



## Petroleum hydrocarbons and their effects on fishery species in the Bohai Sea, North China

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### Abstract

Systematic studies on the changes in concentrations of petroleum hydrocarbons (PHs) and their effects on fishery species in the Bohai Sea during 1974–2004 are presented. Changes in PHs concentrations were closely related to Yellow River runoff. Concentrations of PHs accumulated in fish and shrimp increased by about 0.712 mg/kg dry weight when trophic level of fish and shrimps increased by 1. Attention should also be paid to the high PHs concentrations in mollusks along the coastal waters of the Bohai Sea. Average concentration of PHs in the adjacent coastal waters of Tianjin City during 1996–2005 decreased the population growth rates of fish, crustaceans and mollusks in the Bohai Sea by 2.58%, 6.59% and 2.33%, respectively. Therefore, PHs have significantly contributed to the decline in fisheries in the Bohai Sea, and they must be reduced to realize the sustainable fisheries.

**Key words:** petroleum hydrocarbons; fishery resources; population growth rate; toxicity; Bohai Sea

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### Introduction

The Bohai Sea (117°32'E–122°08'E, 37°07'N–40°55'N) is the main area for ocean petroleum exploitation in China and it has received a large amount of oil-containing sewage from the surrounding regions in recent decades (Song, 2009; Zhang et al., 2009). Moreover, oil spills are frequent and petroleum hydrocarbons (PHs), as the primary components of oils, are the main pollutants in the Bohai Sea (SOAC, 2004–2008).

PHs in seawater can result in fisheries decline by affecting the growth, reproduction, and survival of fishery species. For example, the Exxon Valdez oil spill resulted in a significant reduction (52.3%) in Pacific herring larval production in Prince William Sound in 1989 (Brown et al., 1996). Bohai Sea fisheries resources have declined considerably in recent decades (Tang et al., 2003), and it has become imperative to understand the quantitative effect of PHs on their decline.

Previous studies have mainly focused on the toxic effects of PHs on molecule-, cell-, tissue- and individual-levels of marine organisms (Carls et al., 1999; Colavecchia et al., 2004; Georgiades et al., 2006; Heintz et al., 1999; Martins et al., 2005; Pollino and Holdway, 2002; Reynaud

and Deschaux, 2006; Tang and Zhang, 2000; Thomas et al., 2007; Wu et al., 2007). Despite the fact that PHs are one of the main pollutants in the Bohai Sea, few studies have examined the quantitative effects of PHs pollution on population dynamics of fishery species in the Bohai Sea, mainly due to the following reasons: (1) most toxicological tests have focused on the effect of individual PHs fractions on marine organisms, despite various PHs coexisting in the field marine environment; (2) PHs concentrations used in previous toxicological tests were often much higher than PHs concentrations in the Bohai Sea; (3) most studies utilized acute toxicity tests, and there was a distinct lack of detailed data of long-term chronic toxicity tests; (4) fishes in the Bohai Sea were seldom used in previous studies on toxicological effects of PHs; and (5) population-level effects of PHs on fishery species were complicated because of various influencing factors, such as life history characteristics and feeding habits of the organisms. These factors made it very difficult to assess the population-level effects of PHs on fishery species in the Bohai Sea. Nevertheless, it was necessary to promote the transition to determine the effect of PHs on fishery resources from qualitative to semiquantitative method, then to quantitative method.

On the basis of the above analysis, we studied the

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changes of PHs concentrations in the Bohai Sea during 1974–2004, and assessed effects of PHs on fishery species in the Bohai Sea using existing toxicity data. The objectives of our study were to: (1) discuss the changes and influencing factors of PHs concentrations in the Bohai Sea in recent decades; (2) determine the accumulation characteristics of PHs in fishery species in the Bohai Sea; and (3) assess roughly the semiquantitative effects of PHs on population growth rates of several common fishery species in the Bohai Sea.

## 1 Materials and methods

### 1.1 Regional characteristics of the Bohai Sea

The Bohai Sea, with an area of about  $7.7 \times 10^4$  km<sup>2</sup>, is composed of Bohai Bay, Laizhou Bay, Liaodong Bay, and the Central Part. It is a semi-enclosed marginal sea connected to the Yellow Sea through a 50-mile wide mouth in the southeast. The exchange ability of water between the Bohai Sea and the Yellow Sea was poor, and the environmental capacity of the Bohai Sea is limited. Nevertheless, many pollutants have been discharged into the Bohai Sea from the surrounding regions, and its coastal waters have become polluted (Zhang et al., 2010). The coastal waters of the Bohai Sea are the main spawning and nursery area for fish and shrimp. With the deterioration of the environment, fisheries resources in the Bohai Sea have declined obviously in recent decades.

### 1.2 Data sources and methods

The PHs concentrations in seawater of the Bohai Sea were mainly determined by ultra-violet spectrophotometry and fluorospectrophotometry following China's Marine Monitoring Standards. The determination principles, detection limits, and standard oils between the two determination methods were different. Therefore, it was necessary to assimilate the PHs concentrations determined by the two methods to enhance the comparability of PHs concentrations over the years. Wang and Li (2006) assimilated PHs concentrations to ultra-violet spectrophotometry in the Bohai Sea during 1974–2004, which were used in this study.

Concentrations of PHs accumulated in fishery species were obtained from Cui (1991). Trophic levels of fish and shrimp were obtained from Yang (2001a, 2001b). Toxic effects of PHs (South China Sea (SCS) crude oil) on *Acanthopagrus schlegelii* juveniles, *Penaeus japonicus* juveniles, and *Scapharca subcrenata* were from Jia et al. (2000). We adopted these toxicity results as they have the same toxicants and similar test methods and the test animals are common fishery species in the Bohai Sea.

## 2 Results and discussion

### 2.1 Changes of PHs concentrations in seawater of the Bohai Sea

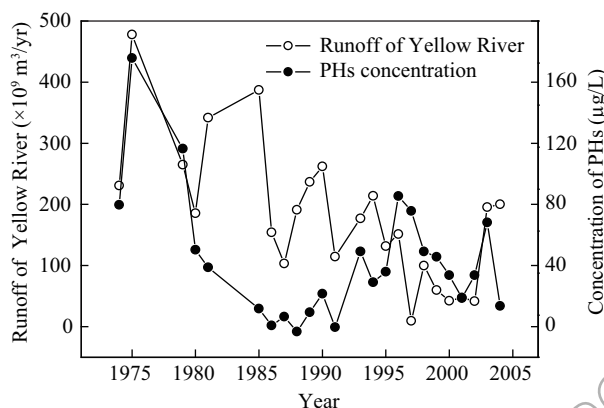
The average concentration of PHs was about 46.5 µg/L in the Bohai Sea seawater during 1974–2004. Changes in

PHs concentrations during 1974–2004 were divided into three periods: (1) decreasing obviously from 1974 to 1987 ( $R_{1974-1987} = -0.834$ ,  $p < 0.05$ ,  $n = 8$ ); (2) increasing gradually from 1988 to 1995 ( $R_{1988-1995} = 0.775$ ,  $p < 0.05$ ,  $n = 7$ ); and (3) decreasing from 1996 to 2004 ( $R_{1996-2004} = -0.670$ ,  $p < 0.05$ ,  $n = 9$ ).

Yellow River runoff may influence PHs concentrations in seawater of the Bohai Sea. We found close positive correlations between PHs concentrations in seawater of the Bohai Sea and the Yellow River runoff during 1974–1995 ( $R = 0.575$ ,  $p < 0.05$ ,  $n = 16$ ) and during 1996–2004 ( $R = 0.860$ ,  $p < 0.05$ ,  $n = 7$ ; except in 1997 and 2004). One possible reason was that PHs were discharged into the Bohai Sea with Yellow River runoff. About 1610 tons of PHs were discharged into the Bohai Sea with Yellow River runoff in 2003 (Liu, 2010); in 2004, petroleum was also a main pollutant in Yellow River runoff at Lijin Hydrologic Station (the nearest station to the Yellow River mouth) (YRCC, 2004). Yellow River runoff has shown a decreasing trend in recent decades (Fig. 1), which has resulted in increasing salinity in seawater of the Bohai Sea (Wu et al., 2004; Zhang et al., 2006). With increasing salinity, more PHs in seawater can be absorbed by sediments (Xia, 2005), and PHs concentrations decreased in seawater of the Bohai Sea. Therefore, a positive correlation was observed between Yellow River runoff and PHs concentrations in seawater of the Bohai Sea.

Yellow River runoff decreased obviously from 1996 to 1997, whereas PHs concentrations in seawater of the Bohai Sea decreased only slightly. This was probably related to frequent oil spills in 1997, with four large oil spills occurring during the short period from March 30th to April 8th in 1997 in the Bohai Sea. Yellow River runoff was a little larger in 2004 than in 2003, while PHs concentrations in the Bohai Sea decreased obviously from 2003 to 2004. Reasons for this are unknown, and further study is needed.

The PHs concentrations were higher in the adjacent waters of Tianjin City than in the Bohai Sea during 1996–2004 (Fig. 2). The PHs concentrations in the adjacent waters of Tianjin City were determined by fluorospectrophotometry, while those in the Bohai Sea were values assimilated to ultra-violet spectrophotometry. PHs concentrations determined by fluorospectrophotometry are lower



**Fig. 1** PHs concentrations in seawater of the Bohai Sea and the runoff of Yellow River during 1974–2004.

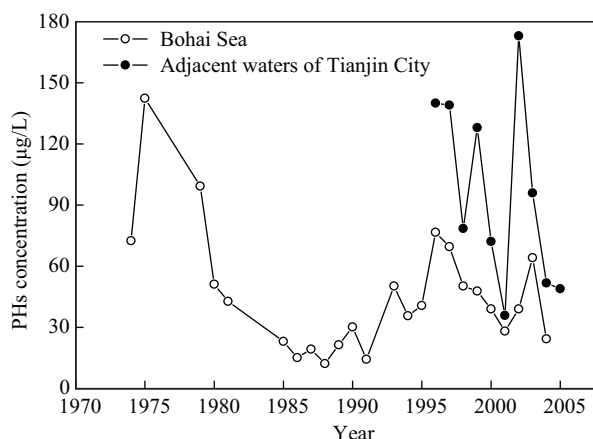


Fig. 2 PHs concentrations in seawater of the Bohai Sea and in the adjacent waters of Tianjin City.

than those determined by ultra-violet spectrophotometry when they are lower than dozens of  $\mu\text{g/L}$  (Zhao et al., 2006). Therefore, PHs concentrations in the adjacent waters of Tianjin City should be much higher than those in the seawater of the Bohai Sea. The Bohai Sea coastal waters, especially the water of Bohai Bay and Laizhou Bay, are important spawning and feeding sites for fishery species. Thus, PHs concentrations in the coastal waters of the Bohai Sea should be used to assess the toxic effects of PHs on fishery species other than PHs concentrations in seawater of the whole Bohai Sea.

Sewage discharge and oil spills have important effects on PHs concentrations in seawater of the Bohai Sea coastal waters. In the adjacent waters of Tianjin City, the average PHs concentration during 1996–2005 (Li et al., 2010) was much higher in sampling sites near the Dagu Drainage River mouth than in other sampling sites, indicating the important impact of sewage discharge on PHs concentrations in the coastal waters of the Bohai Sea. The PHs concentrations in 2002 were significantly higher in November than in May and August in the adjacent waters of Tianjin City (Li et al., 2010). The maximum PHs concentration in November 2002 was up to  $1400 \mu\text{g/L}$ , while the maximum PHs concentrations in May and August were lower than  $100 \mu\text{g/L}$  and  $200 \mu\text{g/L}$ , respectively. This was probably related to the November 2002 oil spill in the adjacent waters of Tianjin City, which covered an area of about  $4.6 \text{ km}$  long by  $2.6 \text{ km}$  wide and resulted in the high PHs concentrations in the adjacent waters of the Tianjin City.

## 2.2 Accumulation of PHs in organisms

There was significant positive correlation between the trophic levels and PHs contents accumulated in *Harengula ovalis*, *Ablennes anastomella*, *Engraulis japonicus*, *Scomberomorus niphonius*, *Sphyrna pinguis*, *Argyrosomus argentatus*, *Pseudosciaena polyatis*, *Lateolabrax japonicus*, *Clupanodon punctatus*, *Sepiella maidroni*, *Loligo japonica*, *Penaeus orientalis*, and *Trachypenaeus curvirostris* (Stimpson) (Cui, 1991) (Fig. 3). Due to the lack of trophic levels of *Loligo japonica* and *Penaeus orientalis*, the trophic levels of their similar species, *Loligo beka* and

*Penaeus chinensis*, were used as those of *Loligo japonica* and *Penaeus orientalis*, respectively. The linear equation between them was as follows:

$$Y = 0.712X + 1.557 \quad R = 0.673, p < 0.05, n = 13 \quad (1)$$

where,  $Y$  (mg/kg dry weight) is contents of PHs accumulated in fish and shrimp and  $X$  is trophic level of fish and shrimp. Based on Eq. (1), contents of PHs accumulated in fish and shrimp increased by about  $0.712 \text{ mg/kg dry weight}$  when the trophic level increased by 1.

PHs contents in mollusks were higher than  $15 \text{ mg/kg wet weight}$ , which was the upper limit of PHs contents in the first class marine mollusks (GB18421-2001) in the adjacent waters of Laohuwei, Xiaobijiashan, Laohekou, Huludao, Nanpaiwuhe River mouth, and Laizhou Bay. This indicated that these areas were heavily polluted by PHs.

Contents of PHs accumulated in mollusks were closely related to PHs concentrations in the water. Within the range of  $1\text{--}400 \mu\text{g/L}$  in seawater, PHs concentrations were proportional to PHs contents in mussels in the waters of Zhoushan (Wang et al., 2008b). PHs concentrations in the Bohai Sea were also in the range of  $1\text{--}400 \mu\text{g/L}$ , and thus we supposed that PHs contents in mollusks were proportional to those in seawater of the Bohai Sea during 1974–2004. Therefore, the average PHs content in mollusks during 1992–2004 was estimated to be about 2.1 times as high as that in mollusks during 1990–1991 based on PHs contents in mollusks during 1990–1991 (Table 1) and the average PHs concentration in seawater of the Bohai Sea during 1990–1991 ( $22.3 \mu\text{g/L}$ ) and during 1992–2004 ( $47.1 \mu\text{g/L}$ ). Using the value 2.1, the average PHs content in most mollusks along the coastal waters of the Bohai Sea was estimated to be higher than  $15 \text{ mg/kg wet weight (ww)}$  during 1992–2004. This should be paid attention to during marine environmental management and protection of the Bohai Sea.

## 2.3 Lethal effects of PHs on marine organisms

Early life history stages are often considered to be the time when recruitment is at least coarsely established for many marine species (Fogarty et al., 1991; Rose,

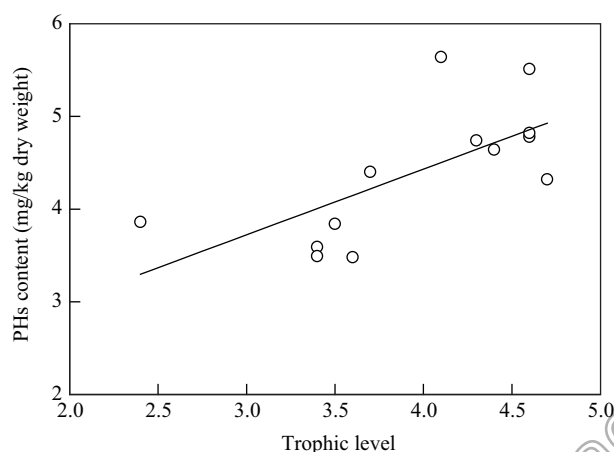


Fig. 3 Relationship between contents of PHs in fish and shrimps and their trophic levels.

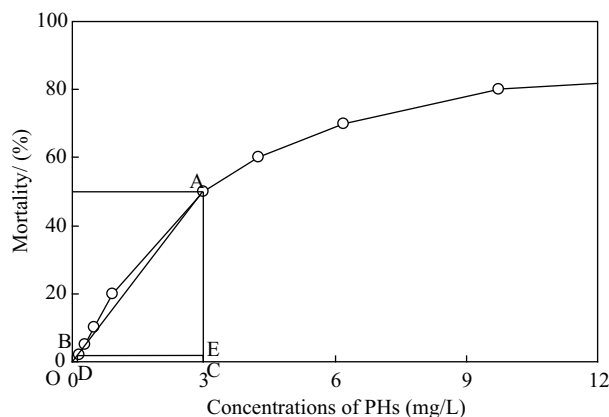
**Table 1** PHs contents in mollusks in the coastal waters of the Bohai Sea (1990–1991) (Zhao et al., 2006)

Sampling site	Species	Concentration (mg/kg ww)
Longwangtang, Lvshun	<i>Mytilus edulis</i>	5.8
Laohuwei, Lvshun	<i>Ruditapes philippinarum</i>	23.2
Baigangzi, Lvshun	<i>Mytilus edulis</i>	11.1–11.8
	<i>Ruditapes philippinarum</i>	
Beihai, Lvshun	<i>Ruditapes philippinarum</i>	7.30–7.90
	<i>Mytilus edulis</i>	
Fuzhou Bay	<i>Mytilus edulis</i>	9.5
	<i>Ruditapes philippinarum</i>	
Xiongyue	<i>Ruditapes philippinarum</i>	8.8
Bayuquan	<i>Mytilus edulis</i>	2.50–13.1
	<i>Ruditapes philippinarum</i>	
	<i>Scapharca subcrenata</i>	
Liaoh River mouth	<i>Meretrix meritrix</i>	9.30–11.3
	<i>Scapharca subcrenata</i>	
Xiaobijiashan, Jinzhou Bay	<i>Ruditapes philippinarum</i>	25.3–40.3
	<i>Scapharca subcrenata</i>	
Laohekou, Jinzhou Bay	<i>Ruditapes philippinarum</i>	34.5
Huludao, Jinzhou Bay	<i>Ruditapes philippinarum</i>	16.8–46.1
	<i>Scapharca subcrenata</i>	
Shanhaiguan	<i>Ruditapes philippinarum</i>	14.1–17.1
	<i>Scapharca subcrenata</i>	
Hangu, Bohai Bay	<i>Mactra quadrangularis</i>	12.6–53.0
	<i>Scapharca subcrenata</i>	
Nanpaiwuhe, Bohai Bay	<i>Scapharca subcrenata</i>	30.2
Majiahekou, Bohai Bay	<i>Crassostrea gigas</i>	14.4
The Yellow River mouth	<i>Meretrix meritrix</i>	7.80–14.0
	<i>Mactra quadrangularis</i>	
Laizhou Bay	<i>Scapharca subcrenata</i>	16.6

2000). Early life stages of most fish species are sensitive to changes in environmental conditions (McKim, 1985). We chose several common fishery species in the Bohai Sea, such as *Acanthopagrus schlegel* juvenile, *Penaeus japonicus* juvenile and *Scapharca subcrenata* to assess lethal effects of PHs on fishery species in the Bohai Sea. The average PHs concentration in the adjacent waters of Tianjin City during 1996–2004 (0.0963 mg/L) was used in the assessment.

LC50 is the most common indicator used to reflect the toxicity of pollutants. Details on the mortality corresponding to PHs concentrations used in toxicity tests to calculate LC50s are scarce, which makes it very difficult to assess the lethal effects of PHs on organisms in the Bohai Sea.

We used 96 hr LC50s to estimate the mortality of organisms exposed to PHs at the concentration of 0.0963 mg/L. The method is as follows. Changes in mortality exhibit an “S” curve with toxicant concentrations (Arufe et al., 2004; Colavecchia et al., 2004; Wang et al., 2008a) (Fig. 4). The coordinates of points A, B, C, D, E and O are (LC50, 50), (0.0963, y), (LC50, 0), (0.0963, 0), (LC50, y) and (0, 0), respectively. Points A and B are located in the “S” curve. Points C and D are projections of points A

**Fig. 4** Method for assessment of organisms' 96 hr mortality.

and B on the X axis, respectively. Point E is the projections of point B on the line AC. Because LC50s in Table 2 are significantly higher than 0.0963 mg/L, the slope of line AB is approximately equal to that of line AO. In right-angled triangle ABE, the length of line AE is equal to the length of line BE multiplied by the slope of line AB. The length of line AE is equal to 50 minus y, the length of line BE is equal to the length of line CD (that is LC50 minus 0.0963), and the slope of AB is equal to 50 divided by LC50. Therefore, an equation can be obtained:

$$50 - y = (LC50 - 0.0963) \times 50 / LC50 \quad (2)$$

From Eq. (2), y can be obtained as follows:

$$y = 50 \times 0.0963 / LC50 \quad (3)$$

where, y is the mortality of organisms exposed to PHs at the concentration of 0.0963 mg/L and the unit of LC50 is mg/L. The mortality of organisms exposed to PHs at the concentration of 0.0963 mg/L can be obtained using Eq. (3) (Table 2).

Organisms in the Bohai Sea have experienced long-term exposure to PHs. Thus, a 96 hr mortality measurement is insufficient to quantify the long-term toxic effects of PHs. There are, however, few relevant long-term exposure toxicity tests on common fishery species in the Bohai Sea. Due to the limited toxicity data, the relationship between 96 hr mortality and long-term mortality of juvenile rainbow trout exposed to naphthalene was used to estimate the long-term mortality of several common fishery species exposed to PHs at the concentration of 0.0963 mg/L. Based on the study of Millemann et al. (1984), we estimated the relationship between mortality of juvenile rainbow trout under long-term exposure to naphthalene as follows:

$$Y = 1.64 \log X + 6.54 \quad (4)$$

**Table 2** Estimation of 96 hr mortality, long-term mortality and reduction in population growth rate ( $\lambda$ ) of organisms exposed to PHs at the concentration of 0.0963 mg/L

Species	LC50 (mg/L)	Toxicant	Test method	96 hr mortality (%)	Long-term lethality (%)	Reduction in $\lambda$ (%)
<i>Acanthopagrus schlegel</i> juvenile	5.89	SCS crude oil	96 hr static water test	0.82	12.04	2.58
<i>Penaeus japonicus</i> juvenile	2.4	SCS crude oil	96 hr static water test	2.01	6.59	6.59
<i>Scapharca subcrenata</i>	6.76	SCS crude oil	96 hr half static water test	0.71	2.33	2.33

where,  $Y$  (mg/L) is the probability unit of mortality and  $X$  is naphthalene concentration. Using Eq. (4), the mortality of juvenile rainbow trout was estimated to be approximately 44.18% when exposed to 0.0963 mg/L of naphthalene over a long period. Based on the Eq. (3) and DeGraeve et al. (1982), the 96 hr mortality of juvenile rainbow trout was about 3.01% when exposed to 0.0963 mg/L of naphthalene. Therefore, the mortality of rainbow trout under long-term exposure to naphthalene (0.0963 mg/L) was about 14.68 times as high as the 96 hr mortality of rainbow trout exposed to naphthalene (0.0963 mg/L). Using the value 14.68, the rough estimation of the long-term mortality of *Acanthopagrus schlegelii* exposed to PHs (0.0963 mg/L) is given in Table 2.

Relationship between the long-term mortality and 96 hr mortality of *Rhithropanopeus harrisii* was used to estimate the long-term mortality of crustaceans and mollusks due to the limited toxicity data. The ratio of long-term mortality to 96 hr mortality of *Rhithropanopeus harrisii* was estimated to be about 3.28 in this study based on Laughlin et al. (1978). Using the value 3.28, the rough estimation of the long-term mortality of juvenile *Penaeus japonicus* and *Scapharca subcrenata* exposed to 0.0963 mg/L of PHs is given in Table 2.

The mortality of *Scapharca subcrenata* was lower than that of juvenile *Penaeus japonicus* (Table 2). However, this did not indicate that the toxic effect of PHs on mollusks was less than that on crustaceans. The reason for the lower mortality of *Scapharca subcrenata* was probably that the juvenile *Penaeus japonicus* were in the early life history stage, whereas *Scapharca subcrenata* were not (the shell length of *Scapharca subcrenata* was about 21–30 mm). Organisms in their early life history stages are especially sensitive to pollutants. Therefore, mortality of the juvenile *Scapharca subcrenata* was probably much higher than 0.71%.

The juvenile mortalities of fishery species are not equal to the reduction in their population growth rate due to various factors, such as different life histories and feeding habits. We assessed roughly the reduction in population growth rates of *Acanthopagrus schlegelii*, *Penaeus japonicus* and *Scapharca subcrenata* using the relationship between reduction in juvenile survival rate, and reduction in population growth rates of fish and benthos (Fig. 5). The population growth rates of *Acanthopagrus schlegelii*, *Penaeus japonicus* and *Scapharca subcrenata* were estimated to be reduced by 2.58%, 6.59% and 2.33% (Table 2), respectively, when they were exposed to 0.0963 mg/L of PHs over a long period.

The long-term mortality of *Scapharca subcrenata* in Table 2 was used as the juvenile mortality of *Scapharca subcrenata*. However, juvenile mortality is often higher than adult mortality when they were exposed to the same concentration of toxicants. Therefore, the population growth rate of *Scapharca subcrenata* might be underestimated in this study. Moreover, SCS crude oil was used as the toxicant in this study. In general, toxicity of crude oil is lower than that of other PHs (Anderson et al., 1974; Jia et al., 2000). The toxicity of PHs in seawater of the Bohai

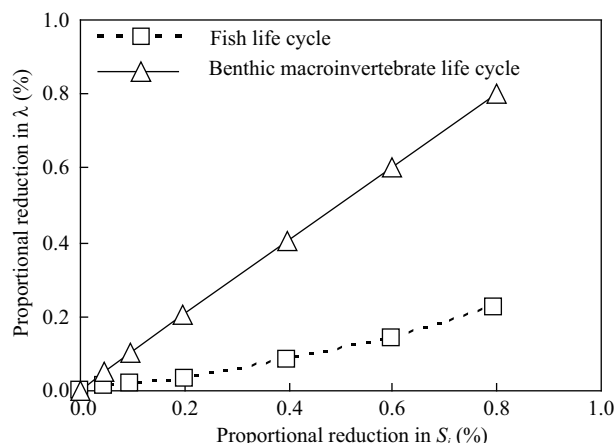


Fig. 5 The proportional reduction in population growth rate ( $\lambda$ ) resulting from a given proportional reduction in juvenile survival rate ( $S_j$ ) for different life-cycle types according to (Forbes et al., 2001).

Sea was probably higher than that of SCS crude oil. This may relate to the fact that the annual PHs concentrations in the adjacent waters of Tianjin City during 1996–2005 was averaged from spring, summer and autumn in every year, and thus PHs in seawater should have undergone weathering in the adjacent waters of Tianjin City. The proportion of PAHs, which were more toxic than other PHs in oils, was higher in more weathered oil than in less weathered oil (Carls et al., 1999). Thus, PHs in seawater of the Bohai Sea probably had more toxic components than SCS crude oil. We may, therefore, have underestimated the toxic effects of PHs on fishery species in the Bohai Sea. Population growth rates of fishery species should be higher than the values shown in Table 2. Nevertheless, the results indicated that PHs played an important part in fisheries decline in the Bohai Sea.

### 3 Conclusions

Average PHs concentration was approximately 46.5  $\mu\text{g/L}$  in seawater of the Bohai Sea during 1974–2004. Changes of PHs concentrations exhibited a decreasing trend during 1974–1987, an increasing trend during 1988–1995, and then a decreasing trend during 1996–2004. Yellow River runoff showed a close positive correlation with PHs concentrations in Bohai Sea seawater. Sewage discharge and oil spills also greatly impacted PHs concentrations in the coastal waters of the Bohai Sea.

Contents of PHs accumulated in fish and shrimp exhibited an increasing trend with increasing trophic levels. Content of PHs accumulated in fish and shrimp increased by about 0.712 mg/kg dry weight when the trophic level increased by 1. Particular attention should be paid to the high PHs contents in Bohai Sea mollusks after the 1990s.

PHs played an important part in fisheries decline in the Bohai Sea. Average concentration of PHs in the adjacent waters of Tianjin city during 1996–2005 (96.3  $\mu\text{g/L}$ ) resulted in the reduction of population growth rate of several common fish, shrimp, and mollusk species by 2.58%, 6.59% and 2.33%, respectively. Therefore, PHs

concentrations in seawater of the Bohai Sea, especially in the coastal waters, should be reduced to promote sustainable fisheries in the Bohai Sea.

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