBs in human breast milk in Dalian and Shenyang, China (Kunisue et al., 2004).

Assessments of risks to human health have been undertaken worldwide to examine the potential health risk due to exposure to toxic contaminants in various environmental media and foodstuff (NRC, 1993). Food consumption databases have been established to provide the necessary information for assessing the health risks associated with consumption of contaminated food in countries, such as the U.S. (Dougherty et al., 2000) and Canada (Berti et al., 1998). There has been a tendency for risk assessors in countries that do not have comprehensive food consumption databases to adopt the American food consumption data for risk assessment. By way of an example, a study aimed to estimate the health risk of coastal populations in China due to seafood consumption was undertaken using American food consumption data, e.g. 16 g/person per day for shellfish (Fung et al., 2004). To provide a more accurate assessment of the risks, it is necessary to establish a specific food consumption database for the Chinese population so that the health risks to the target populations can be meaningfully assessed, and the risks effectively managed.

The coastal regions of China have been subjected to intense industrial activities and urban development, and thus environmental impacts are likely to be most severe along the Chinese coastline. People in coastal cities have a greater chance of exposure to toxic contaminants via the consumption of seafood than inland populations. To this end, a survey was conducted to collect information on dietary composition from a population in a 'typical' coastal city in China, Zhoushan. Zhoushan Island is located on the Chinese coast, near Hangzhou, Ningbo and Shanghai. The offshore area is an important fishing ground, where the total length of the continental coastline is 1500 km and the total sea area of the fishing ground is about 100 000 km^2. The annual fish catch is about 800 000 t, representing one third of the national total (Liu et al., 1991). Specifically, this study aims to (1) measure the concentrations of OCs (chlorinated pesticides, PCBs, and dioxin-like compounds) in selected food fish species collected in the East China Sea; (2) derive relevant cancer benchmark concentrations for a Chinese coastal population based on the local diet consumption data/pattern from Zhoushan; and (3) assess the potential health risks to human consumers due to dietary fish consumption in a coastal city in China.

2. Materials and methods

2.1. Sample collection

Five species of fish were purchased from a local market in Zhoushan in October 2003. Details of the sampling locations were obtained from the fishermen who caught the fish from two stations in the East China Sea [Station (St.) 1 and St. 2], and a coastal station (St. 3) near Zhoushan. Sampling locations and details of the fish samples are shown (Fig. 1 and Table 1). Fish were wrapped in aluminum foil, placed in polyethylene bags, and then stored frozen at −20 °C until analyzed.

2.2. Sample preparation and extraction

Prior to analysis, filleted muscle tissues (with skin and no bone) of individual fish were freeze-dried, ground into a powder, and Soxhlet extracted for 18 hr using a mixture of dichloromethane and hexane (3:1 by volume; 400 mL). The extract was rotary evaporated at 40 °C, and the lipid content gravimetrically determined

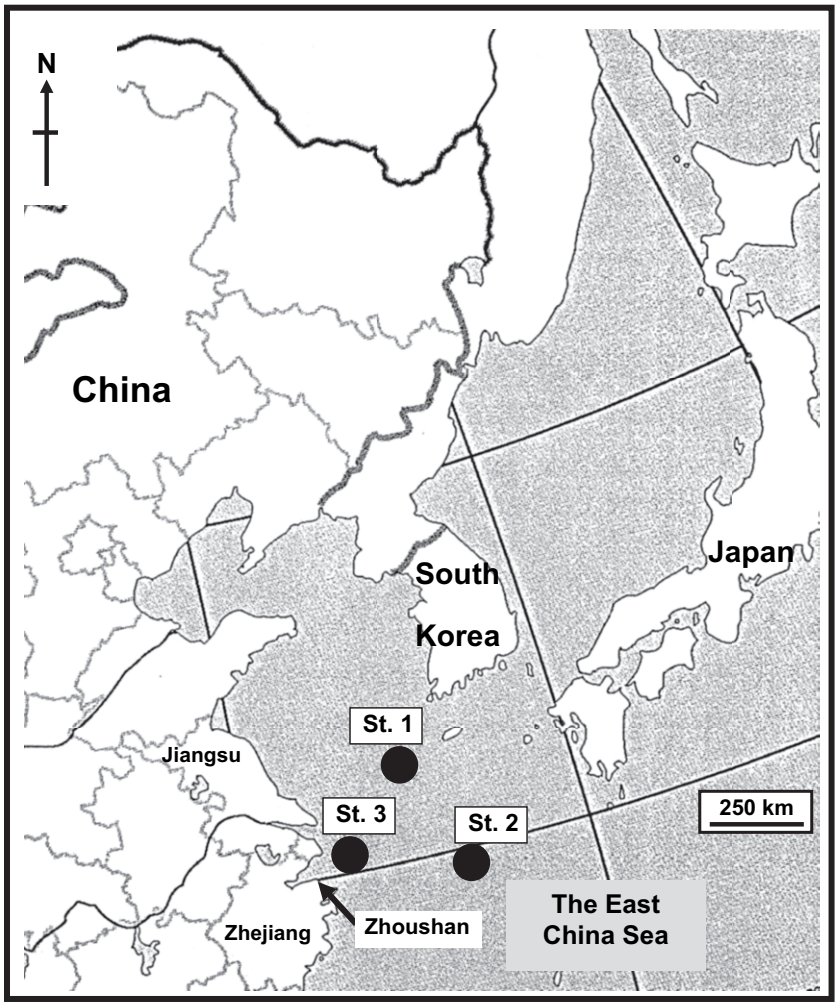

Fig. 1. Sampling locations in the East China Sea.

Table 1
Characteristics of fish samples collected in the East China Sea, near Zhoushan

Fish species	Code	St. 1		St. 2		St. 3	
		Weight (g)	Length (cm)	Weight (g)	Length (cm)	Weight (g)	Length (cm)
White mouth croaker (*Argyrosomus argentatus*)	WC1	200	25	16	23	120	22
	WC2	160	20	170	22	130	21
	WC3	170	25	220	26	110	21
	WC4	180	23	200	26	110	20
Small yellow croaker (*Pseudosciaena polyactis*)	YC1	73	20	61	19	97	23
	YC2	59	18	64	19	120	24
	YC3	54	17	72	21	84	21
	YC4	64	19	55	18	100	23
Cinnamon flounder (*Cynoglossus robustus*)	CF1	120	26	150	27	150	29
	CF2	230	32	170	30	170	30
	CF3	160	28	150	29	150	32
	CF4	75	22	140	29	120	30
Silvery pomfret (*Pampus argenteus*)	WP1	240	23	170	19		
	WP2	190	23	180	22		
	WP3	220	21	200	23		
	WP4	200	23	210	21		
Conger pike (*Muraenesox cinereus*)	PE1					380	62
	PE2					370	59

from an aliquot of the extract following Khim et al. (1999a,b, 2000).

2.3. Instrumental analysis

The detailed procedures have been described in Zheng and Quinn (1988) and Connell et al. (2003). Briefly, a fractionation column consisting of 6 g activated silica gel (maintained at 450 °C for 8 h) was washed twice with 15 mL dichloromethane and then 15 mL hexane. The extraction aliquot (~1 mL) was added to the column. After the aliquant entered the silica gel, 12 mL of hexane was eluted to obtain Fraction 1 containing petroleum hydrocarbons. A mixture of hexane and dichloromethane (80:20) was eluted to collect OC pesticides and PCBs (Fraction 2). Each fraction was reduced to 100 μL prior to quantitation by a Hewlett Packard 6890 series GC-ECD (Wilmington, DE) equipped with an auto injector and sampler (Hewlett Packard 7683 series). The GC column employed was a 30 m HP-5 capillary column coated with 0.25 μm film thickness of 100% dimethylpolysiloxane (J&W Scientific Co., Folsom, CA). Individual compounds, including 28 PCB congeners and OC pesticides such as DDT and its metabolites (DDTs: *p,p′*-DDT, *p,p′*-DDD, *o,p′*-DDT, and *p,p′*-DDE), dieldrin, aldrin, endrin, kepone, chlordane compounds (CHLs: *trans*-chlordane, *cis*-chlordane), hexachlorocyclohexane isomers (HCHs: α-HCH, β-HCH, γ-HCH, and δ-HCH) and hexachlorobenzene (HCB) were measured. Organochlorine concentrations were calculated from the peak area of the sample to a corresponding external standard. The PCB standard used for quantification was a mixture with known composition and content, containing 28 congeners from monochloro- to decachlorobiphenyls (SRM 2262). In addition, accuracy and precision were determined by analysis of standard reference materials (SRM 2978). Concentrations of individually resolved peaks of PCB isomers and congeners were summed to obtain total PCB concentrations. Recoveries of target analytes through this analytical method were 90±2.0% for PCBs, 99±7.7% for DDTs, 100±7.7% for HCHs, 97±4.7% for CHLs and 93±5.4% for HCB. Concentrations were not corrected for recoveries. A procedural blank was analyzed with every set of 6 samples to check for interfering compounds and correction was made, if necessary.

2.4. H4IIE cell-based bioassay

The concentrations of dioxin-like compounds in the fish extract were measured with the in vitro H4IIE-luc bioassay by use of methods described elsewhere (Sanderson et al., 1996; Khim et al., 1999a,b). Briefly, cells for bioassay were plated into 96-well culture plates (250 μL/well) and incubated overnight, prior to dosing. Test and control wells were dosed with 2.5 μL of raw extract of fish or solvent. Standard wells received different concentrations of 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TCDD), while blank wells received no dose. Luciferase and protein assays were conducted after 72 h of exposure. For screening purposes, significant responses were defined as those outside the range defined by 3 times the standard deviation (expressed in %-TCDD-max) of the mean response in the solvent control (0%-standard-max).

2.5. Dietary survey

A questionnaire-based dietary survey was conducted at Zhoushan in 2001, by randomly selecting and surveying 160 healthy adults from the general population. All the participants were local residents (living >10 years in one place). The referent period for the interview was the year prior to the year of the interview. Dietary data for the referent year were collected during a detailed face-to-face interview. The questionnaire included 12 food categories: meat, fish, shrimp, clam, crab, seaweed, egg, bean, vegetable, creamery, oil and rice. Each group comprised about 2–25 food items. Data collected for each food item included frequency of consumption (number of times per day, week, month or year) and the quantity consumed on each occasion. Some food models, measuring cups and bowls were shown during the interview to facilitate the quantification of food intake. The seasonality of consumption for certain food types was also surveyed. Only fish fillets were included in this survey, while whole shrimps, clams and crabs were considered. Shrimps, clams and crabs accounted for <13% of total dietary intake. Daily intake (in g) for each food item was computed for each individual. Statistical analyses were conducted using SPSS for Windows (Chicago, IL), and all tests were considered significant at $p<0.05$.

2.6. Risk assessment

Percent cumulative probabilities on a probabilistic scale were plotted against concentrations on a logarithmic scale for each contaminant (Connell et al., 2003). An equation describing the relationship was obtained by linear regression, from which concentrations corresponding to the 50th and 95th centiles were calculated to provide measures of exposure (Solomon et al., 2000).

To screen for potential public health significance of the estimated exposures, concentrations of exposure were compared to benchmark concentrations for each contaminant. A benchmark concentration, representing a daily concentration below which there is a high probability of no adverse health effect, is different from a benchmark dose, which is a statistically derived value used in setting a Reference Dose (RfD) for non-cancer health effects. Risks associated with cancer and

non-cancer health effects due to exposure were considered separately. The benchmark concentration for carcinogenic effect was derived using USEPA cancer slope factor and the benchmark concentration for non-carcinogenic effect was USEPA Reference Dose. Risk assessments were conducted based on the concentrations of OC pesticides, PCBs and dioxin-like compounds in fish tissues.

Hazard ratios (HRs) were calculated by dividing the average daily exposure by the benchmark concentrations. A hazard ratio greater than unity indicates that the average exposure level exceeds the benchmark concentration (Dougherty et al., 2000).

$$\text{Hazard ratio } (HR) = \frac{\text{Average daily exposure}}{\text{Benchmark concentration}} \quad (1)$$

$$\text{Benchmark concentration} = \frac{\text{Risk} \times \text{Body weight}}{\text{Fish consumption} \times \text{Slope factor}} \quad (2)$$

where risk is the probability of lifetime cancer risk, and slope factor is cancer slope factor obtained from the USEPA Integrated Risk Information System (IRIS) for each contaminant (http://www.epa.gov/iris/). The benchmark concentration is derived by setting the cancer risk to one in one million due to lifetime exposure.

For each contaminant, the average daily exposure level for a population was calculated by:

$$\text{average daily exposure } (\mu\text{g/kg body weight}) = \text{Fish consumption (g/kg body weight)} \times \text{Contaminant concentration } (\mu\text{g/g}) \quad (3)$$

Fish consumption was expressed as daily consumption divided by body weight, which was set at 60 kg for an adult.

Two HRs were estimated to assess the potential health risk to humans. One was based on the 50th centile exposure (fish tissue) concentrations, while the other was based on the 95th centile exposure concentration. The two HRs provided a simple way for screening chemicals that might require a more detailed analysis. When 95th centile HR was greater than unity, a refined risk assessment would subsequently be conducted to further ascertain the real risk. When both 50th and 95th centile HRs were greater than unity, an initiation of appropriate management strategies may be considered.

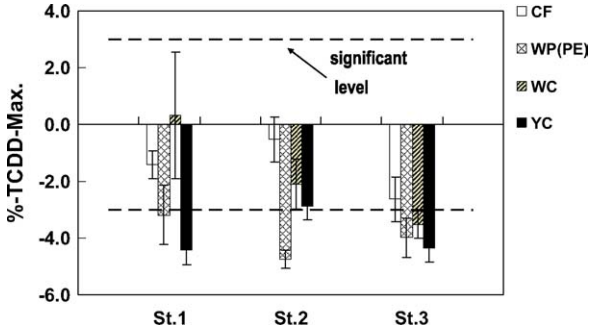

Fig. 2. Luciferase induction in the H4IIE-luc (dioxin-responsive) cell bioassay elicited by Zhoushan fish raw extracts at three sampling locations. Response magnitude presented as percentage of the maximum response observed for a 30 ng 2,3,7,8-tetrachlorodibenzo-*p*-dioxin standard (%-TCDD-Max.). WC, white mouth croaker; YC, small yellow croaker; CF, cinnamon flounder; WP, silvery pomfret; PE, conger pike.

3. Results and discussion

3.1. Dioxin-like compounds

Results of the in vitro H4IIE-luc bioassay on raw tissue extracts of fish collected from different sampling locations are summarized (Fig. 2). The limit of detection (LOD) and ED_{50} for luciferase induction by 2,3,7,8-TCDD were 0.16 pg/well (0.48 fmol/well) and 1.8 ± 0.48 pg/well (0.57 ± 0.15 fmol/well), respectively. No positive significant response (induction) was observed even for the undiluted extracts (greatest concentration). Instead, a negative response relative to 2,3,7,8-TCDD activity was observed for almost all fish species from different sampling locations. Although there are known hotspots of dioxin contamination in China, such as near Ya-er Lake in Wuhan (Wu et al., 2002), there is little information on the extent and degree of dioxin contamination in the Chinese coastal environment. The present study is the first investigation on concentrations of dioxin-like compounds in fish collected in the East China Sea. The results indicate that the concentrations of dioxin-like compounds in fish collected from the East China Sea were low (<0.16 pg TCDD equivalent/well), and that the extracts contained some chemicals that could inhibit the luciferase activity, a response that has been observed previously in other locations (Khim et al., 2000). One recent study reported that the concentrations of dioxin-like compounds, such as PCDDs/PCDFs and coplanar PCBs, in human milk in two north China cities were 3.5–5.7 and 2.5–3.2 pg TEQs/g lipid wt., respectively (Kunisue et al., 2004), while another one reported average TEQ values ranging from 58 to 97 pg/g in a south China city (Lai et al., 2004). Both studies reported total dioxin (PCDDs + PCDFs + Co-PCBs) concentrations in Chinese human breast milk lower than those in the

Table 2
Concentrations [mean (SD) in ng/g wet weight] of organochlorine pesticides in fish collected from East China Sea

	St. 1				St. 2				St. 3			
	CF	WC	WP	YC	CF	WC	WP	YC	CF	WC	PE	YC
Lipid wt./wet wt.%	2.7	7.7	4.7	4.5	0.89	4.0	4.9	5.3	0.65	5.2	10	4.6
α-Chlordane*#	0.88 (0.59)	0.70 (0.34)	0.57 (0.29)	0.61 (0.24)	0.10 (0.07)	0.46 (0.27)	0.61 (0.26)	0.55 (0.16)	0.06 (0.02)	1.9 (0.40)	4.1 (1.2)	1.5 (0.38)
Aldrin	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
γ-HCH	ND	0.01 (0.01)	ND	0.01 (0.02)	ND	ND	ND	ND	ND	ND	ND	ND
Dieldrin*#	ND	0.01 (0.01)	ND	ND	ND	ND	ND	ND	0.01 (0.01)	0.01 (0.02)	0.07 (0.09)	0.05 (0.01)
Endrin-kepone*#	ND	0.02 (0.04)	ND	0.02 (0.01)	ND	ND	ND	0.03 (0.01)	ND	0.01 (0.03)	0.29 (0.40)	0.03 (0.004)
HCB*#	0.29 (0.57)	0.42 (0.28)	0.44 (0.45)	0.68 (0.50)	0.07 (0.09)	0.14 (0.10)	0.11 (0.08)	0.23 (0.22)	0.12 (0.16)	0.52 (0.12)	2.0 (0.75)	0.26 (0.25)
Heptachlor epoxide	ND	0.01 (0.01)	ND	ND	ND	0.01 (0.01)	ND	ND	ND	ND	ND	ND
Heptachlor	ND	0.03 (0.05)	ND	0.01 (0.01)	ND	ND	ND	ND	0.01 (0.01)	ND	0.03 (0.04)	ND
Kepone	ND	ND	ND	ND	ND	0.09 (0.19)	ND	ND	ND	ND	ND	ND
Mirex*	ND	ND	ND	ND	ND	ND	ND	ND	0.03 (0.03)	ND	ND	ND
p,p′-DDE*#	4.1 (3.4)	6.1 (3.1)	2.1 (0.94)	2.5 (0.80)	0.91 (0.67)	4.1 (2.0)	1.9 (0.45)	2.8 (1.0)	0.43 (0.17)	8.5 (2.5)	5.8 (4.5)	8.0 (2.0)
p,p′-DDD+*o,p′*-DDT*#	ND	0.05 (0.06)	ND	ND	ND	0.03 (0.02)	0.01 (0.01)	ND	ND	0.02 (0.01)	ND	0.04 (0.03)
p,p′-DDT*#	0.09 (0.06)	0.30 (0.28)	ND	0.05 (0.02)	0.03 (0.03)	0.08 (0.09)	0.02 (0.02)	0.02 (0.05)	0.02 (0.01)	0.09 (0.02)	0.18 (0.26)	0.09 (0.07)
DDTs*#	4.2 (1.4)	6.4 (2.1)	2.1 (0.70)	2.6 (0.85)	0.94 (0.31)	4.2 (1.4)	1.9 (0.64)	2.8 (0.93)	0.45 (0.15)	8.62 (29)	6.0 (2.0)	8.1 (2.7)
OCs*#	5.4 (1.4)	7.7 (1.8)	3.1 (0.63)	3.9 (0.72)	1.1 (0.29)	4.9 (1.2)	2.6 (0.54)	3.6 (0.79)	0.67 (0.13)	11 (2.4)	13 (2.1)	10 (2.2)

ND indicates samples with OC concentrations below detection limit. The limit of detection is 0.05 ng (0.009 ng/g wet wt.) for individual OC, and 0.5 ng (0.09 ng/g wet wt.) for total OC. CF, cinnamon flounder; WC, white mouth croaker; WP, silvery pomfret; YC, small yellow croaker; PE, conger pike. *There is a significant difference ($p<0.05$) among different sampling locations. #There is a significant difference among different species.

Japanese population (average: 250 pg/g lipid wt.; range: 117–634 pg/g lipid wt.) (Takekuma et al., 2004).

3.2. OC pesticides and PCBs

3.2.1. DDTs

The concentrations of DDTs in the fish samples analyzed in this study are shown in Table 2. Relatively great residue concentrations of total DDTs were found in all five fish species. Mean concentrations of total DDTs varied among species, and were greatest in White Mouth Croaker (WC, *Argyrosomus argentatus*) in all three sites (6.4 ng/g wet wt. in St. 1, 4.2 ng/g wet wt. in St. 2 and 8.6 ng/g wet wt. in St. 3), and lowest in Cinnamon Flounder (CF, *Cynoglossus robustus*) in St. 3 (0.45 ng/g wet wt.). There were significant differences in total DDT concentrations among the five species or among the three sites ($p<0.05$). Average DDT concentrations in St. 1, St. 2 and St. 3 were 3.8, 2.5 and 5.8 ng/g wet wt., respectively. The greatest DDT concentrations were observed at St. 3 (closest to Zhoushan) as compared to St. 1 and St. 2 (stations in the open sea). Notwithstanding, the DDT concentrations in fish collected at St. 3 were still smaller than those measured in fish collected near Shanghai (9.1 ng/g wet wt.) and in coastal waters of Xiamen (<0.5 to 220 ng/g wet wt.) (Klumpp et al., 2002; Nakata et al., 2002). These concentrations were smaller than the concentrations in mussels collected along the Chinese coast (830–54 000 ng/g lipid wt.) (Hong et al., 2000; Monirith et al., 2003). A review by Tanabe et al. (2000) on the health impacts of organochlorine (DDTs, HCHs, chlordanes, and PCBs) residues in fish in Asian developing countries did not include China due to lack of information.

Results of the present study revealed that *p,p′*-DDE was the main contaminant detected in fish tissues, with an average concentration of 3.9 ng/g wet wt., accounting for over 95% (98% on average) of total DDTs (Table 2). *p,p′*-DDE is a metabolite of *p,p′*-DDT, and generally occurs as the main DDT residue in fish samples (Muir et al., 2003). The estimated environmental half-life for DDT, based on studies of bivalves, is 10–20 years (Sericano et al., 1990). In this process, DDT is transformed to DDE and DDD. In the Great Lakes, where an effective ban on DDT use has been in force since the 1970s, DDE accounts for 50–70% of total DDT burden in the 1990s (Newsome and Andrews, 1993). The compositions and residue concentrations of DDTs found in fish samples in this study probably point to the past agricultural usage of DDTs as the main source of these contaminants.

3.2.2. CHLs

The residue concentrations of CHLs in fish tissues collected from Zhoushan are given (Table 2). Again, greater concentrations of CHLs were observed in fish

from St. 3 (0.06–4.1 ng/g wet wt.), and lesser concentrations were observed at the more remote locations (St. 1 and St. 2). These concentrations were slightly greater than those reported by Nakata et al. (2002).

3.2.3. HCB

The average concentrations of HCB in all fish species were less than 1.0 ng/g wet wt., except Conger Pike (*Muraenesox cinereus*) at St. 3 (2.0±0.75 ng/g wet wt.) (Table 2). A similar concentration of 1.3 ng/g wet wt. was reported in mussel samples in the same coastal region of China (Monirith et al., 2003). HCB was not only used as a fungicide, but was also generated as a by-product during the production and usage of several agrochemicals and industrial chemicals, and had been released into the environment by waste incineration (van-Birgelen, 1998). Additionally, HCB was known to have a rather volatile nature (Kannan et al., 1995). The occurrence of relatively small concentrations of HCB in fish tissue in the present study might be a reflection of limited sources and the volatile nature of this compound.

3.2.4. HCHs

Extremely small concentrations of HCHs were observed in fish samples analyzed in this study, with most concentrations smaller than the limit of detection (0.01 ng/g wet wt.). Greater concentrations of HCHs were found in sediment from agricultural areas (Wu et al., 1999), and in mussel samples (2.1–110 ng/g lipid wt.) collected from the coastal waters of China (Monirith et al., 2003). In our study, the fish samples were collected mainly from the open sea, and thus smaller concentrations were expected. γ-HCH was the predominant isomer found in fish samples, probably reflecting the historical use of technical HCH mixtures.

3.2.5. Total OCs

The average concentrations of OC pesticides in fish samples are summarized (Fig. 3 and Table 2). The major contaminants in the fish samples were in the order of *p,p′*-DDE, chlordane, HCB, and *p,p′*-DDT. For different sampling locations, the greatest concentrations of OC pesticides were observed in St. 3. The concentrations of α-chlordane, dieldrin, endrin-kepone, HCB, mirex, *p,p′*-DDE, *p,p′*-DDD, *o,p′*-DDT, *p,p′*-DDT and total OCs were significantly different among the three sampling sites ($p<0.05$). Concentrations of total OCs observed in fish samples collected from St. 3 were greater than those from the other two sites (Table 2). Except for Cinnamon Flounder, the average total OC pesticide concentration in St. 3 (8.6 ng/g wet wt.) was greatest among the three stations (5.0 ng/g in St. 1 and 3.1 ng/g in St. 2). The observed inter-station differences are likely to be attributed to the proximity of St. 3 to the Chinese coast.

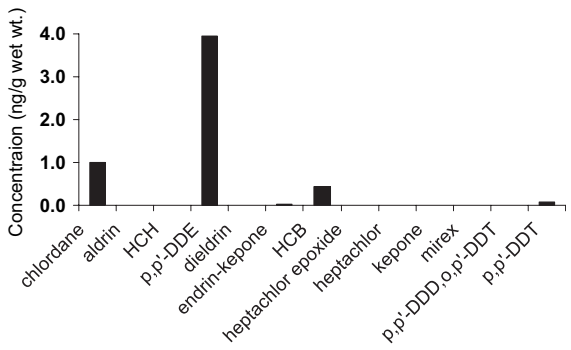

Fig. 3. Average concentrations of OC pesticides in fish samples collected from the East China Sea.

3.2.6. PCBs

In this study, a mixture of 28 PCB congeners was used as standards for the quantification of total PCBs, and thus some congeners that are potentially important in fish tissues may not have been included in the analysis. The concentrations of individual and total PCBs in fish samples collected from the East China Sea are summarized (Table 3). There were no significant differences among different fish species and sampling locations ($p > 0.05$). Total PCB concentrations ranged from 0.24 to 1.4 ng/g wet wt., which are similar to the PCB concentrations recorded in fish (0.57 ng/g wet wt.) from Shanghai, China (Nakata et al., 2002). A relatively greater average PCB concentration was detected in the Conger Pike from St. 3, while slightly smaller concentrations were detected in the White Mouth Croaker from St. 1. The Conger Pike was expected to accumulate greater amounts of lipophilic compounds, e.g. organochlorine compounds, because of its greater fat content (Larsson et al., 1990), and its habit of feeding on bottom sediments (Larsson, 1984). Moreover, owing to their lipophilicity and low biodegradability, PCBs are known to bioaccumulate along the food chain in the aquatic ecosystem (Bressa et al., 1997). The smallest concentration was detected in the Cinnamon Flounder at St. 2. The mean level of total PCBs in the fish samples was 0.71 ng/g wet wt., which points to relatively few local sources and low usage of PCBs in China. Similar results were also reported in mussel samples (2.5 ng/g wet wt.) by Monirith et al. (2003). In general, small residue concentrations of PCBs had been reported in surface water and coastal sediments in China, lending support to the general pattern of low level of PCB usage in China (Zhou et al., 2000).

3.3. Risk assessment

3.3.1. Dietary survey

Among the 160 people interviewed, the mean age was 54±14 years (range: 26–82), and more than half

Table 3
Concentrations [mean (SD) in ng/g wet weight] of polychlorinated biphenyls in fish collected from East China Sea

	St. 1				St. 2				St. 3			
	CF	WC	WP	YC	CF	WC	WP	YC	CF	WC	PE	YC
PCB101*#	0.19 (0.14)	0.19 (0.09)	0.10 (0.08)	0.17 (0.09)	ND	0.14 (0.08)	0.17 (0.04)	0.48 (0.46)	ND	0.24 (0.09)	0.52 (0.74)	0.44 (0.09)
PCB118	0.21 (0.20)	0.43 (0.35)	0.19 (0.29)	0.11 (0.04)	0.05 (0.02)	0.11 (0.08)	0.04 (0.05)	0.11 (0.08)	0.04 (0.02)	0.20 (0.03)	0.43 (0.61)	0.10 (0.12)
PCB126,187	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB128	ND	0.03 (0.06)	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB153#	ND	0.07 (0.10)	ND	ND	ND	0.03 (0.07)	ND	ND	ND	ND	0.13 (0.19)	0.03 (0.07)
PCB18*	0.08 (0.11)	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB180*#	ND	0.15 (0.13)	ND	0.06 (0.03)	0.04 (0.01)	0.09 (0.05)	ND	0.04 (0.03)	ND	0.07 (0.01)	0.12 (0.17)	0.07 (0.02)
PCB188	ND	0.05 (0.10)	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB194*#	0.02 (0.05)	0.06 (0.03)	ND	0.04 (0.02)	0.02 (0.01)	0.04 (0.01)	ND	0.06 (0.04)	ND	0.02 (0.02)	ND	0.02 (0.01)
PCB195	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB200	ND	0.02 (0.03)	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB44	ND	0.05 (0.04)	ND	ND	0.05 (0.09)	ND	0.04 (0.04)	0.03 (0.05)	0.05 (0.09)	0.03 (0.03)	0.09 (0.13)	0.02 (0.02)
PCB77*	ND	0.05 (0.06)	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB1	ND	0.03 (0.06)	ND	0.05 (0.05)	ND	ND	0.46 (0.90)	ND	0.10 (0.17)	ND	ND	ND
PCB50,28#	ND	ND	ND	0.04 (0.03)	ND	ND	ND	0.04 (0.05)	ND	0.03 (0.04)	ND	0.04 (0.02)
PCB52	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB206	0.23 (0.19)	0.05 (0.06)	0.14 (0.12)	0.07 (0.03)	0.07 (0.01)	0.06 (0.02)	0.06 (0.07)	0.09 (0.06)	0.03 (0.03)	0.07 (0.06)	0.05 (0.08)	0.17 (0.19)
PCBs	0.75 (0.11)	1.2 (0.14)	0.43 (0.09)	0.57 (0.05)	0.24 (0.03)	0.51 (0.05)	0.80 (0.22)	0.87 (0.15)	0.26 (0.05)	0.66 (0.08)	1.4 (0.23)	0.89 (0.12)

ND indicates samples with OC concentrations below detection limit. The limit of detection is 0.1 ng (0.02 ng/g wet wt.) for individual PCB, and 0.5 ng (0.09 ng/g wet wt.) for total PCBs. CF, cinnamon flounder; WC, white mouth croaker; WP, silvery pomfret; YC, small yellow croaker; PE, conger pike. *There is a significant difference ($p<0.05$) among different sampling locations. #There is a significant difference among different species.

Table 4
Daily consumption of various fish species in the Zhoushan population

Species	Daily consumption (g/person)	Percentage of total consumption (%)
Drab filefish (*Navodon septentrionalis*)	0.54	0.51
Swamp eel (*Monopterus albus*)	0.77	0.73
Freshwater fish	1.9	1.8
Cinnamon flounder (*Cynoglossus robustus*)	2.6	2.5
Cuttlefish (*Sepia esculenta* and *Loligo chinensis*)	3.4	3.2
Conger pike (*Muraenesox cinereus*)	5.9	5.6
Silvery pomfret (*Pampus argenteus*)	7.7	7.3
White mouth croaker (*Argyrosomus argentatus*) and Small yellow croaker (*Pseudosciaena polyactis*)	14	14
Giant seacatfish (*Arius thalassinus*)	16	15
Ribbonfish (*Trichiurus haumela*)	24	23
Bombay duck (*Harpodon nehereus*)	29	27
Total	105	100

(63.8%) were men. According to the dietary survey, a healthy adult ate 105±182 g fish meat each day. There was a gender-specific difference in the rate of fish consumption. Men took more fish (132 g/day) than women (57 g/day). Daily fish consumption in the coastal city of Zhoushan was greater than the average rate of consumption of marine products in China as a whole (23 g/person per day) (Chen and Gao, 1993).

In this survey, eleven species of fish were examined for individual dietary consumption (Table 4). Marine fish accounted for about 98% of total fish consumption in the coastal city, and the top four species consumed were in the order of Bombay duck (*Harpodon nehereus*), Ribbonfish (*Trichiurus haumela*), Giant seacatfish (*Arius thalassinus*), and White mouth croaker (*Argyrosomus argentatus*) and Small yellow croaker (*Pseudosciaena polyactis*).

3.3.2. Risk characterization

The oral RfD values and cancer benchmark concentrations for various organochlorines are summarized (Table 5). Relevant oral RfDs and slope factors were obtained from USEPA's Integrated Risk Information System (IRIS). It should be noted that the RfD of PCBs refers to the value of nondioxin-like PCBs, and the cancer slope factor for PCBs refers to the upper-bound slope factor for the food chain. Comparing the cancer benchmark concentrations derived from a coastal population in China (this study) with those from USEPA (Dougherty et al., 2000), almost all cancer benchmark concentrations for the coastal population were lower than the USEPA's criteria/values. The major reason for this was the higher daily fish consumption rates in the Chinese coastal city as compared to its counterpart in the USA. The cancer benchmark concentration for PCBs for the Chinese coastal population was higher because the upper-bound slope factor was applied for screening purpose.

The 50th and 95th centile concentrations were not estimated for aldrin and HCHs as the concentrations of these two chemicals were below detection in many of the fish samples. The 95th centile concentration for DDTs (DDT and its metabolites) was greatest among the organochlorines (12.9 ng/g wet wt.), followed by chlordane, total PCBs, and HCB. Concentrations of all of the organochlorine compounds in fish were less than their corresponding oral RfD values. Similar results were found in a previous study undertaken in the Chinese cities of Dalian and Shenyang, which reported that the daily intakes of HCHs and DDTs did not exceed the tolerable daily intake (TDI), 20 μg/kg per day, proposed by the World Health organization (WHO) (Kunisue et al., 2004).

An evaluation of the noncancer and cancer risks to human health associated with the consumption of fish containing OC contaminants was undertaken and the results are summarized (Figs. 4 and 5). The HRs of noncancer risk based on 50th and 95th centile concentrations were all less than unity (Fig. 4), due to the relatively small concentrations of OCs in the fish tissue (Table 2). Similarly, a mussel-based health risk assessment in coastal cities in China also indicated that the non-cancer risk quotients (RQs) for DDTs, CHLs and PCBs were all less than 1.0 (Fung et al., 2004).

In contrast, the cancer risks associated with fish consumption based on 95th centile concentrations of HCB, dieldrin, chlordane, DDTs and PCBs were all

Table 5
National average exposures and benchmark concentrations for contaminants in fish

Contaminants	Oral RfD (μg/kg day)	Cancer slope factor [per (mg/kg/day)]	Cancer benchmark concentration (μg/kg day)	50th centile measured concentration (50th MEC) (ng/g wet wt.)	95th centile measured concentration (95th MEC) (ng/g wet wt.)
HCB	0.80	1.6	0.00036	0.13	2.5
Dieldrin	0.05	16	0.00004	0.01	0.05
Chlordane	0.06	0.35	0.0016	0.51	3.5
DDTs	0.50	0.34	0.0017	2.7	12.9
PCBs	0.02	2.0	0.00029	0.45	2.8

Oral RfDs and cancer slope factors were obtained from USEPA's Integrated Risk Information System (IRIS).

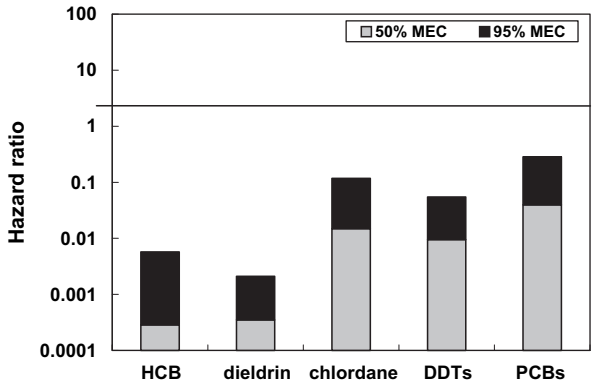

Fig. 4. Noncancer hazard ratios for daily fish consumption by people in Zhoushan. MEC, measured concentration.

greater than unity, suggesting that daily exposure to these contaminants due to fish consumption had a lifetime cancer risk of greater than one in one million. An estimation of the lifetime cancer risk based on the 50th centile concentrations of DDTs and PCBs were also greater than unity. The results indicate that certain contaminants, namely DDTs, PCBs, and chlordane, may be of particular concern. In this study, the limits of detection were used in estimating the exposure concentrations for fish tissue samples containing concentrations of specific contaminants below detection limits, thus the exposure concentrations may have been overestimated. This coupled with the use of a cancer risk of one in one million in calculating the benchmark concentration (Dougherty et al., 2000) may lead to an over-conservative risk assessment. It is believed that such a conservative approach may be appropriate as an initial screening of potential risks.

There are a number of important limitations in this study. For example, this investigation did not consider (1) potentially different risks to separate age groups; (2) possible interactions among various toxic chemicals; (3) risks to populations residing in areas close to sources of contamination. Overall, despite the limitations associated with the analysis, the assessment undertaken indicates a potentially high cancer risk due to OC contaminants in fish, and represents an important step toward a more comprehensive understanding and evaluation of human health risks associated with organochlorine exposures via marine fish consumption in coastal cities in China. With an established dietary database for a Chinese coastal city, more comprehensive risk assessment can be conducted when contaminant concentrations in different food types become available. Research along this line is currently in progress.

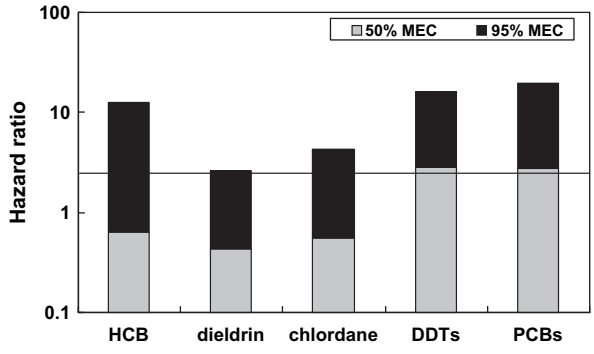

Fig. 5. Cancer hazard ratios for daily fish consumption by people in Zhoushan. MEC, measured concentration.

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