

Distribution, Source, and Ecological Risk Assessment of Polycyclic Aromatic Hydrocarbons in Surface Sediment of Liaodong Bay, Northeast China

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Abstract. Sixteen polycyclic aromatic hydrocarbons (PAHs) were determined in surface sediments from Liaodong Bay, northeast China. The concentration levels of total PAHs ($\Sigma 16\text{PAHs}$) in sediment were 11.0~249.6 ng•g⁻¹ dry weight (dw), with a mean value of 89.9 ng•g⁻¹ dry weight (dw). From the point of the spatial distribution, high PAHs levels were found in the western areas of Liaodong Bay. In the paper, sources of PAHs were investigated by diagnostic ratios, which indicated that pyrogenic sources were the main sources of PAHs in the sediment of Liaodong Bay. Therefore, selected PAH levels in sediments were compared with Sediments Quality Guidelines (ERM–ERL indexes) for evaluation probable toxic effects on marine organism.

Keywords: PAHs, Sediment; Source; Ecological risk assessment; Liaodong Bay.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs), defined as a group of aromatic hydrocarbons with two or more fused benzene rings, which are one of the most important classes of hydrophobicity organic contaminants. PAHs have been proved to be the main components responsible for effects on organisms and received special attention, due to its high toxicity and bioaccumulation, chronic dispersion, semi volatile and persistence [1]. The United States Environmental Protection Agency (USEPA) has included 16 polycyclic aromatic hydrocarbons (PAHs) as a priority controlled organic pollutant. For decades, the source and distribution of PAHs have been the focus of many researches.

Polycyclic aromatic hydrocarbons (PAHs) can be derived from nature, but mainly comes from human activities. PAHs are mainly found in marine areas such as harbor areas and estuaries exposed to anthropogenic input, i.e., industrial process, urban runoff, vehicle exhausts, and spillage of fossil fuel–pyrogenic and petrogenic sources [2]. Pyrogenic PAHs are formed by incomplete combustion of some organic compounds, such as wood, fossil fuels, asphalt and industrial wastes. Petroleum PAHs are usually found in crude oil and refined oil. Once produced, PAHs can be transferred to sea water in many ways, such as the discharge of industrial wastewater, surface runoff, atmospheric particulate deposition and oil leakage etc [3]. In the marine environment, PAHs are adsorbed on the surface of the sediments because of their high hydrophobicity and less degradation. The content of PAHs in sediments can reflect the local pollution level, so it is necessary to study the content of PAHs in sediments.

Liaodong Bay locates in the northeast of China. In the past 20 years, with the national emphasis on economic development in the northeast, the continuous development of Liaodong Bay coastal industry, a large amount of industrial wastewater discharged into the marine environment are integrated into the marine sediments by physical, chemical and biological processes. At the same time, a large number of



people have been gathered because of the economic development, which has brought about great influence on the surrounding environment. These series of socio-economic activities will lead to a wide range of PAHs pollution.

Certain studies of PAHs in aquatic environment have been documented in China, such as The Yellow River [4], Yangtze River [5], Pearl River [6], and East China Sea [7]. However, little studies of these compounds have been well documented in marine area from Northeast China. The contents of PAHs are investigated in surface sediment in Liaodong Bay, Northeast China, the objectives of which are 1) to investigate the concentration and spatial distribution of PAHs in sediment, 2) to determine the main source of PAHs in this region, 3) to determine the potential risk to aquatic organisms of PAHs-Ecological risk assessment of PAHs.

2. Materials and Methods

2.1. Sampling

Surface sediment (0~10cm) samples were collected at 58 sites from marine area in Liaodong Bay, Northeast China, in July, 2011. A map with locations of sampling sites is shown in Figure 1. The main cities around the sampling points include Huludao, Jinzhou, Panjin and Yingkou, while the Bay and the rivers are: Jinzhou Bay, Daling River, Xiaoling River, Shuangtaizi River and Liao River. 58 sampling points are represented by S01~S58. In consideration to the shoreline outfall, the location of the sampling sites along the coast line were around the main outfall within 500 m². After collection with the clamshell sediment sampler, all samples were packed in clean glass bottles, were refrigerated in ice and were sent to the laboratory of the International Joint Research Center for Persistent Toxic Pollutants (IJRC-PTS), Dalian Maritime University, Dalian, China and stored at -20°C until the time for extraction.

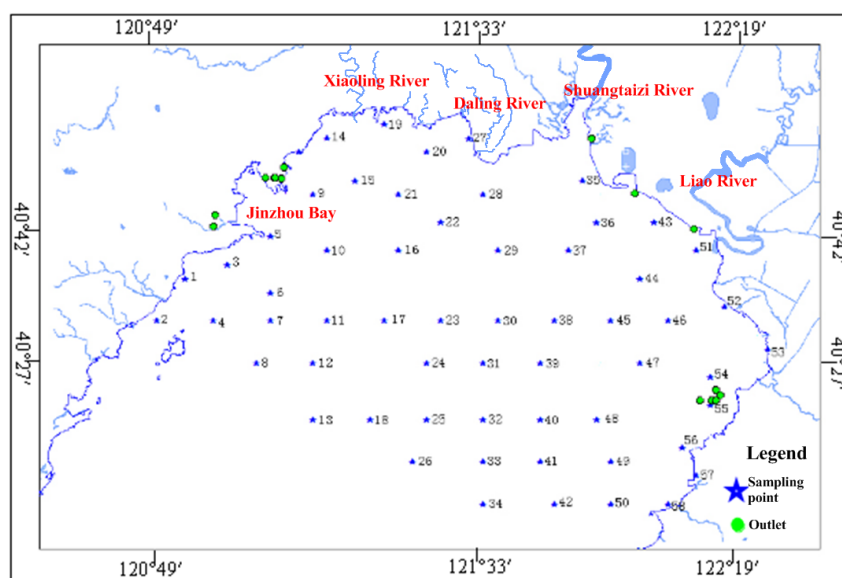


Figure 1. Distribution map of PAHs sampling position

2.2. Extraction and Analyses

The samples are placed in a freeze dryer drying for 36h and ground into fine powder, mixed the 5 g treated sample, 2 g anhydrous sodium sulfate (except water) and 1g copper (except sulfur) evenly, wrapped with extracted filter paper, placed in Soxhlet extractor, and adding standard (recovery indicator).

After mixing, samples were extracted for 24 h with 200 ml mixed solvent (hexane/ acetone, 1:1 v: v) by Soxhlet extractor. Extracts were then rotary-evaporated to 1 ml. The 1 ml extracts were passed through a 5.5 g silica gel column after a 25 ml hexane pre-rinse and eluted with 40 ml of hexane/dichloromethane mixture (1:1 v/v). The extract was rotary-evaporated to 2 ml, then blown to

0.5 mL with purified nitrogen flow, added 100 μ l internal standard substance (PAHs standard solution) and 2 ml isooctane solvent (improving the stability of samples) successively, blowed to 1 ml with purified nitrogen flow again, sealed in a brown bottle and placed at 20 degrees below zero for gas chromatography mass spectrometry (GC-MS) analysis.

The extracts were analyzed by gas chromatography mass spectrometry (GC-MS). All PAHs were identified and quantified using a Thermo Trace gas chromatograph (Thermo TRACE 2000) coupled with a Polaris Q mass spectrometry. Splitless injection was used (1 μ l), the inlet temperature was 250°C, along with a DB-5 column (HP 60 m \times 0.25 mm i.d. \times 0.25 μ m film thickness). The mass spectrometer was operated in electron impact mode (70eV). Helium gas was used as a carrier at constant flow 1.0 ml/min. The column oven temperature was programmed at a rate of 15°C/min from an initial temperature of 60°C to 160°C (1-min hold), then programmed at a rate of 1.5°C/min to 230°C (15-min hold), programmed at a rate of 20°C/min to 280°C (10-min hold) at last.

2.3. Quality Assurance/Quality Control

Strict quality assurance and quality control were put in place to monitor the analytical process, including method detection limits, blank labeling, method blank and added recovery indicator. The surrogate standard (recovery indicator) recoveries (shown in Table 1) ranged from 78% to 96% in all samples. Spike and blank samples were included at a rate of one for every five samples by clean sediment (solvent extracted five times) that was treated as real samples through entire procedures, the recoveries of all 16PAHs were ranged from 82% to 113%. The method detection limit (MDL) for the 16 PAHs ranged from 0.003 to 0.128 ng•g⁻¹ dw. All results were expressed on the basis of dry weight. Before use, the glassware was washed with n-hexane and dried at a temperature of 105°C in the oven, other materials were prewashed with ultra-pure water and acetone.

Table 1. The average recoveries of recovery indicators

Recovery indicators	Nap-D8	Flu-D10	Phe-D10	Pyr-D10	Per-D12
Average recoveries	85 \pm 7%	89 \pm 5%	89 \pm 7%	88 \pm 7%	88 \pm 5%

The recovery indicator is a substance that does not exist in the sample, including Naphthalene-D8, Flourene-D10, Phenatherene-D10, Pyrene-D10and Perylene-D12. They were used as the standard to determine the extracted rate of the target compounds. As is shown in the Table 2, for all the samples, the average values of the 4 recoveries are above 85%, indicating that the extraction of the target compounds can meet the requirements of the analysis.

At the same time, the standard solution of PAHs was added after nitrogen blowing, containing 16 PAHs homologues. By means of gas chromatography-mass spectrometry (GC-MS), 16 kinds of monomers were obtained according to the peak time sequence: Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene, Anthracene, Fluoranthene, pyrene, Benzo (a) anthracene, Chrysene, Benzo (b) fluoranthene, Benzo(k) fluoranthene, Benzo (a)pyrene, Indeno (1,2,3-cd) pyren, Dibenzo (a, h) anthracene, Benzo (g, h, i) perylene. Gas chromatogram of the PAHs standard solution Concentration of 100 ng•g⁻¹ is shown in Figure.2.

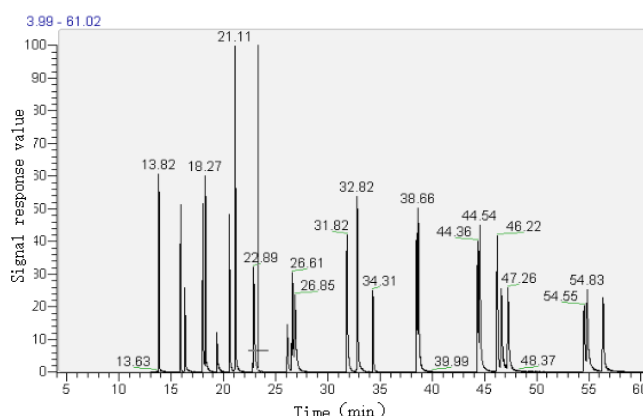


Figure 2. Gas chromatogram of the PAHs standard solution Concentration of $100 \text{ ng}\cdot\text{g}^{-1}$

2.4. Organic Carbon Content Determination

Organic compounds are carbon compounds, and organic matter is the material basis of life. Ten grams of sediment samples were isolated for percent moisture determination and total organic matter (TOM) measurement. Sediment samples were first oven-dried at 105°C for eight hours to a constant weight, then cooled and weighed. After moisture eliminated, the samples were placed in a muffle furnace and baked at 550°C for five hours. After cooling, the difference weight between the before and the after is the organic matter content (g). The levels of TOM are ranged from 2.96 to 9.92 percent. Organic carbon fraction (OCF) for each sediment sample can be calculated by assuming $\text{OCF} = \text{TOM}/1.8$.

3. Result and Discussion

3.1. Distribution Characteristics of PAHs in Sediments

The results of statistical analysis of PAHs and their contents are shown in Table 2. The concentrations of $\Sigma 16\text{PAHs}$ in sediment ranged between 11.0 to $249.6 \text{ ng}\cdot\text{g}^{-1}$ dry weight (dw), with a mean value of $89.9 \text{ ng}\cdot\text{g}^{-1}$ dw. It is obvious that the pollution level of Liaodong Bay is close to Leizhou Bay ($21.72\sim 319.61 \text{ ng}\cdot\text{g}^{-1}$) [12], higher than Laranjal ($7.3\sim 93.2 \text{ ng}\cdot\text{g}^{-1}$) [17], but significantly lower than some densely populated and industrialized estuarine coastal zones, such as Australian Port of Sydney ($<100\sim 380000 \text{ ng}\cdot\text{g}^{-1}$) [16] and the South Carolina of United States [15] ($570\sim 159041 \text{ ng}\cdot\text{g}^{-1}$). In China, the pollution level of Liaodong Bay is lower than Xiamen Port ($203.7\sim 1590.5 \text{ ng}\cdot\text{g}^{-1}$) [13], Daya Bay ($340\sim 710 \text{ ng}\cdot\text{g}^{-1}$) [14] and Southern Yellow Sea ($29.2\sim 1029.10 \text{ ng}\cdot\text{g}^{-1}$) [11]. In surface sediments from 58 sites, only the concentration of Acenaphthylene appears below detection limit for unknown concentration. The concentrations of PAHs in sediment exhibited a rather uniform distribution with a variation of about one order-of-magnitude. Overall, the pollution level of Liaodong Bay is not high, but the economy development process of surrounding city and human activities have y caused a negative impact to Liaodong Bay, which should cause enough attention to relevant management.

About the composition of PAHs of the various stations, the concentration range of the six rings PAHs was the lowest, which is $0.027\sim 18.259 \text{ ng}\cdot\text{g}^{-1}$. The concentration range of the four rings PAHs was the highest, which is $3.588\sim 92.393 \text{ ng}\cdot\text{g}^{-1}$. The concentration range of PAHs' species composition from high to low in order is: four rings > three rings > five rings > two rings > six rings. The percentage of the total PAHs content is: four rings 36.8%, three rings 27.7%, five rings 22.2%, two rings 8.5% and six rings 4.8%. This shows that the main pollutants of the investigated area are four rings PAHs, which are Fluoranthene, Pyrene, Benzo [a] anthracene and Chrysene.

About spatial distribution, the concentrations of PAHs from the western areas of Liaodong Bay were much higher than those from eastern area. The western areas were closed to Hebei Province and City of Tianjin, where were both the heavy industry base area, and were also the potential source of PAHs along the shore area of the study region.

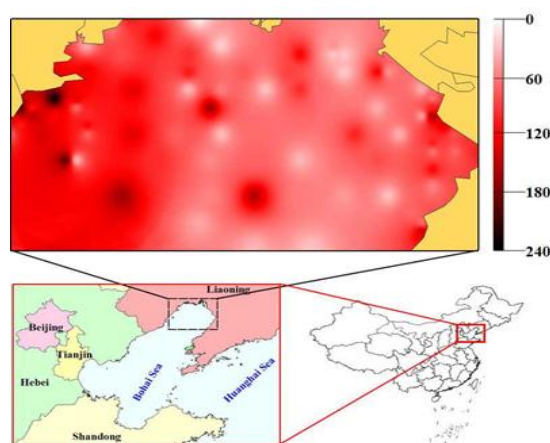


Figure 3. The location of sampling site and spatial distribution of PAHs in surface soil of Liaodong Bay

Table 2. Composition PAHs and their concentrations in Liaodong Bay surface sediment

PAHs	Ring no.	Concentration range (ng·g ⁻¹)	Average(ng·g ⁻¹)	SD
Nap	2	1.685~15.329	7.651	3.195
Acy	3	BDL ~4.553	1.210	0.788
Ace	3	0.011~0.861	0.116	0.190
Flo	3	0.052~10.285	3.888	2.234
Phe	3	2.482~37.082	14.269	5.651
Ant	3	0.665~10.649	5.447	2.419
Flu	4	0.988~30.874	11.411	7.368
Pyr	4	0.235~22.382	7.747	5.270
BaA	4	1.125~22.175	7.864	5.177
Chr	4	1.240~12.520	6.103	3.054
BbF	5	1.238~21.807	8.731	5.650
BkF	5	0.054~11.886	4.239	4.120
BaP	5	0.869~6.587	3.331	1.755
IncdP	5	0.345~10.766	3.625	2.656
DibahA	6	0.012~7.905	1.921	1.854
BghiP	6	0.015~10.354	2.365	2.393
2-Ring		1.685~15.329	7.651	3.195
3-Ring		3.149~ 63.811	24.930	11.08 7
4-Ring		3.588~92.393	33.124	20.80 8
5-Ring		2.506~59.797	19.926	14.10 5
6-Ring		0.027~18.259	4.287	4.240
Total PAHs concentration		10.955~249.589	89.918	53.15 2

note:BDL: below detection limit

Previous studies have shown that concentrations of PAHs in sediments are influenced by a number of factors including total organic carbon (TOC) content, mean sediment grain size, clay content, currents and so on [8,9]. In general, the content of PAHs in sediments is affected by the content of organic matter in sediments. There is a positive correlation between the contents of organic matter in sediments and the contents of PAHs. The organic matter and PAHs contents' logarithm were calculated, and the two values were compared and analyzed. The correlation analysis is shown in Figure.3.

In Figure.4, the change range of the horizontal axis LogTOM is 0.2~0.9, the change range of the vertical axis Log Σ PAHs is 0.6~0.3.7. The relationship between the contents of organic matter and PAHs is: $y = -0.259x + 1.9479$. The results show that there is no correlation between the two ($R^2 = 0.0041$). The correlation analysis between the two shows that the organic matters' contents in marine sediments are not the main factor to control the concentrations of PAHs.

Generally, the two main factors which influence the concentration of PAHs in sediments are the emission source and the physicochemical properties of the sediments [10]. The organic matter fraction has been implicated as the key physicochemical property influencing PAHs content in sediment. However, in this study, no significant correlation was found between PAHs concentrations and OMF

contents in sediment ($r=0.3$, $p>0.1$). It seems that the emission source along with OMF played more important roles in governing the distribution of PAHs in sediment around Liaodong Bay.

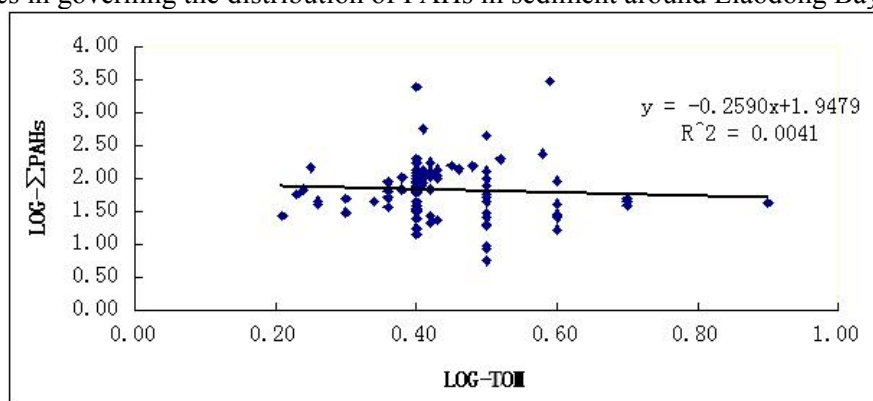


Figure 4. Correlation between LogTOM and LogΣPAH concentration

3.2. The Source Analysis of PAHs in Surface Sediment

Sources distinguish and controls are important to study the fate and transportation of PAHs in air and to reduce the environmental risk caused by PAHs. The possible sources of PAHs in sediment may be identified by diagnostic ratios of individual PAHs compounds. The ratio values of Fluoranthene/(Fluoranthene+Pyrene) and Anthracene/(Anthracene+Phenanthrene) are always used for identifying petrogenic and pyrogenic sources [18].

The ratio of Fluoranthene/(Fluoranthene+Pyrene) >0.5 suggests grass, wood and coal combustion (pyrogenic sources), where as <0.5 indicates petrogenic sources [20]. The ratio of Anthracene/(Anthracene+Phenanthrene) can be used to identify pyrogenic and petrogenic sources. Anthracene/(Anthracene+Phenanthrene) ratio <0.1 likely implies petroleum, and ratio >0.1 implies pyrogenic sources [19]. In the present study, the values of Anthracene/(Anthracene+Phenanthrene) ranged from 0.153 to 0.352 higher than 0.1, indicating a predominant contribution of pyrogenic sources in Liaodong Bay. In addition, the values of Fluoranthene/(Fluoranthene+Pyrene) were higher than 0.5, with average value of 0.578 ± 0.119 , also indicating the pyrogenic sources may be the most important sources of PAHs in Liaodong Bay. Cross plot of Anthracene/(Anthracene+Phenanthrene) ratio and Fluoranthene/(Fluoranthene+Pyrene) ratio is shown in Figure.5.

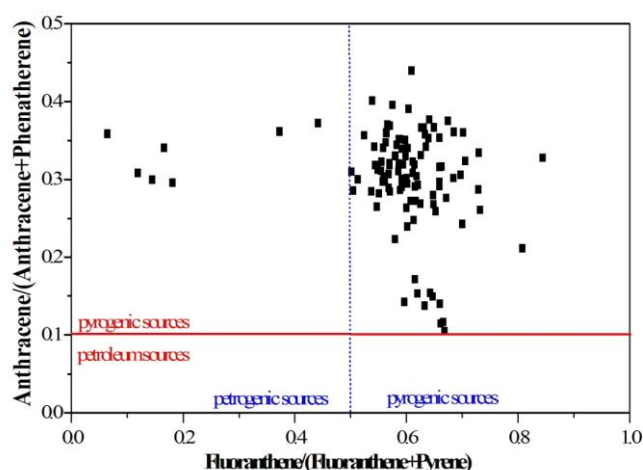


Figure 5. Cross plot of Anthracene/(Anthracene+Phenanthrene) and Fluoranthene/(Fluoranthene+Pyrene) in sediment from Liaodong Bay

To further subdivide the combustion type, Fluoranthene/(Fluoranthene+Pyrene) ratio and Indeno(1,2,3-cd)pyrene/(Indeno(1,2,3-cd)pyrene+Benzo(g,h,i)perylene) ratio were calculated in this

paper. Among them, $\text{Flu} / (\text{Flu} + \text{Pyr}) \leq 0.4$, was due to oil pollution; $\text{Flu} / (\text{Flu} + \text{Pyr}) \geq 0.5$, mainly derived from biomass and coal combustion; the ratio $0.4 \sim 0.5$ indicates combustion sources of oil and its refined products. $\text{InP} / (\text{InP} + \text{BgP}) \leq 0.2$, was mainly caused by oil pollution emissions, $\text{InP} / (\text{InP} + \text{BgP}) \geq 0.5$, was mainly wood, biomass and coal burning pollution, value $0.2 \sim 0.5$ was oil burning pollution [18]. Cross plot of Indeno (1,2,3-cd)pyrene/(Indeno (1,2,3-cd)pyrene+Benzo (g,h,i) perylene) ratio and Fluoranthene / (Fluoranthene+Pyrene) ratio is shown in Figure.6. It can be seen from Figure.6, the $\text{Flu} / (\text{Flu} + \text{Pyr})$ and $\text{InP} / (\text{InP} + \text{BgP})$ ratio are greater than 0.5, indicating that the wood, biomass and coal combustion is the main source of PAHs in Liaodong bay.

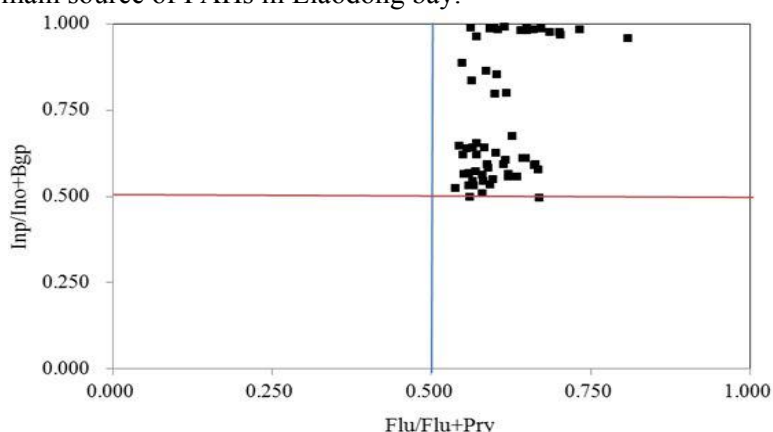


Figure 6. Cross plot of Indeno(1,2,3-cd)pyrene/(Indeno(1,2,3-cd)pyrene+Benzo(g,h,i)perylene) and Fluoranthene/(Fluoranthene+Pyrene) in sediment from Liaodong Bay

Generally speaking, the characteristics of PAHs in Liaodong Bay sediments show two kinds of mixed sources, which are incomplete combustion of fossil fuel and biomass. Exhaust emission of port ships and motor vehicles in surrounding cities should be the main sources of Liaodong Bay PAHs, the input of wood and coal combustion in the northern area also has a great contribution to it.

Table 3. The composition PAHs and their concentrations in Liaodong Bay surface sediment

PAHs	ERL	ERM	Range (ng g ⁻¹)	Average(ng g ⁻¹)	SD
Naphthalene	160	2100	1.685~15.329	7.651	3.195
Acenaphthylene	44	640	aBDL ~4.553	1.210	0.788
Acenaphthene	16	500	0.011~0.861	0.116	0.190
Fluorene	19	540	0.052~10.285	3.888	2.234
Phenanthrene	240	1500	2.482~37.082	14.269	5.651
Anthracene	85.3	1100	0.665~10.649	5.447	2.419
Fluoranthene	600	5100	0.988~30.874	11.411	7.368
Pyrene	665	2500	0.235~22.382	7.747	5.270
Benzo(a)anthracene	261	1600	1.125~22.175	7.864	5.177
Chrysene	384	2800	1.240~12.520	6.103	3.054
Benzo(b)fluoranthene	--	--	1.238~21.807	8.731	5.650
Benzo(k)fluoranthene	--	--	0.054~11.886	4.239	4.120
Benzo(a)pyrene	430	1600	0.869~6.587	3.331	1.755
Indeno(1,2,3-c,d)pyrene	--	--	0.345~10.766	3.625	2.656
Dibenzo(a,h)anthracene	63.4	260	0.012~7.905	1.921	1.854
Benzo(g,h,i)perylene	--	--	0.015~10.354	2.365	2.393
Σ16PAHs	4022	44792	10.955~249.589	89.918	53.152

Note:BDL: below detection limit

4. Ecological Risk Assessment

Through numerous modeling, laboratory and field studies, Long et al matched biological and chemical data, and then two guideline values (an Effects Range-Low and an Effects Range-Median) were proposed for the potential ecological risk of PAHs in marine and estuarine sediments. This method is being used as a basis for the development of Canadian sediment quality guidelines [21]. SQGs provide

two target values: effects range low (ERL) and effects range median (ERM), which are established using the 10th and 50th percentiles, respectively, in a database of increasing concentrations associated with adverse biological effects. In the light of sediment quality guidelines (SOGs), adverse biological effects are expected to occur rarely ($< \text{ERL}$), occasionally ($\geq \text{ERL}$ and $< \text{ERM}$), and frequently ($\geq \text{ERM}$) [21]. Concentrations of several target PAHs in sediment were compared to existing corresponding sediment quality guidelines (the ERL/ERM values) and results were presented in Table 3. In this paper, the concentrations of all PAHs were less than suggested SOGs, indicating there is no potential toxic effect on benthic organisms.

5. Conclusion

(1) The concentration levels of total PAHs ($\Sigma 16\text{PAHs}$) in surface sediment from Liaodong Bay were $11.0\sim 249.6 \text{ ng}\cdot\text{g}^{-1}$ dry weight (dw), with a mean value of $89.9 \text{ ng}\cdot\text{g}^{-1}$ (dw). The composition is mainly composed of 3 and 5 rings PAHs, 2 and 6 rings PAHs are relatively less.

(2) The characteristics of the isomer ratio reflect that PAHs in sediments from Liaodong Bay are mainly from fossil fuel and biomass burning, may be due to ship and automobile exhaust emissions, and coal burning internal heating system. In some parts of the region, the leakage of petroleum fuel and the discharge of oily sewage may also cause PAHs pollution. It should be emphasized that due to the busy traffic in the region, the source pattern of PAHs may change constantly.

(3) Biological risk assessment based on sediment quality standards indicates: PAHs in the surface sediments of Liaodong Bay aren't more than ERL, and no harmful effects on marine organisms.

6. Acknowledgement

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7. References

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