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Polychlorinated biphenyls in sediments of the Yellow Sea: Distribution, source identification and flux estimation

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ABSTRACT

Although there is no direct major riverine input, a large quantity of waste produced in mainland China and Korea is transported continuously to the Yellow Sea (YS) through atmospheric deposition, currents and tides; therefore, the environment is distinctly influenced by man-made pollution. This study focuses on the associated pollutant transport mechanisms and fluxes by sampling polychlorinated biphenyls (PCBs). PCBs (Σ_{24}PCB) in YS sediments ranged between 99 pg/g and 3.13 ng/g of dry sediment (with a mean value of 715 pg/g). PCBs produced unintentionally by industrial and other processes appeared to be the major source of PCBs in the sediments, accounting for 60.5%. Industrially synthesized PCBs with 3Cl and 5Cl accounted for 15.5% and 24.0%, respectively. PCBs were mainly from atmospheric deposition (84.5%), followed by continental runoff (15.5%). The average atmospheric deposition flux of technical PCBs was 789 ng/(m^2 a) and flux of surface runoff was 2.27 ng/L.

1. Introduction

In China, ca. 10,000 tons of polychlorinated biphenyls (PCBs) were produced from 1965 to 1974 (production was banned in 1974), with 9000 tons being trichlorobiphenyls and 1000 tons being pentachlorobiphenyls (Mai et al., 2005). Meanwhile, PCBs were produced and used in relatively small quantities on the Korean peninsula (Breivik et al., 2002a). PCBs were primarily used for insulating liquids in power capacitors and additives in paint. Following the ban on production and use, most of the outdated PCB-containing equipment (i.e., equipment filled with PCBs as dielectric fluid) were removed from use and stored. However, large PCB amounts entered the environment during their manufacture and use (Wong and Poon, 2003). PCBs can now be found in low concentrations in virtually every sphere of the environment (Beyer and Biziuk, 2009). Recent studies have also shown that a large amount of PCBs is unintentionally produced by human activity (Aries et al., 2006; Hong et al., 2005; Liu et al., 2009). Additionally, evidence from studies (Lohmann et al., 2007; Mai et al., 2005) indicates that the composition and concentration of PCBs in the environment is directly related to anthropogenic pollution.

Most PCBs ultimately settled on the continental shelf after their release to the environment (Jönsson et al., 2003). Hence, environmental cycling of PCBs and measured levels can be significantly influenced by organic geochemical conditions (Colomboa et al., 2007; Hu et al., 2011; McCuskera et al., 1999). The Yellow Sea (YS) lies between the Chinese mainland and the Korean Peninsula. Although the Yellow River and Yangtze River (YR) in China do not run directly into the YS, the rivers noticeably influence the sediment sources in the YS (Hu and Su, 1999; Lim et al., 2007). Moreover, a large amount of municipal and industrial toxic waste materials generated by land-based activities are constantly discharged to the ocean. According to previous investigations (CSOA, 2007; MEPPRC, 2007), approximately 7.6% of this waste ends in the YS. In Korean coastal areas, PCB contamination is closely related to shipping and industrial activities (Hong et al., 2005, 2006).

A few studies have focused on surveying PCBs in the YS (Zhang et al., 2007). According to Hu et al. (2011), shelf mud deposition and large rivers have a dramatic impact on the fate of pollutants in the sediments of the YS. Zhang et al. (2007) investigated PCBs and their coupling with eco-environments in YS surface sediments; they obtained evidence that organic carbon was the most important factor in PCB levels. While that study has advanced our understanding of the PCB levels in the YS, a significant gap exists in our knowledge of the sources and fluxes of PCBs. The objectives of this work were to assess the spatial distribution pattern of PCBs in the YS, identify PCB sources and further estimate the PCB flux.

2. Methods

2.1. Sampling

We were particularly interested in the influences of atmospheric deposition and surface runoff on transporting PCBs to the

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YS. Therefore, we collected surface sediment samples from mud areas. A description of the sampling stations is shown in Fig. 1. Samples were collected in April 2011 with a stainless steel box-corer. Efforts were made to minimize disturbance of the surface sediment layers. All of the samples were wrapped in aluminum foil after collection and immediately stored at −20 °C until analysis.

2.2. Chemical analysis

A detailed description of the procedures for extraction, purification, and measurement of PCBs in sediment samples has been provided elsewhere (Duan et al., 2012, 2013). Twenty-four individual congeners were quantified. According to the IUPAC nomenclature, these are PCB5, 18, 28, 31, 44, 52, 66, 77, 87, 101, 110, 112, 118, 138, 141, 151, 153, 155, 170, 180, 183, 187, 198 and 206.

2.3. Source assignment

Partial least-squares regression (PLSR) was applied to the source apportionment of PCBs. A detailed account of the algorithm was given by Wold et al. (2001).

3. Results and discussion

3.1. Occurrence and concentration of PCBs

The concentration of total PCBs (Σ_{24}PCB) ranged between 99 pg/g and 3.13 ng/g (with a mean value of 715 pg/g; Fig. 2). The values were comparable with the concentration range of most surface sediments collected within the Yangtze River estuary (YRE) (Xu et al., 2000; Zhang et al., 2009) and the Yellow River estuary (Ma et al., 2001), but much lower than the concentration in sediments from highly urbanized areas such as the Pearl River Delta (11.5–48.3 ng/g; Kang et al., 2000) and coastal sediments (3.5–25.1 ng/g) around Hong Kong (Connell et al., 1998; Hong et al., 1999). Seven indicator PCB congeners (Σ_7PCB, CB28, 52, 101, 118, 138,153 and 180) accounted for 9.3–61.6% of Σ_{24}PCB; the mean value was 33.3%. The congener profile of the indicator PCBs was dominated by CB52 and CB101 (with mean values of 89 pg/g and 63 pg/g, respectively), which belong to the tetra-PCBs and penta-PCBs, respectively. Another five indicator congeners accounted for an average of 8.0% of Σ_{24}PCB, which was far below the contribution of CB52 and CB101.

PCB concentration increased from the edges to the central study area. The highest PCB concentrations were primarily at 35°N and 36°N latitudes. The PCB distribution pattern was similar to those of polycyclic aromatic hydrocarbons (PAHs; Kang et al., 2009) and organo-chlorine pesticides (OCPs; Hu et al., 2011). The high Σ_{24}PCB in the central YS was associated with small particle size, high TOC and TON contents (Fig. 2a, b, d and e).

Over the entire study region, the minimum PCB level appeared at 34°N. The TOC/TON ratio was between 5.4 and 7.7 (Fig. 2f). In general, the ratio varies from 4 to 10 in the organic matter (OM) released by phytoplankton, whereas the ratio exceeds 10 in terrestrial OM. Therefore, TOC/TON is extensively used to reflect the sources of OM in sediments (Marinari et al., 2006). As indicated by TOC/TON ratios, the OM mainly originated from phytoplankton in our study region. In the area near 34°N and neighboring water

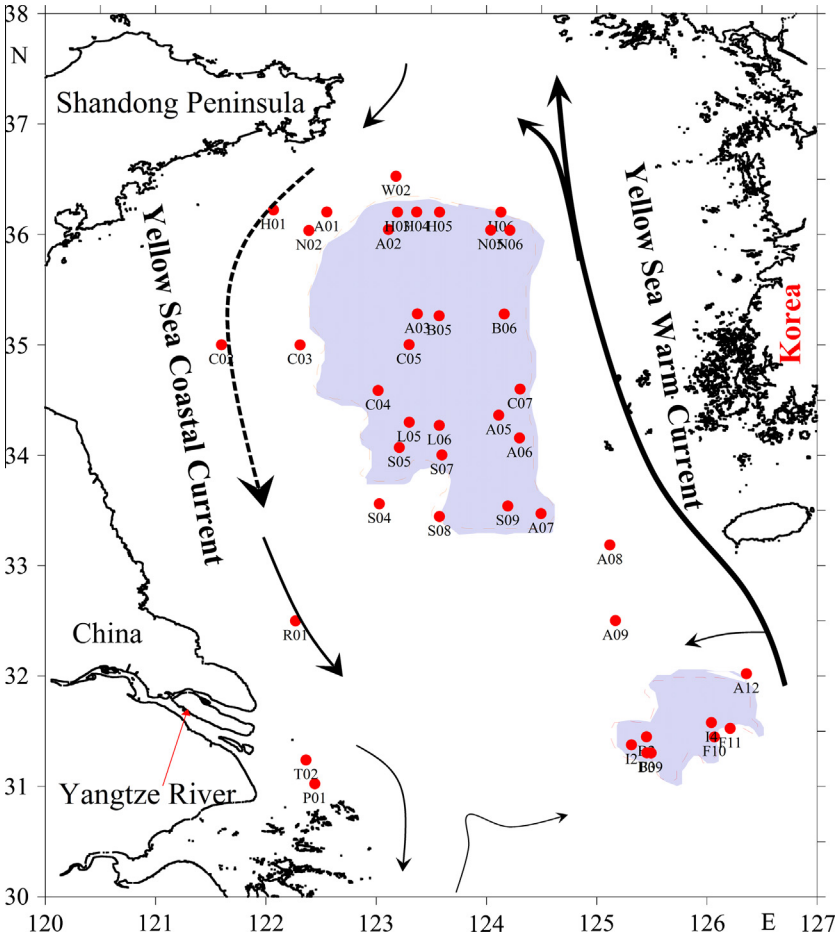

Fig. 1. Location of sampling sites (dots). Circulation systems and mud areas (in light gray) follow Liu et al. (2007) and Cheng et al. (2004).