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Status of POPs accumulation in the Yellow River Delta: From distribution to risk assessment

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ABSTRACT

The Yellow River Delta (YRD) is a large region of China with complex pollution sources and a long history of environmental deterioration. Despite this, relatively little data exists on the status of important contaminants of concern in this region. Here, we review the literature on the status of key persistent organic pollutants (POPs) of concern including organochlorine pesticides (OCPs), polycyclic aromatic hydrocarbons (PAHs), and polychlorinated biphenyls (PCBs) in the YRD. Sources, source identification methods, and spatial distribution patterns are presented. Additionally, POPs contamination levels reported in the literature were evaluated against popular regulatory limits worldwide to form a basis for overall environmental health. Our review determined that OCPs in the YRD originated mainly from current pesticide use and past agricultural pesticide application. Sources of PAHs included petrochemical inputs, coal fired plants, and wood combustion. PCB levels were impacted by the petrochemical industry as well as waste disposal of PCB containing equipment. OCPs exhibited a spatial distribution pattern that increased along the urban–rural gradient, while the opposite was seen for PAHs and PCBs. Comparisons of POPs contamination levels in the YRD with popular regulatory limits suggest that the extent of PCB contamination all mediums (sediment, soil, water, and biota) exceeded that of PAHs and OCPs. Overall pollution levels in the YRD seem to be in control; however, levels from heavily polluted point sources raise numerous concerns about the ecological health of the region and require more attention from regulatory authorities.

The Yellow River Delta (YRD) is a typical ecotone of the Yellow River and Bohai Bay (Xie et al., 2012a) suffering from enormous environmental deterioration from industrialization. With an area of 2200 km^2, the watershed of the YRD is the largest and youngest wetland in the warm temperate zone of China (Yuan et al., 2012), and is also the youngest land in the world. Extremely high sediment loads (>1000 million tons per year, Mt/year) have been discharged to the sea over thousands of years as a result of poor agricultural practices in the Loess region (Oliva et al., 2015). High sediment loading from the river and transport by hyperpycnal flows into the estuary have made considerable contributions to the deltaic depositional system (Fan et al., 2009). In the environment, persistent organic pollutants (POPs) tend to adsorb organic matter due to their chemical properties. Once they enter aquatic environments, they preferentially sorb to organic particles and are deposited in sediments. Thus, large quantities of organic pollutants enter the Yellow River through this sediment loading (Yang et al., 2009). In China, dichlorodiphenyltrichloroethane (DDT) and hexachlorocyclohexane (HCH) were the most widely used and produced organochlorine pesticides (OCPs). These OCPs were produced for use in agriculture, forestry, and public health for more than four decades (Cai et al., 2008).

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Due to their bioaccumulation potential, persistence, toxicity and long-range environmental transport ability, POPs are of great concern to scientists, environmental quality managers and policy makers (IPCS, 2000; Keita-Ouane et al., 2001; UNEP, 2003; Peterson et al., 2010). Measurement of their concentrations and evaluation of ecological risk in a large-scale region is of significant importance to protect environmental health (Sudaryanto et al., 2011). A large amount of work has been carried out regarding state of POPs globally. The United Nations Environment Program (UNEP) (UNEP, 2003), International Program on Chemical Safety (IPCS) (IPCS, 2000), Intergovernmental Forum on Chemical Safety (IFCS) (Peterson et al., 2010), Inter-Organization Program for the Sound Management of Chemicals (IOMC) (Keita-Ouane et al., 2001) and other international organizations are actively involved in the research of characteristics, hazards, fate, etc. of POPs.

With the rapid economic development in China over the last few decades, environmental pollution caused by POPs has been identified in several delta areas, e.g., the Mekong Delta (Cenci et al., 2004), the Pearl River Delta (Yang et al., 2007), and the Yangtze River Delta (Yang et al., 2009). Most investigations of pollutants in the YRD have focused on three main POPs of concern: OCPs, polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) (Fan et al., 2009; Hu et al., 2010; Liu et al., 2010). It is important to note that the Shengli Oil Field, the second largest oilfield in China, is located in the YRD. Given the scale of mining and oil refining activities occurring here, it is of significance to assess the ecological risk of petroleum contaminants in this area. A comprehensive investigation of the distributions of POPs in the YRD will be useful to determine the sources, evaluate ecological risks, and create remediation strategies.

This review is organized as follows: Firstly, previous usage of POPs and potential pollution sources in the YRD are described. Secondly, environment occurrence is introduced on a matrix-specific basis, i.e., soil, sediment, water, and organisms. Thirdly, environmental sources are identified mainly focusing on historical agriculture usage, technical production, atmospheric deposition, and riverine transport. Finally, potential environmental effects are examined through comparisons between environmental quality guidelines and levels of POPs in environmental matrices.

The YRD is a rapidly developing petrochemical and agricultural area of China with a long history of pollution and complex sources of contamination (Fig. 1). From 1960 to 1990, large amounts of lindane, dicofol, and technical DDT in China were produced as well as widely used for pest control. It has been estimated that approximately 258,000 tons of HCH and DDT were used in China in 1990, accounting for 74.0% of all pesticide use (Tao et al., 2007). Agricultural usage of HCHs and DDTs lasted from 1953 to 1993 in the YRD (Tao et al., 2006).

PAH contamination is derived from petroleum sources (i.e., petrogenic sources) and from incomplete combustion of carbonaceous materials (i.e., pyrogenic sources). These sources occur naturally and as a result of human activities (Edwards, 1983). Cracking and refining of petroleum and incomplete combustion of fossil fuels are known to produce large amounts of PAHs (Xie et al., 2012a). Crude oil cracking sites, oil wells, and petrochemical industrial activities are point sources of PAHs, which can increase regional levels of PAH contamination through short-range and long-range transport (Wilcke, 2007). The Shengli Oilfield is arguably the largest point source of PAH contamination in the YRD. Oil well blowouts, leaks and spills from underground tanks, pipelines and illegal disposal threaten the ecosystem in this area. Manufacturers in the YRD have by-produced PAHs (Hu et al., 2005). Effluents from several production facilities were treated ineffectively and wastewater discharged from these facilities contributed large amounts of PAH contamination (Li et al., 2006). Sewage irrigation was used at a very large scale in this area for over 30 years because of the shortage of water resources (Gong et al., 2004).

PCBs have a long history of extensive production and use in China. It has been estimated that 10,000 tons of PCB 3 (9000 t) and PCB 5 (1000 t) were produced in China from 1965 to 1974 (Xing et al., 2005). PCB 3 was used primarily in power capacitors applied in electricity production, distribution and transmission, and PCB 5 was used mainly as a paint additive in China (China SEPA, 2003). Furthermore, following the ban on the production and use of PCBs, most of the outdated PCB-containing equipment (equipment filled with PCBs as dielectric fluid) was removed from use and stored. Potential POPs' pollution sources at YRD are listed in Table 1.

Soil is the largest sink in the environment and significantly influences human and environmental health. In China, OCPs, PAHs and PCBs were the most frequently studied POPs in soil (Xing et al., 2005), and concentrations of POPs in soil have been increasing for 100–150 years, especially in urban areas (Cai et al., 2008). In the YRD, more than 400 soil samples collected from different cities and regions have been analyzed for the occurrence of OCPs, PAHs and PCBs.

Soil OCPs have been investigated in at least 10 studies. DDTs (*p,p′*-DDT, *o,p′*-DDT, *p,p′*-DDE, *o,p′*-DDE, *p,p′*-DDD, *o,p′*-DDD) and HCHs (α-HCH, β-HCH, γ-HCH, δ-HCH) were the main contributors. Occurrences of OCPs in soil in various parts of YRD are described in Table 2 (all concentrations are normalized to dry sample weight). Concentrations of DDTs ranged from 0.05–3819 ng/g (Fig. 2). Spatial distribution varied greatly and rendered a clear upward trend along the urban–rural gradient (Chen et al., 2011; Xie et al., 2012b). DDT concentrations in rural areas varied between 4.10–3818.76 ng/g (with a mean of 315.65 ng/g), while that in the city varied from 3.34–533.53 ng/g (with a mean of 63.81 ng/g) (Chen et al., 2011; Yuan et al., 2011). Among DDTs, the predominant compound was *p,p′*-DDE, which accounted for 63.0%–82.7% of the DDT concentration, followed by *p,p′*-DDT, and lastly was *p,p′*-DDD.

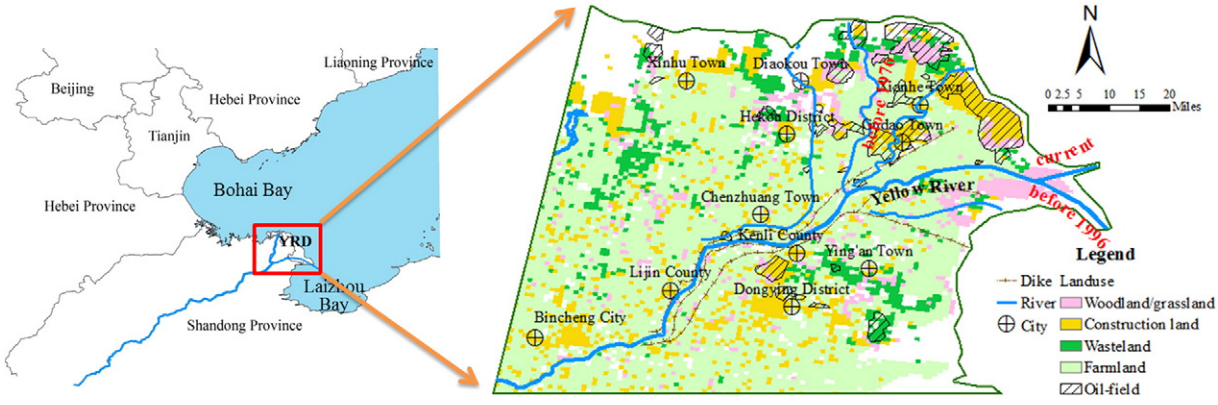

Fig. 1. The location of YRD and the shifting history of the Yellow River.