



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

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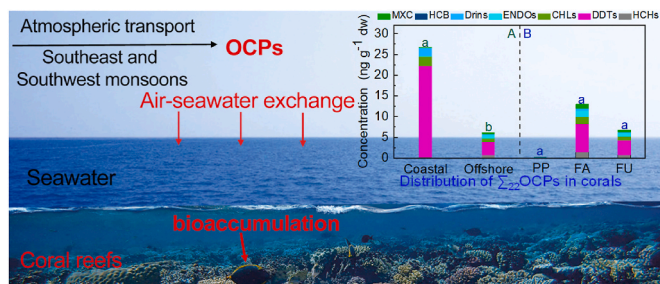
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HIGHLIGHTS

- Pioneer OCPs study of coral tissues from the South China Sea (SCS).
- Corals presented a higher bio-accumulative ability to OCPs.
- Ocean currents and monsoons played essential roles in the OCPs' spatial distribution.
- Net atmospheric deposition of OCPs occurred in the coral reef regions (CRRs), SCS.
- OCPs may have a migration pattern of atmosphere-ocean-corals in the CRRs, SCS.

GRAPHICAL ABSTRACT



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ABSTRACT

The levels, fate, and potential sources of 22 organochlorine pesticides (OCPs) in coral tissues and the surrounding air-seawater system from the South China Sea (SCS) were elucidated for the first time. $\sum_{22}\text{OCPs}$ (total concentration of 22 OCPs) ($16.1\text{--}223\text{ pg L}^{-1}$) was relatively higher in coastal seawater than in offshore seawater, which may be the widespread influence of coastal pollution inputs under the western boundary current. The atmospheric $\sum_{22}\text{OCPs}$ were predominantly distributed in the gas phase ($48.0\text{--}2264\text{ pg m}^{-3}$) and were mainly influenced by continental air mass origins. The air-seawater exchange of selected OCPs showed that OCPs tended to migrate from the atmosphere to seawater. The distribution of $\sum_{22}\text{OCPs}$ in coral tissues ($0.02\text{--}52.2\text{ ng g}^{-1}\text{ dw}$) was significantly correlated with that in air samples, suggesting that OCPs may have a migration pattern of atmosphere-ocean-corals in the SCS. Corals exhibited higher bioaccumulation ability ($\text{Log BAFs: }2.42\text{--}7.41$) for OCPs. Source analysis showed that the new application of technical Chlordanes (CHLs) was primarily responsible

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for the current levels of CHLs in the surrounding environment over the SCS, while historical residues were the primary sources of other OCPs.

1. Introduction

Organochlorine pesticides (OCPs), legacy persistent organic pollutants (POPs) under the Stockholm Convention, have been of worldwide concern due to their persistence, bioaccumulation, adverse ecological effects, and toxicity in humans and wildlife (Wang et al., 2008b; Zhang et al., 2007). They have been widely used to improve agricultural productivity worldwide, predominantly during the 1950–1980s. Nevertheless, only 1% of the implemented pesticides target their pests, while 99% of the pesticides are discharged into the global environment through surface runoff, animal migration, long-distance atmospheric transport, etc. (Wania and MacKay, 1996; Zhang et al., 2011b). Additionally, due to their chemical stability and lipophilicity, OCPs tend to accumulate in biological tissues and biomagnify in the food chain (Jensen, 1966; Sun et al., 2017). Although the prohibition of the use of OCPs has been executed in many nations and the primary emissions of OCPs are restricted or banned entirely in most parts of the world, recent inputs of several OCPs have still been recorded in the environment (Ali et al., 2014). For instance, the input of technical Chlordane and DDTs have been recorded in air and sediments from China, Vietnam, and Pakistan, respectively. (Ji et al., 2015; Syed et al., 2014; Wang et al., 2016). Moreover, the use of DDTs is permitted to control malaria/mosquitoes in India and Vietnam under the Stockholm Convention (Hung and Thiemann, 2002).

The South China Sea (SCS) has an area of approximately 3.5 million km² and is surrounded by subtropical-tropical developing countries in Southeast Asia (Sun et al., 2017). A large quantity of OCPs have been used for agriculture, public health, industry, and so on in these countries and have been considered potentially relevant sources of OCPs (Loganathan, 1994; Minh et al., 2006). Under the limit of primary sources, secondary sources are becoming predominant (Gioia et al., 2012). The oceans were believed to be the most significant sink of OCPs, but they may also be converted into a secondary source of particular POPs (e.g., α -HCH, *p,p'*-DDE, and *p,p'*-DDT) to the atmosphere (Iwata et al., 1993) (Lin et al., 2012). The environmental processes of OCPs, such as air-seawater exchange and dry and wet deposition, have also been studied in many global oceans and seas, such as the Arctic Ocean (Lohmann et al., 2009), Atlantic Ocean (Jaward et al., 2004), Southern Ocean (Bigot et al., 2016), and Equatorial Indian Ocean (Huang et al., 2013). As previously reported, the Global Distillation/Fraction Effect Hypothesis proposed that part of the global POP burden generated at more temperate latitudes is transported to polar regions via atmospheric and oceanic processes (Wania and Mackay, 1993). Thus, the dynamics of air-seawater exchange and the within-ocean processing of OCPs are critical to their global fate and behavior. However, studies on the air-seawater exchange of OCPs in the SCS and northern SCS dated back to three decades ago and fifteen years ago, respectively (Iwata et al., 1993; Zhang et al., 2007).

Coral reefs, the largest ecological reservoirs of marine biodiversity, create the most productive and varied ecosystem in the ocean (Ko et al., 2014) and provide many goods and services to humans (Zhao et al., 2006). However, the amount of living coral coverage (LCC) has severely degraded over the past 50 years under the influence of ocean warming, ocean acidification, local environmental pressure, and intense human activity (Qin et al., 2019; Wilkinson, 1996; Yu, 2012; Zhao et al., 2014). Coral reefs are vulnerable to many toxic substances, such as metals and organic chemicals (Meehan and Ostrander, 1997). Previous studies have indicated the extensive occurrence of OCPs in soft corals (*Sarcophyton glaucum* and *Sinularia gravis*) from coastal coral reef regions (CRRs) in South Africa (Porter et al., 2018), hard coral (*Porites evermanni*), including tissues and skeletons from Tern Island and Bikini Atoll in the

Pacific Ocean (Wang et al., 2008a), and a minor occurrence in hard coral (*Montastraea annularis*) from the Virgin Islands (Bargar et al., 2013). Adverse effects, including reduced dinoflagellate densities and bleaching, impaired photosynthesis, partial colony mortality, reduced fecundity, and reduced larval metamorphosis and settlement, were suspected in coral at multiple life stages. However, to date, the presence of OCPs in corals from the SCS has not been reported. In summary, investigated the levels and fate of OCPs in corals and ambient air-surface seawater system in warranted in the SCS. Understanding the dynamics of air-seawater exchange and the within-ocean processing of POPs in the SCS is critical to obtain better insight into their global fate and behavior.

This study aims to (1) investigate the occurrence, congener profiles, and spatial distribution of OCPs in coral tissues and ambient seawater and air from the SCS; (2) identify potential sources of OCPs in the SCS; and (3) estimate the air-seawater exchange and bioaccumulation of target OCPs.

2. Materials and methods

2.1. Sampling

A total of 29 air samples (Nos: 1–29), 23 seawater samples (Nos: W01 – W23) and 9 coral samples were collected from May to August 2015. The other seven air samples (Nos: 23–29) and five coral samples were taken from April to May 2016 (Text S1). Particulate and dissolved OCPs in seawater were collected in glass fiber filters and polyurethane foam columns (PUFs). Particulate and gas OCPs in air were collected in quartz fiber filters and PUFs. All coral samples were captured using a hammer and chisel by divers (Han et al., 2020; Zhang et al., 2019). More details of sampling, sample information, and preparation were published previously (Zhang et al., 2018, 2021b) and are shown in Texts S1–S2, Fig. S1, and Tables S1–S4 of the Supplementary Materials.

2.2. Extraction and analysis

Sample extraction of target OCPs in PUFs, filters, and coral tissues was similar to that described previously (Ding et al., 2019) and detailed in Text S2. Twenty-two OCPs, including six dichlorodiphenyltrichloroethanes (DDTs) (*o,p'*-DDT, *p,p'*-DDT, *o,p'*-DDE, *p,p'*-DDE, *o,p'*-DDD, *p,p'*-DDD), four hexachlorocyclohexanes (HCHs) (α -HCH, β -HCH, γ -HCH, δ -HCH), three Drins (Aldrin, Endrin, and Dieldrin), five Chlordanes (CHLs) (trans-Chlordane (TC), cis-Chlordane (CC), Heptachlor, Heptachlor epoxide (HCE) (A), and Heptachlor epoxide (HCE) (B)), two Endosulfans (ENDOs) (α -Endosulfan and β -Endosulfan), Hexachlorobenzene (HCB), and *p,p'*-methoxychlor (MXC) were selected as target compounds. Information on samples, including physicochemical properties, suppliers, and handling procedures of all target compounds and chemicals, are given in Text S3 and Table S5.

Measurements of OCPs were carried out with Agilent 7890 gas chromatography-tandem 7000C triple quadrupole mass spectrometry (GC-MS/MS) in electron impact/multiple reaction monitoring (EI/MRM) mode. An HP-5MS capillary column (length: 30 m; i.d.: 0.25 mm; film thickness: 0.25 μ m) was used (Zhang et al., 2021b). The specific operation and parameter settings of the instrument method are given in Text S2.

2.3. QA/QC

As published previously (Zhang et al., 2021b), field and laboratory blanks, replicate samples, and detection limits of GC-MS/MS were analyzed. Target OCPs were not present or were below detection limits

in the laboratory and field blanks. The recoveries of 20 ng surrogate standards 2,4,5,6-tetrachloro-m-xylene (TCMX), PCB30, and PCB204 in air, seawater, and coral samples were in the ranges of 63.5%–110%, 74.0%–110%, and 63.2%–90.5%, respectively. The final reported OCP concentrations in this study were not corrected by surrogate recoveries. The limits of instrument detection (IDLs) were estimated as the concentration corresponding to a signal-to-noise (S/N) ratio of 3. The limits of method detection (MDLs) were calculated by dividing IDLs by the sample's average mass. The MDLs for seawater, air, and coral tissues were 0.0028–0.0564 pg L⁻¹, 0.0009–0.0185 pg m⁻³, and 0.001–0.0358 ng g⁻¹ dw (dry weight), respectively (Table S6).

2.4. Data analysis and air mass back trajectories

The Shapiro-Wilk test and Levene's test were used to test the normality and homogeneity of a data set, respectively. Independent *t*-test (IBM SPSS 17.0) was applied to various variables to examine the statistical significance of the differences between subgroups when the data are normally distributed; otherwise, a non-parametric test was used. The statistical test results were considered statistically significant and highly significant when $p < 0.05$ and $p < 0.01$, respectively. Person's correlation and Heat Map Clustering Analysis (HM-HCA) can be used to infer the provenance correlation of OCPs in the SCS. The Heat Map Clustering Analysis (HM-HCA) was plotted using the R version 4.0.2. The ArcGIS 10.2 software was used for plotting map (Environmental Systems Research Institute, Redlands, US). The calculation of the air mass back trajectories were published previously (Zhang et al., 2021b) and detailed in Texts S4.

3. Results and discussion

3.1. Occurrence and spatial distribution of OCPs in seawater and air samples

3.1.1. Occurrence and concentration levels

In this study, only dissolved phase OCPs in seawater were reported due to the loss of particulate phase samples. Seventeen of the 22 target OCPs were detected in seawater samples, and all the targeted OCPs were detected in air samples (Table 1, Table S6 and S7), indicating that OCP contamination was prevalent in the ambient environment of the corals from the SCS. β -HCH, δ -HCH, *o,p'*-DDT, *p,p'*-DDT, *o,p'*-DDD, *p,p'*-DDD, β -Endosulfan, Endrin, and MXC were detected in all seawater and air samples.

The \sum_{22} OCPs ranged from 16.1 to 223 pg L⁻¹ (mean: 59.0 \pm 43.8 pg L⁻¹) in seawater. CHLs, HCB, DDTs, and Drins were the dominant chemicals, with average percentages of 26%, 25%, 22%, and 20% of \sum_{22} OCPs in seawater, respectively (Fig. S3). A significant positive correlation was observed between the logarithm (log) concentrations of \sum CHLs and \sum drins ($p < 0.01$), likely indicating a similar source of the

two dominant contaminants in seawater (Table S8). Heat map clustering analysis (HM-HCA) further characterized the relationship between CHLs and drins (Figs. S4–A). The \sum_{22} OCPs ranged from 50.3 to 2268 pg m⁻³ (mean: 796 \pm 615 pg m⁻³) in the air samples. In the gas phase, the average \sum_{22} OCPs (778 \pm 605 pg m⁻³) was significantly higher than that in the particle phase (mean: 17.3 \pm 28.1 pg m⁻³) (nonparametric test, $p < 0.01$). In the gas phase, the major OCP congeners were CHLs and Drins, which accounted for 55% and 39% of \sum_{22} OCPs, respectively (Fig. S3). A significant positive correlation was also observed between the log concentrations of \sum CHLs and \sum drins ($p < 0.05$), indicating a similar source of the two dominant contaminants in the gas phase (Table S8, Figs. S4–D). In the particle phase, DDTs, HCB, CHLs, ENDOs, and HCHs were the dominant compounds, with contributions of 31%, 19%, 14%, 12%, and 11% for \sum_{22} OCPs, respectively (Fig. S3). Moreover, significant positive correlations were also observed between the log concentrations of each OCP congener, except for \sum CHLs, with HCB and MXC in the particle phase (Table S8).

Historical data for OCPs in the CRRs from the SCS were fairly limited, associated mainly with HCHs and DDTs, and with only a few focusing on other OCP congeners. The compositional profile of the OCPs was different from previous reports in the SCS, which generally predicted higher levels of DDTs and HCHs than CHLs in seawater and gas phase samples (Lin et al., 2012; Zhang et al., 2007). These results may indicate that the SCS has been continuously exposed to relatively higher levels of CHLs. Chlordane, used as a termiticide for woods in many places of the world, was detected at high levels (Murayama et al., 2003). Additionally, elevated concentrations of TC and CC were observed in South China's air (Li et al., 2007). Compared with global areas, the levels of HCHs and DDTs in seawater and air samples from the SCS were at the lower end of the global range. Regarding CHLs, concentrations of TC and CC were much higher in the SCS than in South Korea and the Equatorial Indian Ocean (Huang et al., 2013; Jaward et al., 2005). Compared with previous data in the same sampling regions, the \sum DDTs, \sum HCHs, TC, and CC showed an increasing and then decreasing trend (Iwata et al., 1993; Zhang et al., 2007). The residual levels of \sum DDTs and \sum HCHs were found to substantially decrease compared to these previous studies, which may be due to the restriction or prohibition of these compounds since the early 1970s under the regulations of the Stockholm Convention (Gioia et al., 2012). However, TC and CC were not found to decrease substantially compared to the previous study, indicating their continuous release from the surrounding environment.

3.1.2. Spatial distribution

In the seawater, relatively high \sum_{22} OCPs were detected in W13 and W14, which are located off Luhuitou (LHT) on Hainan Island (Fig. 1, Figs. S5–A). Hainan Island has experienced fast regional urbanization, industrialization, and a historically high agricultural application of OCPs (Wang et al., 2005). Coastal seawaters were particularly affected by inputs of OCPs via discharges of sewage and seagoing rivers with

Table 1
Summary of selected concentrations of OCPs in air, seawater, and coral tissues from the South China Sea.

Compounds	Air (particle phase)		Air (gas phase)		Seawater		Coastal coral		Offshore coral	
	pg m ⁻³		pg m ⁻³		pg L ⁻¹		ng g ⁻¹ dw		ng g ⁻¹ dw	
	Range	Mean \pm SD ^a	Range	Mean \pm SD	Range	Mean \pm SD	Range	Mean \pm SD	Range	Mean \pm SD
HCHs	0.12–15.6	1.90 \pm 3.48	0.18–18.4	2.44 \pm 4.25	0.47–1.68	0.81 \pm 0.30	0.03–0.92	0.28 \pm 0.33	nd ^b –3.06	0.70 \pm 1.12
DDTs	0.90–32.5	5.42 \pm 7.55	5.00–67.7	17.8 \pm 18.8	3.96–96.2	12.7 \pm 18.2	6.32–49.8	22.0 \pm 15.1	nd–14.1	3.23 \pm 5.06
CHLs	nd–21.6	2.33 \pm 5.40	27.8–1369	431 \pm 347	1.15–99.7	15.3 \pm 21.1	0.09–10.3	2.18 \pm 4.04	nd–3.40	0.84 \pm 1.26
ENDOs	0.09–14.7	2.05 \pm 3.71	2.06–41.9	14.2 \pm 10.2	0.13–6.30	1.42 \pm 1.33	nd–0.75	0.18 \pm 0.29	nd–3.42	0.79 \pm 1.19
Drins	nd–10.0	1.28 \pm 2.36	10.7–826	303 \pm 246	1.71–62.0	12.0 \pm 12.9	nd–9.35	1.96 \pm 3.70	nd–0.90	0.22 \pm 0.34
HCB	nd–27.0	3.22 \pm 6.96	nd–25.7	7.27 \pm 6.17	3.49–37.7	14.9 \pm 6.67	nd–0.32	0.06 \pm 0.13	nd–0.14	0.02 \pm 0.04
MXC	0.40–5.37	1.06 \pm 1.00	0.41–8.83	1.91 \pm 1.81	1.19–3.20	1.78 \pm 0.48	nd–0.18	0.04 \pm 0.07	nd–2.00	0.45 \pm 0.73
\sum_{22} OCPs	1.69–115	17.3 \pm 28.1	48.0–2264	778 \pm 606	16.1–223	59.0 \pm 43.8	6.57–52.2	26.7 \pm 18.4	0.02–27.0	6.26 \pm 9.70

^a Standard deviation.

^b Not detected.

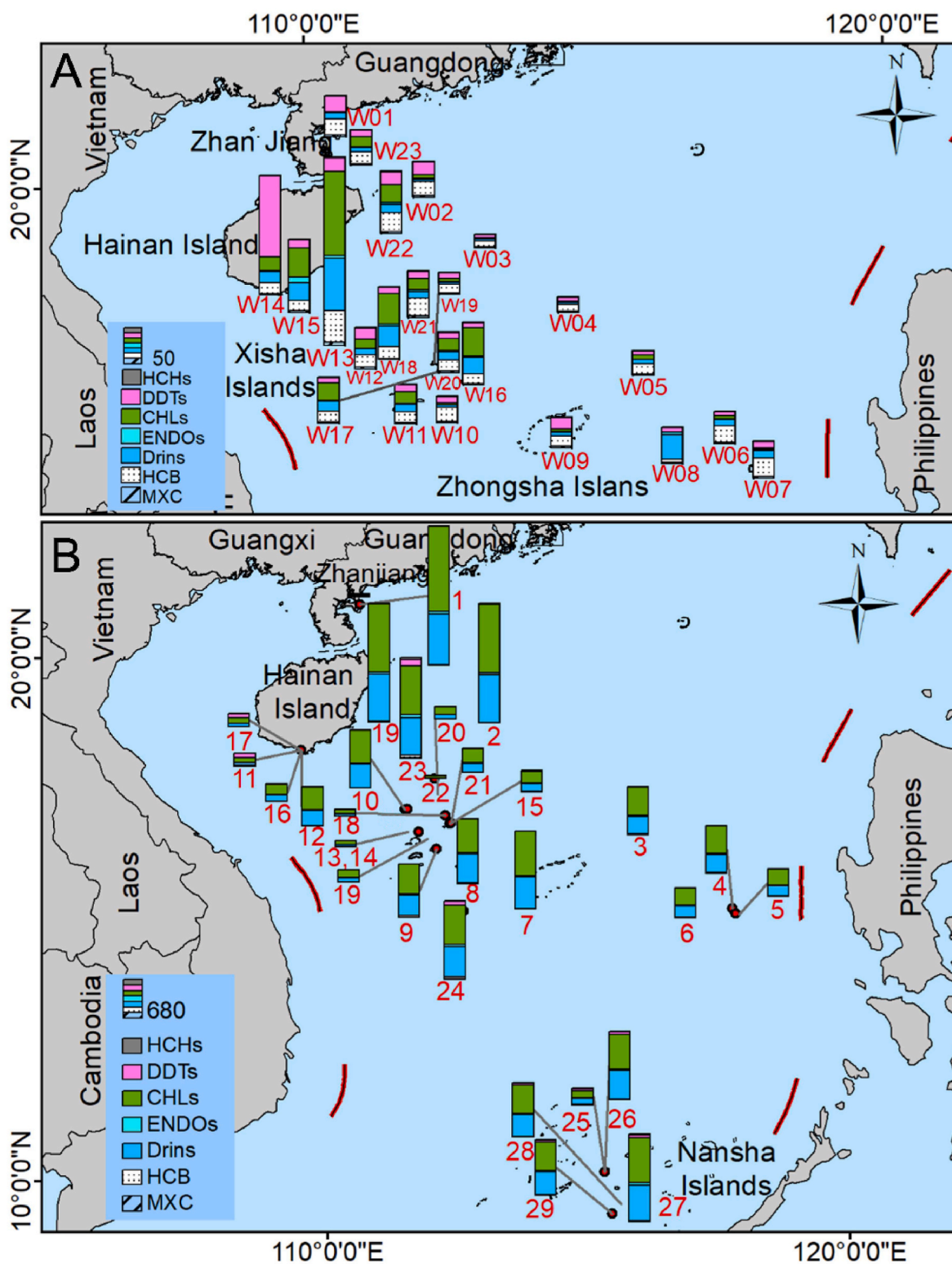


Fig. 1. The spatial distribution of OCPs in seawater (pg L^{-1}) (A) and air samples (pg m^{-3}) (B) in the SCS.

industrial effluents (Lin et al., 2012). In addition, the Zhong-Xisha Sea areas may be the confluence of the west boundary current on the outer edge of the Sunda Shelf and the North Equatorial Current (Hwang and Chen, 2000; Morimoto et al., 2000), accepting pollution sources from the coasts of the Indo-China Peninsula in May–June. Previous studies also found that OCPs were ubiquitously distributed in the Singapore Strait marine environment and the organic farms of the Cameron

Highlands, Malaysia (Farina et al., 2018; Zhang and Kelly, 2018). Thus, the western boundary current may transport OCPs from coastal areas of some Southeast Asian countries to the north of the SCS (Fig. S7).

For individual OCP congeners, β -HCH, δ -HCH, o,p' -DDT, o,p' -DDE, p,p' -DDD, α -Endosulfan, and MXC concentrations were evenly distributed, with the highest/lowest ratios of 6.08, 3.57, 5.87, 2.08, 6.21, 7.62, and 2.68, respectively, which may be related to the nationwide ban or a lack

of fresh input sources and their relatively quick degradation processes in the environment (Lin et al., 2012) (Table S6) (Figs. S6–A). In contrast, other OCPs generally showed a spatial distribution of sites close to land over distant seas, which showed the diffusion of terrigenous pollutants from the mainland to the ocean. It is worth noting that the highest concentration of Aldrin and the lowest concentration of TC were both observed at W07; the highest concentration of β -Endosulfan and the lowest concentration of Aldrin were both observed at W15; the highest concentration of heptachlor and the lowest concentration of MXC were both observed at W23, which may indicate different sources and transport pathways for individual OCPs in different regions.

As shown in Fig. S5, S6-C and D, significant spatial distributions of OCPs were observed in the particle and gas phase samples along the sampling route. For example, the highest/lowest value of Endrin in the gas phase was up to 2058 (Table S6). As described before, potential sources were investigated by examining air mass back trajectories, which is a widely accepted tool to support the identification of source areas (Zhang et al., 2007). In the sampling period dominated by the southwest monsoon, relatively high \sum_{22} OCPs in the gas phase were detected at Nos. 1, 2, 15, 16, and 18 (Figs. S5–D). Back trajectory analysis revealed that the air masses during these sampling points passed through Vietnam, Cambodia, Thailand, Malaysia, and Laos. Additionally, during the sampling period dominated by the southeastern monsoon, the seven sampling stations (Nos. 9, 14, 23, 25, 26, 27, and 29) displayed higher concentrations of \sum_{22} OCPs. The air masses passed through the Philippines at these sampling stations. Based on the above discussion, it can be concluded that the origins of the air masses from the continental outflow in the surrounding countries played important roles in controlling the OCP concentrations in the atmosphere in the SCS. Likewise, an earlier study reported that the changing trends in the levels of OCPs on the sampling route also appear to be controlled by emission sources and air mass origins (Zhang et al., 2007).

3.2. Occurrence and spatial distribution of OCPs in coral tissues

OCP concentrations in coral tissues were reported on a dry weight (dw) basis except where indicated. All targeted OCPs were detected in coastal coral tissues, while 21 OCPs were detected in offshore corals (Table S7). \sum_{22} OCPs was significantly higher (nonparametric test, $p < 0.05$) (Fig. 2) in coastal corals (mean: $26.7 \pm 18.4 \text{ ng g}^{-1} \text{ dw}$; range: $6.57\text{--}52.2 \text{ ng g}^{-1} \text{ dw}$) than in offshore corals (mean: $6.26 \pm 9.70 \text{ ng g}^{-1} \text{ dw}$; range: $0.02\text{--}27.0 \text{ ng g}^{-1} \text{ dw}$). In the offshore CRRs, it is worth noting that the \sum_{22} OCPs were higher in coral tissues from the NS Islands ($11.5 \pm 9.63 \text{ ng g}^{-1} \text{ dw}$) than in those from the XS Islands ($3.31 \pm 8.38 \text{ ng g}^{-1} \text{ dw}$). The regional distribution pattern of \sum_{22} OCPs in coral tissues was similar to that in air samples, indicating the possible impact of OCPs in the corals' surrounding environment. For high levels of OCPs in coral

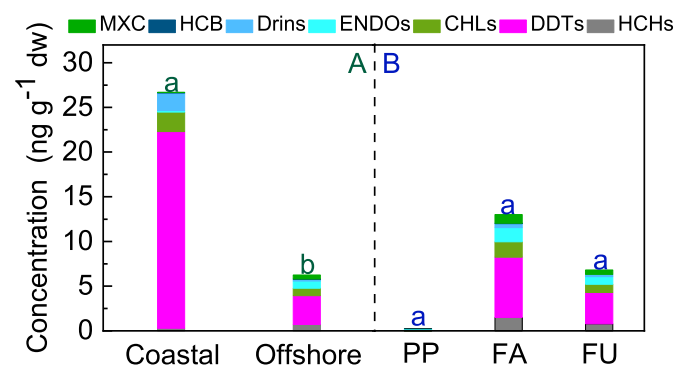


Fig. 2. Concentrations of OCPs in coral tissues from different coral reef regions (A) and coral families (B) (PP: Pocilloporidae; FA: Faviidae; and FU: Fungiidae). For each panel, bars that do not share the same letters are significantly different ($p < 0.05$).

tissues from the NS Islands, ocean currents, tropical monsoons and cyclones, and rainfall may play an important role in their distribution (Zhang et al., 2007). Moreover, the log concentrations of \sum HCHs and \sum DDTs in corals were significantly correlated with the corresponding OCP congeners in the gas phase and seawater samples, respectively, indicating the atmospheric deposition of these compounds in coral reef regions (Table S11).

The composition profiles of OCPs were similar among the coastal and offshore CRRs (Fig. S3). DDTs and CHLs were the dominant contributions to \sum_{22} OCPs, with average contributions of 90% and 65% for coastal and offshore corals, respectively. The predominance of DDTs was also found in marine biota from Yongxing Island (Ding et al., 2019) and Xuande Atoll (Sun et al., 2017). The relatively high levels of DDTs in coastal coral tissues may be partially related to its extensive historical usage in countries surrounding the SCS (Minh et al., 2008; Zhang et al., 2007) or current usage in fishery and agricultural activities in coastal CRRs (Shi et al., 2016). DDT antifouling paints are still used in coastal regions of China to prevent the adhesion of barnacles to the hulls of fishing ships (Zhang et al., 2007).

Globally, there is very little information on the concentrations of OCPs in corals. The average \sum_{22} OCPs recorded in hard and soft corals worldwide was $760 \text{ ng g}^{-1} \text{ dw}$, as summarized by a meta-analysis (Snedaker et al., 1999). The average \sum_{22} OCPs in coastal and offshore coral tissues in this study were lower than the average value (Snedaker et al., 1999). As the dominant OCPs, the average \sum DDTs found in our study had a comparable level to the coral reef skeleton taken from the Egyptian Red Sea and was slightly higher than those found in the mixed matrix of coral tissues and skeletons from Tern Island, Hawaii, and Bikini Atoll in the North Pacific (Table S9) (El Nembr et al., 2004; Wang et al., 2008a). However, \sum_{22} OCPs and \sum DDTs were substantially lower in marginal soft coral tissues from South Africa (Porter et al., 2018). Compared with other aquatic organisms in the SCS, the average \sum_{22} OCPs recorded in our study was significantly lower than those in fish from the Pearl River Delta and Weizhou Island (Ding et al., 2019; Guo et al., 2008) and comparable to those in edible fish from Taihu Lake (Zhao et al., 2013). Additionally, the \sum DDTs were substantially lower than those of mollusks from the Bohai Sea and fish from the Pearl River Delta (Guo et al., 2008; Wang et al., 2008b).

The differences in OCPs among the coral families are shown in Fig. 2. The differences among coral families in coastal coral reefs were not examined because the sample numbers were not sufficiently large enough for meaningful statistical analysis. Comparing among coral families, the average \sum_{22} OCPs in offshore corals was in the rank order of Faviidae (FA) ($13.0 \pm 11 \text{ ng g}^{-1} \text{ dw}$, $n = 5$) > Fungiidae (FU) ($6.80 \pm 9.51 \text{ ng g}^{-1} \text{ dw}$, $n = 3$) > Pocilloporidae (PP) ($0.30 \pm 0.18 \text{ ng g}^{-1} \text{ dw}$, $n = 3$). ANOVA showed no significant differences in \sum_{22} OCPs among coral families (all p values > 0.05). The limited number of samples may have caused this insignificant statistical result. However, it was noted that the massive corals (Faviidae and Fungiidae) showed higher enrichment with OCPs than the branched corals (Pocilloporidae). The difference in the distribution of \sum_{22} OCPs among these coral families was consistent with the concentration distribution of PAHs and PCBs in coral tissues from the same regions in our previous study (Han et al., 2020; Zhang et al., 2021b) and Hainan Island (Xiang et al., 2018) in the SCS.

3.3. OCPs sources/fingerprints

Composition differences of OCP congeners in the environment could indicate contamination sources and historical usage of OCPs and thus have been extensively applied in previous studies (Lin et al., 2012; Zhang et al., 2007). In the present study, we only discussed compounds with a detection frequency higher than 50% at the sampling stations.

In the present study, α -HCH and γ -HCH were not detected in seawater, and discussion only include compounds detected at frequency higher than 50% in samples. β -HCH and δ -HCH were the dominant PCBs

in the seawater and air samples, which were also observed in the soils and sediments in Xinghua Bay in Southeast China (Zhang et al., 2011a). β -HCH is the most stable isomer and is relatively resistant to microbial degradation (Li et al., 2016). High temperatures during the summer in the SCS likely improved the preferential volatile gradient of OCPs from the soils to the atmosphere, and it resulted in transporting OCPs to the SCS via atmospheric deposition and exchange (Ripszám et al., 2015). However, the isomer profiles of HCHs were different from those reported by Zhang et al. over the same oceanic regions with a relatively high proportion of α -HCH and γ -HCH (Zhang et al., 2007). According to the α -HCH/ γ -HCH ratios (range: 1.47–1.55 for gas phase; 0.46–1.55 for particle phase), lindane was dominant at all sampling points during the measurement period (Iwata et al., 1995) (Figs. S8–A). According to the β -/(α + γ -) HCH ratios, the HCHs were mainly from historical use of lindane after a period of degradation (Figs. S8–A) (Liu et al., 2012; Zhao et al., 2013). It is worth noting that obviously lower ratios of β -/(α + γ -) HCH were observed at NO.2 (0.17) in the gas phase samples, indicating the presence of a possible sporadic source of lindane from surrounding countries (Wang et al., 2011, 2016).

All the ratios of (DDE + DDD)/DDT were >0.5 in seawater and air samples, indicating that DDTs were derived from historical use instead of fresh inputs in the SCS (Sun et al., 2014). The ratios of o,p' -DDT/ p,p' -DDT in seawater ranged from 0.39 to 2.86, and 17% of the ratios were >1.3 , suggesting a mixed source from technical DDTs and dicofol (Sun et al., 2014). The ratios of o,p' -DDT/ p,p' -DDT in the particle and gas phase samples were similar, with a range from 0.56 to 1.16, also indicating a mixed source from technical DDTs and dicofol in air samples (Figs. S8–B) (Qiu et al., 2005).

Chlordane is mainly used as a termiticide against agents in tropical and subtropical timber (Ji et al., 2015). In the present study, the ratios of TC/CC in seawater samples ranged from 0.18 to 1.82, 65% of which were >1 and 40% of which were >1.2 , suggesting that the new application of technical CHLs was primarily responsible for the current levels of CHLs in seawater over the SCS (Fig. S3) (Li et al., 2007) (Bidleman et al., 2002). The TC/CC ratios showed a slight difference between the gas and particle phase samples (2.23 ± 0.44 versus 2.30 ± 1.29 , respectively) (Fig. S3 and S8–C). As described previously, relatively high levels and fresh input of technical Chlordane were also observed in the Indian Ocean, East China Sea, Guangdong Province, and South China Sea (Huang et al., 2013; Iwata et al., 1993; Ji et al., 2015). In conclusion, the new application of technical CHLs was primarily responsible for the current levels of CHLs in the surrounding environment over the SCS.

Technical Endosulfan contains 70% α -Endosulfan and 30% β -Endosulfan (Sultana et al., 2014). In all seawater samples, the β -Endosulfan was the predominant compound of \sum ENDOs. In air samples, the ratios of α -Endosulfan/ β -Endosulfan were in the range of 0.21–0.55 and 0.12–15.8 in the particle phase and gas phase, respectively. A majority of the ratios in the gas phase (55%) were higher than 2.33, indicating recent and old sources of technical Endosulfan (Figs. S8–D) (Chakraborty et al., 2010; Khuman and Chakraborty, 2019). The different composition patterns that Endosulfan different exhibits in seawater, and the atmosphere may be due to its different properties (Peterson and Batley, 1993). In air samples, the α -Endosulfan has a higher vapor pressure and Henry coefficient than β -Endosulfan, which means that it is more volatile and, therefore, more likely to exist in the gas phase. This phenomenon was also observed in North America and Vietnam. (Shen et al., 2005; Wang et al., 2016). Additionally, α -Endosulfan is more subject to oxidation than β -Endosulfan in aquatic systems (Walse et al., 2003), resulting in lower α -Endosulfan/ β -Endosulfan ratios than technical grade (7:3), indicating historical use and no fresh input. In terms of the above results, there were no newer inputs of Endosulfan in the seawater. Fresh usage of an Endosulfan technical mixture and physico-chemical properties of compounds are the two main causes of the distribution patterns of ENDOs in air samples.

Correlations between target chemicals in the air may indicate common sources and atmospheric processes. As described before, HCB

showed a significant positive correlation with PAHs and almost entirely originated from the combustion process ($>90\%$) in the air from Zhejiang Province, East China (Mao et al., 2020). Our previous study indicated that many PAHs are mainly from combustion sources (Zhang et al., 2021a). However, according to Pearson's correlation coefficient (Table S12), HCB showed weak correlations with PAHs, suggesting a smaller influence of combustion sources on the HCB in this study area. Thus, the manufacturing and application of pesticides and chemicals were the primary environmental HCB sources.

3.4. Gas-particle partitioning and air-seawater exchange of selected OCPs

3.4.1. Gas-particle partitioning

According to our previous study, the ratio (R_g) of gas phase concentration (C_g) to the total concentration of gas phase and particle phase (C_{g+p}) was used to express the gas-particle partitioning of OCPs (Zhang et al., 2021b). The average values of all OCP congeners were both >0.5 (Figs. S9–A). At many sampling stations (62%–100%), the R_g values of individual compounds were higher than 0.5, except for the compounds that were not detected in either particle or gas phases (Figs. S9–B). The OCPs in the gas phase were dominant over those in the particulate phase, which was also detected in Singapore's atmosphere (He et al., 2009). OCPs are typical volatile organic pollutants. In this study, the OCPs were predominantly distributed in the gas phase, and this phenomenon may be due to the semivolatile properties of OCPs and the relatively high atmospheric temperature in the SCS (Yang et al., 2008). High atmospheric temperature could increase the vapor pressure of chemicals, thus enhancing the chemical volatilization of particulate matter in the air and changing the gas-particle partitioning of chemicals. Some studies have also reported that air humidity, particulate matter properties, and individual pollutant properties play essential roles in the distribution of semivolatile organic pollutants between the gas and particle phases (Gregoris et al., 2014; Qiao et al., 2019). In the particle phase, the log average concentration of individual OCPs had obviously positive relationships to $\log K_{OA}$ ($R^2 = 0.2295$, $p < 0.05$), and the average log average concentrations of individual OCPs had significantly negative relationships to \log vapor pressure ($R^2 = 0.1810$, $p < 0.05$) in the particle phase (Fig. S10).

3.4.2. Air-seawater exchange

Fugacity ratios (ff) and air-seawater exchange fluxes ($F_{a/w}$, $\text{ng m}^{-2} \text{d}^{-1}$) were calculated only for analytes found in $\geq 50\%$ of the samples in both the gas and dissolved phases (Text S5). A total of 16 paired air-seawater samples were used to assess the equilibrium status of OCPs between the air and seawater and the direction of air-seawater gas exchange. The detailed methods for determination ff and $F_{a/w}$ are described in Text S5. The results of ff and $F_{a/w}$ are presented in Table S13 and Fig. 3. The ff values of all selected OCPs except MXC were obviously <0.5 at all the selected stations, indicating that the air-seawater system along our transect in the SCS was far from equilibrium, with net deposition for the compounds investigated. The most common methods to quantify $F_{a/w}$ use a modified version of Deacon's boundary layer model (Mackay and Yeun, 1983). This model has been applied in many previous studies for POPs (Huang et al., 2014; Qiu et al., 2008). The $F_{a/w}$ of all selected OCPs showed negative net fluxes in the targeted areas, another sign that the target OCPs are currently being deposited from the air to the seawater.

The negative $F_{a/w}$ of \sum DDTs (o,p' -DDT, p,p' -DDT, o,p' -DDD, p,p' -DDD and p,p' -DDE) ranged from -2199 to $-215 \text{ ng m}^{-2} \text{d}^{-1}$ (mean: $-917 \text{ ng m}^{-2} \text{d}^{-1}$) in this area, which indicated that large quantities of DDTs transferred from the air into the seawater. The air-seawater exchange direction of DDTs from air to water was also found in the North Atlantic and Arctic Ocean areas (Lohmann et al., 2009, 2012). However, this result is different from those observed in the Indian Ocean and the Southern Ocean, where net volatilization was found for o,p' -DDT, p,p' -DDT, and p,p' -DDE (Bigot et al., 2016; Huang et al., 2013). As

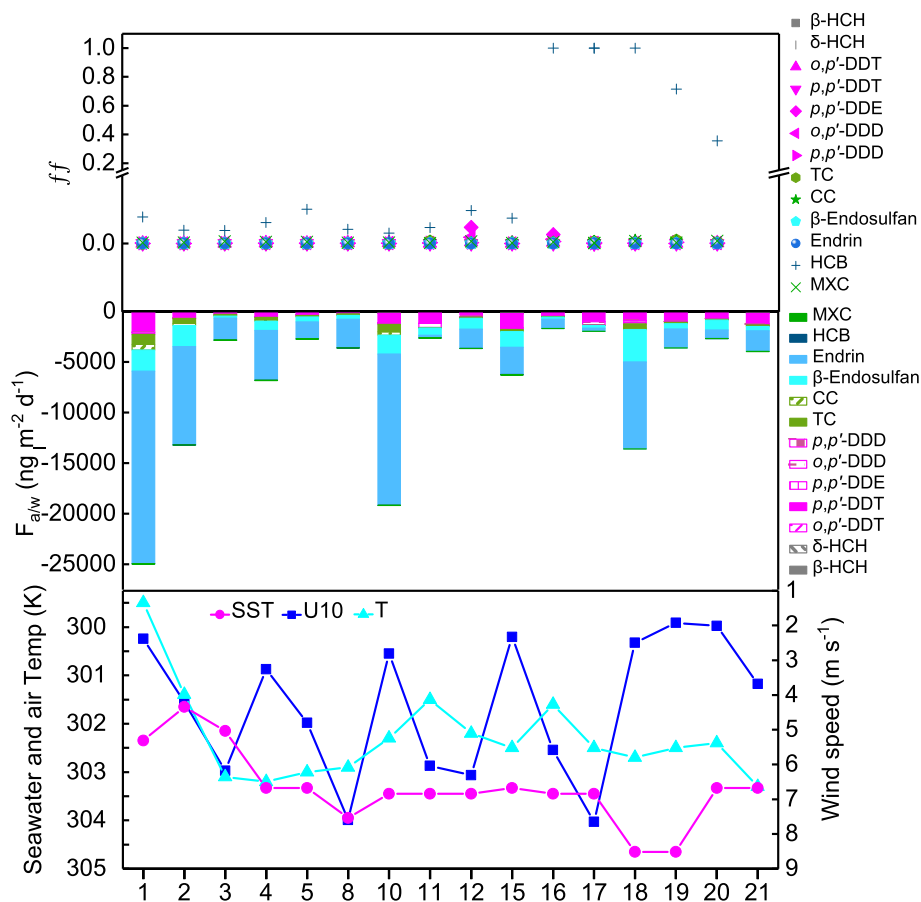


Fig. 3. Fugacity ratios and air-seawater exchange fluxes of selected OCPs during the sampling cruise.

described before in the same study area 10 years ago, the air-seawater exchange direction of p,p' -DDE was mainly from seawater to air, while o,p' -DDT and p,p' -DDT diffused from seawater to air in proximity to land and underwent net deposition in the remote Ocean (Zhang et al., 2007). Compared with a previous study reported in the same study area 30 years ago, \sum DDTs (p,p' -DDT and p,p' -DDE) (mean: $-9.1 \text{ ng m}^{-2} \text{ d}^{-1}$) were obviously lower than those in our study (Iwata et al., 1993). The continuous input through long-range atmospheric transport to the SCS from surrounding developing countries may cause negative DDT fluxes. Additionally, the variation in wind speed and the extent of the biological pump driven by primary productivity could contribute to the large variability observed in fluxes. In the summer, the strong monsoon pushes surface water to flow in a circulative pattern, and pollutants imported from atmospheric deposition and fluvial transport could be quickly dissipated by the surface current (Lin et al., 2012). As mentioned earlier, the increase in the biological pump may decrease the levels of OCPs in seawater, and it further enhances the vertical sinking fluxes of OCPs during high-productivity summer months (Bigot et al., 2016; Dachs et al., 2000).

For \sum CHLs, apparently negative fluxes (range: -1550 to $-25.6 \text{ ng m}^{-2} \text{ d}^{-1}$; mean: $-350 \text{ ng m}^{-2} \text{ d}^{-1}$) were observed in this study. The air-seawater exchange fluxes of TC and CC were consistent with previous results in the same study area (Iwata et al., 1993; Zhang et al., 2007). Stations close to equilibrium or net evaporation of TC, CC, and Heptachlor were found in the North Atlantic (Zhang et al., 2012) and most sampling stations from the Indian Ocean (Huang et al., 2013). It should be noted that the average negative flux of \sum CHLs in our study was significantly higher than that in the same study area observed 30 years ago (mean: $-3.6 \text{ ng m}^{-2} \text{ d}^{-1}$). The $F_{a/w}$ values of HCB ranged from -14.5 to $0.69 \text{ ng m}^{-2} \text{ d}^{-1}$, with a mean value of $-3.64 \text{ ng m}^{-2} \text{ d}^{-1}$. As described before, there is no known process that could degrade HCB in

surface seawaters, and the exportation of HCB associated with particles was the main removal process, but the removal process was considered weak for HCB and was consistent with relatively low $F_{a/w}$ values in our study (Barber et al., 2005; Lohmann et al., 2009). Pearson's correlation analysis suggested that fluxes of OCP congeners were mostly determined by the air-seawater concentration gradient ($C_w - C_{aRT}/H$) (Fig. S11). This is consistent with the result in the Pacific Ocean, where HCB fluxes were mostly driven by the concentration gradient (Huang et al., 2014).

3.5. OCPs' bioaccumulation in coral tissues

The phenomenon of bioaccumulation reflects the absorption of compounds through respiration (such as passive diffusion between the dissolved phase of water and cells of the biota) and dietary intake (Arnot and Gobas, 2006). The bioaccumulative factors (BAFs, in L Kg^{-1}) were defined as the ratio of OCP concentrations (mg kg^{-1} wet weight (ww)) in the coral tissues to those dissolved in ambient seawater (mg L^{-1}). The conversion between the dry and wet weight of OCPs was carried out by the moisture content of the coral tissues (Table S14). The detailed calculation method is described in Text S6. To calculate coral BAFs in the NS Islands, the OCP concentrations in seawater from the northern SCS were used because the seawater samples of the NS Islands were not analyzed. Their MDLs were applied if a few OCP concentrations were lower than the MDLs in seawater (Zhang et al., 2021b). Chemicals are considered to be bioaccumulated if BAFs in L Kg^{-1} are higher than 5000 (Log BAF 3.7) and potentially bioaccumulated if BAFs range from 2000 (Log BAF 3.3) to 5000 L kg^{-1} (Log BAF 3.7) in biota (Agency, 2017).

As shown in Table S15 and Fig. 4, the average Log BAFs of individual OCPs were in the ranges of 2.65–6.87 and 4.65–7.17 in coastal and offshore coral tissues, respectively. Most of them were higher than 3.7, indicating that coral tissues are highly bioaccumulative to OCPs. In

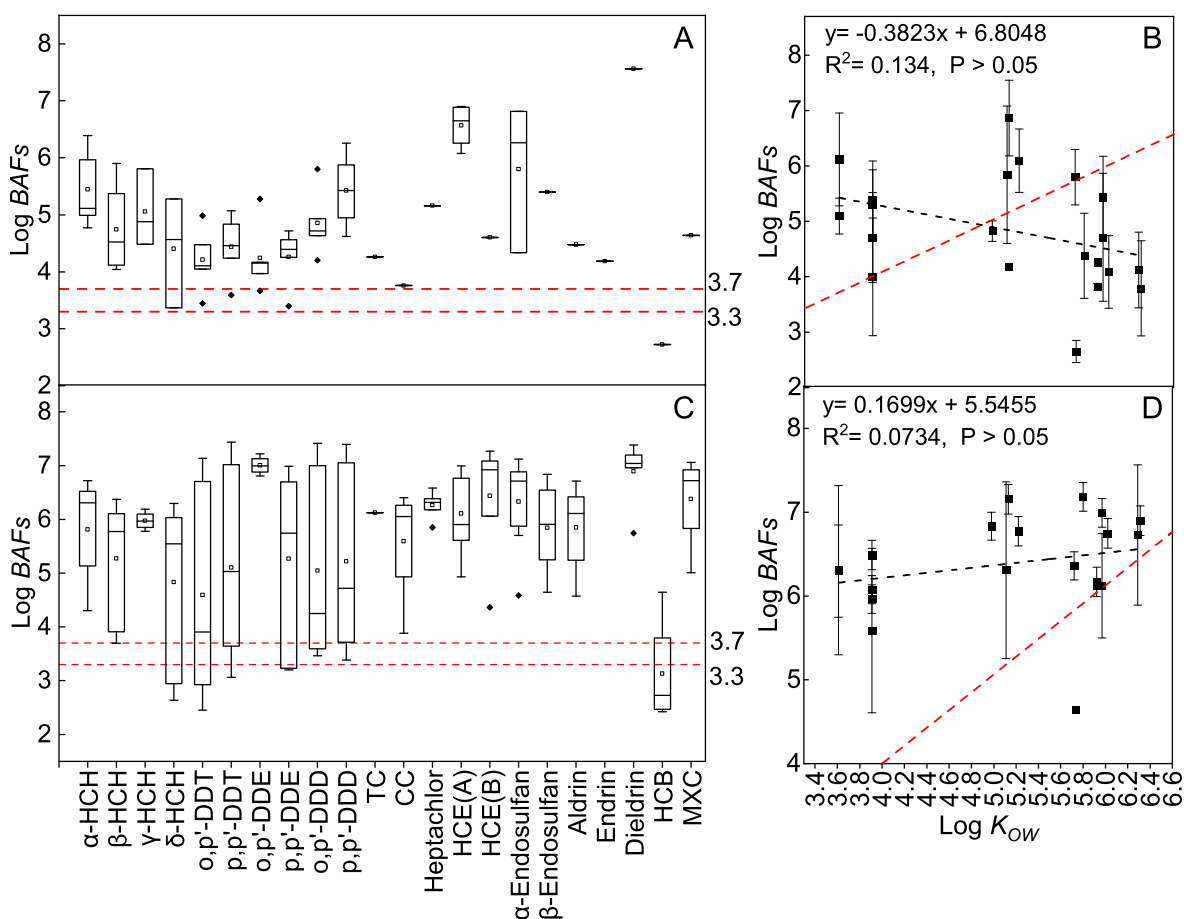


Fig. 4. The Log BAFs and the relationships between the Log BAFs and Log K_{OW} of the detected individual OCP congeners in coastal (A and B) and offshore (C and D) coral tissues. In the box plots, horizontal lines denote the 25th, 50th, and 75th percentile values. The squares denote mean values. The rhombuses mean outliers.

general, the Log BAFs were lower in coastal coral tissues than in offshore coral tissues. This may be caused by the effects of coral mucus in the coastal corals. As described previously, coastal corals may secrete more mucus than offshore corals in response to heavier environmental stress, and mucus may reduce the accumulation of coral tissues to pollutants (Zhang et al., 2019). The Log BAFs of OCPs in the present study were higher than those reported in fish (0.99–4.93) from Weizhou Island (Ding et al., 2019) and comparable to those in fish from the Singapore Strait (3.66–7.90) (Zhang and Kelly, 2018).

The Log BAFs were plotted versus Log K_{OW} for OCPs in Fig. 4. The 1:1 line between Log K_{OW} and Log BAFs is used for comparison (Wang and Kelly, 2018). Log BAF values above the 1:1 line indicated that the partitioning behavior of the compounds between water and lipids was unbalanced, further suggesting that bioaccumulation may be due to dietary intake (Borgå et al., 2005). Meanwhile, Log BAF values less than the Log K_{OW} values suggest disequilibrium of partitioning, potentially caused by degradation. The Log BAF values of all DDT isomers, CHL isomers, Drins isomers, HCB, and MXC in coastal corals were both lower than those of Log K_{OW} , suggesting that these compounds potentially degraded in corals and that other OCPs take up from both partitioning behavior and diet in coastal and offshore corals. For OCPs, a weak negative correlation occurred between the Log BAFs and Log K_{OW} in coastal corals ($R^2 = 0.13$, $p > 0.05$). This relationship was opposite to the previously reported positive correlation between Log BAFs and Log K_{OW} for fish samples from Weizhou Island, Qiantang River, Lake Chaohu, and an urban catchment in Singapore (Ding et al., 2019; Liu et al., 2016; Wang and Kelly, 2018; Zhou et al., 2008). BAF considers all accumulation routes, including ingestion of food and particulate matter to which chemicals adhere (Zhang et al., 2019). Thus, the correlation between Log BAFs and

Log K_{OW} is probably caused by the reduced bioavailability of high K_{OW} chemicals due to their sorption to particulate matter or dissolved organic carbon in seawater (Wang and Kelly, 2018). Coral mucus disturbance may be another reason for this correlation.

4. Potential effects of OCPs on reef communities

Coral reef ecosystems are very biologically productive and diverse but are very fragile and endangered. Human-made stresses to coral reefs have exceeded their regenerative capacity in many locations worldwide, causing dramatic shifts in species composition and resulting in severe economic loss (Bellwood et al., 2004). In the SCS, coral reefs have also suffered a dramatic decline, owing primarily to overharvesting, pollution, disease, and climate change (Burke et al., 2002; Yu, 2012). The water quality degradation caused by toxic substances, such as agrochemicals and industrial toxins, often poses a more significant degree of influence on corals (Lewis et al., 2009; Meehan and Ostrander, 1997). Additionally, the effects of anthropogenically derived chemical pollutants, which can enter reef systems through various pathways, are recognized as one of the main contributing factors to the decline in coral reef systems (Lewis et al., 2009; Ross et al., 2015). Among these pathways, in addition to enrichment and direct ingestion of OCPs by corals from the body surface, OCPs can also enter the corals through ingestion of carriers carrying OCPs, such as plastic debris (Koelmans et al., 2013; Montano et al., 2020). Indoor simulation experiments have proven that plastics may be important agents in the transport of hydrophobic contaminants to benthics (Koelmans et al., 2013; Teuten et al., 2007). The concentrations of OCPs on microplastics in the northern South China Sea were up to 174.4 times those in coastal corals (Dasgupta et al., 2021).

Therefore, the impact of compound pollution on corals should be further investigated in the future.

However, there is less information on the toxicity of these compounds to reef invertebrates to assess the chronic potential effects of OCPs on reef communities. A previous laboratory study demonstrated that Endosulfan at low concentrations (0.3–1.0 $\mu\text{g L}^{-1}$) in ambient water reduced the settlement and metamorphosis of the coral *Acropora millepora* by 50%–100% after 18 h of exposure (Markey et al., 2007). DDT and dieldrin are known to affect bird calcium deposition, resulting in reduced eggshell weight and thickness (Peakall, 1970; Porter and Wiewmeyer, 1969). As such, DDTs and dieldrin could be other factors influencing calcification and the maintenance of reef communities. Additionally, OCPs are likely to have a high affinity for lipid-rich eggs, leading to mortality or abnormalities of the eggs and larvae with reduced survival (van Dam et al., 2011). Furthermore, Markey et al. (2007) proposed some suspected adverse effects of OCPs on corals, including reduced dinoflagellate densities and bleaching, reduced larval metamorphosis and settlement, impaired photosynthesis, mortality in some populations, and reduced fecundity. Although the specific effects of these compounds on corals are currently unclear, these OCPs are likely to be one of the reasons for the decline in the number of corals from the SCS in recent years.

5. Conclusion

In this study, a significant decline in OCP concentrations was observed since 2005 in the seawater and air of the SCS. Σ_{22} OCPs were at a global low or medium level in the corals from the SCS. The seawater and coral samples collected near the continent displayed higher concentrations than those from the offshore coral reef regions. This may be related to land-based emissions and western boundary currents. The massive corals showed higher enrichment with OCPs than the branched corals. Dietary uptake and partitioning behavior were two modes of coral enrichment with OCPs. Coral mucus may play an important role in reducing the accumulation of OCPs in coral tissues. The calculated isomer ratios of OCPs indicated the ongoing usage of CHLs in the surrounding environment of the SCS, while other OCPs mainly came from historical usage. The OCPs were predominantly distributed in the gas phase, mainly attributed to the high atmospheric temperature and high volatility of these pollutants. Deposition from the atmosphere to seawater is the primary process of OCPs in the CRRs from the SCS. OCPs have a migration pattern of atmosphere-ocean-coral, increasing the environmental pressure on coral reef ecology.

The present study provides baseline data for future studies of OCPs in CRRs. Nevertheless, studies linking pollution monitoring and ecotoxicology are needed to assess the potential environmental effect of OCPs on corals in future studies. Additionally, analyzing the level of OCPs on a larger temporal-spatial scale is needed.

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, Resource, Validation, Funding Acquisition. Minwei Han: Investigation, Data curation, Editing. Yinghui Wang: Project Administration, Resource. Xueyong Huang: Investigation and Project Administration. Ruixiang Wang: Investigation, Data curation. Fang Liu: Investigation, Data curation.

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.

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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.2021.131711>.

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