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Organochlorine pesticides (OCPs) in corals and plankton from a coastal coral reef ecosystem, south China sea

Yaru Kang [a,1], Ruijie Zhang [a,b,*,1], Kefu Yu [a,b,**], Minwei Han [a], Jiying Pei [a], Zhenghua Chen [a], Yinghui Wang [a]

[a] Guangxi Laboratory on the Study of Coral Reefs in the South China Sea; Coral Reef Research Center of China; School of Marine Sciences, Guangxi University, Nanning, 530004, China

[b] Southern Marine Science and Engineering Guangdong Laboratory (Zhuhai), Zhuhai, 519080, China

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ABSTRACT

Recent studies have indicated that coral mucus plays an important role in the bioaccumulation of a few organic pollutants by corals, but no relevant studies have been conducted on organochlorine pesticides (OCPs). Previous studies have also indicated that OCPs widely occur in a few coral reef ecosystems and have a negative effect on coral health. Therefore, this study focused on the occurrence and bioaccumulation of a few OCPs, such as dichlorodiphenyltrichloroethanes (DDTs), hexachlorobenzene (HCB) and *p,p′*-methoxychlor (MXC), in the coral tissues and mucus as well as in plankton and seawater from a coastal reef ecosystem (Weizhou Island) in the South China Sea. The results indicated that DDTs were the predominant OCPs in seawater and marine biota. Higher concentrations of OCPs in plankton may contribute to the enrichment of OCPs by corals. The significantly higher total OCP concentration ($\sum_8$OCPs) found in coral mucus than in coral tissues suggested that coral mucus played an essential role in resisting enrichment of OCPs by coral tissues. This study explored the different functions of coral tissues and mucus in OCP enrichment and biodegradation for the first time, highlighting the need for OCP toxicity experiments from both tissue and mucus perspectives.

1. Introduction

Organochlorine pesticides (OCPs), which are intensively used for pest control, have been of worldwide concern because of their long-range transport, persistence, bioaccumulation tendency, and biotoxicity (Zhang et al., 2007). Dichlorodiphenyltrichloroethanes (DDTs), Hexachlorobenzene (HCB), and *p,p′*-methoxychlor (MXC) listed under the Stockholm Convention are three representative OCPs. From 1950 to 1983, the total mass of DDTs (0.4 million tons) used in China accounted for 20% of the global usage during the same period (Hua and Shan, 1996). HCB was first introduced in 1933 as a fungicide, and agricultural uses acoounted for the highest amount of HCB emissions during the 1950s and 1960s (Courtney, 1979). MXC, utilized as a replacement for DDT, has been restricted in several countries for over 15 years (UNEP, 2021). Both DDTs, HCB, and MXC are easily bioaccumulated in the food web due to their strong lipophicity, making them threaten to the ecological environment (Adeola, 2004). Additionally, OCPs can be transported locally as well as globally via various pathways such as atmospheric exchange, ocean currents, and animal migration leading to the prevalence of OCPs in the air, seawater, lakes, sediments, and organisms (Lohmann et al., 2007; UNEP, 2021; Wang et al., 2010). Due to restrictions on the production and use of OCPs, most of them have shown a declining trend over the past decade. Available data indicated that the environmental levels of most OCPs declined in recent years, which may be due to the decrease in major primary sources associated with production and use (Wang et al., 2005). However, secondary emissions, such as release from old stockpiles (sediments, soil, and water, etc), still exist, and a few old types of equipment containing OCPs are still used (Ding et al., 2019; Nizzetto et al., 2010). Therefore, it is necessary to analyze OCPs in the oceans, which serve as important reservoirs of

* Corresponding author. Guangxi Laboratory on the Study of Coral Reefs in the South China Sea; Coral Reef Research Center of China; School of Marine Sciences, Guangxi University, Nanning, 530004, China.

** Corresponding author. Kefu Yu. Guangxi Laboratory on the Study of Coral Reefs in the South China Sea; Coral Reef Research Center of China; School of Marine Sciences, Guangxi University, Nanning, 530004, China.

E-mail addresses: 1827301008@st.gxu.edu.cn (Y. Kang), rjzhang@gxu.edu.cn (R. Zhang), kefuyu@scsio.ac.cn (K. Yu).

[1] Co-first authors (contribute equally).

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contaminants (Iwata et al., 1993).

Reef-building corals are important component of the diverse coral reef ecosystems (Hedouin et al., 2016). However, they are vulnerable to threats from numerous environmental stressors, in which anthropogenically-derived chemical pollutants have received substantial attention (Fonseca et al., 2017; van der Schyff et al., 2021; Zhang et al., 2019, 2021a). Several studies have delved into the environmental occurrence of various anthropogenic chemicals in the global coral reef areas. For example, OCPs are widely distributed in corals off the coast of South Africa (Porter et al., 2018) and Tern Island and Bikini Atoll in the Pacific (Wang et al., 2008). They were detected not only in the coral tissue but also in the coral skeletons. OCPs monitoring in the South China Sea (SCS) has shown that the contamination of coastal corals is more severe than that of the offshore corals and that OCPs tend to converge from the atmosphere to the ocean (Kang et al., 2022). Other chemical pollutants, such as PCBs (El Nemr et al., 2004; Miao et al., 2000; Ranjbar Jafarabadi et al., 2018), antibiotics (Zhang et al., 2019), OPEs (Ding et al., 2020), PCBs (Zhang et al., 2021b), and PAHs (Zhang et al., 2021a), also ubiquitously occurred in corals as well as in the air and seawater from the coral reef regions. A previous study illustrated the toxicological effects of OCPs on corals (Chesher and McCloskey, 1971). Field surveys of OCPs occurrence in corals and the surrounding environments can lay the foundation for policy-making regarding coral reef ecosystem protection and management.

Weizhou Island is a typical coastal coral reef ecosystem in the northwestern part of the SCS. It is situated off the coasts of Vietnam, Guangxi, Guangdong, and Hainan Island and is a famous tourist area that has attracted approximately 600,000 visitors annually in recent years (Yu, 2018). Together with industrial and agricultural activities, the vast human activities have taken a heavy toll on the local coral reef ecosystem. It has been reported that live coral cover decreased from 70% to 17.6% in Weizhou Island between 1991 and 2019 (Huang, 2019). Anthropogenic stressors, including plastics, excess nutrients, heavy metal pollution, organic pollutants, etc., could influence the health and fitness of resident biotas in the coastal marine environment (Adams, 2005; Díaz-Mendoza et al., 2020; Porter et al., 2018; Schulte, 2007). Most contaminant levels in coastal coral reef ecosystem are usually higher than those in offshore coral reef areas (Zhang et al., 2019, 2021a). However, knowledge of the occurrence of OCPs in the coral reefs of Weizhou Island remains scant.

Additionally, scleractinian corals are benthic invertebrates with carbonate skeleton and soft tissue that exudes mucus. The mucus layer plays a vital role in the physiological processes of coral, such as nutrition supply, disease defense, and information transmission (Brown and Bythell, 2005). Our previous study found higher PAH concentrations in coral mucus than those in coral tissues (Han et al., 2020). However, the differences in the occurrence of OCPs in coral tissues and mucus remain unclear.

In this study, two hypotheses are proposed: i) OCPs are widespread in the coastal coral reef ecosystem of Weizhou Island; and ii) coral mucus plays an important role in the bioaccumulation of OCPs by corals. Therefore, eight representative OCPs were selected as the target compounds (Kang et al., 2022). The aims of the paper were to i) investigate the residual levels and composition profiles of OCPs in seawater, pelagic plankton, and benthic corals from Weizhou Island and analyze their potential pollution sources; and ii) explore the differentiated role of coral tissue and mucus in OCPs enrichment and degradation.

2. Materials and methods

2.1. Study site and sample collection

Weizhou Island (20°54′ – 21°10′ N, 109°00′ – 109°15′ E), located in the Beibu Gulf, northern South China Sea, is the youngest and largest volcanic sea island in China (Fig. 1). Comprehensive sampling was carried out during April and May 2018 at a coastal coral reef ecosystem around Weizhou Island (Fig. 1). Six seawater samples, six plankton (including three phytoplankton and three zooplankton), and 24 coral samples were collected. All the samples were collected and processed as described in our previous study and detailed in Text S1 and Table S1 (Ding et al., 2020; Zhang et al., 2019, 2021b).

Each surface seawater sample (~50 L) was collected and stored in precleaned stainless steel barrel. All seawater samples were filtered with a peristaltic pump through glass fiber filters (GFFs, 150 mm, 0.7 μm pore

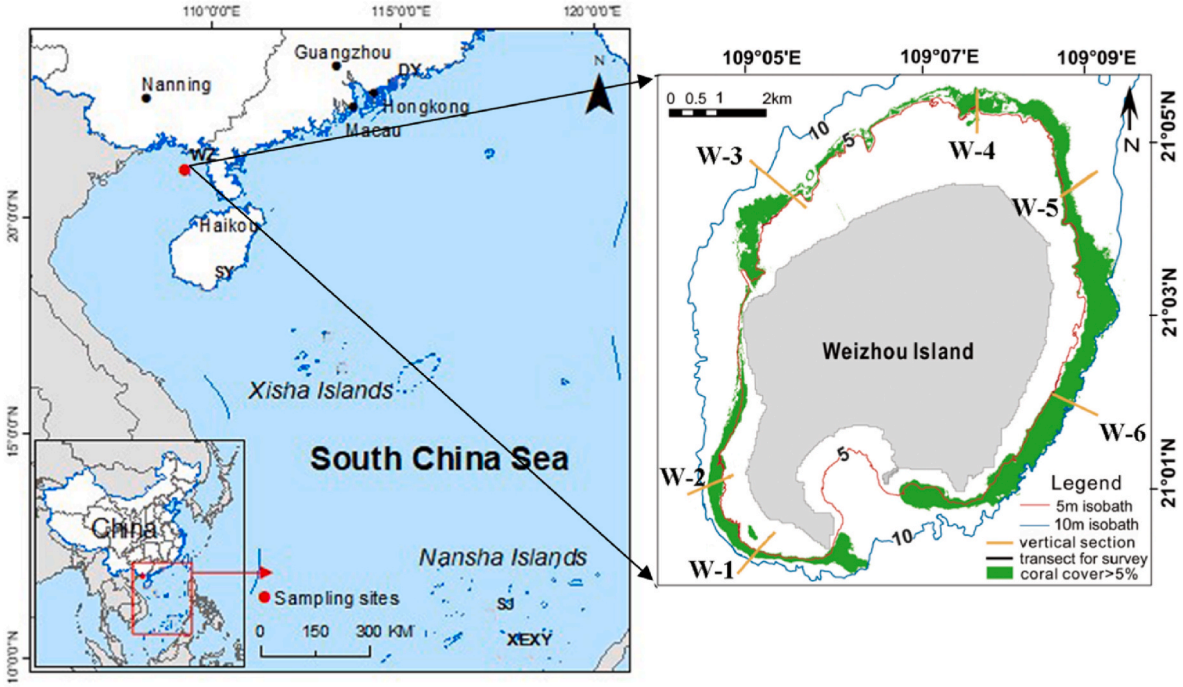

Fig. 1. Map of Weizhou Island and sampling sites. The right panel is cited from Yu et al. (Yu et al., 2019). The seawater sample sites (W-1, 2, 3, 4, 5, 6) are presented in the right picture.

size) for the water particulate phase combined in series with two tandem precleaned polyurethane foam columns (PUFs) (diameter 60 mm, length 100 mm, density 142 mg cm^3) for water dissolved phase at 1 L min^{-1}. Plankton samples were collected by vertical hauls with a conical plankton net with a 22 μm mesh, manually operated within the upper 20 m of the water column (the vertical tow speed was approximately 0.5 m s^{-1}) until the volume of the samples collected was sufficient for analysis. Bulk samples were fractionated through a nylon screen to separate the phytoplankton fraction (<98 μm) from the zooplankton fraction (>98 μm). Plankton samples were filtered with precombusted and preweighed GF/F filters, subsequently wrapped in precombusted aluminum foil, and stored at −20 °C in airtight plastic bags. Plankton biomass was estimated by weight differences of the filters after being freeze-dried (Table S1).

Coral samples including sixteen Faviidae, two Oculinidae, three Poritidae, one Pectiniidae, and one Mussidae were collected by divers using a hammer and chisel. Their sizes ranged from 6 cm × 6 cm–9 cm × 11 cm. According to a previous method (Zhang et al., 2019), coral tissues were separated from the coral skeleton by applying a Waterpik (Ultra Water Flosser, Jiebi Limited, China) with 1 L of synthetic seawater (Fig. S1). Then, using settlement and filtering (0.7 μm, GFFs, Whatman®, England), coral tissues were separated from seawater (Fig. S1). Tissue moisture content was calculated based on the difference in tissue weight before and after lyophilization. Coral mucus was in the filtrate, and filtrate samples were extracted by liquid–liquid extraction using dichloromethane. To quantify the dry weight of coral mucus, the method of Han et al. was used (Han et al., 2020).

All samples were placed on preheated (450 °C) aluminum foils, held on ice when in the field, transported back to the lab, and then stored at −20 °C until analysis. After being lyophilized and ground, the GFFs, PUFs, and marine biotas after being lyophilized and ground were packed in aluminum foil and stored at −20 °C until analysis.

2.2. Sample preparation and analysis

The approaches of Zhang et al. (2021a) were used to prepare the seawater and marine biotas. The freeze-dried coral, GFF, and PUF samples were spiked with 20 ng 2,4,5,6-tetrochloro-m-xylene(TCMX), PCB30, and PCB204 as surrogate standards to monitor the recovery before extracting the target compounds. Briefly, glass fiber filters and tandem precleaning PUFs after filtering seawater samples were Soxhlet extracted with dichloromethane for 36 h, purified with ENVI™-Florisil cartridges (500 mg, 3 mL), and cleaned up on a multilayer alumina/silica column filled with 3 cm neutral alumina, 3 cm acid silica gel (50% sulfuric acid), and 1 cm anhydrous sodium sulfate from bottom to top. The marine biotas were analyzed using a similar method for filter and PUF specimens. However, the solvent used for Soxhlet extraction was changed to a mixture of hexane and acetone (1:1, v:v), and the extraction time was prolonged to 48 h. Agilent 7890 gas chromatography-tandem 7000C triple quadrupole mass spectrometry (GC–MS/MS) equipped with an HP-5MS capillary column (30 m, 0.25 mm i.d, 0.25 μm film thickness) was used for OCPs analysis. More instrument parameter settings are provided in Text S1. Eight OCPs including six dichlorodiphenyltrichloroethanes (DDTs) (*o,p′*-DDT, *p,p′*-DDT, *o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDD, and *p,p′*-DDD), Hexachlorobenzene (HCB), and *p,p′*-methoxychlor (MXC) were selected as target compounds. See Text S2 and Table S2 for detailed information on the physicochemical properties, suppliers, and handling procedures of all target compounds and chemicals.

2.3. QA/QC

Concentrations of target OCPs were quantitatively analyzed by the internal standard method. Procedural blanks (solvent with surrogate standards) and control samples were included in each batch of ten samples to evaluate possible contamination in the whole instrument testing process and monitor instrument status. No target OCPs were detected in procedural blanks. Additionally, target OCPs were not observed or below the detection limits in the laboratory and field blanks. The reliability of the whole analytical procedure was assessed by using surrogate standards for OCPs. Tetrachloro-m-xylene (TCmX), PCB30, and PCB204 were selected as surrogates, and their recovery ranged from 74% to 110%. The limits of instrument detection (IDLs) were calculated based on three times the signal-to-noise (S/N). The limits of method detection (MDLs) were calculated by dividing IDLs by the sample's average mass. Concentrations of OCPs below detection (MDLs) were denoted as "nd". The MDLs of OCPs in seawater and marine biotas are listed in Table S3.

2.4. Statistical analysis

Normality and homogeneity of variance were checked using the Shapiro–Wilk test and Levene's test, respectively. One-way analysis of variance (ANOVA) demonstrated significant variation in OCPs concentrations in various environmental samples. Pearson's correlation coefficient test and Spearman's correlation coefficient test were performed on normally and nonnormally distributed data, respectively. The statistical analyses were conducted using the SPSS (version 17.0, IBM). The ocean surface current chart was plotted using the MATLAB R2018b.

3. Results and discussion

3.1. Occurrence and levels of OCPs in seawater, plankton and corals

The full dataset for the OCPs concentrations in surface seawater (pg L^{-1}) and marine biotas (ng g^{-1} dw) are shown in Fig. 2, Table 1, and Table S4. In general, the total concentrations of all OCPs ($\sum_8$OCPs) in seawater and marine biotas ranged from 83.0 to 242 pg L^{-1} (mean: 138 ± 56.7 pg L^{-1}) and from 0.05 to 7.02 ng g^{-1} dw (mean: 1.75 ± 1.56 ng g^{-1} dw), respectively. $\sum_8$OCPs exhibited higher concentrations in plankton (mean: 3.28 ± 2.34 ng g^{-1} dw) than in whole corals (mean: 1.35 ± 1.02 ng g^{-1} dw) (Table S4).

Surface seawater: Five OCPs (*o,p′*-DDT, *p,p′*-DDT, *o,p′*-DDD, *p,p′*-DDD, and HCB) were detected in seawater with detection frequencies (DFs) ranging from 83% to 100%, indicating the ubiquitous occurrence of OCPs in the coral reef area of Weizhou Island. DDTs were the most abundant OCPs, accounting for 52% of the $\sum_8$OCPs on average (Fig. S2). DDT served as a common agricultural insecticide historically by virtue of its low cost and superior insecticidal effect. It has been reported that 280–320 thousand tons of DDT used in China were mainly used in agriculture during the period of 1951–1983 ((UNDP), 2008; Wei et al., 2007). In addition, 250 tons of DDT were used annually to produce anti-fouling paints to protect ships from 1950 to 2005 in China (Yu et al.,

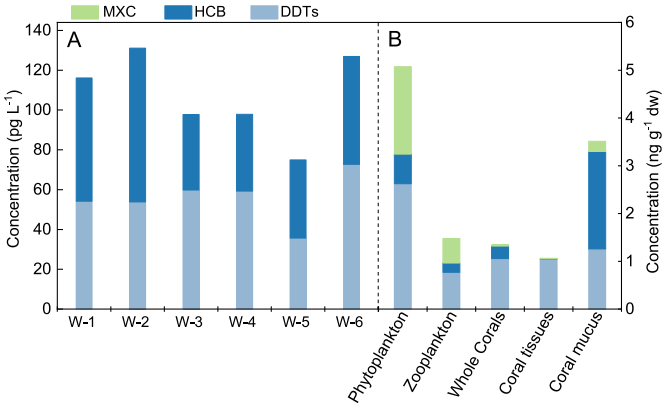

Fig. 2. Composition profiles of OCPs in seawater (A) and aquatic species (B) from Weizhou Island, SCS.

Table 1
OCPs concentrations in seawater (pg L^{-1}) and biota samples (ng g^{-1} dw) from Weizhou Island, SCS.

Chemicals	Seawater (pg L^{-1})		Phytoplankton (ng g^{-1} dw)		Zooplankton (ng g^{-1} dw)		Whole corals (ng g^{-1} dw)		All biotas (ng g^{-1} dw)	
	range	Mean ± SD [a]	range	Mean ± SD	range	Mean ± SD	range	Mean ± SD	range	Mean ± SD
o,p′-DDT	nd[b]–1.10	0.75 ± 0.42	0.24–0.55	0.42 ± 0.17	0.05–0.13	0.09 ± 0.04	0.01–0.61	0.12 ± 0.13	0.01–0.61	0.15 ± 0.16
p,p′-DDT	10.6–33.9	24.9 ± 8.23	0.27–1.02	0.56 ± 0.40	0.06–0.34	0.23 ± 0.15	0.01–2.26	0.25 ± 0.46	0.01–2.26	0.28 ± 0.44
o,p′-DDE	0.24–0.34	0.28 ± 0.04	0.02–0.07	0.05 ± 0.03	nd–0.01	nd	nd	Nd	nd–0.07	0.01 ± 0.02
p,p′-DDE	9.30–14.4	12.3 ± 1.87	0.14–0.49	0.33 ± 0.18	0.11–0.17	0.14 ± 0.03	nd–1.88	0.55 ± 0.48	nd–1.88	0.50 ± 0.45
o,p′-DDD	0.63–3.33	1.40 ± 1.05	nd	Nd	nd	nd	nd–0.21	0.03 ± 0.05	nd–0.21	0.03 ± 0.05
p,p′-DDD	12.6–22.2	16.3 ± 3.36	0.63–1.69	1.26 ± 0.56	0.28–0.35	0.31 ± 0.04	nd–0.56	0.08 ± 0.14	nd–1.69	0.22 ± 0.41
HCB	nd	nd	0.26–0.87	0.62 ± 0.32	0.14–0.29	0.20 ± 0.08	nd–1.21	0.26 ± 0.34	nd–1.21	0.29 ± 0.33
MXC	10.3–183	81.9 ± 58.5	1.00–2.97	1.83 ± 1.02	0.44–0.61	0.51 ± 0.09	nd–0.10	0.03 ± 0.03	nd–2.97	0.27 ± 0.62
∑DDTs	35.7–72.7	55.9 ± 12.0	1.31–3.38	2.62 ± 1.14	0.55–0.94	0.77 ± 0.20	0.04–3.91	1.06 ± 0.97	0.04–3.91	1.19 ± 1.04
$\sum_8$OCPs	83.0–242	138 ± 56.7	3.10–7.02	5.07 ± 1.96	1.13–1.73	1.48 ± 0.31	0.05–3.92	1.35 ± 1.02	0.05–7.02	1.75 ± 1.56

[a] Standard deviation.
[b] Not detected; All the "nd" were regarded as zero in the calculation of mean and standard deviation (SD).

2011b), and antifouling paints were reported to be an important source of DDT in Chinese fishing ports (Zhou et al., 2014). Although technical DDT and additives in antifouling paints have been banned by the Stockholm Convention and related official policies implemented in China in 1983 and 2014, respectively, there are still four major sources of DDT: legacy environmental reservoirs of historical DDTs such as soil and oceans, antifouling paints containing DDTs, malaria control, and dicofol containing high levels of DDT impurity (Lv et al., 2020). DDTs were the dominant contaminants consistent with findings in the intertidal sediment of China (Lv et al., 2020) and fish from the South China Sea (Yu et al., 2020). As such, DDT remains an environmental pollution problem in China due to its historical residues and various illegal uses.

The spatial distribution of $\sum_8$OCPs in the surface seawater is shown in Fig. 2. The sampling sites W-2 and W-6 are close to two villages that plant large amountsof sugarcanes and bananas. Agricultural activities may be one of the emission sources of OCPs. The sampling site W-3 is located near the port, and the frequent activities of boats may be an important source of DDTs (Zhou et al., 2014). Additionally, hydrological transport and biogeochemical transportation may play important roles in the spatial distribution of pollutants (Kang et al., 2022; Zhang et al., 2021a) (Fig. S3).

Marine biotas: Eight individual OCPs were detected in marine biotas with the detection frequencies (DFs) of 17–100%, also indicating the extensive distribution of OCPs in Weizhou Island, which is consistent with the previous studies (Ding et al., 2019). Among them, *p,p′*-DDT, *o,p′*-DDT, *p,p′*-DDD, and MXC were detected in all biota samples (Table S4). Moreover, phytoplankton, zooplankton, and corals showed similar composition profiles of OCPs, with DDTs being the dominant pollutants (Fig. 2). DDTs contributed to 52%, 52%, and 78% of the $\sum_8$OCPs for phytoplankton, zooplankton, and whole corals (sum of coral tissues and mucus), respectively.

The spatial distribution of OCPs in corals collected from different sites (W-1 – W-5) in Weizhou Island is shown in Fig. S4. With corals of Faviidae as an example, HCB and MXC did not show significant differences in corals from different sites. Interestingly, *p,p′*-DDE was significantly higher in both coral tissue and mucus at the W-2 sampling location than at other sampling sites (ANOVA, $p = 0.02$, 0.04, 0.04, and 0.01 for W-1, W-3, W-4, and W-5, respectively). This may be due to the high ingestion of *p,p′*-DDT from the surrounding environment by the corals at location W-2 and further biotransformation in corals.

The $\sum_8$OCPs showed a significant difference between biotas, presenting in descending order as follows: phytoplankton (5.07 ± 1.96 ng g^{-1} dw) > zooplankton (1.48 ± 0.31 ng g^{-1} dw) and whole corals (1.35 ± 1.02 ng g^{-1} dw) (ANOVA, $p = 4.25 \times 10^{-4}$ and 7.63×10^{-6}, respectively) (Table 1). ∑DDTs (ANOVA, $p = 0.02$ and 0.01, respectively) and MXC (ANOVA, $p = 5.76 \times 10^{-6}$ and 7.63×10^{-6}, respectively) were significantly higher in phytoplankton than in zooplankton and whole coral. The proportion of MXC in phytoplankton and zooplankton (36% and 35%, respectively) was higher than that in whole corals (2%) (Fig. S2). Previous studies indicated that biological uptake of compounds by phytoplankton also has strong effects on the transport of hydrophobic organic contaminants (HOCs) in aquatic ecosystems (Galban-Malagon et al., 2013; Qiu et al., 2017). Some studies have also reported that plankton are more susceptible to contaminants, have a higher accumulation capacity for pollutants, and transfer these contaminants to higher trophic levels through ingestion (Ding et al., 2021; Yeo et al., 2020). Corals are planktivorous organisms, and plankton are a significant food source for reef-building corals and many reef organisms (Bargar et al., 2013). The trophic level (TL) positions for individual organisms can be determined using a stable isotope of nitrogen (i.e., increase in ^{15}N from the diet to the consumer, $\delta^{15}N$) (Fig. S5). The detailed method of TL is described in Text S4. The $\delta^{15}N$ and TLs of all the marine biotas are listed in Table S5. On average, TLs increased as follows: phytoplankton (2.0) < zooplankton (2.05) < Faviidae (2.08) < Pectiniidae (2.13) < Poritidae (2.15). As such, OCPs in pelagic plankton were not the only contributing source of contaminants in corals. Additionally, corals have abundant symbiotic zooxanthellae. Significantly higher concentrations of polychlorinated biphenyls were detected in zooxanthellae than in coral tissues (Ranjbar Jafarabadi et al., 2018). The impact of coral symbiosis on coral bioaccumulation of OCPs should be addressed in future studies. Additionally, various biological factors, including dietary exposure, metabolic capability, age, sex, nutritional level, and habitats, can influence the accumulation extent of pollutants in biota, especially for organisms at higher trophic levels (Table S1) (Borgå et al., 2004; Zhu et al., 2020). These elements need to be taken into consideration when we further analyze the coral accumulation pattern of OCPs in the future.

The different distribution patterns of OCPs in marine organisms may be influenced by the climate of the region and morphology of the organism. Weizhou Island has a tropical oceanic monsoon climate with an average temperature of 22.6 °C, which is suitable for plankton growth (Jiang et al., 2020). Previous study found that larger plankton had a higher surface-to-volume ratio, leading to more efficient cellular uptake of hydrophobic pollutants (Ding et al., 2021; Qiu et al., 2017). This phenomenon has been reported in previous studies. For example, PCBs and PAHs were detected at higher concentrations in zooplankton than in various soft and hard corals collected from the Persian Gulf of Iran (Ranjbar Jafarabadi et al., 2018), and halogenated flame retardants (HFRs), personal care products (PCPs), and OCPs were also detected at markedly higher concentrations in plankton than in Tilapia and Apple snails (Wang and Kelly, 2018). Additionally, the susceptible response of zooplankton to pollutants may be related to their short lifespan and rapid equilibration to the surrounding environmental fluctuations (Bettinetti et al., 2012). Zooplankton also have also been found to potentially enrich OCPs from water and food potentially at a much more rapid speed than fish (Borgå et al., 2005). In addition, the higher bioaccumulative factor values (Log *BCFs*) of the HCB and MXC obtained from the samples may indicate their weaker biotransformation in

plankton. In conclusion, plankton play an essential role in the cycling of persistent organica pollutants (POPs) in oceans and lakes (Galbán-Malagón et al., 2012; Ren et al., 2017).

Comparison with other studies: A large number of studies on OCPs residues in surface seawater and living marine organisms have been reported globally (Table S6). In seawater, the ∑DDTs in Weizhou Island (35.7–72.7 pg L^{-1}) were far below the strictest limits of the *Sea Water Quality Standard of China* (*GB3097-1997*) (50 ng L^{-1} for ΣDDTs). Compared with other studies, the concentrations of the target DDTs measured in surface seawater were lower than those in the Pearl River Delta, Hong Kong's coast, the northern South China Sea, China's marginal seas, and the equatorial Indian Ocean (Huang et al., 2013; Lin et al., 2012; Luo et al., 2004; Zhang et al., 2007), but higher than that those between Australia and Antarctica (Bigot et al., 2016). The levels of DDTs did not show any significant decline compared to values from 1989 in the marginal seas of China (Iwata et al., 1993), which indicated that there may be potential pollution sources of OCPs near Weizhou Island. For the DDTs in corals, the concentrations in our study were comparable to those in the mixed matrix of coral tissues and skeletons from Tern Island, Hawaii, and Bikini Atoll in the North Pacific (Wang et al., 2008) but lower than those in the coral reef skeleton taken from the Egyptian Red Sea and marginal soft coral tissues from South Africa (El Nemr et al., 2004; Porter et al., 2018). However, the concentrations of DDTs found in our study were substantially higher than those of hard corals from remote Mascarene islands (van der Schyff et al., 2021).

3.2. DDTs compositon profiles in seawater, plankton and corals

The relative abundance of isomers in DDT mixtures offers insight into their possible sources and degradation processes (Sun et al., 2020; Wu et al., 2016). DDT can be biodegraded to DDE and DDD under aerobic and anaerobic conditions, respectively (Yu et al., 2011a). Generally, (DDD + DDE)/DDTs >0.5 proved that the dominant pollution source was from historical uses, while (DDD + DDE)/DDTs <0.5 proved the occurrence of relatively "fresh" inputs (Ding et al., 2019; Sun et al., 2020). The average (DDE + DDD)/DDT in seawater was 0.55 ± 0.08 (Fig. S6), indicating that DDT residues in Weizhou Island were primarily derived from historical use, in accordance with previous studies (Ding et al., 2019; Sun et al., 2017b, 2020) (Fig. S6). The ratios of *o,p′*-DDT/*p,p′*–DDT were considerably less than 0.2 in seawater suggesting that sources from technical DDTs were the predominant contributors (Sun et al., 2018) (Fig. S6).

For individual OCPs, *p,p′*-DDT, *p,p′*-DDD, and *p,p′*-DDE were the dominant compounds, accounting for 45%, 29%, and 22% of ∑DDTs in seawater, respectively (Fig. 3). *p,p′*-DDD and *p,p′*-DDT were the dominant substances in marine biotas, with sum of the two contaminants contributing 69% and 70% of ∑DDTs in phytoplankton and zooplankton, respectively (Fig. 3). The substances often found in whole corals were *p,p′*-DDE (54%) and *p,p′*-DDT (24%). The higher percentage of parent DDTs in seawater than in marine biota may be due to the biotransformation of DDTs in organisms (Corsolini et al., 2014), which is one of the most important elimination mechanisms for chemicals in the environment. The *p,p′*-isomers were the most abundant contaminants in marine biotas, which also indicated the potential biotransformation and metabolism of DDT in organisms. Notably, the main isomers of DDT differ between plankton and corals, suggesting that the two species may have different pathways to metabolize DDTs (Wang and Kelly, 2018). DDT metabolites were also found to be predominant in other marine biotas from the Singapore Strait (Zhang and Kelly, 2018), Pearl River Estuary (Sun et al., 2014, 2015), Yongxing Island (Sun et al., 2014), and Xuande Atoll (Sun et al., 2017b). The coral tissues exhibited *p,p′*-DDE (59%) and *p,p′*-DDT (24%) predominance among the congeners of DDTs. Special attention should be given to coral mucus, due to the existence of *o,p′*-DDT (35%) and *p,p′*-DDT (30%) (Fig. 3). This may indicate that the coral mucus has a weaker biotransformation capacity than coral tissue or that coral mucus is situated outside of coral tissues and is more exposed to the parent DDTs in the water column and food such as plankton.

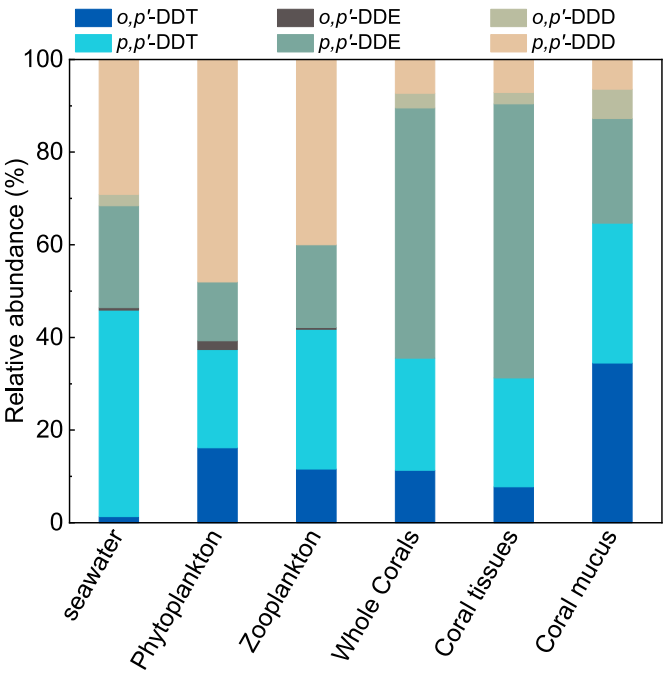

Fig. 3. Compositional profile of DDTs homologs in seawater, plankton, and corals from Weizhou Island.

3.3. Tissue-mucus distribution of OCPs in corals

Coral mucus has a gel-like consistency. It is an organic mixture secreted by coral tissue and includes organic substances such as polysaccharides, proteins, and lipid complexes (Brown and Bythell, 2005; Huettel et al., 2006). The mucus layer plays an important role in the physiological processes of coral, such as nutrition supply, protective mechanisms against pathogens, sedimentation, biofouling, desiccation, and ultraviolet radiation and information transmission (Brown and Bythell, 2005). All OCPs were detected in coral tissues and mucus except for *o,p′*-DDE (Table S7). In coral tissues, ∑DDTs (mean: 1.05 ± 1.02 ng g^{-1} dw) were the most abundant, followed by MXC (0.01 ± 0.01 ng g^{-1} dw) and HCB (0.004 ± 0.02 ng g^{-1} dw). In contrast, HCB was detected at the higher concentrations (2.04 ± 2.71 ng g^{-1} dw), followed by ∑DDTs (1.26 ± 1.16 ng g^{-1} dw) and MXC (0.21 ± 0.26 ng g^{-1} dw) in the coral mucus (Table S7). The HCB and MXC showed significantly higher levels in the coral mucus than in coral tissues (Nonparametric test, p = 0.000 and 0.001, respectively). The ∑DDTs in the coral mucus had slightly higher concentrations than those in the coral tissues (ANOVA, p > 0.05). Collectively, the $\sum_8$OCPs were significantly higher in the coral mucus (mean: 3.52 ± 3.31 ng g^{-1} dw) than in coral tissues (mean: 1.06 ± 1.02 ng g^{-1} dw) (Nonparametric test, p = 0.004) (Table S7). The different composition patterns of OCPs in coral tissue and mucus suggested that various biocomponents of corals may have different enrichment and metabolism modes for OCPs in response to environmental pressures. Notably, the total mass of OCPs may be higher in coral tissues than in coral mucus owing to the difference between coral mucus and tissue weight. This phenomenon has been observed in other studies (Han et al., 2020; Zhang et al., 2019). We used mass fractions of OCPs in coral tissues ($f_{tissues}$) and mucus (f_{mucus}) to describe the tissue-mucus distribution of OCPs in whole corals (Han et al., 2020). The weight of tissue and mucus to the whole coral weight averaged 0.84 ± 0.08 and 0.16 ± 0.08, respectively. The tissue-mucus distribution of OCPs differs between individual OCPs. The $f_{tissues}$ averaged 0.63 ± 0.22 and f_{mucus} averaged 0.37 ± 0.22 for $\sum_8$OCPs in all coral samples

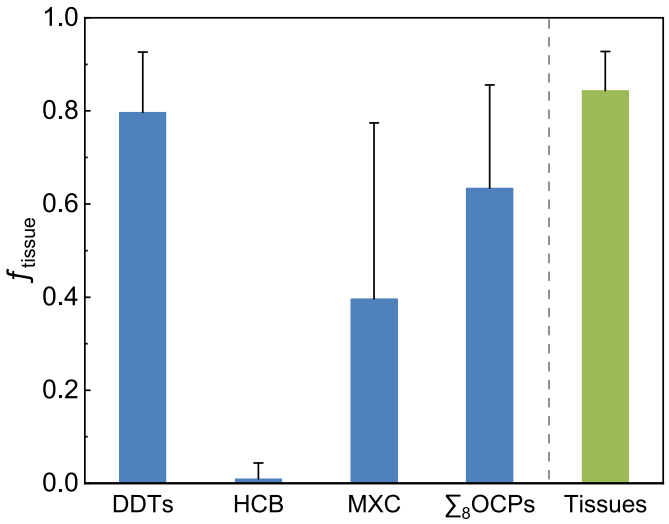

Fig. 4. The average mass fraction of coral tissues and OCPs concentrations in coral tissues.

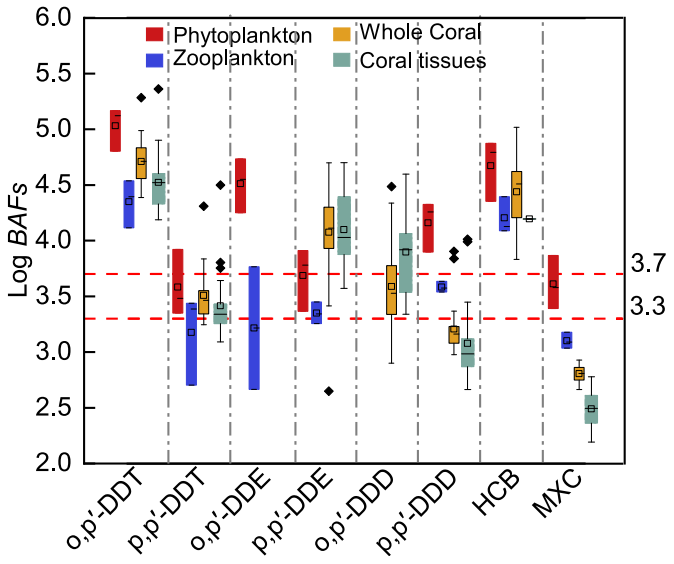

Fig. 5. The Log *BAFs* values of OCP homologs in marine biotas from Weizhou Island, SCS (box: 25–75 percentiles; whiskers: 10–90 percentiles; square: mean; point: outlier).

(Fig. 4). The ∑DDTs were mainly enriched in tissues with ∑DDTs in tissues averaging 0.80 ± 0.13. Levels of HCB and MXC were both significantly higher in mucus than in tissues (Nonparametric test, $p = 3.7 \times 10^{-5}$and 0.03, respectively), with average f_{mucus} values of 0.99 ± 0.03 and 0.60 ± 0.38, respectively. These results suggest that the resistance of corals to the OCPs stress may be remarkably elevated in tissue-mucus partitioning (Ranjbar Jafarabadi et al., 2021).

Coral mucus is an important diagnostic tool for detecting seawater quality and acceptability thresholds of risks for maintaining healthy corals (Lipp and Griffin, 2004). As described previously, corals could elevate fluid mucus production under other stress conditions (e.g., polluted water, increased temperature, and turbidity) (Bessell-Browne et al., 2017; Huettel et al., 2006). In our laboratory, we found that the mucus layer of coastal coral samples was thicker than that of offshore coral samples (Zhang et al., 2019). More antibiotics and PAHs accumulated in the mucus than in the tissues (Han et al., 2020; Zhang et al., 2019). Therefore, under the increasing environmental pressure around Weizhou Island, the coral mucus layer may have thickened and enriched more OCPs from the ambient environment, which could also be the reason for the higher concentration of OCPs in coral mucus than coral tissues in our study. In summary, coral mucus had a higher potential for adsorbing OCPs, indicating that coral mucus may play an essential role in the accumulation of OCPs by corals. Further studies on the mechanisms of accumulation and metabolism by different coral biocomponents are warranted.

3.4. Bioaccumulation of OCPs in biotas

The bioaccumulation phenomenon reflects the uptake of chemicals via both respiration (such as passive diffusion between the dissolved phase of water and cells of the biota) and food intake (i.e., active advection) (Arnot and Gobas, 2006). Chemicals represent very bioaccumulative substances if bioaccumulative factors (BAFs) are greater than 5000 (Log *BAF* 3.7). Chemicals are considered to be potentially bioaccumulative if BAFs are between 2000 (Log *BAFs* 3.3) and 5000 L kg^{-1} in biotas (Conder et al., 2008). The BAFs were calculated as the ratio of OCPs concentrations (mg kg^{-1}) in marine biotas on wet weight basis (ww) to the dissolved OCPs concentrations in the ambient seawater (mg L^{-1}). During BAFs calculations, the MDLs were used when OCPs concentrations in seawater were lower than the MDLs. The detailed calculation method is described in Text S3.

The detected Log *BAFs* of the target compounds in different marine biotas are presented in Table S8 and Fig. 5. The average Log *BAFs* of individual OCPs were 3.35–5.17, 2.67–4.54, and 2.65–5.28 in phytoplankton, zooplankton, and whole corals, respectively, which indicated great bioaccumulation potential of OCPs. Interestingly, phytoplankton showed higher Log *BAFs* than other biotas. This is likely ascribed to the fact that phytoplankton as the primary producer could absorb OCPs and these compounds will partly occur as the source for pollutants in biotas at higher trophic levels (Ren et al., 2017). The average Log *BAFs* of DDTs in the present study (3.74 ± 0.31) were nearly the same as those reported in fish (3.72 ± 0.29) from Weizhou Island, while HCB showed somewhat higher average Log *BAFs* in our study (4.42 ± 0.34) than in fish from around Weizhou Island (3.12 ± 0.23) (Ding et al., 2019). Additionally, the Log *BAFs* of DDTs and HCB in our study were less than those reported in fish from the Singapore Strait (4.79 ± 0.28 for HCB; 6.97 ± 0.44–7.68 ± 0.44 for *p,p′*- DDT and *p,p′*- DDE, respectively) (Zhang and Kelly, 2018).

The Log *BAFs* of the selected OCPs varied in different marine biotas. For example, phytoplankton had higher Log *BAFs* of DDTs and MXC than other marine species and MXC was not bioaccumulative in other marine biotas. Factors such as body length, body weight, age, sex, and environmental temperature can cause this discrepancy in the bioaccumulation of these chemicals in organisms (Wang et al., 2007). Hydrophobic (or lipophilic potentials) and metabolic ability controlled the bioaccumulation ability of compounds in organisms (Gobas et al., 2009).

The octanol-water partition coefficient (Log *Kow*) is generally considered to be a reasonable surrogate phase for lipophilic potential in biological organisms (Arnot and Gobas, 2006; Huettel et al., 2006). The average Log *BAFs* of OCPs in all marine biotas were ranked in descending order as follows: HCB (4.42) > DDTs (3.72) > MXC (2.92). Therefore, we can infer that the bioaccumulation of MXC is less significant than that of other OCPs with higher Log *Kow* (Table S2) (Kim et al., 2021). The relationship between observed Log *BAFs* and Log *Kow* of the various OCPs in marine biotas is shown in Fig. S7. The 1:1 line of Log *Kow* and Log *BAFs* is plotted for comparison. The Log *BAFs* and Log *Kow* had positive correlations in all biotas, which were both not significant ($p > 0.05$). The small number of samples may weaken the correlation between Log *BAFs* and Log K_{OW}. Of course, there would be other uncertainties concerning the achievement of equilibrium of contaminants between biota and water in the field (Chiou, 2002). The Log *BAFs* values of all marine biotas were both in the lower zone below the 1:1 line (Fig. S7). This indicates organism-seawater disequilibrium and OCPs

that have experienced degradation and slow uptake kinetics in marine organisms (Wang and Kelly, 2018). Additionally, environmental factors, such as dissolved organic matter, environmental temperature, and spatial location, may be other complex factors affecting the bioaccumulation potential of OCPs in marine organisms (Ding et al., 2020; Sun et al., 2017a). Likewise, Ren et al. (2017) suggested that dietary pattern is an important factor for the bioaccumulation potential of OCPs in biotas. Therefore, the influencing factors of coral enrichment pollutants need to be further explored and analyzed through indoor laboratory experiments.

4. Conclusion

In this study, $\sum_8$OCPs in seawater (83.0–242 pg L^{-1}), plankton (1.13–7.02 ng g^{-1} dw) and corals (0.05–3.92 ng g^{-1} dw) in a coastal coral reef ecosystems from the South China sea were reported for the first time. Plankton, which play an important role in the bioaccumulation and transport of OCPs, may have effects on OCPs enrichment in corals. The isomer ratios of DDTs indicated that DDTs in this area were mainly from the historical use of DDT-infused antifouling paints. The Obviously higher $\sum_8$OCPs in coral mucus than in coral tissues suggested that coral mucus may play an essential role in resisting the enrichment of OCPs by coral tissues and weaken the biotransformation of OCPs. In conclusion, this study provides insight into the baseline information for the bioaccumulation of OCPs in the coral reef ecosystems, especially in coral tissues and mucus. Of course, the synergistic effects of different environmental factors and biological characteristics on the distribution of OCPs in corals require further analysis.

Credit author statement

Yaru Kang: Investigation, Data analysis, Writing- Original draft preparation. Ruijie Zhang: Conceptualization, Investigation, Methodology, Data curation, Writing-Reviewing and Editing, Funding Acquisition. Kefu Yu: Conceptualization, Funding Acquisition, Resource, Validation. Minwei Han: Investigation, Data curation, Editing. Jiying Pei: Data curation, Writing-Reviewing and Editing. Zhenghua Chen: Methodology, Data curation. Yinghui Wang: Project Administration, Resource.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2022.114060.

References

Adams, S.M., 2005. Assessing cause and effect of multiple stressors on marine systems. Mar. Pollut. Bull. 51, 649–657. https://doi.org/10.1016/j.marpolbul.2004.11.040.

Adeola, F., 2004. The environmental and health impacts of persistent organic pollutants (POPs). Hum. Ecol. Rev. 11 (1), 27–35.

Arnot, J.A., Gobas, F.A.P.C., 2006. A review of bioconcentration factor (BCF) and bioaccumulation factor (BAF) assessments for organic chemicals in aquatic organisms. Environ. Rev. 14, 257–297. https://doi.org/10.1139/a06-005.

Bargar, T.A., Garrison, V.H., Alvarez, D.A., Echols, K.R., 2013. Contaminants assessment in the coral reefs of Virgin Islands national park and Virgin Islands coral reef national monument. Mar. Pollut. Bull. 70, 281–288. https://doi.org/10.1016/j.marpolbul.2013.03.001.

Bessell-Browne, P., Fisher, R., Duckworth, A., Jones, R., 2017. Mucous sheet production in Porites : an effective bioindicator of sediment related pressures. Ecol. Indicat. 77, 276–285.

Bettinetti, R., Quadroni, S., Manca, M., Piscia, R., Volta, P., Guzzella, L., et al., 2012. Seasonal fluctuations of DDTs and PCBs in zooplankton and fish of lake maggiore (northern Italy). Chemosphere 88, 344–351. https://doi.org/10.1016/j.chemosphere.2012.03.009.

Bigot, M., Muir, D., Hawker, D.W., Cropp, R., Dachs, J., Teixeira, C.F., et al., 2016. Air-seawater exchange of organochlorine pesticides in the southern ocean between Australia and Antarctica. Environ. Sci. Technol. 50, 8001.

Borgå, K., Fisk, A.T., Hoekstra, P.F., Muir, D.C.G., 2004. Biological and chemical factors of importance in the bioaccumulation and trophic transfer of persistent organochlorine contaminants in arctic marine food webs. Environ. Toxicol. Chem. 23, 2367–2385. https://doi.org/10.1897/03-518.

Borgå, K., Fisk, A.T., Hargrave, B., Hoekstra, P.F., Swackhamer, D., Muir, D.C.G., 2005. Bioaccumulation factors for PCBs revisited. Environ. Sci. Technol. 39, 4523–4532. https://doi.org/10.1021/es050376i.

Brown, B.E., Bythell, J.C., 2005. Perspectives on mucus secretion in reef corals. Mar. Ecol. Prog. Ser. 296, 291–309. https://doi.org/10.3354/meps296291.

Chesher, R., McCloskey, L.R., Chesher, R.H., 1971. (With L.R. McCloskey)"Effects of man made pollution on the dynamics of coral reefs. In: Miller et. al. TEKTITE II, vol. 6. U. S. Department of Interior VI, pp. 229–237, 229 237. Tektite II. 1971.

Chiou, C.T., 2002pp. Partition and Adsorption of Organic Contaminants in Environmental Systems. John Wiley & Sons Inc., Hoboken, pp. 82–85.

Conder, J.M., Hoke, R.A., Wolf, W.d., Russell, M.H., Buck, R.C., 2008. Are PFCAs bioaccumulative? A critical review and comparison with regulatory criteria and persistent lipophilic compounds. Environ. Sci. Technol. 42, 995–1003. https://doi.org/10.1021/es070895g.

Corsolini, S., Ancora, S., Bianchi, N., Mariotti, G., Leonzio, C., Christiansen, J.S., 2014. Organotropism of persistent organic pollutants and heavy metals in the Greenland shark Somniosus microcephalus in NE Greenland. Mar. Pollut. Bull. 87, 381–387. https://doi.org/10.1016/j.marpolbul.2014.07.021.

Courtney, K.D., 1979. Hexachlorobenzene (HCB): a review. Environ. Res. 20, 225–266. https://doi.org/10.1016/0013-9351(79)90001-X.

Díaz-Mendoza, C., Mouthon-Bello, J., Pérez-Herrera, N.L., Escobar-Díaz, S.M., 2020. Plastics and microplastics, effects on marine coastal areas: a review. Environ. Sci. Pollut. Res. 27, 39913–39922. https://doi.org/10.1007/s11356-020-10394-y.

Ding, Y., Wu, Z., Zhang, R., Yu, K., Wang, Y., Zou, Q., et al., 2019. Organochlorines in fish from the coastal coral reefs of Weizhou Island, south China sea: levels, sources, and bioaccumulation. Chemosphere 232, 1–8. https://doi.org/10.1016/j.chemosphere.2019.05.199.

Ding, Y., Han, M., Wu, Z., Zhang, R., Li, A., Yu, K., et al., 2020. Bioaccumulation and trophic transfer of organophosphate esters in tropical marine food web, South China Sea. Environ. Int. 143, 105919 https://doi.org/10.1016/j.envint.2020.105919.

Ding, Q., Gong, X., Jin, M., Yao, X., Zhang, L., Zhao, Z., 2021. The biological pump effects of phytoplankton on the occurrence and benthic bioaccumulation of hydrophobic organic contaminants (HOCs) in a hypereutrophic lake. Ecotoxicol. Environ. Saf. 213, 112017 https://doi.org/10.1016/j.ecoenv.2021.112017.

El Nemr, A., El-Sikaily, A., Khaled, A., Said, T.O., Abd-Allah, A.M.A., 2004. Chlorinated pesticides and polychlorinated biphenyls in the coral reef skeleton of the Egyptian Red Sea Coast. Bull. Environ. Contam. Toxicol. 72, 1195–1202. https://doi.org/10.1007/s00128-004-0370-8.

Fonseca, J.D.S., Marangoni, L.F.B., Marques, J.A., Bianchini, A., 2017. Effects of increasing temperature alone and combined with copper exposure on biochemical and physiological parameters in the zooxanthellate scleractinian coral Mussismilia harttii. Aquat. Toxicol. 190, 121–132. https://doi.org/10.1016/j.aquatox.2017.07.002.

Galbán-Malagón, C., Berrojalbiz, N., Ojeda, M., Dachs, J., 2012. The oceanic biological pump modulates the atmospheric transport of persistent organic pollutants to the Arctic. Nat. Commun. 3, 862.

Gobas, F.A., de Wolf, W., Burkhard, L.P., Verbruggen, E., Plotzke, K., 2009. Revisiting bioaccumulation criteria for POPs and PBT assessments. Integrated Environ. Assess. Manag. 5, 624–637. https://doi.org/10.1897/IEAM_2008-089.1.

Han, M., Zhang, R., Yu, K., Li, A., Wang, Y., Huang, X., 2020. Polycyclic aromatic hydrocarbons (PAHs) in corals of the South China Sea: occurrence, distribution, bioaccumulation, and considerable role of coral mucus. J. Hazard Mater. 384, 121299 https://doi.org/10.1016/j.jhazmat.2019.121299.

Hedouin, L.S., Wolf, R.E., Phillips, J., Gates, R.D., 2016. Improving the ecological relevance of toxicity tests on scleractinian corals: influence of season, life stage, and seawater temperature. Environ. Pollut. 213, 240–253. https://doi.org/10.1016/j.envpol.2016.01.086.

Hua, X., Shan, Z., 1996. The production and application of pesticides and factor analysis of their pollution in environment in China. Advanced in Environmental Science 4, 33–45.

Huang, H., 2019. In: Report on the Status of the Coral Reefs in China. Coral Reef Branch, Chinese Pacific Society, pp. 1–106.

Huang, Y., Xu, Y., Li, J., Xu, W., Zhang, G., Cheng, Z., et al., 2013. Organochlorine pesticides in the atmosphere and surface water from the equatorial Indian ocean: enantiomeric signatures, sources, and fate. Environ. Sci. Technol. 47, 13395–13403. https://doi.org/10.1021/es403138p.

Huettel, M., Wild, C., Gonelli, S., 2006. Mucus trap in coral reefs: formation and temporal evolution of particle aggregates caused by coral mucus. Mar. Ecol. Prog. Ser. 307, 69–84. https://doi.org/10.3354/meps307069.

Iwata, H., Tanabe, S., Sakai, N., Tatsukawa, R., 1993. Distribution of persistent organochlorines in the oceanic air and surface seawater and the role of ocean on their global transport and fate. Environ. Sci. Technol. 27, 1080–1098. https://doi.org/10.1021/es00043a007.

Jiang, W., Yu, K., Wang, N., Yang, H., Yang, H., Xu, S., et al., 2020. Distribution coefficients of trace metals between modern coral-lattices and seawater in the northern South China Sea: species and SST dependencies. J. Asian Earth Sci. 187, 104082 https://doi.org/10.1016/j.jseaes.2019.104082.

Kang, Y., Zhang, R., Yu, K., Han, M., Wang, Y., Huang, X., et al., 2022. First report of organochlorine pesticides (OCPs) in coral tissues and the surrounding air-seawater system from the South China Sea: distribution, source, and environmental fate. Chemosphere 286, 131711. https://doi.org/10.1016/j.chemosphere.2021.131711.

Kim, J.-T., Choi, Y.-J., Barghi, M., Kim, J.-H., Jung, J.-W., Kim, K., et al., 2021. Occurrence, distribution, and bioaccumulation of new and legacy persistent organic pollutants in an ecosystem on King George Island, maritime Antarctica. J. Hazard Mater. 405, 124141 https://doi.org/10.1016/j.jhazmat.2020.124141.

Lin, T., Li, J., Xu, Y., Liu, X., Luo, C., Cheng, H., et al., 2012. Organochlorine pesticides in seawater and the surrounding atmosphere of the marginal seas of China: spatial distribution, sources and air–water exchange. Sci. Total Environ. 435–436, 244–252. https://doi.org/10.1016/j.scitotenv.2012.07.028.

Lipp, E.K., Griffin, D.W., 2004. Analysis of coral mucus as an improved medium for detection of enteric microbes and for determining patterns of sewage contamination in reef environments. EcoHealth 1, 317–323. https://doi.org/10.1007/s10393-004-0132-4.

Lohmann, R., Breivik, K., Dachs, J., Muir, D., 2007. Global fate of POPs: current and future research directions. Environ. Pollut. 150, 150–165. https://doi.org/10.1016/j.envpol.2007.06.051.

Luo, X., Mai, B., Yang, Q., Fu, J., Sheng, G., Wang, Z., 2004. Polycyclic aromatic hydrocarbons (PAHs) and organochlorine pesticides in water columns from the Pearl River and the Macao harbor in the Pearl River Delta in South China. Mar. Pollut. Bull. 48, 1102–1115. https://doi.org/10.1016/j.marpolbul.2003.12.018.

Lv, M., Luan, X., Guo, X., Liao, C., Guo, D., Miao, J., et al., 2020. A national-scale characterization of organochlorine pesticides (OCPs) in intertidal sediment of China: occurrence, fate and influential factors. Environ. Pollut. 257, 113634 https://doi.org/10.1016/j.envpol.2019.113634.

UNEP, 2021. In: Methoxychlor, Draft Risk Management Evaluation, First Draft POPRC.17. https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/979984/methoxychlor-draft-risk-management-evaluation.pdf.

Miao, X.S., Swenson, C., Yanagihara, K., Li, Q.X., 2000. Polychlorinated biphenyls and metals in marine species from French frigate shoals, North Pacific ocean. Arch. Environ. Contam. Toxicol. 38, 464–471. https://doi.org/10.1007/s002449910061.

Nizzetto, L., Macleod, M., Borgå, K., Cabrerizo, A., Dachs, J., Guardo, A.D., et al., 2010. Past, present, and future controls on levels of persistent organic pollutants in the global Environment. Environ. Sci. Technol. 44, 6526–6531. https://doi.org/10.1021/es100178f.

Porter, S.N., Humphries, M.S., Buah-Kwofie, A., Schleyer, M.H., 2018. Accumulation of organochlorine pesticides in reef organisms from marginal coral reefs in South Africa and links with coastal groundwater. Mar. Pollut. Bull. 137, 295–305. https://doi.org/10.1016/j.marpolbul.2018.10.028.

Qiu, Y., Zeng, E.Y., Qiu, H., Yu, K., Cai, S., 2017. Bioconcentration of polybrominated diphenyl ethers and organochlorine pesticides in algae is an important contaminant route to higher trophic levels. Sci. Total Environ. 579, 1885–1893. https://doi.org/10.1016/j.scitotenv.2016.11.192.

Ranjbar Jafarabadi, A., Riyahi Bakhtiari, A., Aliabadian, M., Laetitia, H., Shadmehri Toosi, A., Yap, C.K., 2018. First report of bioaccumulation and bioconcentration of aliphatic hydrocarbons (AHs) and persistent organic pollutants (PAHs, PCBs and PCNs) and their effects on alcyonacea and scleractinian corals and their endosymbiotic algae from the Persian Gulf, Iran: inter and intra-species differences. Sci. Total Environ. 627, 141–157. https://doi.org/10.1016/j.scitotenv.2018.01.185.

Ranjbar Jafarabadi, A., Mashjoor, S., Riyahi Bakhtiari, A., Cappello, T., 2021. Ecotoxico linking of phthalates and flame-retardant combustion byproducts with coral solar bleaching. Environ. Sci. Technol. 55, 5970–5983. https://doi.org/10.1021/acs.est.0c08730.

Ren, J., Wang, X., Wang, C., Gong, P., Wang, X., Yao, T., 2017. Biomagnification of persistent organic pollutants along a high-altitude aquatic food chain in the Tibetan Plateau: processes and mechanisms. Environ. Pollut. 220, 636–643. https://doi.org/10.1016/j.envpol.2016.10.019.

Schulte, P.M., 2007. Responses to environmental stressors in an estuarine fish: interacting stressors and the impacts of local adaptation. J. Therm. Biol. 32, 152–161. https://doi.org/10.1016/j.jtherbio.2007.01.012.

Sun, Y., Hao, Q., Xu, X., Luo, X., Wang, S., Zhang, Z., et al., 2014. Persistent organic pollutants in marine fish from Yongxing Island, South China Sea: levels, composition profiles and human dietary exposure assessment. Chemosphere 98, 84–90. https://doi.org/10.1016/j.chemosphere.2013.10.008.

Sun, Y., Zhang, Z., Xu, X., Hu, Y., Luo, X., Cai, M., et al., 2015. Bioaccumulation and biomagnification of halogenated organic pollutants in mangrove biota from the Pearl River Estuary, South China. Mar. Pollut. Bull. 99, 150–156. https://doi.org/10.1016/j.marpolbul.2015.07.041.

Sun, R., Luo, X., Tang, B., Chen, L., Liu, Y., Mai, B., 2017a. Bioaccumulation of short chain chlorinated paraffins in a typical freshwater food web contaminated by e-waste in south China: bioaccumulation factors, tissue distribution, and trophic transfer. Environ. Pollut. 222, 165–174. https://doi.org/10.1016/j.envpol.2016.12.060.

Sun, Y., Hu, Y., Zhang, Z., Xu, X., Li, H., Zuo, L., et al., 2017b. Halogenated organic pollutants in marine biota from the Xuande Atoll, South China Sea: levels, biomagnification and dietary exposure. Mar. Pollut. Bull. 118, 413–419. https://doi.org/10.1016/j.marpolbul.2017.03.009.

Sun, R., Luo, X., Li, Q.X., Wang, T., Zheng, X., Peng, P., et al., 2018. Legacy and emerging organohalogenated contaminants in wild edible aquatic organisms: implications for bioaccumulation and human exposure. Sci. Total Environ. 616–617, 38–45. https://doi.org/10.1016/j.scitotenv.2017.10.296.

Sun, R., Yu, J., Liao, Y., Chen, J., Wu, Z., Mai, B., 2020. Geographical distribution and risk assessment of dichlorodiphenyltrichloroethane and its metabolites in Perna viridis mussels from the northern coast of the South China Sea. Mar. Pollut. Bull. 151, 110819 https://doi.org/10.1016/j.marpolbul.2019.110819.

UNDP, 2008. U.N.D.P. Improvement of DDT-Based Production of Dicofol and Introduction of Alternative Technologies Including IPM for Leaf Mites Control in China. https://www.cn.undp.org/content/china/en/home/operations/projects/environment_and_energy/improvement-of-ddt-based-production-of-dicofol-and-introduction-.html.

van der Schyff, V., du Preez, M., Blom, K., Kwet Yive, N.S.C., Klánová, J., Přibylová, P., et al., 2021. Chlorinated and brominated persistent compounds in hard coral, soft coral, and parrotfish from remote Mascarene islands. Chemosphere 267, 129316. https://doi.org/10.1016/j.chemosphere.2020.129316.

Wang, Q., Kelly, B.C., 2018. Assessing bioaccumulation behaviour of hydrophobic organic contaminants in a tropical urban catchment. J. Hazard Mater. 358, 366–375. https://doi.org/10.1016/j.jhazmat.2018.06.070.

Wang, T., Lu, Y., Zhang, H., Shi, Y., 2005. Contamination of persistent organic pollutants (POPs) and relevant management in China. Environ. Int. 31, 813–821. https://doi.org/10.1016/j.envint.2005.05.043.

Wang, Y., Li, X., Li, A., Wang, T., Zhang, Q., Wang, P., et al., 2007. Effect of municipal sewage treatment plant effluent on bioaccumulation of polychlorinated biphenyls and polybrominated diphenyl ethers in the recipient water. Environ. Sci. Technol. 41, 6026–6032. https://doi.org/10.1021/es070913u.

Wang, D., Miao, X., Li, Q., 2008. Analysis of organochlorine pesticides in coral (Porites evermanni) samples using accelerated solvent extraction and gas chromatography/Ion trap mass spectrometry. Arch Environ Con Tox 54, 211–218. https://doi.org/10.1007/s00244-007-9016-2.

Wang, G., Lu, Y., Han, J., Luo, W., Shi, Y., Wang, T., et al., 2010. Hexachlorobenzene sources, levels and human exposure in the environment of China. Environ. Int. 36, 122–130. https://doi.org/10.1016/j.envint.2009.08.005.

Wei, D., Kameya, T., Urano, K., 2007. Environmental management of pesticidal POPs in China: past, present and future. Environ. Int. 33, 894–902. https://doi.org/10.1016/j.envint.2007.04.006.

Wu, Y., Wang, X., Ya, M., Li, Y., Hong, H., 2016. Distributions of organochlorine compounds in sediments from jiulong River Estuary and adjacent western taiwan strait: implications of transport, sources and inventories. Environ. Pollut. 219, 519–527. https://doi.org/10.1016/j.envpol.2016.05.081.

Yeo, B.G., Takada, H., Yamashita, R., Okazaki, Y., Uchida, K., Tokai, T., et al., 2020. PCBs and PBDEs in microplastic particles and zooplankton in open water in the Pacific Ocean and around the coast of Japan. Mar. Pollut. Bull. 151, 110806 https://doi.org/10.1016/j.marpolbul.2019.110806.

Yu, K., 2018. Introduction to the Science of Coral Reef. Science press, Beijing.

Yu, H., Bao, L., Liang, Y., Zeng, E.Y., 2011a. Field validation of anaerobic degradation pathways for dichlorodiphenyltrichloroethane (DDT) and 13 metabolites in marine sediment cores from China. Environ. Sci. Technol. 45, 5245–5252. https://doi.org/10.1021/es2006397.

Yu, H., Shen, R., Liang, Y., Cheng, H., Zeng, E.Y., 2011b. Inputs of antifouling paint-derived dichlorodiphenyltrichloroethanes (DDTs) to a typical mariculture zone (South China): potential impact on aquafarming environment. Environ. Pollut. 159, 3700–3705. https://doi.org/10.1016/j.envpol.2011.07.010.

Yu, W., Wang, W., Yu, K., Wang, Y., Huang, X., Huang, R., et al., 2019. Rapid decline of a relatively high latitude coral assemblage at Weizhou Island, northern South China Sea. Biodivers. Conserv. 28, 3925–3949. https://doi.org/10.1007/s10531-019-01858-w.

Yu, X.X., He, Q.Y., Sanganyado, E., Liang, Y., Bi, R., Li, P., et al., 2020. Chlorinated organic contaminants in fish from the South China Sea: assessing risk to Indo-Pacific humpback dolphin. Environ. Pollut. 263 https://doi.org/10.1016/j.envpol.2020.114346.

Zhang, H., Kelly, B.C., 2018. Sorption and bioaccumulation behavior of multi-class hydrophobic organic contaminants in a tropical marine food web. Chemosphere 199, 44–53. https://doi.org/10.1016/j.chemosphere.2018.01.173.

Zhang, G., Li, J., Cheng, H., Li, X., Xu, W., Jones, K.C., 2007. Distribution of organochlorine pesticides in the northern South China Sea: implications for land outflow and air-sea exchange. Environ. Sci. Technol. 41, 3884–3890. https://doi.org/10.1021/es070072r.

Zhang, R., Yu, K., Li, A., Wang, Y., Huang, X., 2019. Antibiotics in corals of the South China Sea: occurrence, distribution, bioaccumulation, and considerable role of coral mucus. Environ. Pollut. 250, 503–510. https://doi.org/10.1016/j.envpol.2019.04.036.

Zhang, R., Han, M., Yu, K., Kang, Y., Wang, Y., Huang, X., et al., 2021a. Distribution, fate and sources of polycyclic aromatic hydrocarbons (PAHs) in atmosphere and surface water of multiple coral reef regions from the South China Sea: a case study in spring-

summer. J. Hazard Mater. 412, 125214 https://doi.org/10.1016/j.jhazmat.2021.125214.

Zhang, R., Kang, Y., Yu, K., Han, M., Wang, Y., Huang, X., et al., 2021b. Occurrence, distribution, and fate of Polychlorinated Biphenyls (PCBs) in multiple coral reef regions from the South China Sea: a case study in spring-summer. Sci. Total Environ. 777, 146106 https://doi.org/10.1016/j.scitotenv.2021.146106.

Zhou, S., Yang, H., Zhang, A., Li, Y.-F., Liu, W., 2014. Distribution of organochlorine pesticides in sediments from Yangtze River Estuary and the adjacent East China Sea: implication of transport, sources and trends. Chemosphere 114, 26–34. https://doi.org/10.1016/j.chemosphere.2014.03.100.

Zhu, C., Sun, Y., Li, D., Zheng, X., Peng, X., Zhu, T., et al., 2020. Evidence for complex sources of persistent halogenated compounds in birds from the south China sea. Environ. Res. 185. https://doi.org/10.1016/j.envres.2020.109462.