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Bioaccumulation and trophic transfer of PAHs in tropical marine food webs from coral reef ecosystems, the South China Sea: Compositional pattern, driving factors, ecological aspects, and risk assessment

Minwei Han [a], Haolan Li [a], Yaru Kang [a], Huanxin Liu [a], Xueyong Huang [a], Ruijie Zhang [a,b,*], Kefu Yu [a,b,**]

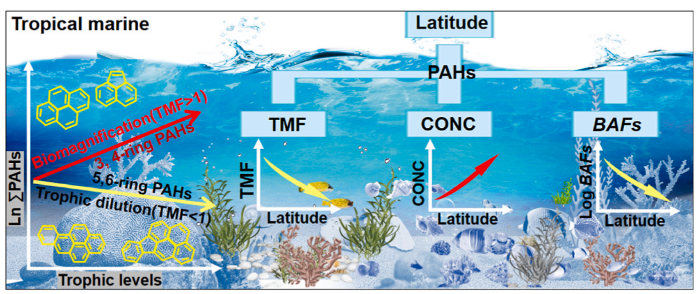
[a] Guangxi Laboratory on the Study of Coral Reefs in the South China Sea, Coral Reef Research Center of China, School of Marine Sciences, Guangxi University, Nanning, 530004, China

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

HIGHLIGHTS

- Reported PAHs' occurrence in the coral reef food web for the first time.
- Latitude affected PAHs' occurrence and bioaccumulation in the organisms.
- Latitude drove the PAHs' trophodynamics in aquatic ecosystem food webs.
- PAHs underwent trophic amplification in the food webs of coral reef ecosystems.

GRAPHICAL ABSTRACT



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ABSTRACT

Multiple environmental pressures caused by global warming and human activities have aroused widespread concern about PAHs pollution in tropical marine coral reef regions (CRRs). However, the trophodynamics of PAHs in the food webs of the CRRs and the related influence factors have not been reported. This study investigated the occurrence, trophic amplification, and transmission of PAHs in various organisms selecting between at least representative species for each level in CRRs of the South China Sea (SCS); revealed their driving mechanisms; and explored the trophodynamics of PAHs in the food web of the coral reef ecosystem. Results showed that more PAHs can be accumulated in the mantle tissue of Tridacnidae, and the proportion of mantle tissue of Tridacnidae increases with the increase of latitude ($y = 0.01x + 0.17$, $R^2 = 0.49$, $p < 0.05$). Latitude drives the differential occurrence level and bioaccumulation of PAHs in tropical marine organisms, and also affects the trophodynamics of PAHs in aquatic ecosystem food webs. PAHs undergo trophic amplification in the food webs of tropical marine ecosystems represented by coral reefs, thus further aggravating the negative environmental impact on coral reef ecosystems. The cancer risk caused by accidental ingestion of PAHs by

* 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. Southern Marine Science and Engineering Guangdong Laboratory (Zhuhai), Zhuhai, 519080, China.

E-mail addresses: 1727301003@st.gxu.edu.cn (M. Han), 2115392036@st.gxu.edu.cn (H. Li), 1827301008@st.gu.edu.cn (Y. Kang), 1432922513@qq.com (H. Liu), huangxueyong@gxu.edu.cn (X. Huang), rjzhang@gxu.edu.cn (R. Zhang), kefuyu@scsio.ac.cn (K. Yu).

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humans through consumption of seafood in CRRs is very low, but we should be alert to the biomagnification effect of PAHs.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are carcinogenic, teratogenic, and mutagenic persistent organic pollutants with a wide range of sources. Their ecological effects and potential toxicity have been reported worldwide (Bandowe et al., 2014; Liang et al., 2007; Lohmann et al., 2009). PAHs have been confirmed to be transmitted in various ecosystems in the aquatic environment through transformation, bioaccumulation, distribution, and exchange, and due to their strong hydrophobicity, they are preferentially assigned to the sediment, which greatly increases their half-life (Doick et al., 2005; Hu et al., 2014; Lin et al., 2013; Wan et al., 2007a). The resuspension of sediment under as a result of the physical disturbances or bioturbation caused by bottom-burrowing animals may promote the re-release of PAHs from the sediment to the water. Similarly, in tropical marine ecosystems, PAHs in sediments and water environments inevitably affect corals, benthos, and high trophic organisms through bioaccumulation (Carrasco Navarro et al., 2013; Ding et al., 2020a). Previous studies have confirmed that PAHs and other chemicals can be transferred to higher trophic organisms through food chains and webs (Ding et al., 2020a; Goutte et al., 2020; Loi et al., 2011). Laboratory studies have found that PAHs adversely affect marine organisms and disrupt normal biological metabolism (El-Alawi et al., 2002; Engraff et al., 2011). Previous studies on the transfer of PAHs between aquatic food webs mainly focused on freshwater ecosystems and high latitude marine ecosystems (Goutte et al., 2020; Loi et al., 2011; Qadeer et al., 2019; Wan et al., 2007a), and the results consistently showed that PAHs underwent trophic dilution among various food webs. However, there is a lack of systematic understanding of the migration and biotransformation of PAHs in the food webs of tropical marine ecosystems, especially in typical coral reef ecosystems. Recent studies have mainly focused on organophosphates (OPEs), organochlorine pesticides, metals, polychlorinated biphenyls, and novel brominated flame retardants (Ding et al., 2020a; Dromard et al., 2018; Fey et al., 2019; Hou et al., 2022) in tropical marine ecosystems, confirming that similar substances show different transmission and transformation modes in food webs in these ecosystems. According to previous studies on middle latitudes (An et al., 2020, Goutte et al., 2020; Loi et al., 2011; Qadeer et al., 2019; Wan et al., 2007a), a decrease in latitude (31.1–48.7° N) caused the increase of trophic amplification factors (TMFs) of PAHs in the food web, whereas for the food web of tropical waters, the TMF values are still unknown. Therefore, systematically revealing the TMFs and their driving factors in tropical waters is of great significance for indicating the ecological effects of PAHs in low-latitude waters.

Coral reefs are among the most important and typical tropical marine ecosystems, with extremely rich biodiversity and complex food webs. Food webs with complex nutritional relationships composed of various marine organisms, including corals, fish, benthos, and algae, jointly maintain the entire coral reef ecosystem. Unfortunately, the ecological function of coral reef ecosystems faces unprecedented challenges owing to the dual environmental pressure of continuous global warming and disruptive human activities (Hughes et al., 2017; McCulloch et al., 2012; Pandolfi et al., 2011). Various chemical pollutants continuously migrate into tropical marine ecosystems (Ding et al., 2020b; Han et al., 2019; Zhang et al. 2018, 2020), causing the entire coral reef ecosystem to continue to deteriorate, and the trace and ecological impacts of PAHs have also been reported (Jafarabadi et al. 2019, 2021; Jiang et al., 2018; Pandit et al., 2006). There are many coral reefs in the South China Sea (SCS) that are mainly distributed in the Xisha, Zhongsha, and Nansha islands. The complete ecosystem has important scientific and ecological value, which is unmatched by other marine ecosystems. Previous studies have found traces of PAHs in various environmental media of coral reef ecosystems in the SCS (Han et al., 2019; Li et al., 2019). Therefore, it is particularly important to perform trophic transfer of PAHs in the tropical marine ecosystem represented by coral reefs, which is of great significance for the ecological risk of early warning of PAHs in the entire tropical marine ecosystem. This study also investigated the trophic transfer and biotransformation of PAHs in low-latitude tropical marine ecosystems and endeavoured to systematically understand their nutrition dynamics in the food web from a global perspective. This study aimed to: (1) investigate the distribution of PAHs in fish and benthos in the CRRs of the SCS, including the Xisha, Zhongsha, and Nansha islands; (2) reveal the trophic transfer potential and trophodynamics of PAHs in the tropical marine food web; and (3) explore the driving mechanism of latitude on the occurrence and trophodynamics of PAHs in aquatic ecosystems. This study provides data for the first time on PAH trophodynamics in the coral reef ecosystem of a typical tropical marine food web and provides a new perspective for studying the driving factors of the trophic transformation mechanism of these chemicals on a global scale.

2. Materials and methods

2.1. Study area and sample collection

From May to August 2016, a total of 85 reef fish samples were collected from Xisha Islands (15°40′–17°10′ N and 111°00′–113°00′ E) and Huangyan Islands of Zhongsha Island (15°05′–17°13′ N and 117°40′–117°52′ E) in the SCS, and a total of 132 benthos were collected from Xisha Islands, Huangyan island, Xinyi Reef (10°13′ N, 114°12′ E) and Sanjiao Reef (10°54′ N, 114°03′ E) of Nansha Islands in the SCS. The detailed sampling points are shown in Fig. S1, and sample information is listed in Table S4 and Table S5.

Xisha Islands are distributed in about 5.0×10^4 km^2 of sea area with a total of 40 islands and reefs, and have the largest land area in the South China Sea (SCS) (Yu, 2012; Zhang et al., 2018). Coral reefs in the Xisha Islands are world-famous high-productivity areas, providing a habitat for 432 species of reef fishes, 41 species of benthonic crustaceans and 26 species of benthonic mollusks (Wang et al., 2011). The Xisha Islands are located in the tropical sea area, with a large number of corals and unique habitats, which makes the reef fish and benthos here diverse, many of which have high economic value. Huangyan Island is a large atoll with an isosceles triangle. It is 15 km long from east to west, 15 km wide from south to north, and 55 km coastline, covering an area of 150 km^2 (including the Lagoon). In the sea area near Huangyan Island, all kinds of hermatypic corals grow well and fish resources are rich, which is an excellent natural fishing ground. Xinyi Reef is heart-shaped, 1.40–1.85 km long, and nearly 2 km^2 (Yu, 2012). Sanjiao Reef is a large-scale atoll with an area of 16.1 km^2.

All samples of benthos and coral reef fish were obtained by diving and fishing. After sampling, each sample was photographed on a whiteboard with a scale to identify the species, and their length and weight were recorded. Details of marine biological samples are shown in Table S3 and Table S4. All marine organism samples were packed with aluminum foil and packed in polyethylene sealed bags, then put in cold storage, and then transported back to the laboratory for storage at −20 °C until analysis.

2.2. Sample preparation and analysis

The detailed pretreatment process of all biological samples is described in Text S2. All samples were treated as a whole except that the

Tridacnidae sample was divided into two parts. Each gram of dried marine biological samples was spiked with an 80 ng mixture of five surrogate standards including naphthalene-D8, acenaphthene-D10, phenanthrene- D10, chrysene-D12, and perylene-D12 (o2si, Charleston, United States) before extraction with a mixture of n-hexane and acetone (1:1, v:v) for 48 h using Soxhlet apparatus according to the previous method (Text S2) (Han et al., 2019; Zhang et al., 2021).

Sixteen PAHs from the US EPA priority-controlled list were selected as target compounds and were analyzed by Agilent 7890 gas chromatograph coupled to a tandem 7000C triple quadrupole mass spectrometer system (GC–MS/MS) (Text S2). Targeted PAHs with their physicochemical characteristics are shown in Table S1. The detailed GC and MS/MS parameters are shown in Table S2.

2.3. Quality assurance/quality control

In the analysis of samples, procedural blanks, experimental blanks and repeated samples were carried out for every 12 samples to ensure quality control. Experimental blanks were added to check for possible background contaminations during the experiment. The five deuterium-labeled PAHs (80 ng/sample) were added to each sample to monitor the recoveries. The instrument detection limit (IDL) is calculated according to the 3:1 signal-to-noise ratio of the corresponding analyte in the minimum standard. All target compounds in the procedure blank and experimental blank were not detected or lower than the detection limit of the instrument, which excluded the possible contamination of the biological sample pretreatment process and the filter itself. The relative standard deviation (RSD) between the replicate samples was 0.1%–23.9%. Recoveries of the surrogates generally ranged from 50% (naphthalene-D8) to 126% (chrysene-D12) of the spiked concentrations (Table S3). The recovery rate of naphthalene-D8 in some samples is too low, so this study will not be reported. The method detection limit (MDLs) of target PAHs was 0.03–0.27 ng/g.

2.4. Statistical analysis

In this study, Shapiro-Wilk test was used to check the normality of the data. When the normal test result is normal distribution, independent samples *t*-test (IBM SPSS Statistics 24.0) was used for each variable to test the difference between statistically significant; Otherwise, independent sample nonparametric test was used. The p-value of <0.05 and < 0.01 indicate significant and extremely significant respectively.

3. Results and discussion

3.1. Occurrence and bioaccumulation of PAHs in the coral reef ecosystem in the SCS

All 15 target PAHs (except NAP) were widespread in fish and benthos in various CRRs in the SCS. Combined with our previous studies on PAHs in corals (Han et al., 2019), the total concentration of the 15 PAHs ($\sum_{15}$PAHs, ng/g, dw) in corals (70.5 ± 108 ng/g, n = 16) > benthos (51.4 ± 28.7 ng/g, n = 110) > fish (43.9 ± 35.2 ng/g, n = 85) (nonparametric test, $p < 0.01$) (Fig. 1 and Fig. 2-a, b). Terrestrial PAHs from the periphery of the SCS migrate to the waters of the coral reefs through ocean currents and the atmosphere, enter the corals through bioaccumulation, and may enter other marine organisms in the coral reefs in a similar way.

Fish. Ten PAHs were detected in coral reef fish. The $\sum_{15}$PAHs ranged from 11.8 to 298 ng/g in coral reef fish in the SCS and were mainly composed of three-ring PAHs, accounting for 84 ± 4% (Fig. 2-a). As shown in Fig. S3, the coral reef fish of the Xisha islands presented significantly higher $\sum_{15}$PAHs (43.9 ± 35.2 ng/g, n = 76) than those of the Zhongsha islands (32.0 ± 8.49 ng/g, n = 9) (*t*-test, $p < 0.01$). Consistent with previous studies on the occurrence characteristics of PAHs in the water environment, they were controlled by human activities and terrigenous PAH pollution (Han et al., 2019; Hu et al., 2017; Lin et al., 2011; Zhang et al., 2021). The $\sum_{15}$PAHs were significantly different in different reef fish (Fig. 1), and these values were relatively low compared to other sea areas around the world (Table S9), including the Persian Gulf, Bohai Sea, and Hong Kong (Jafarabadi et al., 2019; Liang et al., 2007; Wan et al., 2007b). Consistent with previous studies (Jafarabadi et al., 2019), PAH concentrations in coral reef fish were significantly ($p < 0.01$) positively correlated with the lipid content (Fig. S4-b), while negatively correlated with the moisture content (Fig. S4-a). The $\sum_{15}$PAHs (38.7 ± 13.1 ng/g) was significantly higher in carnivorous fish than in omnivorous fish (28.6 ± 15.1 ng/g) (nonparametric test, $p < 0.01$) (Fig. S7-B).

Benthos. All 15 target PAHs were detected in benthos. The $\sum_{15}$PAHs ranged from 6.48 to 224 ng/g, with an average concentration of 51.0 ±

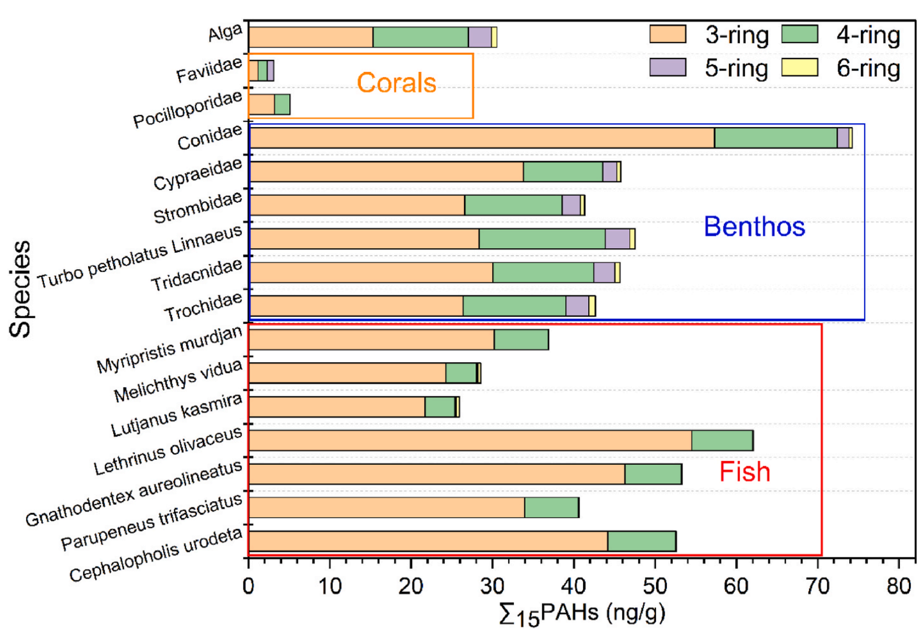

Fig. 1. Composition characteristics of average PAHs in various organisms in the CRRs of the SCS, the $\sum$15PAHs of corals are from our previous study (Han et al., 2020).

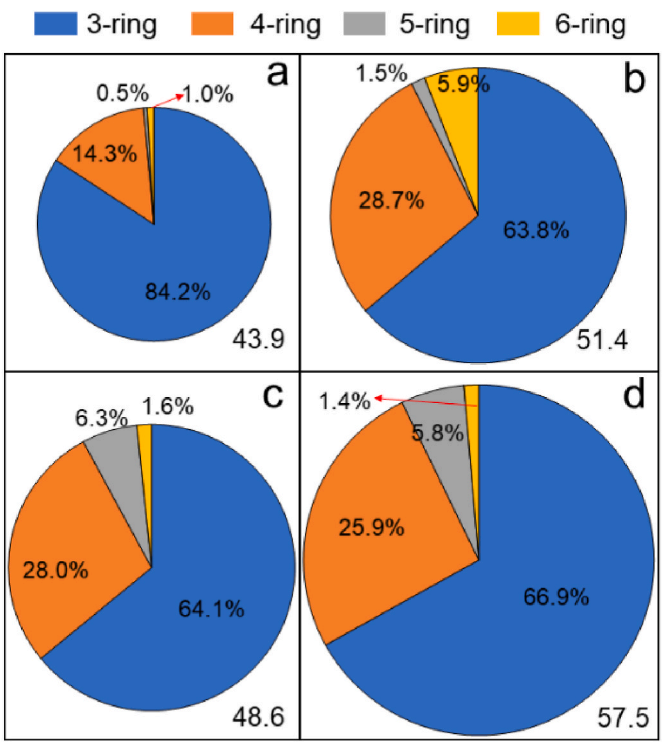

Fig. 2. Average $\sum_{15}$PAHs (ng/g) and composition of PAHs in fishes (a), benthos (b), and two tissues (c: general tissues; d: mantle tissues) of *Tridacnidae* in the same CRRs (Xisha Islands). The size of the pie chart represents the concentration level, which is marked in the lower right corner of the picture.

27.5 ng/g (Table S6). Similar to the fish described above, three-ring PAHs were the main contributors of $\sum_{15}$PAHs, but the proportion was slightly lower than that in fish, reaching 64 ± 9% (Fig. 2-b). The vast majority of benthos have low trophic level (TLs), single habitat and food habits, and live at the bottom of CRRs throughout the year. Benthos are not easily affected by seawater and other exogenous land-based PAH pollution (Billett et al., 1993; Solan et al., 2004), but unlike coral reef fish, they are usually greatly affected by sediments. The $\sum_{15}$PAHs in different benthos showed the following gradient: *Conidae* (74.2 ± 53.0 ng/g, n = 4) > *Trochidae* (52.9 ± 19.6 ng/g, n = 26), *Tridacnidae* (49.3 ± 14.0 ng/g, n = 14), *Turbo petholatus* Linnaeus (48.7 ± 23.1 ng/g, n = 11) > *Cypraea* (45.8 ± 12.6 ng/g, n = 6) > *Strombidae* (39.0 ± 11.3 ng/g, n = 5) (nonparametric test, $p > 0.05$). These values are relatively low compared with those of other sea areas around the world (Table S9), such as Great Wall Bay, Guadeloupe, Bohai Bay, and north China (Ding et al., 2021; Guo et al., 2015; Ramdine et al., 2012). The $\sum_{15}$PAHs were much higher in *Conidae* than in other benthos (Fig. 1), which is related to their feeding habits and lipid content (Fig. S4) (Cheung et al., 2007; Jafarabadi et al., 2020b; Zhou et al., 1999). As shown in Fig. S4d, the $\sum_{15}$PAHs in benthos were significantly positively correlated ($p < 0.05$) with the lipid content. Interestingly, the $\sum_{15}$PAHs were differentiated in various *Tridacnidae* tissues (Fig. 2-c, d). Twenty-five *Tridacnidaes* were divided into general and mantle tissue for their PAH analysis, and the $\sum_{15}$PAHs were higher in mantle tissues (57.5 ± 23.1 ng/g) than in general tissues (48.6 ± 24.2 ng/g) (nonparametric test, $p < 0.05$). Similar to corals, internal symbiotic zooxanthellae may be the primary factor affecting the difference in PAHs concentrations between the two tissues. A structure called the vitreous body at the edge of the mantle of *Tridacnidae* can gather light and provide good conditions for mass reproduction of zooxanthellae (Yan et al., 2011). Previous studies have confirmed that the presence of zooxanthellae increases the concentration of PAHs and other chemical pollutants in corals (Caroselli et al., 2020; Chen et al., 2021; Han et al., 2019). Zooxanthellae may adsorb more PAHs through bioaccumulation, and the photoinduced toxicity of PAHs reduces photosynthesis (Arossa et al., 2019; Jafarabadi et al., 2018; Rinkevich and Loya, 1983), which is not conducive to the synthesis of organic carbon, thus affecting the normal life activities of the host.

Bioaccumulation was used to characterise the enrichment effect of organisms on organic pollutants (Ding et al., 2020a; Han et al., 2019; Jafarabadi et al., 2020a; Krikech et al., 2022) and was calculated using the method described in Text S3. Based on the regulations of the European Chemicals Agency, calculated bioaccumulation factors (BAFs) showed that benthos have higher bioaccumulation capacity for PAHs than fish and corals (Han et al., 2020). The log BAF of $\sum_{15}$PAHs ranged from 1.72 to 4.29 (2.89 ± 0.41) in benthos, and 1.91 to 3.80 (2.70 ± 0.34) and 0.73 to 3.79 (2.34 ± 0.79) in fish and corals, respectively. Although the log BAF value of coral is much smaller than that of fish, coral is more likely to enrich high-molecular-weight PAHs (HMW-PAHs) than fish because of the presence of mucus (Han et al., 2020). In this study, eight PAHs showed bioaccumulation in benthos, whereas only three showed bioaccumulation in fish (Fig. S10). The occurrence level of HMW-PAHs in coral reef fish was much lower than that in benthos, which directly affected the bioaccumulation ability of fish. Thus, differentiated habitats may be the main driving factors of PAH concentration. Benthos can absorb PAHs in the surrounding water body, and a large number of high-ring PAHs are settled by suspended particles in seabed sediments, which cooperatively increases the bioaccumulation of HMW-PAHs by benthos. Consistent with a previous study (Bandowe et al., 2014; Han et al., 2019), the log BAF of fish and benthos showed a significant positive correlation ($R^2 > 0.306$, $p < 0.05$) with the log of the octanol-water partition coefficient (Kow) of PAHs (Fig. S11).

3.2. Driving mechanism of PAHs in organisms from the CRRs in the SCS

The occurrence level of PAHs in organisms from the CRRs in the SCS was uneven (Figs. 1 and 2), showing certain spatial and interspecific differences. There were also obvious differences in organism tissues across the regions. PAH levels in reef organisms are not only affected by individual species and habitats, but also by other potential factors, such as lipid levels and feeding habits (Cheung et al., 2007; Han et al., 2019; Li et al., 2019; Zhou et al., 1999). As mentioned above, both fat and water content are important drivers of PAH concentration differences in organisms of CRRs in the SCS (Fig. S4). Moreover, coral reefs in the SCS are located at different latitudes, and offshore distances and trophic modes may also be important factors affecting differences in PAHs. Latitude is one of the most important factors affecting the environmental occurrence of persistent organic pollutants (POPs), which controls the biogeochemical cycle and environmental fate of POPs in the surface system. We found that PAH levels in three coral reef fish (*Cephalopholis urodeta*, *Parupeneus trifasciatus*, and *Gnathodentex aureolineatus*) were related to latitude to a certain extent. In particular, PAHs in *Parupeneus trifasciatus* showed a significant positive correlation with latitude ($R^2 = 0.9791$, $p < 0.01$). The $\sum_{15}$PAHs or PAHs with different ring numbers in fish increased significantly with increasing latitude (Fig. 3a, e, i). The distance from the mainland, solar radiation, and temperature are likely to affect the PAH concentrations in fish. Affected by terrigenous PAHs and the diffusion of ocean currents, the Xisha islands with higher relative latitudes in the SCS are affected by more intensive human activities such as fishing, drilling, and ship navigation, so their waters may be polluted by heavier PAHs. Solar radiation is stronger at lower latitudes than at higher latitudes; therefore, PAHs in lower altitude waters are more prone to photolysis and become decreased (Nadal et al., 2006; Pereira et al., 2015). In addition, sea areas with relatively high temperatures are more likely to produce a trend of PAH volatilisation from the water body to the atmospheric environment, which is also one of the reasons for the low concentration of PAHs in low-latitude CRRs (Han et al., 2019; Zhang et al., 2021).

Although the level of PAHs in benthic organisms is also closely related to latitude, we found that this correlation is inextricably related

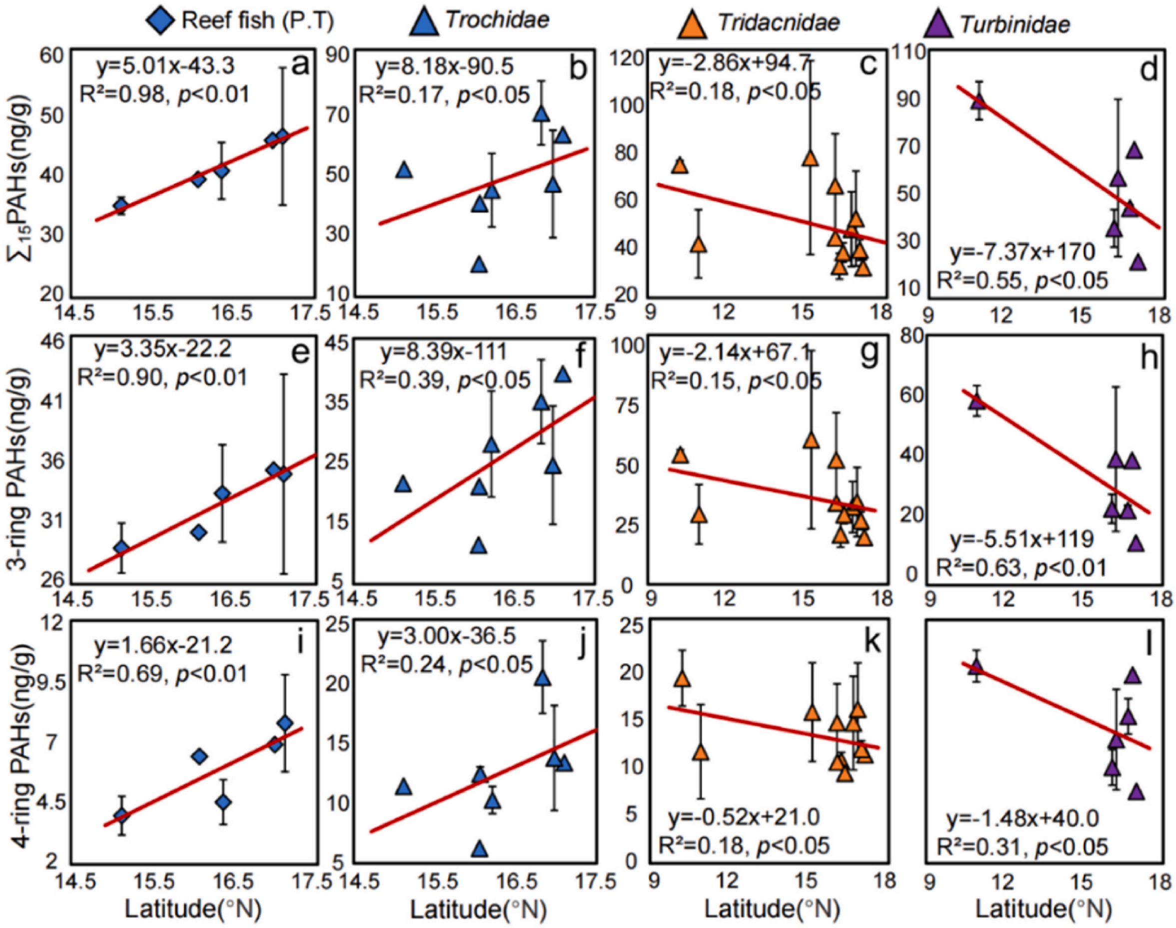

Fig. 3. Relationship between the organisms' $\sum_{15}$PAHs and the latitude of their sampling site.

to their species. The PAH concentration in *Trochidae* showed an obvious positive correlation with latitude ($R^2 = 0.1684$, $p < 0.05$) (Fig. 3b, f, j), whereas the relationships in *Tridacnidae* and *Turbinidae* were significantly negatively correlated ($R^2 = 0.1771$, $p < 0.05$, and $R^2 = 0.5488$, $p < 0.01$) (Fig. 3c, d, g, h, k, l). *Trochidae* generally crawl at the bottom of the ocean or between reefs and mainly feed on algae. Turf algae and macroalgae are likely to be the main foods of *Trochidae* because they are widely distributed at the bottom of the ocean or between reefs. Previous studies have confirmed that the distribution of these algae in the CRRs of the SCS increases significantly with an increase in latitude (Liao et al., 2021; Liao et al., 2019), and the food source of *Trochidae* is more abundant in the Xisha islands with their higher relative latitude, which undoubtedly increases the occurrence level of PAHs. *Turbinidae* are generally omnivorous, and not only prey on other mollusks, but also feed on large algae (Son et al., 2020; Zhou, 2014). The distribution of macroalgae at higher latitudes in the CRRs of the SCS is much higher than that at lower latitudes (Liao et al., 2021). Therefore, *Turbinidae* mainly feed on algae in the Xisha islands with their relatively high latitudes, whereas in the Nansha islands, relatively low latitudes may be more dominated by other benthos (Liao et al., 2021; Son et al., 2020). Intuitively, the $\sum_{15}$PAHs were much higher in benthos than in algae (Fig. 1), and the PAHs ingested by *Turbinidae* through feeding and exposure naturally increased. The Luzon Peninsula is close to the Nansha islands and the influence of terrigenous pollutants is unavoidable. Interestingly, the proportion of mantle tissue-storing zooxanthellae in *Tridacnidae* increased significantly with latitude ($y = 0.01x + 0.17$ $R^2 = 0.49$, $p < 0.05$) (Fig. 4), and the density of zooxanthellae also increased, which is consistent with previous studies on corals (Qin et al. 2019, 2020). Previous studies have found that zooxanthellae increased PAH

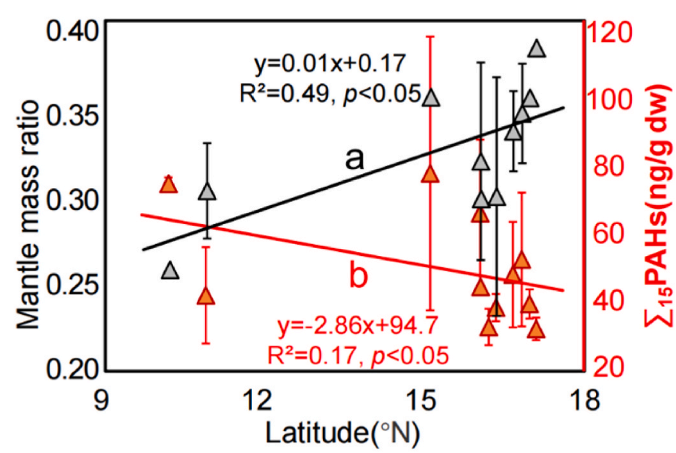

Fig. 4. Correlation between the mass proportion of mantle tissue in *Tridacnidae* and latitude (a) and the relationship between PAHs concentration level in the whole *Tridacnidae* and latitude (b).

concentrations in corals (Caroselli et al., 2020; Han et al., 2019), which contrasts the results of the lower concentration of PAHs in *Tridacnidae* in the CRRs at higher latitudes (Fig. 4). This anomaly is likely to be controlled by the strong human activity at the low latitudes of the SCS. Terrigenous PAHs related to oil spills and oil combustion in the nearby Luzon island migrate into the low-latitude waters of the SCS driven by monsoons and ocean currents (Zhang et al., 2021). As mentioned earlier, latitude partly affects the levels of PAHs in reef organisms, thereby affecting the bioaccumulation of PAHs by these organisms. As shown in Fig. S12, latitude may affect the bioaccumulation of coral reef fish in water, but it does not affect the benthos. The concentration of PAHs in coral reef fish increased with increasing latitude (Fig. 2a, e, i). Previous studies have found that the higher the concentration of PAHs in the CRRs in the relatively high latitudes of the SCS, the more PAHs enter the water environment from the atmospheric environment through water-air exchange (Zhang et al., 2021). In contrast, the lower the latitude, the faster the metabolism of PAHs by marine organisms, and when more PAHs are consumed by metabolism, a phenomenon occurs where the bioaccumulation ability of the organisms themselves is weakened, which may explain the negative correlation between log BAFs and latitude.

Human activities, climate factors, latitude differences, the fat content of organisms in the CRRs, and adequacy of habitat and food sources are all the driving factors leading to difference in PAHs concentrations in organisms. Under certain conditions, they will produce synergistic or antagonistic effects, and jointly dominate the occurrence and distribution of PAHs in coral reef organisms. In the context of global warming, if the problem of destructive environmental pressure caused by continuous human activities cannot be solved as early as possible, PAHs and other chemical pollutants will also threaten the whole coral reef ecosystem without exception. This not only has potential toxic effects on coral reef organisms but can also seriously affect the health of the tropical marine ecological environment.

3.3. Trophic structure of the food web in tropical marine coral reef ecosystems

Coral reef ecosystems are famous for their high productivity and biological richness. The coral reef biome is the most abundant and diverse in the ocean, with a very complex and diverse food chain and web. Similar to other ecosystems, abiotic factors such as sunlight, water, and other organisms in different reef areas constitute coral reef ecosystems. Many previous studies have focused on stable carbon (δ^{13}C) and nitrogen (δ^{15}N) isotopes of biological samples to determine the TLs of various organisms (Ding et al., 2020a; Goutte et al., 2020; Loi et al., 2011). A detailed analysis method is presented in Text S3. We measured the δ^{13}C and δ^{15}N isotopes of all coral reef samples to reveal the TLs of different organisms in coral reef regions (Table S10). The δ^{13}C values were 3.87 ± 0.42‰ (seaweed), 3.79 ± 0.50‰ (corals), 4.38 ± 1.53‰ (benthos), and 9.39 ± 0.94‰ (coral reef fish), and the δ^{15}N values were −15.5 ± 1.29‰ (coral reef fishes), −15.5 ± 2.45‰ (benthos), −11.2 ± 2.94‰ (corals), and −6.21 ± 0.43‰ (seaweed). The trophic status of food web organisms was analyzed using the stable nitrogen isotope, with seaweed (TL = 2.00 ± 0.08), corals (TL = 2.02 ± 0.12), benthos (TL = 2.15 ± 0.39), sponge (TL = 2.34 ± 0.15), and coral reef fishes (TL = 3.47 ± 0.24). δ^{13}C values are commonly used to identify food sources of organisms, and the δ^{13}C and δ^{15}N values of various organisms are significantly correlated, indicating that they probably belong to the same food web (Goutte et al., 2020; Loi et al., 2011). As shown in Fig. S13, δ^{15}N and δ^{13}C values showed significantly different groupings among species at different TLs. The feeding habits of organisms determine their δ^{13}C features, and the characteristics of δ^{13}C values in CRRs can correspond well to the feeding behaviour in the current food web. Combining the feeding and living habits of each organism and the calculated TLs, we determined the nutritional pyramid among organisms in the CRRs of the SCS (Fig. S14). Generally, marine animals in CRRs with phytophagous or filter-feeding habits, including corals, *Tridacnidae*, and other benthos, are at the lowest end of the pyramid. The TLs of *Tridacnidae* and hermatypic corals are almost the same, which is closely related to their living habits and the symbiotic zooxanthellae in their bodies. Hermatypic corals and *Tridacnidae* both have heterotrophic and autotrophic lifestyles (Goh and Todd, 2010; Waters et al., 2016). Their TLs were slightly lower than those of other benthos, which may be due to the reduction in the overall TLs of the hosts by a large number of symbiotic zooxanthellae in their bodies. The food sources of corals and benthos are relatively simple in composition, mainly including photosynthetic autotrophy and filter-feeding; therefore, they are easily controlled by solar radiation, temperature, and other abiotic factors. The food sources of high-TL coral reef fish are extensive and rich, mainly carnivorous, and are not easily or directly affected by these abiotic factors. Even if a food source is accidently affected by certain factors, it can be supplemented by other foods.

3.4. Trophodynamics in tropical marine coral reef ecosystems

Various chemicals in the marine food web undergo complex trophodynamic processes, including biomagnification, biotransformation, and biological metabolism (Fan et al., 2017). They are vulnerable to environmental factors, predator-prey relationships, and laboratory conditions, resulting in several uncertainties (Hou et al., 2022; Walters et al., 2016). TMFs are very useful for evaluating the bioaccumulation potential of high-trophic organisms. Therefore, this study further calculated the TMFs (Text S4) according to the method reported in a previous study (An et al., 2020; Goutte et al., 2020; Wan et al., 2007a). Linear regression was used to evaluate the correlation between PAH concentrations and TLs in coral reef organisms. $\sum_{15}$PAHs were significantly correlated with TLs, with a TMF value of 1.98 (Fig. 5a). Among the 15 PAHs analyzed, the concentrations of three-, four-, and five/six-ring PAHs showed significant associations with TLs, and the TMF values of the low-molecular-weight PAHs (LMW-PAHs) were greater than 1 (TMF = 8.43 for three-ring PAHs, and 4.49 for four-ring PAHs). The results show that these PAHs are in a trophic amplification state in a food web composed of algae, corals, benthos, and coral reef fish in the CRRs of the SCS (Fig. 5b and c). In contrast, five/six-ring PAHs undergo trophic dilution in a food web composed of algae, corals, and benthos (Fig. 5d). In principle, compared with LMW-PAHs, HMW-PAHs have higher Kow and physicochemical stability. They accumulate more easily in organisms with higher fat content or higher TLs. However, the occurrence level of HMW-PAHs in coral reef fish is much lower than that in benthos, which may be closely related to the degradation of PAHs and the metabolic level of organisms. In tropical surface sea water, PAHs are prone to photodegradation and biodegradation, whereas the biological metabolism level with higher TLs is higher, and the biological dilution or metabolism of highly liposoluble pollutants may be faster. Because HMW-PAHs were rarely detected in coral reef fish, it is impossible to predict the impact of coral reef fish on this result. The correlation results between the log values of the individual PAH concentrations and TLs are shown in Fig. S15. Five PAHs, including ACE, ACEY, FLU, FIUA, and PYR, undergo trophic amplification in the ecosystem food web of the CRRs in the SCS, whereas PHE, ANTH, CHR, and BkF undergo trophic dilution. These results show that PAHs in tropical marine food webs not only experience trophic dilution but can also undergo trophic amplification, and the change in latitude may explain to the differences observed with previous studies (An et al., 2020, Goutte et al., 2020; Qadeer et al., 2019; Wan et al., 2007a). Based on relevant research on the TMFs of PAHs in various food webs of aquatic ecosystems (An et al., 2020, Goutte et al., 2020; Qadeer et al., 2019; Wan et al., 2007a) at different latitudes around the world (Table S11), we found that latitude directly affects TMF values. As shown in Fig. 6, there was a significant negative correlation between latitude and TMF values of PAHs in the food web of each aquatic ecosystem. Therefore, PAHs are more prone to trophic amplification in tropical marine ecosystems, whereas PAHs in middle and high latitudes mainly manifest as a result of trophic dilutions. This phenomenon reminds us that we should always pay attention to the ecological harm and amplification effect of environmental problems caused by PAHs in various food webs of low-latitude and tropical marine ecosystems. Importantly, corals play an extremely important role in tropical marine ecosystems. High productivity has promoted the evolution of tropical marine ecosystems, even at low TLs of the food web (Xu et al., 2021). Influenced by the global migration

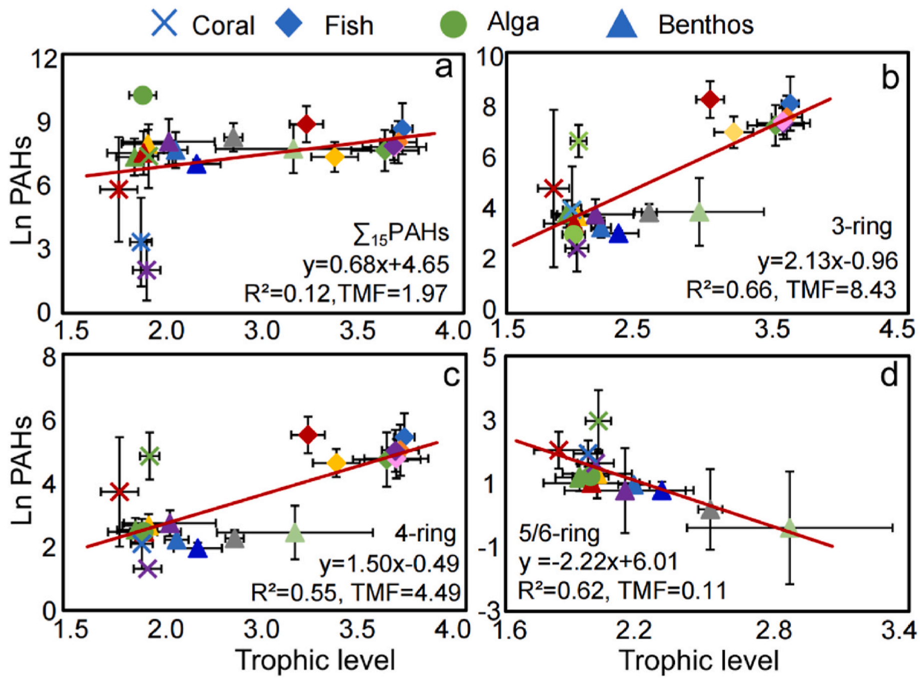

Fig. 5. Correlation between PAHs concentration and biological trophic levels (TLs), and trophic amplification factors (TMFs) of PAHs in the food web of CRRs, the SCS.

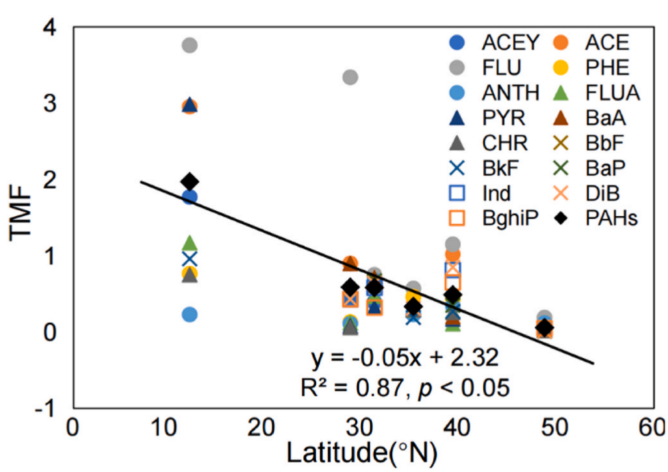

Fig. 6. Correlations between latitudes and PAHs magnification factors (TMFs) in aquatic ecosystem food webs around the world.

phenomenon and the distillation effect of PAHs, environmental problems and ecological effects have been confirmed in many previous studies. If PAHs pose a certain threat and ecotoxicity to corals, coral ecosystems at low latitudes are more vulnerable to PAHs owing to the amplification of PAHs that usually occurs at low latitudes. In coral reef ecosystems at relatively high latitudes, PAHs are likely to show trophic dilution, and previous studies have also found that OPEs show trophic dilution (Ding et al., 2020b), which is undoubtedly of great positive significance to coral reef ecosystems.

However, most PAHs undergo trophic amplification in low-latitude tropical marine ecosystems. Studies have confirmed that TMF is closely related to the hydrophobicity, biotransformation rate, and biological metabolism of PAHs (Goutte et al., 2020; Wan et al., 2007a). As shown in Fig. S16, there is a significant negative correlation between the TMF value and log Kow of PAHs, which indicates that the TMF of PAHs in the ecosystem of CRRs in the SCS is greatly affected by log Kow. Even if latitude directly leads to a difference in temperature, laboratory research has shown that the Kow of PAHs rarely depends on changes in temperature (Lei et al., 2000; Paasivirta, 2006). Therefore, the internal factors driving TMF changes in latitude should be further discussed in terms of biotransformation and biological metabolism (Xu et al., 2021). Corals may play a vital role in the food web of tropical marine ecosystems and their presence in the food web may largely change the nutritional mode of PAHs. Future research on PAHs in the food web of tropical marine ecosystems should fully consider the biotransformation and biological metabolism of corals on PAHs. This would help to fully understand the compound impact of driving factors related to PAH nutritional models in tropical marine ecosystems.

3.5. *Source and risks assessment*

In this study, the correlation between average concentration of 15 individual PAHs and the $\sum_{15}$PAHs in water, fish and benthos samples in the CRRs of the SCS. As shown in Fig. S15, correlation results showed that there was a significant correlation between the concentrations of 15 individual PAHs and the $\sum_{15}$PAHs in the three media in the CRRs of the SCS ($R^2 > 0.82$, $p < 0.05$), indicating that the source of PAHs in coral reef organisms is closely related to the PAHs in the surrounding environment, and organisms can enrich PAHs in the surrounding water through bioaccumulation. Therefore, based on previous studies, we used molecular diagnostic ratio and principal component analysis (PCA) to reveal the source of PAHs (Chen et al., 2012; Han et al., 2021; Hu et al., 2017; Kavouras et al., 2001; Lin et al., 2011; Yunker et al., 2002; Zhang et al., 2021). As shown in,Fig. S16 the diagnostic ratio results show that PAHs had different sources in fish and benthos. The ratio of ANTH/(ANTH + PHE) were mostly less 0.1, and the ratio of FLUA/(-FLUA + PYR) were less 0.5 for fish, suggesting that petroleum and petroleum combustion (Chen et al., 2012; Han et al., 2022b), the ratio of ANTH/(ANTH + PHE) and FLUA/(FLUA + PYR) were greater than 0.5, indicating that the PAHs in benthos are mainly contributed by combustion sources (Han et al., 2022a; Yunker et al., 2002). The source of PAHs was quantified using principal component analysis and multiple linear regression (PCA/MLR) by SPSS (Table S12). For fish, ACEY, ACE, PYR, and FLUA have a sizeable positive load on PC1, BkF and BghiP had large positive loads on PC3, indicating spilled oil and petroleum combustion, PC2 was mainly composed of FLU, PHE and ANTH, suggesting biomass combustion (Han et al. 2021, 2022b; Larsen and Baker, 2003). For benthos, PC1 and PC4 was mainly loaded with CHR, BaA, BbF, BaP, Ind, BaP and BghiP, attributed to petroleum-related products (combustion of liquid fossil and solid), PC2 and PC3 was mainly composed of FLU, PHE, ANTH, and PYR, which produced by the biomass combustion (Sofowote et al., 2008; Zhang et al., 2021). The corresponding PCs of the two types of samples were used as independent variables, and $\sum_{15}$PAHs was used as the dependent variable for MLR analysis using SPSS. The results shown PAHs produced by spilled oil and petroleum combustion has reached 72.7% and the biomass combustion accounted for 27.3% in coral reef fish, while petroleum-related products sources reached 74.6%, biomass combustion accounted for 25.4% in benthos. Although the above source analysis method can reflect the potential source of PAHs in the organisms in the CRRs of the SCS to a certain extent, the biological sample matrix is complex, and the use of the above method needs special caution.

Risk assessment can provide information on the potential health risks to humans through the consumption of reef fish and benthos in the CRRs of the SCS (Bandowe et al., 2014; Ding et al., 2012; Jafarabadi et al., 2019). Detailed methods are described in Text S5. The BaP_{eq} of $\sum_{15}$PAHs was 0.03 ± 0.07 ng/g (rage: 0.01–0.56 ng/g) in the fishes, and 0.61 ± 1.61 ng/g (rage: 0.01–18.5 ng/g) in the benthos in the CRRs of the SCS, most of them are lower than the maximum levels of 2.0 ng/g ww set by the European Union (Table S13 and Table S14). Considering the differences in seafood consumption, we used the EDI concept to assess and compare the health risks of toxic PAHs. The EDI of the average $\sum_{15}$PAHs via coral reef fish and benthos consumption were 112–692 ng g^{-1} d^{-1} and 91.4–489 ng g^{-1} d^{-1} in the CRRs of the SCS, respectively (Table S15 and Table S16). As the main seafood for human consumption, the unintentional intake of PAHs caused by eating fish is relatively higher than that of other coral reef organisms (Fig. 7). The EDI of PAHs caused by *Lethrinus olivaceus* consumption is the highest, followed by *Gnathodentex aureolineatus*, which reaches 266–1053 ng g^{-1} d^{-1} and 196–809 ng g^{-1} d^{-1} in different age groups (Table S17). The accidental intake of PAHs in the human body is directly related to daily consumption, and the daily intake of PAHs is determined by people of

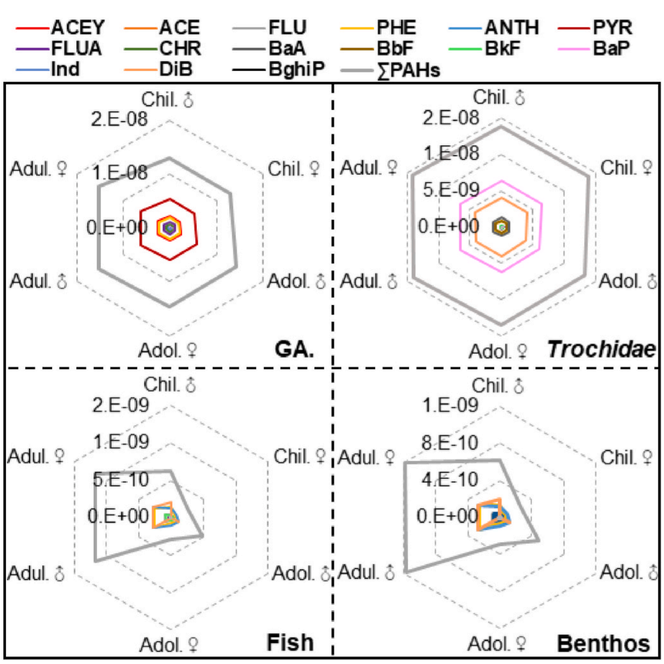

Fig. 7. Carcinogenic risk caused by consumption of seafood in the CRRs of the SCS ("♂" and "♀" represent male and female; GA. and *Trochidae* are the organisms with the highest carcinogenic risk among fish and benthos, respectively).

different ages and gender. Regardless of fish species, regional differences and other factors, the daily consumption of fish by adults is much higher than that of children and adolescents, and the accidental intake of PAHs by fish consumption is the highest. The benthonic organisms in this study are a few shellfish. Given the chance of people eating these shellfish, it is difficult to accurately estimate people's daily intake of each kind of benthos. As shown in Fig. 1, the concentration of PAHs in the *Conidae* is much higher than that of other benthos. Accidental PAHs intake caused by consumption of *Conidae* was the highest among all benthos, reaching 141 ng g^{-1} d^{-1} in children to 755 ng g^{-1} d^{-1} in adolescents. The EDI reflects the accidental intake of PAHs by different populations through the consumption of seafood. We further calculated the risk of Excess Cancer Risk (ECR) to evaluate the impact of accidental intake of PAHs on human health (Text S5). The ECR produced by lifetime exposure to the 15 PAHs via fish and benthos consumption in CRRs was calculated and compared with the acceptable guidance value of 1×10^{-6} set by EPA (Bandowe et al., 2014; Ding et al., 2012). The average ERC values of PAHs ingested by different ages consumers through consumption of fish from the CRRs of the SCS are from 2.5×10^{-10} to 1.2×10^{-9}, while the ERC values by consumption of benthos are from 6.0×10^{-9} to 1.4×10^{-8} (Fig. 7). Therefore, the ERC values are far lower than the acceptable guidance value of 1.0×10^{-6}. Even if the fish GA. and *Trochidae* with the highest BaP_{eq} in the same category are consumed together, these values (2.7×10^{-8} – 2.9×10^{-8}) are also lower than the acceptable guideline value of 1.0×10^{-6}. Hence, the excess carcinogenic risk from eating the fish and benthos from the CRRs of the SCS range from 27 to 29 persons per 1,000, 000, 000 people. The carcinogenic risk coefficient caused by accidental intake of PAHs in seafood in this area is very low and can be safely eaten.

4. Conclusion

This study investigated the occurrence and bioaccumulation of PAHs in tropical organisms of the CRRs, and discussed the trophodynamics of PAHs in the food webs of the CRRs, as well as the related influencing factors. The results showed that PAH concentrations in coral reef organisms in the SvdCS were relatively low compared with those in other sea areas worldwide. The difference in $\sum_{15}$PAHs in different organisms was caused by the synergistic effects of human activities, food habits, and lipid levels, which had a significant impact. More PAHs accumulated in the mantle tissue of *Tridacnidae*, and the proportion of mantle tissue increased with the increasing of latitude ($R^2 = 0.49$, $p < 0.05$). Benthos showed higher bioaccumulation potential than reef fish and corals. The vast majority of PAHs undergo trophic amplification in the food webs of tropical marine ecosystems represented by coral reefs. PAHs in coral reef organisms in the SCS mainly originate from a mixed source of oil spills and combustion (>72.7%). Risk assessment shows that the cancer risk to humans caused by consuming reef organisms is within the safety threshold, but considering the biomagnification effect of PAHs, greater attention should be paid to this issue going forward.

Credit author statement

Minwei Han: Formal analysis, Visualization, Methodology, Data analysis, Writing-Original draft preparation. Haolan Li: Investigation, Sample pretreatment, Editing. Yaru Kang: Editing, Sample pretreatment, Writing-Reviewing and Editing. Huanxin Liu: Investigation, Sample pretreatment, Editing. Xueyong Huang: Investigation, Sampling. Ruijie Zhang: Conceptualization, Methodology, Data curation, Validation, Supervision, Writing-Reviewing and Editing. Kefu Yu: Conceptualization, Funding Acquisition, Project Administration Resource, Supervision.

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.chemosphere.2022.136295.

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