Science of the Total Environment 598 (2017) 12–20

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Spatial-temporal variability and transportation mechanism of polychlorinated biphenyls in the Yangtze River Estuary

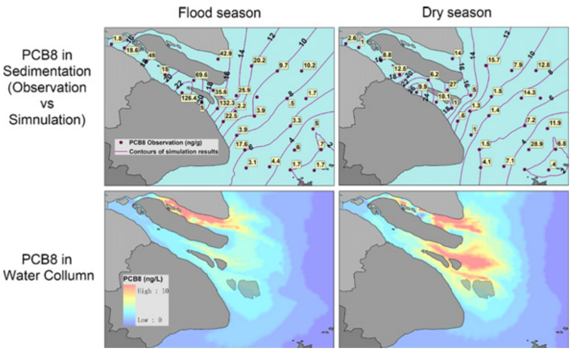
Lei Chen [a], Ye Yang [a,b], Jing Chen [a], Shuohan Gao [a], Shasha Qi [a,c], Cheng Sun [a], Zhenyao Shen [a,*]

[a] State Key Laboratory of Water Environment, School of Environment, Beijing Normal University, Beijing 100875, PR China
[b] Department of Civil Engineering, The University of Hong Kong, Hong Kong, PR China
[c] China Offshore Environmental Services LTD, Beijing, PR China

HIGHLIGHTS

- The temporal-spatial variability of PCBs were illustrated in the Yangtze estuary.
- The transportation mechanism of PCBs were explored by numerical simulation.
- The impacts of upstream Dam on estuarine PCBs were quantified.
- The importance of hydro-sediment conditions on PCBs pattern were highlighted.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 3 December 2016
Received in revised form 27 February 2017
Accepted 11 April 2017
Available online 20 April 2017

Editor: D. Barcelo

Keywords:
Polychlorinated biphenyls (PCBs)
Sediment
Yangtze Estuary
Three Gorges Dam
Environmental Fluid Dynamic Code (EFDC)

ABSTRACT

Although the variability of polychlorinated biphenyls (PCBs) is strongly dependent on the hydro-sediment pattern, the quantification of this interaction is still not well described, especially for estuary areas. In this study, both chemical analyses and numerical simulation were conducted to explain the temporal-spatial variability and transportation mechanism of PCBs in the Yangtze River Estuary (YRE). The impacts of the upstream Three Gorges Dam (TGD) on estuarine PCBs were also addressed with a simulated scenario. The results showed that the PCBs levels in the YRE were relatively low or moderate and the highest levels were related to the maximum turbidity zone. The spatial variability of PCBs is strongly dependent on the hydrological circulation, which resulted in a declining trend from the inner YRE to the adjacent sea. The seasonal variability of PCBs could be due to the joint influence of the current and the erosion/deposition environment. The opposite temporal trends of the overlaying water and sediment are driven by the seasonal characteristics of hydro-sediment patterns. The simulated results also indicated that the distribution, fluxes and transport ability of PCBs in the South Branch changed as a result of the sediment discharge reduction after construction of the TGD.

1. Introduction

Polychlorinated biphenyls (PCBs) are a classic group of persistent organic pollutants that have been used extensively for industrial applications since 1930 (Combi et al., 2016). Owing to their bioaccumulation and toxicity (Fu et al., 2003; Yuan et al., 2015),

---

* Corresponding author.
E-mail addresses: chenlei1982bnu@bnu.edu.cn (L. Chen), zyshen@bnu.edu.cn (Z. Shen).

http://dx.doi.org/10.1016/j.scitotenv.2017.04.069
0048-9697/

PCBs have been designated as a global environmental problem and also prior pollutants by the Stockholm Convention (Wang et al., 2015). Despite the prohibition of their production in most countries, PCBs continue to be detected because of their high persistence, thermodynamic stability and environmental mobility, and their levels are not expected to decrease significantly in the next few decades (Gao et al., 2013).

The pattern of PCBs has been researched worldwide, indicating the temporal-spatial variability of PCBs differs over large domains (Muir et al., 1988; Nellier et al., 2015; Reijnders, 1986; Shoeib and Harner, 2002; Zhang et al., 2004). Thus, explaining the variability of PCBs is a challenging task because of the inherent complex physical processes (Chen et al., 2015a). Comparatively, the variability of PCBs in estuarine ecosystems has been studied to a lesser extent, usually within delimited regions (Granek et al., 2016; Hong et al., 1995; Kaiser et al., 2016). First, estuaries, as the transition zones between land and ocean, are strongly susceptible to the exchanges between upstream river and the adjacent sea (Granek et al., 2016; Kaiser et al., 2016). The ocean currents, waves, tides and seawater influx often have a strong, overlapping effect on the fate of PCBs. Second, due to the high hydrophobicity and molecular mass, PCBs are often absorbed onto particulate matter and accumulate in bottom sediments (Hawker and Connell, 1988). Surface bottom sediments, which reflect the current status of PCBs, are considered an optimal medium to evaluate the variability of PCBs. Previously, most investigations were conducted by analyzing dated sediment or peat cores in coastal systems (Breivik et al., 2002). However, because of the water circulation, sediment movements in estuarine areas are more complex than in other water bodies. During specific seasons, bottom-deposited PCBs can even be transported to the open sea (Combi et al., 2016). Moreover, bottom sediment functions as a reservoir that releases PCBs into the water column through erosion and diffusion (Connolly et al., 2000). Despite widespread recognition of the direct and indirect pathways, the impacts of hydro-sediment conditions on estuarine PCBs are still poorly known.

The Yangtze River is one of the largest rivers in the world by discharge volume and the Yangtze River Estuary (YRE) is its lower, tide-affected section, which provides key services for global ecosystems and local economic growth (Qian et al., 2016; Wang et al., 2015). In the YRE, the formation and evolution of PCBs have been investigated, highlighting the impacts of specific factors such as sediment grain sizes and organic carbon content (Gao et al., 2013; Qian et al., 2016; Wang et al., 2015). However, these studies fail to explain the majority of the variability in PCBs because the variability was closely related to complicated local hydro-sediment processes. First, the fate of PCBs is influenced by the interaction of the Yangtze Diluted Water and the Taiwan Warm Current, in addition to other chemical-biological processes (Chen et al., 2015a; Ye et al., 2015). The intrusion of seawater occurs frequently because of the local bottom topography. Although the monitoring and assessment of PCBs have been reported in the YRE, there is no comprehensive investigation on their transportation mechanism. Second, the river flow and sediment discharge from the upstream Yangtze River have changed greatly in recent years, largely owing to the construction of the upstream Three Gorges Dam (TGD) (Gao et al., 2013; Lu et al., 2011; Zhang and Lou, 2011). It was reported that the impoundment of the TGD trapped 60% of the sediment influx during flood seasons (Lu et al., 2011). Thus, the TGD might have far-reaching effects not only on the regime of downstream flow and sediment but also on the characteristics of PCBs in the YRE. To date, there is no direct evidence for the status of PCBs associated with the regulation of the TGD.

In this study, we provide the first identification of the spatial-temporal variability and transportation mechanism of PCBs in the YRE. Generally, a hydrological model acts as a valuable supplement to field sampling; thus, both sediment sampling and numerical simulation were conducted to investigate the factors regulating PCBs and the impacts of the TGD.

2. Materials and methods

2.1. Description of the Yangtze River Estuary

The YRE, located in the eastern region of China (30°40′N–31°40′N, 120°55′E–123°00′E), extends from the upstream non-flood tidal boundary to the subaqueous delta, where the interaction of fresh water and saline water occurs (Fig. 1a). As a branching estuary, the YRE is characterized by a tree-tier bifurcation with four openings to the outer East Sea. First, the YRE is divided into the North and South Branches by Chongming Island. Then the South Branch is separated into the North and South Bays, and the South Bay is further divided into the North and South passages by an intertidal region. The YRE is in the eastern subtropical monsoon climate region with distinctive seasons. Owing to the variation of topography, a symmetric/irregular asymmetric semi-diurnal tide is observed outside/inside the river mouth. The largest tidal range (5.80 m) occurs in the North Branch because of its trumpet-shaped channel.

As shown in Fig. 1b, a large domain was selected that encompasses the entire YRE, Hangzhou Bay, and the adjacent area of the Yellow Sea and the East China Sea. The upstream boundary was set at Datong Station, which is the key hydrological station that records approximately 95% discharge of the Yangtze River Basin. The daily discharge and sediment data recorded at the Datong and Qiantang River Stations was collected from the Yangtze Water Resources Committee. Since the rate of water withdrawn by Shanghai city is <0.5% of the Yangtze River discharge, it was not considered. To reproduce the PCBs dynamics, the northern, southern, and eastern open sea boundaries were set at approximately 32.5°N, 29.25°N and 125°E, respectively, covering a domain area of 144,817 km^2. The water levels at the outer sea were derived from the GOTIC module and used to configure the open boundary of the inner numerical simulation (Matsumoto, 2001). The bathymetry data were obtained from the Maritime Safety Administration of People's Republic of China. The sea water temperature and salinity at the open boundary were derived from the Marine Atlas of East China Sea. The six-hourly atmospheric data, including wind speed, wind direction, atmospheric pressure, precipitation, air temperature, solar radiation, relative humidity and cloud cover, were collected from the NCEP Reanalysis Project of NOAA's National Centers for Environmental Prediction to set for boundary conditions. A flat free water surface was set as initial condition for cold start, while the spatial distribution of salinity initial condition was estimated based on the Marine Atlas of the Bohai Sea, Yellow Sea, and East China Sea.

2.2. Sample collection and analysis

To assess the variability of PCBs, thirty locations were selected, 20 sites (sites No. 1–18, 27, 28) within the inner YRE and the adjacent sea, while 10 sites were around the −10 m and −20 m isobaths in the sea (Fig. 1c). At least one sediment samples were collected from each location in August 2010 (flood season) and February 2011 (dry season) so over thirty representative samples were obtained from the YRE during each period, respectively. A global positioning system was used for locating these sites. Using a specially designed cylindrical grab sampler, the top 5-cm layer of sediment was collected to represent the current depositional environment. All samples were placed into precleaned glass jars with aluminum foil liners in the lid to avoid potential leaching and stored at −20 °C during transportation to the laboratory and until processing and analysis.

The PCBs in freeze-dried samples were extracted as the procedure described in our previous study (Gao et al., 2013). Briefly, 20 g of sieved sediment sample was ground and homogenized with 200 mL of hexane/acetone (1:1, V/V) for 24 h. The extract was concentrated using a rotary