Distribution of Polycyclic Aromatic Hydrocarbons in Farmed Oysters (*Crassostrea gigas*) around Tongyeong, Korea

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ABSTRACT

To evaluate the culture conditions in oyster-farming waters, chemical and biological measurements were made in seawater and oysters from six bays around Tongyeong in November and December 2003. Nutrient levels in the seawater were higher in the western area than in the eastern area, in contrast to particulate organic matter and dissolved oxygen levels. The mean total polycyclic aromatic hydrocarbon (∑PAH) content of the oysters was 194.5–375.9 ng/g dry weight, with four-ring compounds constituting 34.1%–79.6% of PAH. Despite wide temporal variations, a "western > eastern" spatial distribution of PAH was apparent. These low concentrations of PAHs indicate that Tongyeong waters are pristine in terms of PAH contamination. Among the hemocytic biomarkers, only lysosomal activity was significantly reduced in Hansan-Goje Bay, but did not correlate closely with PAH content. This finding indicates that the impact of PAH on cultured oysters is negligible around Tongyeong waters.

Keywords: Pacific oyster; Polycyclic aromatic hydrocarbon; body burden

Introduction

Marine bivalves, such as mussels and oysters, have been widely used as sentinel organisms to monitor the levels of polycyclic aromatic hydrocarbons (PAHs) because of their sessile lifestyle and limited ability to metabolize PAHs compared with that of fish (James 1989). Considering the variable toxicities of PAHs, monitoring these compounds is crucial to public health. In the 1990s, the Hazard Analysis and Critical Control Points (HACCP) system was implemented to increase food safety and quality (Hielm *et al*. 2006).

Tongyeong is a suburban area in the southeast of Gyeongsangnam-do, Korea. It includes the central and southern parts of the Goseong Peninsula, together with 151 islands (43 of which are inhabited). With the formation of good fishing grounds and nursing zones, Tongyeong's fishing industry has developed and improved over hundreds of years, making it one of the most productive fishing areas in Korea.

The present study was conducted in the six bays in which intensive oyster farming has been carried out since the 1960s. Jaran Bayis rectangular and enclosed by Saryang Island and 12 islets. Of the 7600 ha coastal area, 684 ha are occupied by 113 oyster farms with an annual production estimated to be -6982 metric tonnes (National Fisheries Research and Development Institute 2003). Because of continuous culturing and dense farming, Jaran Bay has experienced red tides and oxygen-deficient water masses.

Goseong Bay, located in Goseong County in the western part of Tongyoeng, has been under culture for the last three decades. It has a relatively stable coastal environment and is semi-enclosed, with a shallow water depth (predominantly < 10 m). Of the 1750 ha of coastal area, 169 ha are occupied by 35 oyster farms, with an estimated annual production of -2068 metric tonnes (Gyeongnam Province 1997). By the 1990s, oxygen-deficient water masses and red tides had emerged as major problems in Goseong Bay.

Farming of oyster by hanging culture has been

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undertaken over the last four decades in Bukman Bay, located in the middle of the city of Tongyeong.

The urbanization of the coastal area began in the 1980s, and domestic discharge has affected the water quality (Jeong 1998). Recently, oyster farms located in the inner part of the bay were moved to the outer bay. In the inner bay, none of the culture facilities are in use because of chronic red tides and eutrophication, and most of the culture facilities are currently located in the outer part of the bay. Of the 1090 ha of coastal area, 161 ha are occupied by oyster farms, which produce 1602 metric tonnes annually.

Hansan-Geoje Bay, enclosed close to Geoje Island and Hansan Island, is one of major areas of oyster culturing around Tongyoeng. In this bay, the main species cultured are oysters and an ascidian, with annual productions of 5794 and 3930 metric tonnes, respectively. Since 1974, 2121 ha of this area have been designated by the U.S. Food and Drug Administration (FDA) as a production area for shellfish for export. Unlike the other bays around Tongyeong, Hansan-Geoje Bay has been subjected to intensive bacteriological and sanitary surveys (National Fisheries Research and Development Institute 2003).

Semi-enclosed Wonmun Bay, located in the southwestern part of Jinhae Bay, has been under intensive oyster farming for the past several decades.

According to Lee (1992), the water column in this bay begins to stratify at the beginning of spring and then develops a strong oxygen-deficient water mass during summer. As a consequence of eutrophication resulting from increased domestic discharge and a weak tidal current, red tides and oxygen-deficient water masses have developed almost every year since the 1990s. Of the 830 ha of coastal area, 100 ha are used to farm the Pacific oyster, with a relatively small annual production because of low primary production rate (Kang *et al*. 1993).

Anjeong waters, located in the western part of Jinhae Bay, have been intensively farmed for oysters. However, beginning in the 1960s, increased pollution loads from urbanization and industrialization in the coastal zone and self-contamination from the cultured organisms have accelerated the eutrophication of these waters. Since the first report on this area, red tides have occurred with increasing frequency and for longer periods (Cho 1979).

Although many data are available regarding the PAH distribution in the coastal ecosystem around Korea (Choi 2000, Khim *et al*. 1999, Kim *et al*. 1999, 2001, Korean Ministry of Environment 1999; Korea Ocean Research and Development Institute 1999, 2003, Lee *et al*. 1998, Ministry of Marine Affair and Fisheries 1999, Moon *et al*. 2001, 2003, 2004, 2005, Nam 2001, Noh and Lee 2000, Yim 1998), data are lacking regarding their distribution relative to oyster culture grounds and/or cultured oysters around Tongyeong. Considering the importance of the coastal oyster fisheries in Korea and the quantities of oysters produced and consumed, possible PAH contamination of the culture waters and/or cultured oysters must be monitored in the interest of public health.

This study investigated the PAH content of cultured oysters collected from the six bays discussed above and measured the levels of several hemocytic biomarkers known to be sensitive to organic contaminants, with the aim of understanding the impact of PAHs on these cultured oysters. This comparative study of the PAH body burden of oysters and hemocytic biomarkers was used to evaluate the present status of PAH levels in oyster farms around Tongyeong.

Materials and Methods

Seawater was collected in Niskin water bottles (Ocean Test Equipment, Inc., Fort Lauderdale, Florida, USA) from 12 oyster farms located in six bays around Tongyeong (two stations per bay) in November and December 2003 (Fig. 1), from the surface (1 m below the surface) and bottom waters (1 m above the bottom). PO_4-P, dissolved inorganic nitrogen (DIN; sum of NO_2-N, NO_3-N, and NH_4-N), and particulate organic matter (POM) were analyzed following standard methods (APHA 1989), and water temperature and dissolved oxygen (DO) were measured with a SBE-19 profiler (Sea-Bird Electronics, Bellevue, Washington, USA). All the

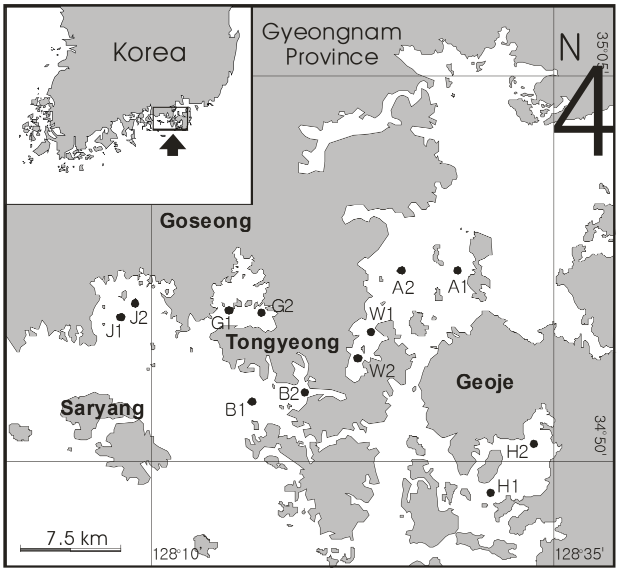

Fig. 1. Sampling sites for seawater and oysters around Tongyeong. J: Jaran Bay; G: Goseong Bay; B: Bukman Bay; W: Wonmun Bay; H: Hansan-Geoje Bay; and A: Anjeong Waters.

chemicals used for the analysis were of analytical reagent grade or above.

For measurement of biomarkers, oysters with a range of shell heights (80–100 mm) were collected at the same stations as the seawater. The hemolymph was collected from the pericardial cavity with a 3 ml syringe. The total hemocyte count was measured microscopically using a hemocytometer, and esterase activity was measured spectrofluorimetrically, according to Dolbear (1979, using fluorescein diacetate (Sigma F7378, USA). Lysosomal activity (LYS) was also measured spectrofluorimetrically, according to Lowe *et al*. (1992) using acridine orange. Phenoloxidase activity (PHE), peroxidase (PO) activity, and alkaline phosphatase activity (ALP) were localized immunocytochemically on a hemocyte smear, according to Xing *et al*. (2002).

For PAH analysis, 2.0 g of freeze-dried oyster was ultrasonically extracted three times in 30 ml of dimethyl chloride ($MeCl_2$). After centrifugation at 2500 rpm for 20 min, the pooled supernatants were concentrated to 2 ml using a K-D concentrator. The concentrated extract (2 ml) was purified by column chromatography on a silica–alumina column. Briefly, a glass column (300 19 mm i.d.) was packed with 20 g of deactivated silica gel (5% water) and 10 g of alumina with 2.5 g of anhydrous sodium sulfate on top. The extract (2 ml) was applied to the top of the column and eluted with 25 ml of n-pentane and then 250 ml of n-pentane: $MeCl_2$ (1:1). The first 10 ml was discarded and the remaining fraction was concentrated to about 1 ml with the K-D concentrator. Further cleanup was performed with a Sep-Pak Plus Silica cartridge. The elute was dried under nitrogen, dissolved in 0.5 ml of acetonitrile, and finally analyzed with a high-performance liquid chromatograph (Agilent 1100 series) equipped with a diode array detector (254 nm). A Supelco LC–PAH column (250 4.6 mm, 5 m particle size) was used. The mobile phase was an acetonitrile (AceCN)/water gradient, with the following program: 60% AceCN initially, 5 min hold, 25 min linear gradient to 100% AceCN, and 100% AceCN for 15 min. The flow rate was 1.5 ml min^{-1}, and the injection volume was 50 l. The peaks were identified by comparing their retention times with those of standards (Supelco 48743). To evaluate the accuracy of the analysis, a standard reference material from National Institute of Standard and Technology (NIST SRM 1974a; organics in mussel tissue) was analyzed. The calculated concentration of the PAH burden was within 29.6%–94.8% of the SRM certified concentrations (Table 1). The 16 target PAHs analyzed in oysters were naphthalene, acenaphthylene, acenaphthene, fluorine (Fl), phenanthrene (Phen), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benzo[*a*]anthracene (BaA), chrysene (Chr), benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene (BaP), indeno[1,2,3-*cd*]pyrene, dibenzo[*a,h*]anthracene, and benzo[*g,h,i*]perylene. Total PAH (∑PAH) was calculated as the sum of the 16 target PAHs. Individual compound concentrations below the detection limit of the method were assumed to be zero for the summation of ∑PAH in each sample.

All data are given as mean ± standard deviation. The data at each station were subjected to a Kolmogorov–Smirnov normality test to ensure that they were drawn from a normally distributed population. One-way analysis of variance (ANOVA) was then performed to test for the homogeneity of

Table 1. Results of analyses of the certified reference material (SRM 1974a mussel tissue)

PAH	Certified[a] (Mean ± STD)	Measured[a] (Mean ± STD)[b]	Recovery rate (%)
Naphthalene	24.0 ± 1.2	7.6 ± 1.7	31.7%
Acenaphthylene	4.7 ± 1.2	3.4 ± 1.8	72.3%
Acenaphthene	2.7 ± 0.53	0.8 ± 0.2	29.6%
Fluorene	4.88 ± 0.36	2.0 ± 0.42	41.0%
Phenanthrene	25.5 ± 1.1	11.6 ± 0.34	45.5%
Anthracene	5.20 ± 0.71	2.5 ± 0.58	48.1%
Fluoranthene	169 ± 7	76.9 ± 8.76	45.5%
Pyrene	178 ± 6	76.7 ± 8.88	43.1%
Benzo[*a*]anthracene	46.8 ± 5.2	22.2 ± 4.71	47.4%
Chrysene	62.2 ± 9.9	48.3 ± 6.94	77.7%
Benzo[*b*]fluoranthene	63.8 ± 5.8	32.2 ± 5.77	50.5%
Benzo[*k*]fluoranthene	31.2 ± 1.8	17.8 ± 1.27	57.1%
Benzo[*a*]pyrene	27.6 ± 3.8	15.9 ± 3.98	57.6%
Dibenzo[*a,h*]anthracene	3.23 ± 0.31	1.7 ± 0.31	52.6%
Benzo[*g,h,i*]perylene	30.8 ± 3.3	16.5 ± 4.06	53.6%
Indeno[1,2,3-*cd*]pyrene	21.1 ± 1.1	20.0 ± 1.46	94.8%

[a]Values are given in ng/g (dry weight). [b]STD = standard deviation (n = 2).

the means. If the data violated the presumption of homogeneity of variance, SNK pair-wise comparisons were used on significant ANOVA results. Statistical analyses were performed using Sigmastat 3.1 (Systat Software, Inc.).

Results and Discussion

During the investigation, the water temperature ranged from 6.6 to 14.6°C and the pH from 7.9 to 8.4, without significant local differences. Fig. 2 shows the mean values of the water analysis in each bay: 7.2–7.9 mg L^{-1} for DO, 0.05–0.13 mg L^{-1} for phosphate, and 0.05–0.15 mg L^{-1} for DIN. Nutrient levels were higher in the western waters (such as Goseong and Bukman Bays) than in the eastern waters (Wonmun and Anjeong Bays), and POM and DO were higher in the eastern waters than in the western waters. However, no significant differences were observed in these parameters among stations (P < 0.05).

Differences in the sizes and ages of the oysters are not expected to have affected the following results.

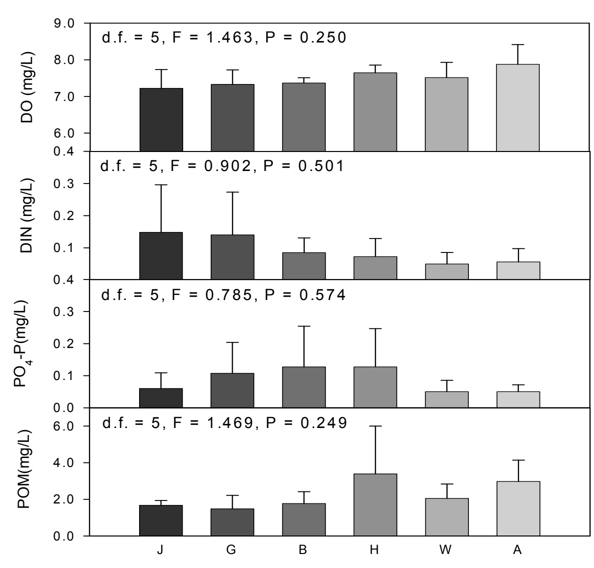

Fig. 2. Spatial variation in the mean water qualities during the present study. For abbreviations of the names of sampling sites, refer to Fig. 1. DO: dissolved oxygen; DIN: dissolved inorganic nitrogen; and POM: particulate organic materials.

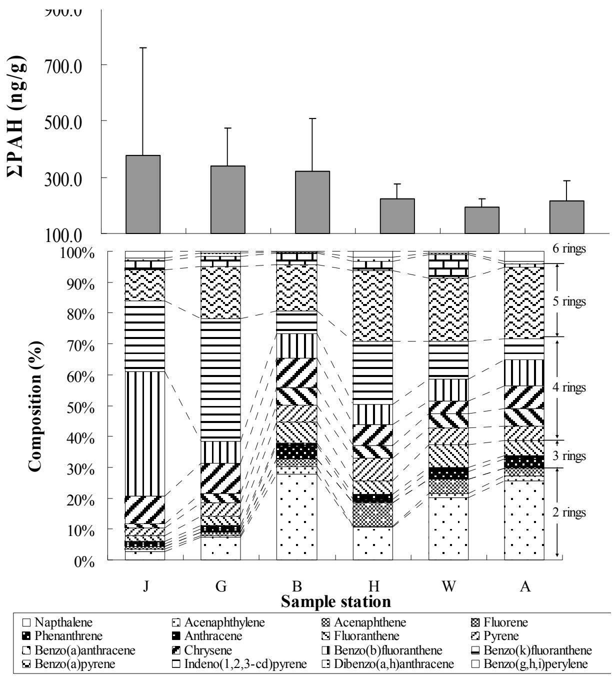

Fig. 3. Spatial distribution and compositions of PAHs in the cultured oysters around Tongyeong. No significant differences were observed among different sites ($P < 0.05$). For abbreviations of the names of sampling sites, refer to Fig. 1.

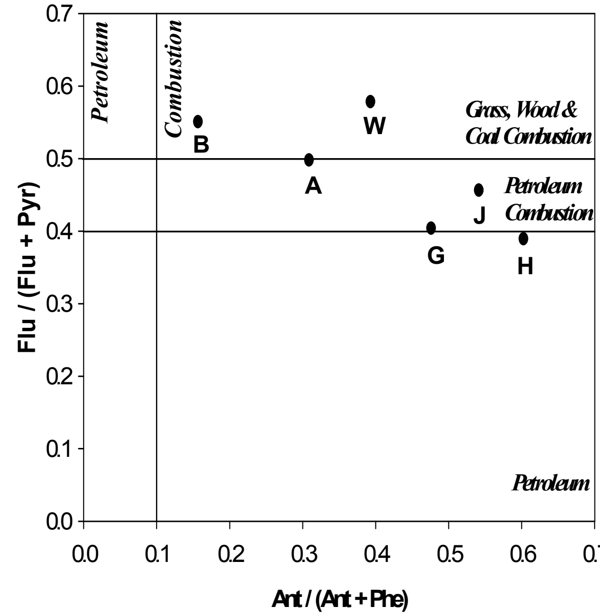

Fig. 4. PAH crossplots for Ant/ (Ant + Phen) vs. Flu/ (Flu + Pyr). The data represent the ratio of the averages of two indices in each bay. For abbreviations of the names of sampling sites, refer to Fig. 1. Ant: anthracene; Phen: phenanthrene; Flu: fluoranthene; and Pyr: pyrene.

Fig. 3 shows the PAH contents of the oysters according to the number of PAH rings. The mean ∑PAH ranged from 194.5 to 375.9 ng/g dry weight. The major components were the four-ring PAHs, which made up 34.1-79.6% of ∑PAH (generally four-ring > two-ring > five-ring > three-ring > six-ring PAHs). However, in the western waters, the concentrations of five-ring PAHs were higher than those of two-ring PAHs. Despite wide variations in the PAH concentrations, the apparent spatial distribution of ∑PAH was observed to be "western > eastern," as with the pattern observed for nutrients.

A source analysis was performed to identify the sources of the PAHs detected in this study. Several molecular ratios (e.g., Nap/FL, Phen/Ant, FL/Pyr, Chr/BaA, and Pyr/BaP) have been proposed for this purpose (Götz *et al*. 1998, Soclo *et al*. 2000, Kavouras *et al*. 2001, Yunker *et al*. 2002, Doong and Lin 2004). Possible PAH sources were identified using the Ant/(Ant + Phen) and Flu/(Flu + Pyr) ratios. PAHs for which Ant/(Ant + Phen) < 0.1 are derived mainly from petroleum contamination, whereas those for which Ant/(Ant + Phen) > 0.1 are typical of combustion sources. PAHs with Flu/(Flu + Pyr) > 0.5 are derived mainly from the combustion of grass, wood, and coal; PAHs in which 0.5 > Flu/(Flu + Pyr) > 0.4 are derived mainly from the combustion of petroleum, and those in which Flu/(Flu + Pyr) < 0.4 are indicative of petroleum contamination.

The obtained Ant/(Ant + Phen) values ranged from 0.31 to 0.60. The values in Goseong and Jaran Bay were within the range of petroleum combustion (0.4-0.5), and those in Hansan-Geoje Baywere in the range of petroleum contamination. All the values were assigned to the "combustion" region. The molecular ratio analysis of the input sources of PAHs indicated that petroleum combustion was the main source of PAH input in the western waters, whereas the PAHs in the eastern waters around Tongyeong were derived from natural sources (Fig. 4). This pattern might be attributable to the presence of industrial complexes in Gwangyang and Yeochun and a steam power plant at Sacheon, all located to the west of Tongyeong. It is possible that the prevailing westerly winds might

Table 2. Comparison of PAH content in the oyster, *Crassostrea gigas*, from Tongyeong and other waters

Location	∑AH (ng/g dry wt)	Contamination levelb	References
Tongyeong waters, Korea	194.5–375.9	Moderate	This study
San Francisco estuary (California, USA)	184–6,899	Moderate to very high	Oros and Ross (2005)
South coast, Korea	144–664	Moderate	Yim et al. (2002)
Kamak Bay, Korea	1,520–4,170	High	Ministry of Marine Affair and Fisheries (1999)
Masan intertidal zone, Korea	550.2–1750.0a	Low to moderatec	Noh and Lee (2000)
Intertidal zone on western coast, Korea	167.5–2,824.8	Low to high	Choi (2000)

[a]Unit: ng/g wet wt.
[b]Based on the scheme proposed by Baumard et al. (1998)
[c]Based on 80% water content.

transport the contaminants to adjacent waters. This tendency was also evident in the spatial distribution of ∑PAH (western > eastern).

To evaluate the extent of PAH contamination, the ∑PAH values calculated in this study were compared with those from other regional studies (Table 2). According to the criteria of Baumard *et al*. (1998) for PAH contamination, the levels of PAH in this study are considered to be "moderate" (100–1,000 ng/g dry weight, Oros and Ross 2005). Although the PAH concentrations in the seawater were not measured around Tongyeong, the PAH concentrations in the oysters from this area suggest that Tongyeong waters are, to some extent, pristine in terms of PAH contamination.

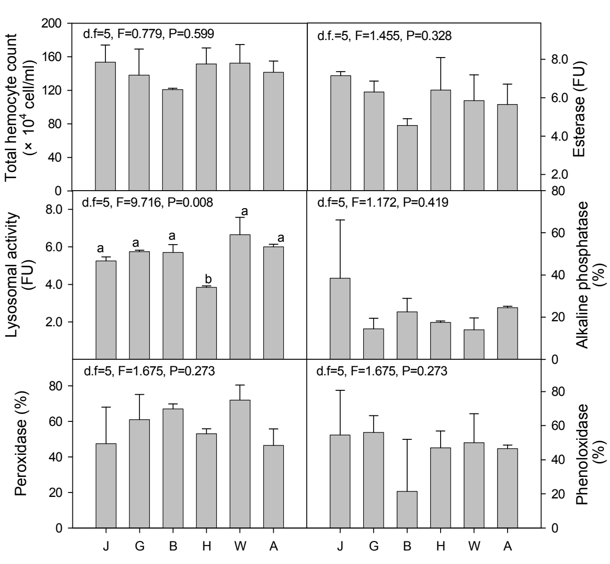

Fig. 5. Results of measurements of hemocytic biomarkers in Pacific oysters collected from sites around Tongyoeng. No significant differences were observed among sampling sites, except in terms of lysosomal activity. For abbreviations of the names of sampling sites, refer to Fig. 1.

To understand the effects of the PAH body burden on cultured oyster, various hemocytic biomarkers that are well documented as sensitive tools for biomonitoring pollutants of the Pacific oyster (Ministry of Marine Affair and Fisheries1999) were also measured. The hemocytic biomarkers and PAH body burdens were then compared. No spatial differences were observed in the average values for the hemocytic biomarkers (Fig. 5), except LYS ($P < 0.05$). LYS showed a significant reduction in Hansan-Geoje Bay ($P = 0.008$), which is the site of oyster production for export, as designated by the FDA.

A Pearson product moment correlation test was performed on the means of each parameter of seawater quality from the six sites, ∑PAH concentration, and hemocytic biomarkers to evaluate

Table 3. Pearson product moment correlation analysis of water qualities, hemocytic biomarkers, and ∑PAH concentrations

	DIN	P	POM	THC	E	LYS	ALP	PER	PHE	PAH
DO	- 0.785	- 0.220	**0.850***	0.111	- 0.244	- 0.029	- 0.292	- 0.289	- 0.030	**- 0.835***
DIN		0.161	- 0.643	- 0.007	0.539	- 0.189	0.465	- 0.267	0.344	**0.930****
P			0.050	- 0.518	- 0.296	- 0.604	- 0.322	0.208	- 0.480	0.240
POM				0.308	0.008	- 0.515	- 0.162	- 0.427	- 0.004	- 0.732
THC					**0.822***	- 0.205	0.143	- 0.327	0.793	- 0.291
E						- 0.365	0.363	- 0.529	**0.873***	0.244
LYS							- 0.148	0.480	- 0.022	- 0.096
ALP								- 0.643	0.031	0.551
PER									- 0.355	- 0.154
PHE										- 0.001

*P < 0.05, ** P < 0.01.
DO: dissolved oxygen; DIN: dissolved inorganic nitrogen; P: phosphate; POM: particulate organic matter;
THC: total hemocyte count; E: esterase; LYS: lysosomal activity; ALP: alkaline phosphatase activity;
PER: peroxidase activity; PHE: phenoloxidase activity.

the influence of the body burden of PAH on hemocytic homeostasis in cultured oyster (Table 3). ∑PAH correlated positively with DIN (P = 0.007) and negatively with DO (P = 0.039), whereas no correlation was found with the hemocytic biomarkers (P > 0.05). This finding indicates that PAHs have yet to have a significant influence on cultured oyster in the waters around Tongyeong. However, given the possibility of increased PAH input from anthropogenic sources, there is a growing need for intensive monitoring of these toxic and harmful compounds. Such monitoring is necessary for the sake of public health and to maintain the maximum sustainable yield of the oyster culture around Tongyoeng.

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