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Organochlorines in fish from the coastal coral reefs of Weizhou Island, south China sea: Levels, sources, and bioaccumulation

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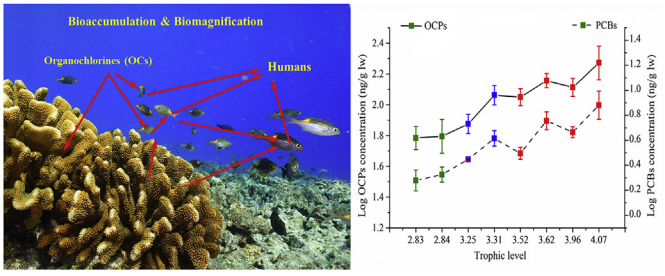
HIGHLIGHTS

- We studied OCs in the coastal coral reef ecosystems of the South China Sea. .
- Organochlorines from historical agricultural sources persisted in the fish muscle.
- The TMFs for organochlorines were lower than those from high latitude regions.
- Consumption of the coral reef fish may not pose health risk to human at present.

GRAPHICAL ABSTRACT



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ABSTRACT

Eight fish species were sampled from a coastal coral reef ecosystem near Weizhou Island, South China Sea, to investigate the composition profiles and bioaccumulation of organochlorines (OCs). The total concentrations of 18 organochlorine pesticides (OCPs) and 22 polychlorinated biphenyls (PCBs) were found to be 26.5–452 ng/g lw and 0.87–19.8 ng/g lw, respectively. The contaminant distribution pattern indicated that agrochemical sources were more important than industrial sources, and that historical residues remain the primary source of OCs in Weizhou Island. Bioaccumulation factors (BAFs) indicated that dichlorodiphenyltrichloroethanes was bioaccumulating with log BAFs ranging from 3.53 to 5.21. Some congeners diverged from the general trend predicted by the logarithm octanol-water partition coefficient (log K_{ow}); this was mainly attributable to differences in the bioaccumulation potentials of these congeners in the studied samples. Trophic magnification factors demonstrated that aldrin, endrin, and dieldrin undergo significant trophic dilution, while the other six OC compounds undergo trophic magnification in the food chain. The presence of OCP congeners was also probably affected by their metabolism in fish tissues. The estimated daily intakes of OCPs via fish consumption by residents ranged from 0.05 to 5.45 ng/kg body weight/day, which is below the acceptable daily intake recommended by the FAO/WHO.

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1. Introduction

Organochlorine compounds (OCs), including polychlorinated

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biphenyls (PCBs) and organochlorine pesticides (OCPs), are a group of persistent organic pollutants (POPs) of global concern owing to their persistence, bioaccumulation, and potential negative impacts on humans and wildlife (Zhang et al., 2007). OCPs were used globally in agricultural and industrial products prior to being banned under the Stockholm Convention (2001), but are still used in some developing countries owing to their low cost and versatility (Xia, 2011). Although PCBs have been banned worldwide for several decades, they remain a problem due to leakage, illegal disposal, and historical residues in sediments (Lakshmanan et al., 2010). China is a 2001 signatory to the Stockholm Convention and has made considerable achievements in pollution control and reduction in past decades. However, as a significant producer, user, and emitter of POPs, China must still control emissions, preserve waste POPs, and contend with environmental and health risks. OCs have been found in different aquatic environments, including river water, sludge, and surface sediments, and in aquatic organisms (Mai et al., 2005; Yang et al., 2013; Robinson et al., 2016; Sun et al., 2017). In aquatic systems, fish are exposed to pollutants via either their gills and/or diet. Since OCPs and PCBs have high octanol-water partition coefficient (K_{ow}) values, they can be bio-accumulated into higher trophic levels via the food chain, and eventually ingested by humans, posing adverse effects to health.

Coral reef ecosystems are hotspots of marine ecology. Currently, there are 632 coral reef fish species in Xisha Archipelago and Zhongsha Archipelago in the South China Sea (SCS), accounting for 5.27% of the world's 12,000 species of marine fish (Li et al., 2006). Studies have shown that a healthy coral reef ecosystem produces 35 tons of fish per square kilometer per year, and 10% of the global fisheries production comes from coral reefs (Yu, 2018). At every trophic level, reef fish reflect the ecological status of coral reefs. Sun et al. (2014) reported that the levels of PCBs and dichlorodiphenyltrichloroethanes (DDTs) in the muscles of coral reef fish from Yongxing Island, SCS, were at the lower end of the global range, with concentrations of 6.3–199 and 9.7–5831 ng/g lw (lipid weight). Compared with offshore coral reef areas, the levels of many contaminants in coastal coral reef waters are usually higher (Guo et al., 2017; Zhang et al., 2018). Therefore, the level of OCs might also be higher, increasing the food risk to humans compared to offshore fish.

Weizhou Island is a typical coastal coral reef ecosystem in the northwestern part of the SCS. It is famous as a tourist area, having attracted about 600,000 visitors annually in recent years, with corresponding anthropogenic impacts (Yu, 2018). It is also the northernmost coral reef ecosystem in China and has significant scientific and ecological value. Weizhou Island is situated off the coasts of both China and Vietnam, which are considered potentially significant sources of POPs (Zhang et al., 2007). Historically, both countries used DDTs in applications in high quantities, with 45,900 tons used in China from 1950 to 2003 and 24,042 tons used in Vietnam from 1957 to 1990 (Hu et al., 2007; Minh et al., 2008). Studies indicate that consumption of bass taken from Fangchenggang (a coastal city in the northern area of the SCS) could pose a potential risk to human health (Pan et al., 2016).

Though a great deal of research has investigated marine fish in the SCS, most studies have focused on offshore fish, and coastal populations are less well considered. In this study, eight common coastal coral reef fish species, five of which are of high local food value, were sampled for OCs. We aimed to investigate the levels and composition profiles of the target OCPs and PCBs in these fish, to analyze the bioaccumulation and trophic magnification of OCs, and to estimate the health risk to local residents posed by PCBs and OCPs via fish consumption.

2. Materials and methods

2.1. Sample collection and preparation

A total of 46 fish samples and six seawater samples were collected in April 2018 in the coral reef regions of Weizhou Island (Fig. S1). Eight species of fish were sampled: *Chaetodon octofasciatus* (C.O., n = 5), *Labracinus cyclophthalmus* (L.C., n = 6), *Cephalopholis boenak* (C.B., n = 7), *Diploprion bifasciatum* (D.B., n = 6), *Sebastiscus marmoratus* (S.M., n = 4), *Selaroides leptolepis* (S.L n = 6), *Terapon theraps* (T.T n = 6), *Siganus fuscescens* (S.F n = 6) (Fig. S2). Five of these species (C.B T.T S.L S.M S.F.) have high value as local food ingredients. The length and weight of each fish were measured after collection (Table S1). All fish were then stored at −20 °C. Water samples were immediately filtered after being transported to the laboratory.

Muscle tissue without skin was obtained from between the vent and pectoral fin, then lyophilized, ground to a fine powder for homogenization, wrapped in aluminum foil, and stored at −20 °C prior to analysis. The muscle moisture content was determined gravimetrically during lyophilization (Table S1).

2.2. Analytical procedures

Seawater samples were extracted according to Streets et al. (2006) with minor modifications. Each seawater sample (200 L) was passed through glass fiber filters (0.7 μm, 90 mm, Whatman) and two tandem pre-cleaning polyurethane foam columns (diameter 60 mm, length 100 mm, density 142 mg/cm^3) at 1 mL/min, as described.

Fish samples were analyzed according to Sun et al. (2014). Briefly, 1 g of dried fish muscle was spiked with 20 ng of 2, 4, 5, 6-tetrachloro-m-xylene (TCMX), PCB30, and PCB204 as surrogate standards, then Soxhlet-extracted with a mixture of n-hexane and acetone (1:1, v:v) for 36 h. The extraction was concentrated to 4 mL and solvent-exchanged to n-hexane. About 0.5 mL of the extract was used for the gravimetric determination of lipid content. The remaining extract was purified using a permeation chromatography gel column to remove lipids, and further cleaned on a multilayer silica column packed with neutral alumina (3 g, 3% water deactivated), acid silica gel (3 g, 50% sulfuric acid), and anhydrous sodium sulfate (1.5 g), from bottom to top. The column was eluted with 20 mL of a mixture of n-hexane and dichloromethane (1:1, v:v). The extract was concentrated to 100 μL under a gentle nitrogen stream and 20 ng $^{13}C_{12}$-PCB138 was added as an internal standard before analysis.

Forty OC congeners were selected as target analytes and quantitatively analyzed using an Agilent 7890B gas chromatograph-7000C tandem mass spectrometry (GC-MS/MS) system in an electron impact ionization (EI) mode. These included six DDTs (o,p', p,p'-DDD, -DDE, -DDT), four hexachlorocyclohexanes (HCHs: α, β, γ, δ-HCH), three drins (aldrin, endrin, dieldrin), chlordanes [CHLs: heptachlor, trans-chlordane (TC) and cis-chlordane (CC)], p,p'-methoxychlor (MXC), hexachlorobenzene (HCB), and 22 PCBs (numbers 18, 28, 44, 52, 66, 77, 81, 101, 105, 114, 118, 123, 126, 128, 138, 153, 157, 169, 170, 180, 187, and 189). An HP-5MS GC column (30 m × 0.25 mm × 0.25 μm, Agilent Technologies Inc.) was used to separate the different analytes. The column oven temperature was programmed as follows: 80 °C for 0.5 min, increasing to 160 °C at a rate of 20 °C/min followed by 4 °C/min to 240 °C for 2 min, then increasing to a final temperature of 295 °C at a rate of 20 °C/min for 2 min. Helium was used as the carrier gas. One μL of sample bulk was injected at a port temperature of 250 °C.

2.3. Quality assurance and quality control

Standardization of instruments was carried out daily using calibration standards. Procedural blanks, spiked blanks (n = 4), and spiked matrices (n = 4) were set to ensure method quality control. The spiked compounds included 18 OCP congeners and 22 PCB congeners. Each set of 12 sample analyses was accompanied by a procedural blank, in which no target OCs were detected. The average recoveries of the three surrogates in all samples were 68.6 ± 10.3%, 72.3 ± 8.4%, and 92.4 ± 0.78% for TCMX, PCB30, and PCB209, respectively. Recovery of OC standards was 73.6–94.1% in the spiked blanks and 79.2–101.5% in the matrix-spiked samples, with a relative standard deviation of <15%. The reported concentrations were not adjusted based on the surrogate recoveries. The limit of detection (LOD) and limit of quantification (LOQ) for the OCs were defined as the concentrations corresponding to a signal-to-noise (S/N) ratio of 3 and 10, respectively. The LODs and LOQs for the fish samples were 0.006–0.155 ng/g lw and 0.021–0.515 ng/g lw, respectively, while they for the seawater were 0.03–0.775 pg/L and 0.105–2.575 pg/L, respectively.

3. Results and discussion

3.1. OC levels

3.1.1. Seawater

The dissolved phase seawater concentrations of individual OCP congeners were higher than those for individual PCB congeners (Table S2). In the seawater samples, the average OCPs value was an order of magnitude higher than the average PCBs value (Table 1). The OCP and PCB content in the study region was lower than those in most of China's coastal seawaters (Luo et al., 2010), and higher than or similar to those in some of the offshore seawaters (Lohmann et al., 2009; Zhang et al., 2012).

3.1.2. Fish

Detection frequencies of OCPs in all samples ranged from 92.1 to 100% of the total OCPs content. DDTs had the highest median level in all fish, followed by MXC, HCB, HCHs, and other OCPs (Fig. S3 and Table S3). DDTs and MXC were the dominant contaminants in all fish. The DDTs concentrations were one to two orders of magnitude higher than the HCHs concentrations, and generally higher than the PCBs concentrations (Table S2). MXC was detected in all samples with concentrations ranging from 8.7 to 142 ng/g lw. DDTs concentrations recorded in this study were higher than those in fish collected from Natuna Island (Hao et al., 2014), but slightly lower than those in fish collected from the Marshall Islands (Wang et al., 2011) and marine fish from coastal China (Qiu et al., 2005; Guo et al., 2010; Pan et al., 2016). The residual levels of the other OCPs (HCB, HCHs, Drins, and CHLs) determined in the present studysite were relatively low compared to other coastal areas, including the Nansei Islands, Norwegian coast, Aleutian Islands, and Pearl River Delta (Table S4). The relatively high content of DDTs and MXC in fish may be related to fishery and agricultural activities in the region. The high DDTs level could also be due to its extensive historical utilization and high lipophilicity (Xia, 2011). Antifouling paints used on fishing boats contain technical DDT at around 500 μg/g, making them a valuable source of contamination (Li et al., 1998; Lin et al., 2009). Furthermore, MXC has been widely used as a DDTs substitute in agricultural pest control.

Table 1
Organochlorine concentration in seawater and fish samples from Weizhou Island, SCS.

	Seawater (pg/L)		Fish (ng/g lw)	
	Median (range)	Mean ± SD	Median (range)	Mean ± SD
DDTs	56.7 (35.7–72.7)	55.9 ± 12.1	54.0 (7.70–401)	75.5 ± 77.3
MXC	nd [a]	nd	30.3 (8.66–142)	40.2 ± 26.7
HCB	46.8 (38.0–77.3)	51.6 ± 16.0	2.2 (0.77–5.12)	2.55 ± 1.14
HCHs	59.4 (56.1–76.8)	64.1 ± 9.61	1.6 (1.12–2.61)	1.66 ± 0.36
Drins	349 (271.9–440)	352 ± 58.2	1.6 (1.04–4.59)	1.67 ± 0.54
CHLs	380 (349–526)	403 ± 65.7	1.5 (1.04–2.40)	1.53 ± 0.30
OCPs	**888(818~1174)**	**927 ± 126**	**111(26.5~452)**	**123 ± 80.9**
PCBs	**51.8(48.3~89.5)**	**59.9 ± 16.1**	**3.5(0.87~19.8)**	**4.5 ± 3.66**

[a] Non-detected. In this study, for targets with concentrations below LOD, was set to non-detected; if that below LOQ, a value of 1/2 LOQ was used for data analysis.

Detection frequencies of PCBs in all samples were 76.1–100%. Compared to other studies undertaken worldwide, the PCB levels observed in fish sampled in this study were lower than those in fish collected from Natuna Island, the Marshall Islands, and elsewhere in Asia (Wang et al., 2011), Europe, and North America (Johnson-Restrepo et al., 2005; Naso et al., 2005; Storelli et al., 2009; Hardell et al., 2010; Greenfield and Allen, 2013). These results indicated that PCBs concentrations in fish from Weizhou Island were at the low end of the global range (Table S4). This may be due to the limited historical use of PCBs in southwestern China and Vietnam (Sun et al., 2014); both regions are less developed than other areas of China, with limited industrial activity.

High concentrations of OCPs and PCBs were present in carnivorous fish species, which could be attributable to their different diets and living habits. Among the species studied, the levels of traditional OCs (except MXC) were significantly higher in carnivorous than in herbivorous or omnivorous species ($p = 0.026$) (Fig. S4). S.M. is a bottom-dwelling fish inhabiting muddy or sandy environments, which may increase its exposure to contaminants in the benthic sediment. Previous studies have also reported that feeding and living habits can influence the OC levels in fish (Adu-Kumi et al., 2010; Sun et al., 2014; Robinson et al., 2016). Furthermore, the concentration of OCPs in all fish samples was significantly higher than the corresponding PCB concentrations, indicating that agrochemical sources of pollution were probably more important than industrial sources in the study area.

3.2. Source analysis of OCs in fish samples

The highest contributions of DDTs were found to be from p,p'-DDE (67.9%) and p,p'-DDD (14.0%) followed by p,p'-DDT (13.0%), o,p'-DDD (2.43%), o,p'-DDT (2.04%) and o,p'-DDE (0.63%) (Fig. 1a). Different combination ratios of parent compounds and their metabolites can indicate possible sources of DDTs. The expression (DDE + DDD)/DDTs > 0.5 can be interpreted as indicating DDTs derived from historical use rather than from new input (Yu et al., 2014). In present study, this ratio in fish ranged from 0.79 to 0.95, indicating that historical residues were the main source of DDTs around Weizhou Island. DDE is a compound formed by the elimination of hydrogen chloride by DDT and is one of the main metabolites of DDT. The ratio o,p'-DDT to p,p'- DDT can be used to distinguish DDTs pollution caused by technical DDT from Dicofol (Qiu et al., 2005). Zhou et al. (2006) report the major components of technical DDT to be p,p'-DDT (75%), o,p'-DDT (15%) and p,p'-DDE (5%), with other metabolites contributing < 5%. The values of o,p'-DDT/p,p'-DDT range from 0.2 to 0.3 in technical DDT and 1.3 to 9.3 or higher in Dicofol (Qiu et al., 2005; Yu et al., 2014). Since the mean value of the ratio in this study is 0.22, technical DDT was probably the main contributors to DDTs in this study area.

The eight fish species presented similar HCH congener patterns (Fig. 1b). The compositional profile of HCHs showed β-HCH (44.09%) to be dominant, followed by α-HCH (26.79%), γ-HCH (16.17%), and δ-HCH (12.95%); this result differs from the compositional pattern of HCHs in bass from Fangchenggang, China (Pan

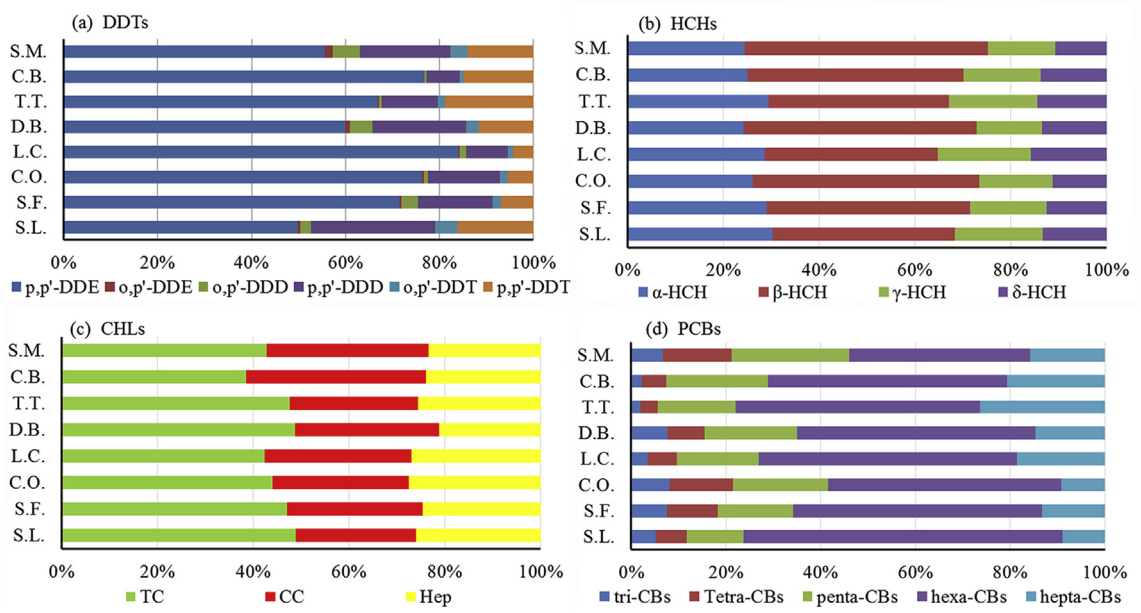

Fig. 1. Compositional profile of different organochlorines in coral reef fish from Weizhou Island.

et al., 2016). The ratio α-HCH/γ-HCH is usually relatively stable, with a value of 4.64–5.83 for technical HCHs, and nearly zero for lindane. However, this ratio may change, since α- and γ-HCH can transform to β-HCH, which shows more stability and poorer degradation and metabolic capacity than other HCH congeners in the environment (Yu et al., 2014). Therefore, the β-HCH detected in the fish samples probably reflects the historical use of technical HCH mixtures and their transformation from homologs. The α/γ-HCH ratios of all coral reef fish samples from Weizhou Island were in the range of 1.47–1.81, indicating that they may be derived from industrial residues and a possible source of lindane (Yu et al., 2014; Pan et al., 2016).

CHL patterns showed a comparatively higher contribution of TC at 44.8%, followed by CC (30.3%) and heptachlor (24.9%) (Fig. 1c). Low concentrations of HCB in the fish muscle may be attributed to its past application as a fungicide and its emission as a byproduct during the manufacturing process and usage of various industrial and agrochemicals (Adu-Kumi et al., 2010; Ameur et al., 2013). Moreover, HCB has a volatile nature that accounts for its transport in the environment (Kannan et al., 1995).

PCB homologs are classified according to the degree of chlorination in fish (Fig. 1d). Hexa-CB (50.06%) was the predominant homologue in all fish samples, followed by penta- (19.54%), hepta- (17.96%), tetra- (7.86%), and tri-CBs (4.58%), except for in L.C. and T.T., in which the contribution of hepta-CBs was greater than penta-CBs. A substantial accumulation of hexa- and penta-CBs may indicate extensive use of Aroclor1254 and 1260, since more highly chlorinated homologs are resistant to metabolism. PCB153 and PCB138, in particular, are likely to accumulate in fish (Storelli et al., 2009). This distribution pattern was consistent with previous studies for coral reef fish from the SCS (Hao et al., 2014; Sun et al., 2014) and coastal fisheries in China (Pan et al., 2016).

3.3. Bioaccumulation and trophic magnification factors of OCs in coral reef fish

In order to investigate the degree of bioaccumulation of OCs in coral reef fish from the seawater, apparent bioaccumulation factors (BAFs) were calculated by dividing the concentration of OCs in fish by that in the ambient seawater (based on the median value). According to the European Chemicals Agency, a chemical is bioaccumulative if its BAF ≥5000 L/kg (log BAF ≥ 3.7) and potentially bioaccumulative if 2000 ≤ BAF < 5000 L/kg (3.3 ≤ log BAF < 3.7) in biological samples (European Chemicals Agency, 2012). The results of the estimation of the log BAFs are shown in Fig. 2 and Table S5.

According to these criteria, DDTs are bioaccumulative, with log BAFs ranging from 3.53 to 5.21. OCPs are also potentially bioaccumulative; log BAFs of the other OCs were lower than 3.3. However, OCPs and PCBs are bioaccumulative in some fish samples. The fish in this study generally presented low BAFs compared with fish in the north of Guangzhou, South China (log BAFs: 1.2–8.4) and in lake trout from Lake Michigan (log BAFs: 5.5–8.5) (Streets et al., 2006; Wu et al., 2008). The cause of this discrepancy is likely to be variations in metabolic capacities among species and

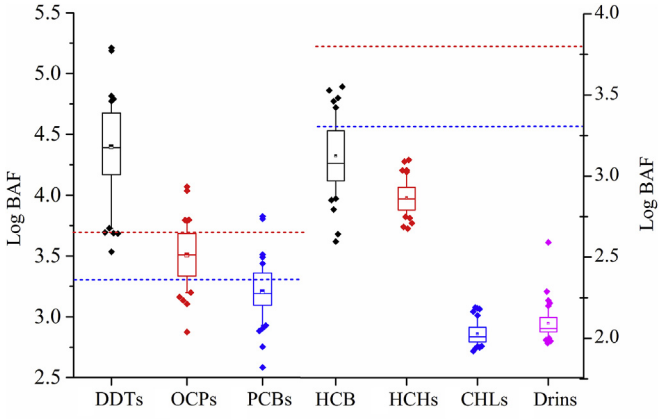

Fig. 2. Bioaccumulation factors (BAFs) of organochlorines in coral reef fish from Weizhou Island, South China Sea (box: 25–75 percentiles; whiskers: 10–90 percentiles; line: median; square: mean; point: outlier). Log BAF values are given in Table S4. A chemical is bioaccumulative if its log BAF is greater than 3.7 (red dotted line), and potentially bioaccumulative if its log BAF is between 3.3 (blue dotted line) and 3.7. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

environmental conditions studied, such as higher water temperature, which could enhance the metabolic rates of OCs in fish (Buckman et al., 2007). In addition, BAF calculations assume a steady state for OCs in apportioning between fish and the surrounding water environment (Tsui et al., 2017). Unknown conditions affecting a steady state could introduce uncertainties to the BAF values reported here.

A correlation analysis revealed a multi-linear relationship between the log BAFs and the logarithm octanol-water partition coefficient (log K_{ow}) of chemicals. Log BAF values for OCP congeners in fish ranged from 0.99 to 4.93. The log BAFs of OCs increased with increasing K_{ow} at log $K_{ow} < 7$ (Fig. 3a). Log BAFs and log K_{ow} for OCPs had a significant positive correlation ($p = 0.048$) when α-HCH and β-HCH were excluded, but the correlation was not significant when they were included. This may be due to differences either among residual levels of OCP congeners in seawater, or in the bioaccumulation potentials of OCP congeners in studied samples (Guo et al., 2017). A highly significant relationship ($p < 0.001$) was found in the plot of log BAFs versus log K_{ow} for PCB congeners, which has also been observed in previous studies (Wu et al., 2008; Wang et al., 2011). This is because PCB congeners with a higher chlorine content have higher biomagnification capabilities, due to their high log K_{ow}, while PCB congeners with low a chlorine content are more easily metabolized or excreted.

Trophic magnification factors (TMFs) for the seven OC compounds were determined based on the slope of regression between log-transformed concentrations of OCs in fish and trophic levels of the corresponding species (Fig. 4 and Table S6). Linear regression analysis indicated a significant positive correlation between trophic levels (TLs) in coral reef fish communities and normalized concentrations of the five compounds (except MXC and Drins) ($p < 0.01$). MXC was also magnified, but not significantly so ($p = 0.396$). The TMF values of the six compounds ranged from 1.18 to 4.54, indicating biomagnification due to predation. Nevertheless, negative relationships were observed for correlations between the TL value and concentration of Drins ($p = 0.196$) and the TMF = 0.90, indicating that Drins undergoes no biomagnification in the food web. The obtained TMFs for OC congeners ranged from 0.65 (CB169) to 6.07 (p, p'-DDT). Although the TMFs for CB169 and aldrin were less than 1, this did not affect trophic biomagnification of the OCPs and PCBs. One study has shown that latitude is a determinant for TMFs, which are higher in arctic aquatic ecosystems; moreover, biodilution, excretion rate, and food web complexity may be essential factors in this phenomenon (Borga et al., 2012). The present study seems to support these findings through comparisons with different latitude studies. TMFs for p,p'-DDE were lower compared with those from high latitudes, which were found to be 5.6 in the Yellow Sea (Byun et al., 2013), 11 in the Arctic Sea (Hallanger et al., 2011), and 14 in Northwater Polynya (Fisk et al., 2010). Similarly, TMFs for PCB congeners were also lower than those in marine food webs of high latitude regions, such as Northwater Polynya (1.7–10.7; Fisk et al., 2010) and Hudson Bay (2.9–11; Kelly et al., 2008). Besides, TMFs for a given pollutant are widely variable across studies, depending on factors, such as food web composition, trophic levels, energetic requirements of organisms, and freshwater versus marine systems (Fisk et al., 2010; Nfon et al., 2008; Walters et al., 2011).

A significant positive correlation ($p = 0.01$) was observed between TMFs and log K_{ow} values of OC congeners (Fig. 3b). A similar relationship was reported by Fisk et al. (2010). Most OCP congeners behave as biomagnified; meanwhile, aldrin was equally likely to be biodiluted, with log $K_{ow} = 6.5$ and TMFs = 0.66. The more significant variability in the relationship among OCP congeners may be due to metabolism or formation. For example, DDE is a compound formed by the elimination of hydrogen chloride by DDTs, which then decreases TMF for DDT and increases TMF for DDE. This study found that TMF for DDE (11.1) was higher than for DDT (10.3). Inaccurate log K_{ow} values might also contribute. A significant parabolic curve was observed between TMFs and log K_{ow} ($p < 0.05$). Negative relationships were observed between TMF and log K_{ow} values of higher chlorinated congeners. The measurement of TMF assumes that sampled organisms are at a steady state in the ecosystem (Burkhard et al., 2013). However, PCB concentrations in coral reef fish in this region may not reach a steady state, due to heavy shipping and fishing-activities. As such, TMFs of more highly chlorinated congeners with high log K_{ow} values take longer to reach a steady state and may not increase with increasing log K_{ow} values.

3.4. Assessment of human dietary exposure risks

Fish consumption has become the predominant point of human exposure to POPs, and it has been confirmed that fish had have higher OC levels than any other food category (Yim et al., 2005). Since fish are an essential source of protein for many coastal

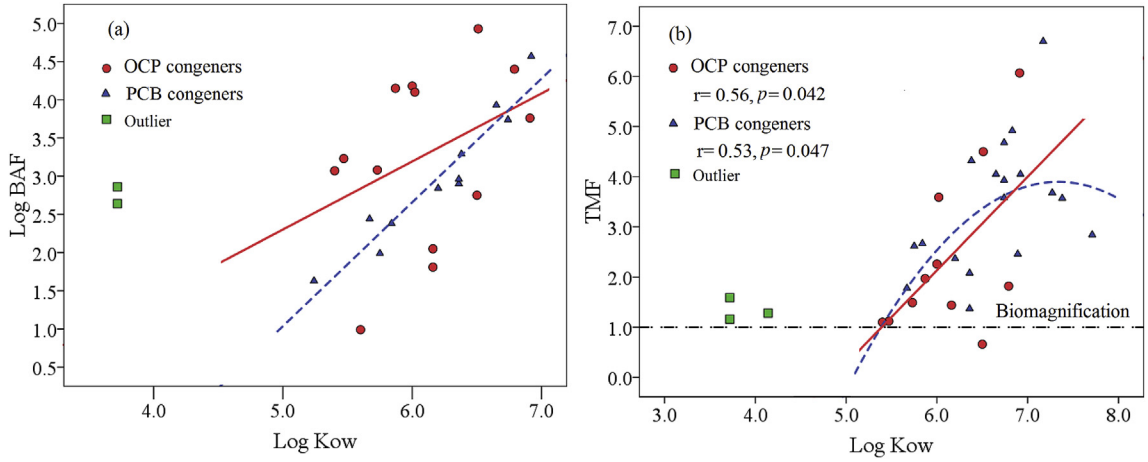

Fig. 3. (a) Log bioaccumulation factor (BAF) versus log K_{ow} for OCP and PCB congeners in coral reef fish. (b) Relationships between trophic magnification factors (TMFs) and log K_{ow} values of the partial OCP and PCB congeners (not counting p > 0.05). Red solid and blue dashed lines are the fit curves for OCP congeners and PCB congeners, respectively; green squares represent outliers and are not in the statistical range. Log $K_{ow} < 5.0$ are represented by green squares with and log $K_{ow} \geq 5.0$ is represented by other points. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

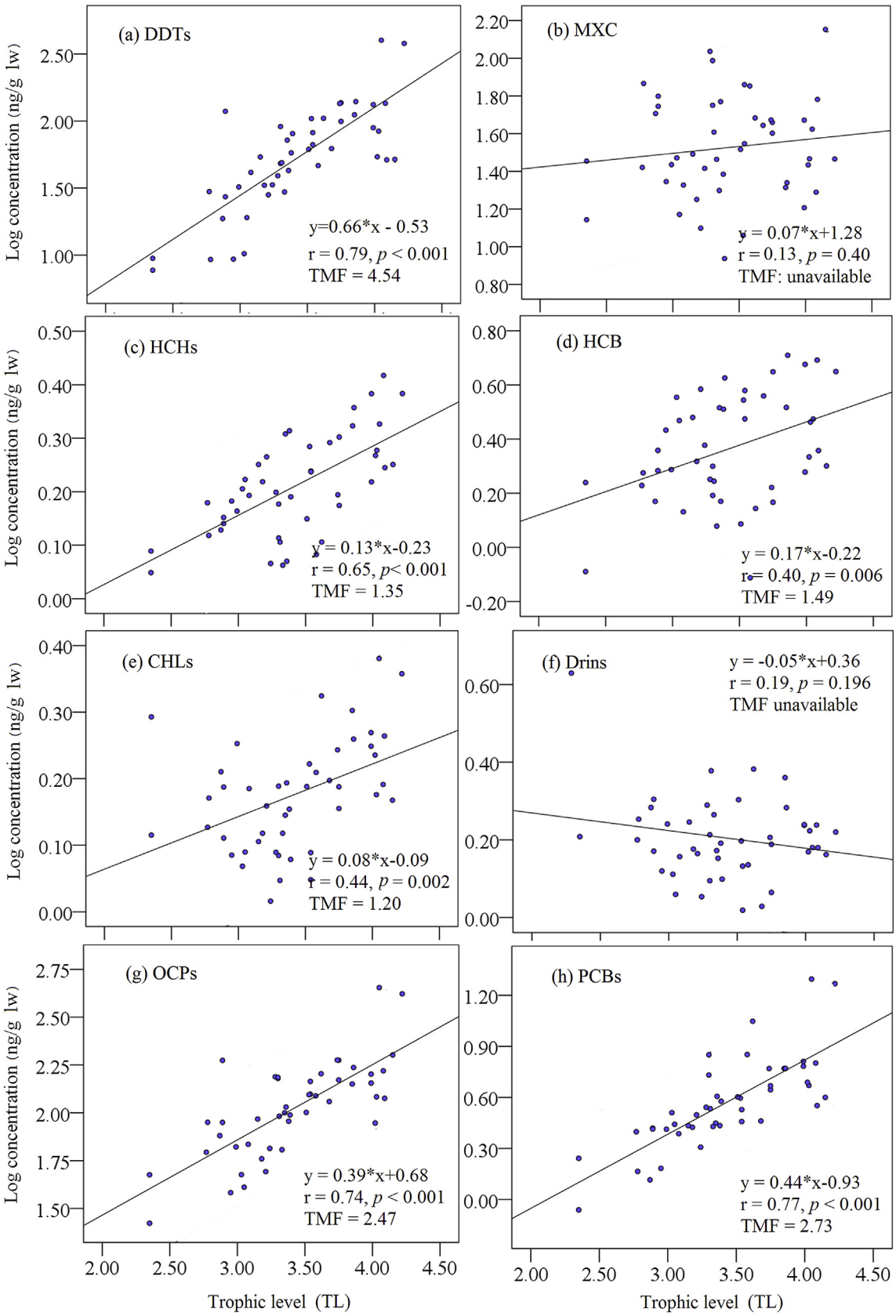

Fig. 4. Relationships between trophic level and log-transformed lipid weight normalized concentrations describing trophic magnification for (a) DDTs, (b) MXC, (c) HCHs, (d) HCB, (e) CHLs, (f) drins, (g) OCPs, and (h) PCBs in the food chain of coral reef fish from Weizhou Island.

residents, such as in Weizhou Island, an assessment of their risk of consumption is warranted.

The measured water contents of muscles in all samples was within 75.0 ± 3.52%. Based on this, data expressed on a lipid weight basis could be used to estimate wet weight content. Using the assessment method described in Text S3 and S4, this study estimated daily intakes (EDIs) for OC compounds following the consumption of edible coral reef fish (Table S7). The EDIs of OC compounds were two to five orders of magnitude lower than the respective recommended acceptable daily intakes (ADIs); indicating the minimum risk posed by these pollutants.

Based on the EDI value and corresponding ADI, hazard quotients (HQ) of OCP compounds were calculated for residents and used to assess potential human health risks from dietary exposure to OCPs (Table S7). A previous study has suggested that HQ < 0.01 indicates a negligible risk, HQ ≥ 0.01 indicates a considerable risk, and HQ > 0.05 is considered a distinct risk (Vragovic et al., 2011). The highest HQ in this study was HCB in C.B. at 3.1×10^{-3}, indicating a negligible risk. This assessment suggests that the consumption of coral reef fish on Weizhou Island might not pose a significant health risk. However, the present study did not systematically investigate daily fish consumption, and OC congeners other than those targeted in the present study may be present in the fish as well. Therefore, long-term comprehensive monitoring for OCs is warranted.

4. Conclusions

The level of OC compounds in coral reef fish from Weizhou Island is at the lower end of the global range, though they have a widespread occurrence and continuous accumulation. OC concentrations showed significant interspecies differences, possibly because of their different food item and living habits, and this would warrant further investigation in future studies. The contaminant distribution pattern indicated that agrochemical source is more important than the industrial sources, and historical residues remain the primary source of OCs. Both BAFs and TMFs may be affected by warmer water temperatures enhancing their metabolism in fish. In addition, intense human activities mean that OC concentrations in the coral reef fish of the region may not reach a steady state. This may introduce uncertainty into the BAF and TMF values reported here. Therefore although the EDIs of the OCs measured in this study suggest that consumption of these fish does not pose a significant health risk to coastal residents of South China, long-term comprehensive monitoring for OCs is still warranted.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2019.05.199.

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