

Accumulation of PAH in bivalves (*Crassostrea gigas* and *Mytilus coruscus*) from Zhejiang coastal, China, and associated human health risk assessment

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Abstract

The paper reported the determination on PAHs concentrations in bivalves (*Crassostrea gigas* and *Mytilus coruscus*) from the Zhejiang coastal of China, and evaluate their composition, and assess their human health risk. The content of Σ PAHs in the two bivalves ranged from 48.30 to 61.08 ng/g. The main component of PAHs was Phenanthrene(11.08–15.79 ng/g). Comparing with HMW-PAHs, PAHs dominated by LMW-PAHs have higher absorption rates and lower purification rates. In the local coastal environment, the pyrolytic sources were an important role of the PAH pollution. At present, it assessed the health risks on consumption the two bivalves species, there was no threat to human health by eating shellfish accumulated PAH intakes, but there were some carcinogenic risks for heavier consumption of this two bivalves. The results will provide a reference value for the shellfish living marine environment in the region.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are typical persistent organic pollutants, which are hydrophobic compounds formed by connecting two or more benzene rings in the form of fused rings or non-fused rings (Chen et al., 2015). PAHs are widely found in the marine environment, it is derived from organic matter and fossil fuels incomplete combustion, oil leakage, industrial wastes, incineration of solid wastes and so on (Kim et al., 2013). PAHs are highly toxic, carcinogenic, mutagenic, teratogenic, and interfere with endocrine effects (Stegeman, 1985, Kim et al., 2013). The biomagnifications effect of PAHs is very limited by transmitting through the food chain, and organisms can enrich PAHs by absorbing PAHs in high concentration water (Barhoumi et al., 2016). With the development of the petroleum and chemical industries, all kinds of water body and aquatic organisms have been polluted by PAHs, including seawater, river water, lake water, and groundwater, which directly affect the quality and safety of human drinking water and food (Cao, 2015, Dong, 2015). Therefore, the concentration distribution of PAHs, pollution source of PAHs, and health risk assessment of PAHs have become the focus and hotspot on persistent toxic pollutants of scholars around the world on (Marrucci et al., 2013, Li et al., 2015, Chen et al., 2015).

Marine bivalve can reflect the local environmental conditions of water body due to their unique physiological and ecological characteristics. The marine mussel can be fixed on hard surfaces such as rocks by the attaching filaments, filtering water for food through the gill tissue, and ingesting other particulate matter. The marine mussels have a strong bioaccumulation effect on fat-soluble pollutants (Coughlam et al., 2002). PAHs are toxic to non-target organisms, among which marine bivalve being the most prominent these (Rantam 1997, Baussant et al., 2001). Shellfish are in the middle of the food chain, the material can transfer threatens the health of even higher organisms and even humans. The bivalve molluscs have a poor mobility, with regarded as monitoring marine chemical pollution indicator organisms, and strong pollutant enrichment capability (Le, 2004). The marine mussels (*Mytilus sp.*), Oysters (*Crassostrea virginica*), and zebra mussels (*Dreissena sp.*) were regarded as model species, monitoring the hazards of chemical pollutants of marine environment, and were used to assessment human health risk with a potential threat (Geyer et al., 1982). There is widespread concern about the bioaccumulation and toxic effects of PAH in these aquatic organisms, and their further spread to humans through the food chain (Moslen, 2019). Health risk assessment is an important basis for formulating food safety policies and regulations and solving international food trade disputes. It estimates potential risk assessments scientifically and reasonably by combining pollutant

concentrations, dietary levels, toxicological effects. As a result, some countries and the World Health Organization (WHO) have established the maximum acceptable concentration of PAHs in certain marine organisms, with beyond the acceptable concentration, the health of human consumers is compromised (Rey-Salgueiro et al., 2009).

Zhejiang province is located in the southeast coastal areas of China and is an important marine fishery base in China. Due to the rapid development of industry and agriculture in recent years, the land-based pollutants discharged into the sea have increased year by year, causing serious pollution such as PAH, heavy metals and so on of the coastal waters (Tientchen, 2008, Wang et al., 2015). *C. gigas* and *M. coruscus* were the most commercially valuable aquatic product species in Zhejiang Province, with high economic value and relevance to human consumption. They are a widely accepted sentinel for the study of seawater chemical pollution and biomarker related to PAH exposure (Wang et al., 2011, Barhoumi et al., 2016). Based on the above considerations, this study selected these two bivalve species and regions for research. Therefore, the purpose of this study was to determine the concentrations of PAH in *C. gigas* and *M. coruscus* obtained from the coast in Zhejiang province, China, and assess the potential health risk concerning the two bivalve consumption. The results obtained can indicate PAHs in the environmental migration of this study area, and provide a reference value for the shellfish living marine environment in the region.

2. Materials And Methods

2.1. Sample collection

The bivalve (*C. gigas* and *M. coruscus*) samples were collected from four regions (ZHOUZHAN, NINGBO, TAIZHOU and WENZHOU) along the coast of Zhejiang, China (Fig. 1). The seafood of the selected areas play a key role in the Zhejiang Province ecosystem and in providing seafood to humans. Samples were obtained every month from March to November. The samples were wrapped in aluminum foil and transported to the laboratory for analysis. The soft tissues from each spaced sample of three shellfish were immediately refrigerated (-20°C) until homogenized for further extractions.

2.2 Extraction and analysis

The method described by Tongo (2018) was used to extract PAHs with some modifications. The homogenized bivalve samples (10g) were thoroughly mixed with anhydrous Na_2SO_4 to dehydrate the sample. During extraction, 20 ml of dichloromethane was added to the sample, which was covered with aluminum foil to prevent evaporation, and then centrifuged to separate the supernatant of the extract. The extract was concentrated using an evaporator, and the extract was purified using a chromatographic column, and appropriately filled with 1 cm glass wool at the bottom. 2g of silica gel and 1 cm of anhydrous Na_2SO_4 were added to the column, and the column was pre-eluted with 20 ml of dichloromethane. Concentrated the extracts and then placed them in a 2 ml vial (Frapiccini et al., 2018, Moslen, 2019). Analytical gas chromatography (HP6890 Series II GC-FID, USA) was used to complete the determination of PAHs in the extracted samples. The instrumental analysis was based on the method by Tongo (2017).

PAH was quantified by external standard calibration (the PAH calibration mixture was from Sigma Corporation). Quality assurance (QA) and quality control (QC) strictly monitored all analytical procedures. The method blank samples, inner indicator samples, parallel samples, and standard sample were analyzed along with every 10 samples. The relative standard deviation (RSD) of parallel samples was less than 8%. Spike recoveries remained from 85–112%. The method blank samples were detected no PAHs concentration. The detection limit of the method was determined 3 times than the signal-to-noise ratio with the ranged from 0.01 to 0.10 ng/g.

2.3 Risk assessment

Human health risk assessment was conducted to estimate the possibility of exposure to PAHs through consumption of contaminated shellfish, and that could adversely affect human health. Different evaluation methods have been proposed the harms to human health of PAHs mixture in food. There are many models for assessing PAH health risks, such as exposure models, toxicity models, risk models, and thresholdless models (Ferrante et al., 2018). Halek(2007) assessed the Dietary Daily Intake of DDI of adult population by PAHs in the bivalves. In order to better assess the risk caused by the consumption of the two bivalves in this study, the average consumption rates for adults were converted based on Chinese averages (24.25×10^{-3} kg/day) (JECFA,2019). Ding(2012) summarized the carcinogenic potencies of individual PAHs (B(A)Pteq) to derive the carcinogenic toxic equivalents (TEQs). Incremental Life Cancer Risk (ILCR) was evaluated to determine non-carcinogenic effects and carcinogenic effects (Oliveira et al., 2018).

2.4. Data analysis

Excel 2010 and SPSS17.0 were used for basic statistics and charts. The PAH concentrations in bivalve tissues was analyzed using the One-way analysis of variance (ANOVA) with Duncan's method. An independent sample t-test was performed to determine the significant difference between the two bivalves analyzed.

3. Results And Discussion

3.1 PAH concentrations in two bivalve species (*C. gigas* and *M. coruscus*)

The quantitative results of PAH homologues in two bivalve species samples were showed in Table 1 from different markets of Zhejiang Province, China. The concentrations of individual PAH homologues in *M. coruscus* were greater profile, followed by *C. gigas*. ANOVA test showed that PAHs concentrations was a significantly difference in the two bivalve species ($p < 0.05$). Among individual concentrations of PAHs, Phenanthrene was the most dominant homologue of the two samples, and its concentration was significantly higher than the other homologues ($p < 0.05$). The average Phenanthrene concentrations of *C. gigas* and *M. coruscus* were 14.50 ± 1.29 and 12.33 ± 1.25 ng/g, accounting for 29.20% and 21.05% of the total PAHs, respectively. However, eight specific PAHs (Acenaphthene, Anthracene, Fluorene, Fluoranthene, Benzo(a)anthracene, Benzo(k)fluoranthene, Indeno(1,2,3)pyrene, Dibenzo(a,h)anthracene and Benzo(g,h,i)perylene) had the lowest mean concentration (ng/g) of ND in bivalve samples examined. The total carcinogenic PAH (sum of BaA, Chr, BkF, BaP, BbF, Ind, DaHA, BgP) concentrations were 9.36 ng/g and 14.60 ng/g in *C. gigas* and *M. coruscus*, respectively (Table 1). The average concentrations for total carcinogenic

PAHs in *C. gigas* accounted were higher (18.85%) than that in *M. coruscus*, but there is no statistically significant difference between the two concentrations ($p > 0.05$, $F = 0.26$).

Table 1
Mean concentrations (ng/g) \pm standard error (SE) of PAH congeners in bivalve samples examined during the study

PAH(ng/g)	Code	Ring number	<i>Crassostrea gigas</i>		<i>Mytilus coruscus</i>	
			Mean	SD(n = 64)	Mean	SD(n = 64)
Naphthalene	Nap	2	4.77	0.34	9.52	0.84
Acenaphthene	Ace	3	ND	ND	ND	ND
Acenaphthylene	Acy	3	10.46	0.51	8.77	0.51
Phenanthrene	Phe	3	14.50	1.29	12.33	1.25
Anthracene	Ant	3	ND	ND	ND	ND
Fluorene	Flu	4	ND	ND	ND	ND
Fluoranthene	Fl	4	ND	ND	ND	ND
Pyrene	Pyr	4	10.56	0.92	13.35	1.78
Benzo(a)anthracene	BaA	4	ND	ND	ND	ND
Chrysene	Chr	4	4.82	0.83	7.34	0.29
Benzo(k)fluoranthene	BkF	5	ND	ND	ND	ND
Benzo(b)fluoranthene	BbF	5	1.03	0.04	1.09	0.03
Benzo(a)pyrene	BaP	5	3.51	0.02	6.17	0.09
Indeno(1,2,3)pyrene	Ind	6	ND	ND	ND	ND
Dibenzo(a,h)anthracene	DaHA	6	ND	ND	ND	ND
Benzo(g,h,i)perylene	BgP	6	ND	ND	ND	ND
Total PAH	Σ PAH		49.65	1.35	58.57	2.51
Total carcinogenic PAH	Σ CPAH		9.36	0.88	14.60	0.39

European Food Safety Authority (EFSA) regards that BaP is not a suitable marker for PAHs in food. The system of two specific PAHs (Σ PAHs₂ = BaP + Chy) or four specific PAHs (Σ PAHs₄ = BaP + BaA + BbF + Chy) or eight specific PAHs (Σ PAHs₈ = Σ PAHs₄ + BkF + BghiP + DahA + Ind) will be the suitable indicators food for PAH, and maintain a separate maximum level for BaP (Ferrante et al., 2018). For two bivalve species, the descriptive statistic were detected in Table 2, which was expressed in ng/g, for PAH₂, PAH₄, PAH₈, PAH₁₆. The same phenomenon was observed in the sum of Σ PAH₂, Σ PAH₄, Σ PAH₈ and Σ PAH₁₆, which were significantly higher than two bivalve species ($p < 0.05$). There was no significant difference among concentrations of PAH₂ and PAH₄ and PAH₈ components ($p < 0.05$), which is due to the lowest concentrations

of BaA, BkF, BghiP, DahA, Ind(Table 2). In the present study we found PAHs8 did not provide much added value, which was consistent with the discovery of two bivalve species with little difference between PAH4 and PAH8 (Moslen, 2019). The concentrations of PAH4 and PAH8 in the two bivalves were lower than the regulatory maximum value of PAH4 (30 µg/kg) for European Commission Regulation (EU) No 835/2011. This indicated that lower risk of carcinogenic potentials for people with consuming the two bivalves on the present study region.

Table 2
PAH2, PAH4, PAH8 and PAH16 concentrations (ng/g) in bivalve

Bivalve		Σ PAH2	Σ PAH4	Σ PAH8	Σ PAH16
<i>Crassostrea gigas</i>	Mean	8.33	9.36	9.36	49.65
	Range	2.57 ~ 15.28	3.12 ~ 17.65	3.12 ~ 17.65	48.30 ~ 69.13
	S.D.	1.29	1.68	1.68	1.35
<i>Mytilus coruscus</i>	Mean	13.51	14.60	14.60	58.87
	Range	10.48 ~ 19.34	9.82 ~ 18.66	9.82 ~ 18.66	35.44 ~ 98.65
	S.D.	2.47	2.64	2.64	2.51

In order to know the compositions and sources of PAH pollution, 16 priority PAHs and 2-, 3-, 4-, 5-, and 6-ring compounds were analyzed in the present study. The lower molecular weight PAHs (LMW) (two to three rings) average concentration of in two bivalve samples were lower than higher molecular weight PAHs (HMW) (four to six rings), accounting for 51.60–59.88% of the total PAH, respectively (Fig. 2). There was no significant difference between the two bivalves samples ($p > 0.05$). It was similar to the PAH profile of mussels collected from the Corral Bay of South Central Chile and the Bizerte lagoon of Tunisia, which was reported by Palma-Fleming et al. (2012) and Barhoumi et al. (2014), respectively. The ratio of low molecular PAHs (two to three rings) to high molecular PAHs (four to six rings) was utilized to predict the pollution sources of different congeners, In general, LMW/HMW > 1 indicates the petrogenic origin, whereas LMW/HMW < 1 indicates the sources of pyrolytic (Yunker et al., 2002). The LMW/HMW ratios obtained for this study was greater than 1, indicating that the PAH of all the samples examined were derived from pyrolysis. The concentration of pollutants in the environment and the physiological and ecological characteristics of shellfish will affect the concentration of pollutants in shellfish (Yim, 2007). Crustaceans are particularly susceptible to contamination because of the reduced bioavailability of PAHs in these species (Law et al., 2002). These four areas were located in the main freight ports of Zhejiang Province, with a large number of cargo ships. The LMW PAHs accumulate in living organisms, which may be related to frequent marine transportation of ships and fuel leaks because of LMW PAHs are mainly derived from crude oil (Haruhiko et al., 2003). This can explain why levels of LMW PAHs in these two bivalves is higher .

For assessment the degree of PAH pollution of the study area, it is necessary to comparison it with other regional studies, in which PAH concentrations in bivalves had been also measured. The concentrations of PAHs in bivalves samples were obtained from different areas were summarized in Table 3. Compared with other marine bivalves, the current results were far lower than the mussels collected from Bizerte lagoon(107.4-

430.7ng/g), and the mussels collected from Tunisia Mediterranean Sea(146.9ng/g), which were two highly polluted areas. On the other hand, PAH concentrations are very different to those measured in bivalves samples collected from the Hainan Island (China), and were similar to that of mussel in the Gulf of Naples, Italy (77.69 ± 9.18 ng/g). The PAHs concentrations of the two bivalve species were higher than that from shellfish in Nigeria (3 to 16 ng/g). Above all, PAH concentrations of the two bivalves of Zhejiang Province are the medium pollution level compared with the reports from the other areas.

Table 3
Total PAHs in bivalves reported by other studies around the world

Bivalve	Total PAH	Reference
<i>Mytilus galloprovincialis</i>	77.69 ± 9.18 ng/g	Perugini et al. (2007)
<i>Mytilus galloprovincialis</i>	146.9ng/g	Mzoughi and Chouba(2012)
<i>Mytilus galloprovincialis</i>	107.4 ~ 430.7ng/g	Barhoumi et al.(2016)
<i>Perna viridis</i>	597.1 ~ 2332ng/g	Wang et al.(2020)
<i>Pinctada martensii</i>	818.5 ~ 215ng/g	Wang et al.(2020)
<i>Arca senilis</i>	3-16ng/g	Moslen(2019)
<i>Crassostrea gigas</i>	48.30 ~ 51.00 ng/g	This study
<i>Mytilus coruscus</i>	56.06 ~ 61.38 ng/g	This study

3.2 Human health risk assessment of PAH in bivalve

C. gigas and *M. coruscus* were regarded as the suitable indicator organisms which could monitor environmental pollution and analyzing the situation of human contact the pollutants through food, as the two bivalve species are consumed generally in Zhejiang Province. Health risk is mainly caused by the consumption of contaminated seafood. For PAH risk assessment of these two bivalve species, the risk of non-carcinogenic and carcinogenic exposure to PAHs in the diet were estimated (Table 4). The results were calculated based on the amount of PAHs by the daily (mg/kg bodyweight/day) amount of shellfish consumption of adult (bodyweight:70kg).

Table 4
 Estimated Dietary daily intake (DDI), Carcinogenic potencies (B(A)Pteq), and Excess cancer risk (ILCR) of PAHs in bivalve species from markets in Zhejiang coastal, China

PAH	TEF	RID	<i>Crassostrea gigas</i>			<i>Mytilus coruscus</i>			
			DDI	B(A)Pteq	ILCR	DDI	B(A)Pteq	ILCR	
			(ng/g/day)	(ng/kg)	(mg/kg)	(ng/g/day)	(ng/kg)	(mg/kg)	
*	Nap	0.001	0.02	115.67	4.77	7.08× 10 ⁻¹³	230.86	9.52	1.41× 10 ⁻¹²
*	Ace	0.001	0.06	ND	ND	ND	ND	ND	ND
*	Acy	0.001	0.06	253.66	10.46	1.55× 10 ⁻¹²	212.67	8.77	1.30× 10 ⁻¹²
*	Phe	0.001	0.03	351.63	14.50	2.15× 10 ⁻¹²	299.00	12.33	1.83× 10 ⁻¹²
*	Ant	0.01	0.30	ND	ND	ND	ND	ND	ND
*	Flu	0.001	0.04	ND	ND	ND	ND	ND	ND
*	Fl	0.001	0.04	ND	ND	ND	ND	ND	ND
*	Pyr	0.001	0.03	256.08	10.56	1.57× 10 ⁻¹²	323.74	13.35	1.98× 10 ⁻¹²
***	BaA	0.1	0.03	ND	ND	ND	ND	ND	ND
***	Chr	0.01	0.03	116.89	48.2	7.15× 10 ⁻¹²	178.00	73.4	1.09× 10 ⁻¹¹
**	BkF	0.1	0.03	ND	ND	ND	ND	ND	ND
***	BbF	1	0.03	24.98	1030	1.53× 10 ⁻¹¹	26.43	1090	1.62× 10 ⁻¹²
***	BaP	0.1	0.03	85.12	351	5.21× 10 ⁻¹¹	149.62	617	9.16× 10 ⁻¹¹
**	Ind	0.1	0.03	ND	ND	ND	ND	ND	ND
**	DaHA	5	0.03	ND	ND	ND	ND	ND	ND
**	BgP	0.01	0.03	ND	ND	ND	ND	ND	ND
Note: TEF values for the PAHs was adopted from (Nisbet and LaGoy, 1992). *Non-Carcinogenic PAHs. **Carcinogenic PAHs. ***Carcinogenic PAH and PAH used to derive the PAH4 Index									
Declarations									

The dietary daily intake (DDI) was determined human health risk by consumption of pollution bivalves. The DDI of individual PAHs of *C.* ranged from 24.98 to 351.63 ng/day, while the DDI was varied from 26.43 to 323.74 ng/day on *M. coruscus* (Table 4). The DDI of individual PAHs was usually lower than the available

reference oral dose, and the DDI value of carcinogenic PAH was higher than that of individual PAHs. The recommended daily intake of BaP is 10ng/kg/day (JECFA, 2019), which is above the DDI concentration of carcinogenic and non carcinogenic PAHs observed in this study. These levels were also lower than those reported on ingestion of bivalve (*Arca senilis*- blood cockles) in Nigeria population (Moslen et al., 2019). The results indicated that the consumption of *M. coruscus* will result in higher risk of exposure to PAHs and carcinogenic PAHs than consumption of *C. gigas*.

The carcinogenic of individual PAH in two bivalve species was different, the (B(A)P)teq of individual PAHs varied from 4.77 to 1090 mg/kg (Table 4). These values were higher than those observed for *Arca senilis* in Nigeria (Moslen et al., 2019). Benzo(b)fluoranthene had the highest carcinogenic potency in *C. gigas* (1030 mg/kg) and *M. coruscus* (1090 mg/kg), which could be a concern that the shellfish alone consumed can not cause the toxic effects of PAHs to the human health (Tongo, 2017).

For cancer risk, the ILCR values of the two bivalve species calculated by the China average ingestion rate were less than 10^{-10} (7.08×10^{-13} - 9.16×10^{-11}) (Table 4), which is below the ILCR concentration observed in Hainan Island, China (Wang et al., 2020). The excess lifetime cancer risk of $\geq 10^{-6}$ was considered an acceptable level, while the cancer risk of $\geq 10^{-4}$ was regarded as a serious level (Mana et al., 2013). Overall, the highest contributors to ILCR are BbF and BaP, which are far below the results of other studies (Wang et al., 2020; Moslen et al., 2019). As a result, consumption of *C. gigas* and *M. coruscus* appears to pose an acceptable risk of cancer.

The present study investigated the PAHs content in the two bivalve species of *C. gigas* and *M. coruscus* from the Zhejiang coastal in China. Our data suggested that high rates of PAHs bioaccumulation in shellfish may be due to marine pollution, which has been examined through the potential health risks by consumption. The concentrations of single PAH homologs and total PAH in the two bivalve samples were minimal compared to regulatory maximum level. However, it was observed that the PAHs dominated by LMW-PAHs are very similar for the two bivalve samples. Compared with HMW-PAHs, the LMW-PAHs have higher water solubility, higher absorption rate and lower purification rates. Furthermore, it should be emphasized that the pyrolytic sources played an important role in PAH pollution of the local coastal environment by the analysis on the proportion of specific PAH compounds. Health index such as DDI, TEQ and ILCR were assessed, respectively. Although the two bivalves have not a potential non-carcinogenic risk, carcinogenic risks may exist for large consumption. However, with protection the public health, human health risks need to be assessed and it is essential to identify as a potential risk of PAHs by consumption the two bivalves.

Declarations

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Conflict of Interest

No conflict of interest exists in the submission of this manuscript, and manuscript is approved by all authors for publication.

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Tables

Table 1 Mean concentrations (ng/g) ± standard error (SE) of PAH congeners in bivalve samples examined during the study

PAH(ng/g)	Code	Ring number	<i>Crassostrea gigas</i>		<i>Mytilus coruscus</i>	
			Mean	SD (n=64)	Mean	SD (n=64)
Naphthalene	Nap	2	4.77	0.34	9.52	0.84
Acenaphthene	Ace	3	ND	ND	ND	ND
Acenaphthylene	Acy	3	10.46	0.51	8.77	0.51
Phenanthrene	Phe	3	14.50	1.29	12.33	1.25
Anthracene	Ant	3	ND	ND	ND	ND
Fluorene	Flu	4	ND	ND	ND	ND
Fluoranthene	Fl	4	ND	ND	ND	ND
Pyrene	Pyr	4	10.56	0.92	13.35	1.78
Benzo(a)anthracene	BaA	4	ND	ND	ND	ND
Chrysene	Chr	4	4.82	0.83	7.34	0.29
Benzo(k)fluoranthene	BkF	5	ND	ND	ND	ND
Benzo(b)fluoranthene	BbF	5	1.03	0.04	1.09	0.03
Benzo(a)pyrene	BaP	5	3.51	0.02	6.17	0.09
Indeno(1,2,3)pyrene	Ind	6	ND	ND	ND	ND
Dibenzo(a,h)anthracene	DaHA	6	ND	ND	ND	ND
Benzo(g,h,i)perylene	BgP	6	ND	ND	ND	ND
Total PAH	ΣPAH		49.65	1.35	58.57	2.51
Total carcinogenic PAH	ΣCPAH		9.36	0.88	14.60	0.39

Table 2 PAH2, PAH4, PAH8 and PAH16 concentrations (ng/g) in bivalve

Bivalve		ΣPAH2	ΣPAH4	ΣPAH8	ΣPAH16
<i>Crassostrea gigas</i>	Mean	8.33	9.36	9.36	49.65
	Range	2.57~15.28	3.12~17.65	3.12~17.65	48.30~69.13
	S.D.	1.29	1.68	1.68	1.35
<i>Mytilus coruscus</i>	Mean	13.51	14.60	14.60	58.87
	Range	10.48~19.34	9.82~18.66	9.82~18.66	35.44~98.65
	S.D.	2.47	2.64	2.64	2.51

Table 3 Total PAHs in bivalves reported by other studies around the world

Bivalve	Total PAH	Reference
<i>Mytilus galloprovincialis</i>	77.69±9.18 ng/g	Perugini et al. (2007)
<i>Mytilus galloprovincialis</i>	146.9ng/g	Mzoughi and Chouba(2012)
<i>Mytilus galloprovincialis</i>	107.4~430.7ng/g	Barhoumi et al.(2016)
<i>Perna viridis</i>	597.1~2332ng/g	Wang et al.(2020)
<i>Pinctada martensii</i>	818.5~215ng/g	Wang et al.(2020)
<i>Arca senilis</i>	3-16ng/g	Moslen(2019)
<i>Crassostrea gigas</i>	48.30~51.00 ng/g	This study
<i>Mytilus coruscus</i>	56.06~61.38 ng/g	This study

Table 4 Estimated Dietary daily intake (DDI), Carcinogenic potencies (B(A)Pteq), and Excess cancer risk (ILCR) of PAHs in bivalve species from markets in Zhejiang coastal, China

PAH	TEF	RID	<i>Crassostrea gigas</i>			<i>Mytilus coruscus</i>			
			DDI	B(A)Pteq	ILCR	DDI	B(A)Pteq	ILCR	
			(ng/g/day)	(ng/kg)	(mg/kg)	(ng/g/day)	(ng/kg)	(mg/kg)	
*	Nap	0.001	0.02	115.67	4.77	7.08×10^{-13}	230.86	9.52	1.41×10^{-12}
*	Ace	0.001	0.06	ND	ND	ND	ND	ND	ND
*	Acy	0.001	0.06	253.66	10.46	1.55×10^{-12}	212.67	8.77	1.30×10^{-12}
*	Phe	0.001	0.03	351.63	14.50	2.15×10^{-12}	299.00	12.33	1.83×10^{-12}
*	Ant	0.01	0.30	ND	ND	ND	ND	ND	ND
*	Flu	0.001	0.04	ND	ND	ND	ND	ND	ND
*	Fl	0.001	0.04	ND	ND	ND	ND	ND	ND
*	Pyr	0.001	0.03	256.08	10.56	1.57×10^{-12}	323.74	13.35	1.98×10^{-12}
***	BaA	0.1	0.03	ND	ND	ND	ND	ND	ND
***	Chr	0.01	0.03	116.89	48.2	7.15×10^{-12}	178.00	73.4	1.09×10^{-11}
**	BkF	0.1	0.03	ND	ND	ND	ND	ND	ND
***	BbF	1	0.03	24.98	1030	1.53×10^{-11}	26.43	1090	1.62×10^{-12}
***	BaP	0.1	0.03	85.12	351	5.21×10^{-11}	149.62	617	9.16×10^{-11}
**	Ind	0.1	0.03	ND	ND	ND	ND	ND	ND
**	DaHA	5	0.03	ND	ND	ND	ND	ND	ND
**	BgP	0.01	0.03	ND	ND	ND	ND	ND	ND

Note: TEF values for the PAHs was adopted from (Nisbet and LaGoy, 1992). *Non-Carcinogenic PAHs. **Carcinogenic PAHs. ***Carcinogenic PAH and PAH used to derive the PAH4 Index

Figures



Figure 1

Sampling locations and sites of sediments in the study areas. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

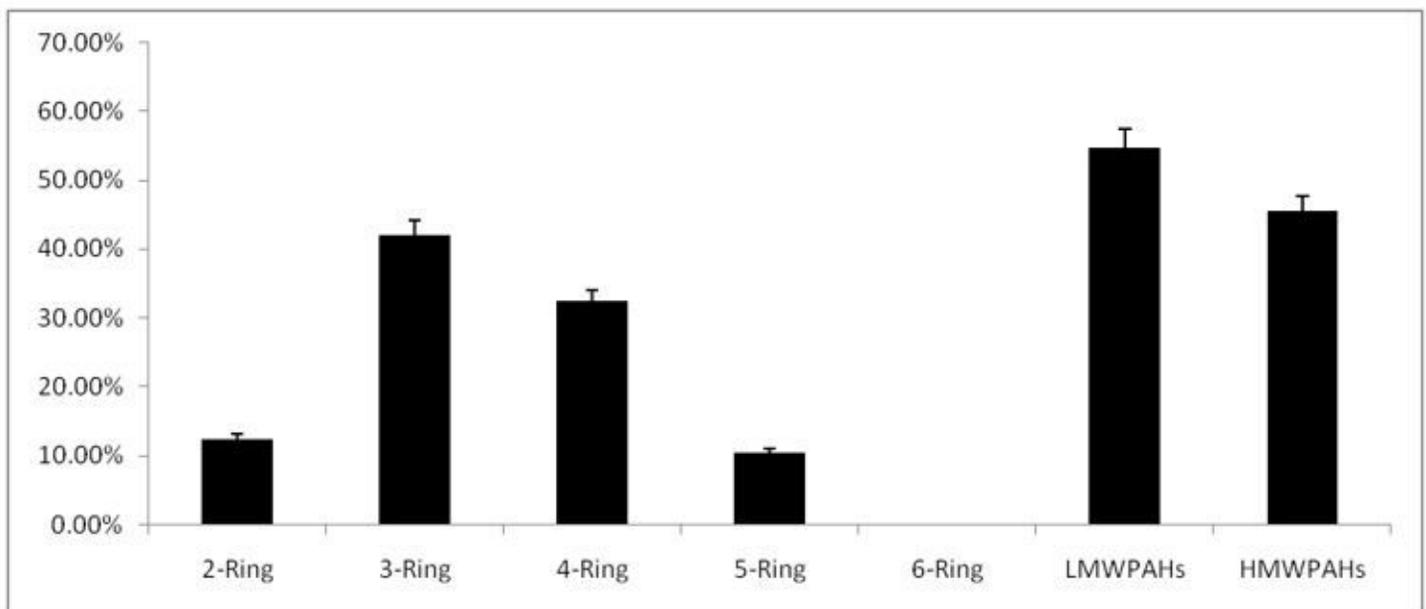


Figure 2

Mean percentage composition of PAHs (%) by ring-type in bivalve species samples from Zhejiang Province of China