



## Biodegradation of polycyclic aromatic hydrocarbons in sediments from the Daliao River watershed, China

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### Abstract

The Daliao River, as an important water system in Northeast China, was reported to be heavily polluted by polycyclic aromatic hydrocarbons (PAHs). Aerobic biodegradations of four selected PAHs (naphthalene, phenanthrene, fluorene and anthracene) alone or in their mixture in river sediments from the Daliao River water systems were studied in microcosm systems. Effects of additional carbon source, inorganic nitrogen and phosphorus, temperature variation on PAHs degradation were also investigated. Results showed that the degradation of phenanthrene in water alone system was faster than that in water-sediment combined system. Degradation of phenanthrene in sediment was enhanced by adding yeast extract and ammonium, but retarded by adding sodium acetate and not significantly influenced by adding phosphate. Although PAHs could also be biodegraded in sediment under low temperature (5°C), much lower degradation rate was observed. Sediments from the three main streams of the Daliao River water system (the Hun River, the Taizi River and the Daliao River) demonstrated different degradation capacities and patterns to four PAHs. Average removal rates (15 or 19 d) of naphthalene, phenanthrene, fluorene and anthracene by sediment were in the range of 0.062–0.087, 0.005–0.066, 0.008–0.016 and 0–0.059 mg/(L·d), respectively. As a result, naphthalene was most easily degraded compound, anthracene was the hardest one. In multiple PAHs systems, the interactions between PAHs influenced each PAH biodegradation.

**Key words:** biodegradation; the Daliao River; microcosm; polycyclic aromatic hydrocarbon; sediment

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### Introduction

The Liao River watershed is located in the region of 40°31'N–45°17'N and 116°54'E–125°32'E, passes through three provinces in Northeast China, which consists of the Liao River water system (Liao River) and the Daliao water system (including the Hun River, the Taizi River and the Daliao River). The mid- and down-stream areas of the Liao River watershed are important bases of oil, chemical and steel-making industries of China. With the rapid development of these industries, the Daliao River was reported to be heavily polluted by organic pollutant, especially polycyclic aromatic hydrocarbons (PAHs). Our previous research showed that total PAHs level in the water, suspended particle matters (SPMs) and sediment of the Daliao River were in the range of 946–13449 ng/L, 318–238519 ng/g and 62–841 ng/g, respectively. Compared with other rivers in the world, the Daliao River has much high PAHs content in water and relatively lower SPMs in sediment (Guo *et al.*, 2007; Ko *et al.*, 2007; Countway *et al.*, 2003; Mitra and Bianchi, 2003; Headley *et al.*, 2001). PAHs could enter environment from fossil fuel combustion, accidental spilling of processed hydrocarbons, domestic and

industrial wastewaters discharge, atmospheric deposition, etc. (Freeman and Cattell, 1990; Witt and Trost, 1999). As PAHs are toxic and suspected carcinogen and mutagen, their presence in contaminated rivers and sediments poses a significant risk to environment and human health. The US Environmental Protection Agency has identified 16 PAH compounds as priority pollutants (Heitkamp and Cerniglia, 1988; Manoli *et al.*, 2000).

Microbial degradation is believed to be one of the major processes to remove PAHs from contaminated environment. PAHs not only could be biodegraded by some pure culture in liquid, but also could be degraded by indigenous culture bound to contaminated sediments or soils. There is an abundance of information on single PAH biodegradation by isolated pure strains (Ravelet *et al.*, 2000; Ramirez *et al.*, 2001; Lee *et al.*, 2003). However, the biodegradation of PAHs by indigenous microbial in sediment samples taken from natural water system was much more complicated than that by a pure strain. It was reported that the biodegradation of PAHs in natural environment is dependent on many factors such as organic content, nutrient, oxygen level, electron acceptor, water temperature and so on (Dan *et al.*, 1999; Hinga, 2003; Lee *et al.*, 2003). For example, the degradation rate of low molecular weight PAHs was reported to correlate with sediment total organic carbon

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(TOC) in marine sediments (Hinga, 2003). In addition, PAHs contaminated environment is generally complicated with various PAHs mixture, effect of substrate interactions on biodegradation is important in understanding the fate of PAHs in intrinsic and engineered system. Therefore, knowing the degradation patterns of PAHs under various conditions is important for scientific program design in remediation of PAHs polluted sites.

The aims of this study were: (1) to measure the effects of several factors including the amendment of carbon source, nitrogen and phosphorus, temperature variation on PAH biotransformation; (2) to compare PAH biodegradation potential in sediments sampled from different locations of the Daliao River water system; (3) to investigate the capability of indigenous microbial in sediments to the biodegradation of a single PAH or their mixture.

## 1 Materials and methods

### 1.1 Chemicals

Naphthalene, phenanthrene, fluorene and anthracene with 99.0% analytical standard were purchased from AccuStandard Inc., USA. All other chemicals were purchased from Beijing Chemical Factory, China and Tedia Co., USA. Deionized water was produced by a Milli-Q system (Millipore Co., USA).

### 1.2 Sediments collection

Surface sediments (1–15 cm) were collected using grab sampler along the Hun River, the Taizi River and the Daliao River (Fig. 1). Samples collected at each site were mixed thoroughly, transferred into sterilized bags, and stored in refrigerator at 4°C. Sediment samples collected at the location of H5, T5 and D1 were used for this study. H5 and T5 represent midstream of the Hun River and the Taizi River, respectively, and are adjacent to lots of discharging sources of wastewater. D1 is the joint point of the Hun River and the Taizi River, and represents upstream of the Daliao River. The physicochemical properties of the site water and sediment from H5, T5 and D1 are presented in Table 1. PAHs were detected in all three sediments with concentration range of 111–817 ng/g (16 PAHs).

### 1.3 Experimental steps

First, the effects of sediment presence, amendment of additional carbon sources (10 mg/L yeast extract and 10 mg/L sodium acetate), ammonium chloride (100 mg/L), sodium dihydrogen phosphate (100 mg/L) and different temperatures (15 and 5°C) on PAH degradation were investigated in microcosm systems. Phenanthrene was selected as the model compound, and water and sediment sample from T5 were used. Microcosm systems were made by adding 1 g sediment, 19 mL site water, 1 mL solution of the nutrient planned (yeast extract, sodium

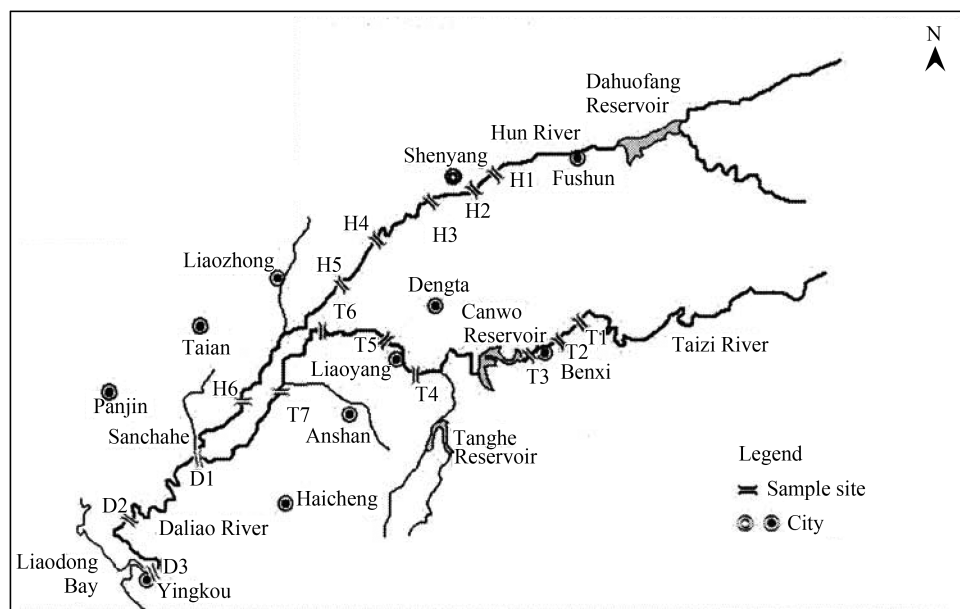


Fig. 1 Sampling points in the Daliao River watershed, China.

Table 1 Physicochemical properties in water and surface sediment of the Daliao River water system

Station	Description	Water				Sediment			
		pH	Eh (mV)	TOC (mg/L)	TC (mg/L)	TOC (%)	Water (%)	Silt-clay (%)	CEC (cmol/kg)
H5	41°29.650'N, 122°57.399'E	7.30	530.1	66.2	113.61	0.15	24.23	48.67	0.95
T5	41°20.710'N, 123°08.584'E	7.53	524	48.17	108.3	0.70	36.42	29.53	2.19
D1	41°00.205'N, 122°24.856'E	7.73	49.29	36.29	82.26	2.56	45.43	34.87	16.30

TOC: total organic carbon; TC: total carbon; CEC: cation exchange capacity.

acetate, ammonium or phosphate), and a certain volume of phenanthrene stock solution (less than 1 mL and dissolved in methanol) into 50 mL sterilized serum bottles. Phenanthrene initial concentration was controlled at about 0.5 mg/L. A killed control microcosm was prepared through adding autoclaved sediment (which was autoclaved twice at 121°C for 30 min) and  $\text{NaN}_3$  (2 g/L, final concentration). Serum bottles were capped with butyl rubber stoppers and wrapped with aluminum foil to prevent photolysis, and oxygen was provided for biodegradation through opening the caps twice a day (about 3 min every time). All treated samples were incubated without shaking at 15°C in the dark except one test controlled at 5°C. Sediment and water mixture was sampled from each reaction system at certain intervals.

Naphthalene, phenanthrene, fluorene, and anthracene were chosen as the targeted PAHs due to their wide distribution in the Daliao River water system. The biodegradations of these four PAHs alone or in their mixture were conducted in microcosms with sediment samples taken from different sites of the Daliao River water system (H5, T5 and D1). Detailed experimental methods were the same as above except that mineral salt medium (MSM) replaced the site water without additional nutrient. The initial concentration of naphthalene, phenanthrene, fluorene and anthracene was 1, 0.5, 1 and 0.5 mg/L, respectively, according to each PAH water solubility and the level in river. A killed control microcosm was prepared for each experiment according to the same method as mentioned above.

For the above experiment system, background PAHs content introduced by adding sediment made up 0.06%–0.40% of the amount of PAHs spiked, therefore, it was too low and can be neglected. Each treatment was conducted in duplicates or triplicates.

MSM contained (g/L):  $\text{K}_2\text{PO}_4 \cdot 2\text{H}_2\text{O}$  4.25,  $\text{NaH}_2\text{PO}_4 \cdot 3\text{H}_2\text{O}$  1.0,  $\text{NH}_4\text{Cl}$  2.0,  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$  0.2,  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  0.012,  $\text{MnSO}_4 \cdot 7\text{H}_2\text{O}$  0.003,  $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$  0.003,  $\text{CoSO}_4 \cdot 7\text{H}_2\text{O}$  0.001.

#### 1.4 Analytical methods

During incubation period, 1 mL sediment and water mixture was sampled at certain intervals. The remaining PAHs was extracted as follows: sediment-water mixture and methanol were added to centrifuge tubes by the ratio of 1:1 (V/V), then mixed on vortex thoroughly and settled for about 1 h. Mixing and settling repeated for three times according to the above method. Finally, samples were centrifuged for 10 min at 5000 r/min. Supernatant liquid was collected and filtered through a 0.22- $\mu\text{m}$  filter, and the filtrated liquid was stored at 4°C for analysis.

Naphthalene, fluorene, phenanthrene and anthracene were analyzed by high performance liquid chromatography (Waters 1525, USA) equipped with UV-Vis detector and C18 reverse-phase column (150 mm  $\times$  4.6 mm). The quantification of PAH performed by the external standard method. The correlation coefficients for calibration curves are all higher than 0.999. Detection wavelengths were set at 254 nm. Mobile phase was methanol and water in the

ratio of 80:20 (V/V). The detection limits of naphthalene, fluorene, phenanthrene and anthracene were 0.01, 0.01, 0.01 and 0.02 mg/L, respectively. The recoveries of the four PAHs in water-sediment mixture ranged from 84% to 98%.

## 2 Results and discussion

### 2.1 Biodegradation of phenanthrene in site water alone or site water sediment combined system

Effect of sediment on the degradation of phenanthrene was investigated through conducting biodegradation in site water alone system, and site water and sediment combined system (Fig. 2). Results showed that the degradation of phenanthrene in site water alone system was much faster than that with sediment presence. Phenanthrene was nearly completely removed within 12 d in site water alone system; while it took 32 d to achieve 95% removal in the presence of sediment. For the sterile control system, phenanthrene remained as high as 83% until day 32. Thus we can deduce that the most of phenanthrene removed in sediments could be attributed to microbial degradation. One possible explanation for the inhibition of sediment on phenanthrene degradation was that the interactions between sediment matrix and PAHs such as their sorption to sediment particles would reduce the bioavailability of PAHs. Similar phenomena were also reported previously (Yuan *et al.*, 2001; Hughes *et al.*, 1997; Ramirez *et al.*, 2001). However, it was also reported that sediments did not inhibit and even enhanced the biodegradation of PAHs (Poeton *et al.*, 1999; Yu *et al.*, 2005). Poeton *et al.* (1999) reported that the degradation rates of phenanthrene and fluoranthene in the presence of sediment were 2.1–3.5 and 2.1–5.3 times faster than that in sediment-free aqueous medium, respectively. Yu *et al.* (2005) investigated the biodegradability of PAHs mixture by a bacterial consortium isolated from mangrove sediments under sediment-free and sediment slurry conditions, and found that fluorene and phenanthrene degradation percentage in sediment slurry was higher than that in liquid medium. Above research results showed that the effects of sediments on the biodegradation of

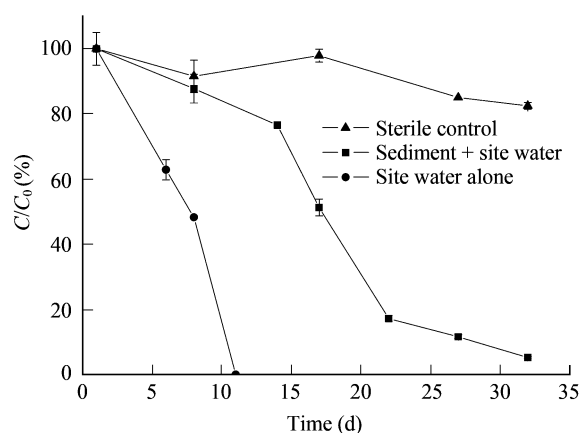


Fig. 2 Biodegradation of phenanthrene in site water alone system and site water sediment combined system.

organic pollutants in liquid system was complicated, which depends on many factors, such as bacteria species and their abundance in sediment and liquid, sediment chemical compositions, types of organic pollutants.

## 2.2 Biodegradation of PAHs in the presence of additional nutrients

Biodegradation of some specific organic pollutant in real contaminated sites is often retarded by the lack of nutrients, such as carbon sources, nitrogen and phosphorus. To boost the growth of indigenous microbial and stimulate their metabolism activity, additional nutrients are generally amended to polluted sites. In this study, sodium acetate, yeast extract, ammonium and phosphate were individually supplemented to microcosm systems and their effects on PAHs biodegradation were investigated (Fig. 3).

Figure 3a indicates that biodegradation of phenanthrene was enhanced by adding yeast extract, but retarded by sodium acetate. The remaining percentage of phenanthrene in the system with sodium acetate and yeast extract addition at day 27 was 19.5% and 4.8%, respectively, while that in the inoculated control system was 11.6%. Inhibition of sodium acetate on phenanthrene biodegradation may be due to that indigenous culture in the microcosm system preferably utilized sodium acetate compared to phenanthrene. As yeast extract could provide multiple nutrients to indigenous culture and increase its metabolic activity, the enhancement on phenanthrene degradation was detected. Yuan *et al.* (2000) also investigated the effects of adding other carbon sources on phenanthrene biodegradation by aerobic mixed culture, and found that phenanthrene degradation was enhanced by the individual addition of yeast extract, acetate, glucose or pyruvate. However, different results were obtained when they did similar biodegradation experiment in river sediment, which phenanthrene biodegradation was not significantly influenced by the addition of carbon sources such as acetate, pyruvate and yeast extract (Yuan *et al.*, 2001). Lee *et al.* (2003) reported that pyruvate could be used as an additional carbon source to stimulate the growth of *Pseudomonas putida* G7 and promoted the degradation of PAH by this strain.

Figure 3b indicates that the biodegradation of phenanthrene in sediment was stimulated by adding ammonium,

but not significantly influenced by adding phosphate, suggesting that nitrogen source was inadequate and phosphorus was abundant for phenanthrene biodegradation in water-sediment combined system. There are research works, in which various mineral salts had been used as additional nutrient to stimulate PAHs biodegradation in sediment. Yuan *et al.* (2001) studied phenanthrene biodegradation in river sediment and found that it was significantly influenced by the addition of ammonium, sulfate and phosphate. Yu *et al.* (2005) also reported that the addition of mineral salt medium could enhance pyrene biodegradation in mangrove sediment slurry. Leia *et al.* (2005) investigated the degradation potentials of PAHs that were present in aged contaminated sediment and found that amendment of inorganic N and P did not enhance the level of degradation. In Ran and Jeffrey's research (2004), three nutrient sources including inorganic soluble nutrients, the slow-release fertilizer Osmocote and Inipol EAP-22, as well as their combinations, were applied to beach sediments contaminated with an Arabian light crude oil, and Osmocote was found as the most effective nutrient source to stimulate the degradation of PAHs.

## 2.3 Biodegradation of PAHs in river sediment under low temperature

As the Daliao River locates at the joint of temperate zone and cold-temperate zone, it has a long cold winter and a hot and rainy summer. The water temperature changes apparently in different seasons. In spring, winter and autumn, the river water temperature in the unfrozen layer ranges from 1 to 5°C; in summer, it ranges from 15 to 20°C. PAHs biodegradation rates at 15 and 5°C are presented in Table 2. Results showed that naphthalene, phenanthrene and fluorene could also be degraded under low temperature (5°C), but relative lower removal rates were obtained compared to that at 15°C, suggesting metabolic activity of microorganisms was inhibited under low temperature.

## 2.4 PAHs biodegradation capability in sediments sampled from different locations

Degradation of naphthalene, phenanthrene, fluorene and anthracene alone in sediment from H5, T5 and D1 is presented in Table 3. For the sterile control, the remaining

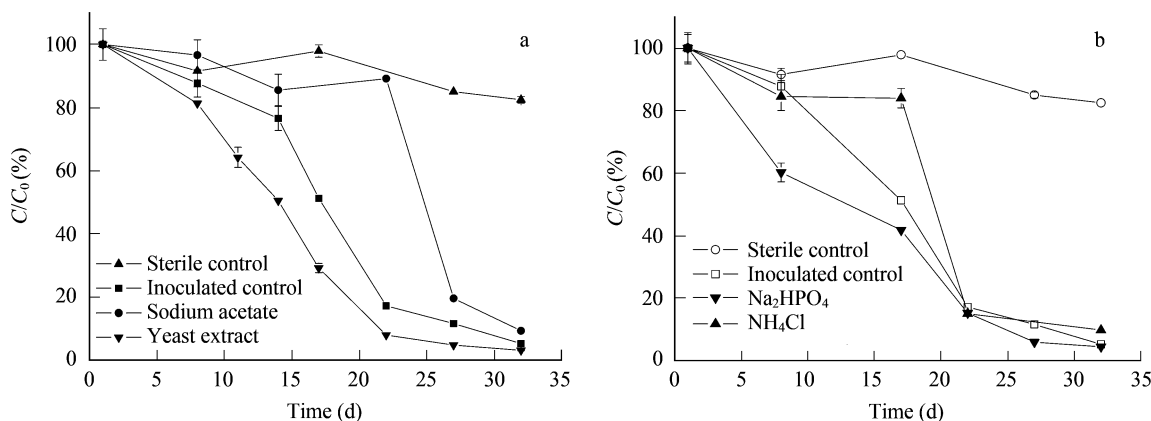


Fig. 3 Effects of carbon source addition (a) and inorganic N and P addition (b) on phenanthrene biodegradation in sediments.

**Table 2** Comparison of PAHs removal rates in T5 sediment under different temperatures

Incubation time	Average removal rate (mg/(L·d) (25°C/5°C)		
	Naphthalene	Phenanthrene	Fluorene
Day 9	0.118/0.080	0.040/0.000	0.036/0.000
Day 15	0.071/0.065	0.021/0.016	0.046/0.027
Day 19	—/—	0.017/0.011	0.038/0.022

—: PAHs were completely removed by day 19, thereby, the average removal rate was not measured. Data in the table represent mean of duplicate experimental results.

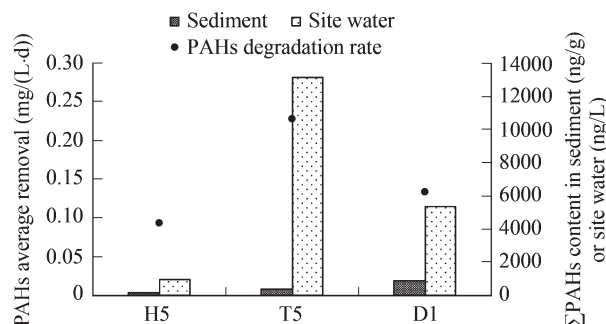
percentage of the four PAHs after a certain period of incubation was much higher than the non-sterilized system, suggesting that the removal of PAHs was mainly attributed to the function of microbial metabolism. The four targeted PAHs were degraded to different extent. Naphthalene was the most easily degraded compound and it was nearly completely removed within 15 d in the three sediment samples; while anthracene was the most poorly-biodegraded one, it could not be degraded in sediment H5 and D1 except sediment T5. However, certain degradation of phenanthrene and fluorene was observed in all sediment samples.

**Table 3** Remaining percentage of PAHs in the microcosm tests after a certain period of incubation

PAHs	Remaining percentage (%)			
	Sterile control <sup>c</sup>	H5 sediment	T5 sediment	D1 sediment
Naphthalene <sup>a</sup>	65–72	1	2	1
Phenanthrene <sup>b</sup>	93–95	15	7	14
Fluorene <sup>b</sup>	95–99	83	63	3
Anthracene <sup>b</sup>	98–100	100	15	100

<sup>a</sup> Remaining percentage at the end of 15 day incubation; <sup>b</sup> remaining percentage at the end of 19 day incubation; <sup>c</sup> sterile controls were made with H5, T5 and D1 sediment samples respectively, the range of remaining percentage is presented here.

Total PAHs average removal rates (4 targeted PAHs) by the three sediment samples were calculated, and were 0.094, 0.228 and 0.134 mg/(L·d) for H5, T5 and D1, respectively. PAHs degradation capacity followed the sequence of T5 > D1 > H5. To know the relationship between site pollution level and PAHs biodegradation capacity, total PAHs (16 types) content in surface water and sediment was measured. Sediments of H5, T5 and D1 contained 110.90, 334.10 and 816.63 ng/g total PAHs, respectively; the corresponding parts in site water were 946.1, 13145.04, 5355.68 ng/L total PAHs. Although T5 had the highest PAHs content in river water among the three sampling points, its sediment PAHs content was not the highest, suggesting PAHs in river water and sediment had not reached equilibrium due to stream turbulence. PAHs pollution level of the three sampling sites followed the sequence of T5 > D1 > H5. The relationship between PAHs content and PAHs removal rates by different sediments is plotted in Fig. 4. It could be found that PAHs biodegradation capacity posed by each sediment was positively related to its PAHs pollution level. A large quantity of PAHs in water detected at T5 probably due to the discharge of wastewater from adjacent chemical

**Fig. 4** Relation between PAHs removal rates in sediments and site pollution level.

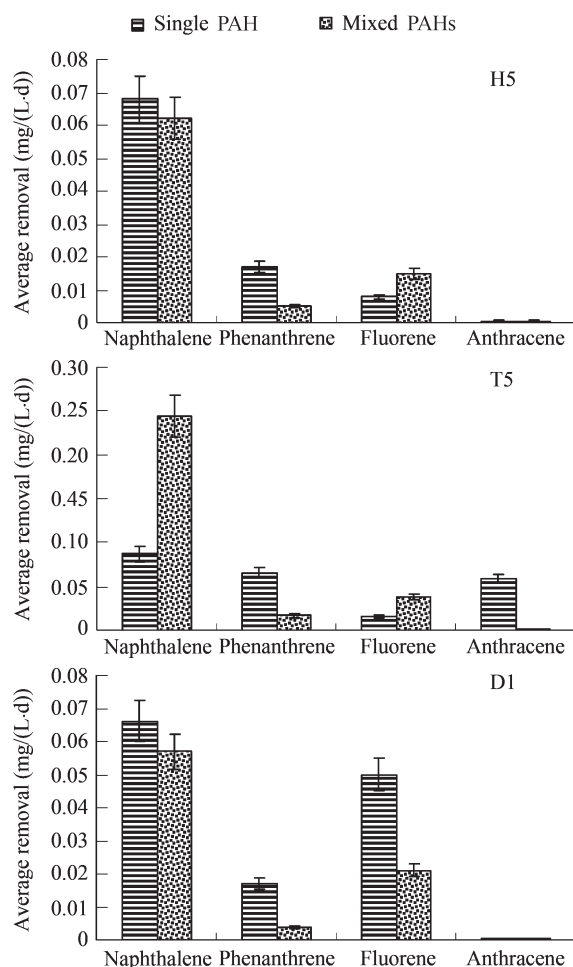
factories in Liaoyang and steel factories in Anshan. H5 received more municipal wastewater and small amount of industrial wastewater from Shenyang City, therefore, it is relatively slightly polluted by PAHs. As D1 is the joint point of the Hun River and the Taizi River, its PAHs level was in middle level. However, PAHs content was the highest in the sediment of D1. One possible explanation was that as the joint point of the Hun River and the Taizi River, the Daliao River at D1 has relative lower flow rate which may lead to a stronger PAHs deposition and distribution in sediment. Above results indicated that pre-exposition to PAHs caused the indigenous microorganisms in sediments acclimated to PAHs and posed the potential for the degradation of these pollutants, and this potential was influenced by pollution history and pollution level.

## 2.5 Biodegradation of PAHs alone or in their mixture

Generally more than one type of PAHs existed in real contaminated environment. Biodegradation of PAHs present as a pure compound may different to that as mixed compound. Therefore, biodegradation of four PAHs in sediment was investigated in single and multiple PAHs systems. The average removal rates are measured and presented in Fig. 5.

For H5 sediment samples, average removal rates of the four PAHs followed the sequence of naphthalene > phenanthrene > fluorene > anthracene when present alone, and were 0.068, 0.017, 0.008, and 0 mg/(L·d), respectively. When the four PAHs coexisted, naphthalene was degraded at first time; phenanthrene and fluorene began degradation simultaneously after about 9 d lag, but anthracene could not be degraded till the end of experiment. In multiple PAHs system, the degradations of naphthalene and phenanthrene was inhibited, but that of fluorene was enhanced. The average removal rate of the four PAHs turned to the sequence of naphthalene > fluorene > phenanthrene > anthracene, and was 0.062, 0.015, 0.005, and 0 mg/(L·d), respectively.

T5 sediments demonstrated a certain degradation ability to all four PAHs when present alone, and PAHs average removal rates followed the sequence of naphthalene > phenanthrene > anthracene > fluorene as 0.087, 0.066, 0.059, and 0.016 mg/(L·d), respectively. When the four PAHs coexisted, naphthalene was also first degraded and when it was removed almost completely, the degradations of phenanthrene and fluorene occurred simultaneously,



**Fig. 5** Average removal rates for the four PAHs existed alone or in their mixture.

but anthracene could not be degraded. Biodegradations of naphthalene and fluorene were enhanced with their average removal rates and increased to 0.244 and 0.038 mg/(L·d), respectively, while phenanthrene and anthracene were delayed greatly. Their average removal rates declined to 0.017 and 0 mg/(L·d), respectively. Average removal rates of the four PAHs followed the sequence of naphthalene > fluorine > phenanthrene > anthracene.

For D1 sediment, PAHs biodegradation rates are in the sequence of naphthalene > fluorine > phenanthrene > anthracene in single PAH system, and they were 0.066, 0.050, 0.017 and 0 mg/(L·d), respectively (anthracene could not be degraded). In multiple PAHs system, although the same degradation order as above was observed, the degradations of naphthalene, fluorene and phenanthrene were delayed, and their average removal rates declined to 0.057, 0.021, and 0.004 mg/(L·d), respectively. Anthracene still could not be degraded.

Above research results showed that interactions among multiple PAHs influenced the removal of each PAH. Some degradation was enhanced, and some was retarded. In addition, interactions among the four PAHs differed greatly in different sediments, and it was hard to correlate to PAHs molecular weight and structure. Biodegradation of multiple PAHs mixture was also investigated by other researchers, but complicated results were reported. Yuan

*et al.* (2001) investigated the PAHs biodegradation in river sediment and found that the biodegradations of anthracene, fluorene and pyrene were enhanced by the presence of phenanthrene, the order of PAHs degradation rate was: phenanthrene > acenaphthene > pyrene > fluorine > anthracene. In the simultaneous presence of all the six PAH compounds, biodegradation rate constants were reduced for phenanthrene and acenaphthene, enhanced for anthracene, fluorene and pyrene. Yuan *et al.* (2000) did a similar biodegradation experiment with mixed culture, and found that degradation rates were delayed for phenanthrene, pyrene, and acenaphthene, but enhanced for anthracene and fluorene when they existed simultaneously. James *et al.* (1988) studied the effects of co-occurring aromatic hydrocarbons on the degradation of individual polycyclic aromatic hydrocarbons in marine sediment slurries, and found that the presence of co-occurring PAHs in acclimated slurries did not appreciably affect the degradation of individual PAH. Above related research works suggest that as different sediment and microbial culture demonstrated different biodegradation pattern and ability to single PAH, their degradation pattern for multiple PAHs also varied greatly.

### 3 Conclusions

The following conclusions were obtained.

(1) Biodegradation of phenanthrene in T5 sediment was enhanced by adding yeast extract and ammonium, retarded by adding sodium acetate, not significantly influenced by adding phosphate. Phenanthrene was biodegraded faster in site water alone system than in sediment and site water combined system. PAHs could also be biodegraded under low temperature (5°C), but their degradation rates were relatively low.

(2) Sediments sampled from different locations of the Daliao River watershed demonstrated different degradation capabilities to the four targeted PAHs, and the order was T5 > D1 > H5. PAHs degradation capacity demonstrated by different sediments was positively correlated to PAHs pollution levels.

(3) For the four targeted PAHs, naphthalene was the most easily biodegraded compound, anthracene was the hardest one, which could not be biodegraded in H5 and D1 sediments. Interactions among the four PAHs influenced the removal of each PAH by sediment, and the interaction varied greatly in different sediments.

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