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Polycyclic Aromatic Hydrocarbons in Sediments of the Yellow Sea

Dong Beom Yang, Jun Yu, Ki Bok Lee, Kyung Tae Kim,
Chang Soo Chung, and Gi Hoon Hong

Marine Chemistry Division, KORDI
Ansan P.O. Box 29, Seoul 425-600, Korea

Abstract: Surface sediment samples collected from the eastern half of the Yellow Sea proper in 1998 were analyzed for polycyclic aromatic hydrocarbons (PAHs), ubiquitous pollutants. Total PAHs concentrations varied from 1.0 to 320.5 ng g^{-1} dw. Relatively high concentrations of PAHs were found in the muddy central part of the Yellow Sea. Sedimentary total PAHs concentrations in the Yellow Sea proper were similar to those of Californian offshores and the central Mediterranean Sea, albeit an order of magnitude lower than the Yellow Sea nearshore areas. Phenanthrene/Anthracene concentration ratio of PAHs in bottom sediments suggested that pyrolytic PAHs might be dominant over petrogenic ones in the eastern Yellow Sea. Downcore depth distributions of PAHs from the relatively undisturbed core samples of the central Yellow Sea showed decreasing PAHs concentrations with core depths and suggested that the Yellow Sea has been increasingly exposed to PAH for decades. Annual total PAH flux to these sediments was estimated to be 166 $\mu g m^{-2} yr^{-1}$ in the central part of the Yellow Sea for the recent decade.

Key words: Yellow Sea, PAH, Pollutant flux.

1. Introduction

A wide range of polycyclic aromatic hydrocarbons (PAHs) enter the natural and human environment in an increasing amount in recent years by direct exposures to petroleum containing industrial wastes, combustion products and automotive exhausts (Hites *et al.* 1980; Wakeham *et al.* 1980; Witt 1995), and by direct contamination through gas and petroleum processing operation and refinery byproducts (Jones *et al.* 1986). Most of these compounds are highly carcinogenic/mutagenic at relatively low levels (White 1986). In general, sources of PAHs in the marine environment are biogenic and diagenetic (e.g., chrysenes, picene, perylene, retene), petrogenic (naphthalenes, fluorenes, phenanthrene, dibenzothiophenes and chrysene), and pyrogenic (e.g. unsubstituted parent compounds of the 2-, 4-, and 5-ring PAHs, anthracene, fluoranthene, pyrene) hydrocarbons (Fernandes *et al.* 1997; Page *et al.* 1998; Tolosa *et al.* 1996). Regardless of origin, the resulting contamination consists of many individual PAHs with varying chemical characteristics. However, combustion temperatures also determine the molecular signatures of PAHs. PAHs formed at high temperatures are dominated by unsubstituted species, while formation at low temperatures favors a higher degree of alkylation. Thus, non-alkylated/alkylated ratios are high for pyrogenic PAH mixtures and low in petrogenic PAHs and often PAHs formed at low temperatures. Pyrolytic processes, either natural or anthropogenic, are extremely variable depending on the regional environmental setting. However, any

relative contribution of these two processes is not known in the waters and air-sheds of the Yellow Sea.

Once PAHs are introduced to the sea, depending on their different water solubilities (i.e. extremely hydrophobic chemicals), and on the size and the organic matter content of particles, PAHs in water tend to associate with particulate matter (Gearing *et al.* 1980). Subsequently, settling of the particulate matter, flocs, and fecal pellets favor the transport of those hydrocarbon complexes to bottom sediments, which act as a long-term reservoir of these toxic contaminants (Landrum and Robbins 1990). The chemical composition of hydrocarbon mixtures in sediments reflects the relative contributions of different natural and anthropogenic sources as mentioned earlier. In the vicinity of highly populated and industrialized areas anthropogenic inputs are prominent, while in remote areas most hydrocarbons are mainly from a natural origin. Thus sediment is the first choice for assessing balance and effects of PAHs in a given ecosystem.

Investigation on persistent organic pollutant inputs to the Yellow Sea started in the 1970s. However, most studies on distribution of contaminants and their bioeffects have been confined to coastal areas, and not much information has been available on the distribution of organic contaminants in the offshore areas of the Yellow Sea. This paper is based on the results of the offshore environmental survey of the Yellow Sea conducted in 1998.

Environmental Settings

Situated between China and Korea, the Yellow Sea is a semi-enclosed, shelf-type shallow basin with reduced water exchanges with the East China Sea. Its total surface area is approximately 380,000 km^2 with the depth averaging 44 m. Recently, environmental degradation of the Yellow Sea became a major concern because of unawareness of pollutant input and its effects on regional marine ecosystem. Possible harmful impact of the significant increases in industrialization along the coastal areas of China and Korea is a nervously anticipated issue. In the north of the Yellow Sea, there are oil fields in the Bohai and West Korean Bay. Petroleum production at Shengli area near the mouth of the Yellow River is approximately 27×10^6t yr^{-1} (Bigot *et al.* 1989). However, petrogenic hydrocarbon contamination due to the exploitation and refinery of petroleum from the southern Bohai Bay appeared to be very low, although the presence of hopane suggested a possible contamination by the activities of the Shengli oilfield (Bigot *et al.* 1989).

The Yellow Sea receives a large amount of sediments from the adjacent lands, largely through the Yellow River in the Bohai Sea (1.0×10^9t yr^{-1}, Yang Z. *et al.* 1998) with lesser extent from a number of smaller rivers which drain China (20 and 9.1 x 10^6 t yr^{-1} for Luanhe and Shuangtaizihe in the Bohai Sea, respectively) and the Korean Peninsula [4.8, ca.12.3, 5.7 x 10^6 t yr^{-1} for Aprock (Yalujiang), Han, and Keum Rivers in the Yellow Sea, respectively(Zhang *et al.* 1997; Hong *et al.* 1995)]. The riverine sediments transported to the sea are mostly deposited in the lower reaches of the river and within the estuary and only less than 1% of the Yellow River sediment (ca. 6 × 10^6 t yr^{-1}; Martin *et al.* 1993) is transported to the Yellow Sea. The Changjiang discharges 4.7×10^8 t yr^{-1} with a factor of two variations (Shen *et al.* 1998). Although the bulk of this sediment is transported southward through the East China Sea by Jiangsu Coastal Current (Milliman *et al.* 1985), a much smaller portion of water and suspended sediment is also transported to the southern part of the Yellow Sea and reaches the Cheju Strait (Ahn *et al.* 1999), however their magnitudes have not yet determined.

The Yellow Sea also receives a significant amount of sediment via atmosphere since it is located along the pathway for the mineral dust (Yellow Sand Storms). Annual atmospheric flux of the mineral dust to the Yellow Sea was estimated to be ca. 53.7 g $m^{-2}yr^{-1}$ (Zhang *et*

al. 1993) which accounts to be ca. 20% of the total annual riverine sediment supply to the Yellow Sea. These estimates are with an uncertainty of a factor of three (Gao *et al.* 1992; Liu *et al.* 1998; Hong *et al.* 1998). Contaminants transported via atmosphere affect the entire sea area unlike the ones transported by the river which are confined mostly in the limited regional estuarine area.

2. Materials and Methods

Sediment samples were collected using a grab sampler aboard R/V Eardo of the Korea Ocean Research and Development Institute in April 1998. Sampling locations are shown in Fig 1. Surface sediments were collected with a Van Veen grab sampler and top centimeters of sediments were carefully subsampled and frozen for analyses of PAHs. Sediment cores were taken with a box corer. More detailed procedures on sampling and analyses are described elsewhere (Yang 1998).

PAH analysis

The samples were stored frozen in solvent-washed jars until analyzed. Extraction of PAHs basically followed the method described by APHA/AWWA/WPCF (1981). Freeze-dried sediments were ground on an agate mortar after eliminating shell residues. Sediments were then sieved through 200 mesh nylon screen. Ten grams of sediment samples spiked with internal standards were placed in 50 ml glass vials. 30 ml of n-hexane/acetone (9:1) was added to each samples. Samples were then sonicated for 60 minutes and centrifuged for 20 minutes. This extraction step was repeated three times and all the extracts were then combined. The extracts were concentrated to 3-5 ml in a rotary evaporator at 40-45°, and then stood overnight with activated copper to remove sulfur. After the cleanup procedure using alumina/silica column, the extracts were finally concentrated to 1 ml under nitrogen stream. A Rexchrom S5-100 ODS (25cm X 4.6mm I.D.) column was

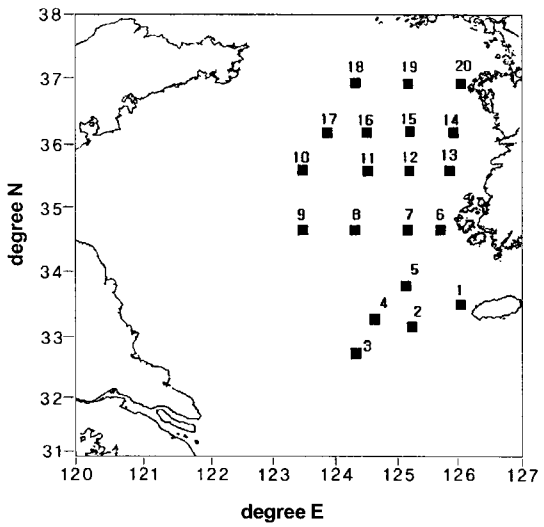

Fig. 1. Sampling stations in the Yellow Sea.

used for the separation of PAH components. 20 ㎕ of extracts were injected to HPLC (SpectraSYSTEM) equipped with an ultraviolet detector (254 nm). After a sample injection, elution was started with 30% aqueous acetonitrile for the first 10min, then 27% aqueous acetonitrile for the next 7min (flow rate of 1.3 ml/min), and finally with 100% acetonitrile for 5min (1.0 ml/min). For column recondition, the column was washed with 20% aqueous acetonitrile for 5 min (1.3 ml/min). Recovery assays for the overall procedure were carried out with spiked standard reference materials (NBS 1941). The results were over 90% for PAHs. Results were not corrected for recovery yields. The average reproducibility (1 σ) of the method varied less than 10% for duplicate samples.

3. Results and Discussion

The PAHs evaluated in this study were naphthalene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, chrysene, benz(a)anthracene, benzo (g,h,i)perylene and dibenzo(a,h)anthracene. In this paper, concentrations of total PAHs represents the sum of the quantified PAHs. Concentrations of individual PAHs below detection limit were excluded from total PAH con-

tents. Phenanthrene, anthracene and total PAH concentrations in surface sediment of the southern Yellow Sea are given in Table 1. Phenanthrene concentrations ranged from <0.5 to 16.3 ng g^{-1} dw. Anthracene concentrations varied in the range of <0.5- 9.7 ng g^{-1} dw. Total PAH concentrations varied from 1.0 to 320.5 ng g^{-1} dw (m = 80.0 ng g^{-1}dw). Total PAH concentrations in surface sediments of the Yellow Sea proper were comparable to that from offshore areas of California (149 ng g^{-1} dw) and the western Mediterranean Sea (179 ng g^{-1} dw). However, our values are an order of magnitude lower than those in the coastal area (Table 2). For example, in Inchon Harbor and its vicinity situated along the west coast of Korea, total PAH concentrations varied from 9.1 to 1400 ng g^{-1} dw with a mean value of 120 ng g^{-1} dw (Koh 1997). The highest concentrations were found in the inner Inchon Harbor due to the large population and high industrial activities in the region. By comparison phenanthrene in the North Sea ranged from 1 to 1500 ng g^{-1} dw and naphthalene varied from 1 to 55ng g^{-1} dw (North Sea Task Force 1993), and total PAH concentrations varied in the range of 32-4120 ng g^{-1}dw in coastal sediments from Arcachon Bay, France (Baumard *et al.* 1998).

Table 1. Distribution of polycyclic aromatic hydrocarbons in surface sediments of the Yellow Sea.

St.	LAT(°E)	LONG(°N)	Phenanthrene (ng g^{-1})	Anthracene (ng g^{-1})	Total PAH (ng g^{-1})
1	126	33.43	1.5	<0.5	6.6
2	125.17	33.02	2.4	0.7	54.1
3	124.33	32.67	1.6	0.7	37.7
4	124.67	33.2	2.6	<0.5	70.2
5	125.17	33.71	1.1	<0.5	3.1
6	125.66	34.58	<0.5	<0.5	1.8
7	125.16	34.58	3.0	0.7	9.6
8	124.33	34.58	3.8	0.7	13.8
9	123.5	34.58	2.1	<0.5	7.6
10	123.5	35.5	5.1	1.2	104.5
11	124.5	35.5	7.1	1.6	320.5
12	125.17	35.5	3.7	<0.5	82.5
13	125.83	35.5	3.7	<0.5	3.7
14	125.83	36.08	3.8	<0.5	281.1
15	125.17	36.08	2.5	<0.5	135.0
16	124.5	36.08	16.3	9.7	260.0
17	123.92	36.08	5.1	1.1	6.2
18	124.33	36.83	4.7	1.0	183.5
19	125.17	36.83	1.0	<0.5	12.3
20	126	36.83	1.0	<0.5	1.0

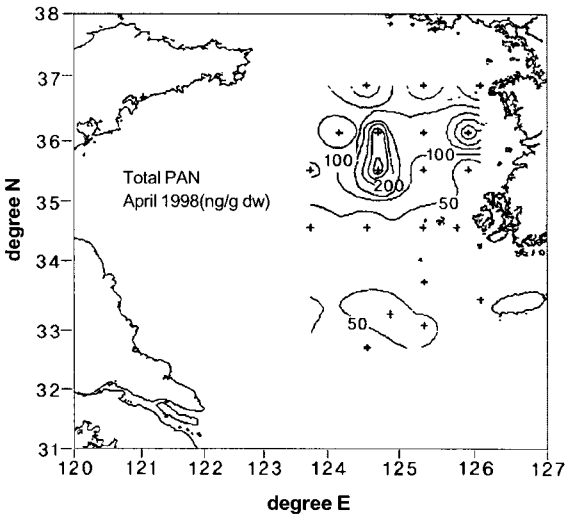

Fig. 2. Distribution of total polycyclic aromatic hydrocarbons in the surface sediments of the Yellow Sea.

Spatial distributions of total PAHs concentrations in bottom sediments of the Yellow Sea proper are shown in Fig. 2. The highest value for total PAHs was observed at St.11 in the central part of the sea. Elevated concentrations for phenanthrene and anthracene were also observed at

Table 2. PAH concentrations in the different areas.

Area	total PAH(ng g^{-1})	Reference
Eastern Yellow Sea	1-320.5	This study
Inchon Harbor (Yellow Sea Coast)	9.1-1400	Koh(1997)
Mediterranean Sea	179-3182	Lipiatou and Saliot(1991)
Bohai Sea	50-150	Zheng and Tang(1997)
Arcachon Bay, France	32-4120	Baumard *et al.*(1998)
San Diego Harbor, USA	5459-12802	NOAA(1991)
offshore area of California (100 km from the coast)	149	NOAA(1991)
Baltic Sea	800-1900 (muddy area)	Witt(1995)
Sedimentary toxic level	4000	Long(1992)

St.11 and St.16. Relatively high concentrations of phenanthrene were found in 1997 in the central part of the Yellow Sea (Yang 1997) as well. Organic carbon and trace elements in surface sediments collected during the same cruise showed elevated concentrations in the central muddy area of the Yellow Sea (Yang 1998; Yang *et al.* 1998). The distribution pattern of total PAHs concentrations in surface sediments generally reflected fine sediment grains characteristics. The central part of the Yellow Sea is covered with silty or clayey mud derived from the Huanghe River and/or Old Huanghe Delta (Lee and Chough 1989; Qin and Li 1986). Dominant bottom sediment is clay, however, progressively higher amount of silt toward the west is observed (Alexander *et al.* 1991). Since the affinity of hydrocarbons with fine sediment fractions is higher than that of coarser fractions, fine sediments in the central part of the Yellow Sea tend to accumulate hydrophobic compounds in much greater extents than sandy sediments in the eastern part of the sea as observed in other areas (Landrum and Robbins 1990; Colombo *et al.* 1989).

Various molecular indices that are source discriminant have been developed to more precisely assess the processes by which PAHs were originated. These indices are useful in distinguishing the relative importance of petroleum-derived petrogenic hydrocarbons versus combustion-derived pyrolytic hydrocarbons (Steinhauer and Boehm 1992). Generally phenanthrene concentration versus anthracene concentration (P/A) ratios of below 10 are indicative of incomplete combustion of the organic matter while the ratios of above 25 imply petrogenic hydrocarbons (Soclo 1986). Latimer and Quinn (1996) reported that, in Narragansett Bay, P/A ratios in sediments were mostly below 7 which were well below the level indicative of fresh petroleum (>50 for No. 2 fuel oil). The low P/A values (1.7-4.7) of PAHs in bottom sediment of the Yellow Sea proper are largely originated from the incomplete combustion of organic matter.

Table 3. Polycyclic aromatic hydrocarbons in sediment core of St. 16.

Depth (cm)	Phenanthrene ($ng\,g^{-1}$)	Anthracene ($ng\,g^{-1}$)	Total PAH ($ng\,g^{-1}$)
0-1	16.3	9.7	260.0
1-2	8.4	1.3	227.5
2-3	6.3	1.6	52.7
3-4	4.3	0.3	62.9
4-5	6.6	1.7	118.0
5-7	4.6	<1.0	4.6
7-9	3.8	0.5	4.2

Downcore depth distribution pattern of PAHs from the St. 16 showed that total concentrations of PAHs increased toward the surface of the sediment (Table 3). It is probable that total PAH inputs to the central Yellow Sea has been increasing in recent decades than the earlier period. Since phenanthrene is pyrolitic and much less subjected to degradation in the marine environment than petrogenic PAHs (Dachs *et al.* 1997), a more detailed time evolution of phenanthrene accumulation in the bottom sediment is presented in Fig. 3. Time constraints for each layer of sediments were given by the ^{210}Pb dating (Yang 1998). Phenanthrene concentrations increased as much as a factor of two in the last decade. Ohta *et al.* (1983) suggested that higher PAH concentrations in the upper layers of sediments compared to the deeper layers also correlated with the increases in the combustion levels of fossil fuels, and increases of petroleum products over coal has resulted in a such distribution pattern of PAHs in the sediment core in

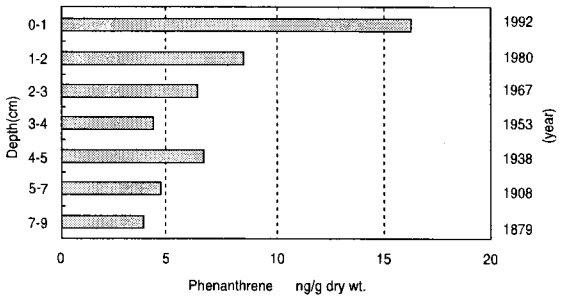

Fig. 3. Phenanthrene concentration in sediment core of St. 16.

Japan. In Korea, petroleum consumption has increased upto 1.5 times during 1991 - 1997 (Ministry of Environment 1998). A nearly quadruple increase of NOx emissions of the atmosphere, largely by combustion of petroleum products is projected from the year 1980 to the year 2020 in Asia (Galloway *et al.* 1994). Therefore, the recent increases of phenathrene accumulation in bottom sediments is most likely. But more core profiles of PAH concentrations in different parts of the Yellow Sea should be available to clearly indicate recent increases of the contaminant.

The annual PAH flux for open-sea core is derived from PAH concentrations and sedimentation rates determined from downcore ^{210}Pb profiles. The ^{210}Pb-derived sediment accumulation rate was 39.8 mg cm^{-2} yr^{-1} (0.04 cm yr^{-1}) at St. 16 (Yang 1998), which was representative values in the eastern central part of the Yellow Sea (Alexander *et al.* 1991). Accordingly, the annual PAH flux at St. 16 was estimated to be 166 μgm^{-2}yr^{-1}. The calculated PAH accumulation rate was comparable to those of other offshore areas. Burns and Villeneuve (1983) reported PAH flux of 690 μg m^{-2}yr^{-1} for a seafloor in 250 m water depth located off Monaco. In the western Mediterranean Sea, the fluxes of PAHs varied from 164 μgm^{-2}yr^{-1} for open-sea sediments, up to 1420 μg m^{-2}yr^{-1} in the estuarine area of Rhone delta (Lipiatou and Saliot 1991). In the relatively pristine remote coastal areas in the northeastern USA, an average flux for individual PAH varied from 8 - 30 μgm^{-2}yr^{-1} (Gschwend and Hites 1981). In urbanized freshwater and marine environment PAH fluxes were much higher, for example, total PAH flux of 900 to 65000 μgm^{-2}yr^{-1} (Latimer and Quinn 1996) in Narragansett Bay, U.S.A. was reported.

4. Conclusions

We have observed the presence of pyrolytic PAHs in bottom sediments of the Yellow Sea and relatively high concentrations in the central muddy area. Annual total PAH flux to the sediment was comparable to other offshore areas of the world. However more information on the water column concentrations, bioaccumulation in the commercially important fish and shellfish species, transport pathways of PAHs including atmosphere and river as well as diagenetic processes in the sediments should be studied to understand the fate and human impact of PAHs in the Yellow Sea System.

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