

PCBs and its coupling with eco-environments in Southern Yellow Sea surface sediments

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Abstract

In this paper, the spatial distribution and source of the PCBs in surface sediments of the Southern Yellow Sea (SYS) and influencing factors, such as the sediment characteristics (components, relative proportions and total organic carbon contents), and hydrodynamic conditions were analyzed. PCB concentrations in the surface sediments ranged from 518–5848 pg/g, with average values of 1715 pg/g decreasing sharply compared to last year. In the study area, the PCB pollution level in the middle area was the highest, followed by that of the east coast and the west coast, respectively. Although the PCB level in the coastal areas was lower than that in the middle areas, it was proven in our study that the Yellow Sea obtained PCBs by virtue of river inputs. There was a positive and pertinent correlation between the clay proportion and PCB concentrations, and the increase of the PCB concentrations was directly proportional to the increase of TOC contents, with $r = 0.61$, but it was contrary to the sediment grain size. Consequently, the factors controlling PCB distribution had direct or indirect relationships with sediment grain size; moreover, the hydrodynamic conditions determined the sediment components and grain size. In conclusion, hydrodynamic conditions of the Yellow Sea were the most important influencing factors effecting the distribution of PCBs in the surface sediments of the SYS.

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1. Introduction

Polychlorinated biphenyls (PCBs) are one group of anthropogenic aromatic congeners with the H atoms of the biphenyl rings substituted for Cl atoms. According to the positions and numbers of Cl atoms on the backbone, PCBs can be composed of any of 210 congeners. By now, 209 congeners have been synthesized, but not all of them are commercially available (Alkhatib and Weigand, 2002). PCB commercial production started in the 1930s, and their unlimited usage was banned by the end of the 1970s. During those 4 decades, it was estimated that about

1,200,000 t of PCBs were synthesized to satisfy the demands of industry and agricultural production (Cummins, 1998). Due to their structures, specifically the strengthened C=X bonds caused by p- π conjugation between single pair electrons and greater π bonds, PCBs are very resistant to acids, bases and oxidants (Li and Xie, 2004). Therefore, although PCBs have not been produced since their usage history, they will remain in the environment for a long time. PCBs tend to be accumulated and magnified through trophic pyramids, and then taken up by humans and animals via the food chain, where they are retained in tissues, blood, and milk by virtue of PCBs' lipophilic nature, inducing some pathological system changes which can even include canceration of the skin, digestive system, neural system, procreation system and the immune system (Zheng et al., 2004). Because of their semi-volatility,

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PCBs can travel long distances and deposit in areas very far from the discharging spots through atmospheric movement, even in the unfrequented polar region; furthermore, PCBs also can be transported by way of animal behaviors, for example, sockeye salmon migration (Krümmel et al., 2003).

PCBs enter air, water, and soil during their industrial manufacture, usage, and improper disposal, from accidental spills and leaks during their transport, and from leaks or fires in products containing PCBs. Once released into the aquatic environment, PCBs bind to sediment particles (Alkhatib and Weigand, 2002). Sediments are types of rather heterogeneous adsorbents, and they act as the main reservoir of various hydrophobic organic pollutants (HOPs) and heavy metals, especially some persistent organic pollutants (POPs), which affect the aquatic environment quality chronically through their partition balance between water and sediment phases (Ma et al., 2001a). The factors controlling PCB distribution remarkably include the sources and modes whereby PCBs entered the environment, the behaviors of transfer, diffusion and agitation, chemo-degradation and bio-degradation, PCB congener characteristics (solubility and volatility), sediment characteristics (grain size, shape, frame and chemical components) and the sediment's environmental characteristics (depth, sedimentation modes of particles, ocean currents and vortex) (Hu, 1994).

The PCBs' sources play an important role in controlling their distribution pattern. The approaches possible for PCBs to enter an aquatic environment include atmospheric transport and deposition, direct and indirect discharge, river input and bio-behaviors (Petrena et al., 2003). It was reported that 85–95% of PCBs in Lake Superior was from atmospheric deposition, which also contributed 58–63% found in Lake Michigan and Lake Houston (Bi and Xu, 2000). However, the Taihu Lake (Zhang and Lin, 2004), Guanting Reservoir (Ma et al., 2001a), and Shiwa Lake (Koh et al., 2005) obtained the majority of their PCBs from river inputs, which were deeply impressed with spot- and line-loading characteristics.

The sediment characteristics also influence the PCB distributions to a large extent. For example, many earlier studies presented evidence that the PCB concentration in surface sediments increased with decreasing of the grain size of the sediment particles, which was contrary to the clay proportions and total organic carbon (TOC) contents (Piérard et al., 1996; Camaho-Ibar and McEvoy, 1996; Petrena et al., 2003). In addition, the characteristics of the sediment's environment affect PCB distribution indirectly; these include the Eh, sheer force, and microorganism population and so on.

Because of their toxicity and persistence, PCBs have been included in many blacklists, such as POPs, priority controlling pollutants and suspected environmental endocrine disrupters (EEDs) (Kavlock et al., 1996). After the implementation of PCB control became an important task following ratification of the Stockholm convention, PCBs

have been of global interest and extensive studies have been conducted on them in the sediments of many rivers, estuaries and lakes, but there is limited published data available for PCBs in the Yellow Sea. The objectives of our study were to: (1) determine the PCB spatial distribution in the surface sediments of the SYS; (2) determine if the level of PCBs in the surface sediments was associated with the sediment characteristics, help the Chinese and Korean governments understand the PCBs' behavior in the sediments and lay a better foundation for understanding PCBs negative effects in the offshore areas environment; and (3) trace the sources of PCBs in the SYS. To achieve these objectives, we studied the PCB distributions, the coupling relationships between PCBs and the sediment components, relative proportions of sediment components, grain sizes, TOC contents, and the hydrodynamic conditions of the Yellow Sea.

2. Material and methods

2.1. Study area

The Yellow Sea is a semi-enclosed epicontinental sea of the northwestern Pacific, connecting with the Bohai Sea in the east. It is located between North China Continent and Korea Peninsula, and is divided into the Northern Yellow Sea and the Southern Yellow Sea by a line linking the Qidong Mouth of the Changjiang Estuary and Cheju Island, whose names come from their positions. The SYS covers 309,000 m², with an average depth 46 m and a maximum depth of 140 m in the northern area of Cheju Island (Qin et al., 1989; Song, 1997). The Yellow Sea receives a vast amount of fresh water from both Chinese and Korean Rivers, such as the Changjiang River, Yellow River, Yalujiang River, Daliaohe River and Haihe River of China and the Han, Kum and Yeongsan Rivers of Korea (Qin et al., 1989). There is good reason to believe that the water exchange between the Yellow Sea and open seas is quite slow because of its semi-enclosed position; thus, most pollutants from river inputs accumulate in the sediments gradually. In order to achieve the projected objective, populations of 33 sample stations on four lines (A, B, C and D) were designated according to their latitude from 33°34.7' N to 36°55.5' N (Fig. 1). In the four lines, "A" was set to evaluate the influence of the Han River and similarly, the "D" line was for the Changjiang River. The sediment samples were collected in the study area with grab samplers and the sea water samples were collected with Go-flow seawater samplers between October 9th and 23rd, 2004. The surface sediments (1–3 cm depth) were removed using a stainless steal spoon and then immediately frozen at –20 °C.

2.2. PCB determination

In the laboratory, the solid samples were frozen dry. Ten grams of dried sediments of every sample was taken, with

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