Science of the Total Environment 847 (2022) 157442

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Environmental fate and effects of PAHs in tropical mariculture ponds near the northern South China Sea: Rainfall plays a key role

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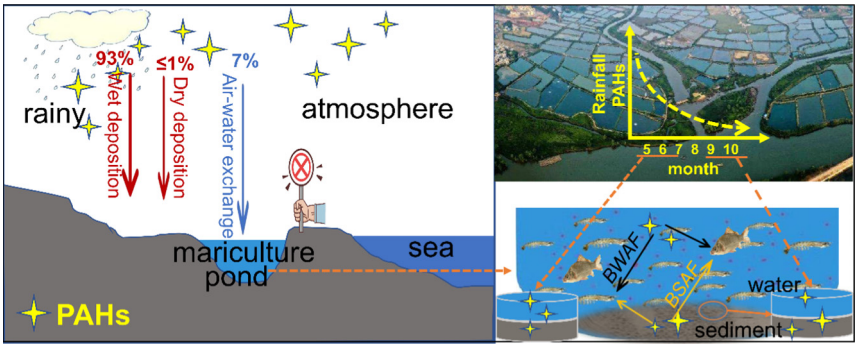
HIGHLIGHTS

- Continuously monitored PAHs in the mariculture ponds for the first time.
- Clarified PAHs' dynamic change, driving factors in tropical mariculture ponds.
- Rainfall played a key role in affecting PAHs' regional environmental behavior.
- Wet deposition dominated PAHs' migration process in mariculture areas.
- Tropical mariculture ponds affected PAHs' regional environmental fate.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Daqiang Yin

Keywords:
PAHs
Mariculture ecosystem
Dynamic change trend
Environmental behavior
Impact mechanism

ABSTRACT

The behavior and fate of PAHs are affected by multiple meteorological factors, but the main factors driving PAHs in tropical mariculture areas are still not clearly understood. This study continuously monitored PAHs in a few tropical land-based mariculture ponds, discussed their dynamic change trend, migration among the multiple media, and the relevant affected factors. Results indicated that PAHs were widely distributed in these environmental media, and the PAHs' concentration showed an obvious attenuation trend in the mariculture cycle. Wet deposition brought overwhelming majority atmospheric PAHs (92 % ± 5.7 %) to the aqueous system, and >72 % of these PAHs came from oil combustion-related sources and biomass combustion. Compared with the natural sea areas in the same region, mariculture ponds sediment could be changed from a sink at the early stage to a secondary release source of PAHs at the late stage of the rainy season, which intensifies the bioaccumulation of PAHs and the risk of edible carcinogenesis of aquatic products. Our research revealed that rainfall drove the occurrence and environmental behavior of PAHs in the tropical mariculture areas, while land-based mariculture ponds ecosystem affected the regional environmental fate of PAHs and weakened their transmission to the marine environment from land.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are typical persistent organic pollutants (POPs) composed of two or more benzene rings (Haritash and Kaushik, 2009; Kim et al., 2013). They come from a wide range of sources, arise from various human activities and natural emission processes, and

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http://dx.doi.org/10.1016/j.scitotenv.2022.157442
Received 25 May 2022; Received in revised form 9 July 2022; Accepted 13 July 2022
Available online 25 July 2022

have existed in various environmental media in the process of continuous migration, transmission, and transformation. Most of them are toxic, genotoxic, mutagenic, and carcinogenic, which has attracted continuous attention all over the world (Idowu et al., 2019; Kim et al., 2013; Xu et al., 2012). Previous studies have found that PAHs can enter the aquatic ecosystem through the exchange phenomenon of water-gas and water-sediment interface, and then expose aquatic organisms to the water environment with a considerable high concentration of PAHs (Chen et al., 2016; Han et al., 2021; Han et al., 2020; Jafarabadi et al., 2017; Li et al., 2006). Due to the characteristics of high-fat solubility, more PAHs will enter aquatic organisms through bioaccumulation, to increase the harm caused by PAHs, and can be transmitted between food webs, resulting in more serious ecological threats and toxic effects (Han et al., 2022; Han et al., 2020; Haritash and Kaushik, 2009; Jafarabadi et al., 2019).

The climate in tropical and subtropical regions is generally humid with sufficient light, and the temperature is the highest from July to August. Meanwhile, annual rainfall in most areas is >800 mm, and the rainy season generally lasts from April to September, and the rainfall is the most concentrated from June to August. Mariculture areas are generally distributed on the coastal shoals of the land-sea interaction area, a typical land-based closed ecosystem, which is mainly engaged in the production activities of marine aquatic economic animals and plants, and are committed to providing basic guarantees for the development of local food industry and the food source of residents. However, with the intensification of human activities, coastal beaches are facing great environmental pressure, which also threatens the ecological security of mariculture areas (Feng et al., 2019). Inadvertent pollutants in industrial and agricultural production activities, wastes caused by people's daily life, and a variety of pollutants inevitably generated in the process of urban development, including PAHs, enter the mariculture area through rainwater scouring, river transportation, land-sea material exchange and atmospheric deposition (Wang et al., 2010b; Zhang et al., 2020a), and then migrate and transform in the mariculture ecosystem, resulting in a serious ecological threat to the regional environment. Pollutants may eventually cause potential harm to people's health through food exposure. Different from other natural aquatic ecosystems, the intuitive advantage of the closed mariculture pond artificial ecosystem is that it can objectively reflect the dynamic environmental fate and change trend of PAHs and other chemicals in a certain period time, reveal their degradation, transformation process and various environmental behaviors in the natural environment, and is a natural laboratory to reveal the biogeochemical cycle of PAHs in the tropical mariculture ecosystem. However, the closeness of the mariculture area also means its fragile stability, and the rainy season in tropical areas can often affect the whole mariculture area system. How does the continuous rainfall drive the environmental behavior and fate of PAHs in the mariculture pond system? Similarly, the characteristics of closeness may also play a role in blocking the diffusion of various pollutants such as PAHs. The length of Beibu Gulf coastline accounts for 8 % of China's total coastline, while the mariculture area in the study area reaches 227 km^2 (Oceanic Administration of Guangxi, 2017). From the perspective of the whole tropical mariculture area, what impact does the local large-scale mariculture area on the ecological effect and environmental fate of PAHs in the regional environment? Notably, the potential impact of rainfall in tropical mariculture areas and the impact of tropical mariculture areas on the fate of regional environmental PAHs are still unknown.

Taking the typical mariculture area of the northern South China Sea (SCS) as an example, this study systematically studied (1) the occurrence characteristics, dynamic changes and driving factors of 16 PAHs in various environmental media (surface water, dustfall, feed and biological samples) during mariculture; (2) the migration and transformation of PAHs in multi environmental media of closed mariculture ecosystem, the dynamic changes of environmental behavior and its driving mechanism; (3) the potential sources of PAHs and the risk of cancer caused by human consumption of seafood; (4) the impact of tropical mariculture area on the environmental fate of regional PAHs. It further provides the most important scientific value for indicating that the organic pollutants represented by PAHs are closest to the biogeochemical cycle in the real state, and provided certain guiding significance for further exploring the environmental fate and ecological effects of various chemical pollutants in the tropical mariculture ecosystem.

2. Material and methods

2.1. Study area and sample collection

Maowei Sea is located northwest of the SCS and is part of the Beibu Gulf. It is a semi-enclosed natural bay, with a total coastline of 120 km, a maximum depth of 29 km, and a total area of 135 km^2 (Wang et al., 2017; Zhang et al., 2020a). Qinjiang River and Maoling River are both rivers entering the sea. The intersection area of rivers and seawater provides sufficient nutrients for the seafood mariculture industry in the Maowei Sea, resulting in the rapid development and commercialization of valuable seafood in the region, such as oysters, prawns and green crabs; Unfortunately, driven by human activities, various harmful substances caused by various urban pollution and industrial and agricultural activities carried by rivers discharge (Hu et al., 2014; Wang et al., 2017; Zhang et al., 2020a), are threatening regional environmental security.

From May to October 2017, four typical mariculture ponds (P1–P4), two river points (R1, R2), a nearshore point (S1) near Maowei Sea were selected as the sampling sites for long-term monitoring of PAHs in the multiple environmental media (Supplementary materials, Fig. S1). Two polyurethane foam passive samplers (PUF-PAS), and two atmospheric dry and wet deposition (i.e., dustfall and rainfall) samplers were installed near the mariculture ponds P1 and P3, and a sediment traps were used to collect the sediment settled over a while (Fig. S2). The sampling interval at each site was about 20 days. The river points were upstream (R2) and downstream (R1) of Qinjiang River respectively. S1 was located in the diversion channel of the mariculture pond. In this study, 30 culture water samples, 18 river water samples, 8 pairs of rainwater samples (including dissolved and granular), 5 sediment samples, 8 atmospheric samples, 13 biological samples, and 2 feed samples were collected (Tables S2–S3). During sampling, water samples were collected in brown glass bottles presoaked in deionized water, and other samples were packed in tin foil sealed bags and stored in a pre-prepared 0 °C icebox, and then transported back to the laboratory as soon as possible and stored in a −20 °C refrigerator.

2.2. Chemical analyses

In this study, 16 preferentially controlled PAHs were extracted as target compounds. Their detailed information are shown in Table S4. Five deuterium-labeled PAHs (naphthalene-D8, acenaphthene-D10, phenanthrene-D10, chrysene-D12, and perylene-D12) were used as analytical surrogates. Before extraction, all the water, filters, PUFs, sediment and feed samples were spiked with 80 ng surrogates to determine the procedural recoveries. All rainwater samples were filtered by a 0.22 μm glass fiber filter membrane, and PAHs in the granular phase and dissolved phase were extracted separately. Detailed operation steps are presented in Text S2. Before analysis, 200 ng of hexamethylbenzene was added to each sample concentrated to 0.5 mL at the internal standard.

All the samples were analyzed using an Agilent 7890 gas chromatograph coupled to a tandem 7000C triple quadrupole mass spectrometer system (GC–MS/MS). The detailed GC–MS/MS operational parameters and temperature program for the PAHs analysis are presented in Table S3 and Text S2.

2.3. QA/QC

Four field blanks were placed at two water sampling points, one rainwater sampling point, and one sediment sampling point respectively. The contamination observed during sample collection, extraction and analysis were detected by adding program blanks, 2 duplicate samples, and 3 experimental blanks. Five deuteriums labeled PAHs were added to each sample before

extraction to monitor recovery. The recovery of NAP-d8 was in the range of 30 % to 45 % in the majority of samples. Therefore, NAP was not quantified in this study. Recoveries of the other four deuterium-labeled PAHs in water samples, sediments, atmosphere and biological samples were 72–115 %, 65–105 %, 70 %–110 % and 68–116 % respectively. The concentrations of 15 target PAHs in all laboratory blank and field blank samples were lower than the instrument detection limits (IDLs) or not detected. The relative standard deviation (RSD) of repeated samples is 8.2–18.5 %. In this study, instrument detection limits (IDLs) and instrument quantitation limits (IQLs) were calculated by 3 and 10 times of signal-to-noise (S/N) ratio, respectively. The method detection limit (MDLs) of 16 targets PAHs were shown in Table S4.

2.4. Data analysis

Data treatment is described in Supporting Information Texts S3–S5, and includes sediment-water distribution and fugacity coefficient of PAHs, flux estimation of air-water exchange, dry and wet deposition, and bioaccumulation factors of PAHs. All of the statistical analyses were performed using the statistical package (IBM SPSS Statistics 24.0), the Shapiro-Wilk test was used to judge the normality of data packets, and then an independent sample *t*-test or independent sample nonparametric test was used for comparisons. The *p*-value of <0.01 or <0.05 was regarded as highly significant or significant.

3. Results and discussion

3.1. Occurrence of PAHs in mariculture pond ecosystem of the Maowei Sea

In this study, 7–15 quantitative PAHs were detected in various environmental media in the Maowei Sea mariculture areas, indicating that PAHs are widely distributed in the Maowei Sea mariculture areas of the northern SCS (Table. 1). In general, >14 PAHs were detected in almost all environmental media, including pond water, river water, rainwater, dustfall, sediment, air and feed samples, and their detection rate ranged from 9 % to 100 %, in which all target PAHs could be detected in sediment, air and dustfall samples, indicating that these environmental media played a vital role in participating in the biogeochemical cycle of PAHs in this area. Seven and 13 PAHs were detected in fishes and shrimps, respectively, with detection rates of 13 %–75 %.

The concentrations of the individual PAHs congeners and the total concentration of 15 PAHs (Σ_{15}PAHs) in various samples are summarized in Table 1 and detailed in Tables S5–S9. For three types of aqueous samples, the Σ_{15}PAHs were significantly higher in mariculture pond water (66.9 ± 70.7 ng L^{-1}) than in the river water (49.5 ± 50.7 ng L^{-1}) and rainwater (42.5 ± 38.5 ng L^{-1}) (nonparametric-test, $p < 0.05$) (Tables 1 and S5–S7). In the diversion channel of the nearshore mariculture pond, the Σ_{15}PAHs (162 ± 108 ng L^{-1}) were much higher than that in pond mariculture water. Affected by obvious urban human activities, the Σ_{15}PAHs in the upper reaches (58.7 ± 59.3 ng L^{-1}) of Qinjiang River were significantly higher than that in the downstream (40.3 ± 38.2 ng L^{-1}) (nonparametric-test, $p < 0.05$) (Figs. 1 and S3-B). The Σ_{15}PAHs were lower in this study than in other mariculture zones in China, such as the northern Yellow Sea (111–997 ng L^{-1}) (Zong et al., 2014), Taizhou Bay (5869–997 ng L^{-1}) (Jiang, 2008). Among the 15 PAHs in these aqueous samples, 3-ring PAHs were dominant, reaching 57.3 % to 70.5 %. For 4 types of solid-phase abiotic samples, the Σ_{15}PAHs (dry weight, dw) in dustfall (1162 ± 948 ng g^{-1}) > mariculture pond sediment (98.0 ± 29.2 ng g^{-1}) > feed (55.0 ± 16.8 ng g^{-1}) (nonparametric-test, $p < 0.05$). The Σ_{15}PAHs ranged from 3.89 to 19.1 ng m^{-3} with an average concentration of 11.5 ± 5.06 ng m^{-3} in air samples. 3-,4-ring PAHs were dominant in sediment and feed, reaching 33.7 % to 40.1 % and 38.1 % to 41.7 %, respectively, while 3-,4-,5/6-ring PAHs were evenly distributed in dustfall (38.4 % ± 12.7 %, 29.2 % ± 3.86 % and 32.3 % ± 8.45 %, respectively). In addition, 3-ring PAHs are dominant in air samples, reaching 80.6 % ± 9.10 %. Compared with the aqueous medium, 5/6-ring PAHs increased significantly in solid-phase abiotic samples. For the organisms, the Σ_{15}PAHs averaged 49.9 ± 75.5 ng g^{-1} (range: nd–220 ng g^{-1}), and in the shrimps (58.6 ± 82.3 ng g^{-1}) were significantly higher than that in the fishes (4.97 ± 5.04 ng g^{-1}) (nonparametric-test, $p < 0.01$) (Table S9). The occurrence level of PAHs in organisms may be greatly affected by the organisms themselves so the concentration of PAHs in organisms varies greatly. 3-ring PAHs are dominant in biological samples, accounting for 62.8 ± 22.9 % of the Σ_{15}PAHs, indicating that mariculture pond seafood may be more likely to accumulate low molecular weight PAHs (LMW-PAHs), and degraduate or metabolize high molecular weight PAHs (HMW-PAHs).

3.2. Dynamic changes and driving factors of PAHs in mariculture ponds

PAHs in various environmental media of the Maowei Sea mariculture ponds show an obvious dynamic change trend (Figs. 1 and 2). Seasonal variation and human interference in the mariculture process may be the main driving factors of this changing trend. Overall, during the whole sampling period, PAHs concentrations in the water environment (mariculture pond and river water) decreased significantly (Fig. S3), which may be closely related to the rainy season in the Beibu Gulf and the change of emission sources, such as combustion sources (fire points) (Fig. S6). As shown by the fire points map (Fig. S6), the density of fire points were high among the dry season (Jan.–Mar. and Oct.–Dec.) and low in the rainy reason (April–September). Besides the decrease of fire points in the rainy season, driven by the rainy season, rainwater scouring and wet deposition brought a large number of land-based PAHs produced by urban industrial and

Table 1
Detection rate and concentration level of PAHs in environmental media in Maowei Sea mariculture areas.

	Pond water (ng L^{-1})	River (ng L^{-1})	Rainwater (ng L^{-1})	Sediment (ng g^{-1})	Feed (ng g^{-1})	Air (ng m^{-3})	Dustfall (ng g^{-1})	Fish (ng g^{-1})	Shrimp (ng g^{-1})
ACEY	85 %[a], 2.29[b]	69 %, 0.9	75 %, 1.55	100 %, 0.38	67 %, 0.58	100 %, 0.03	100 %, 0.01	17 %, 0.07	13 %, 0.05
ACE	73 %, 1.69	69 %, 3.05	75 %, 0.81	100 %, 0.42	67 %, 3.03	88 %, 0.02	57 %, nd	33 %, 1.59	50 %, 0.86
FLU	94 %, 5.28	100 %, 6.81	100 %, 3.82	100 %, 4.20	67 %, 3.51	100 %, 1.03	100 %, 0.03	50 %, 0.68	38 %, 1.70
PHE	100 %, 17.4	100 %, 20.9	100 %, 16.5	100 %, 20.9	67 %, 13.4	100 %, 3.35	100 %, 0.16	67 %, 2.00	75 %, 17.0
ANTH	94 %, 5.11	94 %, 3.45	75 %, 1.41	100 %, 1.15	67 %, 2.23	100 %, 4.82	71 %, 0.01	0 %, nd	38 %, 1.05
FLUA	100 %, 5.99	100 %, 4.49	100 %, 4.94	100 %, 10.4	67 %, 7.21	100 %, 1.25	100 %, 0.05	33 %, 0.33	75 %, 11.3
PYR	10 %, 20.2	100 %, 8.4	100 %, 10.9	100 %, 7.49	67 %, 7.44	75 %, 0.26	100 %, 0.05	33 %, 0.29	75 %, 6.11
CHR	85 %, 0.19	88 %, 0.13	100 %, 0.23	100 %, 2.87	67 %, 3.83	100 %, 0.31	100 %, 0.02	17 %, 0.01	50 %, 2.70
BaA	85 %, 0.78	94 %, 0.7	100 %, 1.28	100 %, 15.7	67 %, 4.80	100 %, 0.27	100 %, 0.04	0 %, nd	50 %, 15.6
BbF	61 %, 0.58	50 %, 0.33	100 %, 0.76	100 %, 13.2	67 %, 3.42	100 %, 0.14	86 %, 0.07	0 %, nd	25 %, 0.14
BkF	18 %, 0.07	13 %, 0.03	38 %, 0.11	100 %, 2.35	67 %, 0.93	100 %, 0.06	86 %, 0.03	0 %, nd	50 %, 0.61
BaP	52 %, 0.4	31 %, 0.09	88 %, 0.17	100 %, 10.9	67 %, 2.29	100 %, 0.03	86 %, 0.04	0 %, nd	38 %, 1.51
Ind	12 %, 0.14	6 %, 0.01	13 %, 0.05	100 %, 5.11	67 %, 1.94	88 %, 0.01	86 %, 0.06	0 %, nd	0 %, nd
DiB	9 %, 0.08	0 %, nd	0 %, nd	100 %, 1.83	0 %, nd	75 %, 0.01	86 %, 0.02	0 %, nd	0.3 %, 0.08
BghiP	52 %, 0.5	39 %, 0.18	100 %, 0.42	100 %, 6.26	67 %, 0.40	88 %, 0.02	100 %, 0.06	0 %, nd	0 %, nd
Σ_{15}PAHs	60.7 ± 62.7	49.5 ± 50.7	42.6 ± 38.6	103 ± 27.8	55.0 ± 16.9	11.5 ± 5.06	0.59 ± 0.38	4.97 ± 5.04	58.6 ± 82.3

[a] The percentage means detection rate.
[b] Represent the mean concentration of PAHs.

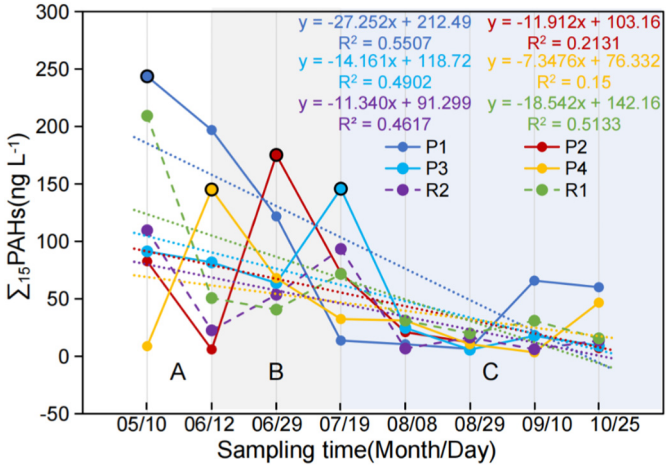

Fig. 1. Dynamic change trend of PAHs in culture water (solid line) and river water (dotted line) in Maowei Sea.

agricultural production activities of residents into various water environments. Due to the lack of material exchange with the external environment in the closed mariculture pond, the PAHs concentration in the pond water during the whole mariculture stage was higher than that in the rainwater and river water environment. As the rainy season continues, the Σ_{15}PAHs in the land surface and atmospheric environment gradually decreased (Figs. 1 and 2). PAHs concentration showed an obvious inflection point in some samples, which may be affected by point source pollution caused by human activities. According to the dynamic change trend of PAHs concentration in the water environment, it can be divided into attenuation period, transitional period, and stability period (Fig. 1). The attenuation period of

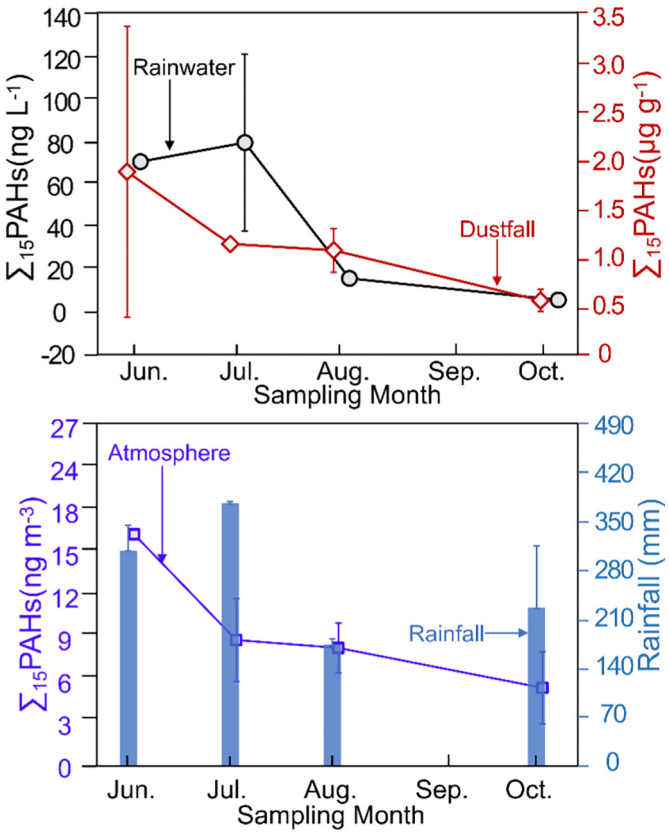

Fig. 2. Dynamic change trend of PAHs in rainwater, dustfall, atmospheric environment in Maowei Sea and the rainfall change in mariculture stage.

PAH concentration was from May 10 to June 12, during which Σ_{15}PAHs in the two water environments showed an attenuation trend. The initial stage of mariculture coincided with the alternation of the dry season and the rainy season. PAHs pollution sources caused by heating and land-based pollution in winter and spring in China's inland urban agglomeration are transferred to southern China driven by northeast wind, and then enter the land surface system through sedimentation and interface exchange, resulting in its seasonal high concentration in various environmental media (Li et al., 2015). Subsequently, the continuously rising temperature and enhanced solar radiation promote the degradation of PAHs in the water environment. The purification effect of the initial rainwater on the atmospheric environment gradually reduces the concentration of dissolved PAHs in the rainwater and plays a strong dilution effect on the surface water (Bae et al., 2006; Chate et al., 2003). The transition period of PAHs concentrations was from June 12 to July 19, which mainly reflected that the PAHs concentration shows an obvious upward trend in the river water body and a differentiation trend in the mariculture pond. Specifically, in the water environment of the four mariculture ponds, the PAHs showed a downward trend in P1 and P4, while an upward trend in P2 and P3. However, the two inflection points of high concentration PAHs on June 29 and mid-July were mainly due to the impact of point pollution. During our sampling period, P2 and P3 underwent changing the water before sampling respectively, and the river water from Qinjiang River caused a significant increase of PAHs concentration in their water bodies. Affected by the synergistic impact of local urban industrial and agricultural activities, scouring and sedimentation caused by the rainy season, and the sharp increase of fishing activities at the end of the closed season, the Σ_{15}PAHs in the river showed a slow upward trend from mid-June to mid-July (Fig. 1). After July 19, PAHs in various water environments showed an obvious attenuation trend, and finally gradually stabilized, but occasionally showed an upward trend, which can be called the stable period of PAHs. After the continuous purification of rainwater, most of the PAHs in the atmospheric environment and the dustfall were washed into the water environment, and entered the sediments or aquatic organisms through sedimentation or bioaccumulation, so that the concentration in the water environment tended to be stable gradually (Bae et al., 2006). Similarly, PAHs concentrations in rainwater, atmosphere and dust also showed an obvious downward trend (Figs. 2 and S5-A), which may be directly driven by the purification effect of rainfall. The rainfall is usually small at the beginning of the rainy season. Most of the PAHs in the atmospheric environment, dustfall and rainwater comes from the volatilization of the land surface system during the whole dry season, the PAHs concentrations are obviously the highest during this period. As the rainy season continues, PAHs in the atmospheric environment migrate to the land surface system with the scouring and sedimentation of rainwater. Purification is accompanied by rainfall, which directly promotes the reduction of PAHs concentration in rainwater, atmosphere and dustfall (Xu et al., 2013). The change of meteorological conditions leads to the change of PAHs concentration in the atmospheric environment, and the decrease of PAHs concentration in the atmospheric environment directly drives the decrease of dissolved PAHs concentration in rainwater. The purification effect of rainfall removes PAHs and other pollutants in the atmospheric environment (Bae et al., 2006; Chate et al., 2003), which is well confirmed in this study.

3.3. Impact mechanism of mariculture ponds on the fate of regional environmental PAHs

3.3.1. Partitioning of PAHs in the sediment-water system

Previous studies have found that the octanol/water partitioning coefficient (Kow) can properly describe the environmental fate of PAHs (Han et al., 2021; Zhang et al., 2020b). The greater the Kow value of PAHs, the easier it tends to adsorb from the aqueous phase to the organic phase. In the sediment-water system, adsorption-desorption is the main driving factor affecting the distribution of PAHs between sediment and water, while diffusion and equilibrium are mainly manifested in the deposition and resuspension of particulate PAHs and the diffusion of dissolved PAHs

between the system interfaces. Estimating the partition coefficient of PAHs in sediments and water can reveal its environmental behavior in the system, and the fugacity coefficient (*ff*) can further systematically reveal the diffusion equilibrium state of dissolved PAHs between water and sediment (Degrendele et al., 2016; Wang et al., 2015). Combining the two methods, this study systematically reveals the distribution of PAHs in the Maowei Sea mariculture water-sediment system. Here, PAHs below the detection limit were replaced by their MDLs.

As described in previous studies (Text S3) (Han et al., 2021, Zhang et al., 2020b), the average value of log *f* (*f* = *field* K_{oc}/*experimental* K_{oc}) was −0.87 to 0.98 in this study. On average, two-thirds of the 15 PAHs were in the adsorption state while the other one-third were in the desorption state (Fig. S8). However, for different mariculture stages, the environmental behavior of PAHs in the sediment-water system showed obvious differences. PAHs generally had the trend of sediment adsorption from the water body in the early stage of mariculture, but after August, it was mainly manifested in the release of sediment to the water environment (Fig. 3-A). As shown in Text S3, the calculation average *ff* values of 15 PAHs in this study were 0.05 to 0.50, and three high molecular weight PAHs (Ind, DiB, and BghiP) were <0.2, which had the trend of sediment enrichment from seawater (Fig. 3-B). Some PAHs (ACEY, FLU, and BbF) were released from sediments to water in August, while sediments were the sink of most PAHs in May and June at the initial stage of cultivation. Two methods consistently showed that PAHs in the Maowei Sea mariculture ponds ecosystem mainly adsorb PAHs in the water environment by sediments in the early stage of mariculture, while in the later stage, it mainly showed the secondary release of sediments to the water environment. Intuitively, this difference was closely related to the concentration difference of PAHs in sediment and the water environment. The concentration of PAHs in the water environment decreased significantly during the attenuation period (Fig. 1), the equilibrium state of PAHs in the sediment-water system was broken, resulting in the desorption of sediments to the water body. In addition, the influence of the rainy season on the distribution balance of PAHs in the sediment-water system cannot be ignored (Fig. 3-A), which may be mainly controlled by the dilution of rainwater.

3.3.2. Migration of PAHs between atmosphere and surface water system

Air-water exchange and atmospheric deposition drive the biogeochemical cycle of PAHs in the land surface system (Tsapakis et al., 2006; Zhang et al., 2006). Due to the closeness of mariculture ponds, compared with other surface ecosystems, the material exchange between them and the atmospheric environment is vulnerable to the surrounding environment and human activities. A classical two-film transport model (Chen et al., 2016; Li et al., 2006; Wu and Tao, 2021; Zhang et al., 2021) was used to estimate the air-water exchange flux of PAHs in this study (Text S4). Through the estimation of net exchange flux in various stages of mariculture, we found that the total flux of 15 PAHs showed negative flux (−635 to −114 ng m^{-2} d^{-1}) (Fig. 4-A and Table S11), indicating that the transportation of PAHs from the atmospheric environment to the mariculture pond area was the main control process of PAHs in this area. In general, with the decrease of atmospheric PAHs concentration, the trend of PAHs entering the water environment through the exchange of water-air interface in the whole breeding stage was gradually weakened, which was directly reflected in the decrease of net flux (Fig. 4-A). The flux of Σ_{15}PAHs was the highest in May at the beginning of breeding, reaching −630 ± 4.81 ng m^{-2} d^{-1}, which was mainly affected by the high atmospheric PAHs concentrations during this period. For individual PAHs, ANTH, PHE, and FLU were dominant in the total exchange flux of PAHs, accounting for >44.5 %, >30.6 %, and >5.87 %, respectively (Fig. 4-A and Table S11), which is consistent with the result that the gas phase PAHs in the atmospheric environment are mainly 3-ring PAHs. Meteorological and hydrological conditions affect the environmental fate of PAHs at the water-air interface and drive the exchange and distribution of PAHs between the interfaces (Wu and Tao, 2021, Zhang et al., 2021). As shown in Fig. 4-A, the rise of air temperature and water temperature in this study has a certain inhibitory or slowing effect on the air-water exchange, while rainfall, PAHs concentration in the atmosphere and the water environment directly promote the air-water exchange process of PAHs.

We also estimated the wet and dry deposition flux based on the method detailed in Text S3, and the results are shown in Table S12. Wet deposition flux (1188–21,824 ng m^{-2} d^{-1}) was significantly higher than the dry deposition flux (14.3–48.9 ng m^{-2} d^{-1}) in the corresponding month (nonparametric-test, $p < 0.01$), indicating that the wet deposition in the mariculture cycle controlled the subsidence of PAHs in this regional atmospheric environment. Dry deposition and wet deposition showed a similar change trend during the whole mariculture period (Fig. 4-B and C), and the maximum dry deposition and wet deposition of the 15 PAHs occurred in May and June, respectively. Dry deposition flux directly confirms the variation trend of PAHs concentrations and temperature in the dustfall of the mariculture cycle. Low temperature accelerates the settlement of PAHs (Cheng et al., 2018), while rainfall directly drives the wet settlement of PAHs (Ma et al., 2013; Tsapakis et al., 2006). The high precipitation in June directly caused the PAHs wet flux to reach the peak, and then with the decrease of rainfall and PAHs concentrations in rainwater, wet deposition gradually decreased and tended to be stable. Dry deposition does not particularly sensitively and positively responded to rainfall, but the purification effect of rainwater also reduces the dry settlement of PAHs, and this mitigation effect gradually weakens with the continuation of the rainy season.

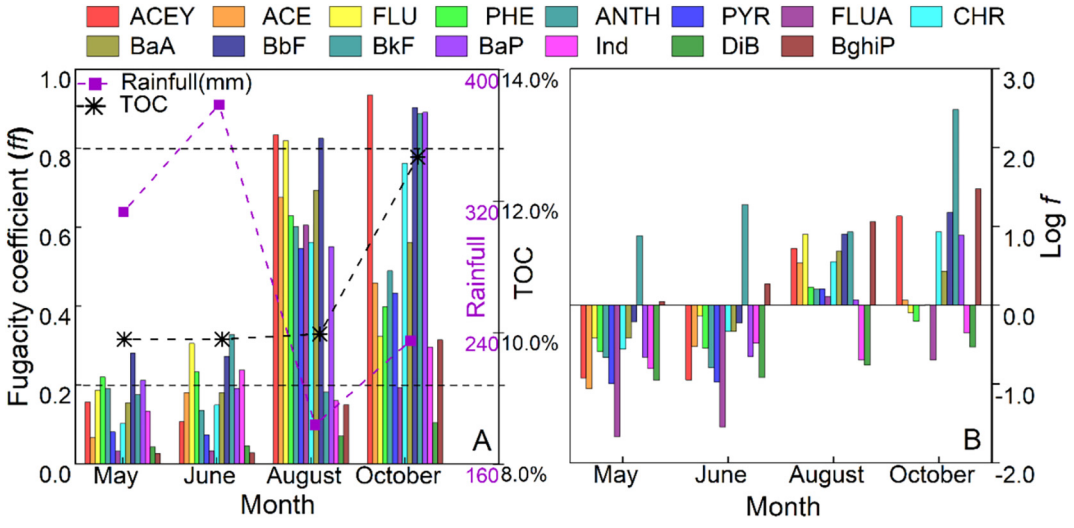

Fig. 3. Calculated *ff* (A) and Log *f* (B) of PAHs in the water-sediment system in mariculture pond of the Maowei Sea.

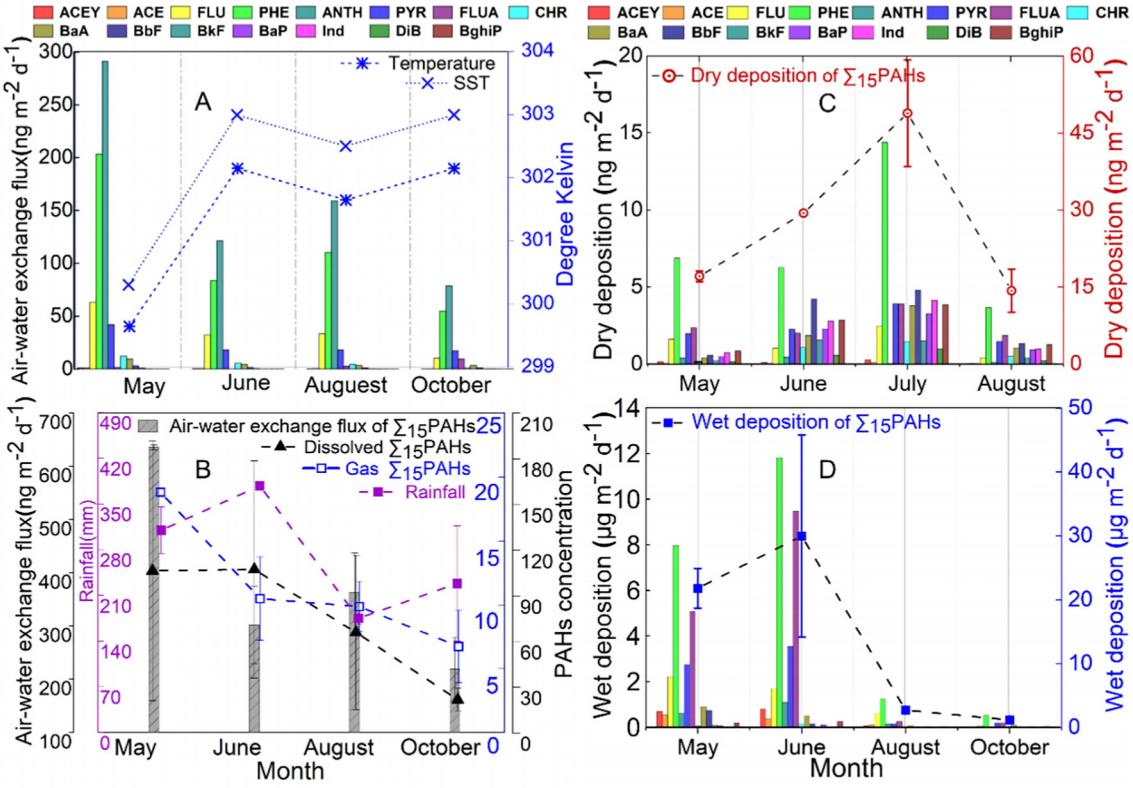

Fig. 4. Air-water exchange flux (A, B), dry deposition (C), and wet deposition (D) of PAHs in mariculture pond of the Maowei Sea.

Among the 15 PAHs, dry deposition and wet deposition flux of PHE were the highest in the whole mariculture cycle. Besides, 5/6-ring PAHs to dry deposition flux (35.2 % ± 11.0 %) was significantly higher than that to wet deposition (4.31 % ± 1.84 %), while the contribution of 3-ring PAHs to wet deposition (59.0 % ± 10.8 %) is significantly higher than that to dry deposition (36.3 % ± 10.2 %), which was closely related to the physicochemical properties of PAHs (Fig. S9). The greater the molecular weight of PAHs, the stronger their hydrophobicity. Therefore, PAHs with high molecular weight in the atmospheric environment are more likely to be adsorbed by suspended particles to produce dustfall precipitation, while LMW-PAHs mainly enter the rainwater in a dissolved state and then migrate to the surface system in the form of wet deposition. Typical areas such as rivers and mariculture ponds become the sinks of PAHs in the atmospheric environment, especially for LMW-PAHs. For the atmosphere-land system, wet deposition (92 % ± 5.7 %) is the key driving factor to control the migration of PAHs from the atmospheric environment to the surface system in the Maowei Sea mariculture areas, while the contribution of air-water exchange (6.9 % ± 5.1 %) is negligible (Fig. 5). However, the contribution of air-water exchange increased from August to October, which was mainly caused by the reduction of wet deposition of PAHs caused by the reduction of rainfall (Fig. 4). Significantly, in the whole mariculture cycle, the mariculture pond area serves as the sink of PAHs. A large number of PAHs in the surrounding atmospheric environment enter the surface ecosystem through sedimentation and interface exchange. Then, various aquatic products could absorb these pollutants from the water body and sediment through biological accumulation and feeding (Jafarabadi et al., 2017; Wang et al., 2010a; Zhang et al., 2020a). These pollutants have proved to affect the normal living activities of organisms (Han et al., 2022; Han et al., 2020; Liang et al., 2007), thus threatening the ecological security of the mariculture areas.

3.3.3. Bioaccumulation of PAHs by aquatic products

Bioaccumulation factors (BAFs, in L/kg) are used to express the enrichment ability of chemical pollutants in organisms, to further understand the transformation and bioavailability of pollutants, which is of great significance to indicate the potential harm of pollutants to organisms (Bandowe et al., 2014; Han et al., 2020). The BWAFs (bio-water accumulation factors) and BSAFs (bio-sediment accumulation factors) were calculated based on

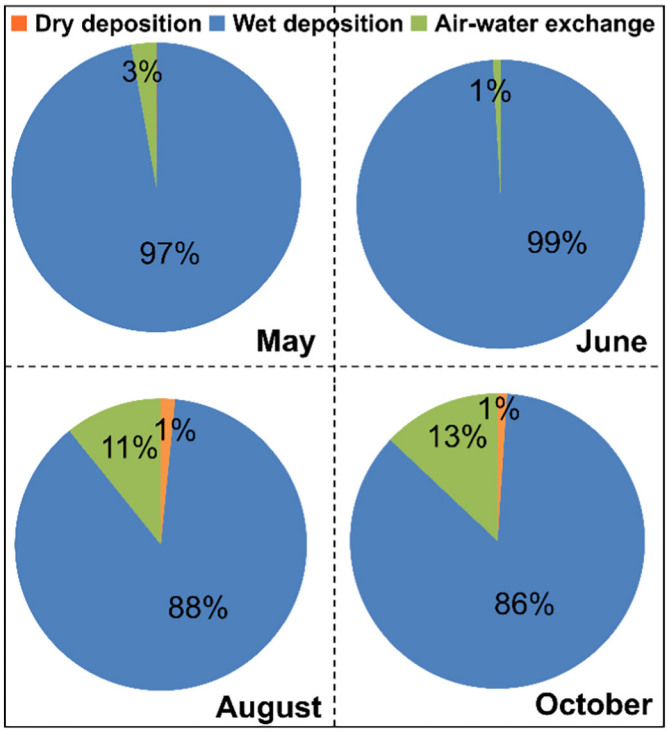

Fig. 5. Ratios of different migration modes of PAHs between atmosphere and surface water system.

the methods shown in Text S5. The calculated Log *BWAFs* (2.48–5.28) and Log *BSAFs* (2.90–5.19) showed that 7 (especially BaP, DiB, BaA, and BkF) and 11 PAHs (notably ACE, ANTH, ACEY, BaP, and FLU are >4) were considered bioaccumulative (Agency, 2012), indicating they can be absorbed by the aquatic products through the water and sediment environment, respectively (Fig. S7-A). Aquatic products showed potential bioaccumulation to 3 PAHs in mariculture water, namely PYR, FLU, and ACE. Overall, LMW-PAHs had relatively higher Log *BSAFs* values, and aquatic products in mariculture areas were more likely to absorb them from sediments, which is consistent with previous studies (Han et al., 2022). Affected by the concentration difference of PAHs in organisms, shrimp had higher bioaccumulation ability to PAHs in the surrounding environment than fish (Table S9). The BAFs have been considered and confirmed to have a significant correlation with the Kow of PAHs (Han et al., 2020; van der Heijden and Jonker, 2009). Pearson correlation coefficient results show that the Log Kow values of 15 PAHs are very consistent with their corresponding Log *BWAFs* ($R^2 = 0.59$, $p < 0.05$) and Log *BSAFs* ($R^2 = 0.45$, $p < 0.05$) values (Fig. S7-B). Compared with the natural sea areas in the same region, more PAHs were considered bioaccumulative, which should be paid more attention.

In general, for the closed mariculture pond ecosystem, the mutation of environmental factors has a significant and direct impact on the environmental behavior of PAHs in the system. Meteorological conditions such as rainfall and temperature synergistically drive the environmental behavior of PAHs in various environmental media of the mariculture pond ecosystem. Although this study only revealed the dynamic change trend of PAHs in mariculture ponds for about half a year, there were obvious differences in environmental fate, environmental behavior and ecological effects between the ponds and open sea areas in the same area, especially, the bioaccumulation capacity of marine organisms to PAHs in natural sea areas was low, and PAHs in sediment-water system only showed the desorption of sediment to water body (Han et al., 2021; Han et al., 2022). Climate change significantly affects the environmental fate of PAHs, but mariculture ponds play a positive role in preventing the migration and transmission of PAHs in the regional environment. The rainy season purified the local atmospheric environment, reduced the diffusion of local PAHs pollution to the surrounding areas, and also drove the migration of PAHs in the atmospheric environment to the surface ecosystem. Fortunately, the closeness of the land-based mariculture ponds made the material and energy exchange between it and the external environment have geographical obstacles, which have become a natural barrier to blocking the migration of PAHs. However, the transformation, bioaccumulation and adsorption/desorption of PAHs in the land-based mariculture ponds ecosystem would inevitably be affected, which may have an irreversible negative impact on aquatic products in the mariculture pond and eventually cause human consumption risk.

3.4. Source apportionment

Diagnostic ratio, principal component analysis and multiple linear regression (PCA/MLR) and probabilistic matrix factorization (PMF) are combined to evaluate the source of PAHs in this study. According to the previous studies (Table S14) (Chen et al., 2012; Han et al., 2021; Yunker et al., 2002; Zhang et al., 2021), we identified the sources of multiple environment media in the Maowei Sea (Fig. S10). Based on multiple molecular ratios (Text S7), the ratio of ANTH/(ANTH + PHE) were >0.1, the ratio of BaA/(BaA + CHR) were >0.35, the ratio of FLUA/(FLUA + PYR) were >0.5, and the ratio of Ind/(Ind + BghiP) were <0.2 in all most aqueous phase samples, indicating that the PAHs in the aqueous phase samples are mainly contributed by coal and biomass combustion and petroleum sources. Similarly, as shown in Fig. S11 and Table S14, PAHs in sediments and settled particles mainly come from combustion sources and petrogenic sources, while PAHs in the atmospheric environment mainly come from petroleum combustion-related sources and biomass combustion.

Quantitative source analysis PCA results of PAHs are shown in Fig. 6. The specific results of each factor are presented in Text S7. Combined with previous studies (Han et al., 2021; Larsen and Baker, 2003; Xu et al., 2012; Zhang et al., 2021), the PAHs produced by the biomass combustion

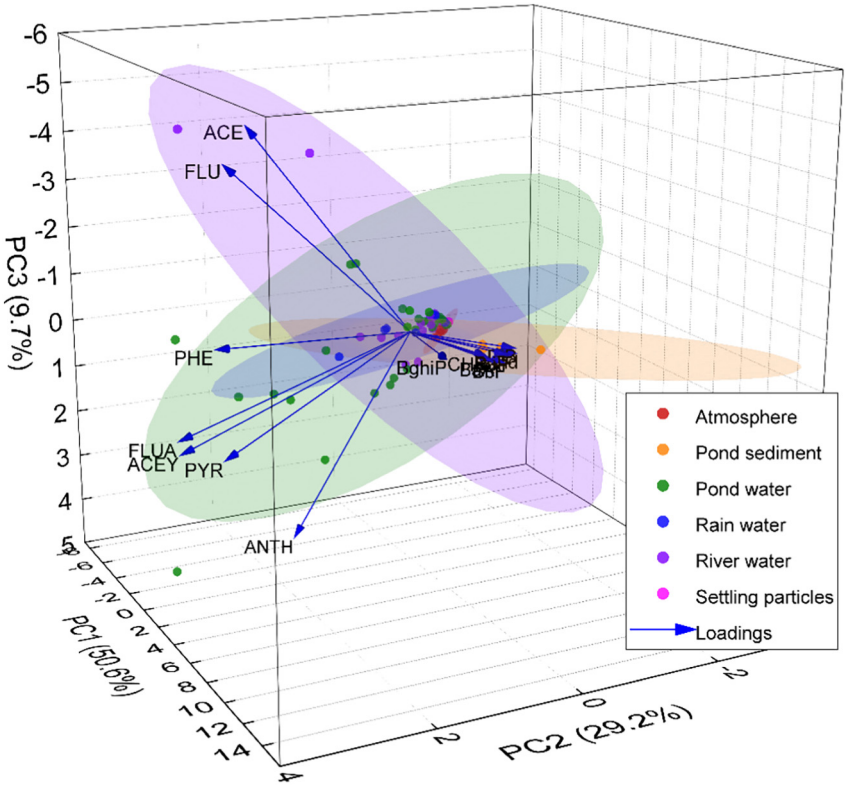

Fig. 6. Principal component analysis (PCA) of PAHs in environmental medias in Maowei Sea mariculture area.

sources have a considerable overall pollution level of PAHs in the region, and the contribution level to the overall PAHs has reached 53.4 %. The PAHs pollution caused by combustion of petroleum-related accounted for 18.8 %, and the potential source of oil pollution accounts for 27.8 %. Combined with previous research on the source of PAHs (Han et al., 2021; Hu et al., 2017; Zhang et al., 2021), the EPA PMF5.0 was also applied to evaluate the potential source of PAHs in this area. The PMF analysis used all 15 PAHs and 77 samples including all water, dustfall, atmosphere and sediment, and an empirical uncertainty of 20 % as the estimation of confidence level in this study (Lin et al., 2011; Zhang et al., 2021). By testing various factors (3, 4 and 5) and the number of seeds, and the solution with 4 factors and 70 seeds was selected to obtain the most stable and interpretable results (Figs. S11–S13). Based on previous studies (Han et al., 2021; Lin et al., 2011; Lin et al., 2013; Zhang et al., 2021; Zhang et al., 2012), four recognized source profiles were selected, including oil sources, biomass combustion, coal combustion and oil combustion-related sources (Text S7). PCA/MLR and PMF quantitative results show that the contribution of oil spill to PAHs in the region is consistent. PMF results reveal coal combustion sources, but the corresponding oil combustion-related sources are significantly reduced compared with PCA/MLR (Fig. S14). However, both coal/wood combustion and oil combustion-related sources belong to pyrolysis sources, and the sum of PMF source 2, 3, and source 4 is consistent with the PC1 and PC2 contribution of PCA/MLR (Fig. 7). Individual models may not fully identify specific sources, which may be the reason for this differentiation.

Three source apportionment models obtained consistent results: oil combustion-related sources, biomass combustion, and oil spill are the culprits of PAHs in the Maowei Sea mariculture areas. Maowei Sea is located at the intersection area of Qinzhou urban area and Qinzhou Bay. A large number of industrial activities in the surrounding environment may be the main driving factors, especially the combustion of local fossil fuels and the emission of a large amount of tail gas from cars and ships. PAHs pollution caused by biomass combustion was mainly due to the pollution of straw returning to the field in local agricultural activities and the adverse impact of forestry activities, especially the controlled burning in the process of Eucalyptus planting (Gerwing, 2010; Yanxian et al., n.d.). In addition, the potential sources of a small amount of oil pollution mainly come from the discharge of local oil drilling platforms, oil spills, and ship ballast water.

3.5. Risk assessment of edible aquatic products

Many previous studies have successfully used the excess cancer risk index to assess the risk of excessive cancer caused by human exposure to PAHs in the seafood diet (Bandowe et al., 2014; Han et al., 2022; Zhao et al., 2014). Using the method described in Text S8, estimated excess cancer risk results of aquatic products are shown in Table S16. The cancer risk caused by consumption of shrimps (8.24×10^{-6}–4.81×10^{-4}) in the Maowei Sea mariculture areas by people of various age groups is much higher than that caused by human consumption of fishes (1.51×10^{-8}–6.65×10^{-7}). Compared with the standards of the U.S. Environmental Protection Agency (EPA, 2018), frequent consumption of shrimp by natives is considered to have potential cancer risk. The excessive cancer risk in different age groups exceeds the minimum risk level but is generally lower than the priority control level. Noticeably, the potential cancer risk of consuming shrimp for boys aged 2–5 years has exceeded the priority control level. Estimated lifetime cancer risk results from accidental ingestion of PAHs due to consumption of mariculture aquatic products are worrying, the lifetime cancer risk of men (5.05×10^{-4}) and women (4.54×10^{-5}) exceeds the priority control level and the minimum safety level respectively (Table S17). These risk values are significantly higher than the natural sea area of the Beibu Gulf, Poyang Lake, and Bangladesh (Habibullah-Al-Mamun et al., 2019; Han et al., 2022; Zhao et al., 2014). However, there are few studies on the carcinogenic risk of PAHs in mariculture areas at present, so it is necessary to carry out long-term monitoring and strengthen comparative research. Fortunately, the daily intake of fish products is much higher than that of shrimp products. We should vigorously promote this eating habit as the health first choice for local people who frequently consume seafood, to ensure that they will not be exposed to the cancer risk of accidental intake of PAHs due to excessive consumption of mariculture aquatic products for a long time.

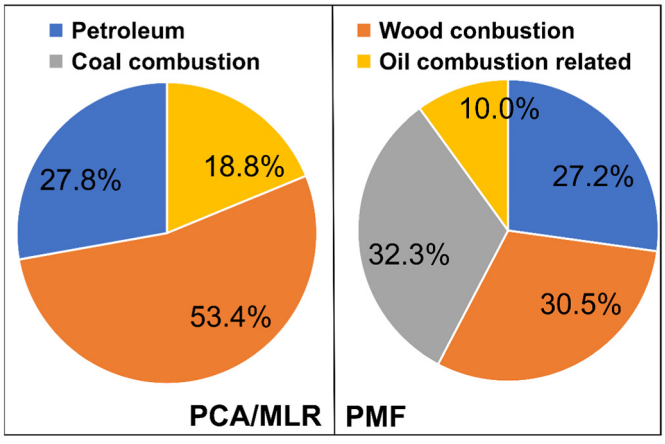

Fig. 7. Source contribution of two source apportionment methods to PAHs in mariculture pond of the Maowei Sea.

4. Conclusion

Through continuous dynamic monitoring of PAHs in a typical mariculture ecosystem of the northern the SCS, we found that meteorological factors, especially rainfall, mainly drive the dynamic attenuation of PAHs in multi environmental media during the mariculture cycle. During the monitoring period, PAHs mainly (>72 %) originated from oil-combustion-related sources and biomass combustion, while mariculture ponds were the sinks of these PAHs, which migrated to the surface system from the atmosphere through air-water exchange, dry and wet deposition. Among them, wet deposition was the predominant control process. Different from the natural waters in the same area, the sediment in the land-based mariculture ponds ecosystem could be the sink and the secondary release source of PAHs at the early and late stages, respectively. The closed land-based mariculture ponds affected the environmental fate of PAHs in the regional environment, blocked the transmission of PAHs in the regional environment, but increased the bioaccumulation of aquatic products to PAHs and their edible risk by human consumption.

CRediT authorship contribution statement

Minwei Han: Formal analysis, Visualization, Methodology, Data analysis, Writing-Original draft preparation

Ruijie Zhang: Conceptualization, Methodology, Data curation, Validation, Supervision, Writing-Reviewing and Editing

Kefu Yu: Conceptualization, Funding Acquisition, Project Administration Resource, Supervision

Annan Yan: Editing, Sample pretreatment, Writing-Reviewing and Editing

Haolan Li: Investigation, Sample pretreatment

Ruiling Zhang: Investigation, Sample pretreatment

Weibin Zeng: Investigation, Sample pretreatment

Zheng-en Zhang: Editing, Data analysis

Fang Liu: Sample pretreatment, Editing

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.

Acknowledgments

This work was supported by the Guangxi Natural Science Foundation (2020GXNSFDA297005), National Natural Science Foundation of China (42030502, 42090041, and 41463011), and the Guangxi Scientific Projects (AD17129063 and AA17204074).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.157442.

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