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Concentrations and profiles of organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) in mussel tissues from Huaniao Island, coastal East China Sea

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Abstract: This study aims to investigate the distribution characteristics and bioaccumulation patterns of persistent organic pollutants (POPs) in different tissues of mussels from Huaniao Island along the coastal East China Sea. The concentrations of hexachlorocyclohexanes (HCHs), dichlorodiphenyltrichloroethanes (DDTs), chlordanes (CHLs), and polychlorinated biphenyls (PCBs) in various tissues (mantle, muscle, gonad, digestive gland, gill, and byssus) were determined using gas chromatography-tandem mass spectrometry (GC-MS/MS). A comparative analysis was conducted based on data normalized to dry weight and lipid weight. Among the target pollutants in mussels, the concentration of Σ DDTs was the highest (865 $\mu\text{g}/\text{kg}$ lipid weight), followed by Σ HCHs (24.3 $\mu\text{g}/\text{kg}$ lipid weight), Σ PCBs (13.8 $\mu\text{g}/\text{kg}$ lipid weight), and Σ CHLs (13.7 $\mu\text{g}/\text{kg}$ lipid weight). On a dry weight basis, concentrations increased from external to internal tissues. Conversely, an opposite trend was observed on a lipid weight basis. Congener-specific ratios [e.g., γ -HCH/ β -HCH, DDT/(DDD+DDE)] indicated that highly toxic substances mainly remained in external tissues. Degradation products or less toxic congeners tended to migrate towards the internal tissues. The fitting analysis between \log BCF and \log K_{ow} showed that the byssus exhibited the strongest bioaccumulation capacity (\log BCF_{max}=6.65), followed by muscle (6.41), while the gonad showed the weakest (6.05). Significant differences were observed in the accumulation patterns of POPs among different mussel tissues. Lipid content was an important factor influencing their distribution. Highly toxic compounds tended to be enriched in the external tissues, while internal tissues were dominated by less toxic metabolites. This study reveals the mechanisms underlying differential POP accumulation across mussel tissues. It provides a scientific basis for accurately assessing the role of organisms in pollutant transport and the associated marine ecological risks.

Key words: mussel tissues; HCHs; DDTs; CHLs; PCBs; BCF; Huaniao Island

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Polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) are a group of legacy persistent organic pollutants of global concern. They can travel long distances, are resistant to degradation, can bioconcentrate in fat-rich tissue and are highly toxic - causing cancer, birth defects, or mutations. PCBs were widely used as dielectric fluids in transformers, capacitors, and other electrical equipment due to their thermal stability and insulating properties^[1]. The historical production of PCBs was estimated at around 1.3 million tons, of which over 70% were tri-, tetra-, and penta-chlorinated biphenyls^[2]. OCPs such as DDTs, HCHs, and CHLs were extensively applied in agriculture and public health because of their effectiveness and low production costs^[3]. Globally, the annual OCPs production reached about 2 million tons. The implementation of international environmental policies, particularly the binding measures under the 1983 Stockholm Convention, coupled with stringent national restrictions on the use of PCBs and OCPs, has successfully curtailed major emission sources. Although these organochlorine pollutants (PCBs, DDTs, HCHs, and CHLs in this study) are now rarely used, they remain widespread in the environment. On one hand, studies had confirmed that PCBs and OCPs can be carried by atmospheric circulation to regions where they have never been produced or used, such as the Tibetan Plateau^[4], the Arctic and Antarctic^[5]. A global survey of background surface soils revealed large residues of PCBs. This demonstrated the long-range transport potential of PCBs and OCPs from source regions to remote areas. On the other hand, PCBs and OCPs can travel long distances by adhering to air particles in vapor form and deposit onto land through precipitation. When they enter into water, these organochlorine pollutants distribute between the water phase and suspended particles that act as sorbents, ultimately accumulating in sediments and living organisms^[6]. Due to their persistence and semi-volatility, these residues remain widely distributed and cycled within the food chain^[7]. Long-term atmo-

spheric monitoring showed a general decline in global organochlorine pollutant concentrations. This trend had been observed in South Korea^[8], China^[9], the United States^[10] and even the Arctic^[11]. In the Arctic marine environment, the average annual decline of organochlorine pollutants in seawater ranged from approximately 3% to 10%^[12]. However, unlike the concentration attenuation of these organochlorine pollutants in the environment, the concentrations in organisms did not completely show a corresponding downward trend. Some studies showed that the concentration of organochlorine pollutants in organisms has significantly increased in recent years^[13-14]. A Canadian study reported a significant increase in OCPs in marine biota^[12]. Bioconcentration and slow metabolism mean that some species at the top of the food chain remain significantly contaminated. Organisms thus may act as significant sinks of these organochlorine pollutants in aquatic environments.

Mussels are typical filter-feeding bivalves. Due to their sedentary lifestyle and high filtration capacity, they are highly sensitive to suspended particles and dissolved pollutants in water. As a result, they are widely used as indicator organisms for monitoring coastal pollution^[6]. Most existing studies focus on pollutant concentration in whole mussels^[15-17], or support carbon-neutral practices^[18]. However, there is still a limited understanding of how pollutants are distributed among different tissues in the mussel. Clarifying the tissue-specific distribution of pollutants is crucial for revealing their transport, transformation mechanisms, and ecological risks. Therefore, in this study, mussels from the Huaniao Island, coastal East China Sea were collected to determine the concentrations of OCPs and PCBs in their different tissues, and to analyze the composition and distribution characteristics. Bivalves are an important part of the global aquaculture industry, among which mussels are one of the main productive species of this group^[19]. Huaniao Island, located in the core aquaculture area of Zhejiang Province, is an important mussel production area

in China. Based on comparison of concentrations between the dry weight and lipid weight, isomer or congener ratio and enrichment factor in mussel tissues, the relationship between physical/chemical properties and composition/distribution was reflected. The findings will improve the understanding of accumulation, transformation and metabolism characteristics in mussels and their role as indicator species in environmental monitoring for persistent organic pollutants.

1 MATERIALS AND METHODS

1.1 Sampling and sample preparation

Seasonal mussel sampling was conducted in the raft-culture zone of Huaniao Island, coastal East China Sea, from January to November 2021 (a total of five batches were collected). The collected samples were labelled and frozen in sealed bags for storage. They were then transported back to the laboratory for further processing. Size-homogenized mussels per batch were dissected into byssus, mantle, gills, adductor muscle, and gonads, thus identical tissues were pooled to form one composite sample (a total of twenty-five samples were collected). Homogenized tissue composites were transferred into pre-numbered aluminum foil bags for wet weight determination. Samples were then lyophilized for 48h and reweighed gravimetrically to determine dry weights. Samples were pulverized with anhydrous sodium sulfate, then subjected to lipid extraction via solid-phase extraction (SPE) coupled with rotary evaporation and nitrogen blowing for lipid weight quantification. Extracts were purified via gel permeation chromatography (GPC) to remove lipids and co-extractives. The eluate was concentrated to 0.5 mL using rotary evaporation coupled with nitrogen blowing. Final extracts were obtained by passing the concentrate through solid-phase extraction (SPE) cartridges.

All experimental animal protocols in this study were reviewed and approved by the Institutional Animal Care and Use Committee (IACUC) of Shanghai Ocean University. All experimental procedures were strictly conducted in accordance with the Labor-

atory Animal Management Regulations and the Guidance on the Humane Treatment of Laboratory Animals issued by the Ministry of Science and Technology of the People's Republic of China, pertaining to the care and use of aquatic experimental animals, and every effort was made to minimize animal suffering and the number of animals used.

1.2 Instrumental Analysis

OCPs and PCBs were determined using an Agilent 8890 gas chromatograph coupled with a 7000D mass spectrometer (GC-MS/MS). The system was equipped with an electron ionization (EI) ion source. The GC-MS/MS system was operated in splitless injection mode. The injection volume was 1 μ L. High-purity helium was used as the carrier gas. The flow rate was set at 2.25 mL/min. The temperature of the ion source was 280°C. The transfer line temperature was also 280°C. The chromatographic separation was performed on a DB-5MS column. Its dimensions are 30 m in length, 250 μ m in outer diameter, and 0.25 μ m in inner diameter. The oven temperature program was set as follows. The initial temperature was 70°C, and it was held for 1 min. Then, the temperature was increased to 160°C at a rate of 10°C/min. Subsequently, it was raised to 280°C at a rate of 5°C/min and held for 5 min. Finally, it was increased to 300°C at a rate of 20°C/min and held for 5 min. The mass spectrometer was operated in multiple reaction monitoring (MRM) mode. Identification of target compounds was performed according to mass spectrometry scanning parameters. The qualitative and quantitative ion pairs for certain compounds are presented in Tab. 1, and the remaining compounds were also identified by referencing the quantitative ion and retention times of PCB standards. Due to the large number of PCBs, the specific parameters are not detailed here. Target chemicals comprised four groups of organochlorine compounds: hexachlorocyclohexanes (HCHs: α -HCH, β -HCH, γ -HCH, δ -HCH), dichlorodiphenyltrichloroethanes (DDTs: *p*, *p'*-DDD, *p*, *p'*-DDE, *p*, *p'*-DDT), and chlordanes (CHLs: trans-chlordane, cis-chlordane, heptachlor), and polychlorinated biphenyl (PCB). Total 41 PCB congeners were analyzed, cat-

Tab. 1 Mass spectrometry scan parameters of target chemicals (partial)

compound	quantitative ion	qualitative ion	retention time
a-BHC	219.00>183.00	219.00>147.00-219.00>145.00	9.972
r-BHC	219.00>183.00	219.00>147.00-219.00>145.00	10.392
b-BHC	219.00>183.00	219.00>183.00-219.00>145.00	10.582
d-BHC	219.00>183.00	219.00>183.00-219.00>147.00	11.107
Heptachlor	272.00>236.80	272.00>234.80-272.00>140.90	12.251
t-CHL	373.00>263.90	373.00>265.80-373.00>265.90	15.339
c-CHL	373.00>263.90	373.00>265.90-373.00>265.90	15.852
<i>p, p'</i> -DDE	246.00>176.10	246.00>211.00-246.00>150.10	16.723
<i>p, p'</i> -DDD	235.00>165.10	235.00>199.10-235.00>163.10	18.657
<i>p, p'</i> -DDT	235.00>165.10	235.00>199.10-235.00>163.10	20.634
PCB-52	292.00>221.90	220.00>150.10	24.436
PCB-118	326.00>255.90	328.00>256.00	33.148
PCB-138	360.00>289.80	362.00>289.80	34.485
PCB-153	360.00>289.90	360.00>289.90	35.940
PCB-180	394.00>323.80	396.00>325.90	38.745

egorized by chlorination level: Trichlorinated biphenyls (PCB-17, -18, -28, -31, -33), Tetrachlorinated biphenyls (PCB -44, -49, -52, -70, -74), Pentachlorinated biphenyls (PCB -82, -87, -95, -99, -101, -105, -110, -118), Hexachlorinated biphenyls (PCB -128, -138, -149, -151, -152, -153, -156, -158, -169), Heptachlorinated biphenyls (PCB -170, -171, -177, -180, -183, -187, -191), Octachlorinated biphenyls (PCB -194, -195, -199, -205), Nonachlorinated biphenyls (PCB -206, -208), Decachlorinated biphenyl (PCB -209).

1.3 Quality Assurance and Quality Control

Procedural blanks and duplicate samples were included every 12 samples to assess potential contamination and analytical accuracy. Surrogate recovery standards (TCMX and ^{13}C -PCB-208) were spiked, with limits of detection (LOD) calculated as three times the standard deviation of blank samples. Target chemicals were non-detected in either blanks or duplicate samples. Recovery ranged from 60.9% to 139.4% for DDTs, HCHs, and CHLs and from 72.6% to 126.5% for PCBs. In this study, Microsoft Excel 2019 was used for preliminary data sorting and summarization. All statistical analyses were performed using IBM SPSS Statistics 26.0. Graphs and charts

were generated using Origin 2021. All figures were standardized to ensure compliance with academic publication standards.

2 RESULTS

2.1 Concentrations and composition of OCPs and PCBs in mussels

The total concentration of DDTs was 865 $\mu\text{g}/\text{kg}$ lipid weight (lw). The concentrations of *p, p'*-DDT, *p, p'*-DDD, and *p, p'*-DDE were 396 $\mu\text{g}/\text{kg}$ lw (accounting for 46 % of the total DDTs, the same below), 76.3 $\mu\text{g}/\text{kg}$ lw (9%), 393 $\mu\text{g}/\text{kg}$ lw (45%), respectively. The DDT concentrations were lower than those reported in oysters from the Guangdong coast^[20] and in mussels from Japan^[21]. The total concentration of HCHs in mussel from the study area was 24.3 $\mu\text{g}/\text{kg}$ lw (as shown in Fig. 1). The concentrations of α -HCH, β -HCH, γ -HCH, and δ -HCH were 2.45 $\mu\text{g}/\text{kg}$ lw (accounting for 10% of the total HCHs, the same below), 8.75 $\mu\text{g}/\text{kg}$ lw (36%), 6.73 $\mu\text{g}/\text{kg}$ lw (28%) and 6.40 $\mu\text{g}/\text{kg}$ lw (26%) respectively. The HCH concentrations were comparable to those reported in oysters from the Guangdong coastal area^[20], lower than those found in mussels from the Marmara Sea coastal sites^[22]. β -HCH was the predominant congener of

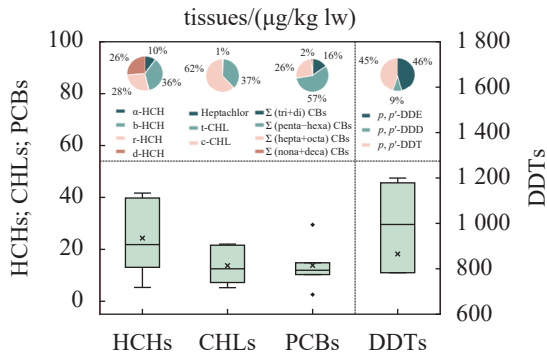


Fig. 1 Concentration of target pollutants in mussel tissues

HCHs in mussels. This is because β -HCH has low water solubility and is resistant to biodegradation. Additionally, α -HCH can be transformed into β -HCH in the environment^[23]. The total concentration of CHLs was 13.7 $\mu\text{g}/\text{kg}$ lw. The concentrations of heptachlor, trans-Chlordane (t-CHL), and cis-Chlordane (c-CHL) were 0.10 $\mu\text{g}/\text{kg}$ lw (1%), 5.07 $\mu\text{g}/\text{kg}$ lw (37%), 8.54 $\mu\text{g}/\text{kg}$ lw (62%), respectively. t-CHL and c-CHL were significantly lower than those found in seafood from the South China coast^[24]. The total concentration of PCBs was 13.8 $\mu\text{g}/\text{kg}$ lw. The concentrations of tri-, tetra-, penta-, hexa-, hepta-, octa-, nona-, and deca-chlorobiphenyls were 0.49 $\mu\text{g}/\text{kg}$ lw (accounting for 4% of the total PCBs, the same below), 1.67 $\mu\text{g}/\text{kg}$ lw (12%), 3.25 $\mu\text{g}/\text{kg}$ lw (23%), 4.58 $\mu\text{g}/\text{kg}$ lw (33%), 2.36 $\mu\text{g}/\text{kg}$ lw (17%), 1.20 $\mu\text{g}/\text{kg}$ lw (9%), 0.20 $\mu\text{g}/\text{kg}$ lw (1%), 0.06 $\mu\text{g}/\text{kg}$ lw (0.4%). PCBs in mussels in the study area were at a low level compared to other coastal cities in China^[25] and South Korea^[26] twenty years ago. This is related to the low industrial development in the study area and the effective ban over the past two decades regionally. The concentration of 7 Indicator-PCBs (PCB-28, -52, -101, -118, -138, -153, -180) was determined to be 0.039 $\mu\text{g}/\text{kg}$ lw (accounting for 18% of the total PCBs). Among the detected congeners, PCB138 (0.008 $\mu\text{g}/\text{kg}$ lw) predominated, corresponding to 4% of the total PCBs in mussels. It was followed by PCB110 (0.007 $\mu\text{g}/\text{kg}$ lw), PCB101 (0.006 $\mu\text{g}/\text{kg}$ lw), and PCB153 (0.006 $\mu\text{g}/\text{kg}$ lw). This congener distribution pattern aligns with that reported in mussels from the Iberian Peninsula^[27]. All these congener concentrations were lower than the levels

reported in mussels from the French Mediterranean^[28].

Overall, in this study, organochlorine pollutant residues in mussels are composed primarily of DDTs, with concentrations up to two orders of magnitude higher than other ones (HCHs, CHLs and PCBs). This finding is consistent with studies of bivalves from the South China coast^[20] and the mussel watch program in the Sea of Japan^[21]. The historical usage of HCHs far exceeded that of DDTs, the distinct physicochemical properties of HCHs—including higher volatility, rapid degradability, and low persistence—account for their considerably lower residual concentrations in mussels. China has ceased the production and use of DDTs. However, they were previously used as intermediates in the production of dicofol and antifouling paints for fishing vessels until 2009. During application, DDTs can adsorb onto colloidal particles and oil droplets, gradually settling into sediments where they accumulate. Meanwhile, they are stored in organisms in substantial quantities through bioconcentration. In China, both CHLs and PCBs were used for a shorter duration and at a much smaller scale compared to DDTs. CHLs had a relatively limited application range in China and were banned under the Stockholm Convention in 2009. PCBs were mainly used as industrial dielectric fluids with relatively low levels of production and use during 1960s-1970s. Besides, the equipment contained PCBs were buried in a centralized manner. Therefore, their residue levels in mussels are much lower than those of DDTs. As a whole, the concentrations of these organochlorine pollutants were well below the maximum allowable levels (MPLs) recommended in China and the US FDA action level for food and the Italy/EU EQS_biota standard^[29].

The isomer or congener ratio is commonly employed to identify the sources of target pollutants in the environment. In general, the value of α -HCH/ γ -HCH is often used to determine the source of HCHs. If the ratio is between 4 and 7, it indicates that the pollution comes from the historical residues of industrial Technical HCH. If the ratio is less than 1, it may be due to the recent input of the pesticide Lindan or the degradation of historical residues of industrial Technical HCH. In this study, the range of α -HCH/ γ -HCH

in the mussel tissues is 0.21-2.31, with an average value of 0.36. It indicates the possible recent input of Lindan. However, it is different from previous studies, in which historical residues rather than fresh input of both industrial Technical HCH and Lindan contributed to HCH load in soil. Considering the relatively "active" physicochemical properties of HCH congeners, significant compositional changes to source (α -HCH and γ -HCH) will occur during their transport and exchange among the atmosphere, water and organisms. Therefore, the ratio of α -HCH/ γ -HCH in mussels cannot be used to indicate the source of HCHs, as it does in soil. α -HCH and γ -HCH will gradually be converted into β -HCH in the environment through physical, chemical and biological processes such as volatilization, photolysis, hydrolysis and microbial degradation. The ratio of (α -HCH+ γ -HCH) to (β -HCH+ δ -HCH) can be used to determine the degradation (or residue) of α -HCH and γ -HCH in the environment. If the ratio is greater than 3, it indicates that there is still a large amount of α -HCH and γ -HCH in the environment. If the ratio is less than 3, it indicates that α -HCH and γ -HCH have undergone long-term degradation. The range of (α -HCH+ γ -HCH)/(β -HCH+ δ -HCH) is 0.43-1.38, with an average value of 0.75. It is indicated that the HCHs in the mussels of Huaniao Island mainly result from the pesticide residues in the early usage. This is consistent with previous studies of HCHs in the water and soil of Huaniao Island^[30]. DDTs in the environment mainly come from the use of industrial Technical DDT. The ratio (p, p' -DDE+ p, p' -DDD)/ p, p' -DDT suggests whether the DDT is a historical use residue (ratio > 1) or recent input (ratio < 1). In this study, the range of (p, p' -DDE+ p, p' -DDD)/ p, p' -DDT in the mussel tissues is 0.52-2.93, with an average value of 1.71. The ratios in the majority of tissue samples are larger than 1, indicating that DDT originated from historical residues. Historically, CHLs mainly come from Technical Chlordane. The ratio of t-CHL to c-CHL can be used to determine the degradation of CHLs in the environment. The range of t-CHL/c-CHL in the mussel tissues is 0.41-0.78, with an average of 0.62. The detected CHLs were largely due to historically depos-

ited residues. These residues have undergone substantial environmental transformation, where aerobic biodegradation acts as the dominant process. In this study, PCBs in mussel tissues were dominated by penta- to hepta-chlorobiphenyls, followed by tetra- and octa-chlorobiphenyls. Previous studies have indicated that PCBs in marine organisms along China's coast are dominated by penta- to hexa-chlorinated biphenyls^[31]. This pattern is not consistent with the specific commercial mixtures of PCBs production and usage during China's relatively short application history. In the mussel tissues, penta-chlorinated biphenyls, hexa-chlorinated biphenyls and hepta-chlorinated biphenyls account for more than 70% of total PCBs. Differently, previous studies have shown that PCBs in the water and sediments in the study area are mainly composed of tri-chlorobiphenyls, which may be contributed by the Yangtze River runoff and atmospheric deposition from historical use in China. Therefore, these ratio results appear to be more indicative of degradation/transformation processes rather than of the original pollution source.

In conclusion, the reliability of the ratio criteria established based on the source signals in environmental media (soils and water) to interpret the data for organisms is questionable. The composition in the environmental medium is mainly governed by physical and chemical processes. The enrichment characteristics within organisms are mainly governed by the processes of biological absorption and metabolism. The differences between biological and abiotic processes lead to deviation in their composition^[32]. Furthermore, selective absorption and distribution within different tissues in organisms can significantly alter the original composition^[33]. Thus, when using indicator organisms such as mussels for source apportionment, the interference of the hidden biogeochemical processes must be fully considered.

2.2 Concentrations of OCPs and PCBs by dry and lipid weight in mussel tissues

Fig. 2 illustrates the concentrations and trends of DDTs, HCHs, CHLs, and PCBs in tissues from the study area, expressed as lipid weight and dry weight,

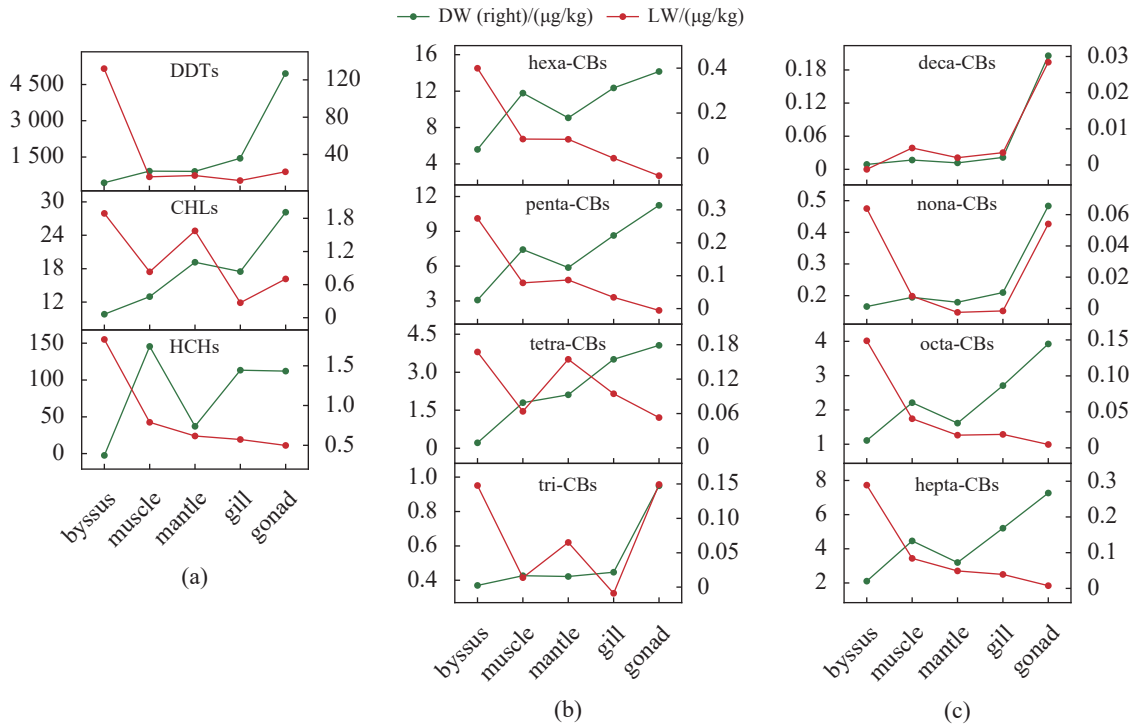


Fig. 2 The dry and wet weight concentration variations of the target pollutant among different tissues of mussels

respectively. Based on the result of concentration by lipid weight, DDTs were predominant in all tissue samples, with *p, p'*-DDE being the major contributing congener. The highest concentration of DDTs was found in the byssus (5 130 $\mu\text{g}/\text{kg}$ lw), followed by the gonads (962 $\mu\text{g}/\text{kg}$ lw). The concentrations were 737.44 and 683.03 $\mu\text{g}/\text{kg}$ lw detected in the mantle and muscle, respectively, while the lowest value 433 $\mu\text{g}/\text{kg}$ lw occurred in the gills. The highest concentration of HCHs was observed in the byssus (155 $\mu\text{g}/\text{kg}$ lw), significantly decreased in the muscle (43 $\mu\text{g}/\text{kg}$ lw), mantle (24 $\mu\text{g}/\text{kg}$ lw) and gills (19.2 $\mu\text{g}/\text{kg}$ lw), and reached the lowest one in the gonads (11.2 $\mu\text{g}/\text{kg}$ lw). Similarly, CHLs showed the highest concentration in the byssus (28 $\mu\text{g}/\text{kg}$ lw), followed by the mantle (24.8 $\mu\text{g}/\text{kg}$ lw), muscle (17.4 $\mu\text{g}/\text{kg}$ lw) and gonads (16.05 $\mu\text{g}/\text{kg}$ lw), and the lowest concentration in the gills (11.89 $\mu\text{g}/\text{kg}$ lw). PCBs exhibited the highest concentration in the byssus (41.66 $\mu\text{g}/\text{kg}$ lw), followed by the mantle (19.8 $\mu\text{g}/\text{kg}$ lw), muscle (18.6 $\mu\text{g}/\text{kg}$ lw), and gills (13.4 $\mu\text{g}/\text{kg}$ lw), with the lowest level in the gonads (9.34 $\mu\text{g}/\text{kg}$ lw). However, certain congeners (Tetra-, nona- and deca-chlorinated biphenyls) exhibit low overall concentrations, but accumulate to high

levels in internal tissues.

The red line designates wet weight concentrations, referenced to the left scale, while the green line represents dry weight values, scaled against the right axis.

In terms of concentrations by dry weight, the concentration of DDTs in gonads (124 $\mu\text{g}/\text{kg}$ dw) was significantly higher than that in other tissues including the mantle (21.9 $\mu\text{g}/\text{kg}$ dw), gills (29.9 $\mu\text{g}/\text{kg}$ dw), and muscles (22.2 $\mu\text{g}/\text{kg}$ dw) and the lowest one in byssus (9.70 $\mu\text{g}/\text{kg}$ dw). The concentration of HCHs was highest in muscle (1.74 $\mu\text{g}/\text{kg}$ dw) and lowest in byssus (0.37 $\mu\text{g}/\text{kg}$ dw). The concentrations in the mantle (0.74 $\mu\text{g}/\text{kg}$ dw), gills (1.44 $\mu\text{g}/\text{kg}$ dw) and gonads (1.43 $\mu\text{g}/\text{kg}$ dw) were at an intermediate level. The concentration of CHLs was the highest in gonads (1.91 $\mu\text{g}/\text{kg}$ dw), followed by mantle (1.01 $\mu\text{g}/\text{kg}$ dw), gill (0.84 $\mu\text{g}/\text{kg}$ dw) and muscle (0.38 $\mu\text{g}/\text{kg}$ dw). The lowest concentration was observed in byssus (0.06 $\mu\text{g}/\text{kg}$ dw). Similarly, the distribution of PCBs varied considerably among tissues, with the concentration ranging from 1.31 $\mu\text{g}/\text{kg}$ dw in the gonads to 0.11 $\mu\text{g}/\text{kg}$ dw in the byssus. Intermediate levels were observed in the gills (0.97 $\mu\text{g}/\text{kg}$ dw), muscles (0.77

$\mu\text{g}/\text{kg dw}$), and mantle ($0.52 \mu\text{g}/\text{kg dw}$).

For results from lipid weight, the highest concentrations of DDTs, HCHs, CHLs, and PCBs were all observed in the byssus. The lowest concentrations of HCHs and PCBs were found in the gonads, while CHLs and DDTs reached their minimum levels in the gills. Target pollutants showed higher levels in the byssus and decreased from external to internal tissues, indicating that a significant amount of these pollutants remained in the byssus rather than other tissues. Mussels use their byssal threads (foot filaments) to securely attach to submerged surfaces. This pattern is associated with the abundance of catechol structures such as 3,4-dihydroxyphenylalanine (DOPA) in mussel byssal proteins^[34]. The difference in lipid content and the lipophilic nature of organochlorides contribute to the completely opposite trend of concentration between tissues.

To sum up, the organochlorine pollutant distributions in tissues differed between dry weight and lipid weight results and can be categorized into two groups (as shown in Fig. 2). The first group of pollutants (including HCHs and tri-, nona-, and deca-CBs) showed consistent inter-tissue distribution patterns between dry-weight and lipid-normalized concentrations. The second group comprises CHLs, DDTs, and tetra- to octa-chlorinated biphenyls. The latter inter-tissue distribution has an obvious difference between dry-weight and lipid-normalized concentration profiles. Based on dry weight results, concentrations progressively increase from external to internal tissues. In contrast, a decreasing trend is observed based on lipid weight results. It is noted that the pollutants from the second group exhibit higher concentrations and greater toxicity compared with the first group in this study. Their profile more accurately represents the overall variation of organochlorine pollutants in mussels within the studied area. For lipid weight results, the inter-tissue distribution in the second group contradicts the expected mechanism of lipid-driven enrichment. This is evident from the highest concentrations occurring in the lipid-poor byssus and the lowest in the lipid-rich gonad. The inverse relationship suggests that

a congener-selective enrichment mechanism may be active in mussels, reducing the incorporation of highly toxic chlorinated compounds.

3 DISCUSSION

3.1 Distribution and transfer of OCPs and PCBs in mussel tissue

As the active insecticidal component, γ -HCH exhibits greater acute neurotoxicity. The HCH isomer γ -HCH is readily metabolized into the stable β -HCH. In this study, the γ -HCH/ β -HCH ratios in the byssus, muscle, mantle, gills, and gonads of mussels were 0.77, 0.75, 0.88, 0.74, and 0.75, respectively. The ratio of γ -HCH to β -HCH shows no significant difference across mussel tissues from external to internal, indicating a relatively uniform distribution of HCH isomers within the organism. Mussels possess a generally weak metabolic capacity for HCH isomers, which is insufficient to cause differential transformation among tissues, thereby maintaining a stable γ -HCH/ β -HCH ratio. Perhaps it is because HCHs have stronger water solubility and relatively lower toxicity that they flow more uniformly among the different tissues (Fig.3).

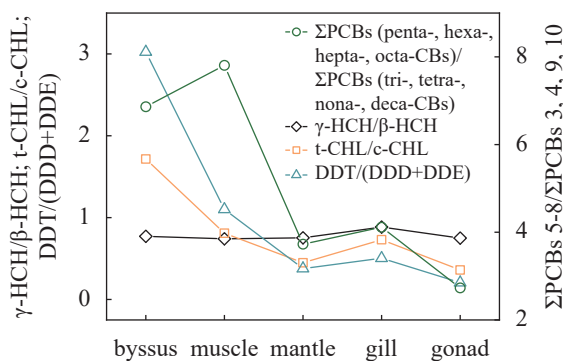


Fig. 3 Tissue-specific distribution characteristics of typical organic pollutant ratios in mussels

The ratios γ -HCH/ β -HCH, t-CHL/c-CHL and DDT/ (DDD + DDE) correspond to the left vertical axis, while the Σ PCBs (Penta-, Hexa-, Hepta-, Octa-CBs) / Σ PCBs (Tri-, Tetra-, Nona-, Deca-CBs) is referenced to the right vertical axis.

Among all its isomers, *p*, *p'*-DDT is the principal active constituent and exhibits the highest toxicity.

Technical-grade DDT historically contained 70%-80% *p, p'*-DDT. Under anaerobic conditions, it undergoes reductive dechlorination to *p, p'*-DDD, while aerobic environments favor oxidation to *p, p'*-DDE. The toxicity of *p, p'*-DDT is significantly stronger than that of its metabolite *p, p'*-DDE^[35]. *p, p'*-DDE exhibits greater environmental persistence than *p, p'*-DDT. In this study, the DDT/(DDD+DDE) ratios in the byssus, muscle, mantle, gills, and gonads of mussels were 3.02, 1.10, 0.38, 0.50, and 0.21, respectively. Maternal *p, p'*-DDT was found at higher proportions in external tissues (byssus and muscle) while being less concentrated in the mantle and gills. Upon reaching internal tissue, the gonad, it was metabolized into the more stable *p, p'*-DDE, or *p, p'*-DDE was preferentially absorbed. Chlordane was historically applied as a pesticide, herbicide, and termiticide. Its primary constituents include *c*-CHL, *t*-CHL and heptachlor. In this study, the *t*-CHL/*c*-CHL ratios in the byssus, muscle, mantle, gills, and gonads of mussels were 1.71, 0.81, 0.45, 0.73, and 0.36, respectively. The *t*-CHL/*c*-CHL ratio was below 1 in the majority of tissue samples. It is consistent with the results of squid (*O. bartramii*) from the Northwest Pacific Ocean^[36]. A situation similar to that of DDT/(DDD+DDE), the ratio of *t*-CHL to *c*-CHL was highest in the byssus, decreased significantly in the muscle and gills, and then reached the lowest values in the mantle and gonads. This suggests a greater presence of *t*-CHL in the external tissues, while more stable and abundant *c*-CHL is concentrated in the internal tissues. Studies have found that *t*-CHL is more prone to epoxidation and dechlorination in organisms than *c*-CHL^[37].

Compared to tri-/tetra-chlorinated and nona-/deca-chlorinated biphenyls, penta- to octa-chlorinated biphenyl congeners exhibited higher concentrations and greater toxicity. In this study, thus, their concentration ratios serve as indicators of the dynamic distribution profile among different tissues. The Σ PCBs (Penta-, Hexa-, Hepta-, Octa-CBs) / Σ PCBs (Tri-, Tetra-, Nona-, Deca-CBs) ratios in the byssus, muscle, mantle, gills, and gonads of mussels were 6.86, 7.81, 3.73, 4.11, and 2.77, respectively. The ratio

decreased from external to internal tissues, mirroring the distribution characteristic of DDTs and CHLs. This distribution profile indicates that the more toxic penta- to octa-chlorinated biphenyls are primarily concentrated in the byssus and muscle, while remaining lower in the gonads. Penta- to octa-chlorinated biphenyls are stable and resistant to degradation. It is still possible that tri- and tetra-chlorinated biphenyls, along with nona- and deca-chlorinated biphenyls, more readily enter metabolic pathways.

3.2 Bioconcentration profiles of OCPs and PCBs in mussel tissues

In this study, the tissue-specific distribution of organochlorine pollutants reveals a clear pattern: highly toxic parent chemicals predominantly accumulate in external tissues, exhibiting a decreasing gradient towards internal tissues. The main components in the gonads are relatively stable degradation products or congeners with lower toxicity. This spatial segregation suggests an active detoxification pathway within the gonads, where pollutants are metabolized into more hydrophilic and less toxic ones. This mechanism likely serves to protect critical reproductive processes, such as gametogenesis and fertilization, from chemical interference^[38].

Apart from the role of processes such as absorption, degradation and transformation, the physico-chemical properties of chemicals should be examined. The enrichment factor can be typically quantified using the Bioconcentration Factor (BCF). This factor is defined as the ratio of the concentration of a specific chemical observed in biota to its corresponding concentration in ambient water under field conditions. In this study, to better understand the extent of bioconcentration, the BCF of target pollutants were calculated. The water data used for calculation were the arithmetic mean concentrations of pollutants in water samples collected during 2020–2021^[39]. Fig. 4 illustrates the relationship between $\log K_{ow}$ and $\log BCF$ across five mussel tissue types, all exhibiting a characteristically parabolic distribution pattern. when $\log K_{ow} < 7$, BCFs increase with increasing K_{ow} , reflecting stronger partitioning into lipid-rich tissues;

beyond $\log Kow > 7$, BCFs decline in all tissues. In the $\log Kow$ range of 5-7, physical partitioning rather than biotransformation predominantly governs tissue distribution in mussels, whereas for $\log Kow > 7$, the large, highly hydrophobic congeners exhibit reduced membrane permeability and lower assimilation efficiencies. Further increases in hydrophobicity impair uptake leading to a disequilibrium between water and biota^[40-41].

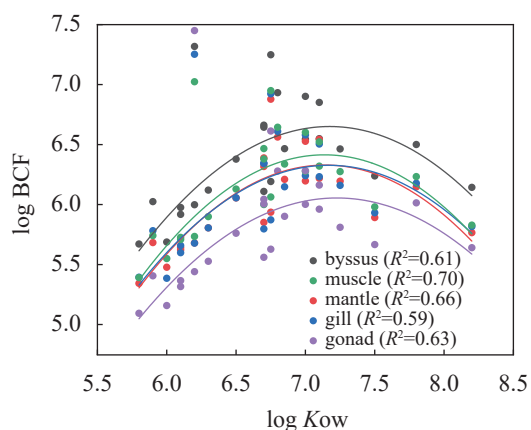


Fig. 4 Tissue-specific relationships between $\log BCF$ and $\log Kow$ for various mussel organs ($P < 0.05$)

The vertical positions of the fitted $\log BCF$ - $\log Kow$ curves for different tissues (as shown in Fig. 4) reflect differential bioconcentration capacities across tissue types. The highest curve trajectory observed in the byssus ($\log BCF_{max} = 6.65$) statistically demonstrates its strongest bioconcentration potential for target pollutants. This phenomenon is likely attributable to the byssus's unique composition, which features dopamine-rich structural proteins, metal-binding sites, and a high-surface-area gelatinous matrix. These properties collectively enhance its adsorption and complexation capabilities toward moderately to highly hydrophobic pollutants. The muscle tissue exhibits the second-highest curve trajectory ($\log BCF_{max} = 6.41$), followed by the gills and mantle at intermediate positions ($\log BCF_{max} = 6.33$). Notably, the gonadal curve maintains the consistently lowest profile ($\log BCF_{max} = 6.05$), reflecting its intrinsically reduced bioaccumulative capacity. It is postulated that mussels may possess a selective adsorption or exclusion mechanism. Through the com-

bined action of tissue structure and metabolic pathways, highly toxic organochlorines (such as CHLs, DDTs, and tetra- to octa-chlorinated biphenyls) are retained in the byssus and muscle tissues. In contrast, less toxic and more stable compounds are preferentially accumulated in the gonad.

The byssus and muscle provide dual protection. As the first point of contact with the external environment, the byssus acts as a primary barrier that captures and isolates highly toxic contaminants. Muscle tissue has a relatively high lipid content and is prone to accumulating lipophilic pollutants. The proportion of highly toxic substances increased^[42]. This leads to an increased proportion of highly toxic substances in these tissues, representing a secondary isolation step. Although the gonad has a high lipid content, it preferentially accumulates stable, low-toxicity metabolites to reduce toxic transfer and physiological damage to offspring, resulting in the lowest levels of highly toxic substances^[43].

4 CONCLUSIONS

Among the target pollutants of mussels in the study area, the concentration of DDTs ($865 \mu\text{g/kg lw}$) was the highest, followed by HCHs ($24.3 \mu\text{g/kg lw}$), while PCBs ($13.8 \mu\text{g/kg lw}$) and CHLs ($13.7 \mu\text{g/kg lw}$) were relatively low. DDTs were the predominant contaminants, with levels two orders of magnitude higher than other ones (HCHs, CHLs and PCBs). DDTs in mussels primarily composed of *p*, *p'*-DDT. β -HCH was the predominant congener of HCHs in mussels. CHLs exhibited *c*-CHL as their dominant component. PCBs were dominated by penta- to hepta-chlorinated biphenyls, with PCB138 being the dominant congener. All measured ratios consistently showed a progressive decline from external to internal mussel tissues. Byssus and muscle tissues are predominantly composed of more toxic congeners. Gonads mainly concentrate relatively stable degradation products or less toxic ones. This pattern indicates that highly toxic parent congeners remain predominantly in external tissues, relatively stable degradation products or congeners with lower toxicity accumulate toward the interior.

Analysis of the log BCF versus log *K*_{ow} relationship curve revealed that the byssus had the highest log BCF value (log BCF max=6.65). The muscle ranked next (6.41). The gonads exhibited the lowest value (6.05). Those results indicate that mussels exist selective sorption or exclusion mechanisms. Through the synergistic effect of tissue structure and metabolic pathways, mussel sequesters highly toxic parent congeners in external tissues, thereby reducing potential damage to internal tissues.

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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华东沿海花鸟岛贻贝组织中有机氯农药 (OCPs) 和多氯联苯 (PCBs) 的含量与特征

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摘要: 【目的】探究东海沿岸花鸟岛贻贝不同组织中持久性有机污染物的分布特征与生物积累规律。【方法】采集贻贝样本, 利用气相色谱-串联质谱 (GC-MS/MS) 检测各组织 (外套、肌肉、性腺、消化腺、鳃、足丝) 中六六六 (HCHs)、滴滴涕 (DDTs)、氯丹 (CHLs) 和多氯联苯 (PCBs) 的浓度, 并基于干重与脂重数据进行对比分析。【结果】贻贝中 Σ DDTs 浓度最高 (865 $\mu\text{g}/\text{kg}$ 脂重), 其次为 Σ HCHs (24.3 $\mu\text{g}/\text{kg}$ 脂重)、 Σ PCBs (13.8 $\mu\text{g}/\text{kg}$ 脂重) 和 Σ CHLs (13.7 $\mu\text{g}/\text{kg}$ 脂重)。干重浓度从外部组织向内部组织递增, 而脂重浓度呈现相反趋势。同系物比值 ([如 γ -HCH/ β -HCH、DDT/(DDD+DDE) 等] 显示高毒性物质主要滞留于外部组织, 降解产物或低毒同系物则向内部迁移。生物富集因子 (\log BCF) 与 \log K_{ow} 的拟合表明, 足丝的生物积累能力最强 (\log BCF_{max}=6.65), 肌肉次之 (6.41), 性腺最弱 (6.05)。【结论】贻贝不同组织对 POPs 的积累模式存在显著性差异, 脂质含量是影响其分布的重要因素; 高毒性物质倾向于富集在外部组织, 内部组织则以低毒代谢物为主。本研究揭示了贻贝各组织对 POPs 差异积累的机制, 为准确评估生物在污染物迁移中的作用及海洋生态风险提供了科学依据。

关键词: 贻贝组织; 六六六; 滴滴涕; 氯丹; 多氯联苯; 富集因子; 花鸟岛

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